Study on Radioactive Contaminated Soil Remediation Technologies and Selection Principles

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Abstract A large number of radioactive materials entered the soil environment by Fukushima nuclear accident, and large area of contaminated soil with radioactive materials became the thorniest problem of Japanese government after the accident. On the basis of the mechanism of radionuclide migration in the soil, such as physical migration, chemical migration, biological migration, and the composite of different migration processes, the series of radionuclide-contaminated soil repair methods is discussed. There are move and replace with out-soil methods aiming at physical migration, there are ion exchange method and oxidation-reduction method aiming at chemical migration, there are microbial remediation and phytoremediation aiming at biological migration. The advantages and disadvantages of different methods and applicable range are illustrated. The final remediation target is the first priority when selecting remediation methods. Then, the influence factors on remediation effect should be considered, including the principles of radiation protection, radioactive pollution levels, soil characteristics, hydrological and meteorological conditions, the radionuclide migration way influence. Finally, there will be a comprehensive comparison from method feasibility, economic affordability, and environmental and legal compatibility. The feasibility of remediation methods includes appropriate repair methods, acceptable repair time, good repair effect, etc. Economic affordability means repair costs can be bearable and as less as better. Environmental and legal compatibility refers that after the restoration the residual risk of nuclides in environment is acceptable and the long-term effect is negligible and meets the requirements of relevant laws and regulations standard.

Keywords Radioactive contamination · Soil remediation · Selection principles · Remediation technology · Factor

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1 Introduction

The potential leakage of radioactive material pollution in nuclear facilities is one of the major environmental problems. The Fukushima nuclear accident resulted in a large number of radioactive substances into the soil environment through the air-borne route, especially ^{137}Cs [1]. It would cause sustained long-term harm to the ecological environment and human health when long half-life nuclides enter into the soil. How to deal with the large area soil by radioactive pollution became the thorniest problem of Japanese government after the accident.

As the number of operational nuclear power plants in China increases continually, it is necessary to precede the scientific evaluation in radioactive contamination of the soil in order to determine the pollution scope and carry out pollution regulation after the nuclear accident. For a long time, a lot of research has been carried on refractory organics, heavy metals, and other aspects of non-radioactive soil bioremediation, while little has been done on soil polluted by radiation systematically. Meanwhile, with the decommissioning of nuclear facilities, the soil will be affected inevitably in a certain extent around those nuclear facilities. As we can see, each environmental remediation technology has its own scope. There will be a huge difference in the renovation cost when using various environmental remediation technologies under the same contaminated sites.

In this paper, based on doing a series of research on soil remediation techniques, having analyzed the advantages and disadvantages of different technologies as well as their scope, we present selective principles for radioactive contaminated site remediation techniques in order to provide technical supports for remediation on the specific contaminated site.

2 Radioactive Contaminated Soil Remediation Technology

There are amounts of radioactive soil remediation technologies, such as shoveling soil of decontamination method, which removed topsoil; deep plowing which means covering with new uncontaminated soil above those polluted soil, thereby rebuilding ecological system; agricultural chemical method, with potassium salt to prevent absorption of 137Cs. Besides, there are suspended soil removal method, phytoremediation method, membrane stripping method, electrochemical method, magnetic method, precipitation, soil washing, ion exchange method, chelating extraction method, flocculation technology, and reverse osmosis ultrafiltration and so on. However, all these methods have its shortcomings. For instance, some of the technology is not mature; some usually requires a huge cost; some is not suitable for the radionuclide contamination process of large areas with low dose, which may easily destroy the structure and texture of contaminated soil causing secondary pollution; on the contrary, some is much lower cost, non-polluting, but inefficient decontamination. The selection process must be based on the analysis of the specific conditions of the soil and the use of selective, or adopt several methods to coexist so as to strive to achieve the lowest cost and expense for maximum removal efficiency. Some decontamination methods are introduced after several radioactive soil contaminations.

