Environmental Pollution and Remediation

Shunitz Tanaka

Contents

Abstract In this Chapter, as the background and introduction of this whole book, the following matters are mainly described. Firstly, it is stated that many environmental pollution problems occur still now in the world by various causes such as industrial activities, our daily and natural activities, and accidents and disasters. In order to solve these environmental pollution problems, the necessity of the development of environmental remediation technology is described. Next, some pollution problems, which Japan experienced previously and is currently facing, are intro-

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duced, for example, Minamata disease, low-concentration pollution problems such as dioxins and endocrine disruptors, and contamination with radionuclides from the explosion of the nuclear power plants caused by the big tsunami. After that, the concept and classification of the environmental remediation technology are described. It is shown that remediation consists of three concepts: containment, separation, and decomposition, and that remediation technologies are also classified into two categories, In-situ and Ex-situ. Several representative techniques for containment, separation, and decomposition are introduced briefly. The combination of some remediation technologies and the evaluation are also described, when these remediation techniques will be applied to the actual contaminated site. It is discussed why further development of remediation technology is necessary and why the design of materials for remediation technology is necessary. Finally, a brief introduction for the structure of this book and chapters is given.

Keywords Concepts for remediation, Containment, Environmental remediation, In-situ and Ex-situ, Low-concentration pollution, Minamata disease, Separation and decomposition

1 Introduction

On the earth, humankind has enlightened and developed its civilization over a long time. According to United Nations; World Population Prospects 2019, 7.7 billion people live on the earth in 2019 [1]. Population Division of UN prospects that the world population will be 9.7 billion in 2050. In order to maintain their lives, we must secure enough amounts of water, foods, and energy for them. For more wealthy life, we have promoted industries to manufacture various kinds of products massively, and circulated them over the world. In order to maintain such our lifestyles and industrial activities, we have always developed the resources of coal and oil as energy and minerals as raw materials from every place on the earth. We have also synthesized new materials that had not existed on the earth. As a result, our lives have become convenient and prosperous. On the other hand, the environments surrounding us have been flooded with every kind of things, and often polluted with harmful substances including natural and synthesized compounds. There are various kinds of pollutants, including heavy metals, harmful organic substances, organic chlorine compounds, asbestos, radioactive substances, and so on. The more sophisticate the current industry becomes, the more diverse of the materials are required there. Consequently, the sorts of pollutant are now diversifying.

Pollution arises from various causes. Pollution often occurs as the results of industrial activities in mines and factories. It also occurs from our daily lives and

Table 1 Some recent pollution problems by accident and disasters

Events	Date	Location and country	Pollutants	Amounts of pollutant
Nuclear power plant accident	April 26th, 1986	Chernobyl, Ukraine	Radionuclides	1.2×10^{19} Bq
Oil spill from a tanker, Nakhodka	Jan. 2nd, 1997	The Japan Sea, near Shimane	Heavy oil	6,240 kL
Chemical plant explosion	Nov. 13th, 2005	The Songhua river, North-east in China	Benzene, aniline, nitrobenzene	100 tons
Nuclear power plant accident	March 11th, 2011	Fukushima, Japan	Radionuclides	0.9×10^{18} Bq
Oil-drilling station accident	April 20th, 2010	The Gulf of Mexico	Crude oil	78×10^{3} kL
Oil spill from a cargo boat, Wakashio	July 25th, 2020	Mauritius, the Indian Ocean	Heavy oil	1,000 ton

natural processes such as volcanic activities. Accidents and disasters sometimes cause severe pollution problems. For example, an explosion of a chemical factory gave rise to the pollution of the Songhua River of China [2]. After the mega earthquake occurred on March 11 of 2011 in Tohoku area of Japan, the big tsunami attacked Fukushima Daiichi Nuclear Plants to lose the electricity for cooling the nuclear reactors, consequently the reactors exploded to pollute the wide areas of Fukushima and surrounding prefectures with plenty of radionuclides [3]. Some pollution problems happened recently by accidents and disasters are summarized in Table 1. And some examples of pollution problems are described in chapter "Pollution Sites Where Need Remediation".

How should we balance economic development with environmental preservation for a sustainable society? This is an unavoidable challenge for our and our future generations to cope with. According to the Sustainable Society Foundation, a sustainable society is defined as the following society: "that meets the needs of the present generation, and that does not compromise the ability of future generations to meet their own needs, in which each human being has the opportunity to develop itself in freedom, within a well-balanced society and in harmony with its surroundings" [4]. At the UN Summit held on Sep. 25 in 2015, the SDGs (Sustainable Development Goals) to achieve a better and more sustainable future for all were adopted. The SDGs consist of 17 challenges including those related to poverty, inequality, climate change, environmental degradation, peace, and justice [5]. The SDGs should be achieved by 2030 in the world, and then every countries and individuals are required to act as aiming these goals. The following six goals seem to be particularly related to environmental remediation and the goals which remediation technologies can contribute to fully (Table 2).

Table 2 SDGs related to environmental remediation

Goal 3	Good health and well-being: ensure healthy lives and promote well-being for all at all ages
Goal 6	Clean water and sanitation: ensure access to water and sanitation for all
Goal 9	Industry, innovation, and infrastructure: build resilient infrastructure, promote sustainable industrialization and foster innovation
Goal 13	Climate action: take urgent action to combat climate change and its impacts
Goal 14	Life below water: conserve and sustainably use the oceans, seas, and marine resources
Goal 15	Life on land: sustainably manage forests, combat desertification, halt and reverse land degradation, halt biodiversity loss

2 Environmental Pollution Problems in Japan

2.1 Occurrence of "Ko-gai": Public Pollution Problems

In Japan, many environmental pollution problems occurred in the 1950–1970s during the economic recovery and growth in Japan after the World War II. Minamata disease, Itai-itai disease, Yokkaichi air pollution, etc., are well known in the world as the representative pollution problems in Japan. These pollution problems were called "Ko-gai" in Japanese, which means "public pollution problems." Japan's economic growth-first policy during this period gave priority to the economic development over the consideration for the environment and consequently it led to many pollution problems. Those problems were not just the pollution which private companies caused but the pollution which Japan's policy during those days caused. Therefore, those pollution problems are considered as the public problems, "Ko-gai." In the 1970s, some countermeasures were conducted, and in 1973, a law, "Basic Law on Pollution Control," which was the first environmental law in Japan, was enacted. Some environmental standards were enforced in this law, and thereafter various countermeasures to keep these standards have been carried out on the basis of this law in Japan. As a result, "Ko-gai"-type environmental pollution problems in Japan gradually decreased. In most cases of "Ko-gai"-type pollution, pollutants were released from the specific industrial process and facility, and then if any countermeasure was taken against the source of pollutants, most of pollution problems could be solved. Figure 1 shows the change in the non-achievement rate for some environmental standards after the enforcement of the environmental law [6]. The non-achievement rate has decreased rapidly after the enforcement and now the rates are below 1% in many items.

Minamata disease was one of the typical "Ko-gai" problems. The pollution problem occurred in Minamata city of Kumamoto prefecture in Kyushu island of Japan. Since the first finding of a patient of Minamata disease in 1956, many patients were found out from mainly fishermen and their family who had been fishing in Minamata Bay. The patients of Minamata disease had a disorder in motor and

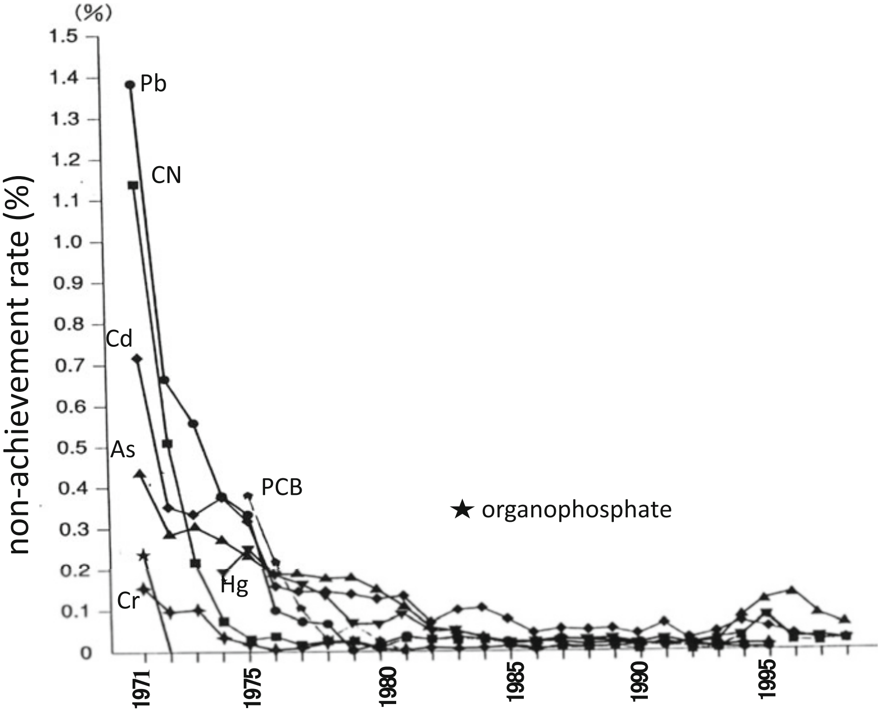


Fig. 1 Change in non-achievement rate for some environmental standards (from The Annual Report of the Environment of Japan (1995) [6]

