

A Radioanalytical Method for Samarium-151 and Promethium-147 in Environmental Samples

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動力炉・核燃料開発事業団

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環境試料中Sm-151及びPm-147分析法の開発

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要 旨

環境影響評価の観点から、環境試料中のSm-151及びPm-147の分析法を開発した。

環境試料中のSm-151及びPm-147は、試料中または添加した安定Ndを分析回収率補正用キャリアーに用い、しゅう酸カルシウム共沈、水酸化鉄共沈、鉍酸-メタノール系陰イオン交換法により精製した後、乳酸-水酸化ナトリウム溶液を溶離液とした高速液体クロマトグラフ装置を用い単離し、それぞれを液体シンチレーションカウンタで定量した。

本分析法による検出下限値は、測定時間 500分で約10mBq/サンプルである。

本報告書は、安全管理部環境安全課における環境安全研究の一環として、これまで検討してきた「環境試料中Sm-151及びPm-147分析法の開発」について、その検討結果をとりまとめたものである。

また、平成2年9月19日から9月21日までの3日間にわたり、英国マンチェスター大学で開催された“6th International Symposium on Environmental Radiochemical Analysis; Royal Society of Chemistry”（第6回環境放射化学分析国際シンポジウム；王立化学協会主催）において発表した本研究報告内容についてもとりまとめた。

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A Radioanalytical Method for Samarium-151 and Promethium-147 in Environmental Samples

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Abstract

A radioanalytical method for low level samarium-151(Sm-151) and promethium-147(Pm-147) in environmental samples has been studied for the environmental assessment around nuclear facilities. In this study, we use the separation method with high performance liquid chromatography (HPLC) to determine Sm-151 and Pm-147 in environmental samples such as sea sediments and marine organisms. Samarium-151 and Pm-147 in environmental samples are coprecipitated with other lanthanoids after adding neodymium(Nd). These nuclides are purified by anion exchange methods in methanol-mineral acid media. After the purification, Sm-151 and Pm-147 are separated with HPLC in lactic acid-sodium hydroxide media, and determined with liquid scintillation counting, respectively. The Nd is determined by inductively coupled plasma atomic emission spectrometry(ICP-AES) to correct chemical recoveries of these nuclides. The detection limits for Sm-151 and Pm-147 in this method are about 0.01Bq/sample.

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**[2] Presentation on 6th International Symposium on Environmental
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**[1] A Radioanalytical Method for Samarium-151 and
Promethium-147 in Environmental Samples**

1. Introduction

Sm-151 and Pm-147 are pure beta emitting nuclides having the half-lives of 91 and 2.62 years and the maximum beta energies of 76 keV and 225 keV, respectively. These nuclides are fission products and have relatively high fission yields among fission products having one-year and longer half-lives.

However the analyses of these nuclides in environmental samples have little been reported, because these nuclides have little contribution to the effective dose equivalents to the public, and partly because it was difficult to separate these nuclides from other lanthanoids.

We developed a radioanalytical method of Sm-151 and Pm-147 in environmental samples, in order to carry out more realistic environmental assessment around nuclear facilities, and from the view point of waste disposal, especially in the case of Sm-151.

In this report, we describe the radioanalytical method for separation of Sm, Pm and Nd with HPLC, and the application to environmental samples.

2. Experimental

2.1 Apparatus and measuring method

2.1.1 High Performance Liquid Chromatography (HPLC) system

Fig.1 shows the schematic diagram of the HPLC system. This system (made by Hitachi Ltd.) mainly consists of a preconcentration column, a separation column, a coulometric detector, a data processor and a fraction collector. The cation exchange resin (Hitachi custom ion exchange resin #2618: grain size 11.5 μ m) was used in the preconcentration and separation column. The separation column was kept at 60°C with a column oven.

2.1.2 Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) system

An ICP-AES system was used to examine the separating conditions for Sm and Nd with HPLC and to determine the concentration of stable neodymium to correct the chemical recoveries of those nuclides.

The ICP-AES system (SPS 1100 by Seiko Electronic Industry) had a plasma power source having a frequency of 27.12MHz and the maximum output of 2.5kW.

The Sm and Nd were determined by using the wave lengths of 359.266nm and 401.246nm, respectively.

2.1.3 Liquid Scintillation Counter

A low background liquid scintillation counter (Model 2260 XL by Packard Instrument Company) was used to determine the activities of Sm-151 and Pm-147,

respectively. Before measuring the samples by means of the liquid scintillation counter, they were prepared by dissolving dried samples in 20ml glass vials with 2ml of 1N HCl diluted by low-tritium water (spring water obtained at 660m underground) and adding 18ml of scintillator (Aquasol-2 by New England Nuclear Corporation) .

The measurement conditions and counting efficiencies of Pm-147 and Sm-151 were set by using standard solutions of Pm-147 and Ni-63(β_{\max} ; 66keV) which were supplied by Amersham Corporation, because the maximum beta energy of Ni-63 is almost equal to that of Sm-151(β_{\max} ; 76keV) .

2. 2 Experiment

2.2.1 Measurement of the concentration of Nd in environmental samples

It is reported that the concentration of Nd is 28 ppm on the earth's crust⁽¹⁾, 28 ppm in igneous rock⁽¹⁾, 13 ppm in sandstone⁽²⁾ and 4.3 ppm in limestone⁽²⁾ .

If the concentration of Nd was high, Pm might be incompletely separated from the Nd which had a broad elution curve.

We also determined the concentrations of Nd in sea sediments, surface soils and ashed seaweeds. The concentrations of Nd were 3.3 to 14ppm in sea sediments, 4.7 to 15ppm in surface soils, and 0.7 to 1.3ppm in ashed seaweeds.

Therefore, to correct the chemical recovery, we used the Nd present for soil samples, and added 1mg of Nd to ashed seaweed samples because of the low concentration of Nd.

2.2.2 Feed conditions into the preconcentration column

To separate Sm, Pm and Nd from each other, it was necessary to concentrate these lanthanoids into the preconcentration column before the HPLC separation.

The effects of feed conditions on the leakage from the preconcentration column were confirmed by measuring Sm and Nd with ICP-AES.

2.2.3 Eluting conditions in HPLC

The separation methods of lanthanoids were reported by cation exchange in lactic acid-NaOH media^{(3),(4),(5)}.

The used eluents were 2 types of 4% lactic acid solution having different pH values (A; 4% lactic acid-0.1 M NaOH (pH=3), B; 4% lactic acid-0.4 M NaOH (pH=5)). These eluents were mixed with the variable gradient patterns.

After we decided the retention time of each element fraction with the coulometric detector, we collected each fraction with the fraction collector controlled by a timer.

2.2.4 Application to environmental samples

Fig.2 shows the radioanalytical procedure of Sm-151 and Pm-147 in environmental samples. This procedure consisted of the coprecipitation of calcium oxalate and iron hydroxide, anion exchange in hydrochloric acid medium, anion exchange in nitric acid medium, methanol mineral acid media, and cation exchange with HPLC in lactic acid sodium hydroxide media.

We usually used the anion exchange method in methanol mineral acid media to purify americium(Am)-241 in environmental samples⁽⁶⁾. In this step, we could separate lanthanoids from Am and curium(Cm). Lanthanoids were eluted with 0.1N HCl-0.5M NH₄SCN-80% CH₃OH eluent and Am and Cm were kept on the column.

Sm, Pm and Nd were separated with HPLC. Sm and Pm were determined by the liquid scintillation counter, respectively. To correct the chemical recoveries of these nuclides, Nd was used as a chemical recovery tracer and the concentration of Nd was determined by the ICP-AES.

The concentration of Nd in the sample was also determined by using a part of the leachate and ICP-AES .

3. Results and Discussion

3.1 Feed conditions into the preconcentration column

The effects of feed conditions such as flow rate, acidity and feed volume on the leakage of lanthanoids from the preconcentration column were examined for 1mg of stable Sm and Nd. The results are given in Table 1.

