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PRELIMINARY INVESTIGATION ON DETERMINATION OF RADIONUCLIDE DISTRIBUTION IN FIELD TRACING TEST SITE

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Preliminary Investigation on Determination of Radionuclide Distribution in Field Tracing Test Site

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Field tracing tests for radionuclide migration have been conducted by using ^{3}H, ^{60}Co, ^{85}Sr and ^{134}Cs, in the natural unsaturated loess zone at field test site of China Institute for Radiation Protection. It is necessary to obtain confidable distribution data of the radionuclides in the test site, in order to evaluate exactly the migration behavior of the radionuclides in situ. An available method to determine the distribution was proposed on the basis of preliminary discussing results on sampling method of soils from the test site and analytical method of radioactivity in the soils.

Keywords: Field Test, Radionuclide Distribution, Migration, ^{3}H, ^{60}Co, ^{85}Sr, ^{134}Cs, Loess Zone, Sampling Method, Analytical Method.

The study was carried out under a cooperative research between Japan Atomic Energy Research Institute and China Institute for Radiation Protection.

* China Institute for Radiation Protection

** Socio-Energy System Co., Ltd.

核種移行野外試験場における放射性核種分布の測定に関する予備的検討

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李　書紳*・神山　秀雄**

（1993年11月9日受理）

中国輻射防護研究院野外試験場の黄土層において、^{3}H、^{60}Co, ^{85}Sr 及び ^{134}Cs を用いた放射性核種野外移行試験を実施してきた。原位置における放射性核種の移行挙動を正確に評価するためには、精度の高い放射性核種の分布データを得ることが必要である。野外試験場からの試料の採取方法並びに採取した試料の分析方法について検討し、分布データを得るための適切な方法を提案した。

本研究は、日本原子力研究所と中国輻射防護研究院との共同研究により実施された。

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1.INTRODUCTION

Understanding the processes of radionuclide migration in hydrogeologic environments is important for the storage or disposal of radioactive waste. In the event that the engineered structures deteriorate, radionuclides be released to local environment. On the subsurface disposal of radioactive waste, the radionuclide migration through saturated and unsaturated environments is an important path way.

In situ field tracing tests for examining radionuclide migration have been conducted in the natural unsaturated loess zone at field test site of China Institute for Radiation Protection, to prove the theory of radionuclide migration, to obtain parameters used for calculation models, and to collect useful data for making a monitoring program. In the *in situ* test, ^{60}Co, ^{85}Sr and ^{134}Cs have been used as radioactive tracers because ^{60}Co, ^{90}Sr and ^{137}Cs are the major components of radioactive waste from nuclear power plants. Also ^{3}H has been used to observe water permeation behavior at the test site.

It is necessary to obtain confident distribution data of the radionuclides during test period, in order to evaluate exactly migration behavior of the radionuclides in the *in situ* test. For this purpose, sampling method of soils from the test pit and analytical method of the radionuclides the soils have been preliminarily discussed.

2.SAMPLING METHOD

2.1 Sampling Interval and Sampling Position

Radionuclide migration tests were designed under both natural and artificial rainfall conditions in test pits A, B, D and E. **Figure 1** shows arrangement of the test pits. Main parameters are summarized in **Table 1.**

In order to evaluate the ^{60}Co, ^{85}Sr, ^{134}Cs and ^{3}H migration in the test pits, two kinds of radionuclide distribution data are required as main information from the test result; one is to observe the change of the radionuclide distribution in the vertical direction during 2 years of test period, and another is to estimate expansivity of the radionuclide distribution in the lateral direction$^{(1)(2)}$.

1) Migrate toward the vertical direction

To measure the change of the radionuclide distribution with lapse of test period,

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1) Migrate toward the vertical direction

To measure the change of the radionuclide distribution with lapse of test period, vertical core sampling was carried out every quarter year in half area of each pit. Sampling position is shown in Fig. 1.

Disturbance of sampling may affect the migratory behavior of the radionuclides and water. To avoid it as possible as one can, following sampling methods were considered.

(a) Top of inserted sampler is covered by cap and the sampler is not taken out until test end.
(b) After pulling out sampler, sampling hole is filled with loess.
(c) After pulling out sampler, another dummy sampler is inserted into the same hole.

In the case of (a), nothing of test results will be obtained until finishing all sampling. As it is difficult to fill to the same condition before sampling, any difference of geological condition may affect on the surrounding soil layer, in the case of (b). Therefore, the case (c) was applied to the project.

2) Expansive toward the lateral direction

Some sampling methods in the left halves of the test pits were proposed and discussed, in order to estimate uniformity of radionuclide distribution in the pits and migration in the lateral direction.

(a) Open−cut sampling:

According to the first proposed method "open−cut sampling", after last sampling the vertical soil core, each pit partially is dug as shown **Fig.2** which presents overlooking and sectional drawings of design for open−cut sampling. Some soil cores are taken toward lateral direction from shaved sampling profiles (1) and (2).