language, and sometimes a problem in brain. It took very long time to clarify the cause of Minamata disease. In 1959, methylmercury discharged from a factory of a chemical company was suggested to be a cause by a research group of Kumamoto University. At those days, the factory had produced acetaldehyde from acetylene by using mercury salt as a catalyst. The inorganic mercury might be converted to methylmercury by the reactions during the production process. However, some authorities such as the academic society, the industrial society, and university in Japan declared other opinions about the cause of Minamata disease. Their opinions confused the clarification of the cause and made it prolong to determine the cause. Later Dr. Nishimura et al. showed the reaction mechanism for methylmercury formation during acetaldehyde production in their book as shown in Fig. 2 [7]. Methylmercury can pass through the placenta and be accumulated in a body of fetus and then many children having the disorder in motor and brain were born. In 1967, Japanese government admitted Minamata disease as a "Koh-gai," a public disease with methylmercury. After many conflicts between fishermen and the company concerning the countermeasures and compensation, finally in 1968, the company stopped the production of acetaldehyde. In 1965, the second Minamata disease was found in Niigata Prefecture in Japan [8].

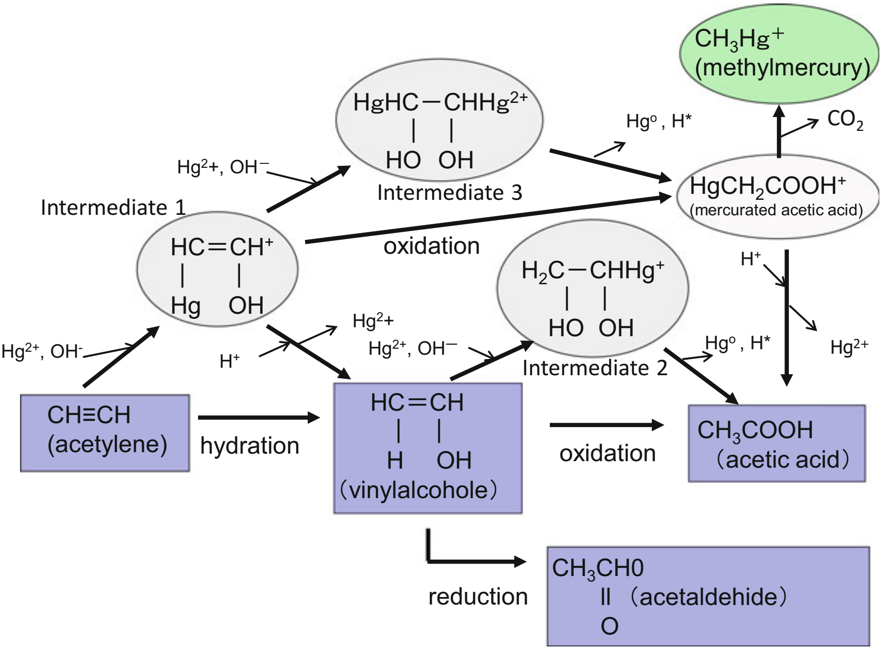


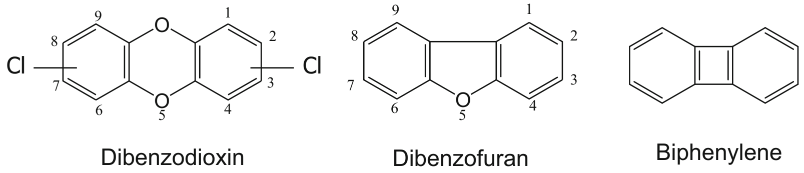
Fig. 2 Reaction mechanism for methylmercury formation during acetaldehyde production [7]

The "Ko-gai" type of the pollution problems like Minamata disease has not been completely solved in Japan. Even now, there are many designated patients of Minamata disease, and also many not-designated patients are still waiting to be recognized as a patient of Minamata disease by the struggle in court and administration. Environmental pollutions with mercury seem to be spreading all over the world, and the third and fourth Minamata diseases may occur or have occurred somewhere in the world. For this reason, the Minamata Convention on Mercury (Minamata Treaty) [9] was adopted at UN meeting held at Minamata city in 2013 with the aim of reducing and controlling the use of mercury worldwide.

In Japan, in order to cope with global environmental problems such as global warming and ozone layer depletion, a new law, the Basic Environmental Law, was established in 1993 instead the old law. In this law, preventive and diverse methods were introduced to in addition to ordinal regulatory methods and the countermeasures based on environmental education and international cooperation are recommended.

2.2 *Low-Concentration Pollution Problem*

In the 1990s, Japan was facing pollution problems with dioxins and environmental hormones (endocrine disrupting chemicals). These pollutions have different



No. of Cl		M.W.	No. of isomers
1	M1CDD	218	2
2	D2CDD	252	10
3	T3CDD	286	14
4	T4CDD	320	22
5	P5CDD	354	14
6	H6CDD	388	10
7	H7CDD	422	2
8	O8CDD	456	1
		total	75

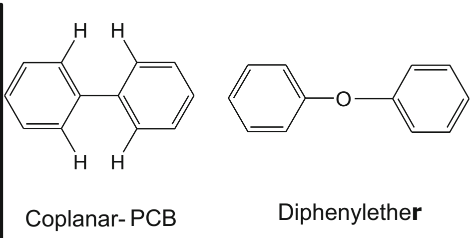


Fig. 3 Structures of dioxin and dioxin analogs

characteristics from previous pollution problems, “Koh-gai,” in the point that the pollutants affect human health and ecosystems at very low concentration. In addition, pollutants are released from unspecific and multiple sources but not from a specific factory like “Koh-gai”-type. Especially dioxins are generated even when garbage routinely discharged from our ordinary households is combusted. Dioxin generally represents polychlorinated dibenzo-dioxin and some analogs such as dibenzofuran and biphenylene are also included in Dioxins (Dioxin group). Each analog has many isomers and congeners. Figure 3 shows the structures of dioxin and dioxin analogs. Among them, 2, 3, 7, 8-dibenzo-*p*-dioxine is known to have the strongest toxicity. Dioxins are unintentionally generated chemicals at the incineration plants in city and also in hospitals, schools, companies, etc. If pollution with dioxins comes from the combustion of our household garbage, the perpetrators of dioxin pollution are ourselves who generate the garbage, and the victims are also us. In “Ko-gai” pollution problems, the perpetrators and victims could be clearly distinguished, but not in the case of dioxins pollution problems.

In Japan, a special law for dioxins (Law concerning Special Measures against Dioxins) was established in 1999 [10], although scientific information was not enough. The criteria for the concentration of dioxins in the emitted gases and water from an incinerator were established. The owner of an incinerator is obligated by this law to report the concentration of dioxins in and around the incinerator. Consequently, most of small incinerators disappeared from hospital, school, and home. People stopped burning garbage in their garden. Local governments replaced an old incinerator with a new one which could burn garbage at the higher temperature not to generate a large amount of dioxins. As a result, the amount of dioxins discharged from the incinerator has been reduced largely to be negligible now in Japan as shown in Fig. 4 [11].