The effect of the flow rate varying from 0.5ml/min to 5.0ml/min was examined with the acidity of 0.04N and the feed volume of 50ml. In these results, no leakage of lanthanoids from the column was detected even at the flow rate of 5.0ml/min.

The effect of the acidity of the feed solution was examined with the acidity varying from 0.01N to 8N, the flow rate of 5.0ml/min and the feed volume of 50ml. It was proved that there was no effluent with these elements from the preconcentration column in the range of acidities up to 0.1N. This result showed that it was necessary to adjust the acidity in the feed solution to 0.1N or less by dilution or neutralization.

The effect of the volume of the feed solution from 50ml to 300ml was examined at the flow rate of 5.0ml/min and the acidity of 0.1N. In these results, the leakage of lanthanoids from the column was not detected in these conditions.

3.2 Eluting conditions in HPLC separation

The separation of stable Sm and Nd was conducted by using 2 types of eluents which were 4% lactic acid-NaOH solutions having different pH values. Fig.3 shows the gradient pattern and the elution curves of lanthanoids. The employed gradient pattern

was that the mixing ratio of the solution B was increased from 20% to 30% in 80 min and from 30% to 40% in next 40 min. The effective separation of three elements was confirmed with the coulometric method for Sm and Nd, and with the liquid scintillation counting for Pm-147.

In this gradient pattern, the retention times were determined by stable Sm and Nd varying from 0.1 mg to 1 mg. The results are given in table 2. From these results, it was confirmed that the retention times varied 2% or less for the coefficient of variation(C.V.).

The reproducibilities of the recoveries of stable Sm and Nd in HPLC separation were also determined in the same gradient pattern. The results are given in Table 3. These results showed that the reproducibilities of the chemical recoveries were 2% or less for the C.V., and the recoveries of Sm and Nd were almost 100%.

The influence of interfering elements in the fractions of Sm and Pm separated with the HPLC were examined. The examined interfering elements were europium(Eu) and Pm-147 for Sm-151 and Sm and Nd for Pm-147. The results are shown as decontamination factors (D.F.) in table 4. These results showed that the D.F. of interfering elements in each fraction was high enough to analyze the concentration of Sm-151 and Pm-147 in environmental samples.

3.3 Application to environmental samples

The chemical recoveries of Sm and Nd before the HPLC separation were determined by using 50g of sea sediment samples. The results showed that the chemical recoveries of Sm and Nd were about 90% and that the Sm/Nd ratios were almost 1. These results proved that, in the analytical processes prior to the HPLC separation, the chemical recoveries of Sm and Nd were very high and that Nd had the same behavior as that of Sm.

The reproducibilities of the activities of Sm-151 and Pm-147, and the chemical recoveries of Nd through the whole analytical procedure were determined by the analyzing of sea sediment samples. The results are given in Table 5. They showed that the coefficients of variation of Sm-151 and Nd were 9.4% and 9.0%, respectively. In addition, the chemical recoveries of Nd were about 85%. But the activities of Pm-147 in the samples could not be detected.

3.4 Measurement of Sm-151 and Pm-147 by liquid scintillation counting

To determine the activities of Sm-151 and Pm-147, the measuring ranges of these nuclides must be discriminated from any influence of alpha emitting nuclides such as Sm-147 and Am-241. Samarium contains 15.0% of natural abundance of Sm-147 (half life; 1.1×10^{11} y, alpha energy; 2.2MeV) and Am-241(half life; 433 y, alpha energy;

5.48MeV) is contained in environmental samples as the daughter of Pu-241. But the influence of Am-241 to the measurement of Pm-147 was thought to be small, because the decontamination factors of Am-241 for the lanthanoid fractions in the anion exchange method in methanol mineral acid media were larger than 8.9×10^3 .

In this radioanalytical method, the detection limits of Sm-151 and Pm-147 were about 0.01Bq/sample, respectively, for 500 min of the measuring time. The counting efficiencies of Sm-151 and Pm-147 with the liquid scintillation counter were about 55% and 70%, and the background counting rates of each nuclides were about 4.5 cpm and 7cpm, respectively.

3.5 Determination of Sm-151 and Pm-147 in environmental samples

We determined the activities of Sm-151 and Pm-147 in sea sediments, surface soils, and seaweeds. The results are given in Table 6. The ranges of the activities of Sm-151 and Pm-147 were 1.2 to 1.3Bq/kg · dry, <0.3 to 0.41Bq/kg · dry, respectively, for surface soil samples, 0.40 to 1.8 Bq/kg · dry, <0.4 to 0.59 Bq/kg · dry, respectively, for sea sediment samples, and <0.04 Bq/kg · fresh, <0.06 Bq/kg · fresh, respectively, for seaweed samples.

The measured activities of Sm-151 and Pm-147 in the surface soil and sea sediments can be regarded as originating from global fallout for surface soil and sea sediment in Japan.

4. Conclusion

Samarium-151 and Pm-147 were separated from lanthanoids and other interfering radionuclides with the HPLC system and the radioanalytical method such as the anion exchange method in methanol mineral acid media, and measured in the discriminated range from the interfering nuclides with the liquid scintillation counting. The detection limits were about 0.01Bq/sample both for Sm-151 and Pm-147 for 500 min of the measuring time. The proposed radioanalytical method is sensitive and reproducible enough to determine low level activities of Sm-151 and Pm-147 in environmental samples.

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Table 1 Effects of feed conditions on the leakage from the preconcentration column

Feed conditions		Leakage (%)	
		Sm	Nd
Flow rate (ml/min)	0.5	<0.1	<0.1
	1.0	<0.1	<0.1
	· Acidity ; 0.04 N	2.0	<0.1
	· Volume ; 50 ml	3.0	<0.1
	5.0	<0.1	<0.1
Acidity (N)	0.01	<0.1	<0.1
	0.05	<0.1	<0.1
	· Flow rate ; 5.0 ml/min	0.1	<0.1
	· Volume ; 50 ml	1	51.4
		3	97.3
	8	100	100
Volume (ml)	50	<0.1	<0.1
	· Flow rate ; 5.0ml/min	100	<0.1
	· Acidity ; 0.1 N	300	<0.1

Table 2 Retention time of Sm and Nd in HPLC separation

Added amounts of Sm and Nd (mg)	Retention time (min)	
	Sm	Nd
0.10	69.6	100.2
0.50	71.3	100.0
0.75	71.9	100.1
1.00	71.5	99.5
Average $\pm 1 \sigma$	71.1 \pm 1.01	100.0 \pm 0.31
Coefficient of Variation (%)	1.4	0.31

Table 3 The reproducibility of the recoveries of Sm and Nd in HPLC separation

Run No.	Separation recovery (%)	
	Sm	Nd
1	92.7	100
2	92.3	102
3	97.1	102
4	94.7	-
5	95.9	100
Average $\pm 1 \sigma$	94.5 \pm 2.05	101 \pm 1.2
Coefficient of variation (%)	2.2	1.2

Table 4 Decomtamination factors of Eu,Sm,Pm-147 and Nd in HPLC separation

Collected fraction	Decomtamination factor			
	Eu	Sm	Pm-147	Nd
Sm fraction	250	-	>700	-
Pm-147 fraction	-	450	-	370

Table 5 The reproducibilities of the activities of Sm-151, Pm-147 and the chemical recoveries of Nd

Run No.	Activity (Bq/kg · dry)		Chemical recovery(%)
	Sm-151	Pm-147	Nd
1	1.08±0.10	N.D.	91.9
2	1.21±0.12	N.D.	76.4
3	1.03±0.10	N.D.	95.7
4	0.959±0.10	N.D.	87.0
5	1.17±0.11	N.D.	81.3
Average±1σ (Standard deviation)	1.09±0.11	-	86.5±7.80
Coefficient of variation (%)	9.4	-	9.0

