

SESSION IV

NATURALLY OCCURRING RADIATION/RADIOACTIVITY AND ITS USE

TUESDAY, AUGUST 24

4.1 Radioactivity of and exposure by the consumer's goods containing NORM

一般消費財中に含まれる放射性物質の存在量

M. Yoshida, T. Ohhata, S. Sato, R. Ohyama and H. Furuya

吉田昌弘, 大畑勉, 佐藤滋郎, 大島柳太郎, 古屋廣高

Nuclear Safety Technology Center, Hakusan 5-1-3, Bunkyo-ku, Tokyo

原子力安全技術センター

「要旨」

自然起源の放射性物質 (NORM) を含有する 20 種類の一般消費財を、多種多様な情報源を用いて、購入、取得した。これらの一般消費財中の放射能濃度を ICP-MS 及び Ge(Li) 半導体検出器で測定した。この結果を用いて、NORM を含む一般消費財が日常生活で使用される時の年間被ばく量を計算した。この結果、一般消費財の中には、国際原子力機関がクリアランスレベルの指標とする放射能濃度 (U, Th で 1Bq/g) を超える消費財もあったが、その年間被ばく量は、EU (ヨーロッパ連合) の放射線防護委員会が NORM による被ばくについて勧告した制限値 (300 μ Sv/y) より小さかった。

1. Introduction

There exist naturally a number of long-lived radioactive nuclides in the earth. The material containing these radioactive nuclides is called "naturally occurring radioactive materials (NORM)", and several kinds of consumer's goods including NORM are circulated in the usual daily life environment.

Based on how to utilize NORM, these consumer's goods are classified into two types. In the first type of consumer's goods, low level of radiation emitted from NORM is important. As a typical good, the health-enhancing product is famous. Many peoples who believe "hormesis" use several kinds of consumer's goods containing uranium and/or thorium such as underwear, bedclothes, body accessories, small size of hot spring source and so on in their daily life.. In addition, the paint containing NORM is coated on the bottom of ship in order to protect the shell from adhering to it.

In another type of consumer's good, the property characteristic of uranium or thorium is important. Uranium or thorium compounds have clearly beautiful color, depending the atomic valence, because there are vacancies of electron in the 5f shell of inner orbital in the atomic core. Thus, NORM containing uranium and/or thorium are utilized as glazers on the pottery or additions to uranium glass.[1] In addition the chemical stabilities and consequently the melting points of thorium dioxide are very high, and then they are utilized as one kind of refractory and an additive to tungsten electrode for the arc welding..

It is so important from the view of point of regulation to know the radioactive concentration or radioactivity in the consumer's goods containing NORM and the radiation exposure when they are used in the daily living life. In this study, 20 consumer's goods

containing NORM were collected. After chemically processing them, the radioactive concentrations in them were measured by an inductively coupled plasma attached with mass spectrometer (ICP-MS) and a γ -ray spectrometer of Ge(Li). In addition, the radiation exposures were calculated in four typical cases where the consumer's goods are generally used in the daily living life.

2. Experimental procedures

2.1 Collection of specimens

20 consumer's goods containing NORM were collected, based on various information

Table 1 list of consumer's goods containing NORM collected

No.	Kind of good	Specimen	Existing state	Analysis(*1)
1	bedcloth	upper bedcloth	fabric, cotton	ICP-MS γ
2		lower bedcloth	fabric, cotton	ICP-MS —
3	underwear	semisleeve underwear	fabric	ICP-MS γ
4	supporter	belly-warmer tie-A	fabric	ICP-MS γ
5		belly-warmer tie-B	ceramic button(#2)	ICP-MS —
6		wrist band	fabric(print)	ICP-MS —
7	socks	socks	fabric	ICP-MS —
8	shoes sheets	shoes sheets-A	fabric	ICP-MS —
9		shoes sheets-B	fabric(print)	ICP-MS —
10		shoes sheets-C	ceramics	ICP-MS —
11	bracelet	bracelet-A	ceramics	ICP-MS —
12		bracelet-B	ceramics	ICP-MS —
13	hot spring source	hot spring source-A	powder	ICP-MS γ
14		hot spring source-B	ceramics	ICP-MS γ
15	wall paper wall sheet paint	minus ion sheet	fabric	ICP-MS γ
16		wall sheet paint	paste	ICP-MS —
17	industrial products	Th added W electrode	metal	ICP-MS γ
18		ship bottom paint	powder	ICP-MS γ
19		muffler catalyst	alloy	ICP-MS γ
20		abrasive	powder	ICP-MS γ

(*1) ICP-MS: measurement of ^{238}U and ^{232}Th contents

γ : γ -ray spectrometry of ^{208}Tl , ^{212}Pb , ^{214}Pb , ^{214}Bi and ^{228}Ac

*2): ceramic button attached on the fabric

sources such as the commercial catalogues, the inspection on the internet by using key words “hormesis”, “minus ion” and others.

The consumer's goods containing NORM similar to each other were sold from different companies. Table 1 shows the list of consumer's goods containing NORM collected. These goods can be divided into 8 kinds of goods used in the daily living life and 4 kinds of goods used in the industry. The bedclothes are shown as a typical example in the Photo 1.



Photo 1 Bedclothes

2.2 Measurement of radioactivity

A part of each collected consumer's good was pulverized or burned in air and then was chemically treated. The standard method was applied to the chemical treatment, which has been already established and used for the analysis of environmental specimen in a number of institutes or companies in Japan.[2,3]

Radioactivity in the consumer's good was measured by two methods. First, the concentration of uranium and thorium were analyzed by an inductively coupled plasma attached to mass spectrometer (ICP-MS) and were converted to their radioactivities by using the value of specific radioactivity. Next, the radioactivities of daughter nuclides of uranium or thorium were measured by a GE-Li semiconductor detector.

2.3 Calculation of exposure

From the results obtained in the measurement of radioactivity, the exposures were calculated when four kinds of consumer's were utilized in the daily living life. A famous computer code “MCNP-4C” was applied to the calculation by using a relatively

conservative boundary as shown in Table 2. A schematic diagram of calculation system is shown in Fig.1. Table 2 shows the boundary conditions of calculation

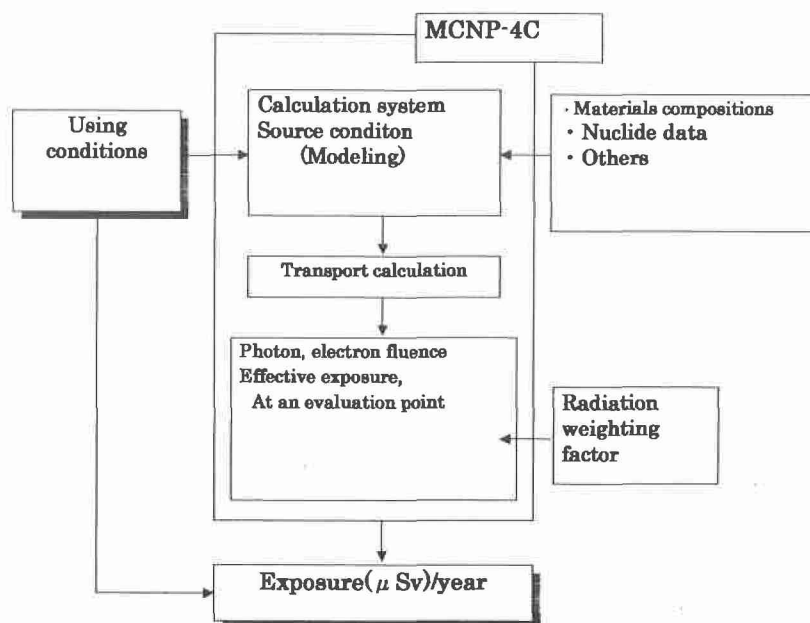


Fig.1 A schematic diagram of calculation system

Table 2 Boundary conditions used the calculation of exposure

NO	NORM goods	Radiation source	Exposure Scenario
a	Bedclothes	O:210x150cm U:210x100cm	8h/d
b	Underwear	26.6cm ϕ 65cm h	24h/d
c	Hot spring source	11x21 cm plane	30m/d At 10cm
d	Wall sheet	270x450cm plane	8h/d At 30cm

3. Results and discussion

3.1 Radioactive concentration of uranium, thorium and their daughter nuclides

Table 3 shows the concentrations of uranium and thorium in the consumer's goods, which were analyzed by an ICP-MS, together with the radioactive concentrations derived from the

measured results. Table 4 shows the radioactive concentrations of daughter nuclides measured by the Ge(Li) semiconductor detector.

Table 3 Radioactive concentration of uranium and thorium in the consumer's goods

No.	Specimen	ICP-MS					
		Concentration (mg/g)		Radioactive concentration (Bq/g)			
		U	Th	²³⁸ U	²³² Th		
1	bedclothes-o	0.0035 ± 0.00005	0.0025 ± 0.00004	0.043 ± 0.0007	0.01 ± 0.0002		
2	bedclothes-u	0.02 ± 0.0003	0.58 ± 0.003	0.26 ± 0.004	2.3 ± 0.01		
3	underwear	0.082 ± 0.0006	2.2 ± 0.04	1 ± 0.01	8.8 ± 0.16		
4	belly-warmer-A	0.076 ± 0.0005	2.1 ± 0.03	0.94 ± 0.006	8.5 ± 0.12		
5	belly-warmer-B	0.43 ± 0.003	8.4 ± 0.06	5.4 ± 0.04	34 ± 0.2		
6	wristband	0.00093 ± 0.00002	0.023 ± 0.0003	0.011 ± 0.0002	0.093 ± 0.0014		
7	socks	0.057 ± 0.0007	1.5 ± 0.03	0.7 ± 0.009	6.2 ± 0.14		
8	shoes sheet-A	0.025 ± 0.0004	0.66 ± 0.005	0.31 ± 0.004	2.7 ± 0.02		
9	shoes sheet-B	0.0069 ± 0.00012	0.16 ± 0.003	0.085 ± 0.0015	0.63 ± 0.012		
10	shoes sheet-C	0.034 ± 0.0006	0.68 ± 0.009	0.42 ± 0.008	2.8 ± 0.04		
11	bracelet-A	0.14 ± 0.002	3 ± 0.04	1.7 ± 0.03	12 ± 0.2		
12	bracelet-B	0.71 ± 0.003	18 ± 0.1	8.8 ± 0.04	71 ± 0.4		
13	hot spring source-A	2.8 ± 0.02	67 ± 0.8	34 ± 0.2	270 ± 3		
14	hot spring source-B	0.83 ± 0.014	20 ± 0.4	10 ± 0.2	81 ± 1.7		
15	wall paper	0.054 ± 0.0003	1.3 ± 0.02	0.67 ± 0.003	5.4 ± 0.1		
16	minus ion paint	0.047 ± 0.0005	0.77 ± 0.011	0.58 ± 0.006	3.1 ± 0.05		
17	Th added W electrode	0.00096 ± 0.00008	21 ± 0.3	0.012 ± 0.001	86 ± 1.3		
18	ship bottom paint	0.97 ± 0.014	20 ± 0.1	12 ± 0.2	81 ± 0.6		
19	muffler catalyst	0.27 ± 0.004	52 ± 0.5	3.3 ± 0.05	210 ± 2		
20	abrasive	0.016 ± 0.0002	0.17 ± 0.002	0.2 ± 0.003	0.7 ± 0.007		

Table 4 Radioactive concentration of daughter nuclide in the consumer's goods

No.	specimen	γ-ray spectrometry					
		radioactive concentration (Bq/g)					
		U-238 series		Th-232 series			— ¹³⁸ La
		²¹⁴ Pb	²¹⁴ B	²²⁸ Ac	²¹² Pb	²⁰⁸ Tl	
1	bedclothes	0.043 ± 0.0003	0.036 ± 0.0003	0.0091 ± 0.00025	0.011 ± 0.011	0.003 ± 0.00007	—
3	underwear	0.97 ± 0.003	0.83 ± 0.003	7.5 ± 0.01	8.5 ± 0.005	2.5 ± 0.003	—
4	belly-warmer tie	0.9 ± 0.004	0.75 ± 0.005	6.9 ± 0.02	8.5 ± 0.008	2.4 ± 0.005	—
13	hot spring source-A	33 ± 0.2	28 ± 0.2	250 ± 0.6	270 ± 0.3	83 ± 0.2	—
14	hot spring source-B	10 ± 0.04	8.9 ± 0.04	72 ± 0.2	82 ± 0.07	24 ± 0.04	—
15	minus ion sheet	0.69 ± 0.006	0.62 ± 0.007	5 ± 0.02	5.7 ± 0.01	1.8 ± 0.007	—
17	Th added W electrode	0.038 ± 0.009	0.061 ± 0.012	42 ± 0.05	25 ± 0.02	11 ± 0.01	—
18	ship bottom paint	13 ± 0.08	11 ± 0.08	86 ± 0.3	97 ± 0.1	29 ± 0.08	—
19	muffler catalyst	44 ± 0.2	37 ± 0.2	160 ± 0.6	220 ± 0.3	63 ± 0.2	—
20	abrasive	0.12 ± 0.002	0.11 ± 0.002	0.66 ± 0.005	0.57 ± 0.002	0.23 ± 0.002	0.21 ± 0.002

As shown in table 3, the concentrations of uranium and thorium varies in several orders. That is, the concentrations of uranium ranges from 0.00093 to 2.8 mg/g, which correspond to the range from

0.011 to 34 Bq/g. On the other hand, the concentrations of thorium scatters in the range from 0.0025 to 67 mg/g, which correspond to the variation from 0.01 to 270 Bq/g. It should be noted that the concentration of thorium is larger than the one of uranium. This reason likely comes from the fact the monazite including thorium is frequently used in the consumer's goods and the Clark number is larger in thorium than in uranium.

It can be seen in table 4 that the radioactive concentration of daughter nuclides is approximately equal to the ones of uranium and thorium except the result of Tl-208. This agreement suggests that the secular equilibrium holds between the uranium or thorium and daughter nuclide.

In Japanese regulation on the raw nuclear materials, the upper limits of radioactive concentration of uranium and thorium approved in solid and liquid are 370 and 37 Bq/g, respectively. Then, the results on the radioactive concentrations of uranium and thorium satisfy the regulation of raw nuclear materials in Japan. However, the present data are larger than the value, 1 Bq/g, cited in the Basic Safety Standard (BSS), which has been recommended as a basis of clearance level by International Atomic Energy Agency (IAEA)[4]. Thus, it is important to evaluate the exposure when the consumer's goods are italicized in the daily living life.

3.2 Evaluation of exposure

When the exposure is theoretically calculated during the utilization of consumer's good, it is necessary to certify the reliability of calculated value. Then, both experimentally measured and calculated values were compared with each other in the water bath shown in fig.2. The exposure rates were measured at the distance of 16, 32, 64, 85 and 90 cm from the radiation source, and then

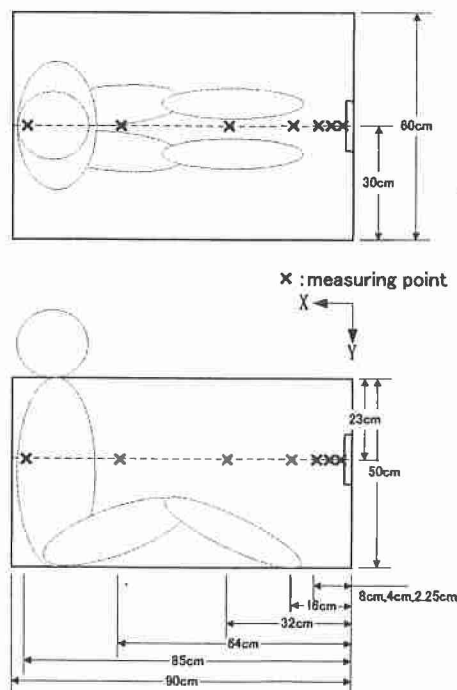


Fig.2 A schematic diagram of water bath for the comparison between the measured and calculated values

were compared with the calculated values. Fig.3 shows the comparison between both measured and calculated results. As shown in this figure, both data agree with each other, and it can be found that the calculation of exposure by "MCNP-4C" code was reliable.

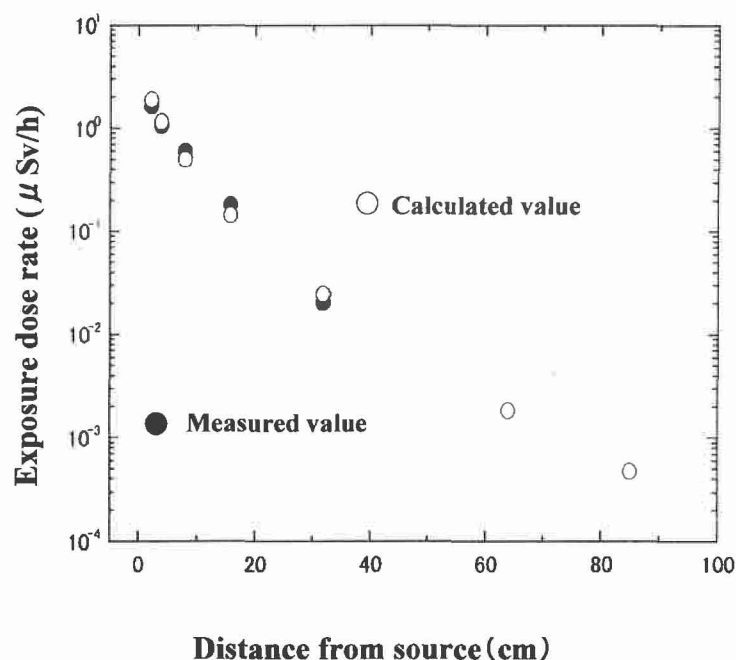


Fig.3 Comparison between the measured and calculated values

After certifying the reliability of calculation, the annual exposures were calculated when the bedclothes, underwear, hot spring source used at home and wall sheet are used under the conditions described in table 2. the results obtained are shown table 5.

Table 5 Calculated results of annual exposures

NO	NORM goods	Radiation source	Exposure Scenario	Results (μ Sv/y)
a	Bedclothes	O:210x150cm U:210x100cm	8h/d	110
b	Underwear	26.6cm ϕ 65cm h	24h/d	220
c	Hot spring source	11x21 cm plane	30m/d At 10cm	110
d	Wall sheet	270x450c plane	8h/d At 30cm	10

In the connection with the radiation protection in environment, there are some kinds of guidelines. In Japan, the exposure of people living around the nuclear facility is limited to be below 1 mSv/y except the exposure due to natural radiation, based on the ICRP recommendation [5]. Recently, EU radiation protection committee has recommended a value of 300 μ Sv/y as the exposure limit due to NORM goods [6].

