Restoration Technology Research Progress of Mercury Polluted Farmland

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Abstract—Soil is an important part of the environment, which is the precious renewable resources. Soil heavy metal pollution is becoming more and more serious, which affects the normal function of soil, causes pollution of food chain, threats to human health and environmental quality. Mercury as one of the persistent toxic pollutants, is also one of the precedence-controlled pollutants, soil mercury pollution problem has aroused high attention of the world. In our country, mercury discharge of about 1.9×108 kg/a into environments, covering 3.2×104 ha contaminated farmland，mercury as a kind of toxic heavy metals is non-necessary for human, posing a great risk to human health. Considering the circumstances of mercury pollution and the remediation demands, six kinds of commonly applied techniques were developed in the following aspects of working principles, researching progress, studying cases, advantages or disadvantages, and application scopes, and so on, supplying the technology support for the remediation plan of the mercury contaminated farmlands.

Keywords-Mercury Pollution;Farmland Soil;Remediation Technology;harm;application

I. INTRODUCTION

Since the last century, accidents of mercury pollution occurred frequently, such as Japan's Minamata Bay, Kumamoto Minamata disease event in 1956, event of four American children poisoned by mercury-fed pork, methyl-mercury dressing food poisoning event in Iraq in 1972. All these events have brought human beings and ecological environment great disaster[1, 2].

Mercury is a potential neurotoxin heavy metal element[3], UNEP, WHO, FAO and other international organizations list mercury as one of the priority control pollutants due to its persistence in the environment and bioaccumulation characteristics. Mercury pollution in soil mainly derive from the exploitation of gold mine and copper zinc ore, chloric alkali industry, wet deposition (dissolved mercury，Hg^{2+}) and dry deposition(particulate mercury, Hg^{0}) of atmospheric mercury generated by burning coal[4-6]. In addition, sewage irrigation and soil improved by city sludge can also contribute to the increase of mercury and methyl-mercury concentrations in soil[7]. Once mercury enters the soil, 95% of it can be fixed by soil or adsorption[8], and it is difficult to be transformed or degraded[9], therefore, the perniciousness of mercury in soils is cryptic, hysteretic and long-term, besides, it can harm human health by crops absorption and beneficiation of food chain transfer[10, 11]. Thus, farmland soil pollution prevention has become the focus of many scholars both at home and abroad[4, 12-14].

According to the study, soil in many domestic and foreign areas have been polluted to varying degrees by mercury[5]. Every year in our country, the content of mercury invaded in the environment is about 1.9×10^8 kg, which leads to 3.2×10^4 ha of mercury-polluted cultivated land[15]. The mercury pollution status in Songhua River basin of Jilin Province and Guizhou Wanshan area is serious[16], human health survey suggests that mean content of mercury in Songhua River fishermen's hair in 1975 is 17.50 μg/g, in 1990 is 3.82 μg/g and in 2000 is 2.15 μg/g, though there is a downward trend, it is still higher than the 1mg/kg limit value enacted by the America Environmental Protection Bureau[17]. Mean content of mercury in residents' hair in Guizhou Wanshan mercury mining area is 837 μg/g[18], which far exceed the standard. The mercury content of soil in six sewage irrigation area in the eastern suburbs, western suburbs and northern suburbs of Xi'an with about 200 km^2 range is between 0.52 mg/kg and 0.90 mg/kg[19]. Soil mercury concentration of sewage irrigation area in Huaiyin has exceeded one times more than mercury concentration in soil clean irrigation area, also exceed the limit value of second ambient soil quality standards (GB15618-1995)[20].

There are mainly two ways to harness mercury pollution in soil. One is to change the mercury speciation in soil, transfer it from the active state to stable state, and reduce its bioavailability and migration activity[21], the other is to reduce Hg content in soil by some measures[22]. At present, there are several technologies frequently-used at home and abroad in remediation of soil contaminated with mercury, such as turning over the soil, soil method, heat treatment method, electric repair method, leaching method, stabilization and solidification method, bioremediation.

II. PHYSICAL REMEDIATION TECHNOLOGY

A. Soil aeration, soil replacement

Soil aeration is to disperse pollutants gathering in the surface soil into the deep. Soil replacement is to remove contaminated soil and cover uncontaminated soil, or to directly mix the contaminated soil and uncontaminated soil, with the aim of pollutants diluting. The two methods can be simply operated, but its workload is large, have a great disturbance on soil structure, and mercury in soil has not been fundamentally removed, so it is not suitable for the situation of heavy pollution or lack of land resources.