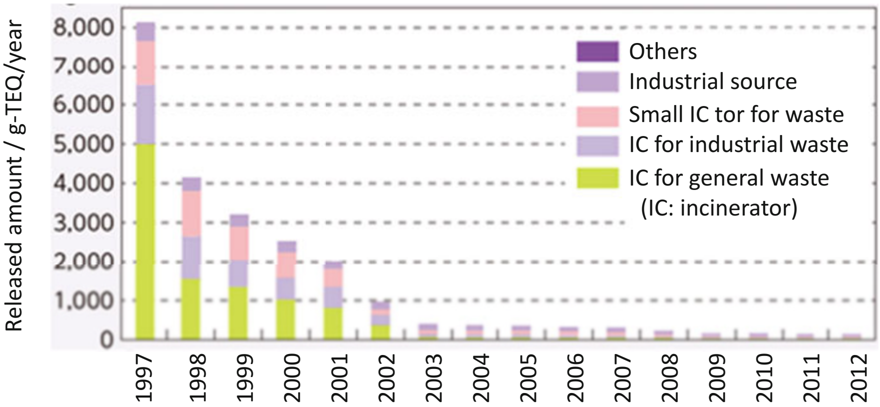


Fig. 4 Released amount of dioxins from various sources (from The Annual Report of the Environment of Japan (2014) [11])

In Japan, the pollution problem by endocrine disrupting chemicals was a notable environmental topic in the 1990s. These chemicals were called "environmental hormones" in Japan because those chemicals acted like hormones and affected human health and ecosystems, especially on reproduction and the next generation, even though at very low concentration. Many researchers joined in the studies on the detection of many kinds of chemicals in environments and the effects on human health and ecosystems. It was like a fever as involving many mass media. Two books of "Silence Spring" by Rachel Carson and "Our stolen future" by Theo Colborn must enhance not a little the concern and fears of people for the chemicals. The Japanese Ministry of the Environment has launched a program, "Strategic Programs on Environmental Endocrine Disruptors (SPEED98)" in 1998 to urgently investigate the effects of endocrine disrupting chemicals on human health and ecosystems [12]. The program concluded that few chemicals showed the endocrine disrupting effects on the human health, a few chemicals showed some effects on ecosystem and there might be still now many things which we did not know. The program was carried on to a new program, EXTEND 2010 program [13]. However, the concerns of people and the interest of the media in the endocrine disrupting chemicals declined rapidly.

Polychlorinated biphenyl (PCB) is an artificially synthesized compound. This compound is very stable chemically and biologically and has high insulated and fire-resistant properties. Therefore, PCB has been used in various purposes, insulated oil for condenser and capacitor, paint, heat-transfer fluid, transfer paper, and so on, and it was called "a dreamy chemical." However, the toxicity of PCB to human health and ecosystem was found out and then the production and use were banned. The more stable and persistent the compound is, the longer the compound can stay in environments. Since PCB is almost insoluble in water, the concentration of PCB in water is usually very low. However, the hydrophobic and persistent properties of PCB facilitate the bio-concentration. Even though the concentration of PCB in water is very low, PCB is accumulated gradually in the living body to be a significant concentration to give some damages. Therefore, PCB pollution problem has a common feature with the low-concentration pollution problem such as dioxins and environmental hormones. PCB has many isomers and congeners with different numbers of chlorine at the different positions. Among these congeners, the PCB isomers having no chlorine at the position of 2, 2’, 6, and 6’ are called “coplanar PCB: Co-PCB” because these have the co-planarity of two benzene rings. The structure of Co-PCBs is similar to that of dioxin and then they have the strong toxicity like dioxins. Therefore, Co-PCBs are regarded as one of dioxins. In Japan, the treatment of PCB oil and the products containing PCB such as condenser and capacitor has been conducted by Japan Environmental Storage and Safety Corporation (JESCO), whose treatment facilities exist at five areas in Japan and all of PCB including high and low concentration will be decomposed by 2027 [14].

The measure for the low-concentration pollution has some difficulties as follows:

1. It is difficult to detect the pollutants and then to know the pollution situation exactly.
2. It is difficult to evaluate the impacts of the pollution because it may take a long time to appear the impacts by the pollution, sometimes the impacts may appear in the next generation.
3. It is difficult to find out the effective countermeasures against the pollutants, because the concentration is too low to apply the conventional treatment methods to them. Pre-collection and accumulation of the pollutants are necessary before the treatment. In order to cope with low-concentration pollution, the novel technologies for high sensitive detection and selective accumulation of pollutants are required.

As described previously, if a pollutant is persistent and stays in the environment for a long time, the pollutant may be accumulated to the level that can affect human health by bio-concentration even at low concentration. In particular, the effects of long-term exposure on children have not yet been clarified. Currently a project for investigation of the effects of the long-term exposure on children is in progress [15]. A part of the results of the project will be described in detail in chapter “Effects of Persistent Organic Pollutants (POPs) in the Ecosystem and HUMAN health: Focusing on Chlorinated Chemicals”.

2.3 Contamination with Radionuclides by Nuclear Power Plant Accident

On March 11 of 2011, an earthquake of the magnitude 9.0 attacked the northeastern Japan followed by a big tsunami. The tsunami over 20 m rushed toward the shore of the wide areas and citizens more than 15,000 died and about 2,500 have been still missing by the earthquake and tsunami [16]. The tsunami also attacked Fukushima Daiichi Nuclear Power Plants and destroyed the electric systems to cool the reactors.

Consequently, four nuclear reactors exploded to release a large amount of radionuclides into environments. The amount of released radionuclides, 0.9×10^{18} Bq, was as large as one-tenth of the amount released in the nuclear plant accident at Chernobyl [17]. The radionuclides firstly released into the atmosphere were carried by the wind to fall down into soil, farmland, and forests throughout Fukushima prefecture and the neighboring prefectures.

Two months after the accident, the Ministry of Education, Culture, Sports, Science and Technology of Japan, some universities and institutes cooperatively conducted an emergency survey of soil pollution with radionuclides within a radius of 100 km from the nuclear power plants. According to the results of the survey, it was found that the wide areas in Fukushima were polluted with radionuclides, especially with ^{134} Cs, ^{137} Cs, and others, The pollution was spreading distinctively in the northwest direction from the site of the nuclear power plants as shown in Fig. 5 [3]. The radioactive Cs has been remaining in the surface soil within several cm because Cs ions bound strongly to clay minerals in soil. The Ministry of the Environment recommended a series of the processes shown in Fig. 6 as a decontamination method for the surface soil. The upper 5 cm of the contaminated surface soil is stripped off and put into the large bags (flexible containers). These bags are stored in a temporary storage site for 3 years and then moved sequentially to an intermediate storage facility for 30 years. Lastly the contaminated soil is moved to the final disposal facility [18]. However, the amount of contaminated soil is too huge

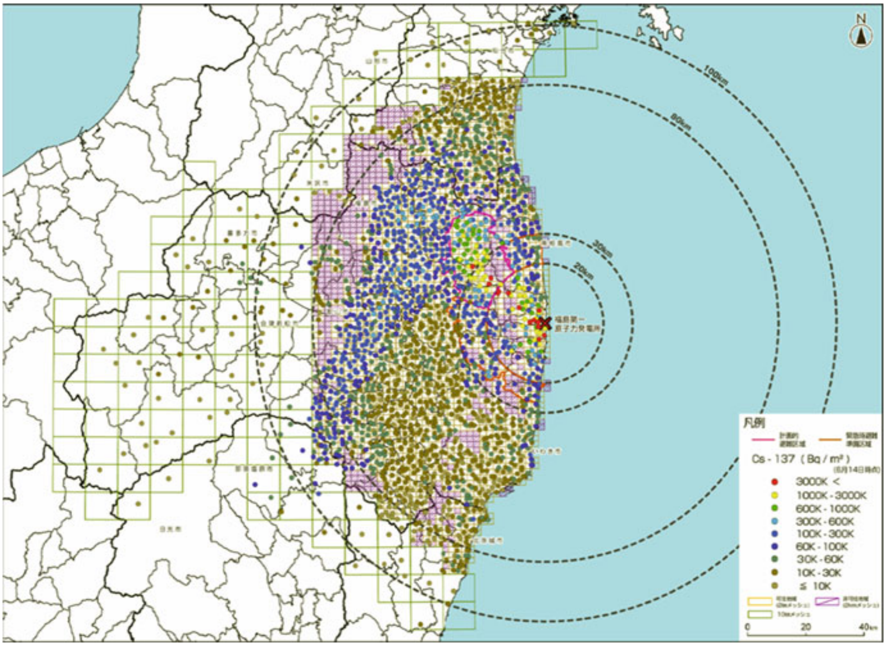


Fig. 5 Distribution of ^{137}Cs in soil around of the NPP [3]

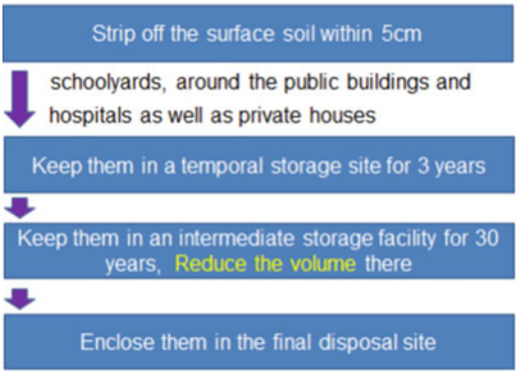


Fig. 6 Flow of decontamination for polluted soils proposed by the Ministry of Environment, Japan

to treat and store them and the construction of the intermediate storage facilities has not so progressed. Moreover, Japanese government has not yet decided the locations for the final disposal facility. It is doubtful whether the decontamination would proceed as planned or not. Even now, 9 years after the accident, there are many residents who cannot return to their homes because the areas around their houses have been designated as the evacuation areas.