As can be seen in table 5, the annual exposures in four cases are in the range from 10 to 220 μ Sv/y. These values are larger than 10 μ Sv/y, which has been recommended as a guideline of clearance level by IAEA, but are smaller than the 1 mSv/y and 300 μ Sv/y described above.

4. Conclusion

Twenty kinds of consumer's goods containing NORM were collected, based on various information sources. The radioactive concentration were measured by ICP-MS and Ge(Li) semiconductor detector, and the annual exposure were evaluated in four cases of daily living life based on the measured results by a computer code, MCNP-4C. The radioactive concentrations of uranium ranged from 0.011 to 34 Bq/g. On the other hand, the radioactive concentrations of thorium scatters in the range from 0.01 to 270 Bq/g. The annual exposure due to the utilization of NORM goods in the daily living life ranged from 10 to 220 μ Sv/y. They were smaller than 300 μ Sv/y, which has been recommended as the exposure limit due to NORM goods by EU radiation protection committee.

Reference

- 1) K. Tomabechi 「Uranium Glass」 Iwanami book service center (1995)
- 2) A Japanese standard on radioactivity measurement, series 13 「Specimen preparation method for instrumental analysis including Ge(Li) semiconductor detector」 (in Japanese), (1982)
ibid series 16 「Sampling method of environmental specimen」 (in Japanese), (1983)
- 3) "RSIC COMPUTER CODE SELECTION, MCNP4, Monte Carlo Neutron and Photon Transport Code System," CCC-200A/B(1991)
- 4) International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources Safety Series No.115(1996)
- 5) Japanese Law [Law on raw nuclear material, nuclear materials, nuclear reactor]
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4.2 Hot Spring and Radioactivity

温泉と放射能

Kimiko HORIUCHI

堀内公子

Otsuma Women's University

大妻女子大学

〒206-8540 Karakida 2-7-1 Tama, Tokyo 206-8540

Tel:042-339-0089 Fax:042-339-0044 E-mail:horichi@otsuma.ac.jp

1. はじめに

温泉とは読んで字の如く温（あたたか）い泉（いずみ）である。しかし昔から温かくは感じないが色や臭い、感触等で普通の水と明らかに区別の出来る水が地中から湧き出すことが知られていた。現在では温度だけからではなくこれら特別の水を普通の水と区別して温泉と呼ぶことが温泉法¹⁾で定められている。

2. 温泉の成立要因²⁾

温泉が湧出するためには、①熱源、②湧出径路、③水源の三つが必要であって、このうち一つが欠けても温泉は成立しない。放射線は温泉成立の必要条件の総てに関係を持っている。

2-1 温泉の熱源

地球は宇宙の塵が重イオン反応や核融合反応を何十億年もの間繰り返した末に誕生した。そのため、誕生以来 46 億年経った現在でも多種類の放射性元素が存在している。

地球内部は高温を示し、地表へ向かって熱の流れがある。その熱源の一つに岩石中に含まれるウラン、トリウム、放射性カリウム等の長寿命放射性元素の原子核崩壊に伴って発生するエネルギーがある。

一方地球内部からの熱の流出は①火山活動、②温泉・地熱、③地震波動、④地下から地表を通過して上昇する熱（地殻熱流量）等によっている。従って温泉の熱源の一部は地球の創成期から地殻に含まれる天然放射性元素に由来しているといえる。地球誕生以来地球内部で岩石の中に貯えられたウランやトリウム等放射性元素の崩壊熱エネルギー量はかなり大きいと見積られている。

2-2. 温泉の湧出径路²⁾

温泉が地表まで上昇してくるためには、地盤に開口割れ目が必要になる。地殻は地質時代より、引っ張り、圧縮、火山、地震、隆起、陥没、その他の原因による変動が加えられ、断層や、大小の割れ目が生じる。こうした割れ目（開口亀裂帯）は温泉の上昇に最も都合の良い径路となる。この割れ目が網目状に発達すると、多量の温泉を蓄える温泉貯留層の役目も果たす事が出来る。

開口亀裂帯は、温泉の上昇と同時に温泉の熱源となっている放射性元素ウラン、トリウム等の崩壊生成物ラジウム、ラドン等が温泉水に溶解したり、温泉水の移動によって運ばれるなどして地表部に到達し、崩壊生成物の各種 γ 線を放出する。

2-3. 温泉の水源

温泉水の大部分は、天水が地下に浸透して地下深部で温められて、それが再び地表あるいは地表近くにまで上昇して来た地下水と考えられている。よって地下水の動きを知る必要があるが、水の天然のトレーサーとして水素の放射性同位体トリチウムが用いられている。トリチウムを含んだ降水が地下に浸透し、熱源に近い所で加熱され、循環対流によって他の開口亀裂帯を通って上昇し、温泉として湧出した時、温泉水中のトリチウム ($T_{1/2}$: 半減期 12.6 年) を測る事により水循環の速度を知ることが出来る。また、有望な温泉を得るためには、亀裂帯などによって、地下水の涵養機構が発達している循環性の地熱貯留層を探し出す必要がある。

3. 温泉探査の手段としての放射能

温泉の開発適地は地下からマグマや熱水が上昇し、利用量が満足出来る水循環条件を持つことである。浅い位置でこの条件に合う場合は温微が明瞭な地点で、既に温泉開発がなされている。よって現在では地下 1000m 以上といった深い位置にこの条件を見出すことが課題とされている。

3-1 ラドン

地層の開口割れ目では揚水能が一般の透水層の 10 倍以上にもなり、また反復揚水による水みちの発達で降水や表層部からの水の浸入量は漸増する。従ってまずは温度の十分な開口割れ目を見出すことが温泉探査の目的となる。開口割れ目を通してラドンが上昇することは早くから知られていて、地表面にあけた小孔中の気体の放射能測定から埋没断層の位置を求める方法は数多く検討されて来た。

3-2 γ 線

1950 年代の末に NaI(Tl)検出器による γ 線スペクトロメトリーが可能になると、ラドンを直接測定せずにラドンと同一行動をしている崩壊生成物の ^{214}Bi の γ 線 1.76MeV の計測が行なわれるようになった。放射能による温泉探査は正確な結果が得られるとして信頼度が高く、次第に地表から迅速に検出し、分析が容易なエネルギーの高い自然 γ 線 $^{208}\text{Tl}(2.61\text{MeV})$, $^{214}\text{Bi}(1.76\text{MeV})$, $^{40}\text{K}(1.46\text{MeV})$ 等の 3 核種を指標とした表層地質の解析が始まった^{3,4)}。

次いでこの 3 核種の一次ガンマ線量は表層の核種の含有量に比例し、その含有量は地層毎に固有値を持つという現象が見出されると、アメリカ、カナダ、ヨーロッパ等の広い平坦な地域では検出器をヘリコプターに乗せて高度 30m をゆっくり飛びいわゆるエアボーン (ヘリボーン) により、表層地質の解析が行われるようになった⁵⁾。しかし地形の起伏が激しく、地層が細かに変わる我が国では自動車や携帯計測器を用いたカーボーンやマンボーンでの精密な探査が極め手の鍵ともなる。

4. 温泉水中の放射能

4.1 ラジウム

我が国のラジウム測定の実史は古く、1915 年パナマ万国平和博覧会を契機に刊行された「The Mineral Springs of Japan」に我が国初の温泉のラジウム測定値が記載されている。全国 477 ヶ所の温・鉱泉と 15 の石油及び天然ガス地域に於ける試料水のラジウム測定データが 1940 年に発表された。これはラジウム測定データとして最もまとまったものであり、時節柄我が国のラジウム資源を鉱泉に求めるには経済的価値の低いことも述べられている。

液体シンチレーションカウンター(以下 LSC)が開発されると、同一試料中のラジウム、ラドンを測定した結果が 119 の温泉につき報告された。従来、温泉水中のラドンは、親核種ラジウムとは非平衡に多く存在することが知られているが、ここでもそれが証明されている。

4.2 ラドン

温泉水の中には主としてラドン(Rn: ^{222}Rn)とその同位体トロン(Tn: ^{220}Rn)が存在するが、半減期が 56 秒と短いトロンのフィールドでの検出はなかなか困難であり、データはあまり多くない。

鉱泉水等環境試料中のラドンの存在は 1903 年に確認され、同時にラドンによる鉱泉水の治療効果の可能性も示唆されている。世界最初の鉱泉水中のラドン測定器はエングラ・ジーベキング泉効計で、我が国でもこの泉効計により本邦最初の温泉の放射能値が報告された。よって、我が国の環境放射能の研究は温泉水中のラドン濃度の測定から始まったとも言えよう。

現在、温・鉱泉水中のラドンは温泉の分析項目の一つであり、温・鉱泉水中のラドン発見以来研究と利用との二つの面から測定され今日に至っている。ラドンは、放射性のガス成分であり、温・鉱泉水中ほとんど他の溶存化学成分との相関はなく単独に存在している。我が国には多種類の泉質の温泉が湧出しているが、それらの中で放射能泉は溶存化学成分プラス放射能というかなり特異的な存在と考えられる。

5. 鉱泉の定義¹⁾

現在我が国の温泉は環境庁自然保護局の所管行政で、国民の保健休養と同時に自然環境の保護も目的としている。長寿社会へと移行し、余暇が増加しつつある我が国で温泉の果たしうる役割は極めて大きいものと期待されている。温泉は温泉法によって保護され利用の適正化と公共の福祉の増進が図られており、泉質は昭和 26 年制定され、53 年に改定された鉱泉分析法指針により分析すべき項目と分析方法が定められている。

指針によれば、放射能による鉱泉の定義は

ラドン(Rn) $20 \times 10^{-10}\text{Ci}$ 以上、常水との区別、鉱泉と認める濃度 (5.5 マッヘ単位以上)

ラジウム塩(Ra として) $1 \times 10^{-8}\text{mg}$ 以上

ラドン(Rn) $30 \times 10^{-10}\text{Ci}$ 以上、特殊成分を含む療養泉 (放射能泉) (8.25 マッヘ単位以上)

である。しかし療養泉 (放射能泉) は温泉法の対象ではなく、温泉医学に於ける臨床経験から医療効果の期待出来る温泉と言う考え方である。

我が国の鉱泉の基準は常水と区別する鉱水の限界値を定めたドイツのナウハイム決議(1911 年)に準じており、温泉の放射能の単位も H. Mache により提案されたマッヘ単位(1 マッヘ M.E.=水

1 l に含まれるラドンによる飽和電離電流が 10^{-3} e. s. u. のとき)が用いられて来た。放射能による鉱泉・放射能泉の定義をベクレル表示 ($1\text{Bq}=27\text{pCi}$, 0.074M.E.) に換算すると、それぞれ 74Bq/l および 111Bq/l 以上となる。

6. 温・鉱泉分布図による、我が国の放射能泉の特徴

我が国は世界で最も泉質の種類に富んでいる。先年まとめられた温・鉱泉分布図⁶⁾によると、我が国の放射能泉は全体の 7.73%(泉質の明記されているもの：総数 3,659)にあたり、全国の放射能泉の数を都道府県別に集計すると花崗岩分布地帯の多いフォッサマグナの西側、主として中国地方に多く分布している。また泉温については 25°C 以下の冷鉱泉が多く全体の 86.5%をしめている。中でも $15\sim 20^{\circ}\text{C}$ が最も多く、地下水の温度領域に近い。

我が国の放射能泉の特徴をまとめてみると、それ以外の泉質の温泉に比べて湧出量は $1/2\sim 1/3$ と少なく、液性は中性から弱いアルカリ性を示し、ラドン以外ほとんど他の溶存化学成分を含まない単純冷鉱泉が主体である⁷⁻⁹⁾。

7. 放射能泉の医療効果

古来、放射能泉はリウマチのリハビリや鎮痛作用等の医療効果が認められ、温泉治療に利用されて来た。温泉治療に利用されているのは、ほとんどラドン泉である。ガス成分のラドンは呼吸により体内に入り、体内に入ったラドンの一部は崩壊し、次々と半減期の短い別の核種に変化して体内に残る。ラドンの崩壊生成物はその濃度と体内にある時間に比例して多くなるが、やがてラドンの崩壊生成物に固有の生物学的半減期によって排泄される。

ラドンの医学効果は臨床医学的に自律神経の鎮静、内分泌系に適度の刺激を与えることによるホルモンや代謝異常の調整、鎮痛、消炎作用などと言うことが出来る。ラドン濃度を増すと鎮痛効果はあがるが、心血管や植物神経系統に反作用が起きるので、他の温泉成分と同様、適切な利用が大切である。また放射能泉の効果は単に放射能だけによるのではなく他の溶存化学成分や泉温等との相乗効果である。

8. 終りに

放射能泉は古くから多くの人達によって農繁期の疲れを癒したり、リウマチや怪我のリハビリ等で利用され続けてきた。世界に誇りうる数少ない天然のエネルギー資源：温泉は限り在る資源でもある。放射線関係の研究の進展につれ、放射能泉と人体の係わり合いのメカニズムも次第に明らかにされつつある。放射線の危険性を正しく理解した上で、放射能泉が愛され有効利用されて行く事が期待される。

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4.3 Natural Radioactivity Distribution Images and Their Practical Uses

自然放射能分布イメージとその活用

Chizuo MORI

森 千鶴夫

Aichi Institute of Technology

愛知工業大学

〒470-0392 愛知県豊田市八草町八千草 1247 E-mail cmori@sc.starcatt.ne.jp

中部原子力懇談会 (Chubu Atomic Power Conference)

〒460-0008 名古屋市中区栄 2 丁目 10-19 Tel: 052-223-6615

The general public does not well know that natural radioactive substances are contained in all natural materials including foods. We obtained the images of natural radioactivity distributions in vegetables, meat, stones, or ornaments by exposing them on the Imaging Plate developed by Fuji Photo Film Co. Ltd. in a lead shielding box for about one month. We made brochures named "Natural Radiations through Eyes" or "Environmental Radiations through Figures", etc. which include the images mentioned above. We sometimes give lectures and experiments on radiations or energy to the middle or high school students using the brochures. After the lectures, most of the students say that the images are effective to realize the presence of natural radioactivity even in our bodies.

1. はじめに

自然放射性物質は、岩石はもちろんのこと、野菜や肉などの食材、装身具などの全ての材料に含まれていることは、一般人にはまだ十分に認識されていない場合が多い。そのようなこともあって、放射線、放射性物質と聞くと、ただその言葉だけで、いたずらに恐怖心を抱くことがある。しかし、身近な食材などにおける自然放射能の分布状態を可視化した像が得られれば、一般の人々にとって、自然放射性物質、自然放射線を実感的に理解する上で大いに役立つと思われる。このような意図をもって野菜や肉などに含まれる自然放射能分布の像を取得した。本報告ではその取得の方法と取得したいくつかの像を紹介すること、および、これらの像を用いて学生や一般人に対して行なっている PA 活動を紹介する。

2. 放射能分布像の取得

2.1 露出時間の推定

Fig.1 に示すように、富士写真フィルム㈱が開発したイメージングプレート (IP) は、X線フィルムの 100~1000 倍の非常に高い感度を持っている¹⁾。これだけの感度があれば私達の身の回りの品々に含まれている自然の放射能の分布状態が得られるのではないかと考えた。

自然放射能イメージングの取得に役立つ自然放射線は、野菜などの食材においては、ほとんどが K-40 からの β^- 線である。K-40 は半減期 1.2×10^9 年で、 β^- 壊変の割合は 89% である。K-40 は食材や人体中には、0.02~0.1Bq/g の割合で含まれている。岩石や装身具においては、やはりほとんどは K-40 であるが、ウランやトリウム系列からの α 線、 β 線、 γ 線もイメージングの取得に役立つ。

Fig.1 の IP の最低検出感度は C-14 に対して約 $(0.15 \text{ dpm/mm}^2) \times 60 \text{ 分} \times 18 \text{ 時間} = 160 \beta / \text{mm}^2$ である。C-14 の

β 線の平均エネルギーは約 50keV であるから、単位面積当りのエネルギー付与は $8 \times 10^3 \text{ keV/mm}^2$ となる。この β 線の最大エネルギーは 155keV で、その飛程は IP の厚さに換算すれば約 $100 \mu\text{m}$ であるので、IP の輝尽性発光体の厚さ $150 \mu\text{m}$ m の中で全てのエネルギーを失うことになる。

一方、野菜などに含まれる K-40 の含有率、 β 線の平均エネルギー 440keV、飛程 0.17 g/cm^2 、等を考慮すれば、 $150 \mu\text{m}$ の厚さの輝尽性発光体の付与されるエネルギーは、 $(0.26 \sim 1.3) \times 10^{-2} \text{ keV/mm}^2 \text{ s}$ となる。したがって、野菜などからの β 線を検出するのに必要な露出時間は $8 \times 10^3 \text{ keV/mm}^2 / (0.26 \sim 1.3) \times 10^{-2} \text{ keV/mm}^2 \text{ s} = 7.2 \text{ 日} \sim 36 \text{ 日}$ となる。

2.2 遮蔽箱の必要性

前述の K-40 β 線の輝尽性発光体 (厚さ $150 \mu\text{m}$ 、密度 3) へのエネルギー付与は $(0.92 \sim 4.6) \times 10^{-12} (\text{J/kg})$ である。一方、バックグラウンド放射線の単位時間当たりの吸収線量は、年間約 1 mGy であることを考慮して、 $3.2 \times 10^{-11} (\text{J/kg})$ となる。したがって、バックグラウンド放射線を 1 桁以上低減する必要があることが分かる。

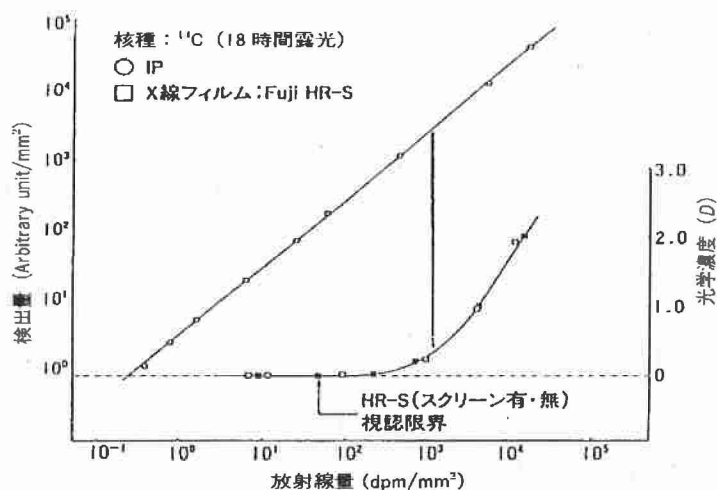


Fig.1 Comparison between the radiation sensitivity of Imaging Plate (IP) and that of X-ray film