Shovel contaminated soil. The soil contaminated by nuclear material (typically topsoil) should be shoveled away, transported to the specialized nuclear disposal sites for processing and disposal, which could prevent the further spread of radioactive elements fundamentally. However, the method is labor-intensive, operators will be easily exposure to radiation, and a lot of shoveling will increase processing and disposal costs. In addition, the topsoil also contains large amounts of organic matters for crop production. Shoveling all soil would further exacerbate the land crisis.

Deep plowing new soil or covered with new soil. New soil is relative to the case of the contaminated local terms. For a relatively high level of radioactive contamination of the region, the replace with out-soil is the use of deep plow method that contaminated topsoil turned to deep soil, while the lower layer were not nuclear-contaminated soil turned to the surface, so as to dilute and reduce surface contamination levels. Covering out-soil is shipped uncontaminated soil directly from the outside to contaminated soil, which, to some extent, can prevent nuclear contaminants from entering the food chain so as to form the internal radiation to human body. The advantage of this approach is to avoid pollution of soil contaminants into the food chain through contaminated soil over a layer of clean soil cover. But we must ensure that the amount of clean soil has a certain thickness, so that plant roots would not extend to the contaminated soil. Disadvantage of this method is that the soil contamination could not be removed, nor can completely rule out the potential hazards of soil contaminants.

Suspended soil removed method. This is mainly for situations, for example, paddy soil has a large amount of water-flooded soil. A thin layer of soil at the top of the water is about to stir into a paste, pumping suspension soil, sediment and then separated from the water, only to precipitate after processing. The advantage of this method is that the amount of secondary waste generated is small; the inadequacies of this approach are also drawn plenty of organic matter in the soil.

Peelable membrane. The cleaning liquid polymer compound with a variety of functional groups would quickly be sprayed by land spraying machinery the film-forming decontamination materials are coated on the contaminants, and quickly fixed nuclear contamination, control transferring and diffusion of pollution in the shortest time. After solidification of the film-forming material, surface contaminants rapid build-molding in the role of nuclear peel adhesive, and then recovered clear to the use of mechanical or manual for forming the membrane, so as to achieve on-site restoration of the natural environment and battlefield environmental purification. The surface decontamination coefficient of this method could reach 100% and have relatively low economic costs. However, there has no effect on the removal of soil penetrate inside the nuclear pollutants.

Agricultural chemical method. Agricultural chemical method refers to the use of general operational measures, so as to minimize the content of radioactive substances in soil and to inhibit the migration of radioactive materials to crops. Due to the similar chemical properties of cesium and potassium, we choose to add potassium and nitrogen into the soil under normal circumstances in general, with a high concentration of potassium and ammonium salts to prevent cesium from transferring to crops, namely food chain. The advantage of this method is that operation is simple, drawing easily, and low cost. To a certain extent, it could reduce the content of radioactive substances in plants. But the disadvantage is that radioactive substances could not be removed from the soil fundamentally, which may leave future troubles.

Phytoremediation. Phytoremediation is a method aiming at accelerating degradation of pollutants in soil through those properties, such as the use of plant accumulation of certain pollutants, plant metabolic processes of certain pollutants' transformation and mineralization, increased microbial activity of plant rhizosphere. Phytoremediation is a kind of environmental control technology, using process of plant roots absorbing water and nutrients to assimilate, transforming the contaminated carrier pollutants in order to achieve clear, repair or governance purposes. Usually we harvest plants grown in contaminated soil, and treat those plants by incineration, landfill, or any other suitable methods. Compared with other repaired techniques, phytoremediation is low cost and has small impact on the environment. It is beneficial for improving the ecological environment. Compared with other traditional chemical remediation, physics and engineering technology, phytoremediation technology is easy to operate with low investment and maintenance costs. What is more, it takes the use of solar energy, so that will not result in secondary pollution. But the disadvantage is that radioactive substances could not be removed from the soil fundamentally, which leaves future troubles.