B. Thermal treatment

Thermal treatment uses heat or hot steam to make the volatile or semi volatile pollutants in the polluted soil into gas, centralized recovered or disposed[23]. Mercury in soil mainly exists in the forms of elemental mercury and compounds such as HgS, HgO and $HgCO_3$, when the temperature gets 600 - 800℃, compounds will breakdown and release elemental mercury vapor, which will recover by off-gas treatment device[24-25].Thermal treatment is widely used in the remediation of mercury contaminated soil and has the advantages of simple operation, high treatment efficiency, the main process is shown in Fig .1.

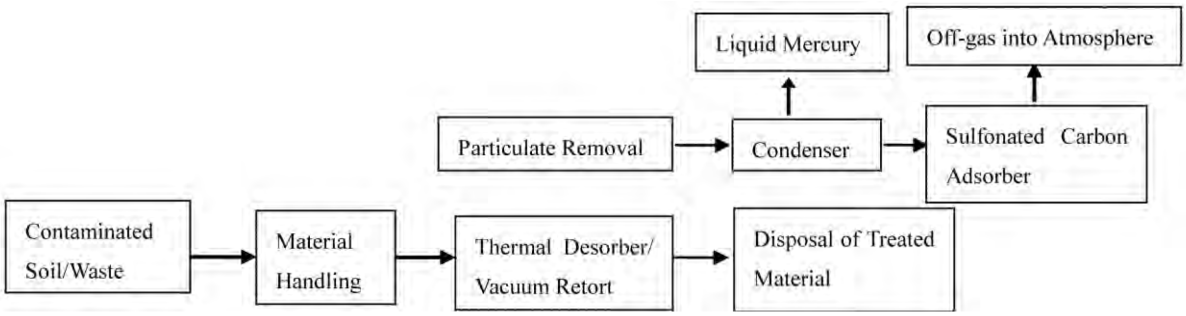

Figure 1. Model of a Thermal Desorption or Retort System (From References [24-25])

Because of the large energy consumption, the high investment and operation costs of thermal treatment, and the truth that more than 500℃ will cause the loss of soil nutrients, destroy the physicochemical properties of soil and soil structure, in recent years, low-temperature pyrolysis technology has been developed and applied. Zhang Qian[26] treated the typical mercury contaminated soil of Wanshan Guizhou in the low-temperature of 370℃. The mercury in soil can be reduced into 10 mg/kg, treatment rate of 95.73%, and residual mercury in treated soil is mainly in slag state mercury. Qiu Rong[8] researched the mercury contaminated farmland soil of Qingzhen Guizhou in low-temperature pyrolysis technology, showed that under 350 ℃, treatment rate could be more than 90%[27]. Under the temperature of 300℃, the mercury contaminated soil around a waste factory of chlor alkali production in Qinghai, with the residence time of 60 min, mercury concentration is reduced to 10 mg/kg, the treatment rate is nearly 90%[28]. Comuzzi disposed mercury contaminated sediment by co-induction using tetrabutylammonium chloride and thermal desorption, treatment rate increased from 24% to 60%[29].

C. Vitrification

Vitrification uses hot heat source such as plasma and electric current, to melt contaminated soil at 1600 - 2000℃, contaminants are pyrolysed or vaporized, cooled melting will form corpora vitreum. It wraps contaminants and reduces the migration of contaminants. Once in treatment with mercury contaminated soil by vitrification technology in the US state of Michigan, the initial concentration of mercury is about 40 g/kg , after the treatment, available mercury concentration is less than 0.23 g/L[30]. This technology can be operated via mobile devices in the field, but costs much.

III. CHEMICAL REMEDIATION TECHNOLOGY

A. Electrokinetic remediation

Electrokinetic remediation is the technology of inserting electrodes on both ends of the contaminated soil. Under the low voltage direct current field, the pollutants are transported into the electrode chamber, and finally collected through engineering methods, the main electrode reactions are as follows (E_0 is standard electrode potential)[31]:

Anodic reaction:

$$2H_2O - 4e^- \rightarrow O_2 \uparrow + 4H^+, \quad E_0 = -1.229\ V \qquad (1)$$

Cathodic reaction:

$$2H_2O + 2e^- \rightarrow H_2 \uparrow + 2OH^-, \quad E_0 = -0.828\ V \qquad (2)$$

