# **Chapter 13 Soil Contamination and Conservation**



Yongfen Wei and Huijuan Shao

**Abstract** Healthy soils are essential to food security and provide significant contributions to the Sustainable Development Goals (SDGs) by conserving biodiversity, improving water quality, and enhancing resilience against extreme weather. However, the introduction of contaminants from various sources into the soil environment has degraded the soil ecosystem and endangered human health. Soil contamination often cannot be visually detected; therefore, soil must be properly utilized, appropriately managed, comprehensively studied, and efficiently conserved. This chapter firstly introduces the types and sources of soil contamination, global soil contamination status, and soil remediation technologies. It then briefly describes the soil contamination practices implemented to address this problem. Furthermore, the results of our previous research on treating Cs contaminated soil using industrial waste materials are introduced. Finally, the efforts and challenges in managing and controlling soil contamination are presented to provide reference for soil remediation.

**Keywords** Soil contaminants · Soil remediation · Cesium · Recycling material · Immobilization · SDGs

# 13.1 Introduction

Humans have considerably changed the soil environment to acquire benefits. These changes have caused unintended negative consequences to soil property and most organisms, thereby challenging their survival. Soil contamination is a classic example of such a scenario because in many cases it is not directly observed, and its effects could become apparent after several decades since its commencement.

Y. Wei (🖂)

H. Shao College of Resources and Environment, Shandong Agricultural University, Tai'an, China

River Basin Research Center, Gifu University, Gifu, Japan e-mail: weiyf@gifu-u.ac.jp

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Unfortunately, 25% of global soils have been reported to be contaminated. Moreover, in many parts of the world, several decades of industrial activities, farming activities, and improper urban waste disposal have saturated and exceeded the capacity of soil to retain contaminants.

To emphasize the importance of soil as an important component of the natural system, as a vital contributor to human commonwealth due to its contribution to food, water and energy security, and as a mitigator of biodiversity loss and climate change, along with the significance of soil conservation, the 68th UN General Assembly adopted a resolution (A/RES/68/232) in December 2013 to officially declare December fifth as the World Soil Day (WSD). After the recognition of the WSD, the International Year of Soils, 2015 (IYS 2015) was also declared by the 68th session of the United Nations General Assembly. Further, in 2018, "Be the Solution to Soil Pollution" global campaign was adopted to create awareness regarding the importance of maintaining healthy soil ecosystems and encouraging societies to combat soil pollution and improve soil health. In the same year as the IYS 2015, the Sustainable Development Goals (SDGs), also known as the Global Goals, were approved by the United Nations as a universal call to action to end poverty, protect the planet, and ensure that by 2030 all people enjoy peace and prosperity. Soil is the at the core of sustainable development; improving soil quality is an integral step towards achieving the SDGs, specifically the Goals on zero hunger (SDG 2), climate action (SDG 13), and life on land (SDG 15).

This chapter describes the types and sources of soil contaminants, global status of soil contamination and soil remediation technologies; moreover, the soil contamination situation in Japan after the Fukushima Daiichi nuclear power plant (FDNPP) accident, and the decontamination practices implemented by the country to deal with the soils contaminated with radioactive <sup>137</sup>Cs are described. The results of our previous research on the methods to in-situ remediate <sup>137</sup>Cs contaminated forest soil using industrial waste materials are also introduced. Finally, efforts and challenges faced in managing and controlling soil contamination are briefly presented.

# **13.2** Soil Contaminants and Global Status of Soil Contamination

### 13.2.1 Soil Contaminants

Soil contaminants can be broadly divided into chemical (inorganic and organic), physical, biological and radioactive types. Chemical contamination is the most common. Inorganic chemical contaminants mainly include acids, alkalis, heavy metals (HMs) and salts, whereas organic chemical contaminants mainly include pesticides, petroleum, synthetic detergents, PAHs and PCBs. Recently, the effects of new soil contaminants such as microplastics, antibiotic resistance genes and e-wastes are also becoming an increasing concern. The sources of soil contamination mainly

Region	Main sources of soil contamination
Asia	East Asia: Industrial activities, mining, coal-burning, and solid waste disposal
	Southeast Asia: Increasing number of chemical industries and weak environmental regulations, along with agriculture and forestation- deforestation
	Central Asia: Mining, oil industries, metal sectors, and agricultural activities
Europe	Released radionuclides from Chernobyl accident, military activities, industrial and commercial activities, mining, waste disposal, agricul- tural activities
Latin America and the Caribbean	Agricultural practices, urban waste management, mining, quarrying and smelting activities, transport exhaust emissions, energy produc- tion, industrial and livestock activities
North America	Mining, industrial and agricultural activities, energy production, solid waste, military activities and nuclear weapons
Near east and North Africa	Dispersal of wind-blown contaminated dust, agricultural and industrial activities, intensive oil production, mining, inappropriate waste disposal
Sub-Saharan Africa	Mining and quarrying, fossil fuel extraction, agricultural and industrial activities, transport, energy sector, waste disposal and processing