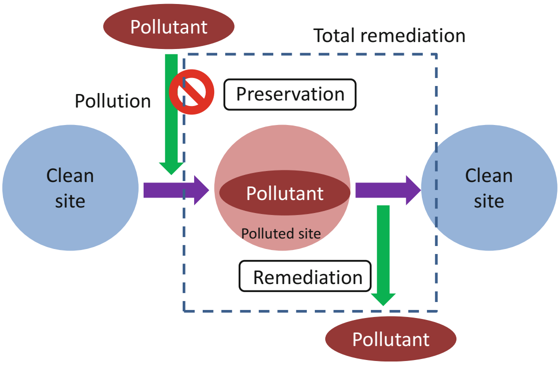
If radionuclides could be removed from contaminated soil by any methods, the volume of the contaminated soil could be reduced to scale down the number and size of the storage facilities. Many researchers have intended to develop various kinds of treatment methods using phytoremediation, adsorption, hydrothermal method, electrochemical method, etc. So far, the practical methods which can be applied to actual contaminated soil have not yet been found out because most of the reported methods have disadvantages in terms of processing time and cost. A Korean researcher, Kim studied on the removal of radionuclides from contaminated soil by a combined method of soil washing and electro-kinetic method [19]. We also investigated EK method to remove Cs ion from soil and some results of these studies are described in chapter “Electrokinetic Remediation”.

3 Environmental Remediation

3.1 Concepts of Environmental Remediation

What we should do firstly to keep the earth’s environments clean is to not release pollutants to environments. In all processes of manufacturing, we have to introduce pollutant-free processes by developing new technologies. These technologies are called conservation/preservation, zero-emission technology, and sometimes green chemistry for the technology in the field of chemistry. On the other hand, there are already many contaminated environments around us. For these environments, it is necessary to restore the previous clean and safe situation by removing the pollutant.

Fig. 7 True remediation technology contains the preservation technology



That process is the environmental remediation and the technology used for that purpose is the remediation technology. The remediation technologies are also called restoration, decontamination, cleaning, and so on. The definitions of these terms are not always clear.

The technologies for preservation and remediation should be complementary to each other. Even though a contaminated site could be restored completely by the remediation technology, as far as the source which releases a pollutant still exists, the site will be again contaminated with the pollutant. Therefore, the true remediation technology should contain the preservation technology which can suppress the release of the pollutant from the source and the injection to the site as shown in Fig. 7. If the ultimate preservation technologies, which can suppress the release of pollutants completely, could be established in all processes, the remediation technologies may be not necessary. So far, unfortunately such preservation technologies have not been established in most fields. The complementary roles of two kinds of the technology are shown in Fig. 8. In the figure, at first, before starting remediation, a certain amount of pollution exists. The amount of the pollution consists of not only already existing pollutants but also newly injected pollutants. The pollution is treated by the remediation technology and the amount is reduced as a function of time. At the same time, the pollution is treated by preservation technologies and the amount of the pollutant is also reduced as a function of time. Finally the remaining pollution is disappeared by the complementary actions of the remediation and preservation.

3.2 *Classification of Remediation Technologies*

The diverse environments of aqueous sphere, atmosphere, and soil can be the targets of remediation. Various kinds of substances such as metals, harmful organic matters, radionuclides, and so on are also the target substances of remediation. The situation of pollution differs from site and site, and the concentration and toxicity of pollutants

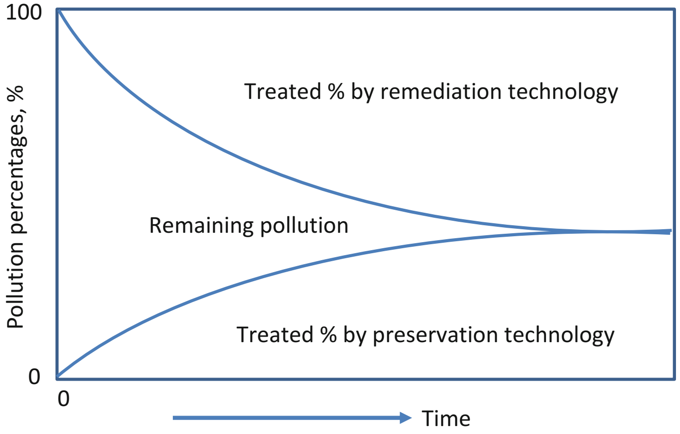


Fig. 8 Complementary relationship between remediation and preservation technologies

and the environments surrounding the polluted site are also different. The removal efficiency of pollutants required depends on the status of the site. Some site requires the complete removal of pollutant but other site not so because the reduction of pollutants by natural attenuation can be expected there after the remediation. For such diverse requirements, we have to prepare many kinds of remediation technologies.

Remediation technologies are classified into three groups depending on these concepts, containment, separation, and decomposition. In addition to these, accumulation technologies are required for the treatment of low-concentration pollutants but these technologies are included in separation. Table 3 shows the classification of some representative remediation technologies [20–22].

In containment, pollutants are converted to stable and immobile substances or enveloped into the stable material to prevent pollutants from spreading more in environments. In separation, pollutants are removed from contaminated site by various kinds of chemical, physical, and biological methods. In decomposition, harmful pollutants are converted to harmless compounds and destroyed completely to form carbon dioxide and water by various chemical and biological processes. A remediation technology may be used independently or some technologies may be combined, for example, a pollutant is firstly contained for a certain period (containment technology), then it is separated from the matrix (separation technology) and finally it is decomposed (decomposition technology).

The remediation methods are also classified into In-situ and Ex-situ methods. In-situ means on-site, that is, remediation is carried out on and in the vicinity of the contaminated site and Ex-situ means off-site, that is, contaminated matters are transported to the treatment facility and treated there as shown in Fig. 9. In a case of Ex-situ remediation for contaminated soil, the action of digging out contaminated soil in order to transport it gives a heavy load to the land, and during digging and transportation of contaminated soil, the pollutant in the soil may spill to

Table 3 Classification of representative remediation technologies [20–22]

Concept	Technology	Main principle	Materials used	In-situ or Ex-situ	Targets materials	Remarks
Containment	Slurry wall	Surrounding with low permeable clay mineral	Clay mineral and cement	In-situ	Heavy metal ions, harmful organics, oil	Low-cost, easy construction
	Deep geological disposal	Storing in deep stable stratum	Glass and canister	In-situ	High-level radioactive waste	Need a long management, no-exposure for current generation
	Vitrification	Enveloping with stable glass material	SiO_2, Al_2O_3, graphite	In-situ and Ex-situ	Heavy metals, radionuclides, harmful organics	Need high current and voltage, 1,000 kWh/ton for In-situ
	Precipitation	Precipitation reaction	Hydroxide, sulfide and xanthate	Ex-situ and In-situ	Heavy metal ions	The secondary pollution by added reagent should be addressed
	Solidification and Stabilization	Encapsulating into solid material	Cement, pozzolanic, thermoplastic	Ex-situ and In-situ	Heavy metals and harmful organics	Some factors may interfere this process
Separation	Solvent extraction	Distribution to organic phase	Organic solvent	Ex-situ	PCB. dioxins, pesticides, and oil	Liquefied gas and critical solution temp. solvents also have been used
	Electro-kinetic remediation	Electrophoresis, electroosmotic flow	EDTA and surfactants and so on are used as an additive	In-situ	Heavy metal ions, harmful organics	Applicable to low permeable clayey soil
	Electrochemical accumulation	Oxidative film formation on electrode	Carbon fiber electrode	Ex-situ	Phenols and aniline	Some organics are accumulated on the electrode as a polymer film
	Soil washing	Mechanically and chemically scrubbing, particle size separation	Acid, base and surfactants	Ex-situ	Oil, organics and heavy metals	Need to treat washing water used
	Soil flashing	Chelating and solubilization	Water, surfactants	In-situ	Hydrophilic and hydrophobic organics	Flushing liquids should be recovered and recycled