Table 6 Activities of Sm-151 and Pm-147 in environmental samples

Sample	Location	Sampling date	Activity	
			Sm-151 (Bq/kg · dry for soil or Bq/kg · fresh for seaweed)	Pm-147
Surface soil-1	Katsuta,Ibaraki-ken	May,90	1.2±0.1	<0.3
Surface soil-2	Rokkasho,Aomori-ken	July,89	1.3±0.1	0.41±0.12
Sea sediment-1	Off-shore Tokai,Ibaraki-ken	June,89	0.40±0.05	<0.3
Sea sediment-2	Off-shore Tokai,Ibaraki-ken	June,89	1.6±0.04	0.51±0.05
Sea sediment-3	Off-shore Tokai,Ibaraki-ken	June,89	0.83±0.06	<0.3
Sea sediment-4	Off-shore Ose,Ibaraki-ken	Apr.,89	1.8±0.04	0.59±0.04
Sea sediment-5	Off-shore Rokkasho,Aomori-ken	July,89	1.1±0.1	<0.4
Seaweed-1	Off-shore Tokai,Ibaraki-ken	June,90	<0.03	<0.03
Seaweed-2	Off-shore Rokkasho,Aomori-ken	Aug.,89	<0.04	<0.04
Seaweed-3	Off-shore Mihama,Fukui-ken	Aug.,88	<0.04	<0.06

