Radioactivity in Books Printed in Japan: Its Source and Relation to the Year of Issue

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$^{226,228}\text{Ra}/^{228}\text{Th}/^{40}\text{K}/^{137}\text{Cs}/\text{Books}$

The radioactivities of the naturally occurring radionuclides ($^{226}\text{Ra}$, $^{228}\text{Ra}$, $^{228}\text{Th}$ and $^{40}\text{K}$) and a fallout nuclide ($^{137}\text{Cs}$) in books produced in Japan in the 20th century were measured by gamma-ray spectrometry to obtain information on radiation emitted from books. The respective concentration ranges of $^{226}\text{Ra}$, $^{228}\text{Ra}$, $^{228}\text{Th}$, $^{40}\text{K}$, and $^{137}\text{Cs}$ were $0.2-6.4$, $0.4-11.2$, $0.3-11.3$, $1-112$, and $0-3.6$ Bq kg$^{-1}$. X-ray diffraction spectra of the papers used in book printing showed that pyrophyllite, talc, kaolinite, and calcium carbonate were contained as fillers. A comparison of the radioactivity contents of the pulp and filler indicated that most of $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ in the books was present in the filler whereas $^{137}\text{Cs}$ was in the pulp. The pattern of the concentration of each nuclide vs. the year of issue of the book was investigated. Patterns for the naturally occurring radionuclides were similar and were explained by the kinds of filler used. The pattern for $^{137}\text{Cs}$ differed from the patterns of the naturally occurring radionuclides, having a marked peak in the mid-1960s.

INTRODUCTION

Measurement of the radioactivity in materials in the environment is very important for estimating the radiation dose to inhabitants produced by environmental radiation. Books are present in large number in libraries. They also are found in large numbers in offices and homes. There is little in the literature on the radioactivity of books. Lalit and Shukla$^1$ measured the radioactivities of $^{226}\text{Ra}$, $^{228}\text{Th}$, $^{40}\text{K}$, and $^{137}\text{Cs}$ in about 250 books produced in several countries. The respective concentrations of $^{226}\text{Ra}$, $^{228}\text{Th}$, $^{40}\text{K}$, and $^{137}\text{Cs}$ in the books were of the order of 4–80, 4–70, 7–240, and 0–7 Bq kg$^{-1}$. These concentrations are comparable to those in rocks, therefore, books may be the main source of radiation in libraries and other places where many books are stored.

It is necessary to determine why books contain as much radioactivity as rocks. Lalit and Shukla$^1$ suggested that the radioactivity in books comes mainly from the plants used as the raw material for pulp which have been contaminated by radionuclides. Their suggestion is questionable because the $^{226}\text{Ra}$ and $^{228}\text{Th}$ contents of plants do not account for the nuclide concentrations
found in books. The concentration of $^{226}$Ra in plants generally is about $0.37 \text{ Bq kg}^{-1}$ ($0.01 \text{ pCi g}^{-1}$)\(^2\), and the concentration of $^{228}$Th in plants may be comparable to that of $^{226}$Ra\(^3\). The sources of $^{226}$Ra and $^{228}$Th as well as the sources of $^{40}$K and $^{137}$Cs must be investigated.

There have been marked advances in papermaking technology; not only the techniques and machines but the raw materials use in papers have been changing. It is necessary to know the relation between the radioactivity content of books and these advances.

I measured the radioactivity concentrations of $^{226}$Ra ($t_{1/2} = 1600 \text{ y}$), $^{228}$Ra ($t_{1/2} = 5.76 \text{ y}$), $^{228}$Th ($t_{1/2} = 1.91 \text{ y}$), $^{40}$K ($t_{1/2} = 1.27 \times 10^9 \text{ y}$), and $^{137}$Cs ($t_{1/2} = 30.1 \text{ y}$) in books produced in Japan during the 20th century. Here I discuss the sources of these radioactivities and the relation between them and the year of issue of a book.

**MATERIALS AND METHODS**

**Sample description**

The samples tested were 67 books produced in Japan that were issued between 1917 and 1993. Books with normal paper (uncoated printing paper) with the following dimensions: 200–220 mm long, 140–160 mm wide, and 27–33 mm thick, were chosen. All but three had a hard-cover. The thickness of the hard-covers was about 2.5 mm.