鉛の厚さ 10cm の遮蔽箱の内面をカドミウムと銅で内張りし、さらにその内面を数 mm の厚さの亚克力樹脂で覆った遮蔽箱はバックグラウンド放射線を約 1/20 に低減する。

2.3 潜像の減衰と長時間露出

IP に蓄えられた潜像が、読取時間までに減衰する特性に関しては多くの報告がある²⁾。この特性を、いろんな放射線源で得た結果を Fig.2 に示す。

5 日後の潜像の強度を PSL(5) として、この値に対する相対強度をフィッティングによって求めれば、次式が得られる。

$$\text{PSL}(t)/\text{PSL}(5)=0.48\exp(-0.32t)+0.52\exp(-0.0385t)\equiv R(t)$$

この曲線を図中に点線で示す。

露出が長時間 T にわたる場合に蓄積された潜像が読取られる強度 $I(T)$ は次式で表される。

$$I(T)=\int_0^T R(t)dt=\frac{a}{b}\left[1-\frac{1}{bT+1}\right]+\left[\frac{1-a}{c}\right]\left[1-\frac{1}{cT+1}\right]$$

ただし、 $a=0.4, b=0.3, c=0.03$ 、 T : 時間 (h) である。

得られた $I(T)$ の結果を Fig.3 に示す。無限の長時間露出した場合に得られるであろう強度の約 75% が 30 日の露出時間で得られる。したがって、実質的には 1 ヶ月以上の露出はあまり意味がないことが分かる³⁾。

3. 得られた像の例

野菜や肉などの場合は遮蔽箱の中でほぼ一ヶ月間の露出が必要であるので、腐敗や変形に注意する必要がある。遮蔽箱から取り出したあとは出来るだけ早く潜像を読み取るのがよい。岩石などの場合の露出は数日でよく、場合によっては遮蔽箱を必

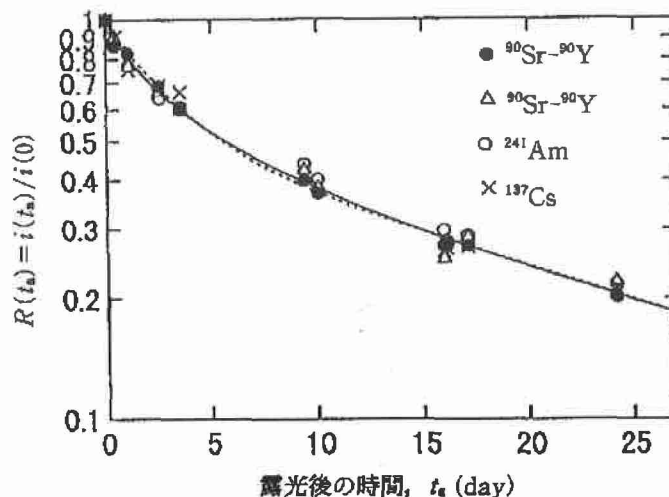


Fig.2 Fading characteristic of the latent image stored in Imaging Plate

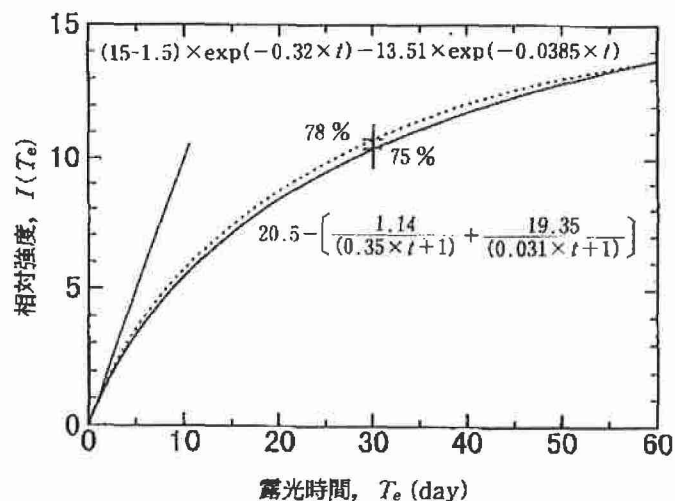


Fig.3 Effective intensity of the image read out after long time exposure

要としない。

Fig.4 の左側に、豚肉、バナナのタテ切り、横切り、およびショウガからの K-40 β 線に

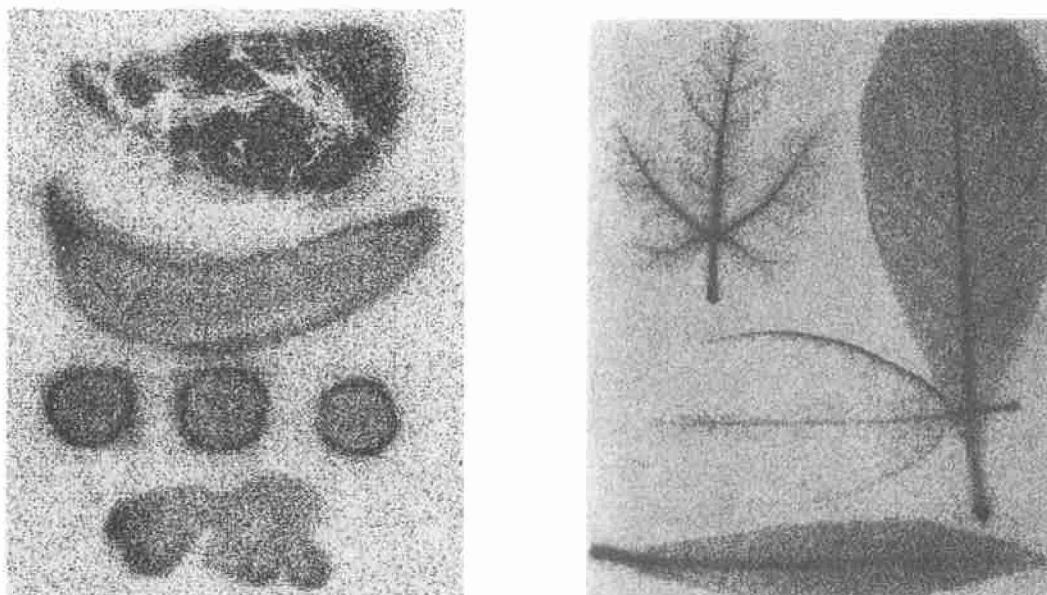


Fig.4 Natural radioactivity distribution images of pork, banana, ginger (left side) and leaves (right side)

よる像を示す。豚肉では、たんぱく質の部分に K-40 が多く、脂肪の部分にはほとんど含まれない。バナナは皮の部分に多い。右側には木の葉の像を示す。葉脈の部分に多いが、葉の端に多く出ているものもある。

次に装飾品やめがねの像を Fig.5 に示す。陶磁器製のブローチやカリウムガラスにはカリ

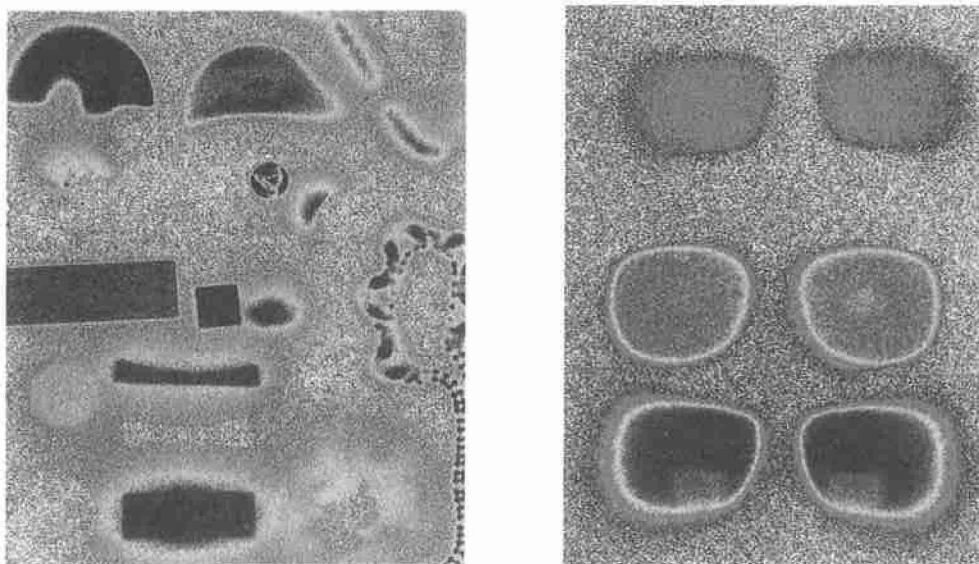


Fig.5 Left: ornaments, glass ware, pottery ware. Right: glasses

ウムが多く含まれていることが分かる。ナトリウムガラスにはほとんどカリウムは含まれていない。めがねでは、サングラスはナトリウムガラスらしくほとんど放射性物質がない。しかし、近眼用めがねや老眼鏡などはカリガラスで K-40 が含まれている⁴⁾。しかし。既知の K-40 含有量の試料による校正測定をし、計算によって眼の放射線被ばくを計算してみると、めがねから受ける放射線被ばくは、日本の低地における自然放射線程度であることが分かる。

Fig.6 に花崗岩の小片の写真(左)とその放射能分布像(右)を示す。黒い部分は K-40 からの β 線によるものである。この白黒の像ではあまり見えないが、カラーでは、雲母に含まれるウランやトリウムからの α 線による強い点状の像が赤く見える。

4. パンフレットの作成とその活用

取得した像を含めて自然放射線に関するパンフレットを作成した。Fig.7 にパンフレットの表紙を示すが、左側は「目で見える自然放射線」で、自然放射線を可視化した様々な図を中心に編集している。右側は「図で見る環境放射線」で、環境放射線強度などを図にして編集している。これらを使って、中学校や高等学校へ放射線やエネルギーなどをテーマにした出張授業を行なっている。授業は講義と実験から成っている。授業の最後にとる生徒のアンケートによれば、野菜や肉の中に放射性物質があることを始めて知ったとか、直接見る事が出来て驚いたとかが述べられていて、像による視覚的な授業の効果は大きいと思われる。

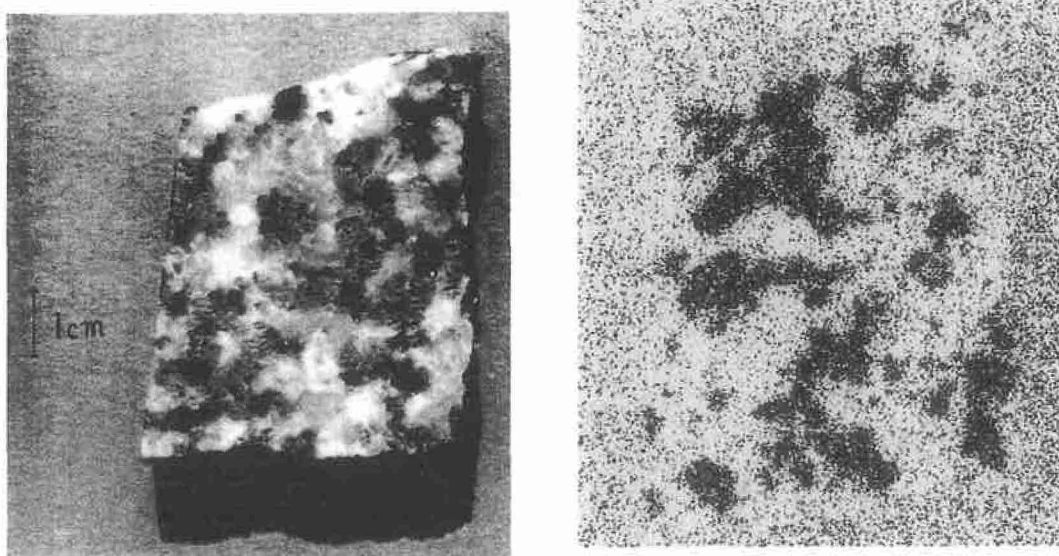


Fig.6 Left: photograph of a piece of granite. Right: Radioactivity distribution image of the granite.

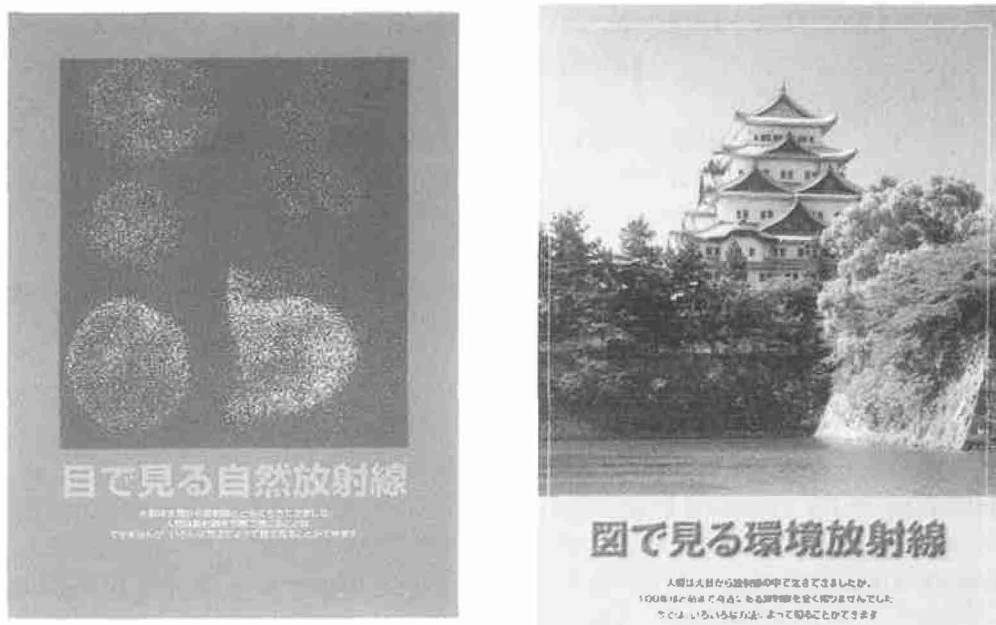


Fig. 7 Brochures containing natural radioactivity images. Left: "Natural radiations looking through eyes". Right: "Environmental natural radiations understanding through figures"

5. まとめ

放射線に対して極めて感度の高いイメージングプレートを用いて、遮蔽箱の中で野菜、肉、木の葉、装飾品、岩石などを露出し、これらに含まれる自然放射能の分布像を得た。これらを使ってパンフレットを作成し、中学校、高等学校、ならびに一般の人々に自然放射性物質が、私達の身体を含む全ての物質に存在し、どのように分布しているかを可視的に示すことに役立てている。アンケートの結果では、生徒や一般の方々において、放射線に対する無理解に伴う必要以上の恐怖感を少なくすることができていると思われる。

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4.4 Radon variations in an active landslide zone from Himalaya: A preliminary study

V.M. Choubey, S.K. Bartarya and R.C. Ramola¹

Wadia Institute of Himalayan Geology,
33 General Mahadeo Singh Road, Dehradun- 249001, India
Email: vchoubey1@rediffmail.com

¹Physics Department, HNB Garhwal University, Badshahi Thaul Campus, Tehri
Garhwal - 249199, India

Abstract

The radon concentration was measured in soil and water samples from an active landslide zone in the Garhwal Himalaya. The landslide is compound in nature i.e. slump in the crown portion and debris slide in the lower part. The measured radon concentration varies from 3.1 to 18.3 Bq/l in water whereas in soil/debris samples it varies from 2.3 to 12.2 kBq/m³. The crown portion (upper portion) show higher radon values in comparison to distal portion. The higher radon concentration in crown portion may be because of the failure plane of landslide associated with high fracturing and crushing whereas, increased porosity of debris of slide does not allow radon to accumulate in soil and water in the lower portion.

Introduction:

Radon is omnipresent radioactive gas and used as a natural tracer in hydrology, geology and geophysics. Dynamic phenomena occurring in the rock mass due to earthquake, volcanic activities, tremors, rock blasting and landslide may be related to outbursts of gas and changing radon concentration (Trique 1999; Wysocka 1999; Kies et al., 2002). Schmid and Wiegand, 1998 have suggested that due to traffic vibration the topmost soil layers lose radon to the atmosphere and as a result the upward migration increased. High radon concentration is generally considered with uranium deposits or with highly sheared fault/thrust zone (Choubey and Ramola, 1997; Purtscheller et al., 1995; Steele et al., 1982).

The investigations in Himalayan region shows that the concentrations of radon in soil and groundwater are largely controlled by the tectonic processes, types of rocks and geohydrological characteristics of the rock mass (Choubey et.al., 1999, 2001). The Himalayan area is prone to landslides due to geodynamic processes, which have been considered as a source of radon (Purtscheller et. al., 1995). The indoor measurement in Koefels landslide (Tyrol, Austria) shows high concentration of radon in houses located on old landslide deposits (Ennemoser et.al., 1994). However, more study is needed to find out the relationship between radon emanation in soil and water with landslide activities. The use of radon as a precursor of landslides is also yet to be investigated in details.

The aim of this study is to analyze the radon characteristics of soil and water in an active landslide zone in relation to its applicability in the study of geological disasters. Keeping in view this, radon was measured in the soil and spring water in and

around an active landslide zone located along Pindar River between Hirmani and Mathera villages in the Chamoli District of Uttaranchal, India. The relationship between landslide activity and observed radon concentration in soil and spring water is reported in this paper.

Radon Measurement Technique:

The radon was measured in water (spring and seepage) and soil/ debris in an active landslide zone. About 750 ml of water sample was taken in radon-tight reagent bottle of 1 litre capacity connected in a close circuit with a ZnS coated detection chamber through a hand operated rubber pump and a glass bulb containing CaCl_2 to absorb the moisture (Fig. 1b).