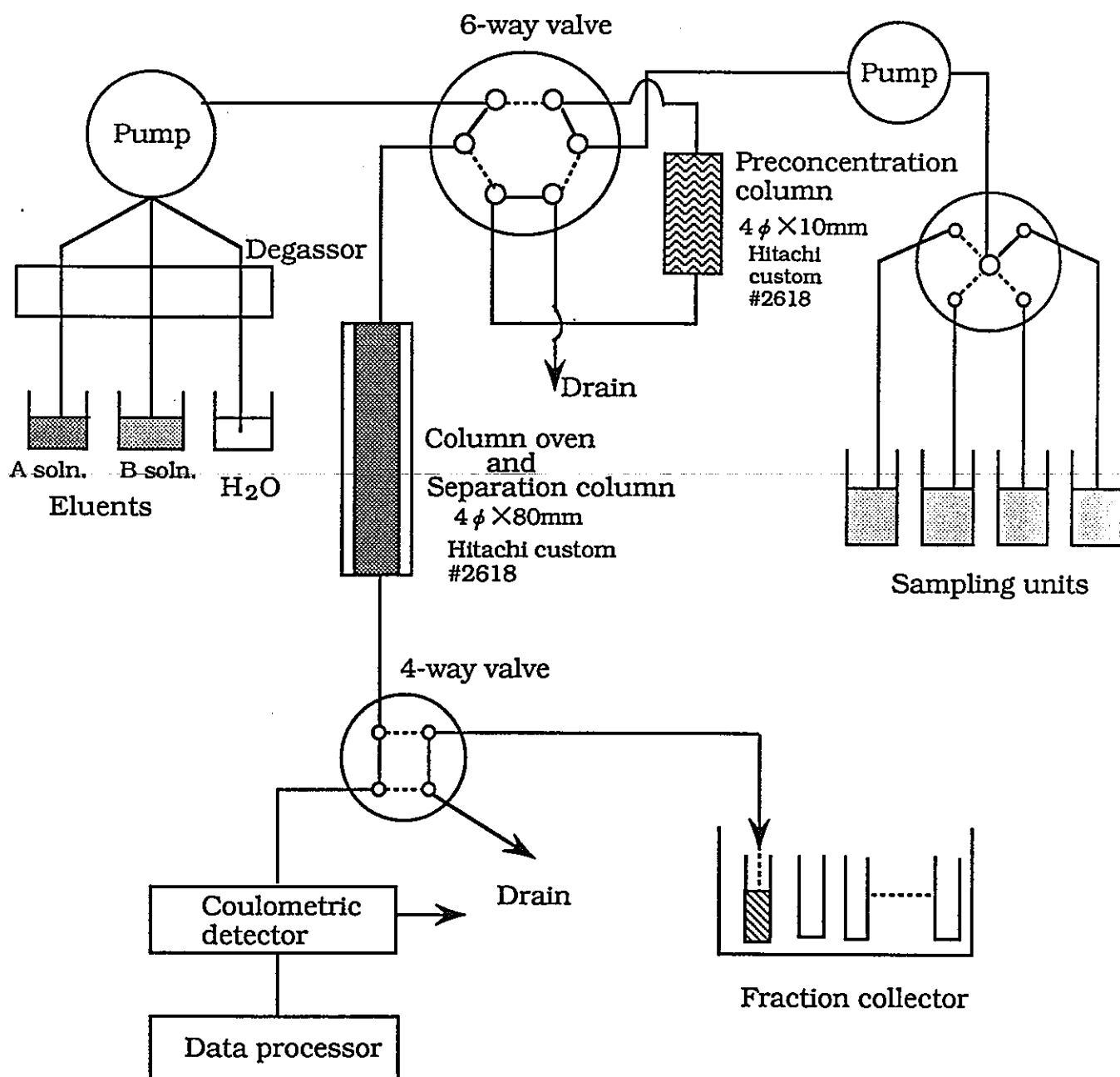


Fig.1 Schematic diagram of HPLC system

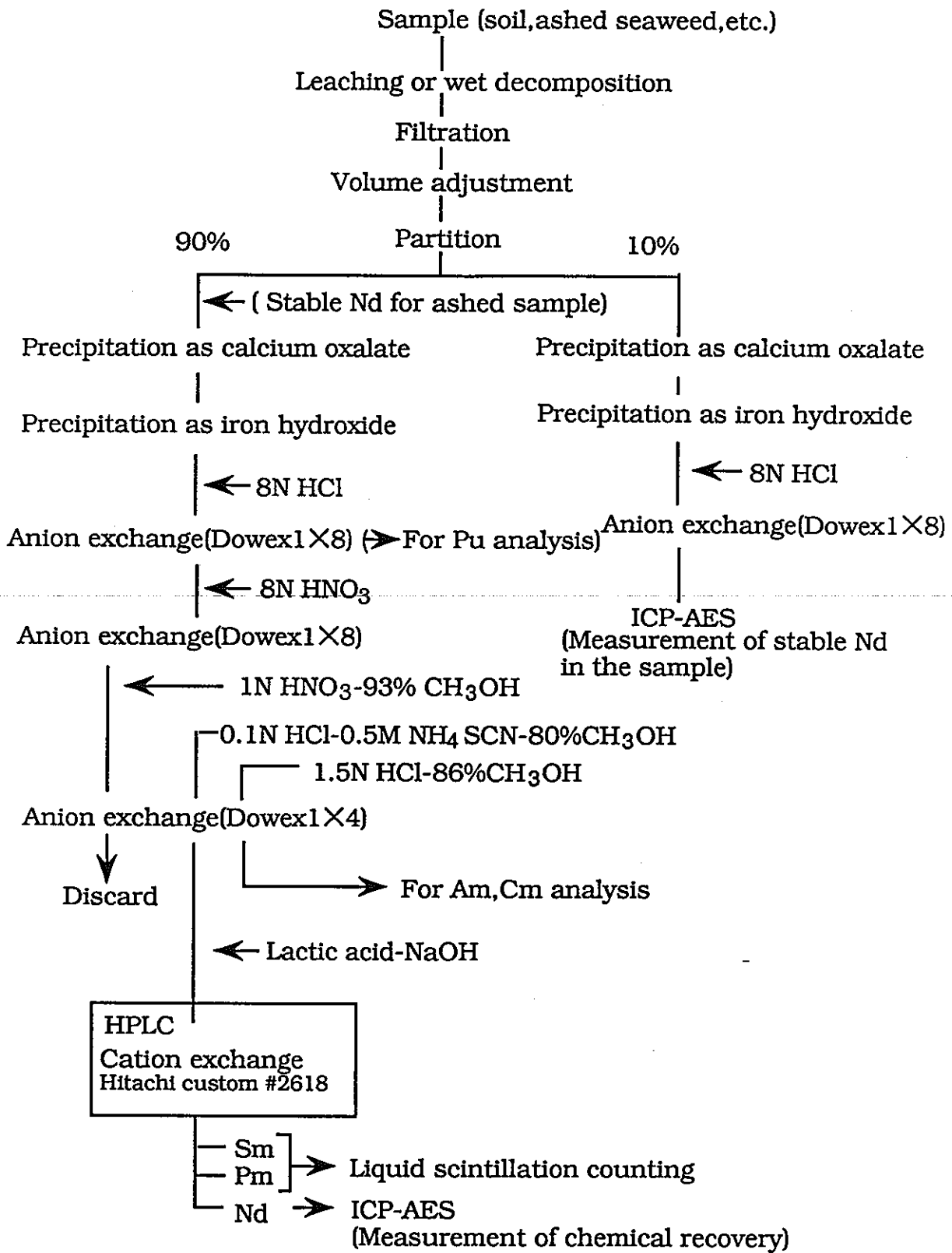
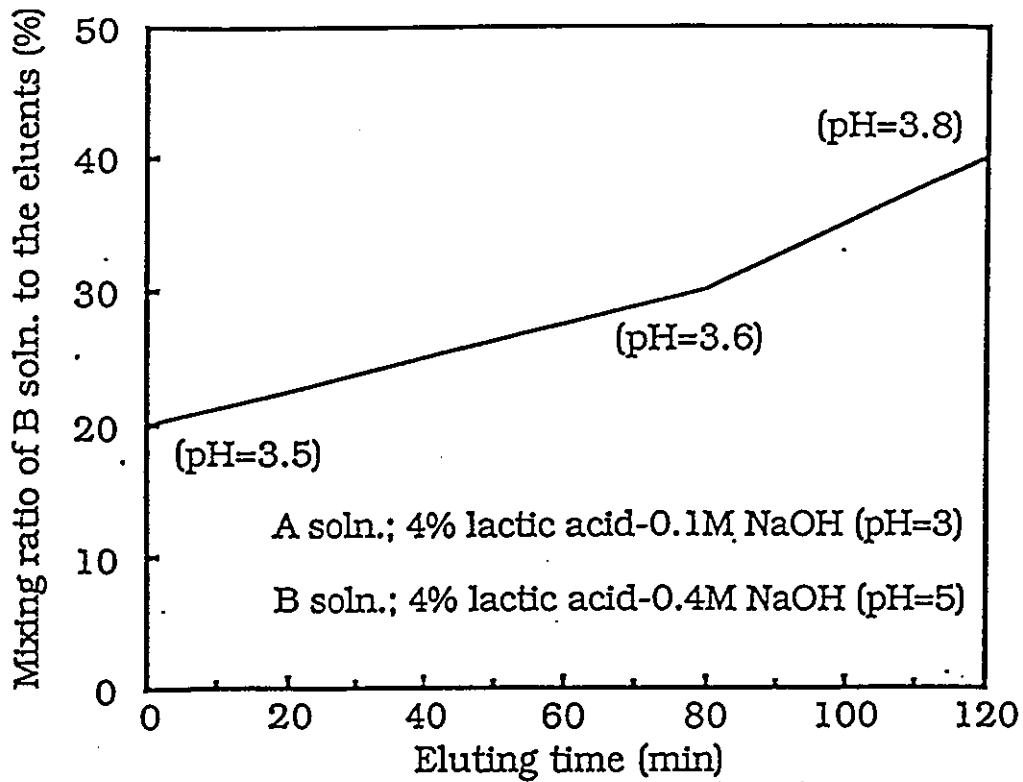
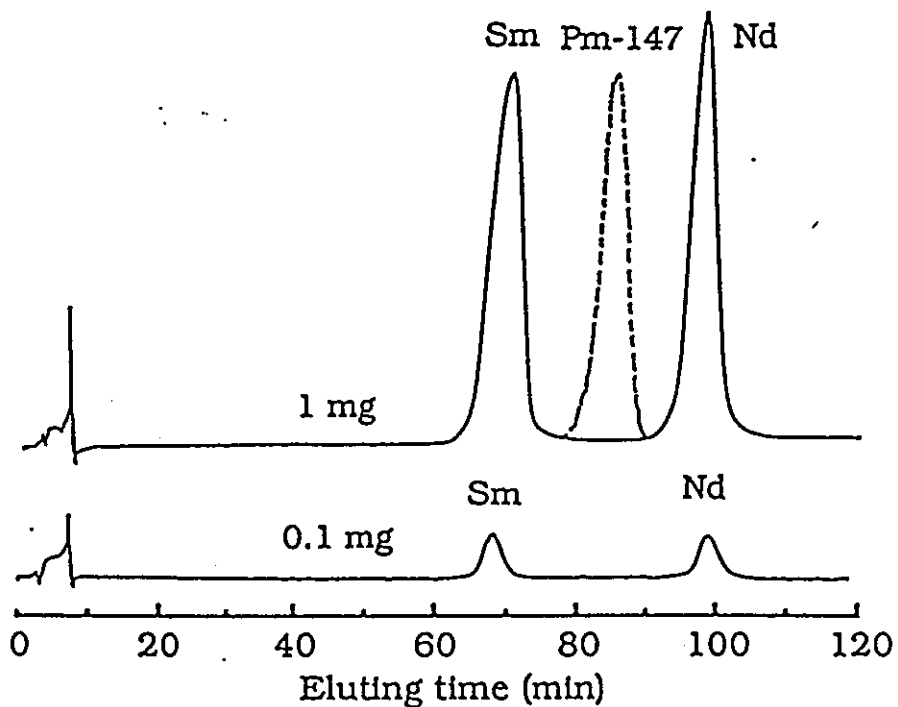


Fig.2 Radioanalytical procedure of Sm-151 and Pm-147 in environmental samples



Gradient pattern of the eluents for the separation of Sm, Pm and Nd with HPLC



Elution curves of Sm, Pm and Nd in HPLC separation

Fig. 3 The gradient pattern of the eluents and elution curves of lanthanoids

**[2] Presentation on 6th International Symposium
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A Radioanalytical Method for Samarium-151 and Promethium-147 in Environmental Samples

Mr.chairman, Thank you for your kind introduction.

Ladies and gentlemen,

It is my great pleasure to be here and talk to you.

I will present a radioanalytical method for samarium-151 and promethium-147 in environmental samples.

Physical properties and fission yields of Sm-151 and Pm-147

Nuclide	Half life (y)	β max. energy (keV)	Formation	Fission yield (%)		Activity in spent fuel (Bq/t)
				U-235	Pu-239	
Sm-151	91	76.4	(n,f) $^{150}\text{Sm}(n, \gamma)$ $^{151}\text{Pm}(\beta)$	0.42	0.77	13TBq/t
Pm-147	2.62	225	(n,f) $^{146}\text{Nd}(n, \gamma)$ $^{147}\text{Nd}(\beta)$	2.26	1.95	4.8PBq/t

This table shows the physical properties and fission yields of samarium-151 and promethium-147.

These nuclides are pure beta emitting radionuclides having the half-lives of 91 and 2.6 years, respectively.

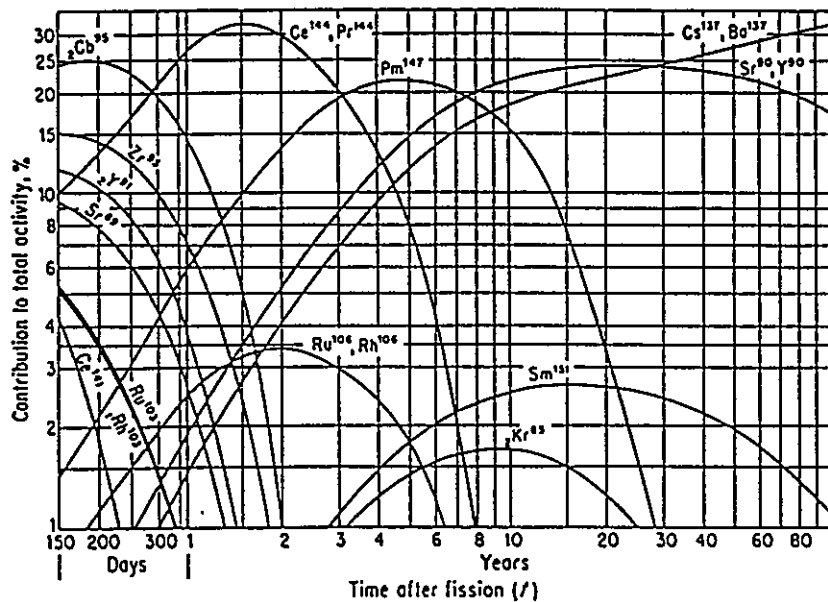
Their maximum beta energies are relatively low, 76 keV for Samarium-151 and 225 keV for promethium-147.