Table 13.1 Main sources of soil contamination in different regions (FAO and UNEP 2021)

include mining, smelting, industrial activity, sewage irrigation, solid waste stacking, chemical fertilizers and pesticides. Table 13.1 lists some examples of the main sources of soil contamination in different regions of the world. As shown, most contaminants originate from human activities such as industrial processes and mining, poor waste management, unsustainable farming practices, accidents ranging from small chemical spills to nuclear power plant damage, as well as the armed conflicts (FAO and UNEP 2021).

### 13.2.2 Global Status of Soil Contamination

According to Kumar et al. (2019), more than ten million sites worldwide have been contaminated. For example, in the United States, more than 100,000 contaminated sites have been reported (Tu et al. 2020). In 2020, 2.5 million sites across Europe were potentially contaminated, according to the European Environment Agency. Further, out of the total number of radioactive contaminated areas (34 million hectares) after the Chernobyl accident in 1986, 20 million hectares of highly contaminated soils were in Europe, 70% of which were in Eastern European countries, including Belarus, Ukraine, and the Russian Federation (Boubriak et al. 2016). In Australia, approximately 80,000 sites were estimated to be contaminated.

In Asia, due to the diversity in the economic development, lifestyles, environmental policies, and recognition of sustainable development in different countries (IMF 2018), the issues of soil contamination are becoming complex and diverse. For example, in Japan, approximately 1.16 million acres of agricultural soil have been contaminated by HMs (Xu et al. 2016). And the soil contamination by radioactive substances, mainly due to the FDNPP accident occurred in 2011 also remains a significant problem. In 2015, the South Korean Ministry of Environment monitored soils at 1000 sites for eight trace elements and 13 other compounds, found that Hg and Zn were the most common contaminants (KOR-MOE 2017). In Thailand, the proportion of contaminated sites is increasing because of not only their domestic wastes but also the imported wastes (Diss 2019). There are approximately 52 major contaminated sites in Thailand, and the main sources of contamination are mining, petrochemical industries, illegal dumping, waste processing facilities, and imported e-wastes (FAO and UNEP 2021). Although the Bengal Delta is one of the most heavily arsenic (As) contaminated areas worldwide, soil conservation has not been prioritized in Bangladesh. In India, soils have been significantly contaminated with trace elements because of rapid economic growth through industrial, agricultural, and mining activities. In Sri Lanka, the use of agrochemicals, such as inorganic fertilizers and pesticides, is one of the main causes of soil contamination. Additionally, the lack of municipal solid waste collection facilities and poor waste management practices continue to cause severely soil contamination in Sri Lanka (Bandara et al. 2008). In China, the Ministry of Environmental Protection estimated that almost 16.1% of the total arable land and 19.4% of the cultivated land exceeded the permissible limit for HMs. Moreover, out of all the contaminated soils, 82.8% were contaminated with trace elements, such as As, Cd, Cu, Pb, and Hg; additionally, 13.7%, 2.8%, 1.8%, and 1.1% of the total cultivated land have been reported to be slightly, mildly, moderately, and heavily contaminated, respectively (Zhao et al. 2015). Notably, trace element contamination is generally more serious in southeast China than that in northwest China.

In summary, soil contamination has become one of the most challenging environmental problems worldwide due to its great harm to the ecological environment and public health. To conserve and maximize the use of limited and difficult-toregenerate soil resources, remediation of contaminated soil is very important and urgent.

# 13.3 Soil Remediation Technologies and Approaches toward Reducing the Risk of Cs Contaminated Soil in Japan

### 13.3.1 Soil Remediation Technologies

Soil remediation technologies tend to remove hazardous substances or reduce their amount accumulated in the soil, and to limit their spread so that the contaminated areas cease to pose a threat to the state of the environment and human health. Generally, soil remediation can be broadly divided into physical, chemical and biological methods (Liu et al. 2018).

#### 13.3.1.1 Physical Remediation Method

Physical remediation is using certain technologies to separate the contaminants from the soil and restore the available value of the soil. Physical remediation methods mainly include landfilling, soil surface capping or encapsulation, vitrification, thermochemical treatment, and electrokinetic remediation. Landfilling is one of the simplest soil remediation techniques through which the contaminated soil is removed from its original site and transported to a secure landfill for disposal. Soil surface capping or encapsulation method refers to covering the contaminated soil with a layer of waterproof material to form a properly designed physical barrier system. This technique is not truly a soil "remediation" method, as no efforts are made to remove or reduce contaminants in the soil. Nevertheless, the method can efficiently eliminate the risk of exposure to the contaminated soil. Vitrification is a thermal remediation technique that uses heat to transform contaminated soil into glass-like solids. Although this method requires high temperature and high pressure which means large energy consumption, it has high remediation efficiency. Thermochemical treatment is using high or low temperatures to heat the contaminated soil to volatilize the contaminants. Notably, thermal treatment technology can only remove volatile soil contaminants. Electrokinetic remediation is to reduce the scope of soil remediation and reduce the amount of engineering. Based on the principles of electrochemistry and electrophoresis, contaminants are moved to the electrode for centralized treatment or separation. It is only suitable for soil with high humidity. It has a certain danger, and the cost is high due to the electric energy consumption.