This technology was first developed by the United States to purify contaminated soil in situ[32], then gradually developed to strengthen electrokinetic remediation technologies, including electrodialysis method, complexing method, acid-base neutralization, cation-selective membrane method, surfactant method, oxidation-reduction method, EK-conjunction with biological and Lasagna™ method. Zheng Shenshen adopted the method of cathode electric supplemented by cation exchange membrane and KI + I_2 solution oxidation complex method to remediate mercury contamination typical paddy soil in Guizhou and the Hg removal efficiency can reach 68.6%[33]. Reddy made the removal efficiency of mercury in the soil increase to 97% by adding 0.1 mol/L KI to kaolin[34]. Cox added I_2/KI lixiviant in cathode and made the insoluble Hg into HgI_4 complex anion, so that the mercury removal efficiency was as high as 99%[35]. Electrokinetic remediation technology is suitable for low permeability soil such as clay, silt soil. This method has high removal efficiency and has no effect on the soil fertility, but the repair cost is high and electrolysis will cause changes in soil pH, repair mechanisms and reaction conditions need further research as well.

B. *Soil leaching method*

Soil leaching method is mainly used to deal with pollutants physically or chemically adsorbed on soil particles. Eluent transfers the inorganic or organic pollutants from the soil solid phase to the liquid phase, then further treatment of the eluent contained pollutants is taken. Soil leaching removes pollutants in two ways: one is using eluent to dissolve contaminants in the liquid phase, adsorption phase and gas phase; another is using water to wash contaminants in soil pore or adsorbed on soil. The former is controlled by pollutant solubility and Henry's law constant, the latter depends on the pressure gradient, soil flushing water viscosity and the concentration of pollutants[36]. The key of Mercury polluted soil leaching technology is to select eluent that both can better absorb mercury, and does not destroy the physical and chemical properties of soil. Some studies found that iodide, EDTA, thiosulfate compounds have good leaching effect[37]. Lin Kai found that in severely mercury polluted soil, the mercury removal effect of eluent is NaOH solution > emulsifier > EDTA > DTPA > tartaric acid > phosphoric acid > glacial acetic acid > hydrochloric acid >nitric acid > citric acid > the agricultural irrigation water > $CaCl_2$ solution > precipitation water > deionized water, and the mercury removal rate in soil is from 6.5% to 37.5%[38]. Chen Zongying[39] found that $Na_2S_2O_3$ solution can efficiently remove the mercury in soil whose initial concentration is 108.76 mg/kg, removal rate is 65.32%. Ray[40] used H_2O_2, $Na_2S_2O_3$, Na_2S to joint repair mercury polluted soil, whose initial concentration is 2100 mg/kg, terminative concentration is 270 mg/kg, mercury removal rate is 87%.

Since the United States GE Wiring Deviees used KI solution as eluent to repair the mercury polluted places in 1992, large-scale heavy metal polluted soil leaching projects have been carrying out one after another. Kuhlman[41] summarized ectopic soil leaching process, whose details are shown in Fig .2.