3 Restoration Technology Selection Principles

For specific contaminated sites, we should not only consider their advantages, respectively, other factors should also be considered such as remediation goal, the final residue levels, and radioactive contamination levels, principles of radiation protection, soil type, hydrological and meteorological conditions. Finally, we take consideration of repair technology from comprehensive standards, including repair technologies feasibility, repair costs affordability, remediation goal acceptability.

3.1 Remediation Goal

Target of soil remediation is an important part of the implementation of environmental management, which is also a key indicator for technical restoration. The remaining level of radioactivity in soil is the most immediate goal of soil remediation. In order to develop the remaining level of radioactivity, we must first clear the source of the radioactive contaminated sites and radionuclide type. Next, we consider the radiation protection principles when repairing radioactive contaminated soil and gain the goal of soil remediation via dose assessment.

The source of radionuclide could be divided into the following aspects: (a) settlement of radioactive substances caused by atmospheric nuclear tests. According to statistics, from July 16, 1945, when the USA conducted its first nuclear test till to the end of 1989, all countries conducted totally 1800 nuclear tests worldwide. Radioactive dusts would gradually settle after the atmospheric nuclear tests, causing pollution of soil, rivers, and oceans. (b) Nuclear accidents. "Ural incident," occurred in September 1957, led to polluted area up to 5000–23,000 km^2; in the "Chernobyl accident," the soil within a radius range of 30 km was polluted by ^{90}Sr, ^{137}Cs, $^{239+240}Pu$, ^{241}Am, and other radionuclides [2]. In Ukraine, 260,000 km^2 area had been contaminated by ^{137}Cs [3]. Fukushima nuclear power plant accident resulted in surrounding soil was contaminated by ^{137}Cs, and the cumulative concentration reached 100,000 and 10,000 Bq/m^3 [4, 5]. (c) Wide use of fertilizer containing radionuclides. The use of phosphate fertilizer containing radionuclides ^{232}Th, ^{238}U, ^{226}Ra, and potash containing ^{40}K is very extensive. The use of these fertilizers containing radionuclides contamination can cause soil pollution. (d) Mineral exploitation. Development and utilization of nuclear energy requires a lot of uranium, and uranium mining and processing would pollute the surrounding soil. According to global estimates, slag volume that caused by uranium mining and uranium tailing reached 9.38×10^8 t [6].

The most immediate goal for soil remediation is remaining level of radioactivity in soil. For soil in the intervention system after accident, the screening scene and selection process should be established according to the environmental conditions surrounded by pollution, social development. Then remaining level of radioactivity in soil will be determined according to the scene. Nuclides screening method is important key to establish a dose contribution, which determines the radionuclide exposure pathways according to the established scene. Finally, according to the evaluation model to determine the relevant calculation parameters, the remaining level of radioactivity is deduced. Only when the overall interests of the radioactive contaminated site remediation more than the cost that paid for, we believe the restoration is justified.

3.2 Factors of Restoration Effect

There are various differences in radioactive contaminated soil on radionuclide types and proportions from different sources. At the same time, degree of soil pollution is also differently affected by weather conditions under such accident. Radionuclide distribution of contaminated sites, soil types, hydrological and meteorological conditions, radionuclide morphology and nature, and the exposure pathways in polluted sites would affect repair effect.

(a) The distribution of radionuclides. Since the source of radionuclides, the difference of nuclides released meteorological conditions, resulting in different types of radioactive nuclides in contaminated sites, scope and concentration of radionuclide distribution in the same contaminated sites. This difference reflected in the distribution of the horizontal and vertical directions of radionuclide: ① in the horizontal direction, location and extent of the high value area of soil surface contaminants; ② location and extent of different pollution concentrations in different soil depths; ③ with longitudinal migration of contaminants from the sources of change; ④ risk analysis, the probability of exceeding a specified threshold concentration. Different repairing technique has a significant difference in terms of the type and concentration of radionuclides applicability.