#### 13.3.1.2 Chemical Remediation Method

Chemical remediation is the process of changing the contaminant structure or reducing the mobility and toxicity of contaminants by means of chemical decomposition or fixation. It mainly includes soil washing, chemical oxidation/reduction, and solvent extraction.

Soil washing has been commonly used to remove organic contaminants from the soil by flushing surfactant solutions or passing extraction fluids through the contaminated soil. Extraction fluids in the form of chemical solutions are typically injected or infiltrated into the soil. This technique is applicable to homogenous coarse-textured soils with high permeability. Although this method is economic and user-friendly, installing solution collection wells or subsurface drains is challenging and expensive. Chemical oxidation-reduction technology purifies the soil by allowing the reaction between the chemical oxidants added to the soil and the contaminants, and chemically converting hazardous contaminants to nonhazardous or less toxic compounds. The most used oxidants are  $K_2MnO_4$ ,  $H_2O_2$  and ozone gases. Solvent



Fig. 13.1 Possible immobilization mechanisms of heavy metals (M) by biochar

extraction remediation is a technology that uses solvents to extract or remove harmful chemicals from contaminated soil. This technology is suitable for organic contaminated soil, and the effectiveness of extracting contaminants from soil depends on the intimate contact between the soil and the solvent mixture.

The above-mentioned conventional physicochemical remediation techniques are highly efficient and can be applied to severely contaminated soils but only at small scales (Dada et al. 2015). Moreover, they exhibit other disadvantages, such as high energy requirements and costs, destruction of soil structure and quality, and secondary contamination due to the released chemical substances during the remediation processes (Cristaldi et al. 2017). Therefore, some safe and effective remediation methods have been developed. For example, soil photocatalytic degradation, a novel deep-soil oxidation remediation technology, can be applied to remediate pesticide contaminated soils. Stabilization/ solidification is a well-developed and effective method for immobilizing potentially toxic elements in contaminated soils via adsorption, hydration, precipitation, or alkaline activation (Li et al. 2021; Shen et al. 2019). Besides, several studies have been conducted to reuse various low-cost or recycling materials, such as biochar, fly ash, blast-furnace slag, incinerated sewage sludge ash, zeolite, red mud and phosphogypsum, to immobilize soil contaminants (Shao et al. 2021; Tian and Sasaki 2019; Wang et al. 2021).

Biochar has been reported to adsorb or passivate soil contaminants and has been used primarily for in-situ remediation because of its large specific surface area, abundant active functional groups, good complexation, and adsorption property (Fig. 13.1). Additionally, biochar immobilizes HMs in soil through physicochemical adsorption with an aim of changing the specific chemical forms of HMs and inhibiting the bioavailability of HMs (Lu et al. 2014). Zheng et al. (2013) reported that adding biochar acquired from rice residues significantly decreased Cd, Zn and



Fig. 13.2 Phytoremediation strategies

Pb concentrations by 71%, 37% and 61% in the plant shoot, respectively. Liu et al. (2020) also revealed that the application of lychee biochar enhanced sunflower growth and immobilized HMs in the contaminated soil.

### 13.3.1.3 Biological Remediation Method

Bioremediation uses plants, microorganisms, or microbial/plant enzymes to detoxify, remove, or degrade contaminants to benign forms or low concentrations (Kumar et al. 2011). Compared to physicochemical methods, bioremediation is an environmentally friendly, safe and inexpensive technology for soil reclamation (Xiao et al. 2019). Among the available bioremediation technologies, phytoremediation is commonly used because it is easily operated, requires fewer human resources, and improves the aesthetic value of the environment. It uses vegetation to remove contaminants from the soil or decrease the availability of contaminants, thereby restoring the soil to a healthy level (Lajayer et al. 2019).