And their chemical characteristics are very similar, because they belong to lanthanoids.

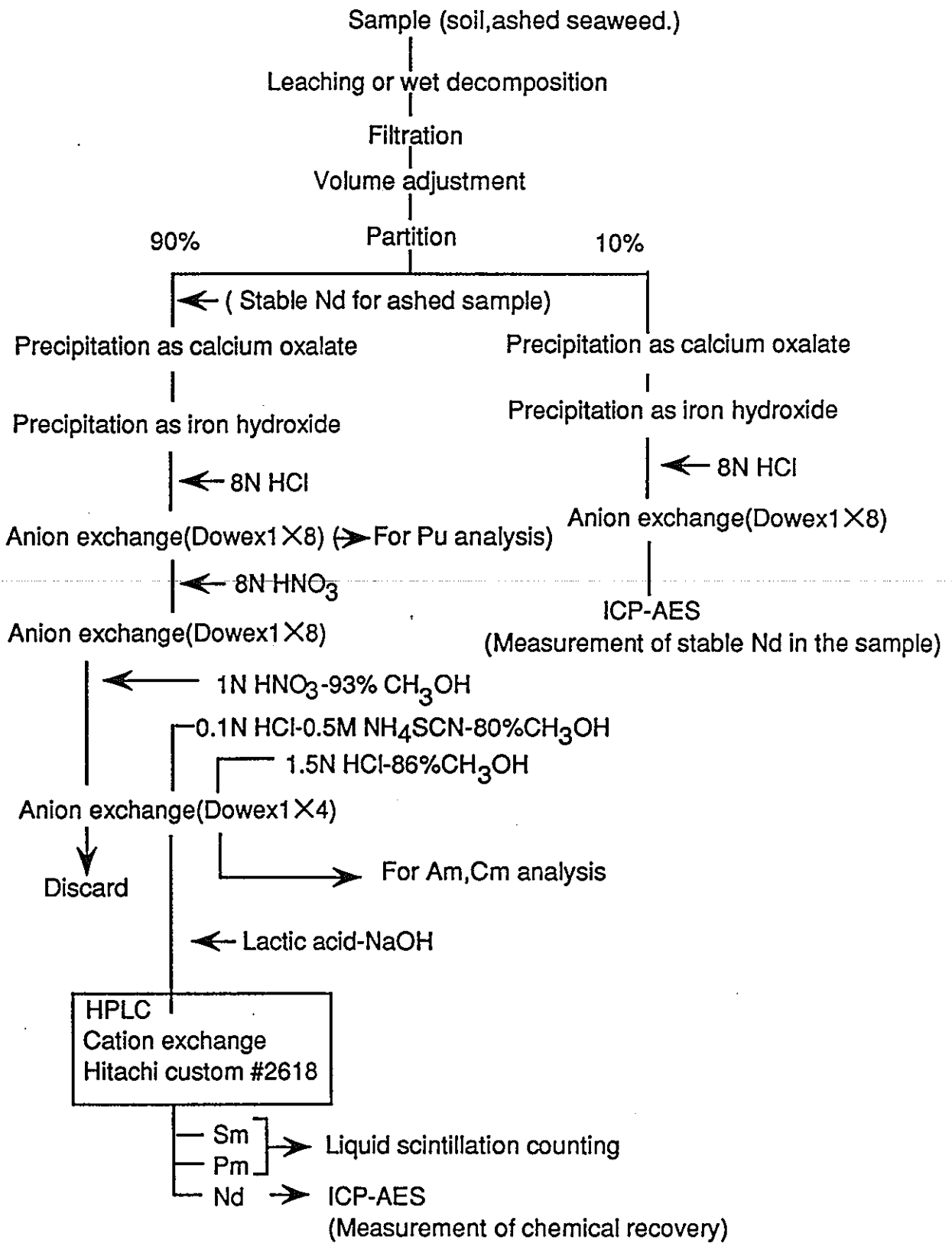
Therefore, the determinations of these nuclides in environmental samples have little been reported, in spite of their relatively high fission yields at irradiation of thermal neutrons.

Today, I will present the application of High Performance Liquid Chromatography (HPLC) to the separation these nuclides from other lanthanoids and the measurement by liquid scintillation counting.

参考



Yields of the principal radionuclides from the slow-neutron fission of ^{235}U .
(From Hunter and Ballou, 1951.)



Radioanalytical procedures of Sm-151 and Pm-147 in environmental samples

This figure shows the radioanalytical procedure of Sm-151 and Pm-147 in environmental samples such as sea sediment and marine products.

This procedure consists of the co-precipitation of calcium oxalate and iron hydroxide, anion exchange in hydrochloric acid medium, in nitric acid medium and methanol mineral acid media, and cation exchange with HPLC in lactic acid sodium hydroxide media.

We usually use the anion exchange method in methanol mineral acid media to purify americium-241 in environmental samples.

In this step, we can separate lanthanoids from americium and curium.

Lanthanoids are eluted with 0.1N hydrochloric acid - 0.5M ammonium thiocyanate - 80% methanol eluent, and americium and curium are kept on the column.

Samarium, promethium, and neodymium were separated each other with HPLC system.

Samarium and promethium were determined by the liquid scintillation counter, respectively.

To correct the chemical recovery of these nuclides, Neodymium was used as a chemical recovery tracer, and the concentration of stable neodymium was determined with the ICP (Inductively Coupled Plasma) Atomic Emission Spectrometry (ICP-AES).

The concentration of neodymium in the sample was also determined by using a part of the leachate and ICP-AES.

At first, we determined the chemical recoveries of samarium and neodymium before the HPLC separation to confirm the propriety to use neodymium as a chemical recovery tracer.

Next, we examined the separation conditions of Sm, Pm and Nd with HPLC system such as feed conditions into a preconcentration column and eluting conditions in HPLC.

Finally, we applied this radioanalytical method to environmental samples.