**Gamma-ray spectrometry**

Samples for gamma-ray spectrometry were prepared by sealing each book in two polyethylene bags (0.1 mm in total thickness) airtight. The samples were allowed to stand for more than 30 days before measurement to ensure there was radioactive equilibrium between $^{226}$Ra and $^{222}$Rn. Gamma-rays were measured with a gamma-ray spectrometer equipped with a 100-cm\(^3\) pure germanium detector and a 4096-channel pulse-height analyzer. The detector was placed in a shielding box made up of layers of 100-mm lead, 50-mm iron, 10-mm copper and 5-mm acrylic resin. The spectrometer was placed in a well ventilated, air-conditioned room to ensure low background counts\(^4\). Measurement of each sample was done for 3 days. $^{226}$Ra, $^{228}$Ra, and $^{228}$Th were determined by the intensities of the gamma-rays from their daughter nuclides in radioactive equilibrium. That is, $^{226}$Ra, $^{228}$Ra, and $^{228}$Th respectively were determined using $^{214}$Bi 609 keV, $^{228}$Ac 911 keV, and $^{208}$Tl 583 keV gamma-rays. $^{137}$Cs and $^{40}$K respectively were determined by the intensities of the 662 keV and 1461 keV gamma-rays.

Two standard samples 210 mm long, 150 mm wide and 30 mm thick were used. They were prepared by diluting accurately weighed potassium chloride (reagent grade, Wako Pure Chemical Ind.) or monazite powder in sucrose in a plastic case. The concentrations of $^{226}$Ra, $^{228}$Ra, and $^{228}$Th in the monazite powder were measured and the radioactive equilibrium of the thorium series nuclides in the mineral confirmed as reported previously\(^3\). The concentrations of $^{226}$Ra, $^{228}$Ra, and $^{228}$Th in the monazite powder also were measured by another method based on a determination of the nuclides present in the powder using the efficiency curve obtained for a $^{152}$Eu solution of known $^{152}$Eu concentration as reported by the Japan Radioisotope Association. The analytical values obtained by this method agreed with those obtained by the method...
reported previously\textsuperscript{3}). The monazite-doped standard sample was sealed airtight and allowed to stand for more than 30 days prior to gamma-ray spectrometry.

**X-ray diffraction measurements**

X-ray diffraction measurements were used to identify the minerals in printed pages of the books. Pieces of pages from each book were stacked to 1 mm thick in a sample holder and subjected to X-ray diffraction measurements in a diffractometer (Rigaku RINT 2000 series) that used copper K alpha X-rays passed through a nickel filter. Minerals were identified by the X-ray diffraction spectra obtained.

**Phloroglucinol color test**

The main component of paper is pulp. The pulp used in book printing has been prepared mainly from wood in the 20th century in Japan. This wood pulp is classified as mechanical or chemical pulp depending on the preparation method. Mechanical pulp, prepared by grinding the wood, contains lignin. Chemical pulp is prepared by chemically separating cellulose from the wood and therefore contains little lignin. Phloroglucinol staining is used to detect the presence of lignin.

The phloroglucinol color test was used to judge whether mechanical pulp was present in the printed pages of the books. Phloroglucinol solution was prepared by dissolving 1.0 g of phloroglucinol in 50 ml of ethanol then adding 25 ml of concentrated hydrochloric acid. All the chemicals used were of reagent grade. A drop of this phloroglucinol solution was placed on a printed page of each book and the degree of red purple coloration checked.

**RESULTS AND DISCUSSION**

**Evaluation of the book samples**

Most of the book samples analyzed were hard-bound technical and literary books, typical of the kinds of books found in libraries throughout Japan. The samples had the ordinary dimensions of most books found in libraries, and therefore, the books analyzed were typical library books. The number of the books analyzed was 67, and they represented 43 different publishing companies. In most cases, when two books were published by the same company, they differed in year of issue by more than 5 years. The papers used in the books are thought to vary widely in origin. Consequently, the analytical data obtained from the books tested should give unbiased information about radioactivity in Japanese library books.

**Concentrations of radioactive nuclides in the books**

The concentrations of the radioactive nuclides in the books are shown in Figs. 1–5. The gamma-ray spectrometry was done during 1991–1994. The \textsuperscript{137}Cs concentrations shown in Fig. 5 are for respective decays corrected to 1 January 1995. As seen in these figures, the respective concentration ranges of \textsuperscript{226}Ra, \textsuperscript{228}Ra, \textsuperscript{228}Th, \textsuperscript{40}K, and \textsuperscript{137}Cs are 0.2–6.4, 0.4–11.2, 0.3–11.3, 1–112, and 0–3.6 Bq kg\textsuperscript{-1}. The respective standard deviations for the data on the \textsuperscript{226}Ra, \textsuperscript{228}Ra,
Fig. 1. Concentration of $^{226}$Ra in books produced in Japan during the 20th century. ▼ indicates samples in which pyrophyllite was detected.