(b) Only vertical sampling:

The second proposed method shows that some vertical sampling procedures are also performed in the left halves of the test pits. In addition, to observe expansivity of the radionuclides, soil cores are vertically taken outside pit, as shown in **Fig.3**. The more core samples are taken, the results are more exact. But analytical samples are also produced in large quantities.

Finally, on the basis of discussion above, five vertical soil core samples were collected respectively from left half sides of pits A, B, D and E, as seen in **Fig.4**.

Figure 5 shows the burial condition of contaminated soil in pits A, B, D and E. In pits A and D the burial depth was 30 cm, in pits B and E it was 100 cm. The interval between the edge of active area and sampling positions No.11 and No.15 was kept at 4 cm, since a steel angle (ca. 30 mm in width) was laid around contaminated soil, as shown in Fig.5.

2.2 Core Sampling Procedure

To take vertical soil core samples from pits A, B, D and E, the sampling tube consisted of stainless steal was inserted into each sampling point, and then it was pulled out with containing soil core(2).

Figure 6 shows insertion method of sampling tubes. The core sample was taken by 2 m in maximum length. Insertion procedure was divided into two processes to evade compression of soil core. As to pits A and D, after a core sample including contaminated layer was taken with a tube of 120 cm at first, a tube of 80 cm was inserted into the deeper layer. On the other hand, as to pits B and E, a tube of 120 cm was inserted after taking a core sample of 80 cm in length.

3.CUTTING METHOD

Radioactivities at some points of the core sample were measured directly and nondestructively from outside tube by scanning with NaI type gamma-ray detector to know roughly radioactivity distribution in the core before cutting.

The core sample was cut into sections of every 5 mm thick in high activity region and every 10 mm thick in desired low activity region(2).

Each soil sample cut was put into a sample box of 75 mm in diameter and 25 mm in height. The sample box was sealed by transparent adhesive tape to avoid evaporation of moisture and dispersion of soil particle. The relative depth in the sampling pit and the weight of each soil sample were recorded at the top of sample box.

During the sampling, cross contamination may rise in insertion procedure of sampling tubes (show **Fig.7**) and in cutting procedure of core samples. In the case of cutting, if one soil particle of the higher region is fallen into the lower region sample, the radioactivity of the lower region sample is significantly affected by it. Because the radioactivity level is several orders different between the lower region and the higher one.

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Therefore, the order of cutting the samples was from lower radioactivity region to higher one, as shown in **Fig.8**.

4.DETERMINING METHOD OF RADIOACTIVITY

4.1 Analysis of Gamma-nuclide

Gamma-nuclide concentration of the soil samples was measured by gamma-ray spectrum analyzer (pure Ge semiconductor type, CANBERRA Co. Ltd.). To determinate exactly the gamma-nuclide concentration, it is necessary to discuss the preparation procedure of analytical sample and influence factors on error(3)-(6).

1) Preparation

Two methods were proposed as preparation procedures of the soil samples for gamma-nuclide analysis(2).

(a) Acid extraction method:

At first, the soil sample is separated into half and half, and stored in two glass containers. One of the half sample is used for gamma-nuclide analysis, the other is used for ^{3}H analysis. The sample for gamma-nuclide analysis is dried in a oven after it is weighed. The moisture content of sample is determined from weight difference between before and after drying. The desired weight of dried sample is transferred into a gamma-ray counting container and it is contacted with 50 ml of 0.1 N HCl in the container to extract the radionuclide species.

(b) Direct counting method:

The sample boxes, in which soil samples cut are packed, are used as gamma-ray counting container. Gamma-nuclide concentration of the soil samples is measured by placing directly the sample box on the detector of gamma-ray spectrum analyzer.

By applying the acid extraction method, the radionuclides are diffused into liquid phase so that localization of radionuclide in the container is able to avoid. In drying process, however, a great deal of tritiated steam is produced from many samples. In addition, this method in which a portion of the sample is used will give lower counting rate, compared with the direct counting method in which all amount of the sample is used.

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It was concluded, therefore, that the direct counting method is more suitable.

2) Correction for soil weight

The specific radionuclide concentration is calculated from the radioactivity and the weight of soil sample in the sample box, but the soil sample includes a moisture in the case of direct counting method. It is necessary to correct a soil sample weight, by using moisture weight obtained experimentally on ^{3}H concentration measurement which is described hereafter, or by using that determined from weight difference by drying soil sample in a oven, or by using that measured directly with neutron moisture meter in the test pits.

3) Calibration and error(7)

In the case of direct counting method, the counting efficiency of the gamma-ray detector was calibrated by using cylindrical soil sources with diameter of 75 mm and thickness of 5 mm, 10 mm, 20 mm.

Table 2 lists the soil sources used in calibration of counting efficiency. Total error of measurement results of soil samples are contributed by factors as follows:

E1: Error of peak area, affected by activity in samples and measurement time.
E2: Error affected by background.
E3: Error of calibration efficiency.
E4: Error in calibration sources.
E5: Error affected by weight and water content in samples.
E6: Error affected by geometry of soil in sample boxes.

