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The Status of Soil and Groundwater Contamination in Japan and Case Studies of their Remediation

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ABSTRACT

Risk and exposure assessment for subsurface environment is very important for both aspects of health and environmental protection as well as making decision of remedial goal for engineering activities. Exposure due to hazardous chemicals in the subsurface environment is essential to assess risk level to individual person, especially from soil and groundwater environmental media. In this paper, the status of soil and groundwater contamination is presented to discuss on the problem for environmental risk assessment. The methodologies of fate and exposure models are also discussed by conducting the case studies of exposure assessment for heavy metals, organic compounds, and dioxin compounds. In addition, the structure of exposure models and available data for model calculation are examined to make clear more realistic exposure scenarios and the application to the practical environmental issues. Three kinds of advanced remediation techniques for soil and groundwater contamination are described in this paper. The most practical method for VOCs is the bio-remediation technique in which biological process due to consortium of microorganisms can be applied. For more effective remediation of soil contaminated by heavy metals we have adopted the soil flushing technique and clean-up system using electro-kinetic method. We have also developed the advanced techniques of geo-melting method for soil contaminated by DXNs and PCB compounds. These techniques are planed to introduce and to apply for a lot of contaminated sites in Japan.

KEYWORDS: soil contamination, risk assessment, exposure analysis, remediation

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1. Introduction

There are a lot of soil and groundwater contamination sites that have appeared in whole regions of Japan, especially in urban and industrial areas. This situation has changed dramatically in these years because the regulations of soil and groundwater criteria have prepared and put into operation by the government. The new law for soil environment protection in Japan will be established in 2002. In the new regulation, both environmental criteria of the leaching ability and the total content of hazardous chemical substances in contaminated soil are due to assess for human health, and also the methodology of risk assessment will be adopted to the case at the first time.

In this paper we present the state of subsurface contamination in Japan, and the new regulation of soil environment control. The methodology, modeling and application of risk and exposure assessment for soil and groundwater environment are also discussed. In addition, some practical cases of remediation techniques are introduced to be able to apply for contaminated sites in Japan.

2. Subsurface contamination in Japan

The number of soil and groundwater contamination sites tends to increase in Japan. (**Fig.1**) Most frequent contaminants are heavy metals and organic chloride compounds. The first regulation for groundwater quality was established in 1991 in Japan. After 1998 the number of contamination increases rapidly because of the reinforcement and the addition of contents in the regulation.

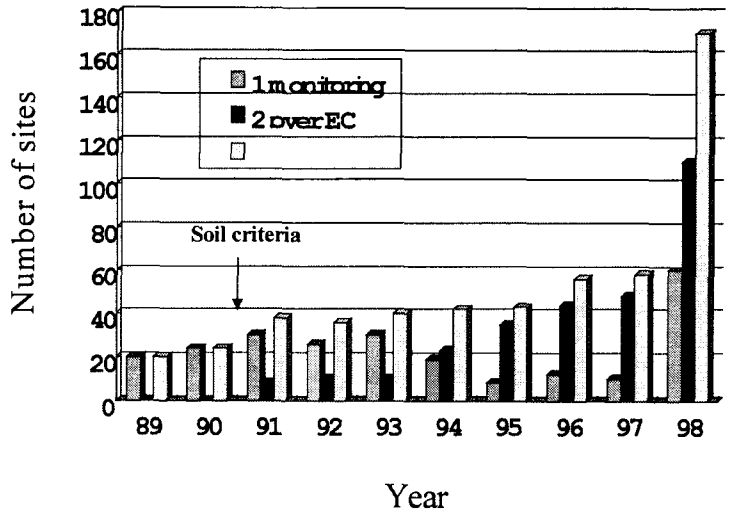

Fig.1 Soil and groundwater contamination in Japan.

The number of land trade and dealing cases has increased in these years. Due to the change in owner and user of lands, the contamination became tangible. The certification of ISO 14000 for monitoring the environment is a chance to disclose the contamination.