Hyperaccumulators tolerate high concentrations of contaminants and possess distinct capabilities to efficiently absorb contaminants from soil, translocate them from roots to shoots, detoxify and sequester contaminants in plant tissues. Phytoremediation of organic-contaminated soils is mainly manifested through the direct absorption of organic pollutants by plants, biodegradation promotion of organic pollutants by various secretions or enzymes released by plants, and enhancement of organic pollutants mineralization by plant rhizosphere. Phytoremediation is usually grouped into several sub classes including phytovolatilization, phytodegradation, phytoextraction, rhizofiltration and phytostabilization (Fig. 13.2). Phytovolatilization is using plants to take up contaminants from soil, convert these toxic elements into less toxic volatile form, and subsequently release

them into the atmosphere by plant transpiration process via the leaves or foliage system (Yan et al. 2020). Phytodegradation is the breakdown of organic compounds by metabolic processes of plants or enzymes. Phytoextraction, also known as phytoaccumulation, is the uptake of contaminants from soil by hyperaccumulator roots and their translocation and accumulation in the aboveground biomass, followed by seasonal harvesting until the concentrations of soil contaminants decrease to an acceptable level. Rhizofiltration is the reclamation of soil pore water, wastewater, surface and groundwater with low levels of contaminants by plant roots. Phytostabilization refers to the decreases in the mobility and bioavailability of contaminants in the soil, as well as the limitation of their leaching into groundwater and entering to the food chain, due to their stabilization from off-site transport with the help of certain vegetations.

Phytoremediation has been recently receiving increasing attention because of its low-cost, feasibility, and eco-friendly natures. However, phytoremediation also shows some challenges, for example, it is time-consuming, proper disposal of contaminated plants is an important concern. Moreover, the remediation efficiency depends on many factors including soil contamination level, soil characteristics, plant growth rate and biomass, root depth, as well as favorable meteorological conditions (Mahar et al. 2016; Cameselle and Gouveia 2019). Besides, phytoremediation is only applicable for medium to low degree of soil contamination, because plant growth can be restricted under a high degree of contamination. Contaminants that are tightly embedded in the soil are difficult to be eliminated through phytoremediation due to their low bioavailability. To increase the bioavailability of contaminants and improve the remediation efficiency, phytoremediation should be supplemented with other physical, chemical and biological methods; therefore, future research should focus on combined soil remediation technology.

# 13.3.2 Practical Approach to Reduce the Risk of Cs Contaminated Soil in Japan

### 13.3.2.1 Fukushima Daiichi Nuclear Power Plant Accident

In March 2011, the FDNPP accident triggered by a large earthquake and tsunami occurred and released 6000–12,000 PBq of <sup>33</sup>Xe, 100–500 PBq of <sup>131</sup>I, 15–20 PBq of <sup>137</sup>Cs, and 15–20 PBq of <sup>134</sup>Cs (Komatsu et al. 2021). This resulted in extensive radioactive contamination and caused detrimental effects to life and property in northeastern Japan, particularly in Fukushima Prefecture.

Among the radionuclides released during the FDNPP accident, radioactive Cs (<sup>134</sup>Cs and <sup>137</sup>Cs) was the most harmful element that affected the current and could affect the future health of the local population. <sup>137</sup>Cs contamination in the environment was more significant than that of <sup>134</sup>Cs, and posed as a long-term hazard. This is because its long half-life (approximately 30 years) could potentially contaminate the environment for long periods and threaten present and future generations of

living organisms. Additionally, its structural analog of an alkali element, such as K, could facilitate its immediate absorption by the plants from the soil, mainly through the K uptake system in the roots during K deficiency, and accumulate in plants following similar pathways, consequently, inhibiting plant growth, further damaging living organisms, and increasing the risks to cancer via the food chain.

To reduce the risk of exposure to radioactive materials, the Japanese government ordered immediate evacuation of the residents living around the FDNPP after the accident on March 12, 2011. Initially, only the areas within a 3-km radius of the FDNPP were evacuated, but as the effects of the accident became severe, areas within a radius of 20-30 km from the FDNPP were ordered to evacuate. Subsequently, on April 22, 2011, the government designated the 20-km radius around the FDNPP as a restricted area and prohibited entry for public, excluding those engaged in emergency response. Moreover, to minimize the effects of radioactive exposure, the contaminated area was divided into different areas based on the degree of contamination such as deliberate evacuation areas, specific areas recommended for evacuation, and evacuation prepared areas in case of emergency. Simultaneously, the government also set the criteria for radioactive Cs (sum of <sup>134</sup>Cs and <sup>137</sup>Cs) concentrations in food as 100 Bq kg<sup>-1</sup> to ensure that annual internal exposure to Cs by food ingestion is less than 1 mSv per year (Ministry of Health, Labor and Welfare 2018). In municipalities where food products exceeded the standard criteria, shipping restrictions were introduced.