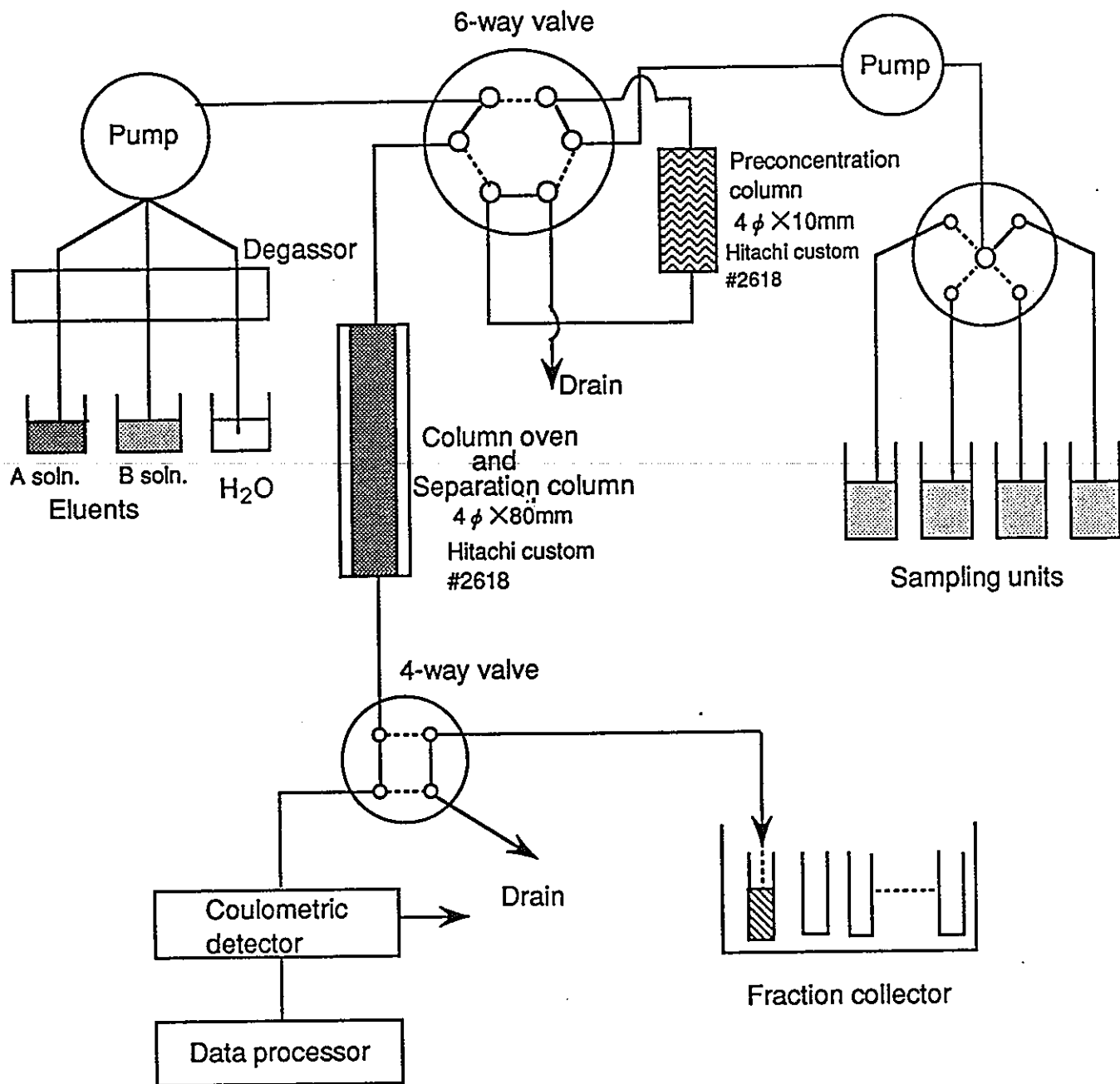
The chemical recoveries of stable Sm, Nd and Sm/Nd ratio in the radioanalytical procedure before the separation with HPLC

Run No.	Chemical recovery (%)		Sm/Nd ratio
	Sm	Nd	
1	91.4	91.4	1.00
2	91.5	89.5	1.02
3	83.5	83.2	1.00

This table shows the chemical recoveries of stable Sm,Nd and Sm/Nd ratios before the HPLC separation.

The chemical recoveries of these elements were about 90% and that the ratios were almost 1.

These results prove those, in the processes before the HPLC separation, the chemical recoveries of these elements are very high and that neodymium has the same behavior as that of samarium.



Schematic diagram of HPLC system

This figure shows the schematic diagram of the HPLC system.

This HPLC system consists of a preconcentration column, a separation column, a coulometric detector, a data processor and a fraction collector.

The cation exchange resin was used in the preconcentration and separation column.

After we decided the retention time of each element with the coulometric detector, we collected each element with the fraction collector controlled by a timer.

Effects of feed conditions on the leakage from the preconcentration column

Feed conditions	Leakage (%)			
	Sm	Nd		
Flow rate (ml/min)	0.5	<0.1	<0.1	
	1.0	<0.1	<0.1	
	• Acidity ; 0.04 N	2.0	<0.1	<0.1
	• Volume ; 50 ml	3.0	<0.1	<0.1
	5.0	<0.1	<0.1	
Acidity (N)	0.01	<0.1	<0.1	
	0.05	<0.1	<0.1	
	• Flow rate ; 5.0 ml/min	0.1	<0.1	<0.1
	• Volume ; 50 ml	1	51.4	38.4
	3	97.3	100	
8	100	100		
Volume (ml)	50	<0.1	<0.1	
	• Flow rate ; 5.0ml/min	100	<0.1	<0.1
	• Acidity ; 0.1 N	300	<0.1	<0.1

To separate samarium, promethium and neodymium from environmental samples with the HPLC, it is necessary to concentrate these lanthanoids into the preconcentration column before the HPLC separation.

This table shows the results of the effects of feed conditions such as flow rate, acidity and feed volume on the leakage of lanthanoids from the preconcentration column by 1 mg of stable Sm and Nd.

The effect of the flow rate varying from 0.5ml/min to 5.0ml/min was examined.

In these results, No leakage of lanthanoids from the column was detected even at the flow rate of 5.0ml/min.

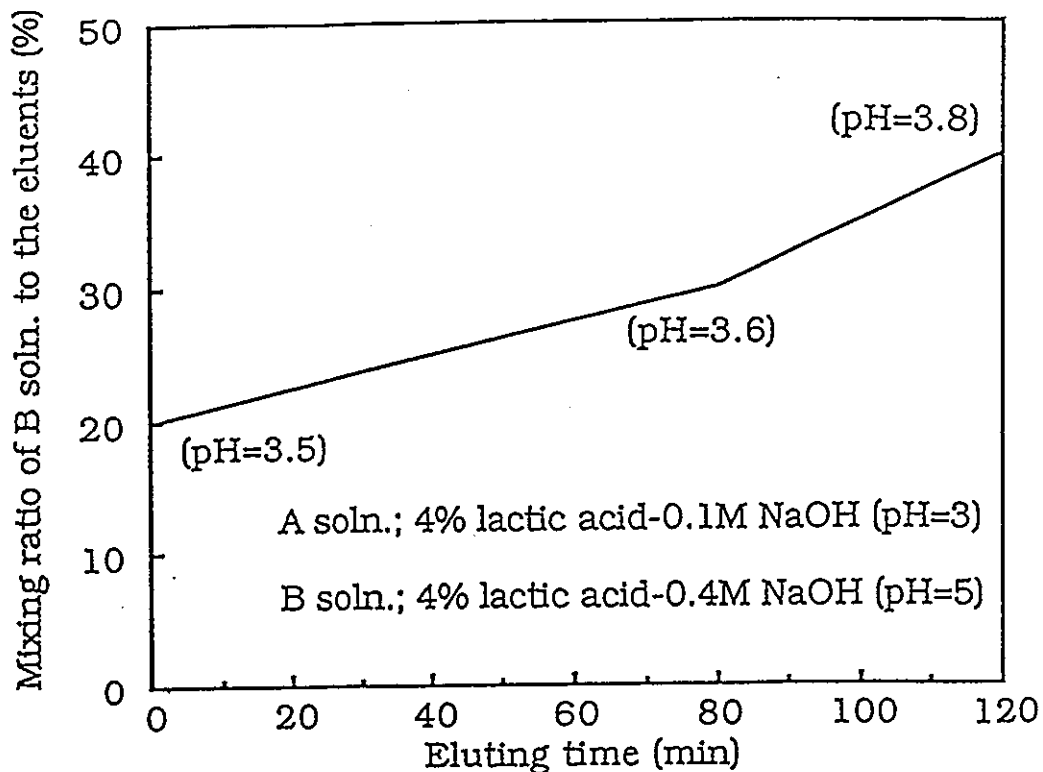
The effect of the acidity of the feed solution was examined with the acidity varying from 0.01N to 8N.

It was proved that there was no effluent with these elements from the column in the range of acidities up to 0.1N.