Based on the social background, Japanese Government has developed the new regulations of soil environmental criteria and law of environmental control in February 2003. Environmental criteria for groundwater and soil are listed in **Table 1** and **Table 2**. The total content of soil has newly regulated as a result of exposure assessment due to direct intake, dermal contact, and injection of heavy metals and other compounds. Both the leaching and total content criteria should be maintained to protect health and environment. In the assessment we assume the average direct intake of soil is 200mg/day and 100mh/day for child and adult, respectively. The methodology of exposure assessment is described in the next chapter.

Table 1 Environmental criteria for groundwater environment.

Heavy Metals	Organic compounds
Hg : 0.0005 mg/L	Benzene : 0.01 mg/L
Cd : 0.01 mg/L	TCE : 0.03 mg/L
Pb : 0.01 mg/L	PCE : 0.01 mg/L
As : 0.01 mg/L	DCE : 0.02 mg/L
Cr : 0.05 mg/L	cis-DCE : 0.04 mg/L
Se : 0.01 mg/L	DCM : 0.02 mg/L
Others	Others
CN : non-ditection	DXNs : investigating
F : 0.8 mg/L	PCB : non-detection
B : 1.0 mg/L	

Table 2 Environmental criteria for soil environment.

Heavy Metals Leaching	Heavy Metals Content
Hg : 0.0005 mg/L	Hg : 9 mg/kg
Cd : 0.01 mg/L	Cd : 150 mg/kg
Pb : 0.01 mg/L	Pb : 150 mg/kg
As : 0.01 mg/L	As : 150 mg/kg
Cr : 0.05 mg/L	Cr(6) : 250 mg/kg
Se : 0.01 mg/L	Se : 150 mg/kg
Others	Others
CN : non-ditection	CN : 50 mg/kg-free
F : 0.8 mg/L	F : 4000 mg/kg
B : 1.0 mg/L	B : 4000 mg/kg

There are some features in subsurface contamination, compared with atmospheric and hydrosphere environments. (**Table 3**) First of all, it is very difficult to make clear the in-situ condition of subsurface contamination. Site-specific features are also important to assess the environment. One of the difficulties of soil and groundwater investigation is that the land is privately owned in many cases. As for the survey and monitoring the contamination, there is very few existing data and documents and it is difficult to make a survey because of operating condition of industries. Since there is no official regulation for environmental assessment, it is necessary to develop the methodologies for exposure and risk assessment, regarding some

aspects of variability and uncertainty. For the more sophisticated approach, risk management system that includes risk/cost analysis, cost effectiveness analysis will be necessary.

Table 3 Features of subsurface contamination.

Social background Difficult to appear, Site specific, Property of land
Survey and monitoring Few monitoring data, Difficulty of detailed survey
Assessment methods and models No official methods, Uncertainty of parameters
Management Criteria, Restoration effect, Risk management

3. Risk assessment in subsurface environment

The procedure of risk analysis is very useful for the assessment of hazardous chemicals in soil and groundwater environment. The methodology and the procedure are called as 'Risk Assessment'. If there is a heavy contamination at a site but one does not have any exposure of chemicals from the site, the risk is regarded to be zero. This means no need of remediation and clean-up activity. However, there may be a possibility of the adverse effects from ecological aspects, such as the effects to plants, earthworm and microbiology. Thus, it is necessary to conduct a risk assessment for each site of contamination.

There is a complicated methodology in exposure and risk assessment for subsurface environment. (**Fig.2**) In the first step of procedure, we need to assign the exposure scenarios to the generic or specific case. The scenario consists of exposure route, exposure condition, and exposure factors. A lot of data and documents related to chemical substances, environmental media, and exposure group are also needed for the exposure assessment. In the second step, we will use three kinds of exposure model to determine the level and distribution of exposure to individual. The final step is the procedure of risk assessment, using both results of exposure and hazard assessments.