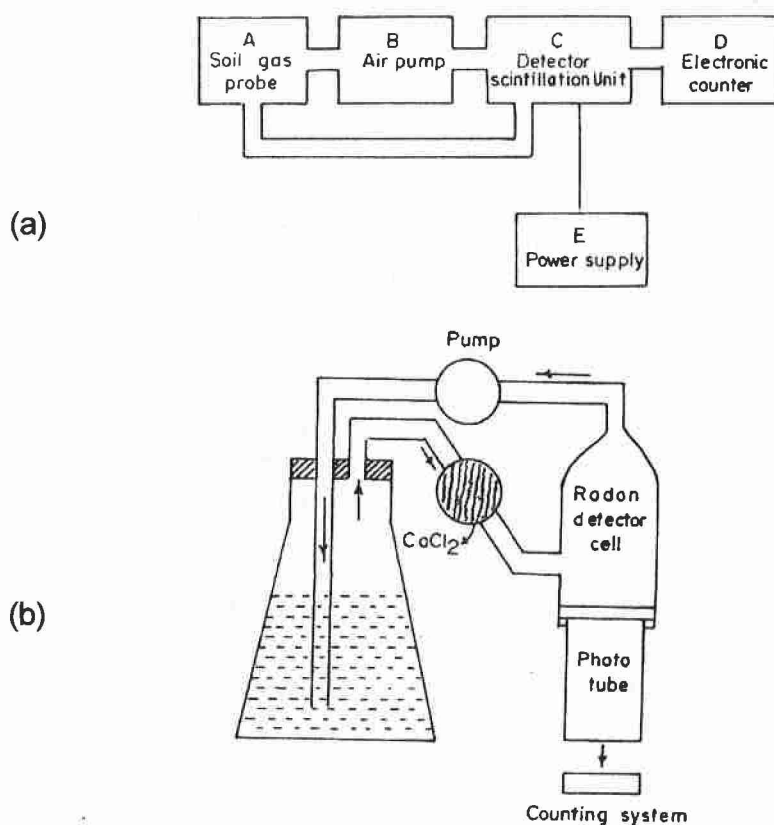


Fig. 1 (a) Block diagram of close circuit technique for radon measurements in soil, (b) Apparatus for the measurement of radon in water.

Air was then circulated in close circuit for a period of ten minutes till the radon forms a uniform mixture with the air and the resulting alpha activity was recorded. The detector was then isolated by clamping both the ends and observations were taken after four hours. A time gap of four hours is necessary to allow alpha emitting daughters of radon, RaA and RaC , to come in equilibrium with their own daughters. Resulting counts were then converted in Bq/l by using an appropriate calibration factor (Choubey et al., 2000).

For soil gas radon measurement, auger holes each 50 cm in depth and 6 cm in diameter were left covered for 24 hours so that the amount of radon and thoron became stable. The soil-gas probe was fixed in the auger hole and formed an airtight compartment (Ghosh and Bhalla, 1981). The rubber pump, soil-gas probe and alpha detector were connected in a close circuit (Fig. 1a).

Air was circulated through a ZnS coated chamber for a period of ten minutes till the radon formed a uniform mixture with the air. The detector ends are clamped and counts are recorded after a time gap of four hours. The resulting number of the alpha counts is then converted into Bq/m³ by using the calibration factor 1 count/min = 66.30 Bq/m³, determined by Choubey et al. (2000) under similar conditions.

Gneissic rock samples close to the landslide zone were also analyzed for their trace elements, including uranium using the sequential X-ray spectrometer (Siemens SRS-3000).

Results and Discussion

The landslide is located on the left bank of the Pindar river in Chamoli district of Garhwal Himalaya. The slide is compound one i.e. slump in the upper portion and slide in middle part of the slide. The slide consists of three parts: *crown portion* (slide scar area consist of fractured and jointed freshly exposed rock mass), the *middle slide mass* and *toe portion* having accumulated slide debris (Fig. 2). The radon concentration was measured in waters of spring and seepages and from soil of the crown, middle portion and toe portion of the slide. The main rock mass consist predominantly of schist and alternating bands of gneiss of the Saryu formation of Almora Group of Precambrian age (Valdiya, 1980). The rocks exposed in the scar area are highly fractured, jointed and sheared. The intensity of fracturing in landslide zone differs from place to place but was high in upper portion of the slide.

The observed radon concentrations in soil and groundwater of the land slide zone are given in Table 1. Soil gas and groundwater radon concentrations were found to vary from 2.3 Bq/m³ to 12.2 Bq/m³ and 3.1 Bq/l to 18.3 Bq/l, respectively (Table 1). It was observed that for both the cases radon concentration is higher in crown area and lower in the toe area of the landslide.

The most important pre-requisite for high radon concentrations in soil air and water are uranium and radium contents, permeability of the soil or rock and the possibility for high emanation rates, determined by certain moisture content as one of the most important factors (Gundersen et al., 1992). Nearly all these factors are closely related with deformations. Since landslide are localized phenomenon and no fault has been recorded with in the landslide zone other possible sources for the enhanced radon in crown area are need to consider. Surbeck (1992) related radon distribution in rock slides to permeabilities but not as source. The other landslides which have shown high radon concentration are those of Langtang Himal of Nepal and Koefal slide of Tyrol (Austria). The present slide show somewhat similarity to the above slides. Though, radon concentrations are far below the values reported in these slide but the present slide show enhanced radon towards crown portion. The high degree of fracturing in the crown portion near the base of the slide scar where possibilities of emanation and

circulation pathways are best exist result in relatively high radon towards slide scar in crown portion. According to Putscheller (1995) the material of the landslide meet all the prerequisites for an elevated emanating power and diffusion coefficient by their heavy fracturing and crushing down to grain and sub grain size, resulting in an increase of active surface porosity.

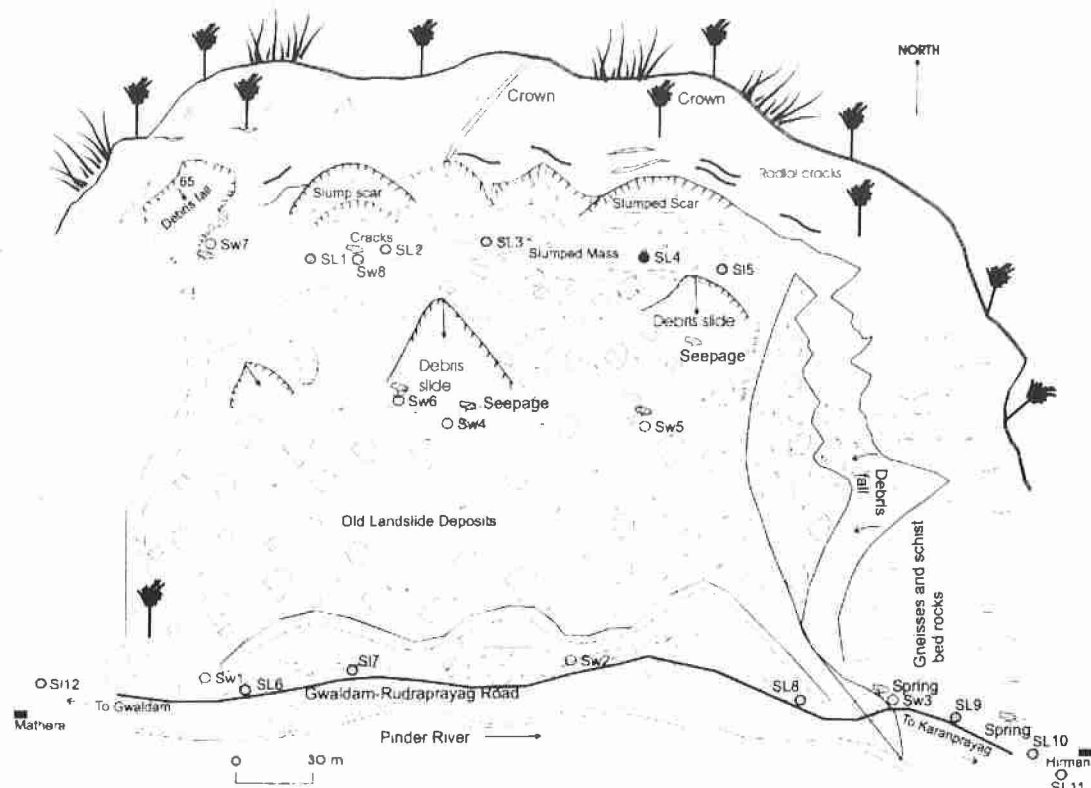


Figure 2. Map of the landslide zone showing geomorphic features and location of the samples for radon measurement. (open circle=water, solid circle= soil samples)

Table 1. Radon concentration in soil and water in the land slide area

Sample No.	Location of sample in slide area	Radon
<u>Soil samples</u>		kBq/m ³
SL1,2,3,4,4a,5,	Crown area (upper portion of slide)	10.3,9.1,12.2, 11.7,4.7,12.1
SL6,7,8,9,10,11,12	Toe area (lower portion of slide)	2.8,2.3,6.3, 5.7,10.5
<u>Water samples</u>		Bq/l
SW 7, 8	Crown area (upper portion of slide)	15.3, 18.3
SW 4,5,6	Middle portion of slide	8.1,9.8,8.4
SW 1,2	Toe area (lower portion of slide)	18.3,3.1

The recorded high value of radon in crown area (slide scar) of the present slide is mainly because of the steeply dipping crosscutting and partly open joints which provide good permeabilities and circulation pathways for soil air. Therefore, the highest radon emanations in the landslide zone occur within landslide scar area where strongly fractured gneisses and schist are exposed. The contribution of radon due to presence of uranium/radium in the debris material is almost negligible. This is reflected in the geochemical analysis of gneisses rocks which are present close to the landslide zone. The uranium concentration varied from 1.8 to maximum of 3.9 ppm. The minimum, maximum and average values of trace elements are given in Table 2.

The low concentration of radon both in soil and water in the toe portion of the landslide may be due to the high porosity and permeability of the debris material, which does not allow radon to accumulate in the soil and water. Wu et al., 2003 observed that the permeability/porosity of the subsurface material and fracture network plays an important role in escape of radon to the atmosphere.

Table 2. Trace element data of rock samples (all values in ppm)

Elements	Minimum	Maximum	Average
Co	9.0	21.0	13.9
Ni	18.0	65.0	34.6
Cu	16.0	39.0	28.8
Zn	59.0	89.0	74.7
Ga	7.0	16.8	12.7
Pb	17.3	34.7	24.5
Th	7.9	22.1	14.2
Rb	78.0	219.0	133.0
U	1.8	3.9	2.9
Sr	33.0	104.0	71.6
Y	18.4	42.1	33.3
Zr	99.0	351.0	218.9
Nb	4.8	16.3	11.5

The preliminary results of the radon measurement in an active landslide zone from the Lesser Himalayan region suggests that the enhanced radon concentration in the landslide zone may be indicative of instability of the slope. Possibly the landslide scar and failure plane of an active landslide may be the source of enhanced emanation of radon in an active landslide zone (Figure 2). The enhanced radon emanation is because of the high active surface area and circulation pathways due to fracturing and crushing of rocks near the landslide scar. However in the toe portion increased porosity of the colluvial debris increased the diffusion of radon in the atmosphere and thus resulting in low radon concentration. However, more measurements from other active landslide would confirm the above inference.

The continuous monitoring of radon in the landslide prone area may also be helpful to predict the occurrence of landslides in the area. Keeping in view the active

nature of Garhwal Himalayas, the measurement of radon in soil and groundwater will be helpful to locate the landslide prone areas and thus to minimize the risk to the local habitants.

Acknowledgements

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4.5 Cosmic rays in space

宇宙空間と放射線

Kazunobu FUJITAKA

藤高和信

National Institute of Radiological Sciences

独立行政法人 放射線医学総合研究所

Project Leader, Space Radiation Protection

宇宙放射線防護プロジェクトリーダー

ABSTRACT

Cosmos is a mysterious space by which many researchers are fascinated for many years. But, going into space means that we will receive extra exposure due to existence of cosmic rays. Cosmic rays are mainly composed of highly energetic protons. It was born in the last stage of stellar life. Understanding of cosmos will certainly bring right understanding of radiation energy, or energy itself. As no one could see the very early stage of cosmic rays, there is only a speculation. But it is better to speculate something based on certain side evidences, than to give up the whole. Such attitude shall be welcomed in the space researches.

Anyway, cosmic rays were born in the last explosion of a star, which is called as Super Nova. After cosmic rays are emitted from the Super Nova, it will reach to the human surroundings. To indicate its intensity, special unit of "dose rate" is used. When a man climbs a mountain, cosmic ray intensity surely increases. It doubles as he goes up every 1500m elevation. It was ascertained by our own measurements. Then what happens when he goes up more? At aviation altitude, where airplanes fly, the dose rate will be increased up to 100times the high mountain cases. And what is expected when he goes up further more, up to space orbit altitude? In this case, the dose rate increases up to 10times the airplane cases.

Geomagnetism affects the dose rate very much. As primary cosmic ray particles are charged particles, they cannot do well with existence of the magnetic field. In effect, cosmic rays can penetrate into the polar atmosphere along geomagnetic lines of forces which stand almost vertical, but penetration of low energy cosmic rays will be banned when they intend to penetrate crossing the geomagnetic lines of forces in equatorial region. Therefore, exposure due to cosmic rays will become large in polar region, while it remains small in equatorial region. In effect, airplanes which fly over the pole will receive larger dose than to fly over the equator.

Only, we have to know that the cosmos has its own lifetime. In other words, lifetime of cosmos is finite. After all, at least human beings are concerned, we cannot ignore this.

誰しも宇宙がどのようなものか、なぜ宇宙が存在し、これから宇宙はどうなるのか、を考えてみたことがあるに違いない。宇宙の来し方行く末を考えるのは宇宙論という分野だが、宇宙論の端くれを理解しておくことが、放射線というエネルギーの形態に理解を深めることになると思える。

そもそもなぜ宇宙にエネルギーがあるのか、は誰も答えられない疑問である。大昔に、誰かが投げ込んだのが、エネルギーの束だったとしか言いようがない。何ごとにも必然性を求め、それを説明するのが自然科学の当然の方法だが、宇宙開闢だけはどうしても説明できない。あるものしかなく、現実にはそれは存在する、としか言いようがない。それで不安を感じるなら、自然科学以外の世界で安心を得るべきである。もともと自然科学とは自然現象の説明学だったのである。ある一定条件の下に、どのような法則性があり、従ってそれから先々何が起きるかを予想することを目的としていた。その条件そのものが変わってしまうなら、話は別なのである。宇宙ではそのような変化もあり得るのである。また宇宙という容器がなければ何も無く、何も起きようが無い。その意味で、宇宙論とは万物の根本を扱う分野である。ただ、シニカルなことを言うなら、役に立つことをしたいという人には勧められない。どうにも役に立ちそうもないからである。

役に立つ分野の代表は医学である。誰も病気になりたくなく、病気から一瞬でも早く解放されたいはずである。その技術を持つのが医者であり、だからこそ医者は特別扱いされて当然とされる。最近航空機医学の雑誌に「良きサマリヤ人」の話がよく出るが、それは医学の本質を付いていると思う。良き医療技術者に過ぎなくても、それでいいのである。看護師も同様であるが、彼らは心理分析家かもしれず、親しい相談相手かも知れない。一方宇宙を扱う物理学では、なぜ、に答えることは出来ないが、どのような形で起きそうかは答えられる。また宇宙の今後を占うことも、或る程度可能である。

それでは宇宙を最も卑近な所から眺める事から始めたい。下部では地殻に含まれる放射性核種から出る放射線が強く出るが、地表面から高い方へ向かって上昇すると、線源から離れるため放射線量は減少し、それが 150m 程度から上昇に転じる。これは航空機で実測したし、また高い鉄塔に上っても確かめた。このようにして、現在なら、宇宙線のせい、と言える。しかしこれは 90 年前にはどうしてそうなるのか、分からなかった。この「上方にある線源」はオーストリアの探検家の Hess が予想し、それから約 30 年後にノーベル賞を受賞した。これを分かり易く言うと、地上から航空機高度へ上ると 100 倍に増え、それが宇宙空間に出るとさらに 10 倍（つまり地表の 1000 倍）になる。これは放射線の線量率を計測した場合である。結論から先に言うと、世の中で「宇宙線」と称するものは、実は多くの粒子群の集合体である。実はそれらは大半が荷電粒子であり、故に磁場の影響を受けている。ところが地球磁場は荷電粒子と仲が悪く、磁場の強い所を荷電粒子は避けたいのである。太陽は一種のプラズマ発生源（荷電粒子群）だから、

この流れの中にある地球は、地球磁気圏という独特な形を保つようになる(Fig.1)。このように、宇宙にあるのは線源に違いないが、それは通常の線源ではない。そもそもそれは約 100 億年前の線源である。だから宇宙線をコントロールしようとしても出来ないのである。これは各所にある放射線施設を監視する機器に関連して、常に頭を悩ました点だったが、それを理解するには天体物理学の情報が必要だった。

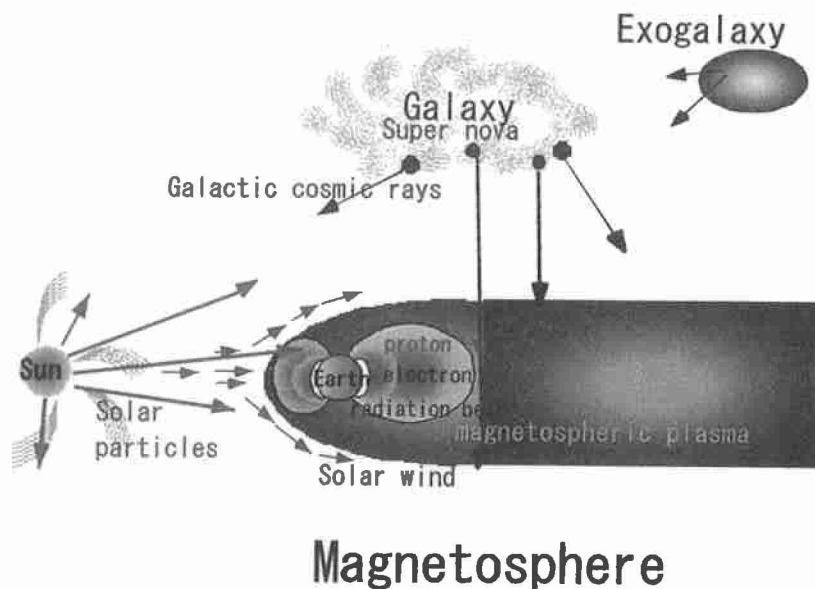


Fig.1 Rough sketch of magnetosphere

今日の知識で言えば、一般環境にある宇宙線とは、実は宇宙空間にある宇宙線そのものでなく、それらが大気と相互作用してできた2次宇宙線のことである。余りに種類が多いため、これらを総称して宇宙線と呼ぶ。宇宙ではほとんどが陽子であるが、これが大気粒子と衝突してそれを破壊すると、構成粒子がバラバラになって飛び出す。この過程のことを「核カスケード」と称する。飛び出すのが陽子かも知れず、あるいは中性子、または中間子かもしれない。

