Note

Naturally Occurring Radionuclides of Paper Ashes and Their Effect in the Environment

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The concentrations of naturally occurring radionuclides (^{226}Ra , ^{228}Ra , ^{228}Th , and ^{40}K) in ashes of papers such as magazines and newspapers were determined from the nuclide concentrations in the papers and the ash contents of the papers. The average ^{226}Ra , ^{228}Ra , ^{228}Th , and ^{40}K concentrations in the 34 ashes were respectively 27, 68, 75, and 75 Bq kg⁻¹. The radium equivalent activities of the ashes were calculated to evaluate the hazard of γ -ray radiation from the ashes in the environment. A copying paper sample showed a high radium equivalent activity of 602 Bq kg⁻¹. However, the average radium equivalent activity was 140 Bq kg⁻¹ and was lower than the level that causes an environmental health problem.

Key Words : paper, paper ash, naturally occurring radionuclides, radioactivity content, radium equivalent activity, municipal waste, incinerator residue, environmental radiation

1. Introduction

Large amount of paper is produced, consumed, discarded, and recycled in developed countries. In Japan, 3.06×10^{10} kg of paper and paper board was produced, and 1.90×10^{10} kg of waste paper was recycled in the year 2008¹⁾. In the country, combustible municipal waste including paper waste is burned to ash in incinerators. Most of the resulting incinerator residue is buried in disposal sites, and a part of the residue is used as raw material of cement (eco-cement)²⁾. In 2008, 3.57×10^{10} kg of combustible municipal waste was incinerated³⁾. The weight of paper waste incinerated is calculated to be 1.1×10^{10} kg, because the proportion of paper waste in combustible municipal waste is estimated to be 30%⁴⁾. City of Nerima, Tokyo⁵⁾ reported the kinds of papers and their contents of combustible municipal waste collected in the

city as follows : magazines, books, wrapping paper, etc., 15.9% ; newspaper and inserted bills, 5.4% ; corrugated cardboard, 3.7%.

The author^{6)–8)} studied the radioactivity levels of naturally occurring radionuclides in papers, and showed that a number of the paper samples examined contained the nuclides as much as rocks. The conversion of paper waste into ash should concentrate radionuclides contained. This implies that ash produced by combustion of paper waste may increase the radiation levels at disposal sites and places where eco-cement is used.

In this article, the author reports the result of determination of naturally occurring radionuclides in ashes of papers and discusses the hazard of radiation from the ashes in the environment.

2. Materials and Methods

2.1 Sample description

Pocket-sized books, weekly photo magazines, magazines full of color pages, weekly general interest magazines, weekly comic magazines, newspapers, and inserted bills printed between 1999 and 2000 in Japan were examined in this work. Printing papers, copying papers, and notebooks sold between 1999 and 2000 in Japan were also examined in this work. The samples examined were representative of papers that were produced or sold in Japan. The details of the samples were described in the previous articles^{8),9)}. The concentrations of naturally occurring radionuclides (^{226}Ra , ^{228}Ra , ^{228}Th , and ^{40}K) in the samples were measured by γ -ray spectrometry and reported in the earlier article⁸⁾. The fillers and coating pigments contained in the samples were examined by X-ray diffraction analysis and also reported in the earlier article⁸⁾.

2.2 Determination of ash contents

Except for the printing paper, copying paper, and newspaper samples, more than one kind of paper were used for the samples tested in this work. The weight of samples that were used for the γ -spectrometry ranged from 0.4 to 1.1 kg. Therefore, samples for determination of ash content were prepared by the following manner: 1.3 – 2.1 g portion was cut out by punching a few holes (6.5 mm diameter) in each γ -ray spectrometry sample in vertical direction to the layer of paper sheets. Each portion obtained was weighed and placed in a porcelain crucible. Then it was burned to ash and ignited at 900 °C in an electric furnace¹⁰⁾. Resulting ash was weighed, and the ash content was calculated.