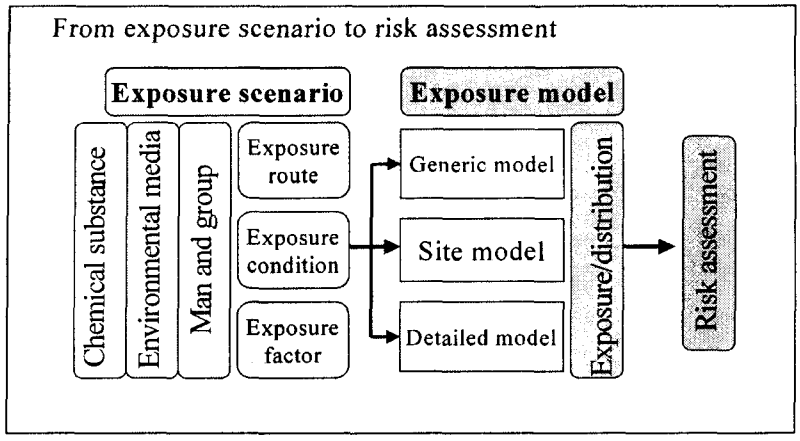

Fig.2 Methodology of exposure and risk assessment.

4. Exposure scenarios

The type of exposure scenario can be summarized into three categories based on the properties of chemical substances; (1) Heavy metals and related substances, (2) Organic chloride compounds and VOCs, (3) Dioxins, PCBs and POPs compounds.

The typical case of the exposure route for (1) heavy metals and related substances is shown in **Fig.3**. The most probable routes are intake of drinking water via groundwater and surface water, and intake from crops, dairy products and fishes. Direct exposure from soil, direct intake, dermal contact, and vapor inhalation, is a possible route for the assessment.

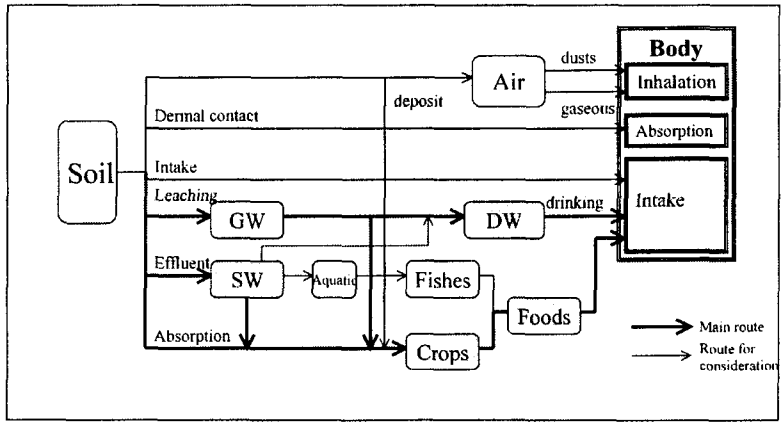

Fig. 3 Exposure from contaminated soil and groundwater (1) Heavy metals.

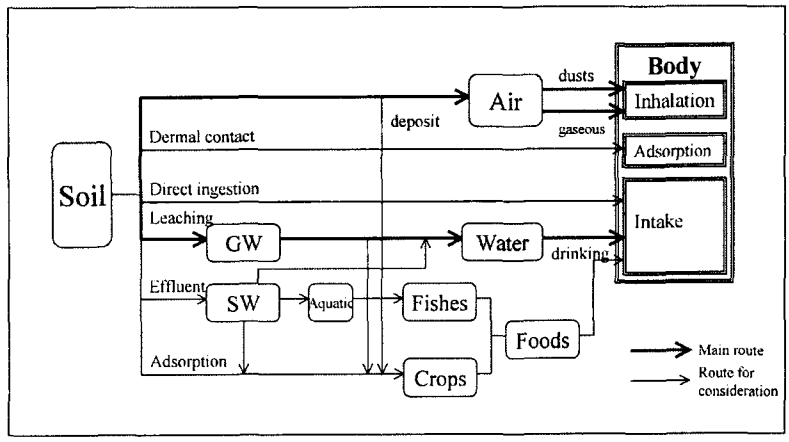

Fig.4 Exposure from contaminated soil and groundwater (1) COCs and VOCs.