このうち、中間子というのは π 粒子であって、それは電荷を帯びない π^0 粒子と、電荷を帯びた π^\pm 粒子とからなる。電荷を帯びない π^0 は即2個の光子になってしまう。光子は動くと電子対を生み、その電子対が動くと光子が生まれる。これは交互に起きることで、実際問題として区別しにくいので、単に「電子カスケード」と称している。航空機高度では電子による被ばくが問題になるが、それはこの理由による。一方、電荷を帯びた π^\pm 粒子からは、もう一つの間接子たる μ^\pm 粒子が生まれる。ただし π^\pm はより寿命が長い (10^{-8} 秒のオーダー) ので、地表やもっと深部まで達してから、 μ 粒子に転換される。今まで最も深い所で見つかったのはインドのコラ鋳山の中である。結果的に地表面

では全体の被ばく源の80%が μ 粒子であり、残りが電子である。要するに地表の環境において、我々が遭遇する宇宙線はほとんど μ 粒子である。

すると最初に生じた宇宙線はどうやってできたか。これは星の進化に伴う超新星の爆発だろうと推定されている。恒星は始め表面にある水素を核融合反応で燃やすが、それが尽きるとその内側にあるヘリウムが燃え出す。我々の太陽はまだそこまで達していない。ヘリウムに点火するとこれは燃焼効率が非常に高いので巨大に膨らみ、太陽系を例にとつて説明すると、地球軌道を呑込む程度まで膨らむ。ところが後が続かない。そして太陽は燃焼による膨張圧力と、縮む重力が競い合い、ついには後者が勝つ。そして星は中心に向かって急激に落ち込み、これが大変な圧力を生む。元々の星の質量が太陽の3倍以下しかない場合は、白色矮星を残し、太陽の3倍を越える時は、中心部に中性子星が残される。元々の星がさらに大きく太陽の30倍もあった時は、中央にブラックホールが生まれると言われる。

いずれも落ち込みが激しく、その際に中性子を含む様々な粒子群が生まれ、それが四方八方に飛散る。それが宇宙線の誕生である。なぜ様々な重さの自然放射性核種があるのか、その訳はこの中性子を考えれば説明できる。この減少は平均して100億年の寿命があるとされるが、我々の太陽はまだその半分程度しか消耗していない。しかし、いずれどうにもならない時が来る。天体物理を知り過ぎるとシニカルになるわけである

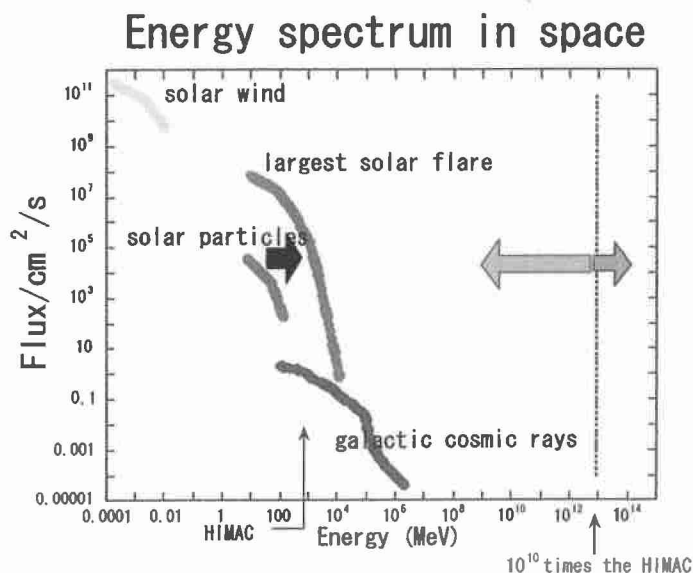


Fig.2 Energy spectra of cosmic rays

宇宙線を両対数グラフ用紙にプロットしてみると、全貌が良く分かる(Fig.2)。ここには太陽風という極めて穏やかなプラズマ流から、太陽フレアと呼ばれる突発的な現象、また普通に宇宙線と呼ぶ高エネルギーの現象の各フラックス分布を示してある。そして太陽起源のものも、銀河起源のものも宇宙線の範疇に入れてある。そしてこの銀河で生まれる最大エネルギーは 10^{13} MeV/核子とされる。我々の銀河の外で生まれた超高エネルギー

一のものも無いわけではないが、それは星雲同士の衝突に起源を持つと考えられている。分かり易くすると、放医研が誇る加速器HIMACの最大出力が800MeV/核子だから、その100億倍のエネルギーが、銀河内部で生まれる最大エネルギーである。人智の作り出す技術の小ささを感じさせる。

それでも地球は磁気圏の存在によって護られていることに触れたい。地球は全体として天然の棒磁石だが、そのモーメントは少しだけ回転軸から傾いている。我々はその中にいる限り、太陽フレアのような突発的な現象の時を除いて、かなり護られているのである。これが最も顕著に現れるのが、航空機で北回り路線と南回り路線のどちらを選ぶか、という問題が起きた時である。同じ欧州に行くとしても、南回りの方が時間がかかる。では線量はどうか。

Comparison of hypothetical flight

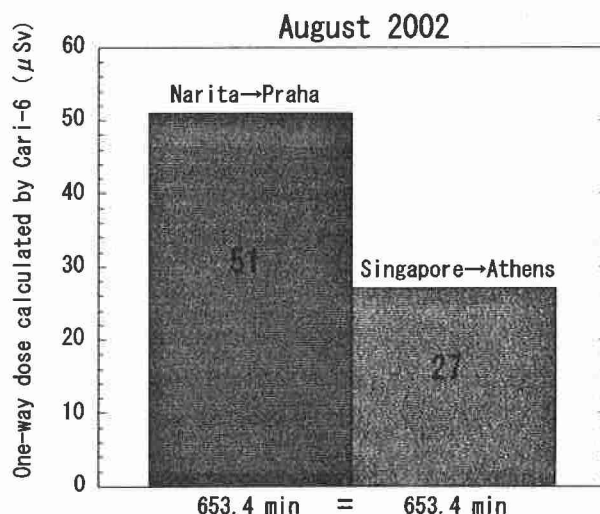


Fig.3 Northern root receives larger dose than southern root

ここにモンテカルロ計算コードCari-6を用いて計算した結果がある。仮想的な飛行ルートで、片方は成田→プラハへ飛び、もう片方はシンガポール→アテネに飛ぶルートである。モンテカルロ計算だから、色々な仮定に左右されるので、限界は自明だが、ここでは370000フィートの一定高度で飛ぶ場合を考えた。但し上昇時と下降時には各30分づつ時間をかけて移行するとした。明らかに成田→プラハの方がシンガポール→アテネより被ばく線量が大きい (Fig.3)。ほとんど2倍の差がある。モンテカルロ計算は便利だから、色々高度を変えた計算もしてみた。すると37000フィートと31000フィートでは、前者が約55 μSv 浴び、後者は約34 μSv を浴びることが分かった。さらに太陽活動の影響を調べるため、Heliocentric Potential という磁場のインデックスを計算したところ、成田発ではニューヨーク便が最大40%の変化を見せ、ベルリンやモスクワもそれに準じた。ところがホノルルやシンガポールでは殆ど変化しなかった (Fig.4)。ここに描いていない国内便のグラフはもっと下になり、形も寝ている。繰り返すが、あくまでこれはモデル

計算なので鵜呑みにはできない。

One-way dose which an airplane from Narita receive

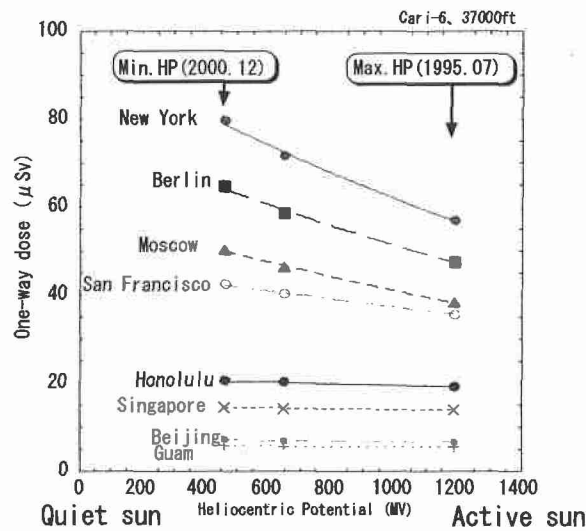


Fig.4 Effect of solar activity variation

太陽と言えは昨年のフレアの時どうなったのか興味あるだろう。これは太陽からの圧力で地球磁気圏が圧縮されて磁場が強くなり、もっと外側からくる銀河宇宙線が跳ね返さ

Solar Flare Effect

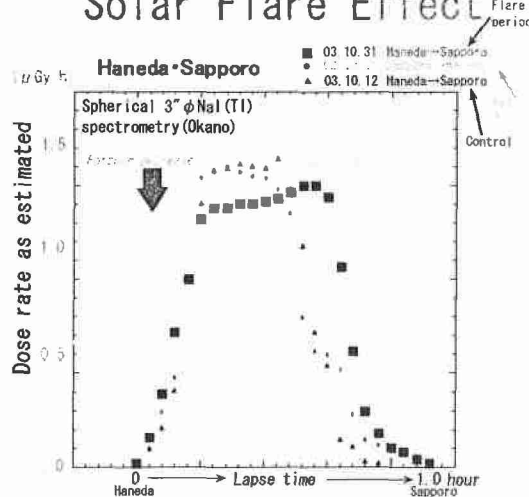


Fig.5 Forbush Decrease

れて、地球近傍の宇宙線は減ったのである。これは Forbush 減少と呼ばれる現象であり、日本のような低地磁気緯度で観測されたことは珍しい。その計測データを持っている。

どこまで飛んだらどれだけ被ばくすることになるかのデータも持っているが、これは簡易型のポケット線量計の結果であり、その限りにおいて、また中性子の評価法もその限

りにおいて、とにかく校正してある。成田から各地で飛ぶ時の片道線量で、ニューヨーク行きが最も大きく、アジア地域では小さい。ただし経験から言うと、同じパリ行でもルートが大きく異なったり、また飛行高度が大きく異なったりした例があった。従って一般的にこうなる保証はないかもしれない。

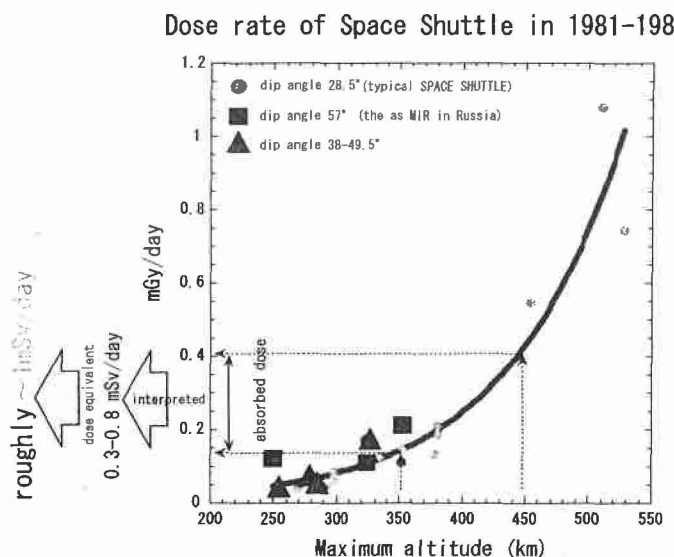


Fig.6 Dose rate in cosmos

そして宇宙である。我々の所では、その被ばく線量計測を調べている。米国 NASA のミッションが中断中なので、ロシアに頼み、プログレス・ロケットで打ち上げて貰っている。また中国からも長征 6 号に搭載して貰う話が進められているところである。今までのところ、磁気圏ない宇宙空間では 1mSv/日というのが線量の値として、世界中で使用されている。図はその内訳と簡略化の方法を示す (Fig.6)。

一方 JAXA(旧 NASDA)の計画している宇宙飛行士の受ける被ばく線量限度が発表されている。これをみると、火星に行くのは、40 歳にならないと 1Sv(シーベルト)を越すような被ばく線量を受け入れられないことになる。この Sv という単位は発がんを意識した単位である。比較すると、如何に火星ミッションが大きな被ばく源になるかが分かる (Fig.7)。この 1Sv という被ばく限界も法的な面でどうなるかは不明だが、その数値に頼って色々な星までの線量を距離から推定してみると、どうも人類は冥王星を越えられないようである。すなわち、太陽系から脱出できない。がんの治療線量になるからである。がん治療では合計 15 回程度に分割して患者に与え、その合計線量が約 70Sv になるが、宇宙でも太陽系の境界まで行くと、その値に近づく。がん細胞を殺す事を期待していたが、同様の線量で正常細胞も死ぬかも知れないのである。これをさらに他の恒星に惑星系があったとして、アルファ・ケンタウルス星まで行くと、10 万 Sv となって到底無理なことが分かる。この線量をどうするか、という議論がさらに深まることを期待したい。

Estimated dose in various missions (mSv)

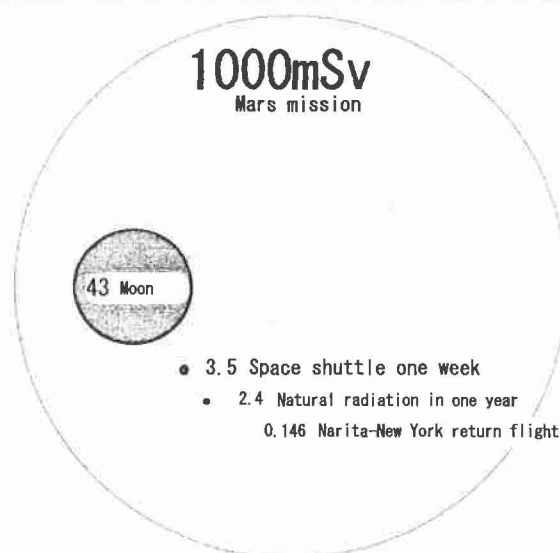
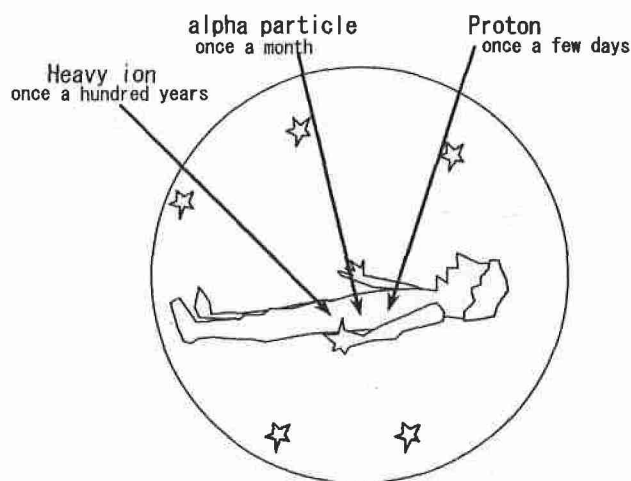


Fig.7 Doses received by various missions

宇宙線は約 92%の陽子と、7%の重粒子、1%の重粒子の合計からなる。これが人体に当たった時、何が起きるだろうか。体の特定細胞を考えるなら、陽子は2〜3日に1回ぶ

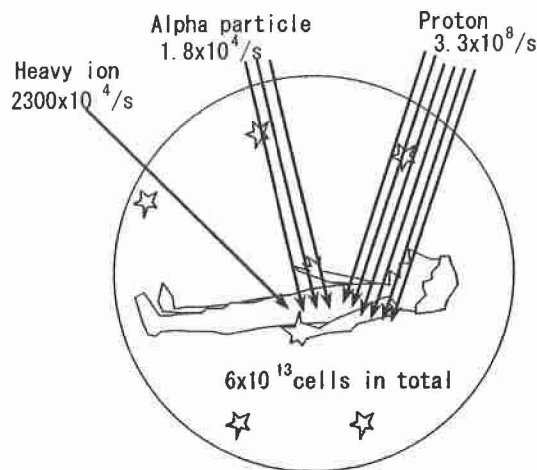


Same cell has a rare opportunity to be hit

Fig.8 Collision of same cells

ぶつかるし、アルファ粒子(ヘリウム原子核)イオンは1ヶ月に1度ぶつかり、また重粒子に至っては100年に一度しかぶつからない(Fig.8)。一生涯当たらない人もいるはずである。ところが体のどこかの細胞にぶつかれば良いとして確率を計算すると、この関係は大きく変わる。即ち陽子は1秒に3.3億回、ヘリウム原子核イオンは1秒に2300万回、

重粒子は1.8万回衝突するのである(Fig.9)。人体には60兆個もの細胞があるためだが、特定細胞でなく、そこから外れた細胞でも可、という所に興味を持たれる。放射線生物学では「バイスタンダー効果(当たっていない細胞が影響を受ける)」が昨今話題を集めているからである。



Some cells have got opportunities to be hi

Fig.9 Collision of different cells

もう一つ指摘しておきたいのは、宇宙が微小重力だという点である。現在遠心分離器の原理に基づき、大型の装置を計画中だが、実際問題として人工重力を作らないと、長期間宇宙へ人間を送ることは困難だと思う。3ヶ月間軌道にあったスカイラブ4号の宇宙飛行士の骨を観察すると、脱カルシウムが5%程度見られたからである。永久に地球に戻らないなら、タコみたいになっても良いと言うなら、影響を無視することも可能だが、元に戻ることを想定するなら、この微小重力対策は重要と思う。今までロシアで行われたボランティアによる長期間の寝たきりテスト結果や、過去にあった1年を越すような長期間宇宙滞在の際の記録を点検する必要があると考えている。

今のところ、宇宙に行くのはある種のボランティアのような、使命感のある人たちだろうと思う。彼らはパイオニアであり、大切にされなければならないと思う。そして地上でも理論研究や加速器利用研究をもっと進めるなど、色々な方面の研究をしないと、宇宙の余りの巨大さに太刀打ちできないだろうと考える。

【参考文献】 NASDA:有人サポート委員会放射線被ばく管理分科会報告書,H13.12 等

4.6 Utilization of Technology Relevant to Radiation and Isotope in the Archaeological Research

考古学における放射線及び同位体技術の応用

Nobuaki Matsuoka and Hidehisa Kawamura

松岡信明、川村秀久

Kyushu Environmental Evaluation Association

九州環境管理協会

Abstract

Many kinds of scientific technology have been used in the archaeological research. Especially the methodologies relevant to radiation and isotope have contributed to archaeology, giving a lot of scientific information. Among these methodologies, the radiocarbon dating, proposed by Willard Libby, has the greatest contribution since 1950. In Japan some scientists introduced this dating method immediately after Libby's proposal. As the result, the start of the Jomon period, in which the rope pattern was applied for decoration of earthenware, was reconsidered to be about 10,000 years ago. Yoshimasa Takashima mastered this technique and did the dendrochronological study at University of Washington, Seattle, from 1960 to 1961. After that he started the radiocarbon dating in Kyushu University, Fukuoka. First he employed the proportional gas counter to measure ^{14}C , requiring the complicated and time-consuming preparation of sample. When he restarted the radiocarbon dating with the authors in 1994 at Kyushu Environmental Evaluation Association (KEEA), he adopted the liquid scintillation counting method combined with the benzene synthesis from sample. Because this method is so convenient, many laboratories have adopted it as the conventional method in Japan. Since 1994, almost 100 samples have been treated every year in KEEA by this method. However this requires considerably much amount of sample, for example 20g in the case of wood. So that, in case of only small amount of sample can be obtained or a valuable sample is subjected to measurement, this method cannot be applicable. To resolve this problem, the accelerator mass spectrometer (AMS) has been used widely. In this method the atoms of ^{14}C are counted directly, getting the high sensitivity and requiring very small amount of sample (order of mg). Recently, in KEEA, the radiocarbon dating using AMS was started under the cooperation with Center for Applied Isotope Studies (CAIS), University of Georgia.