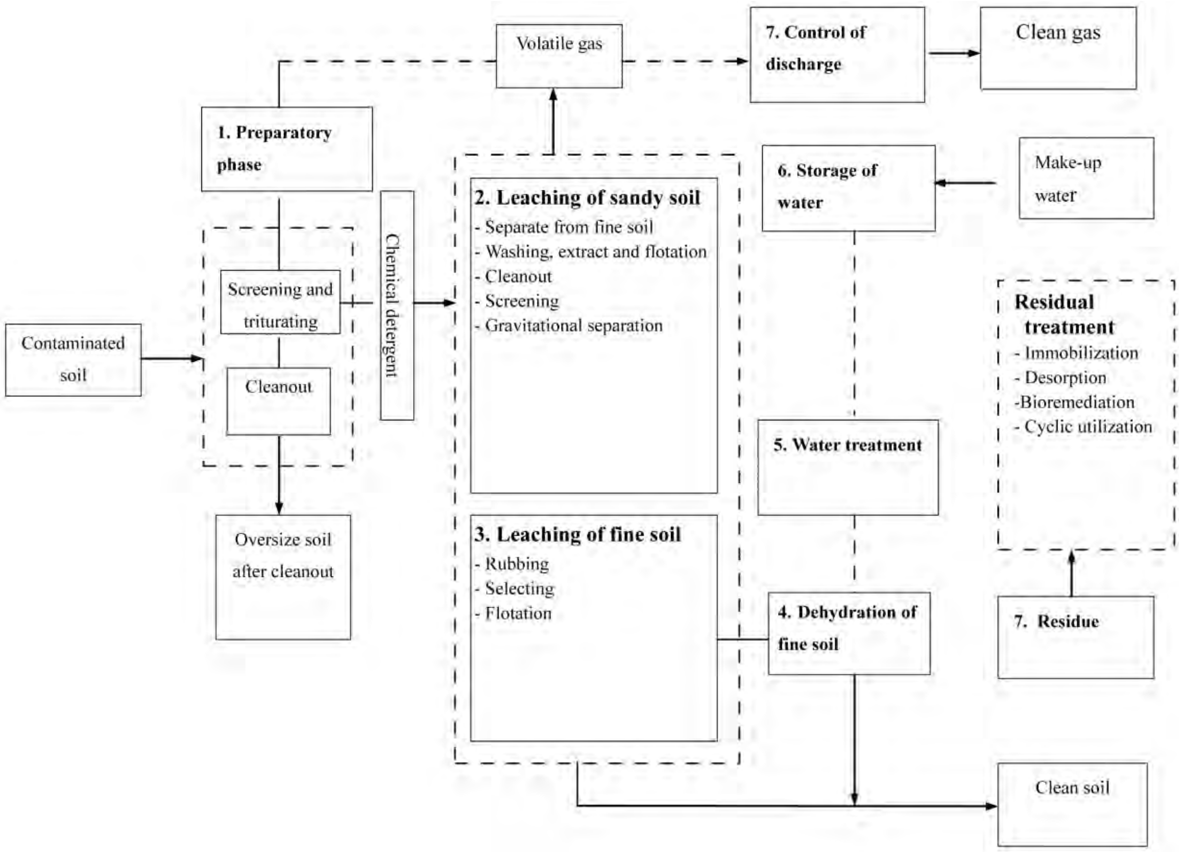

Figure 2. The flow diagram of soil ex-situ washing

Chemical leaching technology can remove heavy metals from the soil efficiently and thoroughly. It is suitable for repairing small area of soil which is polluted by heavy metal. It also can serve as a pretreatment technology, then combined with other methods to repair the contaminated soil polluted by heavy metal[42]. But the cost of chemical leaching technology is too high, and the use of drench lotion will reduce soil biological activity, so the application of chemical leaching technology is still in its infancy in our country.

C. *Solidification/stabilization*

The theory of solidification/stabilization technology is as following: adding reagent into the soil, pollutant reacting with reagent for physic-chemical reaction, turning it into fixed mercury, so as to reduce the mobility of mercury in soil. Cement, ash and many other substances are the commonly used physical curing agents. Cement is economic and the most commonly used, it converted the mercury in soil to mercuric oxide precipitation, and then fixeds it in the concrete block[43]. Commonly used materials for stabilization are s-based compound, zeolite, lime, calcium carbonate, phosphate, silicate, etc. Lime or calcium carbonate was mainly used to improve the pH value of soil, and made Hg and other elements in soil form precipitation of combination state salt of hydroxide or carbonate[44]. Zeolite can decrease the effectiveness of the heavy metals in soil through ion exchange adsorption and specific adsorption[27].

Chao[45] used Na_2S and cement as curing agent to dispose of mercury polluted soil from a chemical company. When the pH value was 8, the mercury concentration in the leaching liquid of curing block was lower than 1 ng/L and achieved the goal of innocent treatment of mercury polluted soil. Wu[46] optimized the ratio of stabilizer and curing agent to repair mercury polluted soil at a chemical enterprise in Yunnan, and improved the economic feasibility of the restoration work, made a significant reduction of mercury concentration in the leaching liquid. Wang[47] used the technology of adding fly ash to dropped mercury concentration in soil significantly.

Using solidification/stabilization technology to repair mercury polluted soil had good effect and low cost, but mercury is still exist in the soil, and the treated soil loses agricultural productivity and other functions.

IV. Bioremediation Technique

A. *Phytoremediation*

Phytoremediation is to use the extraction effect, volatile effect and curing effect of plants to reduce the content of mercury in soil[48].

The key of phytovolatilization technology is to embed mercury invertase genes into the plant bodies, then the plant can compound mercury invertase to transform combining state of mercury into zero-valent mercury when the combining state of mercury is absorbed from the soil into the plant, then the zero-valent mercury can be released by leaf transpiration. Researchers have implanted mercury reductase genes and organic lyase genes into many wetland plants such as typha, rice, rice straw and others. Meagher[49] obtained transgenic plants-arabidopsis whose tolerance ability of organic mercury increased 50 times. He[50] embedded the MerA genes of bacteria into tobacco, which made it can normally grow in $HgCl_2$ medium within 50 μM ~ 350 μM.