	Adsorption	Adsorbing ability, ion-exchange, chelating	Many kinds of adsorbent, activated carbon, ion-exchange resin, chelating resin	Ex-situ and In-situ	Organics, heavy metals and radionuclides	Need to recover adsorbent after adsorption
	Vapor extraction	Volatilization, vapor flow	Hot air, steam	In-situ	Volatile organics and fuels	Injection of hot air or steam facilitates volatilization
	Phytoremediation	Uptake by plants	Hyperaccumulator plants	In-situ	Metals, organics	Need to manage and treat the plant used
Decomposition	Bioremediation	Biodegradation	Microorganisms, bacteria, fungi, actinomyces	Ex-situ and In-situ	Fuel oil, waste oil, pesticides	Need to control moisture, oxygen, pH, and nutrients in process
	Catalytic and photocatalytic reaction	Catalytic and photocatalytic reaction	Heterogeneous, homogeneous catalyst and TiO_2	Ex-situ	Organics, NOx and SOx	Sunlight or UV light is necessary for photo-catalytic reaction
	Zero valent iron	Reduction, and oxidation by hydroxyl radical (Fenton reaction)	Micro- and nano-Fe(0) particles	In-situ and Ex-situ	Chlorinated organics, nitroaromatic organics and others	ZVI is often used in the permeable reactive barriers (PRBs)
	Supercritical water oxidation	Oxidative degradation and dehalogenation near the critical point	Oxygen, hydrogen peroxide	Ex-situ	Halogenated organics, PCBs and pesticides	Need high temp. and pressure and corrosion-resist equipment
	Chemical oxidation	Oxidation reaction	Ozone, hydrogen peroxide, hypochlorite, chlorine	Ex-situ	Amines, chlorinated organics, cyanide, mercaptans, and others	Combining with UV photolysis improves the efficiency of degradation
	Electro-degradation	Electrolysis	SnO_2 electrode	Ex-situ	Organics	Some organics can be decomposed at the electrode
	Thermal degradation	Incineration, pyrolysis	Auxiliary fuel	Ex-situ	Various kinds of organics	More toxic compounds sometimes may be formed under the inadequate incineration condition

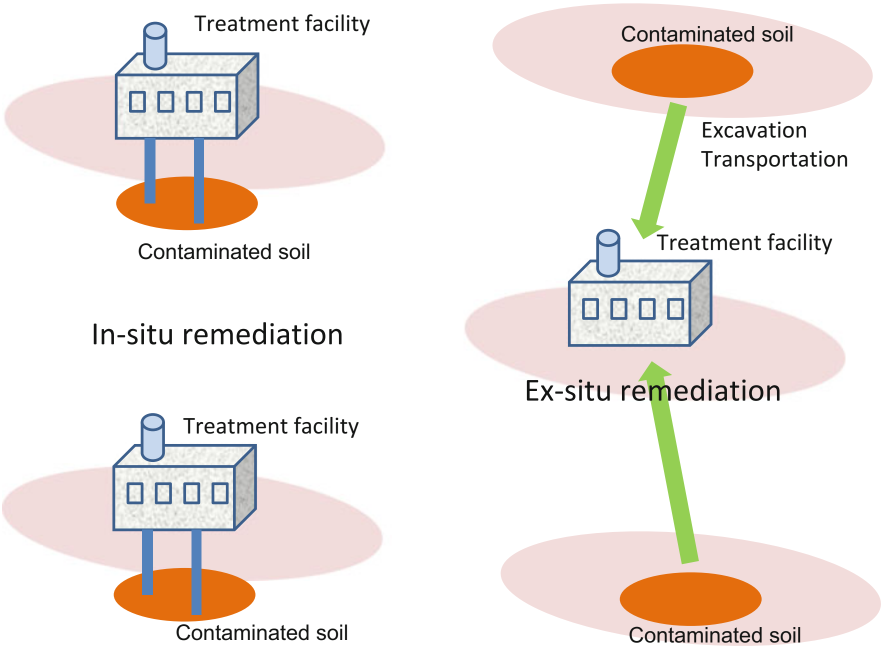


Fig. 9 In-situ and Ex-situ remediation technologies

environments and cause the secondary pollution. Therefore, recently In-situ remediation methods which do not need digging and transportation are preferred more than Ex-situ remediation.

3.2.1 Containment

In containment, to prevent pollutants from spreading in environments, pollutants are confined in a limited area by various chemical and physical methods or converted into immobile forms. The concept of the containment is based on the following relation on the risk of chemicals.

$$\text{Risk} = \text{Severity of Effect} \times \text{Possibility of Exposure}$$

The risk of a pollutant is represented as the product of the severity of the effect from pollutant and the possibility of the exposure of us with the pollutant. Even though a pollutant has a very high toxicity, if the possibility that we are exposed with the pollutant would be almost zero, then the total risk by the pollutant might be almost zero. Containment is a process to minimize the possibility of the exposure with pollutants. Many methods are applied and developed to minimize the exposure with pollutants. One of these methods is to make pollutant itself immobile. Some precipitation reactions are used for the purpose, for example, heavy metal ions such as Cd(II) react with hydroxide and sulfide to form precipitation of $Cd(OH)_2$ and CdS, respectively. These precipitations are water-insoluble and can stay as a solid in soil for a long time.

$$\mathrm{Cd^{2+} + 2OH^- \rightarrow Cd(OH)_2}$$

$$\mathrm{Cd^{2+} + S^{2-} \rightarrow CdS}$$

Hexavalent Cr(VI) is very toxic and carcinogenic. It is so water-soluble that can move easily in environments. While trivalent Cr(III) is one of essential elements and forms the precipitate with hydroxide in neutral pH. Therefore, the reduction of Cr (VI) to Cr(III) contributes to prevent Cr(VI) from spreading in environments. Some natural organic matters such as fulvic and humic acids can be used as a reductant for Cr(VI) [23]. Another method to minimize the spreading of pollutants is to envelop the pollutants with stable materials such as concrete, glass, and clay minerals. For example, heavy metal ions and radionuclides are solidified with concrete and glass. Vitrification is a method to envelop the pollutants into glass. In this method, pollutants such as heavy metal ions and radionuclides are heated with SiO_2 and Al_2O_3 as the raw materials for glass at the higher temperature than 1,100°C. The vitrification can be applied to the actual contaminated site as an In-situ method. Two electrodes are embedded near the contaminated soil and a large amount of the electricity of 4,000 V/400 A is applied between the electrodes, and the soil between the electrodes is heated at 1100-1500°C. Usually natural soil consists of mainly SiO_2 and Al_2O_3 and then soil is vitrified to confine the pollutants in the glass [20].

Clay mineral has a very low permeability for water. If a wall made with slurry prepared by mixing clay mineral, concrete, and water is constructed like surrounding a contaminated site as shown in Fig. 10, the wall can shut pollutants off from

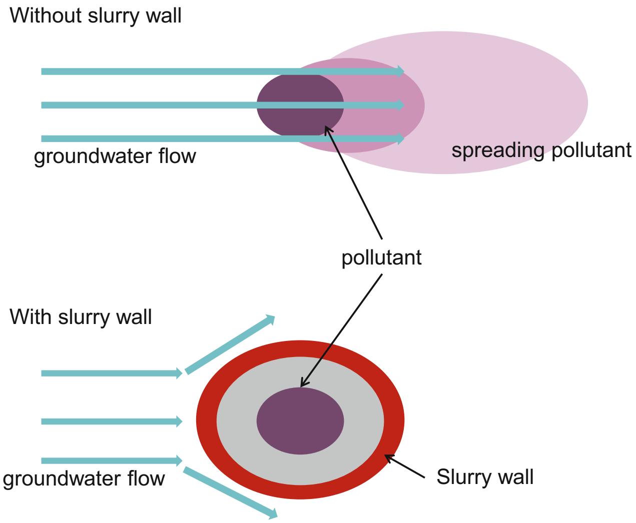


Fig. 10 Slurry wall method

spreading. The wall is called "slurry wall" and from the reason of relatively low cost, the slurry wall is applied to many contaminated sites, especially to the site just after the disclosure of the pollution. The migrating rate of water in the slurry is known to be about 10^{-3}–10^{-6} cm/s and then if 1 m length of the slurry wall with the lowest permeability is constructed around the contaminated site, the pollutants would be stored inside the slurry wall at least for 3 years. During this containing period, we can plan the next treatment of the pollutant and operate actually the process to remove or decompose the pollutants enclosed with the slurry wall. The containment technology is expected most for the deep geological disposal of high-level radionuclides. The effect of radiation from radionuclides on human health is very severe. In order to reduce the risk of high-level radionuclides, it is necessary to minimize the possibility of the exposure with them to almost zero. Therefore, high-level radionuclides are vitrified and then put into a canister made from stainless steel. The canister enveloped with a buffer material such as clay mineral is transported to the deep soil by passing through the vertical shaft and stored in the tunnel built in 500–1,000 m depth as shown in Fig. 11. Under this condition, the possibility that ordinal people could expose with the radionuclides in the tunnel would be almost zero. This method is expected to be a final disposal technology for high-level radioactive waste generated from nuclear atomic plants. In Finland, a deep geological disposal using the facilities of an abandoned mine, which is called "Onkalo," is now in the practical application stage [24]. In Japan, "Horonobe Underground Research Center," one of the research center of Japan of Atomic Energy Agency (JAEA) has conducted the studies on the deep geological disposal for high-level