#### 13.3.2.2 Soil Decontamination

To radically reduce the impact of radioactive Cs on human health and environment and to facilitate quick rehabilitation of the evacuated local population, the government decided to implement soil decontamination projects on an extensive scale. Accordingly, the Japanese Ministry of the Environment adopted the Act on Special Measures concerning the Handling of Environment Pollution by Radioactive Materials in August 2011. The law was established to ensure that the government will be responsible for promoting decontamination and will continually provide the technical information necessary for decontamination, including effective decontamination methods, funds, and considerations, through model projects in each region, especially in areas with particularly high doses of radioactive Cs.

Additionally, the Act on Special Measures for Decontamination was also enacted in November 2011. To supplement these acts, decontamination guidelines were released by the Japanese Ministry of Environment in December 2011 and updated in 2013 by incorporating knowledge and new technologies. The guidelines consisted mainly of three parts: Guidelines for Methods for Investigating and Measuring the Status of Environmental Pollution in Intensive Contamination Survey Areas, Guidelines Pertaining to Decontamination and Other Measures, and Guidelines Pertaining to the Collection and Transfer of the Removed Soil. They outlined the methods necessary for surveying and quantifying the contamination levels, preparing the contaminated areas for remediation, and treating the removed contaminated soil.



Fig. 13.3 Soil decontamination areas (cited from HP of the Ministry of Environment (Japan), https://www.env.go.jp/jishin/rmp/conf/law-joky006/lj06\_mat02.pdf)

Accordingly, under the direct supervision of the government, a decontamination road map (the special decontamination zone decontamination policy) was published in January 2012.

Soil decontamination began in January 2012, mainly in agricultural and residential areas. Decontamination activities were focused on the special decontamination area (SDA), including 11 districts, which were evacuated after the accident, and the intensive contamination survey area (ICSA), including 97 districts affected by lower, but significant radioactivity levels (not evacuated in 2011) (Fig. 13.3).

Decontamination was conducted in accordance with the circumstances of respective areas. In the cultivated areas within the SDA, the soil surface layer was removed till a depth of 5 cm and replaced with "new soil" comprising locally available crushed granite. In areas farther from the plant, materials that could fix or substitute radioactive Cs (zeolite, K fertilizers, etc.) were applied to the soil. In residential areas, ditch cleaning, roof cleaning, and gutter cleaning were conducted, and vegetable gardens were treated as cultivated areas. Moreover, forested areas located within a radius of 20 m of the houses were treated by cutting branches and collecting



**Fig. 13.4** Soil decontamination effects in Fukushima Prefecture (Air dose rate was measured at a height of 1 m from the ground surface. The chart shows the average value of air dose rate in residential areas, agricultural land, forests and roads. Residential areas included schools, parks, graveyards and large-size facilities. Agricultural land included fruit farms. Forests also included slopes, plains and grasslands. Data were acquired from HP of the Ministry of Environment (Japan), http://josen.env.go.jp/area/index.html)

the corresponding litter. However, for difficult-to-return zones (within the SDA area) in the immediate vicinity of the FDNPP, decontamination work was not implemented because these areas were difficult to access.

#### 13.3.2.3 Decontamination Effects and Limitations

According to the Japanese Ministry of the Environment, the entire area covering more than 9000  $\text{km}^2$  was decontaminated. As shown in Fig. 13.4, this decontamination work in the residential areas reduced the radiation dose by 60%, and a subsequent investigation showed that it had further decreased by 76%. In other land types, such as agricultural lands, forests, and roads, evident decontamination effects were also reported.

The decontamination strategy after the FDNPP accident provides valuable insights into the effective removal of Cs from the environment; however, certain limitations exist. For example, the method used to clean the contaminated lands reduced Cs concentrations by 72–76% in treated agricultural and residential areas, but the removal cost of the uppermost part of the soil in cultivated land was approximately 3000 billion Japanese yen. The costs could increase further if the forested areas are decontaminated. In addition, this method generates a significant amount of waste that is difficult to treat, transport, and store before its shipment to final disposal sites. Moreover, according to the reports by Mainichi Japan on May 27, 2021, a survey by the Japanese Board of Audit showed that the radiation doses in almost 12,900 locations subject to decontamination near the FDNPP or around 2%

of the sites (561,232 locations) cleaned after the nuclear disaster, did not fall below the pre-decontamination levels.

# 13.3.3 Experimental Approach to Reduce the Risk of Cs Contaminated Forest Soil

Although the decontamination in agricultural and residential lands has resulted in relatively satisfactory results, decontamination in large contaminated forests is difficult, due to increased costs and labor, destructive impacts on forest ecosystems, and additional expenses required to dispose large amounts of waste soil.

To alleviate the adverse effects of <sup>137</sup>Cs contamination, the Japanese Ministry of Agriculture, Forestry, and Fisheries utilized different methods, such as removing topsoil and fallen leaves, cutting trees, and covering the forest floor with wood chips in the experimental sites in Kawauchi village, Hirono town, and the litate village of Fukushima Prefecture, after the FDNPP accident from April 2012 to June 2013. However, despite a reduction in the dose by 12–14%, the release and diffusion of <sup>137</sup>Cs from the forest ecosystems, especially for those located in sloped areas were confirmed.