Fig. 2. Concentration of $^{228}$Ra in books produced in Japan during the 20th century. ▼ indicates samples in which pyrophyllite was detected.
Fig. 3. Concentration of $^{228}\text{Th}$ in books produced in Japan during the 20th century. ▼ indicates samples in which pyrophyllite was detected.

Fig. 4. Concentration of $^{40}\text{K}$ in books produced in Japan during the 20th century. ▼, ● and ○ indicate samples in which the minerals ▼, sericite; ●, mordenite; and ○, feldspar were detected.
Lalit and Shukla measured the radioactivities of 14 books printed on normal paper in Japan. For comparison, their and my data are given in Table 1. The concentrations of $^{226}$Ra and $^{228}$Th reported by Lalit and Shukla generally are higher than the corresponding data I obtained, but the differences are not large. Their $^{40}$K data are similar to mine. The average $^{137}$Cs content I found for books issued in the period 1964-1968 is comparable to that obtained by Lalit and Shukla. In contrast, the average $^{137}$Cs content I found for books issued in the period 1951-1963 is much lower than that found by Lalit and Shukla. Their data show that the average $^{137}$Cs content of books issued between 1951 and 1963 is much higher than that for books issued between 1964 and 1968. This is incompatible with the fact that the amount of $^{137}$Cs fallout deposition was highest in the first half of the 1960s. Lalit and Shukla also measured the radioactivities in books issued prior to 1950 in the U.S.A., Europe, and India and found that significant amounts of $^{137}$Cs were present in these books which were expected to be free of this nuclide. They attributed the unexpected presence of $^{137}$Cs to contamination of the books by $^{137}$Cs in their institute. The Japanese books issued between 1951 and 1963 that they tested also appear to have been contaminated with $^{137}$Cs.

Both $^{228}$Ra and $^{228}$Th are members of the thorium series. As shown in Figs. 2 and 3, the concentration of $^{228}$Th in each book is equal to that of $^{228}$Ra within the error based on counting statistics. This means that the thorium series nuclides, including the parent $^{232}$Th, are in radioactive equilibrium in the books. As seen from Fig. 2, the $^{228}$Ra concentration is highest in books issued during the 1950s, after which it decreases gradually with the year of issue, becoming fairly constant after 1975. Figures 1 and 2 resemble each other, indicative of a positive
correlation between the concentrations of $^{226}\text{Ra}$ and $^{228}\text{Ra}$. Figures 1–4 show that $^{40}\text{K}$ is similar to the three natural radioactive series nuclides in its pattern of its concentration vs. the year of issue of the book, except that in the period 1975–1993 some samples show high $^{40}\text{K}$ concentrations. $^{137}\text{Cs}$ (Fig. 5) has a completely different pattern from those of the naturally occurring nuclides (Figs. 1–4). Whereas $^{137}\text{Cs}$ was not detected in books issued before 1955, it was present in books issued in and after the second half of the 1950s. The $^{137}\text{Cs}$ concentration is highest in the mid-1960s then decreases, falling below the detection limit after 1980.

Minerals and lignin in the paper used in books

Eighty to ninety percents of the matrix of paper used for book printing is pulp, the remainder (10–20%) predominantly being filler. The amount of other constituents, such as sizing agent and ink, are negligible as compared with the amounts of pulp and filler. Filler increases the smoothness, whiteness, and opacity of paper, thereby improving its suitability for printing. Pyrophyllite, talc, kaolinite, and calcium carbonate have all been used as filler in the paper used in book printing in Japan\(^5\)–\(^7\). There are two kinds of calcium carbonate filler; heavy calcium carbonate prepared by pulverizing limestone, and light calcium carbonate prepared by precipitating calcium carbonate in aqueous solution. Heavy calcium carbonate has been the major calcium carbonate filler used in Japan.

Minerals identified by X-ray diffraction are shown in Table 2. Ten kinds of minerals were found in the papers. No other substances, except cellulose, were detected. Pyrophyllite, talc, kaolinite, and calcite, the minerals used as fillers, have other minerals associated with them. Pyrophyllite generally is associated with quartz\(^5\)\(^,\)\(^6\) and often with sericite\(^5\). This is confirmed by

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**Table 1.** Concentrations of radioactive nuclides in books printed on normal paper. Comparison of data from this study with those obtained by Lalit and Shukla (1984).