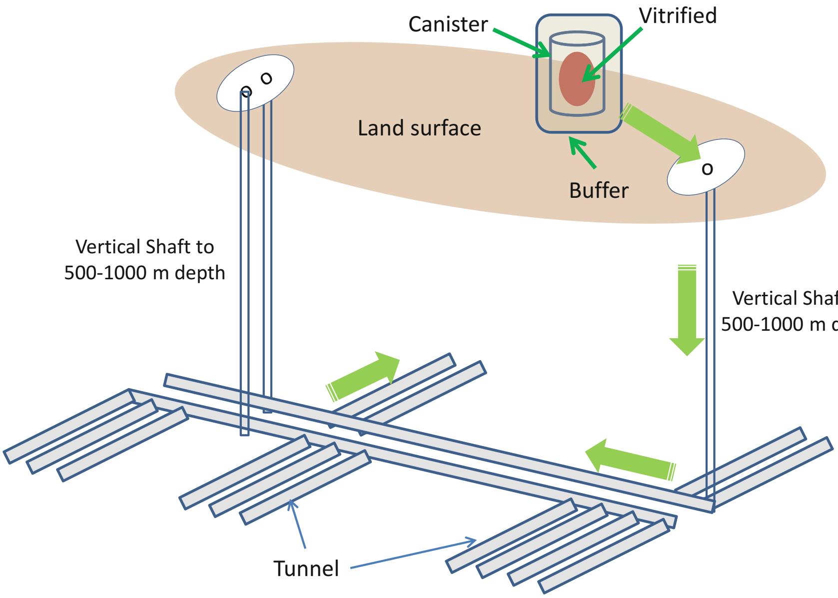


Fig. 11 Deep geological disposal of high-level radionuclides

radioactive waste by using the pits of an abandoned mine at Horonobe in Hokkaido [25]. The purpose of this center is to investigate the behaviors of high-level radionuclides in actual deep geological environments, to find out the factors which can give some effects on the behaviors and finally to verify the reliability of this method for a long period. However, this center will be used only for the research but not for the actual disposal. The national government promised the local residents that after the research completion or after the promised period, the facility will be backfilled and never used for the actual disposal. The area around Horonobe, the northern Hokkaido, is one of the leading agricultural and dairy farming areas in Hokkaido, and if the facility of Horonobe would be used as an actual disposal site, a plenty of high-level radioactive waste will be transported in this area from all over Japan, consequently rumor damages for agricultural products may be concerned.

The deep geological disposal is expected as a practically applicable method for the containment of high-level radioactive waste, but has some problems. Ordinary, the migration of radionuclides in soil is extremely limited (2 cm/year) due to the ion-exchanging capacity of soil. Therefore, even if radionuclides enclosed in the vitrified material would leak from there, soil would prevent the radionuclides from spreading into environments. However, it is known that soil organic matters such as humic substances would act as a carrier to increase the migration distance of the radionuclides in soil [26]. The behavior of radioactive materials in soil probably varies depending on various conditions. In addition, the lifetime of radionuclides often lasts for thousands or tens of thousands of years and more, and then the deep underground disposal facilities must be maintained for a long period corresponding to their lifetime. Therefore, even if the storage facility exists now in deep underground, the possibility that it may appear on the surface of the land in far-off future is not zero because the crustal movements can cause for a long period. The risk of the current generation may be zero, but the risk of later generations is not necessarily zero.

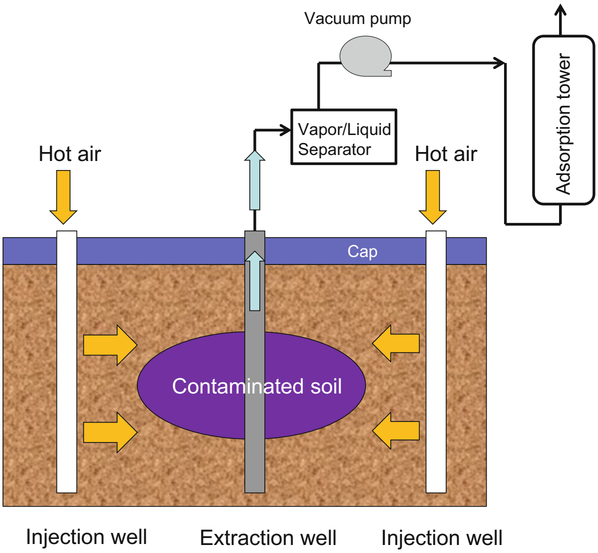
Containment methods can be viewed as the chronal transfer of the risk in a certain sense. That is, a current existing risk is contained for a certain period and transferred to the future. The negative legacy accompanied with the benefits that the current generation has received should not be handed to the later generation without any treatment by the current generation.

3.2.2 Separation

Some containment methods, especially slurry wall method, are relatively low-cost and easy to build up in the contaminated sites. However, contaminants have not been disappeared from that location and continue to stay there although the spreading can be suppressed. There are some threats that someday the materials and wall for the containment may be collapsed to release pollutants into environments. Therefore, it is necessary to plan the next measures during containing pollutants in the wall and to conduct the measures. Separation is a method for separating and removing pollutants from contaminated matters and sites. Various methods have been devised depending on the type of pollutants and the situation of the contaminated site. For the separation of pollutants suspended in an aqueous solution, physical separation methods such as filtration and centrifugation can be applied to, and for heavy metal ions dissolving in the aqueous solution, various chemical and physical methods can be applied to. For example, heavy metal ions are precipitated with hydroxide or sulfide and then the precipitants are filtrated or centrifuged. Many other chemical methods such as acid elution, organic solvent extraction, ion-exchange resin, chelating resin, and adsorption are used as the fundamental separation techniques, and some of them are described in the later chapters.

Some separation techniques are used for In-situ remediation. In-situ vapor extraction method is applied to the remediation of contaminated soil with volatile organic compounds such as jet fuel, gasoline, and so on. As shown in Fig. 12, the pipes or thin wells are inserted or dug in the vicinity of the contaminated soil and volatile pollutants are sucked with a pump through the pipes or wells [21]. After passing through a gas-liquid separation, volatile pollutants are introduced to the adsorption tower filled with adsorbent like activated carbon. Finally pollutants are removed by the adsorption and clean air is exhausted to the atmosphere. By injecting hot air to the vicinity of the contaminated soil, the rapid treatment can be achieved by the acceleration of the volatilization of pollutant [22]. Soil flushing is also an In-situ separation method for contaminated soil, in which washing water is sprayed on the surface of contaminated soil and contaminants in soil are washed away as washing water soaks into the soil. The soil flushing and extraction methods using aqueous solution cannot be applied to clayey soil because the low water permeability of the clayey soil prevents water from soaking into soil. Electro-kinetic remediation is only one method which can move water in clayey soil by the electroosmotic flow. In EK as shown in Fig. 13, if a pollutant has positive or negative charges, pollutants in soil

Fig. 12 In-situ vapor extraction [21]



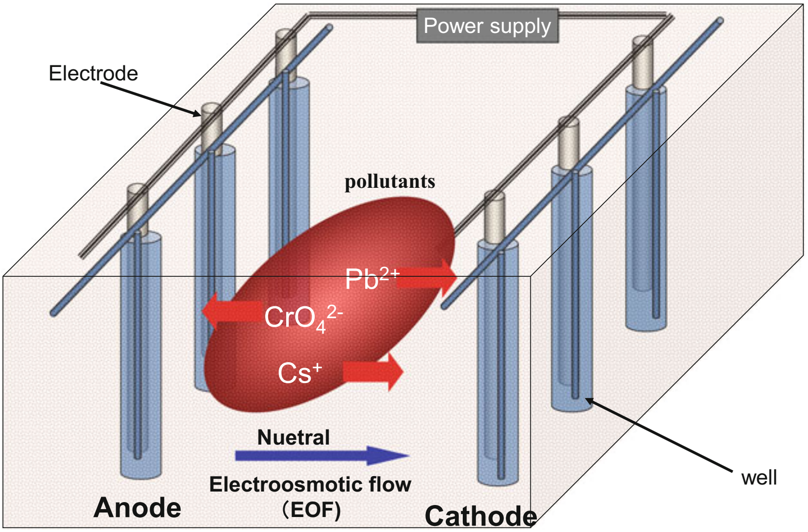


Fig. 13 Image of electro-kinetic remediation

are removed by electrophoretic migration. Even though a pollutant does not have any charges, the pollutant can be moved by the electroosmotic flow generated by applying the electricity between the electrodes inserted near the contaminated soil [27]. The details of the EK method are described in the later chapter “Electrokinetic Remediation”.

Phytoremediation is another separation method using some plants. Plants, which have the ability of the uptake of pollutants in leaves, stems, and roots, hyperaccumulator plants, can be used for removal of pollutants in contaminated soil. Phytoremediation is expected as a low-cost and easy method for treatment. Ideally, all we have to do is just to sow the plant seeds on the contaminated soil and to harvest the grown plants which have taken pollutants into some parts of the plant [28]. However, there are some limitations and disadvantages in phytoremediation. For example, the depth of soil to be able to be treated is limited by the length of the plant root. It usually takes a long time to treat and this method cannot be operated in winter season in the northern area. After harvesting, the management of the plant and some treatments are required to extract pollutants from the plant or store the plant somewhere. The details of phytoremediation are described in the later chapter “Phytoremediation”.