3. Results and Discussion

3.1 Ash contents and concentrations of naturally occurring radionuclides in the ashes

The result of determination of ash contents is shown in Table 1. As seen in Table 1, the ash content varies widely from 3.3 to 29.8% although paper samples of the same category show comparable ash contents. The average ash content is 13.0%.

The concentrations of naturally occurring radionuclides (^{226}Ra , ^{228}Ra , ^{228}Th , and ^{40}K) in the ashes of the paper samples were estimated from the nuclide concentrations in the samples reported in the author's previous article⁸⁾ and the ash content data obtained in this work. The result of the estimation is also shown in Table 1. As shown in Table 1, the average ^{226}Ra , ^{228}Ra , ^{228}Th , and ^{40}K concentrations of the ashes are respectively 27, 68, 75, and 75 Bq kg⁻¹.

3.2 Radium equivalent activities of the ashes

The γ -ray radiation from natural materials is substantially composed of radiations from the following three sources: ^{226}Ra and its descendant nuclides; ^{232}Th and its descendant nuclides; ^{40}K . The intensities of γ -ray radiation of the above three sources can be expressed respectively by the radioactivities of ^{226}Ra , ^{232}Th , and ^{40}K . When the health effect of γ -ray radiation from natural materials is considered, an index is needed to express the total intensity of γ -ray radiations from the above three sources. Radium equivalent activity (Ra_{eq}) is such an index for this purpose¹¹⁾. In this index, the radioactivities of ^{232}Th and ^{40}K are converted to the radioactivity of ^{226}Ra , and Ra_{eq} is calculated by the following equation:

Table 1 Ash contents of paper samples, radionuclide concentrations in paper ashes, and radium equivalents of paper ashes

Kind of samples	Number of samples	Range of ash content (%)	Average ash content (%)	Average concentrations of radionuclides in paper ash (Bq kg ⁻¹)				Radium equivalent of paper ash (Bq kg ⁻¹)
				²²⁶ Ra	²²⁸ Ra	²²⁸ Th	⁴⁰ K	
(A) Pocket-sized book	5	14.8-17.8	16.4	39	131	144	114	254
(B) Weekly photo magazine	4	21.9-29.8	26.3	30	76	77	52	144
(C) Magazine full of color pages	3	21.1-23.9	22.4	29	60	64	53	124
(D) Weekly general interest magazine	4	11.9-12.5	12.1	29	58	62	71	123
(E) Weekly comic magazine	4	4.9-5.6	5.3	21	46	53	95	106
(F) Printing paper	2	9.2-11.5	10.4	11	14	17	21	38
(G) Copying paper	3	3.3-6.6	4.5	35	116	137	114	241
(H) Notebook	4	5.7-10.5	8.5	21	44	51	62	98
(I) Newspaper	3	3.3-3.9	3.7	14	23	29	76	62
(J) Inserted bill	2	19.3-20.5	19.9	28	64	68	45	129
Whole samples	34	3.3-29.8	13.0	27	68	75	75	140

$$Ra_{eq} = A_{Ra} + (A_{Th} \times 1.43) + (A_K \times 0.077) \quad (1)$$

where A_{Ra} , A_{Th} , and A_K are respectively the radioactivity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in the material of interest.

In Equation 1, it is assumed that the descendants of ^{226}Ra are in radioactive equilibrium with ^{226}Ra , and it is also assumed that the descendants of ^{232}Th are in radioactive equilibrium with ^{232}Th . As seen in Table 1, the radioactivity concentrations of ^{228}Ra are somewhat lower than those of ^{228}Th . It means that the radioactive equilibrium of thorium series nuclides is not closely established in the samples tested. The reason for the disequilibrium can be explained by preferential leaching of ^{228}Ra in production of paper or in preparation of raw materials of paper⁸⁾. However, thorium series nuclides are nearly in equilibrium in the samples. Therefore, in this work the concentrations of ^{228}Th in the ashes are adopted as A_{Th} in Equation 1, because the γ -rays of thorium series nu-

clides are chiefly made up of γ -rays from ^{228}Th and its descendant nuclides.