| Year of issue | Sample number | $^{137}\text{Cs}$ (Bq kg\(^{-1}\)) | $^{40}\text{K}$ (Bq kg\(^{-1}\)) | $^{226}\text{Ra}$ (Bq kg\(^{-1}\)) | $^{228}\text{Th}$ (Bq kg\(^{-1}\)) |
|---------------|---------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|
| this study    |               |                                 |                                 |                                 |                                 |
| 1917–1950     | 11            | 0.00                            | 30.5                            | 3.21                            | 5.28                            |
| 1951–1963     | 15            | 0.27                            | 62.0                            | 4.03                            | 7.03                            |
| 1964–1968     | 8             | 0.77                            | 43.8                            | 4.24                            | 5.94                            |
| 1969–1977     | 14            | 0.14                            | 27.1                            | 2.77                            | 4.43                            |
| 1978–1993     | 19            | 0.04                            | 15.9                            | 2.06                            | 2.52                            |
| Lalit and Shukla (1984)* |     |                                 |                                 |                                 |                                 |
| 1951–1963     | 10            | 5.69                            | 64.0                            | 9.29                            | 13.2                            |
| 1964–1968     | 4             | 0.88                            | 12.4                            | 3.81                            | 11.2                            |

* Their measurements were made from 1979 to 1982. Their $^{137}\text{Cs}$ data have been corrected for the decay of $^{137}\text{Cs}$ between the time of measurement and 1 January 1995 (14 y).
| Year of issue of book | pyr | tal | kao | cal | qua | ser | chl | mor | mag | fel | Lignin* |
|----------------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|--------|
| 1917                 | s   | m   | m   | w   |     |     |     |     |     |     | m      |
| 1924                 | s   |     |     |     |     |     |     |     |     |     | s      |
| 1926                 | s   | s   | s   | w   |     |     |     |     |     |     | w      |
| 1934                 | s   | s   | s   | w   |     |     |     |     |     |     | s      |
| 1937                 | s   | s   | s   | w   |     |     |     |     |     |     | w      |
| 1941                 | m   | s   | s   | w   |     |     |     |     |     |     | s      |
| 1942                 | w   | s   | w   |     |     |     |     |     |     |     | m      |
| 1944                 | m   | s   | w   |     |     |     |     |     |     |     | s      |
| 1947                 |     |     |     |     |     |     |     |     |     |     | s      |
| 1949                 | m   |     |     |     |     |     |     |     |     |     | s      |
| 1950                 | w   |     |     | w   |     |     |     |     |     |     | m      |
| 1953                 | s   | m   | s   | m   |     |     |     |     |     |     | m      |
| 1954A                | s   | m   | s   | m   |     |     |     |     |     |     | m      |
| 1954B                | s   | m   | s   | m   |     |     |     |     |     |     | m      |
| 1955                 | m   | m   | s   | w   |     |     |     |     |     |     | w      |
| 1956A                | s   |     | s   | w   |     |     |     |     |     |     | s      |
| 1956B                | m   |     | s   | w   |     |     |     |     |     |     | s      |
| 1957A                | s   | m   | s   | m   |     |     |     |     |     |     | m      |
| 1957B                | m   |     | s   | w   |     |     |     |     |     |     | s      |
| 1959                 | s   | w   | s   | w   |     |     |     |     |     |     | m      |
| 1960                 | m   |     | s   | w   |     |     |     |     |     |     | m      |
| 1961                 | w   | s   | s   | w   |     |     |     |     |     |     | s      |
| 1962A                | s   | w   | s   | w   |     |     |     |     |     |     | m      |
| 1962B                | w   | s   | m   |     |     |     |     |     |     |     | m      |
| 1963A                | m   | s   | m   |     |     |     |     |     |     |     | m      |
| 1963B                | m   |     | s   | m   |     |     |     |     |     |     | m      |
| 1964                 |     | m   | s   | s   | m   |     |     |     |     |     | m      |
| 1965A                | m   |     | w   | s   | w   |     |     |     |     |     | m      |
| 1965B                | m   | s   | s   | w   | w   |     |     |     |     |     | m      |
| 1966                 | m   | s   | s   | w   |     |     |     |     |     |     | s      |
| 1967A                | s   | m   | s   | w   | w   |     |     |     |     |     | m      |
| 1967B                | m   | s   | s   | w   | w   |     |     |     |     |     | m      |
| 1968A                | s   | s   | s   | w   | w   |     |     |     |     |     | m      |
| 1968B                |     | s   |     |     |     |     |     |     |     |     | m      |
| 1969A                | s   | s   | w   |     |     |     |     |     |     |     | s      |
| 1969B                | m   | s   | s   | w   | m   |     |     |     |     |     | m      |
| 1970                 | s   |     |     |     |     |     |     |     |     |     | s      |
| 1971A                | s   |     |     |     |     |     |     |     |     |     | m      |
| 1971B                | m   | s   | w   | s   |     |     |     |     |     |     | m      |
| 1972                 | m   | s   | s   | w   |     |     |     |     |     |     | m      |
| 1973                 | s   | s   | s   | w   |     |     |     |     |     |     | m      |
| 1974A                | m   | s   | s   | w   |     |     |     |     |     |     | m      |
| 1974B                | s   |     |     |     |     |     |     |     |     |     | s      |
| 1975A                | m   | m   | m   |     |     |     |     |     |     |     | m      |
| 1975B                | s   |     |     |     |     |     |     |     |     |     | s      |
| 1976                 | s   |     | m   |     |     |     |     |     |     |     | m      |
| 1977A                | s   |     |     |     |     |     |     |     |     |     | m      |
| 1977B                | s   |     | s   |     |     |     |     |     |     |     | s      |
the results shown in Table 2; every sample that contains pyrophyllite contains quartz. Sericite also was detected in most of the samples that contained pyrophyllite.