Another work of KEEA is the measurement of lead isotope ratios in the archaeological and environmental research. The lead isotope ratios, $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$, give useful information to understand the place or age in which an archaeological relic was produced. For example, as the lead isotope ratios are changeable depending on the difference of lead mine, it becomes possible to estimate the origin of relic containing lead as a component. Especially the origin of ancient bronze

mirror can be estimated by this method. The lead isotope ratios are measured by ICP-MS easily. In addition, the source estimation of lead pollution in the environment can be done in the same manner.

1. Introduction

Although archaeology is classified into the category of cultural or social science, many scientific technologies are applied for investigation in this field. Especially in recent years, new technologies have been introduced into the archaeological research one after another, and participation of scientists has been increasingly requested. In Table 1, scientific technologies, which have been introduced into the archaeology in Japan, are listed. As shown in Table 1, many technologies relevant to radiation and isotope, i.e. radiocarbon dating, thermoluminescence

Table 1 Scientific technologies applied in archaeology

Purpose	Scientific technologies	Relation with radiation and isotope
Dating	Radiocarbon dating	related
	Thermoluminescence dating	related
	Electron spin resonance dating	related
Estimation of relic producing center	Measurement of lead isotope	related
	Chemical analysis	
	X-ray fluorescence analysis	related
	Neutron activation analysis	related
Appraisal of technique and material	X-ray transmission	related
	γ -ray transmission	related
Appraisal of red pigment	Chemical analysis	
	X-ray fluorescence analysis	related
	Electron probed micro analysis	related
Investigation of life and occupation	Pollen analysis	
	Plant opal analysis	
	Fatty acid analysis	
	Measurement of carbon and nitrogen isotope ratios	related
Exploration of remains	Geophysical exploration	
	Aerial photograph	
Conservation of relic and remains	Conservation science	

dating, electron spin resonance dating, activation analysis, X-ray fluorescence spectroscopy and others, have been applied in the archaeology.

Especially the radiocarbon dating, proposed by Willard Libby⁽¹⁾, has the greatest contribution all over the world. For example, it was revealed by the radiocarbon dating that the monuments such as Stonehenge were earlier than the Mycenae civilization. Also in Japan, the start of Jomon age (rope sentence age, 10000–400 BC), when the rope pattern was applied for decoration of earthenware, was reconsidered to be about 12,000 years ago through the radiocarbon dating. This is several thousands years earlier than previously believed. On the early stage of investigation, the gas counting method was mainly used to measure radiocarbon. In the next stage, since the gas counting method is so complicated, the liquid scintillation counting method (LSC) has been introduced and widely used in the world due to its convenient property. In recent years, however, the very high precision is required to evaluate the accurate chronological age, and it is also required to use the very small amount of sample for the test. For these reasons, now the accelerator mass spectroscopy (AMS) is commonly used to measure radiocarbon⁽²⁾.

The naturally occurring lead has the four isotopes, ^{204}Pb , ^{206}Pb , ^{207}Pb and ^{208}Pb . The lead isotope ratios provide the information regarding to the producing center and age of ancient bronze relics excavated in Japan, in particular bronze mirrors. That is, the bronze relics contain a trace amount of lead and the lead isotope ratios are changeable depending on the difference of lead mine. Considering this property it becomes possible to estimate the producing center and age of relics. Commonly the lead isotope ratios are measured by the thermal ionization mass spectrometry (TI-MS) to achieve high precision. On the other hand, the inductively coupled plasma mass spectrometry

(ICP-MS) has come into wide use for trace analysis in the environmental research and can be used also to measure isotope ratios with the sufficient precision. This technique also can be applied to source estimation of environmental pollution. The lead, existing in the environment as the pollutant, has the same isotope ratios as those of source materials. And generally the lead isotope ratios of surrounding environmental materials are different from those of pollutants. So the source estimation of lead pollution can be done easily by comparing the isotope ratios of surrounding materials, pollutants and source materials.

Kyushu Environmental Evaluation Association (KEEA) is the research institution that has been doing the analysis of environmental pollutants, radioactivity, isotope ratios and so on. In its activity, there are many opportunities in which the analyses of archaeological samples are done. In this paper, some examples regarding to the radiocarbon dating and the lead isotope ratios of ancient bronze mirrors are presented.

2. Radiocarbon Dating in KEEA

2.1 Principle

Carbon has the three naturally occurring isotopes, that is, ^{12}C , ^{13}C and ^{14}C . It consists of 99% of ^{12}C and 1% of ^{13}C , but about one part in a 10^{12} of modern carbon is ^{14}C . Unlike ^{12}C and ^{13}C , ^{14}C is unstable and therefore radioactive, emitting β -ray with the energy of 186 keV. As shown in Fig.1, the earth is always exposed to the primary cosmic rays. In the upper atmosphere the primary cosmic rays react with the atmospheric materials, producing the secondary cosmic rays. Neutron is one of the secondary cosmic rays and produces the radiocarbon (^{14}C) through the nuclear reaction of $^{14}\text{N}(n, p)^{14}\text{C}$. Carbon-14 is oxidized into carbon dioxide ($^{14}\text{CO}_2$) and distributed uniformly on the earth through circulation and mixing of atmosphere. Furthermore ^{14}C would be distributed in hydrosphere through dissolution of carbon dioxide into fresh and ocean water, and in biosphere as the results of photosynthesis. Thus a slight amount of ^{14}C exists in our surrounding environment.

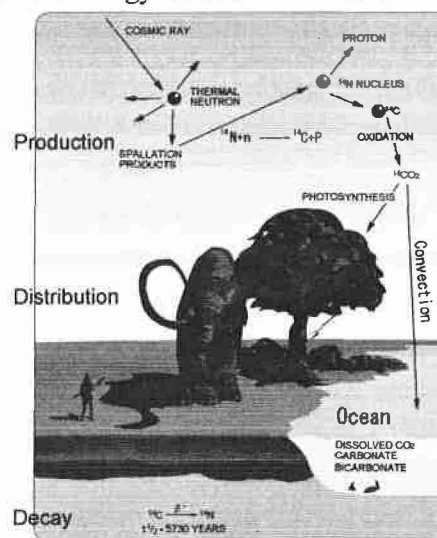


Fig.1 ^{14}C cycle in the environment

While ^{14}C decays with the half-life of 5730 y, it is newly produced in the upper atmosphere as shown above. So the equilibrium between decay and production of ^{14}C has been established in the environment. In the radiocarbon dating, it is the principal condition that the atmospheric concentration of ^{14}C (Bq/g-C) has been constant through the earth history, while actually slight variations due to some reasons, that is, fluctuations of earth's magnetic field strength, variations in sunspot activity and so on, have been confirmed. In principle, there is a constant ^{14}C level in the atmosphere and all living organisms.

Before 1950, the equilibrium concentration above mentioned was 226mBq/g-C in the atmosphere. However, artificial ^{14}C was produced and provided into the environment through the nuclear weapons testing since 1945. Therefore the concentration of ^{14}C in the environment increased abruptly after 1950. In 1963, the year of Partial Test Ban Treaty, the atmospheric concentration of ^{14}C reached the peak of 450mBq/g-C. After 1963 the concentration level has been gradually decreased due to nuclear weapon test ban and mixing of the excess ^{14}C through the carbon exchange cycle. Nevertheless, the present concentration still remains in 260mBq/g-C in the atmosphere. The phenomenon above mentioned is called "Bomb Effect".

On the other hand, the burning of large quantities of fossil fuel such as coal, oil and natural gas started in the 19th century. The fossil fuel laid down so long ago that its ^{14}C has totally decayed away, resulting in "Dead Carbon" that contains no ^{14}C . Accordingly carbon dioxide released through the burning of fossil fuel dilutes the atmospheric ^{14}C relative to ^{13}C and ^{12}C . This phenomenon is called "Suess Effect". It is sure that Suess Effect affects the atmospheric ^{14}C since the Industrial Revolution.

While it becomes rather comprehensive to evaluate the atmospheric ^{14}C concentration due to Bomb and Suess Effects, the radiocarbon dating is based on the principle that the equilibrium concentration of ^{14}C is attained in the environment through the earth's history. However, when a plant or an animal dies, it ceases to participate in carbon exchange with the biosphere and atmosphere, and no longer take up ^{14}C . Therefore, if a tree would be cut off, the initial ^{14}C concentration of A_0 would be decreased according to the half-life ($T_{1/2}$). The ^{14}C concentration of the tree after time t , A , could be related with A_0 by the following equation.

$$t = 1/\lambda \cdot \ln(A_0/A) \quad (1)$$

$$\lambda = \ln 2 / T_{1/2} \quad (2)$$

The best estimate of $T_{1/2}$ is 5730y. For the historical reason, however, the Libby half-life (5568y) is conventionally used as $T_{1/2}$. Using the Libby half-life, the equation (1) could be rewritten as follows.

$$t = -8033 \cdot \ln(A/A_0) \quad (3)$$

This t is the radiocarbon age. The reference material, oxalic acid produced in 1950, is used to determine A_0 , so that the radiocarbon age is presented in the form of years BP, meaning how many years old than 1950. In recent years, the calibration curves such as INTCAL98, are developed to convert the radiocarbon age to the calendar age.

As is understandable from the principle, the datable materials are organic. That is, the materials, which once formed part of the biosphere, are datable.

2.2 Measurement Method of ^{14}C

There are two methods of measurement of ^{14}C . The conventional method detects the activity of sample, that is, β -ray emitted by the decay of ^{14}C . In the early stage of radiocarbon dating, gas counting method was well used to measure β -ray from ^{14}C . In this method, carbon of the sample is finally converted to gases such as carbon dioxide, acetylene and methane. These gases are introduced into a proportional counter as counting gases. Recently liquid scintillation counting method

(LSC) is well used to measure β -ray from ^{14}C instead of gas counting method. In LSC, carbon of the sample is finally converted to benzene or methanol. These are added with small amounts of scintillator and subjected to LSC. Gas counting method is relatively complicate rather than LSC. Furthermore, benzene could contain relatively much more carbon as a component compared to other organic materials, so that LSC could attain the high sensitivity of measurement of ^{14}C . So now LSC is commonly used to measure β -ray from ^{14}C in the radiocarbon dating. As compared with the conventional method, accelerator mass spectroscopy (AMS) is a very much more recent technique and directly detects the number of ^{14}C atoms in the samples relative to ^{13}C and ^{12}C atoms. AMS has the higher sensitivity than LSC and requires only several mg of sample to measure the concentration of ^{14}C . In Japan, however, there are only a few institutes that conduct AMS, and these are not commercially available.

KEEA started the radiocarbon dating in 1993 with LSC. The schematic diagram for dating is shown in Fig.2. Samples subjected to the test are deterged with alkaline and acid solutions. First carbon dioxide is produced through combustion of organic samples or dropping phosphoric acid to carbonatious samples. Next carbon dioxide reacts with lithium metal at the temperature of 700°C , producing lithium carbide. If elemental carbon remains, it reacts also with lithium metal to produce lithium carbide. After cooling down the reaction vessel, water is added to lithium carbide to produce acetylene. Finally acetylene is trimerized to obtain benzene using the chromium(III)-alumina pellet as a catalyst. Approximately 5ml of benzene is obtained by processing several 10g of sample material. The above chemical procedure is done using the apparatus, TASK Benzene Synthesizer, developed by J.E.Noakes⁽³⁾. After adding the scintillator to the benzene sample at a constant ratio, the β -activity of ^{14}C is measured using the liquid scintillation counter, Packard TRI-CARB 1050/LL. The radiocarbon age is calculated according to the equation (3).

In recent years, AMS has been adopted in KEEA. In this case, the gas of carbon dioxide corresponding to 5mg of carbon is produced from a sample and sealed into a glass ampoule. The glass ampoule containing carbon dioxide is send to Center for Applied Isotope Studies, University of Georgia (CAIS). In CAIS, carbon dioxide is converted to graphite and subjected to AMS.

2.3 Examples of Dating in KEEA

2.3.1 Dating of Charcoal

The remains of Shobaru is located in Fukuoka prefecture, northern part of Kyushu Island. In

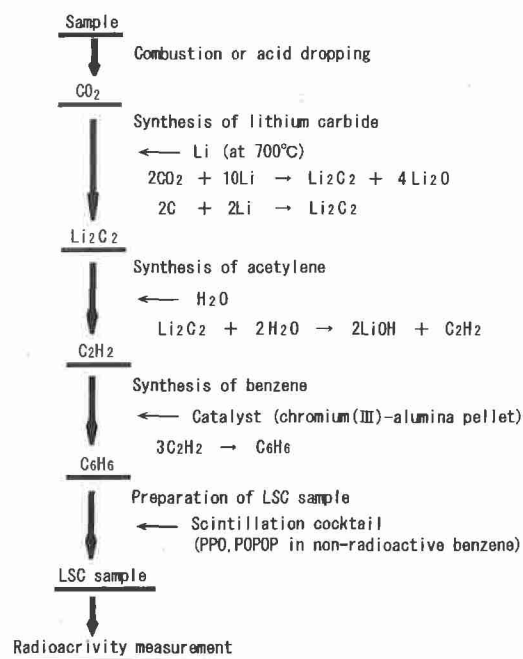


Fig. 2 Schematic diagram of LSC for the radiocarbon dating

those remains, the place, where an old furnace for melting a metal once stood, was discovered in 1995. The charcoal excavated from this place was sorted out by floatation and subjected to the radiocarbon dating in KEEA. The radiocarbon age was estimated by LSC to be 2220 ± 170 years BP, the early stage of Yayoi age. Yayoi age (400 BC – 300 AD) is the age after the rope sentence age (Jomon age) and prior to the ancient tomb age. However, there is a suggestion that the iron artifacts were produced in Shobaru remains. If so, this result is very epoch-making, because so far it has been accepted in Japan that the iron manufacture was started in the later stage of Yayoi age.

2.3.2 Dating of Unearthed Fallen Tree

The remains of Isouda-D is located near the Fukuoka City. Here the fallen trees were unearthed from 5m below ground. These trees had the marks of stoneax cutting and were dated to be 4800 years BP. From these results, it was revealed that a logging field for pit dwellings was situated here in Jomon age.

2.3.3 Dating of Carbonized Tree

The tephra is the general term of the pyroclastic material, which was emitted from the crater on the occasion of an eruption and deposited on surface of the earth. In Japan many tephra layers are distributed. The Aira tephra layer was formed by the eruption of Aira volcano, which was located in the southern part of Kyushu Island, and distributed all over Japan, especially in Kyushu Island. The carbonized tree was unearthed from the Aira tephra layer at Kagoshima prefecture, the southern part of Kyushu Island, and dated to be 23500 ± 700 years BP. This result well agrees with that introduced from the geological study.

2.3.4 Dating of Wooden Block

Kourokan was the guesthouse for greeting the guest from a foreign country for about 400 years from 7th century to 11th century. It was situated at the center of Fukuoka City. From these remains many kinds of relic such as wooden blocks, pieces of charcoal, potteries and others were excavated. These wooden blocks and pieces of charcoal were dated by LSC, and the calendar ages of 480 – 680 AD were obtained. These results seem to be contradictory to the fact of history. Taking the wood problem into account, however, there is no contradiction. That is, generally a wooden relic shows an earlier age than that in which it was used.

2.3.5 Dating of Underwater Remains

Imari Bay is located in the northern part of Kyushu. In 1281 AD, the Mongolian army attacked Imari Bay. At that time many Mongolian's naval fleets were sunk to the seabed of the bay. The main part of the anchor of a naval fleet was made of wood, and the big stones were fastened to it with the bamboo. Recently some anchors were pulled up from the seabed, and wooden and bamboo parts were subjected to the radiocarbon dating. In this case, the results of LSC were compared with those of AMS, resulting in good agreement with each other (Fig.3). These results were also in good agreement with the fact of history. So it can be concluded that not only AMS but also LSC are ap-

plicable for dating the medieval relics. In Fig. 3, the data without dendrochronological calibration are shown. In recent years the data calibrated by the calibration curve such as the INTCAL98⁽⁴⁾ are requested generally. In this case, the calendar age accompanied with certain probability is given.