Contribution of each error to the total error is listed in **Table 3**.

4.2 Analysis of Tritiated Water

To determine accurately tritiated water concentration in low moisture soil sample, a suitable ^{3}H analytical method must be established. We discussed following two analytical methods(8).

(a) Direct distillation method:

A apparatus to separate water from soil is shown in **Fig.9**. The apparatus is the vacuum-heating distillation system in which pressure are kept as low as −600 mmHg by a vacuum pump.

About 5 g of soil sample placed in a heating tube is kept at 105 ℃ in a oven. All moisture from the soil sample is completely collected in a condenser placed inside a cold trap after distillation. Then 10 ml of Insta-gel cocktail is filled in the condenser. The moisture is rinsed out from the condenser wall with cocktail carefully and the mixture is transferred into a counting vial for radioactivity measurement with Packard 2250 CA liquid Scintillation Counter (LSC).

The ^{3}H concentration per soil weight unit or water unit, C_{SH1} or C_{WH1}, in the soil sample can be calculated from the equations:

$$C_{SH1} = A/MR \qquad (Bq/g)$$
$$C_{WH1} = A/(M_0 - M)R \qquad (Bq/ml)$$

where, A: radioactivity measured with LSC (Bq), M_0: weight of soil sample before distillation (g), M: weight of soil sample after distillation (g), R: recovery coefficient of the analysis.

Moisture weight in soil sample can be obtained from weight difference between before and after the distillation.

(b) Extraction and distillation method

After about 2 ml of deionized water is added to 5 g of soil sample to extract and distill easily ^{3}H retained on soil, water is separated from the soil sample by the same distillation as shown in method (a). One ml of the distilled water collected in the condenser is sampled by pipetting and then transferred into a counting vial. The 10 ml of Insta-gel cocktail is mixed with the distilled water in the counting vial and radioactivity was measured with LSC.

The ^{3}H concentration, C_{SH2} or C_{SH2}, in the soil sample can be accurately calculated without uncertain coefficient R:

$$C_{SH2} = A\{W + (M_0 - M)\}/M \qquad (Bq/g)$$
$$C_{WH2} = A\{W + (M_0 - M)\}/(M_0 - M) \qquad (Bq/ml)$$

where, W is amount of deionized water added.

The actual measurement results of recovery coefficient under the direct distillation method are listed in **Table 4**. The ^{3}H recovery coefficient was about 0.9 and rather satisfactory consistency was obtained. The ^{3}H concentration can be confidently determined by making a correction using the recovery coefficient. However, the direct distillation method includes uncertain coefficient R which has 2 % of error and depends on the skill of operators.

On the other hand, the actual results studied for reproducibility of measurement values under the extraction and distillation method are listed in **Table 5**. It is found that the measurement values present within the distribution interval of 1 % to the average (recovery tendency: dpm measured/dpm added = 1) and are rather satisfactorily obtained for 6 soil samples. This results prove that the extract and distillation method can determine more accurate ^{3}H concentration than the direct distillation method.

However, this method may not be useful for extremely low ^{3}H concentration sample, because the detection limit of ^{3}H concentration is reduced by diluting with deionized water and decreasing counting efficiency with increase of quenching. In addition, much more time is required for the distillation. It is concluded, therefore, that the direct distillation method is more suitable for ^{3}H measurement on this project, because of having high recovery coefficient and reproducibility.

5.CONCLUSION

To evaluate exactly the radionuclide migration behavior on the *in situ* field tracing tests, determination methods of the radionuclide distribution in the test pits were preliminarily investigated.

The sampling of several core soils in vertical direction was performed to obtain the information relating to uniformity and expansivity in both the vertical and the lateral direction of the radionuclide distribution in the pits.

Since the cross contamination produced in sampling and cutting procedure of core samples was important problem, the effective method to avoid the cross contamination was proposed.

On the analysis of gamma-nuclide, influences of error were indicated. On the analysis of tritiated water, soil-water separation technique by the direct distillation method was suitable for ^{3}H measurement, because of having high recovery coefficient and reproducibility.

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Table 1 Main parameters of field test.

Parameter	Test pit			
	A	B	D	E
Rainfall condition	Natural	Natural	Artificial	Artificial
Pit size (cm)	200×200	200×200	200×200	200×200
Pit depth (cm)	30	100	30	100
Tracer layer size (cm)	150×150	150×150	150×150	150×150
Tracer layer thckness (cm)	0.7	0.7	0.7	0.7
Test period (year)	2	2	2	2

Table 2 Soil sources used in calibration.

Source No.	Weight (g)	Height (mm)	Nuclide	Activity (Bq)
1	25.65	5	Eu-152	12.07
			Cs-137	46.34
			Co-60	44.32
2	42.85	10	U-238	23.57
			Ra-226	22.70
3	44.15	10	Eu-152	20.76
			Cs-137	79.77
			Co-60	76.29
4	79.15	20	U-238	43.53
			Ra-226	41.91
5	76.85	20	Eu-152	36.14
			Cs-137	138.8
			Co-60	132.8

Table 3 Error analysis of measurement results.