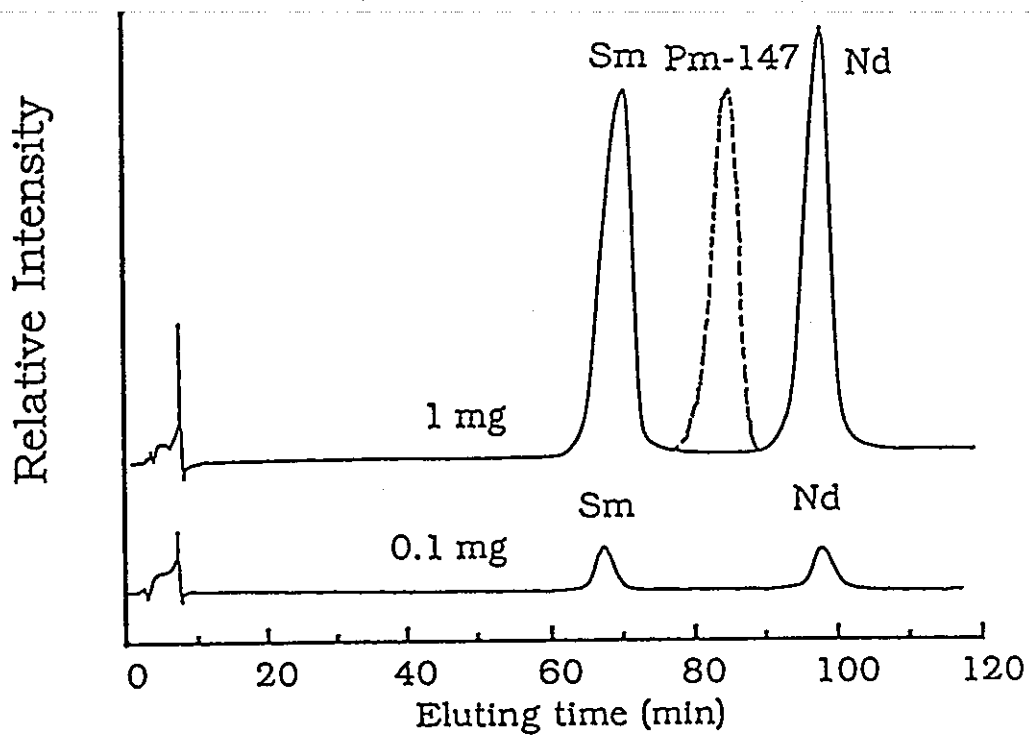
This result showed that it was necessary to adjust the acidity in the feed solution to 0.1N or less by dilution or neutralization.

The effect of the volume of the feed solution from 50ml to 300ml was examined.

In these results, No leakage of lanthanoids from the column was detected in these conditions.



Gradient pattern of the eluents for the separation of Sm, Pm and Nd with HPLC



Elution curves of Sm, Pm and Nd in HPLC separation

The gradient pattern of the eluents and elution curves of lanthanoids

This figure shows the gradient pattern and the elution curves of lanthanoids.

The eluting conditions of lanthanoids were examined by using 2 types of eluents.

A solution was 4% lactic acid - 0.1M sodium hydroxide solution.

B solution was 4% lactic acid - 0.4M sodium hydroxide solution.

These solutions had different pH values and were mixed.

Then mixing ratios of the solutions were gradually changed.

The employed gradient pattern was that the mixing ratio of the B solution was increased from 20% to 30% in 80 min and from 30% to 40% in next 40 min.

In this gradient pattern, the effective separation of three elements was confirmed with the coulometric method for Sm and Nd, and with the liquid scintillation counting for Pm-147.

Retention time of stable Sm and Nd in HPLC separation

Added amounts of Sm and Nd (mg)	Retention time (min)	
	Sm	Nd
0.10	69.6	100.2
0.50	71.3	100.0
0.75	71.9	100.1
1.00	71.5	99.5
Average $\pm 1 \sigma$	71.1 \pm 1.01	100.0 \pm 0.31
Coefficient of Variation (%)	1.4	0.31

In this gradient pattern, the retention times were determined by stable Sm and Nd varying from 0.1 mg to 1 mg.

From these results, it was confirmed that the retention times were as little variable as 2% or less in coefficient of variation(C.V.).

The reproducibility of the recoveries of Sm and Nd in HPLC separation

Run No.	Separation recovery (%)	
	Sm	Nd
1	92.7	100
2	92.3	102
3	97.1	102
4	94.7	-
5	95.9	100
Average $\pm 1 \sigma$	94.5 ± 2.05	101 ± 1.2
Coefficient of variation (%)	2.2	1.2

The reproducibilities of the recoveries of stable Sm and Nd in HPLC separation were also determined in the same gradient pattern.

These results showed that the reproducibilities of the chemical recoveries were as little variable as 2% or less in C.V., and the recoveries of stable Sm and Nd were almost 100%.

Decomtamination factors of Eu,Sm,Pm-147 and Nd in HPLC separation

Collected fraction	Decomtamination factor			
	Eu	Sm	Pm-147	Nd
Sm fraction	250	-	> 700	-
Pm-147 fraction	-	450	-	370

The influences of interfering elements in the fractions of Sm and Pm separated with the HPLC were examined.

This table shows the results as decontamination factors (DF).

The examined interfering elements were europium(Eu) and Pm-147 for Sm-151 and Sm and Nd for Pm-147.

These results showed that the decontamination factors (D.F.) of interfering elements in each fraction were high enough to analyze the concentration of Sm-151 and Pm-147 in environmental samples.

**The reproducibilities of the activities of Sm-151, Pm-147
and the chemical recoveries of Nd in sea sediments**

Run No.	Activity (Bq/kg · dry)		Chemical recovery(%)
	Sm-151	Pm-147	Nd
1	1.08±0.10	N.D.	91.9
2	1.21±0.12	N.D.	76.4
3	1.03±0.10	N.D.	95.7
4	0.959±0.10	N.D.	87.0
5	1.17±0.11	N.D.	81.3
Average±1 σ (Standard deviation)	1.09±0.11	-	86.5±7.80
Coefficient of variation (%)	9.4	-	9.0

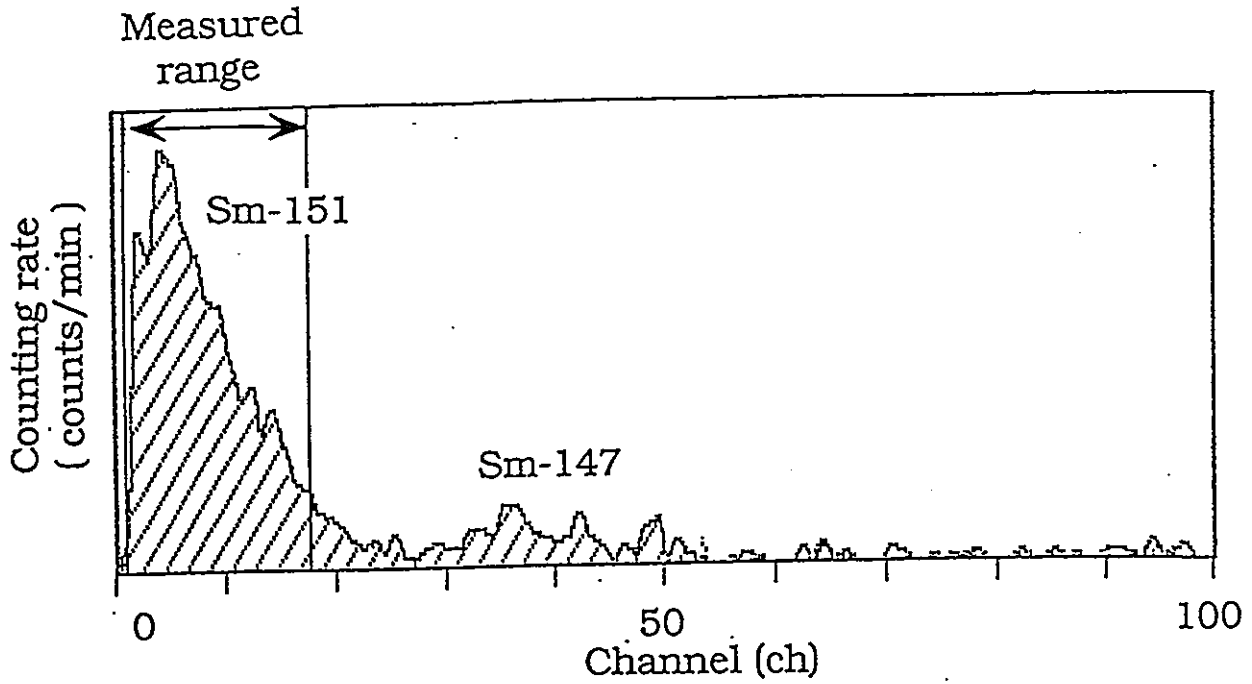
The reproducibilities of the activities of Sm-151 and Pm-147 and the chemical recoveries of stable Nd through the whole analytical procedure were determined by the analyzing of sea sediment samples.