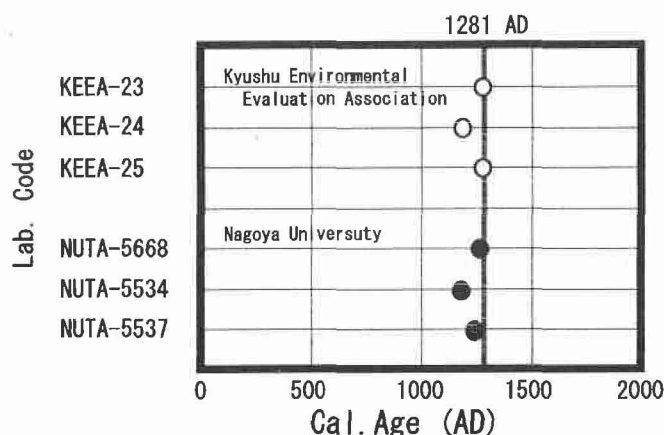


Fig. 3 Comparison between LSC and AMS in radio-carbon dating

Sample : Anchor materials pulled up from the seabed of Imari Bay

KEEA : Results of LSC carried on at KEEA

NUTA : Results of AMS carried on at Nagoya Univ.

*Each column of KEEA and NUTA shows the results for the same sample as each other.

3. Source Estimation of Lead Using Lead Isotope Ratios

3.1 Principle

In nature, there are 4 stable isotopes of lead, ^{204}Pb , ^{206}Pb , ^{207}Pb and ^{208}Pb . Among these, three isotopes except ^{204}Pb are the final products of natural radioactive decay chains of ^{235}U , ^{238}U and ^{232}Th . So, if uranium and thorium coexist with lead in a material, three isotopes of lead increase in quantity with time, resulting in the change of lead isotope ratios such as $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{206}\text{Pb}$, $^{208}\text{Pb}/^{206}\text{Pb}$. The lead isotope ratios are changing with time in a material containing lead, uranium and thorium at once. That is, lead isotope ratios of a certain lead ore are different from those of others depending on the geological and geochemical histories of individual materials. This can be said not only on ore samples, but also on other archaeological and environmental materials. Cumming and Richard studied the lead isotope ratios of earth materials, and proposed the theory of growth curve of isotope ratios⁽⁵⁾. On the basis of their theory, geo-chronological information of rock samples can be obtained by measuring lead isotope ratios. According to the theory of Cumming and Richard, materials having relatively low isotope ratios of $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ are defined as “more radiogenic”, whereas materials with relatively high isotope ratios are defined as “less radiogenic”.

So far, lead isotope ratios have been used for dating of ore and rock samples. However, lead that we encounter in our environment has peculiar isotope ratios, and we can obtain some environmental or archaeological information by measuring lead isotope ratios. Mukai et al. characterized

the sources of lead in the urban air of Asia using lead isotope ratios, and revealed that the exhaust gas of automobile affects the urban airs of Bangkok and Jakarta seriously ⁽⁶⁾. They also revealed that the airs of Korean and Chinese cities have been affected from coal combustion power plants, and the air of Japanese city has been affected from refuse incineration factories. Sturges and Harrison investigated the cross-border air pollution between USA and Canada using lead isotope ratios ⁽⁷⁾. They measured the lead isotope ratios of gasoline in Canada and USA, and of materials emitted from smelters in Canada.

As to archaeology, in Japan the measurement of lead isotope ratios has been applied to estimate the producing center of an ancient bronze mirror. That is, an ancient bronze mirror contains copper, tin and lead as the major component. So it becomes possible to estimate the producing center of mirror through measuring the lead isotope ratios. Mabuchi and Tominaga reviewed the methodology and scientific base ⁽⁸⁾. The bronze mirrors excavated from Japanese ruins were produced mainly in China of West Han dynasty, China of East Han dynasty and Japan. Plotting the data on the lead isotope ratio diagram, it would be estimated where the samples were produced (Fig.4).

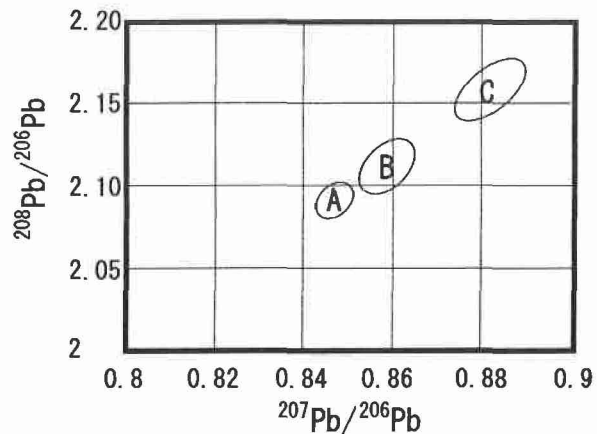


Fig. 4 Lead isotope ratio range of the ancient bronze mirror excavated in Japan

A : Galene produced in Japan
 B : Mirror produced in China of Eastern Han dynasty
 C : Mirror produced in China of Western Han dynasty

3.2 Measurement Method of Lead Isotope Ratios

As the archaeological sample material, a few 10mg of rust was taken from an ancient bronze mirror. As the sample for the environmental research, fifty mg of coal, 100mg of bottom ash, 100mg of soil, 300mg of dried agricultural product and a filter accompanied with SPM were subjected to analysis, respectively. Each sample was taken into a Teflon vessel specially designed for microwave decomposition ⁽⁹⁾, followed by addition of appropriate amount of deionized water and 3 to 4ml of nitric acid. In addition, a few ml of hydrofluoric acid, perchloric acid or hydrogen peroxide was added depending on the compositional situation of sample. The important point is to make a blank sample, to which same kinds and same amounts of reagents are added as well as a true sample. After addition of reagents, the tightly sealed Teflon vessel was placed in a microwave oven for kitchen use and the sample material was heated intermittently (5min × 3times) at the power corresponding to almost 50W/sample. After microwave decomposition, the sample was filtered with the filter paper (ADVANTEC 5C). The filtrate was taken into a volumetric flask and was adjusted to 25 or 50ml with deionized water. This solution was introduced into ICP/MS for measurement of lead isotope ratios. In ICP/MS measurement, true and blank samples were measured simultaneously, and the net ion-counts of individual isotopes were calculated by subtracting the counts of blank sample from those of true sample. From the net counts of individual

sample from those of true sample. From the net counts of individual isotopes, $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ ratios were calculated, and these data were normalized to SRM-PB-981 by measuring the reference material simultaneously.

3.3 Application to the Archaeological and Environmental Studies in KEEA

3.3.1 Producing Center Estimation of Ancient Bronze Mirrors

In Japan there are many old tombs built in the ancient tomb age, and sometimes the ancient bronze mirrors are excavated from these tombs. The archaeologists have the great interest in the producing center of these, because it is much important for archaeological considering where the mirrors were produced.

The Maebaru City is located in the west of the Fukuoka City. In this city there are many ancient tombs built in the ancient tomb age. Two ancient bronze mirrors excavated from this city were subjected to the analysis of lead isotope ratios (Fig.5). From the analytical results, the raw materials of two ancient mirrors were considered to have originated either in China of Eastern Han dynasty, or the Korean peninsula.

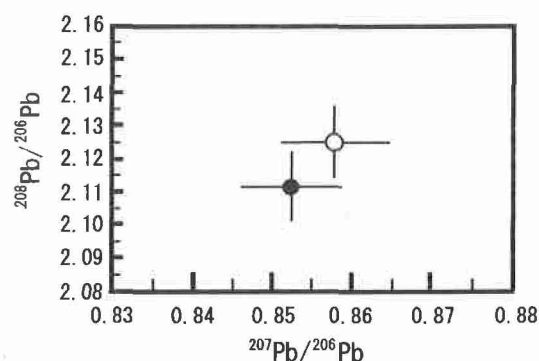


Fig.5 Lead isotope ratios of the ancient bronze mirrors excavated from the Maebaru City, Fukuoka Prefecture

An error bar shows the 2σ region.

○: mirror from Higashi-Makata No. 1 mound tomb
●: mirror from Tateishi No. 1 mound tomb

3.3.2 Source Estimation of Environmental Pollution

Coal combustion power plants release pollutants, such as sulfur oxide (SO_x), nitrogen oxide (NO_x), suspended particulate matter (SPM) and gaseous materials, into the environment. Therefore, it is necessary to monitor these pollutants in the surrounding environment of plant. In Japan, many monitoring stations are arranged around a power plant to monitor SO_x , NO_x and SPM continuously, and furthermore metal elements in air, soil and plants are measured periodically. However, many artificial and natural sources of pollution, such as incineration factories and volcanoes, exist in the surrounding environment of power plant. In this situation, it is very difficult to estimate the influence of power plant exactly using information only on concentrations of pollutants.

Lead is the useful and important indicator for the environmental monitoring of coal combustion power plant, and fortunately has four stable isotopes. Therefore, it enables us to do the environmental evaluation using isotope ra-

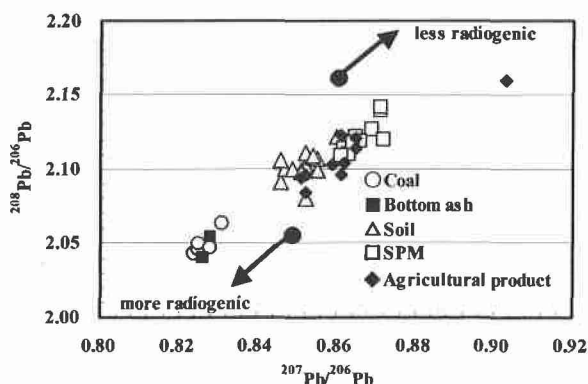


Fig.6 Lead isotope ratios of coal, bottom ash, soil, SPM and agricultural products

tios. So this technique was applied to evaluate the environmental impact of Reihoku coal combustion power station, located in the middle part of Kyushu Island. That is, the samples of coal, bottom ash, soil, suspended particulate matter (SPM) and agricultural product were collected in the surrounding area of station, and were subjected to the analysis of lead isotope ratios ⁽¹⁰⁾. From the results shown in Fig.6, it was revealed that there is no environmental influence of the power station as far as lead pollution is concerned.

4. Conclusions

At the present, many techniques and equipments, relevant to radiation and isotope, are essential for the human life. These have contributed to the developments of industry, agriculture, medicine and others since the end of 19th century. Especially the nuclear power became the main energy source in the later half of 20th century, and is thought to be essential for the sustainable development of the mankind in future. In addition, the nuclear power generation is needed to some extent to prevent the increase of carbon dioxide in the atmosphere.

Considering the fact above mentioned, people should have the correct knowledge about radiation, radioactivity, isotope and nuclear. So it would become more important to educate people, especially young people, in these fields. In the archaeological and environmental science, many techniques, which are relevant to radiation, radioactivity and nuclear, have been used. From the viewpoint of radiation education, it would be very effective to teach about these theories, methodologies and results.

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4.7 Charms of Radiation Research

Mitio Inokuti

Argonne National Laboratory, Argonne, Illinois 60439, U. S. A.

e-mail: Inokuti@anl.gov

Abstract

Most of my professional efforts over nearly five decades have been devoted to radiation research, that is, studies of the physical, chemical, and biological actions of high-energy radiation on matter. (By the term “high-energy radiation” I mean here x rays, γ rays, neutrons, and charged particles of high enough energies to produce ionization in matter. I exclude visible light, infrared waves, microwaves, and sound waves.) Charms of radiation research lie in its interdisciplinary character; although my training was in basic physics, the scope of my interest has gradually increased to cover many other areas, to my deep satisfaction. High-energy radiation is an important component of the universe, and of our environment. It often provides an effective avenue for characterizing matter and understanding its behavior. Near Earth’s surface this radiation is normally present in exceptionally low quantity, and yet it plays a significant role in some atmospheric phenomena such as auroras, and also in the evolution of life. The recent advent of various devices for producing high-energy radiation has opened up the possibility of many applications, including medical and industrial uses. I have worked on some aspects of those uses. At every opportunity to address a broad audience I try to convey a sense of intellectual fun, together with some of the elements of the basic science involved. A goal of radiation education might be to make the word “radiation” as common and familiar as words such as “fire” and “electricity” through increased usage.

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1. Prologue

In August 1945, when World War II ended with the defeat of Japan, I was a sixth grader of a Tokyo elementary school but was living safely in Nikko, a mountain resort 120 km north, owing to the “student evacuation” as a precaution against air raids. A few days earlier the atomic bombs had been dropped at Hiroshima and Nagasaki, and shortly thereafter a science teacher told us students about the science and technology related to the atomic bomb. The lecture, shorter than an hour, made a profound impression and gave me lasting stimulation; I wanted to learn much more about the topic. I am still studying a part of this topic.

The first technical book I ever read through in English, in 1948, was the Smyth Report [1]. It was the first official document published about the atomic bombs, and thus was translated into many languages, and indeed was excellently written. In this book I first saw the name of Argonne Laboratory, without a hunch that later I would work in the successor laboratory for longer than forty years, until today.

My specific interest in radiation and its interactions with matter was aroused by the Fukuryumaru (Lucky Dragon) incident, which occurred after the test of a thermonuclear device (named *Bravo*) in 1954 at the Bikini Atoll. The crew of a Japanese fishing boat was exposed to considerable radiation, and a member died shortly after returning to Japan. At the same time I began to learn about the beneficial use of x rays in medical diagnosis and therapy and was even more intrigued by this. Since then I have maintained a desire to learn about radiation interactions with matter and to contribute something to the increase of knowledge about them.

The charms of radiation science lie in its variety and diversity. High-energy radiation and its actions on matter are related to studies of elementary particles, nuclei, atoms, molecules, plasmas, and condensed matter, as well as accelerators, other radiation sources, and related instrumentation, within the discipline of physics. Some aspects of high-energy radiation belong to various branches of chemistry: physical, organic, inorganic, synthetic, and analytical chemistry. Other aspects fit into biology, medicine, environmental sciences, and some branches of engineering. Therefore, radiation science is a good area of study for someone, like me, whose range of interest is broad. Radiation science is a potpourri of different fields, and yet it represents an intellectual synthesis of encyclopedic knowledge.

Some areas of science originated in physics and eventually settled as a branch of engineering, as exemplified by mechanical engineering and electrical engineering. The twenty-first century may see radiation engineering as a similarly established discipline.

Let me discuss some topics selected to illustrate various aspects of radiation science.

2. Effects of radiation on synthetic polymers

As a graduate student at the University of Tokyo I studied the topic in title. The practical importance of this field does not seem to be as widely appreciated, as it should be in my view. Let me discuss an example that shows the importance.

A commonly available sample of polyethylene consists of large molecules (technically called polymers), each of which is a linear string and consists of many units (technically called monomers) of chemical structure $\text{-CH}_2\text{-}$. Irradiation of polyethylene with high-energy radiation such as x rays or γ rays produces a new chemical bond between neighboring molecules. A chemical bond thus produced is technically called a cross-link, and the process of its formation cross-linking. The cross-linking process accounts for a substantial fraction of the total energy absorbed from radiation, and in this sense it is a main result of radiation action.

After sufficient exposure to radiation, enough cross-links are formed to connect polyethylene molecules into a three-dimensional network (technically called a gel). Then, the sample becomes extremely strong and resistant both mechanically and chemically. This phenomenon led to many industrial applications. One application has to do with the cooling of the engine of an automobile with water. Till the late 1960s I often witnessed the loss of cooling water, leading to overheating of the engine and eventually to disabling of the automobile. This occurred as a result of degradation of rubber tubes and pipes because of oxidation, which is inevitable after repeated changes of temperature and in constant contact with oxygen in air. Young people probably do not know this kind of automobile failure, because it has been completely eliminated by the use of irradiated, cross-linked polyethylene in the water-cooling system.

Interestingly this eminently practical application began in Japan, years after its scientific basis had been discovered by Arthur Charlesby [2, 3] in the U. K. and by Malcolm Dole [4] in the U. S., independently of each other. This history is similar to that of the transistor

and its application to radios and other common devices, and it illustrates different kinds of strengths of different countries.

Effects of radiation on polymers in general depend on the molecular species, their configuration, and other characteristics, and therefore these effects offer a rich field of research still to be pursued [5, 6]. Wishing to learn more about the topic, I became an associate of Dole at Northwestern University in 1962, and thus began my life in the U. S. Gradually my scientific interest went deeper and deeper.

3. Electronic excitation and ionization of molecules

Gases such as air, liquids such as water or gasoline, and solids such as synthetic polymers or biological cells are made of molecules and therefore are technically called molecular substances. Most of them are electrical insulators. The action of high-energy radiation on these substances begins with disturbances of the motion of electrons, which are the main agents of chemical binding between atoms and hence are crucial to molecular structure. Most of the energy of radiation is expended in changing the motion of electrons, because they are the lightest constituents of matter, indeed much lighter than atomic nuclei. As a consequence, electrons may be raised to an orbit of higher energy but still be bound in an atom or molecule; technically we call this event electronic excitation. Alternatively, electrons may be pushed out of an atom or molecule and thus liberated to move far away; technically we call this event ionization, because what remains behind will be a positive ion. An ion is a charged atom or molecule. The radiation that causes ions to form is called *ionizing radiation*.

As a novice in science I wanted to understand fully all the microscopic processes sketched above, but I had no appreciation of the vastness and depth of the subject area. It encompasses electronic structures of atoms, molecules, and their assembly into liquids and solids, as well as collision phenomena. After nearly five decades I continue my study with no prospect of an end.

4. Two teachers I had in Chicago

I was fortunate to become associated with two masters of physics and chemistry related to radiation, U. Fano and R. L. Platzman. One of the benefits of studying any field of science is to be acquainted with persons of high intellectual ability and devotion.

Fano came from Italy and was trained in Rome by Enrico Fermi, one of the greatest physicists of the twentieth century. Fano contributed much to various areas of theoretical physics [7, 8]. I learned from him the basics of radiation's interactions with matter, first from his papers during my student days in Tokyo and later from numerous conversations in Chicago. He taught me many things. However, I remain most impressed with his statement to the following effect: "When we do something in an applied field like radiation science, we must still maintain intellectual standards high enough to command respect by a basic scientist." The truth of this idea has been becoming clearer and clearer to me, as I see more and more of mediocre work in applied sciences.

Platzman was an American, trained first at Chicago by James Franck, one of the greatest chemists of the twentieth century, and later in Copenhagen by Niels Bohr, and grew to be a pioneer in theoretical radiation chemistry [9]. Platzman invented several notions seminal to later developments. In 1963 he hired me as co-worker at Argonne. He taught me much, including several ways to study electronic excitation of molecules, including the use of synchrotron radiation, which is now realized in numerous institutions throughout the world.