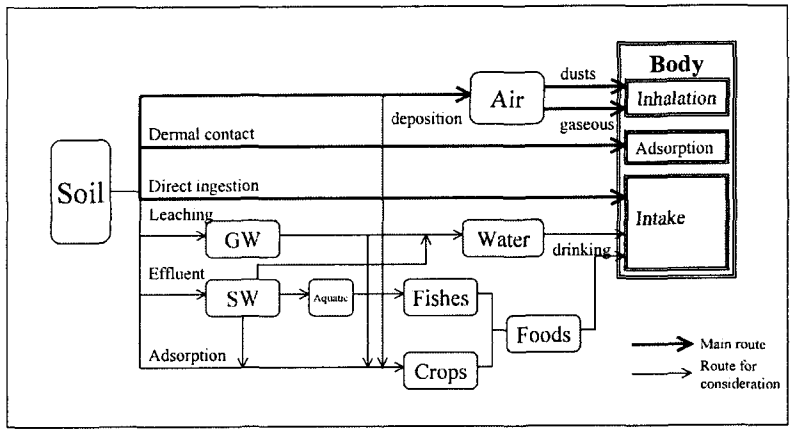

Fig.5:Exposure from contaminated soil and groundwater (3) DXNs and PCB.

On the contrary, the main routs for (2) Organic chloride compounds and VOCs are inhalation from air and intake from drinking water, especially from groundwater. (**Fig.4**) TCE, PCE, and other VOCs are very easy to migrate into subsoil and groundwater environment. (3) Dioxins, PCBs and POPs compounds have a lot of different exposure routes, because of the properties of substances. (**Fig.5**) These are typical persistent organic compounds, so that they migrate and transport through multimedia environment with very low degradation rate.

5. Exposure models for soil and groundwater

Modeling the amount of exposure in every routes and the behavior of chemical substances in subsurface environment is very important process for risk assessment. Some characteristics of soil and groundwater media should be considered in exposure models. There are major differences for subsurface

environment compared with other environmental media, such as heterogeneous properties, multiphase flow in porous media, and the enclosed environment. It is usually hard for chemical substances to be decomposed if they are emitted into subsurface environment because of no light and no oxygen situations. We should know a lot of parameters of environmental behaviors in subsurface environment. The most important parameters are those related to hydrology and geology.

Three types of exposure model have been proposed and developed for different purposes. We are developing each type of exposure model for more advanced exposure assessment and the application to Japanese situation. The screening type model in general condition is called as 'Generic Model'. The typical example of use of Generic model to the case of Dioxin Compounds has been considered. In this assessment we assumed a lot of exposure scenario and exposure routes for getting the total amount of exposure in general condition. Direct intake of soil and food chains are most probable exposure routes for this kind compound. Food chain via fishes, shellfishes and other kinds of diet are important to estimate exposure of DXNs and PCBs compounds. As a result of calculation, it was found that approximately 70 % of exposure is related to direct intake of contaminated soil. In 1999 Japanese Government established the environmental criteria of soil for DXNs compounds, 1000 pg-TEQ/dry g. The value was set on the basis of the exposure and risk assessment.

We can use site-specific type exposure models for the risk based exposure and risk assessment of soil and groundwater. (**Fig.6**) Risk Based Corrective Action (RBCA) system based on ASTM (USA) is very famous and widely used for site-specific risk assessment. A lot of parameter and data are necessary to complete the model calculation of this kind of model. Especially geological and hydrological parameters for the contaminated site are essential. In order to analyze exposure and risk level from soil and groundwater, we consider the chemicals of concern (COCs) for each exposure pathway and amount of exposure by using exposure factors and parameters. Finally we can assess the risk level for overall chemicals of concern by the toxicity data, such as hazard property or slope factor. Through the reverse process it is also possible to estimate the target or clean-up level of contaminated soil

from the acceptable risk level by RBCA system.