Pyrophyllite was mined abundantly in Japan and was the main filler for paper for books until the 1960s. Pyrophyllite filler contains a large amount of quartz. Because quartz is a hard mineral, pyrophyllite filler damages paper making machines. For this reason, pyrophyllite was replaced by talc. The mineral identifications shown in Table 2 are in good agreement with these known facts of the history of pyrophyllite filler. Table 2 shows that pyrophyllite was replaced by talc in the 1960s-1970s. Pyrophyllite was detected in most of the books published before 1975, but not in those published after 1975 except for one sample. Talc was detected in most of the books published after 1960. A mixture of pyrophyllite and talc was main filler used in the 1960s and the first half of the 1970s. Talc also was detected in several books published before the end of World War II, and it may have come from Korea or the northeast region of China.

In recent years neutralized paper has been used for books printed in Japan. Neutralized paper requires calcium carbonate as the filler. As shown in Table 2, calcium carbonate (calcite) was detected in all books printed after 1985 except for one sample. A small proportion of kaolinite usually is contained in pyrophyllite filler. The number of books in which kaolinite filler was used is less than ten of 67 (Table 2).

Results of lignin detection using phloroglucinol also are shown in Table 2. Lignin was detected in 14 samples; indicated by “s”, “m” or “w” in the column for lignin in Table 2 based on the intensity of coloration. For comparison, Japanese newspapers, which are assumed to contain more than 65% mechanical pulp, also were tested. The three book samples denoted by “s” had approximately the same intensity of coloration as the newspapers, and may contain as
much mechanical pulp as the newspapers do. The 11 samples denoted by "m" or "w" do not contain as much mechanical pulp as the newspapers. No lignin was detected in 53 samples, evidence that chemical pulp only was used for these papers. Results of the phloroglucinol color test show that chemical pulp is the main pulp used in papers for printing books.

Source of $^{137}$Cs and relation between $^{137}$Cs content and the year of issue of the book

$^{137}$Cs is a fallout nuclide of nuclear weapon testings. It is diffused in the troposphere and stratosphere, and transported to the earth's surface mainly by precipitation. The first nuclear weapon testing was conducted in 1945, and most of the atmospheric nuclear weapon testings were done from 1954 to 1963. The amount of $^{137}$Cs fallout deposited was largest in the first half of the 1960s, decreasing rapidly after that period\(^8\).

In general, fallout $^{137}$Cs is found in the surface layer of soils because it has high affinity for soil\(^8\). There is little possibility therefore that ores for fillers contain $^{137}$Cs. Perhaps the $^{137}$Cs present in the books is from the pulp used. The radionuclide could have been incorporated into the plants used as raw materials for paper pulp. All, except a few of the books, I studied are hard-cover ones. The estimated weight percentages of the papers used in printing and the hard-covers for the book studied respectively are 83% and 17%. As stated, in 20th century Japan, papers used for book printing have mostly been made from wood pulp. A number of reports\(^10\text{-}15\) have shown that wood is contaminated with fallout $^{137}$Cs. The hard-covers of books mainly are made of book cover board, which in Japan, is a straw board prepared from coarse rice straw pulp. Rice straw collected after the middle 1950s may have been contaminated with $^{137}$Cs\(^16\).

The pattern of $^{137}$Cs concentration vs. the year of publication of the book shows a marked peak in the mid-1960s (Fig. 5). Hirose et al.\(^9\) investigated the annual deposition of $^{137}$Cs from 1959 to 1984 in Japan and found the maximum annual deposition of the radionuclide in 1963. The peak of the $^{137}$Cs concentration in the books I tested is about 2 years behind the maximum annual deposition of $^{137}$Cs. Except for this delay, the $^{137}$Cs concentration pattern for the books (Fig. 5) is similar to the $^{137}$Cs fallout deposition pattern. The time-lag between the collection of plants for paper pulp production and the printing of the books in which the pulp is used is one to three years, average two years\(^1\). This time-lag explains the delay in the peak $^{137}$Cs concentration found for the books as compared to the maximum annual deposition of $^{137}$Cs. Variations in the $^{137}$Cs content of the book in terms of the year of publication (Fig. 5) may reflect changes in the $^{137}$Cs contents of the plants used as raw materials for paper pulp.