Activity (Bq)	E1 (%)	E2 (%)	E3 (%)	E4 (%)	E5 (%)	E6 (%)	Total (%)
10^{-2}	30	15	16	5	10	8	40
10^{-1}	10	10	16	5	10	8	25
10^{0}	5	5	16	5	10	8	22
10^{1}	1	1	16	5	10	8	21
10^{2}	0.5	-	16	5	10	8	21
10^{3}	-	-	16	5	10	8	21
10^{4}	-	-	16	5	5	3	18

Total: =$\sqrt{\Sigma(E_i{}^2)}$, i=1, 2....6

Table 4 Measurement results of recovery coefficient.

Run No.	^{3}H added (dpm)	^{3}H counted (dpm)	^{3}H recovery
1	6404.7	5937.22	0.927
2	6404.7	5750.26	0.898
3	6404.7	5886.13	0.919
4	6404.7	5548.60	0.866
5	6404.7	5611.83	0.876
6	6404.7	5772.14	0.901
7	6404.7	5929.43	0.926
8	6404.7	5788.48	0.904
9	6404.7	5709.09	0.891
10	6404.7	5617.95	0.877
11	6404.7	5797.86	0.905
12	6404.7	5762.45	0.900
Average			0.899±2%

Table 5 Measurement results of reproducibility.

Run No.	^{3}H added (dpm)	^{3}H measured (dpm)	^{3}H measured/^{3}H added
1	6404.7	6392.75	0.998
2	6404.7	6450.26	1.007
3	6404.7	6394.11	0.998
4	6404.7	6320.00	0.987
5	6404.7	6328.45	0.998
6	6404.7	6356.32	0.992
Average		6373.65	0.995±1%