To reduce the pollution risk to the surrounding water and vegetation, the use of immobilization additives is considered as a promising alternative treatment to inhibit the <sup>137</sup>Cs from being washed off and transferred from contaminated soil to vegetation. Several adsorbent materials, including wood charcoal, bamboo charcoal, and zeolite, have been used to fix <sup>137</sup>Cs in soil and thus, inhibit its transfer from the soil to the surrounding environments (Aung et al. 2015; Burger and Lichtscheidl 2018). However, few studies have focused on the inhibitory effect of <sup>137</sup>Cs transfer from soil using biochar and other industrial waste-based recycling material adsorbents; correspondingly, the associated knowledge is also insufficient.

To select effective additives for <sup>137</sup>Cs immobilization, provide valuable references for the sorption and transfer behavior of <sup>137</sup>Cs in water, vegetation, and soil environments, and address <sup>137</sup>Cs contamination problem in forested soil, we collected soil samples from the topsoil of evergreen coniferous forest (0–15 cm depth), which is most widely distributed soil in Japan, from the vicinity of Ijira Lake in Gifu Prefecture. <sup>133</sup>Cs stable isotope was used as the surrogate for <sup>137</sup>Cs because both show similar physicochemical properties. After evaluating the fixation capability of Cs on different recycling materials based on sorption experiments, candidate recycling materials as Cs-fixing additives were selected. Subsequently, through pot cultivation experiments, the inhibitory effects of the selected recycling materials on Cs transfer from contaminated forest soil to vegetation and water penetrated from the potting soil were investigated and are described comprehensively in the subsequent sections.



Fig. 13.5 SEM micrographs of soil and the five recycling materials. *BC* coconut shell biochar, *RHB* rice husk biochar, *ISSA* incinerated sewage sludge ash, *CS* carbonized sludge

### 13.3.3.1 Fixation Capability Comparison of Recycling Materials for Cs Immobilization in Contaminated Forest Soil

Five recycling materials were used for comparison: coconut shell biochar (BC), rice husk biochar (RHB), blast furnace slag (hereafter referred to as slag), incinerated sewage sludge ash (ISSA), and carbonized sludge (CS). BC and RHB were obtained from the TAKII Seed Corporation and KANSAI Corporation, Japan, respectively, and the slag was obtained from the NISSHIN KOGYO Corporation, Japan. ISSA was acquired after P was recovered from the North Wastewater Treatment Plant of Gifu City in central Japan Gifu Prefecture. CS was a product of carbonization using mixed sludge from several small biological wastewater treatment facilities and was obtained from the Japan Environmental Management Center Co. Ltd., Kaizu City, Gifu Prefecture, Japan. The scanning electron microscopy (SEM) images of the recycling materials and the soil used are shown in Fig. 13.5.

Bottle-point batch sorption experiments were conducted to generate isotherm data for Cs after conducting sorption on each recycling material, soil, and recycling material-soil mixtures having different mixing ratios (7:3, 5:5, and 3:7 by weight). The initial Cs concentration varied at 0, 10, 50, 100, 500, and 1000  $\mu$ g L<sup>-1</sup>. The sorbent added to all bottles was fixed to an identical amount of 0.1 g in the dry state. After adding 30 mL of the Cs solution diluted to the designated concentration in each bottle, all bottles were shaken at 20 °C for 48 h to ensure sorption equilibration. Subsequently, the mixed solution in each bottle was centrifuged at 3500 rpm for 5 min, and the supernatant was filtered through a 0.2-µm membrane filter. The filtrate was then subjected to Cs quantification using inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7500 Series, Agilent, USA). The distribution of Cs adsorbed on the sorbent was analyzed using SEM/ energy dispersive X-ray analysis (EDX) (Hitachi SU-3500). In addition, the basic physicochemical properties of the recycling materials, soil, and their mixtures under different mixing ratios were also pre-analyzed.

The acquired isotherm data were described by the Freundlich isotherm model. The Freundlich K and Cs distribution after sorption on the recycling materials analyzed by SEM/EDX indicated that CS followed by BC and ISSA had larger fixation capability for Cs than the sample soil, suggesting that these materials can serve as potential additives for Cs immobilization in contaminated forest soils (Shao et al. 2018). In addition, parametric positive correlations were observed between the fixation capability of Cs and the property-related indexes of the recycling materials (cation exchange capacity, CEC; organic matter content; and K mass concentration).

### 13.3.2 Examination of Inhibition Effects of the Selected Recycling Materials on the Cs Transfer to Vegetation and Water

To examine the inhibitory effects of the BC, ISSA, and CS, on Cs transfer from contaminated forest soils to vegetation and penetrated water, a pot experiment was conducted in a greenhouse at the Gifu University.