Source of $^{40}$K

As stated previously, paper used for printing books mostly is formed from chemical pulp. As judged by the method used to prepare chemical pulp, considerable amounts of inorganic constituents in wood may be lost in paper production, therefore the $^{40}$K content of chemical pulp may be considerably lower than that of wood. Book cover boards are prepared from coarse rice straw pulp which is produced by treating rice straw with hot Ca(OH)\(_2\) solution. The $^{40}$K content of the coarse rice straw pulp therefore may be considerably lower than that of raw rice straw.

The potassium content of wood generally is about 0.1%\(^10,13,15,17\). The $^{40}$K content of wood
is, accordingly, about 32 Bq kg\(^{-1}\). The potassium content of rice straw is about 2%\(^{18}\), and the \(^{40}\)K content about 630 Bq kg\(^{-1}\). Estimation of the amount of \(^{40}\)K contributed by plants to a book might aid the elucidation of the origin of \(^{40}\)K in books. Because potassium is lost in pulping, the upper limit of the concentration of the \(^{40}\)K from plants can be estimated on the basis of the following assumptions: (1) the book is composed of wood pulp (72%), filler (11%), and coarse rice straw pulp (17%); (2) the \(^{40}\)K contents of the wood pulp and coarse rice straw pulp are less than those of the raw materials; i.e. 32 and 630 Bq kg\(^{-1}\), respectively. The estimated limit therefore is 130 Bq kg\(^{-1}\). All the \(^{40}\)K concentrations found for the books tested (Fig. 4) are below this limit. Estimation of the upper limit of the concentration of \(^{40}\)K from plants does not, however, clarify the source of the \(^{40}\)K in the books, the extent of potassium loss in pulping being unknown.

Identification of the minerals in the papers used for printing provides important information about the source of \(^{40}\)K in books. The chemical formulas of the minerals found in these papers are pyrophyllite, \(\text{Al}_2\text{Si}_4\text{O}_{10}(\text{OH})_2\); kaolinite, \(\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4\); talc, \(\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2\); calcite, \(\text{CaCO}_3\); quartz, \(\text{SiO}_2\); chlorite, \((\text{Mg}, \text{Fe}, \text{Al})_{12}(\text{Si}, \text{Al})_8\text{O}_{20}(\text{OH})_{16}\); sericite, \(\text{K}_2\text{Al}_4(\text{Si}_6\text{Al}_2)\text{O}_{20}(\text{OH}, \text{F})_4\); mordenite, \((\text{Na}_2,\text{K}_2,\text{Ca})\text{Al}_2\text{Si}_{10}\text{O}_{24}\cdot 7\text{H}_2\text{O}\); magnesite, \(\text{MgCO}_3\); and feldspar, \((\text{Na},\text{K})\text{AlSi}_3\text{O}_8\) or \(\text{CaAl}_2\text{Si}_2\text{O}_8\). Sericite, mordenite, and feldspar contain potassium as a major constituent. The concentrations of \(^{40}\)K in the minerals calculated from their chemical formulas are sericite, 3060–3090 Bq kg\(^{-1}\); mordenite, 0–2670 Bq kg\(^{-1}\); and feldspar, 0–4420 Bq kg\(^{-1}\). The other minerals contain potassium as a trace constituent. The \(^{40}\)K concentrations in the books studied can be estimated from the potassium minerals present. If a book contains 2% sericite by weight, the \(^{40}\)K content is about 60 Bq kg\(^{-1}\). Figure 4 shows the samples in which sericite, mordenite, or feldspar was found. \(^{40}\)K concentrations clearly are high in samples that contain sericite, mordenite, or feldspar, indicative that the bulk of the \(^{40}\)K in these books is due to the potassium-containing filler. It also indicates that \(^{40}\)K of plant origin makes a relatively small contribution to the \(^{40}\)K concentration in books (less than 5 Bq kg\(^{-1}\)) as compared with its estimated upper limit (130 Bq kg\(^{-1}\)) and that the major part of the potassium in plants is lost during pulping.