The result of calculation of radium equivalent activities is shown Table 1 and Fig. 1. The average radium equivalent activity of each category is listed in Table 1, and the radium equivalent activities of all the ashes are shown in Fig. 1. Large difference of Ra_{eq} value is observed among the samples examined (Fig. 1). A copying paper sample (G3) shows a high Ra_{eq} value of 602 Bq kg⁻¹ whereas a printing paper sample (F1) shows a low Ra_{eq} value of 8. The author⁸⁾ has demonstrated that the naturally occurring radionuclides in the paper samples were mainly brought with kaolinite used as filler or coating pigment in the papers. G3 sample contained kaolinite and talc as filler, while F1 sample contained calcite as filler and did not contain kaolinite⁸⁾. That is the reason for the large difference in Ra_{eq} value between G3 and F1. As shown in Table 1, the average of the radium equivalent activities is 140 Bq kg⁻¹.

Fig. 1 Radium equivalent activities of ashes from paper samples. Kinds of samples are as in Table 1.

Table 2 Radium equivalent activity

	Concentrations of radionuclides			Radium equivalent radioactivity (Bq kg ⁻¹)
	(Bq kg ⁻¹)			
	²²⁶ Ra	²³² Th	⁴⁰ K	
Earth crust	11.2	14.1	282	53
Granite	60	80	1000	251
Limestone	30	7	90	47
Paper ash (this work)	27	75	75	140

3·3 The radioactivity level of the ashes

As is mentioned in the introduction, ash from combustible municipal waste including paper waste is used as raw material of cement in Japan. As for building materials, some criterions for the limit of radionuclide contents have been proposed. Among such criterions, the following^{(11), (12)} is generally accepted: $Ra_{eq} < 370 \text{ Bq kg}^{-1}$. Figure 1 shows that three samples (A2, A4, and G3) represent Ra_{eq} values beyond the criterion value. However, the average Ra_{eq}

value of the paper ashes tested (140 Bq kg^{-1}) is fairly lower than the criterion value.

As already mentioned, ash from combustible municipal waste including paper waste is buried in disposal sites in Japan. It is necessary to discuss the question whether paper ashes increase radiation dose rate at disposal sites. Comparison of the Ra_{eq} values of paper ashes with those of the earth crust and representative rocks is effective for the discussion. The Ra_{eq} value of the earth crust is calculated based

on the average concentration of uranium, thorium, and potassium concentrations in the earth crust¹³⁾. The R_{eq} values of granite and limestone are calculated based on the typical concentrations of ^{238}U , ^{232}Th , and ^{40}K in granite and limestone¹⁴⁾. Granite and limestone are common rocks in the earth crust, and it is known that radiation dose rate is generally higher in granite area than in other rock areas. The result of the calculation is shown in Table 2. As seen in Table 2, the average R_{eq} value of paper ashes examined in this work is higher than those of the earth crust and limestone, but it is fairly lower than that of granite.

In conclusion, the radioactivity level of the ashes of Japanese papers is not so high that the ashes probably will not cause an environmental health problem.

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要 旨

紙類の灰に含まれる天然放射性核種と健康への影響評価

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雑誌, 新聞等の紙類の灰に含まれる天然放射性核種 (^{226}Ra , ^{228}Ra , ^{228}Th 及び ^{40}K) の濃度を, 紙類中のそれらの濃度及び灰分量から決定した。34 試料の灰に含まれる ^{226}Ra , ^{228}Ra , ^{228}Th 及び ^{40}K の平均濃度は, それぞれ 27, 68, 75 及び 75 Bq kg^{-1} であった。環境中における紙類の灰からの γ 線の有害性を評価するために, 各灰試料のラジウム当量を計算した。コピー用紙の一つは, 602 Bq kg^{-1} という高いラジウム当量を示した。しかし, ラジウム当量の平均値は, 140 Bq kg^{-1} であり, 環境的な健康問題を起こすレベルではなかった。