(b) Soil type. Different soil types have a different impact on the adsorption and migration of radionuclides. Colloid composition is different from ordinary farmland, paddy fields, woodland, grass land soil types, resulting in large differences in physical and chemical properties of soil, pH, moisture content, redox potential, and root microorganisms. Studies have shown that the composition of different soil types in repair contaminated soil is one of the main theoretical bases for phytoremediation, because being as the center part, the plant roots gathered a lot of living matters and secretions, such as bacteria, microbes, earthworms, nematodes, which consist of a very unique "ecological restoration unit." Therefore, remediation effect of different soil types may have significant differences.

(c) Hydrological and meteorological conditions. Hydrological and meteorological condition determines the dominant species and its habitat ecosystems, which is of great significance to the local phytoremediation and microbial remediation. Ebbs and others studied [7] on uranium accumulation capacity to absorb the 12 species, finding that there is a huge difference between different plants on ability to accumulate uranium. Bennett [8] evaluated the potential capability of the plant concentrate radionuclides in radioactive contaminated soil. It showed that vascular plants had higher ^{99}Tc (Tc) uptake, and 99Tc also had strong shoots transfer and accumulation trends nuclides form.

(d) The morphology and properties of radionuclides. The migration ability of dissolved cation is low in clay soil. Radionuclides of different oxidation valence state have different plant absorption capacity. Generally, smaller the particle size of radionuclides, more likely to be absorbed by plants.

(e) Exposure pathways. We need to consider the external exposure from radionuclides deposition to the ground and internal exposure from ingestion of crops. We should take into account the internal exposure inhalation of dust also. The following factors would ultimately affect the selection of repair target, environmental media parameters, dose conversion factors, crop consumption and living habits, and so on, thereby affecting the taken remediation.

3.3 Repair Technology Selection Principle

After the accident, the selection method of radioactive contaminated soil remediation program is based on the determination of soil remediation objectives, in terms of technical, economic, environmental, and social indicators. What is more, comparison studies of repairing technology also need to determine the appropriate remediation program method for restoration.

Select the repair strategy. We should confirm the overall goal of soil repair according to the soil pollution survey and the radioactivity level of soil residual. The appropriate repair strategy is determined through preliminary analysis of the repair mode, remediation technology type and application conditions, contaminated soil characteristics, hydrological and geological conditions, the level of technical and economic development.

Optimal scheme comparison of radioactive contaminated soil remediation technology. A reasonable combination of all kinds of feasible technology to achieve the overall objective as well as a potentially feasible selection of remediation programs will be provided through considering the overall objectives of the soil remediation, restoration strategy, pollution status, soil characteristics, hydrogeological conditions, repair results in screening and evaluation.

(a) Technical indicators of operability: reliability of remediation technology, time requirement of repair scheme, availability of necessary equipment and resources, removal efficiency of contaminant, etc.
(b) Economic indicators: start-up costs, operating costs, late stage fees, etc.
(c) Environmental indicators: residual risk, long-term effects, healthy effects, etc.
(d) Social indicators: compliance with current laws and regulations, relevant standards and norms; level of public acceptance.

Using the established 'comparison and selection' index system, detailed analysis of each potentially feasible remediation technology schemes can be done. For the final choice of remediation technology schemes, it can be got through various indicators comparison and comprehensive judgment, also through the expert score mode.

4 Conclusions

Radioactive contamination remediation is a long and arduous task, requiring a lot of manpower and material resources. Many contaminated technologies and measures on soil restoration practiced in Japanese Fukushima are worth for us to carry out further research. For instance, for small-scale soil contamination with a high concentration, we can take shovel soil of decontamination method; for low concentration of water in soil, deep plowing method with out-soil and suspended soil decontaminated method can be used. From the perspective of long-term management, we should pay attention to methods of low cost as well as non-pollution, such as phytoremediation method. In addition, after the accident the first should be done is to investigate and confirm the range and extent of contaminated soil, in order to take appropriate repair techniques. Furthermore, if shovel soil of decontamination method is taken, how to save radioactive soil and how to reduce capacity are both very urgent problems. Finally, soil contamination caused by weather and other factors should also be of concern during the operation.

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