3.2.3 Decomposition

In the decomposition, a harmful substance is transformed to harmless one by various chemical reactions and/or microorganism process. Sometimes harmful organic substances can be completely decomposed into carbon dioxide and water but in most cases they are just transformed to less harmful compounds. In dechlorination, chlorines are liberated from the organic chlorinated compound to form a less or no-chlorinated compound. From the early years, combustion (incineration) has been used widely as the simplest decomposition method for harmful organic compounds. However, the combustion sometimes may transform a harmful compound to more harmful one like dioxins. Dioxins, which have very strong toxicity, are often generated as the by-products of the incineration of garbage and so on. The amounts of released dioxins depend on the condition of the incineration, the lower the temperature of combustion is, the larger amount of dioxins is formed. Recently, some innovated methods to decompose organic substances, using photo-catalyst [29], supercritical water [30], electrochemical method [31], ultra-sonic waves [32], and so on have been developed.

Zero valent iron (ZVI) has a high reactivity. ZVI reacts with oxygen and water to form reactive oxygen species. The reactive species dechlorinate reductively chlorinated hydrocarbon such as tetrachloroethylene and trichloroethylene to form less toxic compounds. The In-situ remediation of groundwater contaminated with TCE has been carried out by a permeable reactive barrier (PRB) containing ZVI. Nano particles of ZVI enable to remediate the groundwater and soil by the direct injection of nano-ZVI. ZVI has also a high ability to adsorb some heavy metal ions. Particularly, arsenic ions selectively adsorb on the hydroxide shell of iron around the core of ZVI [33, 34]. The details of ZVI are described in chapter "Zero Valent Iron and Some Other Nanometal Particles for Environmental Remediation".

Metal ions can be converted from highly harmful species to less harmful. As described previously, hexavalent chromium is a toxic and carcinogenic metal ion, but trivalent chromium is one of the essential metals. Therefore, the reduction of hexavalent chromium to trivalent chromium can be considered as one of the decomposition methods. In addition, when groundwater contaminated with high nitrate ions is continued to be used as drinking water, it will cause methemoglobinemia in human, especially in children. If nitrate ions can be converted into harmless nitrogen using a catalyst, the method using a catalyst can also be a decomposition method. The treatment of nitrate ion using the catalyst is described in detail in chapter "Heterogeneous Catalysts for Environmental Purification". Some chemical reactions are also used for the decomposition of pollutants, for example, ozone reaction [35], Fenton reaction [36], and zero valent iron (ZVI). The most promising method for decomposition is bioremediation method, whose detail is described in chapter "Bioremediation: From Key Enzymes to Practical Technologies".

4 Combination of Remediation Methods

The remediation methods based on each concept are often used as combining several methods, rather than the individual use. For example, when contamination is discovered somewhere, a containment process such as slurry wall is first employed to prevent the pollutant from further spreading. During the containment period, the

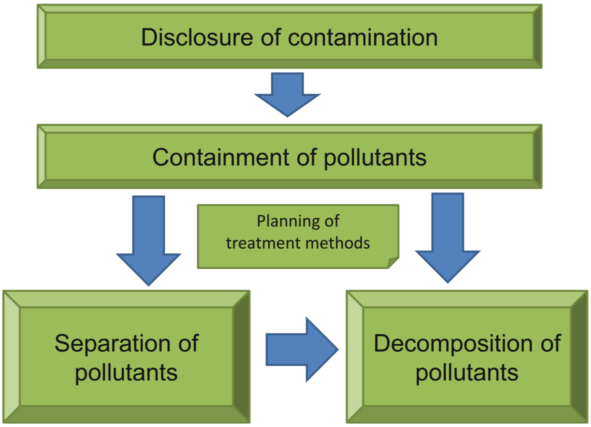


Fig. 14 Combination of remediation technologies

method to treat the pollutant is sought and planned, and then the pollutant is separated from there and/or decomposed to harmless compounds. The flow of these processes is shown in Fig. 14.

In the restoration of paintings and works of art, the reversible restoration is always required. That is, it is desirable that the repaired parts can be removed to retake the state before the restoration. That is because the restoration sometimes undermines the value of the original work of art. The environmental remediation should be also reversible if possible, especially in the remediation based on containment. Since the better remediation methods may be developed in the future, the performed process should be reversible as making the process removed if necessary.

5 Selection and Evaluation of Environmental Remediation Technology

The status of pollution varies from site to site. What should we consider when we choose a remediation technology from many technologies? The first thing to do is to get the accurate information about the pollution situation. What is a causative substance of the pollution, how high is the concentration of the substance at the contaminated site? And how does the possibility of spreading the substance to the environments exist? When the pollution occurs in soil, the rate of spreading the pollution may be very slow, but when the pollution happens in water or air, the pollutant will diffuse rapidly. Depending on the situation of the pollution, we should select a remediation technology which is best matted to the pollution. The toxicity of the pollutant is also an important factor in the selection of remediation technology. When the pollutant is very toxic like radionuclides, sufficient attention must be paid to all of the activities during the remediation process. In addition, the final residual concentration of the pollutants after the remediation needs to be considerably low so that it can ensure the safety. The geological or social geographical information of the contaminated site is also very important to choose the adequate remediation technology. The geological information around the contaminated site will give insights on the direction of pollutant diffusion, and the social geographical information is necessary to obtain the cooperation and agreement with the local people who live around the contaminated site during, before, and after the remediation process. Furthermore, the time required for remediation, the extent of remediation, and the cost of remediation are also important.

In order to evaluate various methods for soil remediation comprehensively, an assessment by scoring to the following criteria; (1) overall cost, (2) ability to clean to an acceptable level, (3) time to complete clean up, (4) reliability and maintenance, (5) data needs, (6) safety, and (7) community acceptability, has been proposed.

6 Design of Environmental Materials for Remediation

Many environmental remediation technologies have been currently developed and some of them are now available for actual contaminated sites. However, there are some specific cases of the pollution that cannot be treated only by the conventional remediation technologies. Therefore, it is necessary to design the novel methods and materials useful for the specific remediation. Many researchers have made much effort to develop newly designed methods and materials to be able to deal with the specific cases and some studies by them are introduced in this book. I had some experiences where I felt the necessity to design newly the methods and materials for the remediation. Some pollution problems that I have experienced are introduced in chapter “Pollution Sites Where Need Remediation” and our developments of the designed materials are described in some chapters, especially in chapters “Magnetic Separation of Pollutants for Environmental Remediation” and “Easily Collectable Floating-up Adsorbents to Remove Pollutants”. Here I would like to introduce one of my experiences that led us to our development of the new designed environmental materials. In November of 2005, an explosion accident occurred at a petrochemical plant in Jilin City of China, where is located in the Songhua River basin, and about 100 tons of benzene, aniline, and nitrobenzene in the plant flowed into the Songhua River. Among these pollutants, nitrobenzene has a specific gravity of about 1.20 heavier than that of water and is slightly soluble in water. Therefore, nitrobenzene was supposed to have flowed down along with the river water at the bottom of the river while dissolving in water to some extent as shown in Fig. 15. I and my colleagues conducted a survey of pollution of the Songhua River at this time with a researcher of Northeast Forestry University of China, as the detail of this pollution is described in chapter “Pollution Sites Where Need Remediation”. However, eventually I could not do anything in the remediation and the recovery of the pollutants in the Songhua River.