**Source of natural radioactive series nuclides**

The \(^{226}\)Ra concentration in plants generally is about 0.37 Bq kg\(^{-1}\) (0.01 pCi g\(^{-1}\))\(^{2}\). The \(^{226}\)Ra concentration in wood is assumed to be comparable to the general \(^{226}\)Ra concentration in plants\(^{15}\). Several reports on the \(^{228}\)Ra and \(^{226}\)Ra contents of plants\(^{3,15,19}\) show that mostly they are comparable. The \(^{226}\)Ra and \(^{228}\)Ra concentrations in most of the books are more than 5 times the general \(^{226}\)Ra and \(^{228}\)Ra concentrations in plants (Figs. 1 and 2), suggesting that the major percentages of the radium isotopes found in the books was not from plants.

In plants the radioactivity of \(^{232}\)Th generally is much less than the radioactivities of \(^{228}\)Ra and \(^{226}\)Ra\(^{3}\) because plants absorb radium preferentially to thorium\(^{2,20,21}\). In contrast, the radioactivity of \(^{232}\)Th in the books tested is comparable to the radioactivities found for \(^{228}\)Ra and \(^{226}\)Ra, as judged from the radioactive equilibrium of the thorium series nuclides in these books. This also indicates that plants are not the major source of the natural radioactive series nuclides in the books.
As reported, pyrophyllite, talc, kaolinite, and calcium carbonate (limestone) are fillers used in the papers for printing books. These materials contain significant amounts of naturally occurring radionuclides. Beretka and Mathew reported the respective $^{226}\text{Ra}$ and $^{232}\text{Th}$ contents of a pyrophyllite to be 115 and 167 Bq kg$^{-1}$, and the average respective $^{226}\text{Ra}$ and $^{232}\text{Th}$ contents of 4 clays (including a kaolinite) to be 63 and 163 Bq kg$^{-1}$. Doi et al. found uranium and thorium concentrations in a talc of 0.5 and 0.4 ppm, respectively; 6.2 Bq kg$^{-1}$ for $^{238}\text{U}$ and 1.6 Bq kg$^{-1}$ for $^{232}\text{Th}$. Rankama and Sahama reported that the respective average $^{226}\text{Ra}$ and $^{232}\text{Th}$ contents of limestones were 15.5 and 5.2 Bq kg$^{-1}$. These concentrations of naturally occurring radionuclides in fillers explain the $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ concentrations in the books, because the $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ in the books appear to be mostly in the filler.

Paper towels contain little filler, being essentially pure pulp. The radioactivity in two paper towel samples was measured using a method similar to that used for the books. Results showed that the contents of $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ in the paper towels were less than 0.6 Bq kg$^{-1}$, indicative that most of the $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ present in the books are in the filler.

Relation between the concentrations of naturally occurring radionuclides and the year of publication of a book

The pyrophyllite deposits in Japan were formed by the hydrothermal alteration of rhyolites. The concentrations of the natural radioactive series nuclides in pyrophyllite are much higher than those in talc and limestone. Figures 1–3 shows that books containing pyrophyllite filler have higher concentrations of $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ than do books that contain no pyrophyllite filler. The concentrations of $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ in the books depend on the kind of filler used. The patterns of the $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ concentrations vs. the year of issue of the book (Figs. 1–3) can be seen by the change in the kind of filler used. Radionuclide concentrations are highest in the 1950s because of the exclusive use of pyrophyllite as filler in this period, then there is a gradual decrease because a mixture of pyrophyllite and talc constituted the main filler from 1960 to 1975, after which, when pyrophyllite was rarely used as filler, the concentrations become relatively constant.

A correlation between the concentration of $^{40}\text{K}$ and the concentrations of $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ was found in my study. Pyrophyllite filler contains much $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$, and a large amount of $^{40}\text{K}$ as well, because it usually contains the potassium mineral, sericite, which explains the above correlation. The $^{40}\text{K}$ concentration present in books also depends greatly on the kind of filler.

As stated, the pattern for $^{40}\text{K}$ (Fig. 4) is similar to the patterns for $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ (Figs. 1–3) but there is a difference: Some books issued after 1975 have high $^{40}\text{K}$ concentrations whereas after 1975 the concentrations of $^{226}\text{Ra}$, $^{228}\text{Ra}$, and $^{228}\text{Th}$ are fairly constant. The samples with high $^{40}\text{K}$ concentrations contain another potassium mineral, mordenite (Fig. 4), which is sometimes associated with talc. This is the reason for the high $^{40}\text{K}$ concentrations.