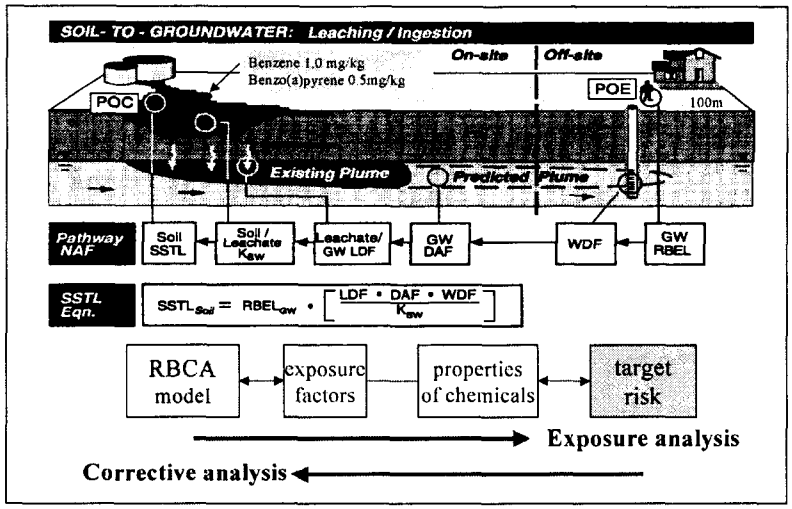

Fig.6 Methodology of site assessment by Risk Based Corrective Action(RBCA)

For the purpose of more detail and accurate exposure assessment, we can use the detailed numerical model (**Fig.7**) and the multimedia exposure model (**Fig.8**). We have developed both types of model in the Center for Chemical Risk Management, AIST, Japan. The detailed model is very useful to estimate the 3-dimensional distribution and time growth of hazardous chemicals in subsurface environment, so that it makes possible to determine the amount of exposure for more realistic environmental conditions, such as intake of drinking water from specified wells and surface water. The assessment costs very high because it may need a lot of data from geological and hydro-geological survey and prospecting, but the calculation will provide more reliable results for exposure assessment. Chemical and biological processes including sorption, degradation, and natural attenuation can be considered in this model calculation.

We are also developing the multimedia exposure model that consists of atmospheric, hydrosphere, subsurface, and ocean environments. For chemical substances widely distributed in the environment and took partitioning into multimedia environment due to the physical and chemical properties, the Multi-grid Multimedia exposure model should be very powerful for exposure assessment. Persistent organic chemicals (POPs), such as DXNs and PCBs

compounds are typical substances that are necessary to carry out multimedia exposure assessment.

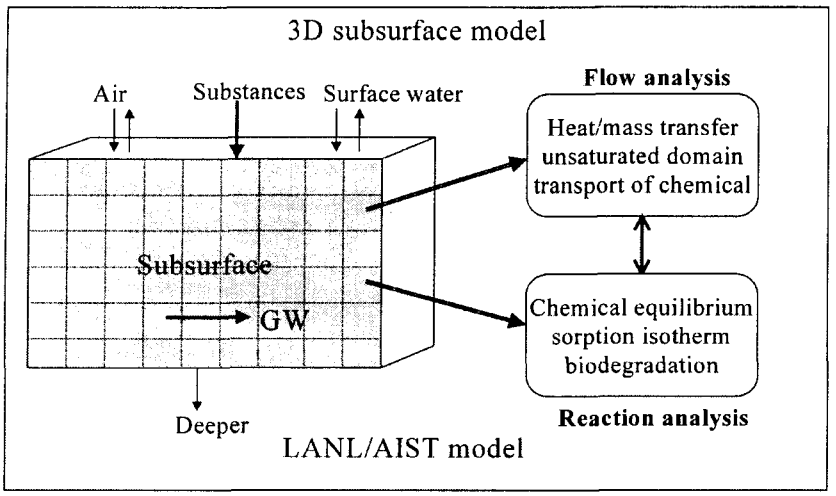

Fig.7 Structure of detailed numerical model for exposure assessment.