The results are given in this table.

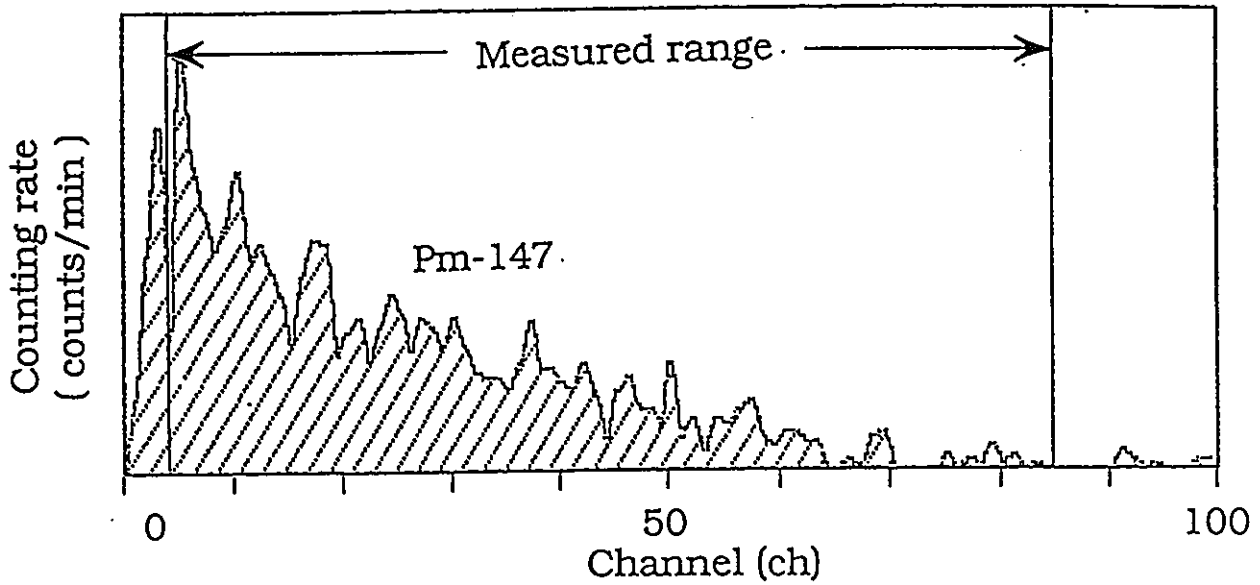
They showed that the coefficients of variation of Sm-151 and Nd were 9.4% and 9.0%, respectively.

In addition, the chemical recoveries of Nd were about 85%.

But the activities of Pm-147 in the samples could not be detected.



Spectra of Sm-151 and Sm-147 on liquid scintillation counting



Spectrum of Pm-147 on liquid scintillation counting

Spectra of Sm-151, Sm-147 and Pm-147 on liquid scintillation counting

This figure shows the spectra of samarium-151 and promethium-147 in sea sediment sample with liquid scintillation counting.

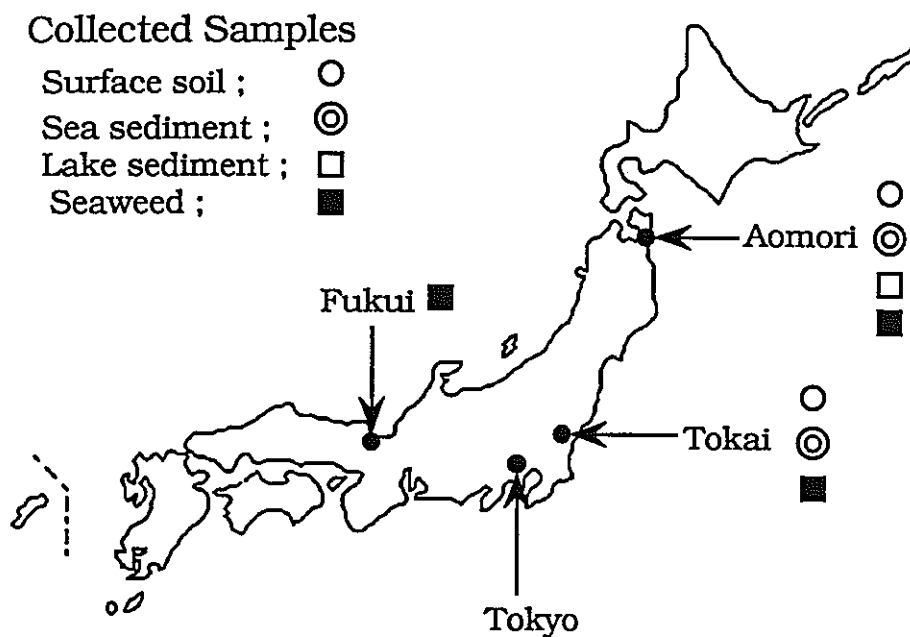
Then, these lines show the measured ranges of these nuclides.

The measurement conditions and counting efficiencies of promethium-147 and samarium-151 were determined by using a standard source of promethium-147 and nickel-63 (Ni-63).

Nickel-63 has the maximum β energy almost equal to that of samarium-151 (β_{\max} ; 76keV).

To determine the activities of Sm-151, measured range of Sm-151 was discriminated from any influence of Sm-147 alpha ray, because stable Sm contains 15.0% of natural abundance of Sm-147 (half life; 1.1×10^{11} y, alpha emitting energy; 2.2MeV).

The detection limits of Sm-151 and Pm-147 were about 0.01Bq/sample, respectively, for 500 min of the measuring time.



Sampling points and collected samples

The ranges of measured activities of Sm-151 and Pm-147 in environmental samples

Samples	Measured activity (Bq/kg · dry or fresh)	
	Sm-151	Pm-147
Soil	0.40-1.8	N.D.-0.59
Seaweed	N.D.	N.D.

N.D. ;Not detected

We determined the activities of samarium-151 and promethium-147 in environmental samples.

This sheet shows the sampling points, the collected samples and the activities.

Our laboratory is located in Tokai here.

Surface soils, sea sediments and seaweeds were collected in 3 points, Tokai, Aomori and Fukui.

The ranges of the activities of Sm-151 and Pm-147 were for soil samples, 0.40 to 1.8 Bq/kg · dry, N.D. to 0.59Bq/kg · dry respectively, for seaweed samples both N. D. .

Conclusion

1. The proposed radioanalytical method is sensitive and reproducible enough to determine low level activities of Sm-151 and Pm-147 in environmental samples.

2. Samarium-151 and Pm-147 were separated from lanthanoids and other interfering radionuclides with the HPLC system and the radioanalytical method, and measured in the discriminated range from the interfering nuclides by the liquid scintillation counting.

3. The detection limits were about 0.01Bq /sample both for Sm-151 and Pm-147 for 500 min of the measuring time.

Finally, our researches are brought to conclusion as follows.

1. The proposed radioanalytical method is sensitive and reproducible enough to determine low level activities of Sm-151 and Pm-147 in environmental samples.

2. Samarium-151 and Pm-147 were separated from lanthanoids and other interfering radionuclides with the HPLC system and the radioanalytical method such as the anion exchange method in methanol mineral acid media, and measured in the discriminated range from the interfering nuclides with the liquid scintillation counting.

3. The detection limits were about 0.01Bq/sample both for Sm-151 and Pm-147 for 500 min of the measuring time.