Perennial evergreen plants, resistant to high concentrations of heavy metals and root developed, could reduce the activity of heavy metals through root decomposition, precipitation, chelation, redox and so on, thus reduce the metal leaching into groundwater or the possibility of further environmental pollution through the air spread, which is referred to as plant curing[51]. The study indicates that the mercury enrichment capacity could reach 10 mg/kg by silver birch, while the maximum mercury absorption accumulation of Canada Populus was about 7 mg/kg[52]. Root is the main tissue part for mercury curing, the mercury content in the leaves of lobular boxwood accounting for 8.6% of that in the root soil[53]. Only 0.45% ~ 0.65% of absorbing mercury by willow root cell wall were transferred to the overground part of plant[54]. Compared with rape, wheat and pea, willow had not been found releasing mercury into the atmosphere[55]. Rice straw had stronger resistance to mercury[56] by converting organic mercury to inorganic mercury, which showed well potentiality of mercury-contaminated soil repairing. Humic acid could increase the residual mercury quantity in soil, and reduce the plant availability of mercury, thus effectively restraining the mercury accumulation in leaf[57-58].

Heavy metal hyperaccumulation plants absorb heavy metals in soil, transfer them to the overground parts to storage, then harvest to removal, which is called as plant extract. Mercury-hyperaccumulation plants have not been found. By artificial induction and adding the chelating agents such as $Na_2S_2O_3$, KI, NH_4SCN, EDTA and $(NH_4)_2S_2O_3$[59], the mercury extraction ability of plant was improved. Plant species is an important factor affecting the absorption of heavy metals. Huckabee[60] pointed out that the mercury enrichment capability of herbaceous plants was obviously higher than that of woody. Liu[61] found that the total mercury content in wetland plant was higher than that in upland crops, and the total mercury content in typical plants were as following: moss > algae > carex > grass > bush. Huang Hui[62] discovered that the addition of $Na_2S_2O_3$ could significantly promote the absorption of various forms of mercury in calendula and mustard, while the mercury enrichment in root was stronger than that in stem leaf part. Wang J also found that after adding thiosulfate in the soil, the content of effective state of mercury in soil was significantly increased, improving the phytoremediation efficiency of mercury-polluted soil by Indian mustard[63]. The mercury contents in the leaf, stem and root of tuber fern under slightly acidic conditions were higher than those under acidic or alkaline conditions[64]. Wet earth pot method research showed that the amount of mercury enrichment of different tissues and organs in bamboo reed were in order of root > stem > leaf[65].

Phytoremediation technology is of low cost and simple operation, which is suitable for a large area governance of mercury-polluted soil. However, the governance cycle is too long, during which the mercury evaporating into the atmosphere will cause certain influence to human health, and plants for heavy metals absorbing should be harvested and harmless timely.

B. *Bioremediation*

The use of microbial activity can adsorb or transform heavy metal affinity in the soil, increase stable-state metal content and thereby reduce the risk of being absorbed by organisms[66-67]. Recent studies have found that a mercury-resistant microbe with mer operon can effectively control mercury pollution in soil and water[68-69], and the evaporation rate of mercury in the soil was proportional to the number of microbes[70]. Spangler[71] inoculated four pseudomonades in a closed volumetric flask containing 50 ml culture media and 25 μg methylmercury bromides. After culturing for 5 days, methylmercury reduced by 50 %, and the upper part of the air in the flask generated methane and mercury vapor. In the experiment, Shariat[72] isolated 40 kinds of bacteria from soil, sediment and sewage treatment substance, among which 21 kinds of bacteria can achieve methylmercury demethylation. In addition, bacterial degradation range for methylmercury under aerobic conditions can be from 20% to 84% and desulfovibrio de-sulfuricans decreased 32% of methylmercury under anaerobic conditions. Japan developed a bioremediation technology that can collect bacteria rich in mercury and to remove mercury by the method of evaporation, activated carbon adsorption[27]. Yang Juhua[73] selected a highly mercury-resistant strain of pseudomonas putida, and after 48 hours, the reduction efficiency of 5 ~ 50 mg/L Hg^{2+} is from 94.3 % to 82.8%. Li Mengjie[74] took use of schizophyllum GGHN08-116 strain and cottonseed hull and corn stalk as substrate for solid-state fermentation to restore soil contaminated by mercury, lead and chromium so that the contents of these exchangeable elements can be decreased, while mercury and lead in carrots undetected.

Although the bioremediation is low in cost, high in efficiency and has little impact on environment, the treatment effect is limited by the type of contaminants and the restoration time lasts long.

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