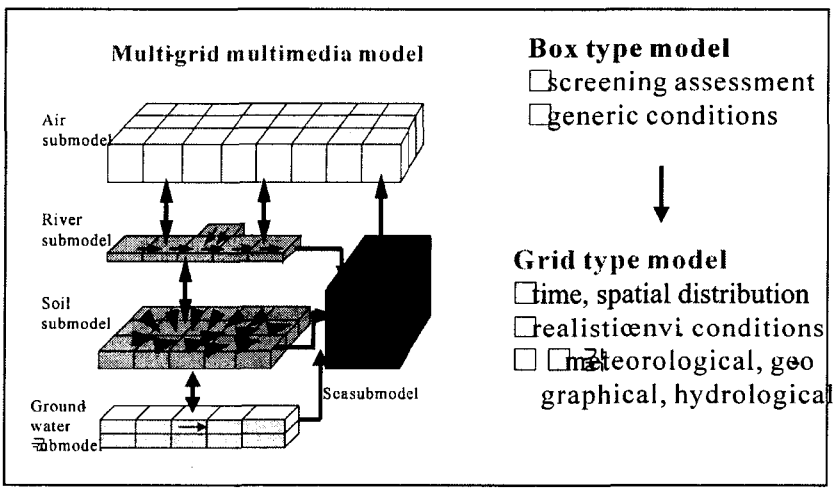

Fig.8 Structure of Multimedia model for exposure assessment.

6. Remediation techniques for soil contamination

There are a lot of remedial activities that have been applied for soil and groundwater contamination. Because the social demand of more effective and more economical ways of remediation, the advanced techniques have been developed and applied for many sites. The three remediation processes as listed in the following are very innovative and possible ways to the practical use.

(1) Bio-remediation method for VOCs; ***TCE, PCE, cis-DCE***

(2) Soil flushing and removal method for heavy metals; ***Pb, As, Hg, Cr***

(3) Soil consolidation and removal method for POPs; ***DXNs and PCBs***

These are all designed as risk reduction options based on exposure control from soil and groundwater environment. Biological method using microbiology is a way of environmental friendly remediation for both soil and groundwater environment. Soil flushing and removal method using electro-kinetic method and special kind of solution is very effective for soil contaminated by heavy metals. Soil consolidation and thermal decomposition by geo-melting method is very much innovative method for soil contaminated by DXNs and PCBs.

(1) Biological remediation technique using ***consortium of microorganisms***.

Bio-remediation is one of the effective clean-up methods for soil and groundwater contamination. We have proposed the advanced method of biological remediation technique, composed of (a) bio-reactor system using microorganisms consortium, (b) the high-pressure injection system into soil column and (c) soil remediation system. **Fig. 9** illustrates the biological remediation system, using consortium of microorganisms. This can be used for both soil and groundwater contamination due to VOCs, such as TCE and PCE. Biological materials and nutrients are added to promote the activity of microorganisms in groundwater. After the treatment of groundwater by bio-reactor system with microorganisms, the solution with chemical and biological functions is injected into soil to clean-up the compounds in situ condition. The treated groundwater can be recycled for the continuous process of bio-remediation. The concentration of TCE was reduced to less than the environmental criteria in a couple of days by using this system. This system has some advantages, very high cost-effectiveness, very small effect to the environment, and the application to operating factory and on-site.