How to collect the oily pollutants which are flowing down with the river water at the bottom of the river? According to the UNEP report, the local government of China injected a large amount of activated carbon and plant materials as adsorbent in

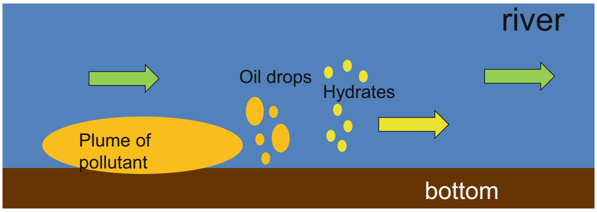


Fig. 15 Expected situation at the Songhua River: the plume of pollutant moves with the river water at the bottom and oily drops and hydrates of the pollutant also move along the river water

the vicinity of the gate for the water intake as the source of drinking water to recover pollutants. Unfortunately the effects of the adsorbents and the recovery amounts of pollutants by the adsorbents were not described in that report [2]. Activated carbon is not so expensive but plenty of activated carbon needs to recover the pollutants released into the river. Therefore, the adsorbent used under such situation should be low-cost and also environmental friendly materials. The enhancement in the function of activated carbon by modification is described in chapter “Preparation and Modification of Activated Carbon Surface and Functions for Environments” and the environmental friendly and low-cost adsorbents using paper sludge, some plants, and natural products are described in chapters “Environmentally Friendly Adsorbents” and “Coal Fly/Bottom Ash, Hydroxylapatite and Hydrotalcite”. The recovery of pollutant flowing down at the bottom of river is one of the special challenges that conventional treatment methods and materials cannot be applied to. It is essential to develop newly designed materials that can cope with such pollution situations. This specific situation gave authors a motivation to develop the adsorbents which can be collected easily after adsorption of pollutants on them. One of such adsorbents was a modified magnetite with hydrophobic compounds and it aimed to collect the adsorbent by a magnet after adsorbing oily materials like nitrobenzene in water. The magnetic separation of oily drops with modified magnetite was firstly developed by the researchers of Kawasaki Heavy Industries Ltd. [37]. Their study was also motivated by an accident of the oil spill from the Russian tanker, “Nakhodka,” near the coast of Shimane prefecture in the Japan Sea in 1997. In this accident, a large amount of fuel oil spilled from the ship and polluted a wide area of the coastline. The modified magnetite with hydrophobic compound can surround around oily drops of fuel oil and the oil droplets surrounded with the modified magnetite can be collected by a magnet. Our challenges were to clarify whether this technology could be applied to the collection of organic solvents such as nitrobenzene and chlorobenzene, furthermore, to expand this method for the collection of heavy metal ions and organic matters by introducing some adsorbing abilities into the modified magnetite. The image of easily collectable adsorbents by a magnet is shown in Fig. 16.

Another challenge is to develop the adsorbent that can float up on the surface of water after adsorbing pollutants. In the case of the Songhua river accident, a heavier pollutant than water such as nitrobenzene would flow down at the bottom of the river as described previously. In order to collect such pollutants with adsorbent, the adsorbent should once sink down at the bottom of the water to make contact with pollutants and stay there to adsorb the pollutants. Finally if the adsorbent could float

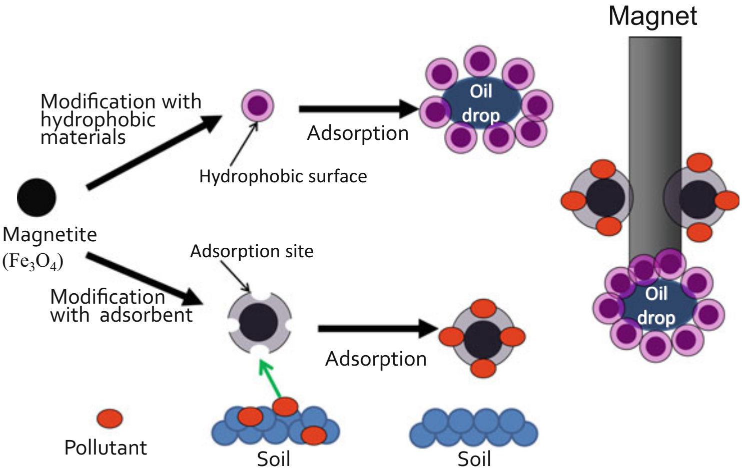


Fig. 16 Modified magnetite and magnetic separation

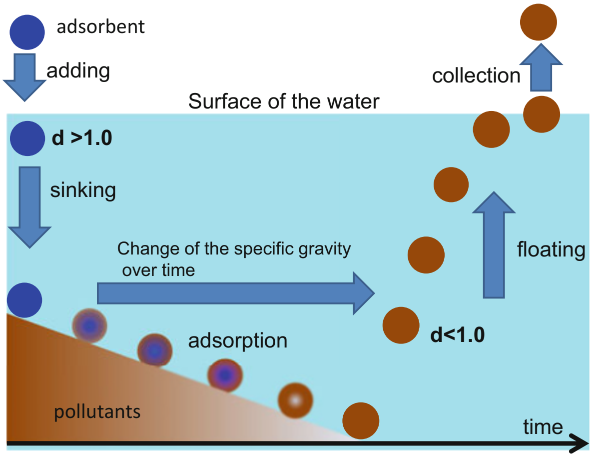


Fig. 17 Floating adsorbent and easy collection

up to the surface of the water, the adsorbent can then be collected relatively easily from the water surface as shown in Fig. 17. In order to develop such an adsorbent, we attempted to introduce a weight and a float to the adsorbent. When a heavier weight than the buoyancy due to a float is initially attached to the adsorbent, the adsorbent will sink down at the bottom of the water. However, if the weight gradually dissolves in water, this adsorbent would float up to the surface of the water after a certain time. In the process of the development of this type of adsorbents, we found out the very interested adsorbent which repeated the floating up to the surface and sinking down to the bottom a dozen and so times. This behavior is suitable for the collection of pollutant diffused in bulk solution. Lastly if the adsorbent could float up the water surface, we can collect the pollutant with the adsorbent easily. The details on this adsorbent are described in chapter “Easily Collectable Floating-up Adsorbents to Remove Pollutants”.

7 Contents of This Book

This book consists of five parts. Part I including two chapters is a background for environmental remediation and the introduction of some pollution problems. The necessity of environmental remediation and the classification of the technologies for remediation are dealt in chapter “Environmental pollution and remediation”. In this chapter, the necessity of the design of the environmental materials and methods for remediation is also described. Some case studies of recent pollution problems which authors have been concerned in are introduced in chapter “Pollution Sites Where Need Remediation”. In Part II including two chapters, the effects of pollutants on human health and ecosystem and the evaluation are described. The cases of heavy metals are described in chapter “Effects of Metals on Human Health and Ecosystem” and the cases of harmful organic substances in chapter “Effects of Persistent Organic Pollutants (POPs) in the Ecosystem and HUMAN health: Focusing on Chlorinated Chemicals”. Part III including four chapters shows some representative remediation technologies such as electro-kinetic remediation (chapter “Electrokinetic Remediation”), phytoremediation (chapter “Phytoremediation”), electrolysis for accumulation and decomposition of pollutants (chapter “Electrochemical Accumulation and Decomposition”) and bioremediation (chapter “Bioremediation: From Key Enzymes to Practical Technologies”) as relating to the design of the new functions and materials which can be applicable to these technologies. In Part IV including eight chapters, the developments of some designed environmental materials are introduced. The preparation and characterization of the developed materials are described and also the application or the possibility of the application is shown in each chapter. The developments of environmental friendly adsorbents are dealt in chapter “Environmentally friendly adsorbents”, and the studies on modified activated carbon (chapter “Preparation and Modification of Activated Carbon Surface and Functions for Environments”), hydro-char and bio-char (chapter “Hydrochar and biochar”), graphene (chapter “Graphene Oxide for Elimination of Dyes”), heterogeneous catalyst (chapter “Heterogeneous Catalysts for Environmental Purification”), adsorbents based on natural products (chapter “Coal fly/bottom ash, hydroxylapatite and hydrotalcite”), solidified DNA and cyclo-dextrin (chapter “Bio-Inspired Materials for Environmental Remediation”), zero valent iron (chapter “Zero Valent Iron and Some Other Nanometal Particles for Environmental Remediation”) are introduced. Finally in Part V including three chapters, easily collectable adsorbents and floating plants are described. The floating adsorbent can be collected easily from the surface of water. The adsorbents with magnetism can be collected easily by a magnet. The floating plants like algae, which can intake some pollutants, can be also collected easily from the water surface. The results of our studies on magnetic separation using the modified magnetite are shown in chapter "Magnetic Separation of Pollutants for Environmental Remediation". The floating adsorbents and plants are introduced in chapters "Easily Collectable Floating-up Adsorbents to Remove Pollutants" and "Remediation by Floating Plants", respectively.

8 Conclusion

An ideal world is one that does not need any remediation technologies. For that purpose, it is necessary that zero-emission technology has been established in every process and no pollutants are emitted into the environment. However, in reality, it will take a long time to develop the complete zero-emission technology. Even if such a technology would be developed, it will take plenty of time and cost to deploy it to every process. In the meantime, pollutants will continue to be discharged into the environment. Moreover, there are many environments around us that have already been polluted. Even if a zero-emission system is completed and deployed, a system failure may occur due to accidents or disasters, resulting in pollution. Therefore, the remediation technology is an essential technology at all time. The inexpensive and efficient remediation methods can replace an expensive preservation technology. However, the establishment of the complete remediation techniques does not mean that the release of pollutants into the environment is permitted if efficient remediation techniques are available. Until the complete zero-emission system will be established, we have to supplement it with remediation technology. As described previously, the remediation and preservation techniques are complementary. Therefore, it seems to be indispensable to develop the remediation technology and to design the useful materials for the remediation as well as the preservation technology.

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