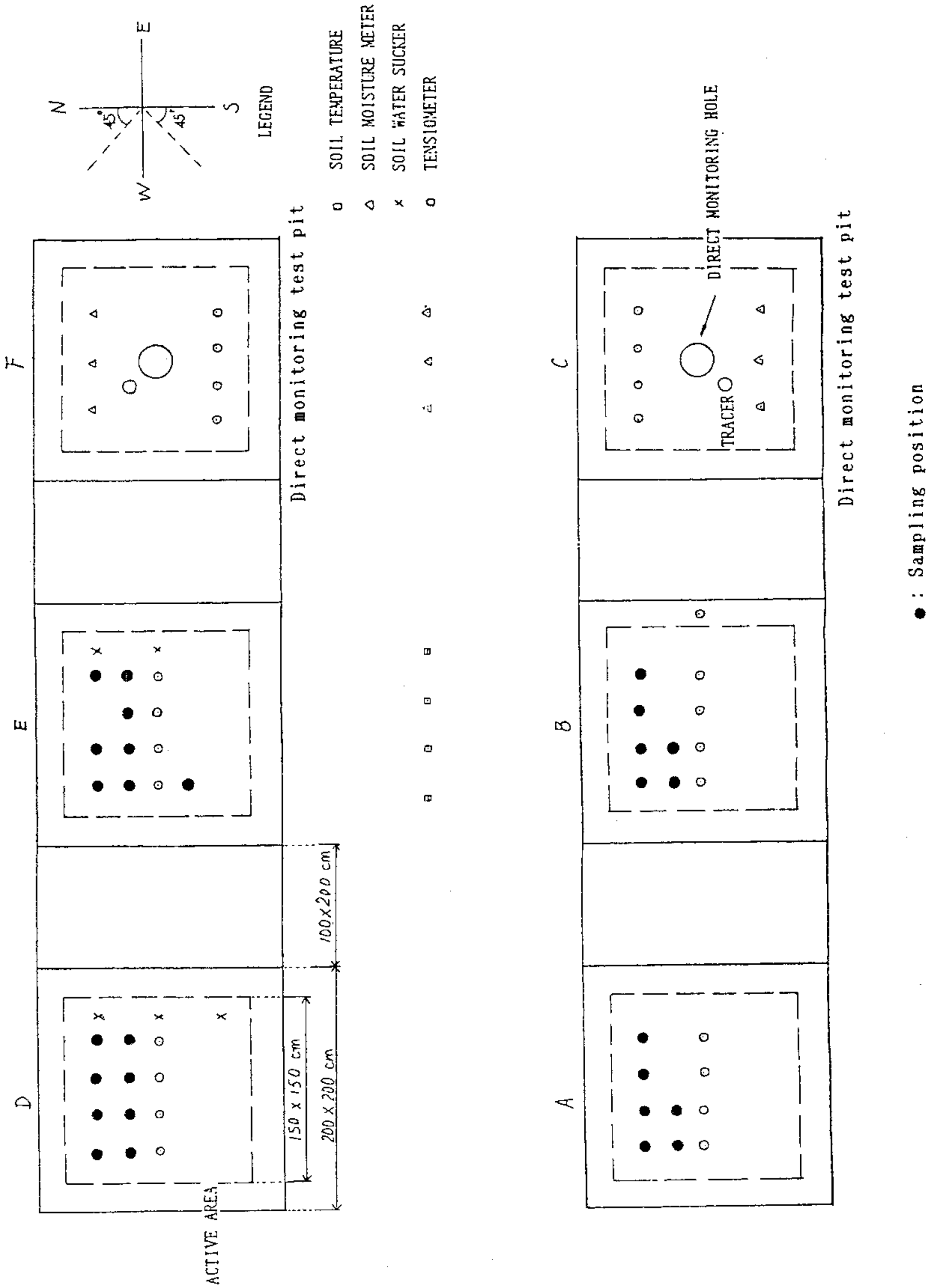
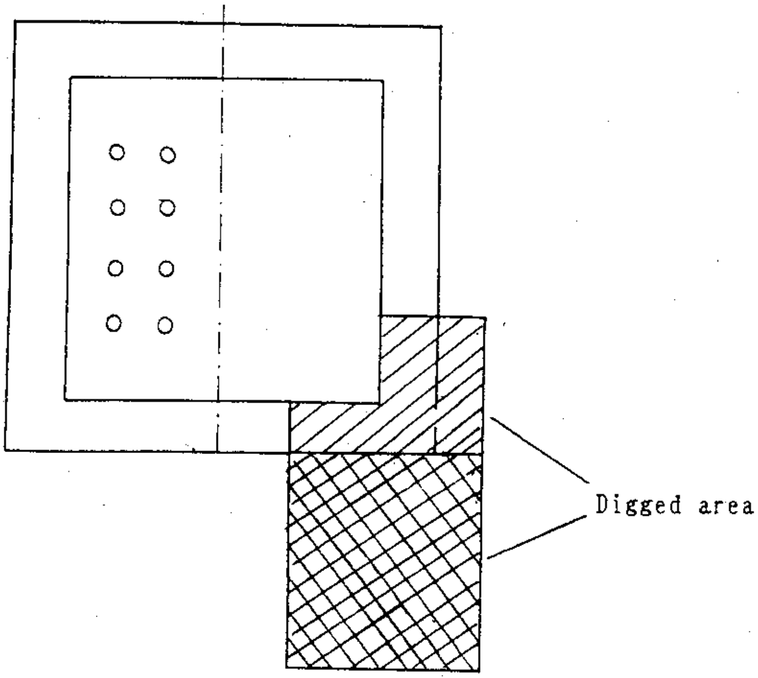
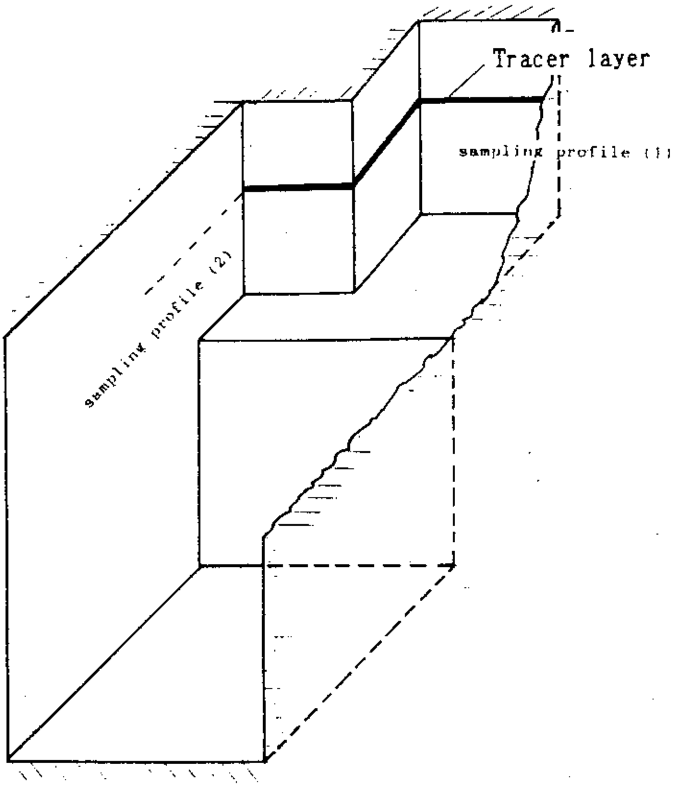