The potting soil in each pot was obtained by separately adding BC, ISSA, and CS at three different concentrations (0, 5%, and 10%, w/w) to the forested soil, which was pre-adjusted to three different <sup>133</sup>Cs contamination levels (0, 25, and 50 mg kg<sup>-1</sup>). Moreover, Napier grass (*Pennisetum purpureum*) was cultivated because it can accumulate high Cs concentrations (Kang et al. 2012).

After 30 d, all grasses were harvested and washed with tap water to remove all soil particles from the surface. This was followed by washing with deionized water. Subsequently, the grasses were separated into different parts (leaf blade, leaf sheath, and root), and freeze-dried under vacuum freeze-drying conditions for 24 h to obtain a constant dry weight. The dried grass samples were crushed using a crusher and digested with nitric acid in an autoclave (Navarrete-López et al. 2012). The Cs and K concentrations of the digested samples were measured using ICP-MS (Agilent



Fig. 13.7 Amount of Cs released from soil (per kg) to the penetrated water. Different letters (a-d) indicate significant differences (p < 0.01) between different additive additions (reproduced from Shao et al. 2019). *BC* coconut shell biochar, *ISSA* incinerated sewage sludge ash, *CS* carbonized sludge



7700x). The distributions of Cs and other elements in the grasses and potting soils were analyzed using SEM/EDX. The penetrated water was collected during the final watering stage, and the Cs concentration in the water was also measured using ICP-MS.

As shown in Fig. 13.6, the value of the soil-to-plant transfer factor, which is an important parameter used in transfer models for predicting the contaminant concentration in plants and for estimating the impacts of internal dose on humans (Ban-Nai et al. 1999), significantly decreased after BC, ISSA, or CS applications, indicating that the Cs transfer from soil to grass was restrained. Thus, it can be concluded that all sorbent materials used in the study could inhibit Cs transfer to the food chain through plants, but the best performance was achieved by BC compared to ISSA and CS.

The Cs amount released from per unit of soil to the penetrated water is shown in Fig. 13.7. Compared to the control group (without additives), all three additives significantly reduced the Cs amount released from the soil to the penetrated water at different Cs contamination levels. At the 25 mg kg<sup>-1</sup> Cs contamination level, adding BC, ISSA, and CS reduced the amount of released Cs by 78.4–84.4%, 76.7–78.0%, and 51.9–69.6%, respectively. Moreover, at the 50 mg kg<sup>-1</sup> Cs contamination level, the released Cs was reduced by 91.9–92.7%, 84.4–88.2%, and 73.7–79.9% for BC,



Fig. 13.8 Proposed interactions among Cs, soil, biochar, and plants (reproduced from Shao et al. 2020)

ISSA, and CS additions, respectively, and the amount of released Cs showed significant differences among the three additives and followed the order: BC < ISSA < CS.

The possible mechanisms of Cs immobilization by biochar are shown in Fig. 13.8. The bioavailability of Cs in plants is influenced by several processes, including the transfer of Cs from soil particles to water, uptake of Cs by plant roots, and translocation of Cs within plants (Yoshida et al. 2004). Biochar can enhance Cs immobilization in soil by increasing the specific adsorption of Cs and inhibiting plant uptake due to competition with K. Cs was adsorbed onto biochar mainly through cation exchange, surface interaction, and complexation with functional groups (such as -COOH and -OH) on the biochar surface (Uchimiya et al. 2011). In addition, soil properties could influence both the Cs availability in the soil and the phase of Cs absorbed by plant roots (Fujimura et al. 2015). For instance, biochar addition could increase the negative charges of soil particles, thereby increasing the adsorption affinity for cations (Jiang et al. 2012). Biochar properties, such as large surface area, high porosity, pH, and CEC, promote the fixation of mobile Cs onto biochar and facilitate the conversion of Cs into more stable forms (Beesley et al. 2011; Xie et al. 2015). Conversely, the increased K concentration in biocharamended soil suppresses Cs uptake by plant roots and Cs transfer from roots to aboveground parts due to the competitive relationship between Cs and K (Shao et al. 2020).

## 13.4 Efforts and Challenges in Managing and Controlling Soil Contamination Worldwide

Most countries have formulated management and control strategies for soil contamination. For example, in the United States, the Comprehensive Environmental Response, Compensation, and Liability Act (commonly known as the Superfund Law) was enacted as the first law on soil pollution in 1980, and it has achieved significant results to address soil contamination. According to this law, approximately 800 substances have been designated as harmful substances. Along with this law, the United States implemented other laws, such as the Resource Conservation and Recovery Act, Safe Drinking Water Act, and Water Quality Purification Act, with an aim to prevent soil and water contamination.