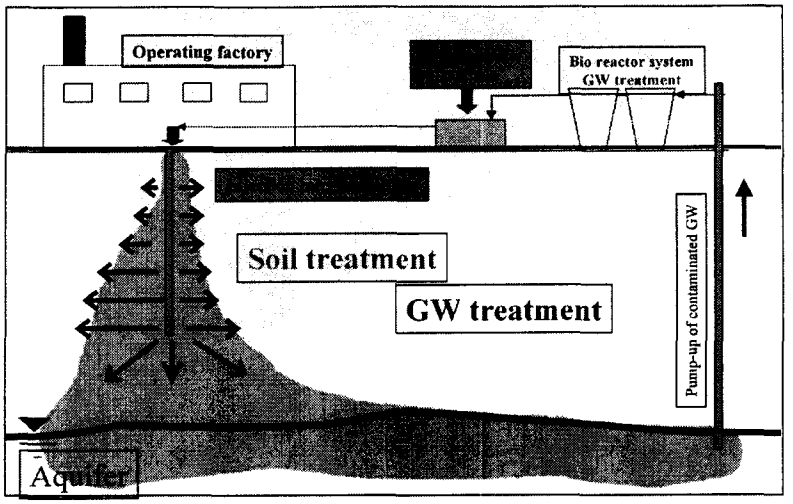

Fig.9 Biological remediation technique using consortium of microorganisms

(2) Soil flushing treatment technique using ***electro-kinetic method***.

It is very difficult to remove heavy metals from contaminated soil under in-site situation. The breakthrough for heavy metal treatment is soil flushing, control of groundwater and in situ treatment of soil. We have developed the coupled method of soil flushing and electrode system for the removal of heavy metals. **Fig.10** illustrates the soil flushing treatment technique using electro-kinetic method. Heavy metals such as Pb, As, Hg, and Cr can be leached out in the process of soil flushing by using functional solution. Citric acid and acetic acid are most effective solutions to promote leaching of Pb and As from contaminated soil. These contaminants can be removed by electrode system with electro-kinetic mechanism. The control of groundwater flow and with contaminant transport is very important for more safe and effective treatment.

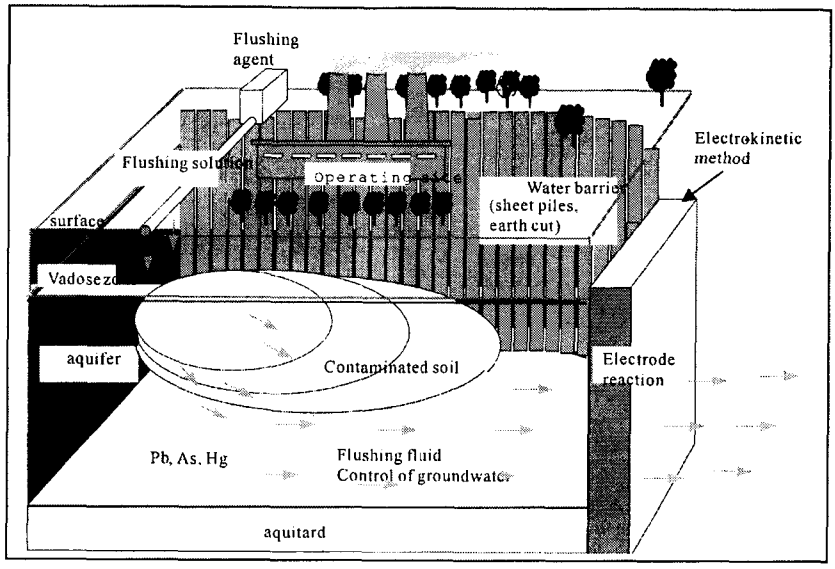

Fig.10 Soil flushing treatment technique using electro-kinetic method.