Fig.1 Arrangement of test pit



(a) Overlook of design for open-cut sampling



(b) Section of design for open-cut sampling

Fig.2 Open-cut sampling

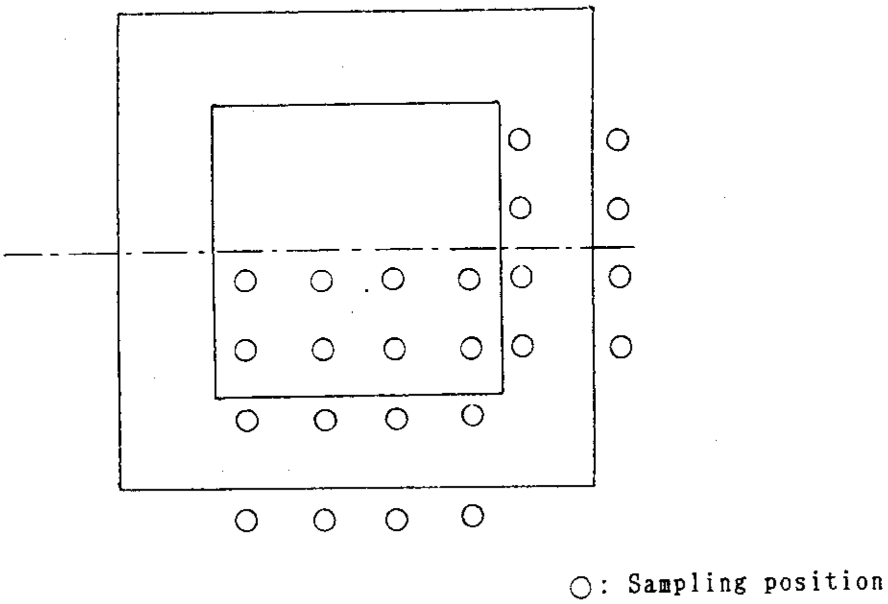


Fig.3 Vertical sampling

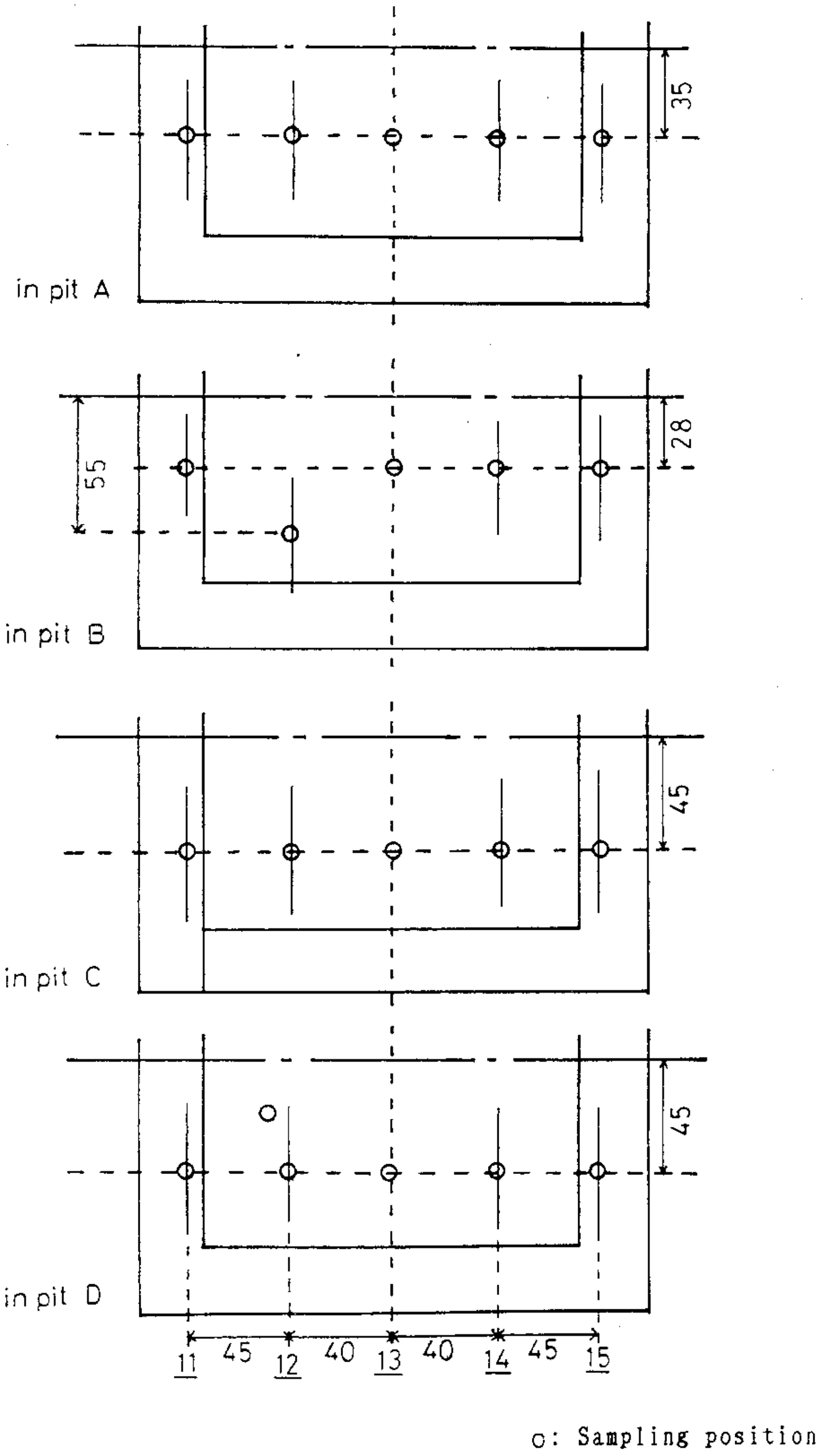