Germany has a long history of industrial development. Apart from soil contamination caused by industrial activities, military-based soil contamination due to defense projects associated with the two World Wars has also become a social problem. As a measure against soil contamination, Germany enforced the Federal Soil Protection Law in March 1999 and the Soil Conservation Contamination Site Ordinance in July of the same year. These laws have established permissible limits for different contaminants according to land use; additionally, according to these laws, purification targets can be determined based on the agreement between regulators and landowners for specific applications in each category. Moreover, the enforcement of these laws unifies disparate soil contamination countermeasures in different regions, thereby leading to increased rational and effective soil conservation efforts.

After Europe and the United States, soil contamination in China has become increasingly serious in recent years. To conserve soil resources and safeguard human health, in 2016, the Chinese central government allocated funds for soil contamination prevention and control and issued 28.5 billion RMB to support source control, risk control, restoration, and supervision of soil contamination; and promote the establishment of funds and multi-channel support for soil contamination prevention and control a the provincial level. According to the global Environmental Research Network, the market capacity of soil remediation industry in China during the 13th Five-Year Plan was 72.45 billion RMB, of which 15 billion RMB, 48.8 billion RMB, and 8.65 billion RMB were allocated for cultivated land restoration, site restoration, and coal mine restoration, respectively. Along with this financial capital, soil remediation began attracting social capital.

However, most countries worldwide still face various challenges regarding soil conservation. For example, data on non-point source emissions at the national level are scarce, especially in the developing countries. In some countries, the primary

cause of soil contamination is lack of awareness among the general population. For example, a survey revealed that more than 68% respondents in Bangladesh were unaware of the hazards of e-waste, while 12% believed that e-waste did not have negative effects (Islam et al. 2016). In addition, lack of information on the extent of contamination in the food chain reduces the understanding of the risks to health and food security. In addition, the soil management capacity varies widely among different countries. The foundation for soil environmental supervision and management is weak, and the technical support is insufficient in many developing countries, all of which have made the prevention and control of soil contamination difficult.

Soil remediation is characterized by a long cycle, high cost and slow effects. Thus, the lack of national-level infrastructure, standard or advanced technology, and systematic treatment make it difficult to ensure effective soil restoration. Moreover, financial resources for soil remediation and environmentally sound waste management are insufficient. For example, although China adopted its first soil contamination law as its latest effort to address environmental problems, strategies to finance the remediation of the existing damages remain unclear. During the 12th Five-Year Plan (2011–2015), only \$4.546 billion was allocated for soil remediation, and mainly in urban areas. Further, the estimated cost of remedial work was approximately \$1.3 trillion for 2016–2020. The Chinese government estimated that this amount possibly includes only a fraction of the total cost.

Restoring contaminated soil is a long-term and arduous task for all countries in the world. Soil remediation strategies are subjected to the concentrations and amounts of the contaminants, geographical conditions as well as financial resources. In addition, for successful soil remediation the integrated implementation of different physical, chemical and biological technologies will be the key.

### 13.5 Conclusion

Soil is an important component of the ecosystem, as most of the food we consume originates from it. Therefore, maintaining soil purity, fertility and its toxin-free characteristic is important. However, with the increase in the impacts of human activities on the environment, soil has become increasingly contaminated. Contaminants such as heavy metals and radioactive <sup>137</sup>Cs which cannot be easily degraded and will persist in the environment for a long duration. Consequently, this may cause serious problems for the next generations.

This chapter introduces the types and sources of soil contaminants, global status of soil contamination, soil remediation technologies, soil contamination situation in Japan after the FDNPP accident, and effective soil decontamination practices to reduce the impact of radioactive <sup>137</sup>Cs on human health and the environment. Moreover, a case study to address <sup>137</sup>Cs contaminated forest soil using different industrial waste materials based on sorption and pot experiments was introduced. We found that the application of biochar significantly decreased the accumulated Cs concentration in vegetation and inhibited Cs transfer from soil to vegetation and

water, thus, showing high performance in reducing the bioavailability of Cs in soil. This finding serves as a valuable reference for addressing the Cs contamination problem in forest areas because biochar as a soil amendment is inexpensive and can improve soil fertility, soil water holding capacity, and aeration status. However, the safety and biological toxicity of biochar needs to be further studied through long-term field experiments before large-scale application in different soil types.

Efforts to address soil contamination have been undertaken for several years. Improving the soil environment is a long-term and arduous task for all countries worldwide. With the improvements in public awareness regarding environmental protection and governmental support, more economical and efficient soil remediation technologies have been recently developed and applied. Furthermore, with the increasing awareness on green and sustainable remediation in many countries, soil contamination will be completely controlled and the sustainable development of soil resources will be achieved by using the most optimum remediation technology.

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