(3) Soil consolidation and treatment technique using ***geo-melting method.***

DXNs and PCBs are very toxic compounds emitted from industrial activities and waste treatment. To clean up soil contaminated by these compounds, it is necessary to remove them with more safe and efficient way. One of the most feasible remediation processes for DXNs is the Geo-melting method. **Fig. 11** illustrates the concept of geo-melting method newly developed by Konoike Construction Co. Ltd. of Japan. Geo-melting method has a couple of functions, consolidation of soil and decomposition of hazardous chemicals. Geo-melting process makes the situation of extreme high temperature, such as 1,600-2,000 C, so that DXNs can be decomposed to safer products. In addition the products can be consolidated or isolated into glass structure formed as high temperature reaction. The decomposed gas products can be trapped and removed by gas collector system. This technique has been applied to the contaminated waste disposal site in Wakayama Pref. in Japan. In the site, there was more than 2,000m^3 of waste material and soil contained DXNs with a concentration of 100ng-TEQ/dry.g, which was 1,000 times more than the environmental criteria of soil. In the case of 100m^3 treatment of contaminated soil, geo-melting process took about 7 days for complete decomposition of DXNs. The concentration of soil could be reduced to less than 0.01ng-TEQ/dry.g.

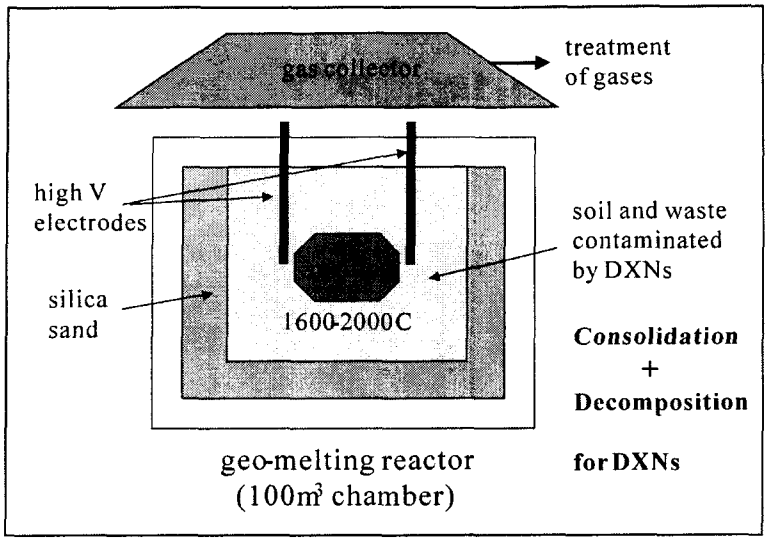

Fig. 11 Geo-melting remediation technique for contaminated soil with DXNs.

7. Conclusions

It is very important to assess exposure and risk caused by contaminated soil and groundwater. Risk based assessment makes it possible to realize the quantitative analysis of environmental risk for health and ecology as well as the cost-effectiveness analysis and socio-economical analysis.

Three types of exposure model for soil and groundwater contamination were summarized and discussed, especially on the application to the practical cases. It is necessary to develop exposure models in accordance with the categories and properties of chemicals, exposure scenario, and the methodology of risk assessment. The research of the exposure and risk assessment in subsurface environment has just started, so that it is needed to compile a lot of factors and parameters for the assessment.

It is very important to develop and apply the remediation techniques for soil and groundwater contamination. Three advanced remediation methods that have been developed in these years were introduced in this paper. These techniques can be widely applied to the practical contaminated sites with various contaminants, such as VOCs, heavy metals, and DXNs compounds.

Risk assessment is an essential element of corrective actions for soil and groundwater contamination, when investigating the problems of risk benefit, cost effectiveness, and information disclosure. Especially for VOCs contaminated sites, such as gas stations and oil spill sites, risk assessment will provide reasonable solution for protecting the environment and for

cost-effective remediation.

The following research works are necessary;

(1) Practical case studies of exposure and risk assessment.

(2) Analysis of remediation process and risk reduction.

(3) Development of more effective remediation techniques.

Acknowledgements

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