Fig.4 Sampling position in left half sides

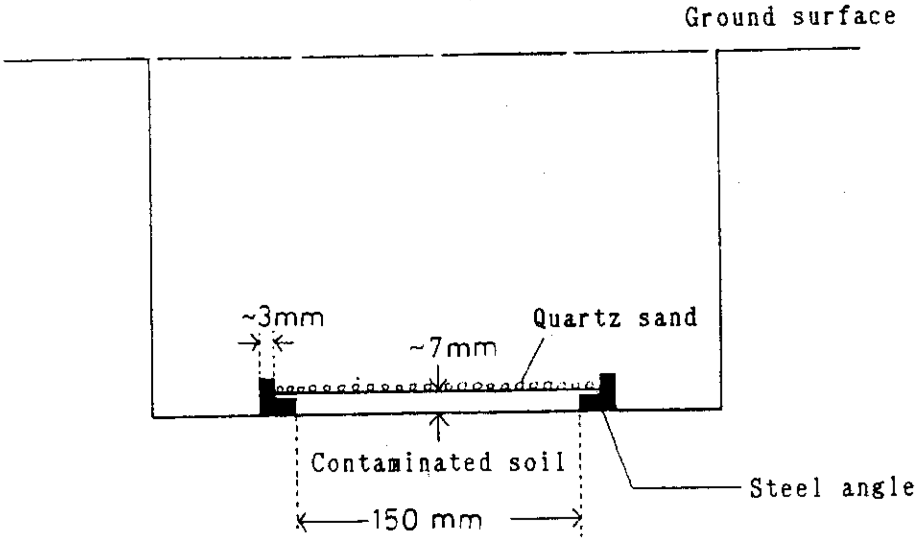


Fig.5 Burial condition of contaminated soil

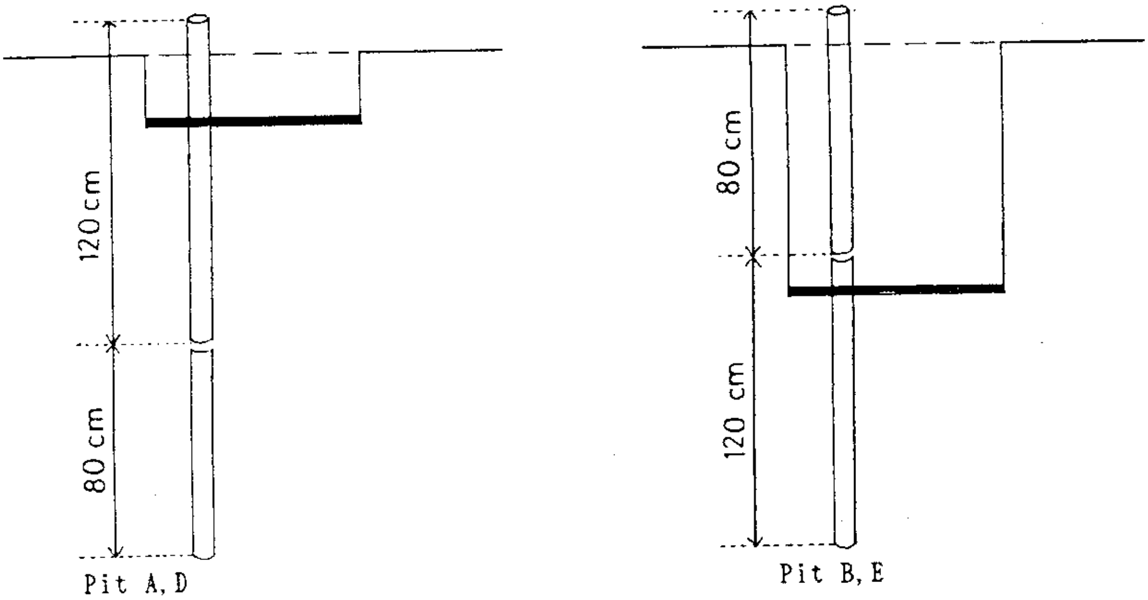


Fig.6 Insertion method of sampling tubes