The best known of Platzman's accomplishments is the 1952 prediction of the hydrated electron. In liquid water under irradiation, ionization occurs abundantly; in other words, many electrons are liberated and move into the water medium, leaving positive ions behind. To discuss what happens to these electrons, we need to recall a property of the water molecule: the outermost electrons in the molecule tend to be closer to the oxygen atom than to either of the two hydrogen atoms. Therefore, when seen from far out, the molecule looks like a dipole, namely, a line segment with one end positively charged and the other end negatively charged. We express the strength of the dipole by the electric dipole moment, defined as the product of the line length and the magnitude of the charge at either end. Thus, we say that the water molecule has an electric dipole moment. Indeed, many characteristics of water are attributable to the large dipole moment. Molecules in

water at room temperature are moving around, are vibrating internally, and most importantly, are rotating or changing the orientation of their dipoles, all as a consequence of thermal motion. If several neighboring dipoles happen to be directed to a central region, then, to an approaching electron, this region appears like a fixed positive charge. Thus, the electron is drawn and might be attached to this region if it loses some of its excess energy. Once the electron stays in the region for a while, then the arrangements of molecular orientations and hence dipoles become more favorable for trapping the electron. Platzman called an electron thus trapped in water the *hydrated electron* and reasoned that it ought to survive long enough to permit spectroscopic detection and also to act as a readily identifiable species in chemical reactions.

In 1962, Hart and Boag [10, 11] observed the absorption spectrum of the hydrated electron. Shortly thereafter I witnessed a conversation among Platzman, Boag, and Hart, sharing the jubilation of the great discovery. Indeed, this discovery is perhaps the most important contribution of radiation chemistry to chemistry in general. Similar states of electrons occurring in many dielectric liquids, such as ammonia and alcohols, are called *solvated electrons*. They act as a reducing species.

5. Effects of radiation on water

Chemical reactions initiated ionizing radiation are important subjects of study in two respects. First, many nuclear reactors use water as coolant. Second, water is the major constituent of the biological cell.

Pioneers in nuclear engineering such as Eugene Wigner were justifiably concerned about the effects of radiation on materials they considered using in reactors. Water is certainly one of such materials, because it is available at low cost, and it contains hydrogen atoms, which are most efficient as a neutron moderator. One may naively imagine that hydrogen molecules (H_2) and oxygen molecules (O_2) would be produced from water (H_2O) under irradiation, leading to the production of hydrogen and oxygen gases. If this occurred, then gas bubbles would emerge from the water coolant in a running reactor and give rise to high gas pressure in the reactor containment, posing a nasty engineering problem, as well as a potential for explosion.

Naturally, studies on chemical changes of water and other materials caused by radiation began early in the Manhattan Project. These studies were soon established as a field of research called *radiation chemistry*, and continue today. It was quickly found that only small amounts of gas come out of water under irradiation. More strikingly, irradiated water rebuilds itself almost completely after a period that is short but long enough to permit numerous collisions between molecules. During this period, diverse chemical reactions occur among hydrated electrons, hydroxyl radicals (OH), and other intermediate species produced by irradiation. Full explanation of all this [12] is highly technical, but it is noteworthy that common molecular substances are usually incapable of reforming themselves after irradiation. For example, irradiation of oil or gasoline leads to very appreciable production of hydrogen, methane, and other gases. As an exception, carbon dioxide CO₂ in gas or liquid is effective in reforming itself after irradiation, and is radiation-resistant.

As for radiation effects on the biological cell, it is important to appreciate that water dominates in the absorption of energy from radiation. Moreover, the atomic composition (more importantly, the electron density) of proteins and other constituent molecules in a cell differs only slightly from that of water; therefore, it is not a bad approximation to consider pure water in place of a cell, as far as energy absorption is concerned. Thus, a part of radiation effects on a cell must stem from energy absorption by water; in other words, hydroxyl radicals and other chemical species are produced from water and react with biomolecules, leading to some radiation effects. These are called *indirect effects*. Alternatively, some of radiation effects are attributable to energy absorption by proteins, histones, DNA, and other molecules in a cell. These are called *direct effects*. Most of experts in radiation biology believe that no consideration based on either indirect or direct effects alone can be complete; in other words, these two pathways are comparable in importance. We will return to radiation biology in Section 8.

6. Radiation effects on metals

One of the charms of radiation research lies in the great differences in radiation effects on different kinds of matter. Metals behave radically differently from molecular substances such as water and polymers, for example, for reasons to be sketched below.

Most of the electrons in a piece of metal are tightly bound to an atomic nucleus. These electrons move around in the immediate vicinity of the nucleus (technically called inner shells) and play little role in determining atomic structure and material properties. Only a modest number of electrons moving far from any atomic nuclei are decisive for atomic structure and material properties related to electricity, magnetism, and optics. These are called conduction electrons because they cause electric conduction. Ionizing radiation gives some energy to all of the electrons in matter, and roughly speaking similarly to each of them. Most of the energy given to the electrons degrades rapidly into a large number of quanta of atomic vibration (technically called *phonons*) and then to thermal energy, without causing irreversible displacements of atoms from their normal positions in the crystalline structure; thus, radiation heats the metal to some extent but causes little changes in material properties.

To observe effects of radiation on metal, one must do something special. One way is to cause a huge amount of x rays or γ rays to be absorbed. Another way is to use protons, α particles, or heavier ions of low speeds, comparable to the speeds of conduction electrons or lower; such slow particles hardly excite electrons, but they give energy to atoms to set them in motion, leading to an appreciable probability of irreversible displacements of atoms and hence to appreciable changes in material properties. Finally, neutrons cause atomic displacements directly, or indirectly through the generation of moving ions. Consequently, considerations of radiation effects on reactor materials usually focus on neutrons, heavy ions, and particles resulting from recoil of them [13].

In a personal conversation near the end of his life, I asked Charlesby, one of the discoverers of the cross-linking of polyethylene I mentioned in Section 2, how he made the discovery. His reply was in effect the following. In 1949, when he was new at Atomic Energy Research Establishment at Harwell, his assignment was to study effects of radiation on major materials used in nuclear reactors, especially, structurally important metals. Earlier studies had shown that metals are not highly sensitive to radiation, but his task was to determine the extent of radiation effects quantitatively. For months he tried to measure changes in properties such as electrical conductivity, heat conductivity, and elastic modulus, but in vain. The changes resulting from a moderate period of irradiation in a Harwell reactor were so minute that measuring devices conventional at that time gave no

reliable results; thus, he felt that the task was both boring and frustrating. Then, on a walk in the city of Oxford, near Harwell, an idea struck him; he saw cheap wristwatches on sale at a store and bought many of them. The wristwatches then, unlike electronic ones today, ran with the unwinding of steel springs. He wound all of the watches fully, set them at the correct time, and put them in the Harwell reactor. He took them out successively after fixed periods of time, and read the time; he found changes of the time readings, appreciable enough to be observed at good precision and also closely proportional to the period of irradiation. He was able to convert the observed results to changes in elastic modulus and other mechanical properties of steel, and he managed to write a report. This anecdote shows his mastery of experimental physics in perceiving a huge enhancement of the sensitivity of measurements by the use of wristwatches.

This experience drove Charlesby to look for a material that would show effects of radiation more easily. Having irradiated many kinds of materials at hand, he was fortunate to hit upon polyethylene, that is, a novel material at that time.

7. Measurement of radiation

In telling the story of wristwatch irradiation by Charlesby (Section 6), I mentioned an effect (changes of the time reading) occurring in proportion to the time of irradiation. Let us consider this point at some depth. For simplicity, let us assume that the power of a reactor stays the same. This means that the energy produced per unit time interval is constant, as is the number of nuclear-fission processes per unit time interval and also the number of secondary charged particles produced per unit time interval. We are tempted to say that a wristwatch in the reactor receives a constant “amount of radiation” per unit time interval. What does the “amount of radiation” mean? The wristwatch in the reactor is bombarded by particles of different kinds and different kinetic energies. We may choose to observe some quantity as an indicator of radiation action on matter. Which quantity is the most suitable? Pioneers of radiation science a century ago stumbled over this problem.

We now use as an index the energy of radiation absorbed by matter of unit mass, called *absorbed dose*. The unit of this quantity is the gray or joule/kilogram. With abbreviations, we write $1 \text{ Gy} = 1 \text{ J/kg}$. We apply the idea of absorbed dose to any kind of radiation and to any kind of matter. This notion certainly reflects some empirical knowledge. According

to measurements, cross-links in polyethylene, atomic displacements in a metal, charge carriers in a semiconductor, and ions in a gas are all produced in numbers closely proportional to the absorbed dose, under certain conditions. It is not simple to describe the conditions completely, but they always include an upper limit for the absorbed dose and hence for the number of product species. Typically, the number of product species departs from the proportionality to the absorbed dose when the latter is very high.

Can we derive from theory the proportionality of the number of product species to the absorbed dose? I have tried to solve this question. However, my answer is not very simple, and it certainly is too technical to explain here [14]. All I can say here is that the proportionality is only approximate and is subject to several rather restrictive assumptions about microscopic processes involved. Consequently, we see that the use of the absorbed dose is not unequivocally dictated by science but is in essence a stipulation adopted for convenience.

Despite the above qualification, experimental and theoretical studies on the absorption of radiation energy by matter form a branch of radiation science called *dosimetry*, which is important in many practical contexts in medicine, industry, and law, and therefore is pursued by many workers. Current studies in dosimetry concern ever more kinds of available radiation and also quantities far more detailed than the absorbed dose [14].

The goal of the International Commission on Radiation Units and Measurements (ICRU) is to provide, starting with the absorbed dose, international standards concerning quantities related to radiation, techniques for their measurement and calculation, data necessary for such tasks, reporting of results, and applications to medicine, industry, and research in other fields. The ICRU was formed in 1925 to address needs of medical radiologists. I have been a member since 1985. The main job of the ICRU is to select from the broad topical area of radiation measurements a timely theme suitable for a report, to find standards of knowledge and judgment reflecting an international consensus, and to publish a report describing the standards. For this purpose, scientists best qualified to prepare a manuscript are appointed to form a report committee. After work for several years, the report committee submits a manuscript to the Commission. Most often the manuscript is revised after the Commission's deliberations and is eventually adopted for issue as a report. So far 71 reports have been produced and made available to the public.

Since 2001 I have been serving as the Scientific Editor, and have worked on the final stages for Report Nos. 65-71. For a full account of the ICRU activities, I recommend the web site at www.icru.org, which includes an electronic journal entitled *ICRU News*.

8. Biological effects of radiation

Following up Section 5, I now briefly discuss biology. The cell of soft tissue consists of many kinds of molecules and has a hierarchical structure of many levels. According to most experts, biological effects stem from changes in molecular bonds and structure in DNA (deoxyribonucleic acid) in the cell nucleus, leading to perturbations of normal functions. Biological effects include the cell death, mutation or transformation of genes, and malfunctions or diseases of organs. Carcinogenesis, the most notable of the effects, is attributable to a failure of healthy cell replication and programmed cell death (technically called apoptosis). However, the cell contains many components other than DNA such as histones, other proteins, and molecules forming cell membranes. Radiation must cause a great deal of physical and chemical change in those components; consequences of these changes remain little studied, as far as I know.

In contrast, radiation-induced changes in DNA have been studied extensively. Molecules of DNA in a cell are present as the well-known double-helix structure, which is different from many synthetic polymers. Thus, scientists working on this topic distinguish between single-strand breaks and double-strand breaks and tell us that the latter kind of breaks should lead to more severe consequences than the former kind. It has been also well established that a majority of the breaks or damages of DNA are detected by enzymes and immunity agents and are repaired; no similar provision is present in inanimate matter.

Another remarkable aspect is that the amount of a biological effect is usually not proportional to the absorbed dose, even when the absorbed dose is small. Often the amount of an effect appears to consist of two parts, one of which is proportional to the absorbed dose and the other to the square of the absorbed dose. What does this mean? First, one reason for the departure of the amount of some effect from proportionality is physical and is related to the non-uniformity of energy absorption over a scale (about a micrometer or less) that happens to be comparable to the scale of microscopic structures of constituents of the cell. Second, a biological effect we observe is often not a response

of a single molecule or even a single cell, but is a coordinated response of a system of cells, as has recently been recognized increasingly clearly [15].

Even the first (that is, physical) reason alone points to notable consequences. One is that the amount of an effect for a given absorbed dose depends on kinds and kinetic energies of particles of radiation, because we know from physics that the spatial distribution of energy absorption in matter is different for different kinds and kinetic energies of particles. Another consequence is that the relation between the absorbed dose and its effect depends on how the intensity of radiation varies with time.

To kill half of the cells in a specific culture might require an absorbed dose of 2.0 Gy when we use x rays, but only 0.8 Gy when we use neutrons. It is customary to consider x rays as standard and to express the above finding by saying that the *relative biological effectiveness* for neutrons is $2.0/0.8 = 2.5$.

In medicine and legal regulations, one uses operational quantities called *dose equivalent* or *equivalent dose*, that is, the product of the absorbed dose and a modifying factor. The value of this dimensionless modifying factor is determined through analysis of data starting with the relative biological effectiveness and the kind and kinetic energy of every particle making up the radiation under consideration. The unit for the operational quantities can be J/kg, but a special name sievert (abbreviated as Sv) is recommended to emphasize the distinction from gray. The limit of exposure of a person of the general population is 1 mSv /y. The limit of exposure of a radiation worker is 20 mSv/y, on the average, over five years.

Examples of some larger values are as follows. A member of a crew on an international space station normally receives about 1 mSv/d, chiefly due to energetic protons from the Sun and cosmic rays. A large explosion on the surface of the Sun, called a solar flare, increases the exposure by nearly a factor of 10. A standard goal for killing cancer cells in therapy is 60 Sv. Exposure of 100 Sv over the whole body is fatal. (Note that thermal energy of 100 J/kg delivered to a human body will raise the temperature by 0.024 °C only.)

It is important to set standards for exposures of humans and the environment. Considerations necessary for this purpose, called *radiation protection*, extend far beyond natural science and include elements of economics, law, and decision-making strategy.

One issue often discussed in this connection is whether the risk of an adverse effect increases in proportion to the absorbed dose when it is very small or whether it is negligible up to some value (called a threshold) of the absorbed dose. Questions of this kind are difficult to answer when considered from the point of view of pure science; indeed, they are not precise enough to permit a clear-cut scientific answer. An answer to a question of this kind cannot be inconsistent with the scientific knowledge within its current limitations, but usually involves a decision or a policy related to the management in a broad sense.

The International Commission on Radiological Protection (ICRP) is responsible for issuing recommendations related to radiation protection. The ICRP and the ICRU maintain cooperative relations; often the ICRP raises issues to be considered, and the ICRU provides a scientific basis for considerations necessary to resolve the issues.

9. Role of physics in radiation research and applications

The greatest contribution to human well-being among applications of radiation is medical diagnosis by x rays. Initiated by Röntgen over a century ago, x-ray medical imaging has now developed into the computerized tomography (CT), which represents a feat in imaging technology. It is often said that the CT machine has saved more human lives than any other machine. The imaging technology with ionizing radiation or with other agents (such as ultrasonic or low-frequency electromagnetic waves) has many applications also in materials science, mechanical and structural engineering, geosciences, astronomy, and military technology.

In connection with diagnosis, I like to point out a wonderful advance I witnessed. The largest contributor to the total exposure of the general public in the 1960s was x rays used for medical diagnosis. Now the largest contributor is natural radioactivity. An appreciable reduction in human exposure was achieved by improvements in machines and detectors such as photographic films.

A reason for the success of the imaging by radiation is that this technology is based on firm knowledge of largely established physics. To clarify the foregoing statement and to show the role of physics, let me present a general observation. I find it appropriate to separate radiation-physics problems into two classes. Problems of Class I concern the fate

of radiation after interaction with matter. Problems of Class II concern the fate of matter after interaction with radiation. A typical problem of Class I is the degree of attenuation of high-energy photons (that is, x rays or γ rays) penetrating matter and its dependence on the particular kind of matter. Unless the photon energy is extremely low or high, and unless the atomic number of the matter is very high, we have reliable answers to this problem from theory and experiment, which serve as a basis for the imaging technology. Problems of Class I are easy to study experimentally, because one needs merely to analyze radiation after transmission through matter. Indeed, most of the problems of Class I have been solved in principle, apart from exceptional cases. (An example is when numerical results of extremely high precision are required. Another is when matter is highly inhomogeneous.)

Any problem of Class II is difficult to study, because a complete solution would require in principle analysis of all the major product species present in irradiated matter. Techniques available to us now do not meet this requirement. In other words, any irradiated matter represents a new material in the sense that we do not know all the component species, and a complete solution would mean full characterization of all these species. In addition, any irradiated matter is not in thermal equilibrium, and therefore its properties should be varying with time. Consequently, no complete solution of a Class II problem has been obtained.

I have personally devoted several years to a simple-looking problem: to characterize theoretically all the major pathways by which radiation energy is absorbed by pure hydrogen gas and to evaluate the probability of each pathway. The solution I obtained with the help of several co-workers turned out to be useful for some astronomers, because pure hydrogen gas serves as a basis for some considerations about cool regions of interstellar clouds and about the atmosphere of Jupiter.

Another Class II problem is old, well known, and much more complicated. This is to ask in effect what happens in a Geiger counter. It contains a gas and a pair of electrodes, and the passage of an ionizing particle causes a minute discharge that is amplified into a current that is measurable externally. According to empirical work, it is good to use a gas mainly made of argon, with a small amount of an alcohol, an ether, or a halogen-containing molecule. I cannot explain why and how this gas composition is optimal.

Finally, I wish to add an observation about the role of physics in radiation research. It is clear from an analysis [14] that contributions of physics have been substantial and decisive in the development of instrumentation in a broad sense, including radiation sources, dosimetry, and methods for detecting and analyzing some particular kinds of product species. However, contributions of physics to the elucidation of mechanisms of radiation action on matter remain limited and tentative. This trend is likely to continue.

10. Epilogue

I hope that the foregoing discussion conveys a sense of the research I have been participating in. Limitations of my ability and allotted space prevented me from treating a broader range of related topics. Some of them concern radiation and radioactivity as ubiquitous components of the environment and of the universe, as excellently surveyed by Draganić, Draganić, and Adloff [16]. Other topics are related to nuclear power and nuclear weapons. A recent book by Garwin and Charpak [17] presents a masterly survey of the topics.

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Note: In view of the non-technical character of the present article, references are limited to materials of general interest, leaving out the standard scientific literature except for a few items of historical significance.