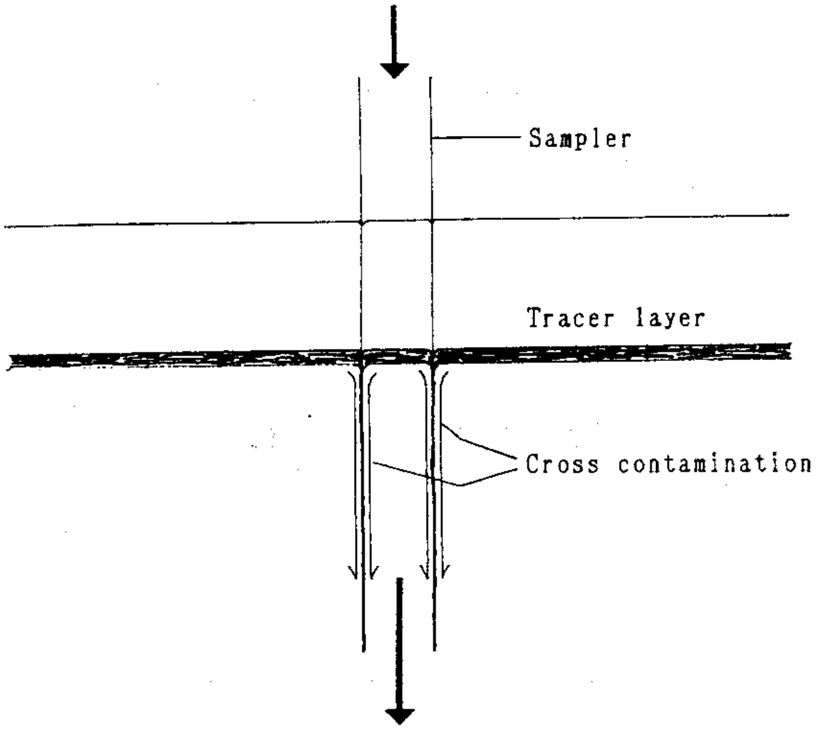


Fig.7 Cross contamination under collecting core samples

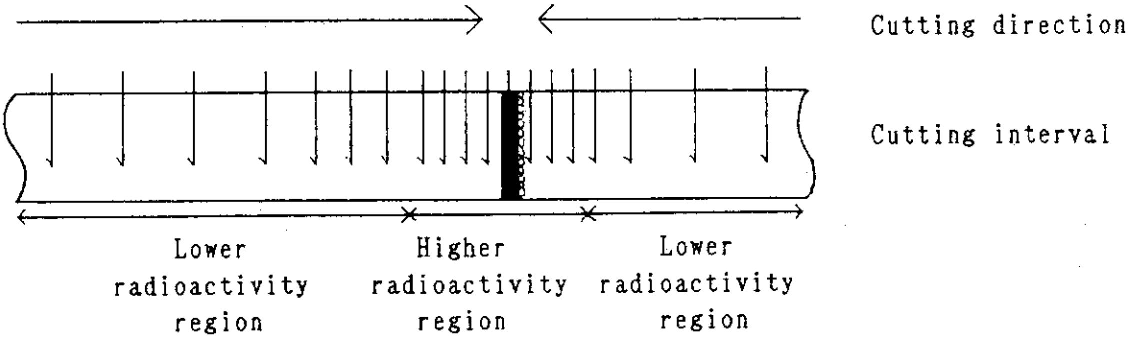


Fig.8 Cutting method of core samples

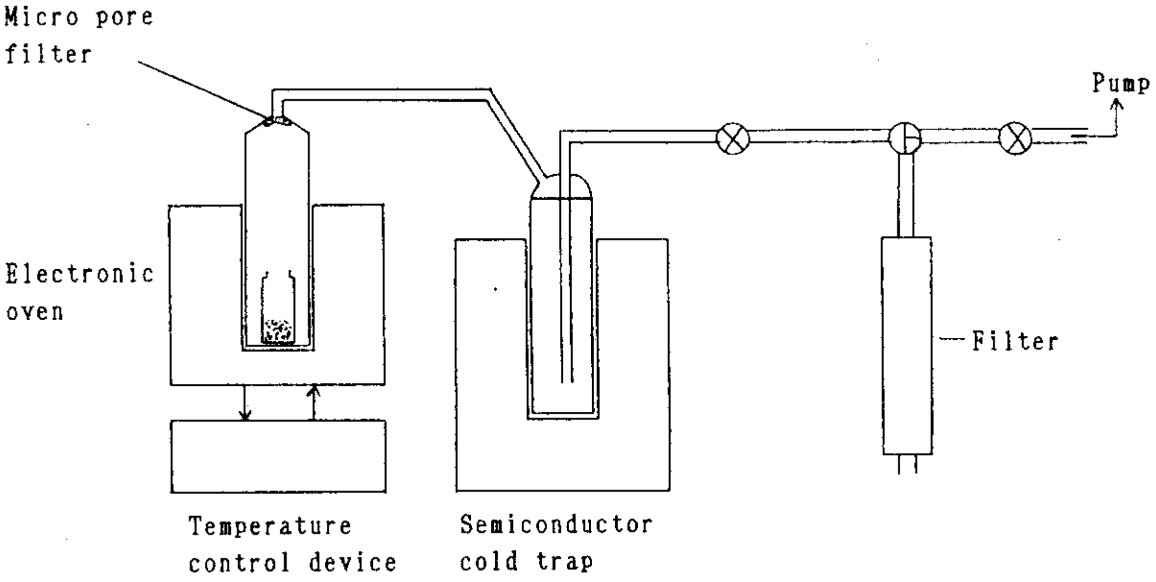


Fig.9 Vacuum-heating distillation system