The radioactivity of $^{137}\text{Cs}$ in books is lower than the radioactivities of $^{226}\text{Ra}$, $^{228}\text{Ra}$, $^{228}\text{Th}$, and $^{40}\text{K}$. The concentrations of naturally occurring radionuclides in books depend on the kind of filler used. Consequently, the radioactivity in books is dependent on the kind of filler. In Japan, the change in the type of filler from pyrophyllite to other materials decreased the radioactivity
Increase of the dose equivalent rate caused by radioactivity in books

The radioactivity in books may produce an increase in the radiation experienced in libraries. Whether there is an increase in radiation dose depends on several factors; the number of books, the radioactivity content of books, and the arrangement of the books. Nevertheless, the magnitude of the increase in radiation dose can be estimated by postulating a model of a library book stack. The increase in the dose equivalent rate due to radioactivity in Japanese books was calculated using the model described and the following assumptions (1)–(3):

1. Books were compactly stacked on shelves arranged on the floor in a circle 2 m in diameter. The stack of books was 2 m high and 0.3 m thick. The increase in the 1 cm dose equivalent rate at a point 1 m above the center of the circle was estimated.

2. The density of the books was 0.78 g cm\(^{-3}\) (the average density of the books in this study).

3. The respective concentrations of \(^{226}\text{Ra}, \; ^{232}\text{Th}, \; ^{40}\text{K},\) and \(^{137}\text{Cs}\) in the books were 3, 5, 40, and 0.2 Bq kg\(^{-1}\) (the average concentrations of the nuclides in the books in this study). Thorium series nuclides were in radioactive equilibrium in the books as was \(^{226}\text{Ra}\) and its descendants, and all the radionuclides were uniformly distributed.

Under the above conditions, the total weight of the books was 2940 kg, and the respective total radioactivities of \(^{226}\text{Ra}, \; ^{232}\text{Th}, \; ^{40}\text{K},\) and \(^{137}\text{Cs}\) in the books were 8820, 14700, 118000, and 588 Bq.

The 1 cm dose equivalent rate, \(D\), at the distance of \(r\) from the radiation source of a radionuclide is given by

\[
D = \frac{A \times C \times T}{r^2} \tag{Eq. 1}
\]

where \(A\) is the radioactivity of the nuclide, \(C\) the 1 cm radiation dose equivalent rate constant of the nuclide, and \(T\) the transmittance of a 1 cm dose equivalent of the nuclide. The increase in the 1 cm dose equivalent rate caused by the books can be calculated by Eq. 1. The values of \(A\) for \(^{226}\text{Ra}, \; ^{232}\text{Th}, \; ^{40}\text{K},\) and \(^{137}\text{Cs}\) have been given above. The values of \(C\) for \(^{226}\text{Ra}, \; ^{232}\text{Th}, \; ^{40}\text{K},\) and \(^{137}\text{Cs}\) respectively are \(2.52 \times 10^{-7}\), \(3.28 \times 10^{-7}\), \(2.08 \times 10^{-8}\), and \(9.10 \times 10^{-8}\) \(\mu\text{Sv} \; \text{m}^2 \; \text{Bq}^{-1} \; \text{h}^{-1}\) (the values for \(^{226}\text{Ra}, \; ^{232}\text{Th},\) and \(^{137}\text{Cs}\) include the contributions of their descendants in radioactive equilibrium). Radiation from the books was shielded on the average by a 15 cm thick (11.7 g cm\(^{-2}\)) matrix of the books. Taking into account the elemental compositions of this matrix of books and concrete, the 11.7 g cm\(^{-2}\) thick matrix of books is approximately equivalent to 11.7 g cm\(^{-2}\) thick concrete with respect to its shielding ability; therefore, the values of transmittance for a 1 cm dose equivalent to 11.7 g cm\(^{-2}\) thick concrete were adopted as the values for \(T\) in Eq. 1. The values of transmittance for the 1 cm dose equivalents for \(^{226}\text{Ra}, \; ^{232}\text{Th}, \; ^{40}\text{K},\) and \(^{137}\text{Cs}\) through 11.7 g cm\(^{-2}\) (5.1 cm) thick concrete are approximately equal (0.83)\(^{26}\). The average distance from the side surface of a cylinder 2 m in diameter and 2 m high to the center of that cylinder is 1.15 m. This value was adopted for \(r\). By substituting the values for \(A, \; C, \; T\) and \(r\) of each radionuclide in Eq. 1, the dose equivalent rate for each radionuclide in the books could be calculated. The increase in the dose equivalent rate due to the books at the point...
of estimation was calculated by summing the dose equivalent rates for $^{226}\text{Ra}$, $^{232}\text{Th}$, $^{40}\text{K}$, and $^{137}\text{Cs}$ in the books. The increase in the 1 cm dose equivalent rate caused by the books was 0.0060 $\mu$Sv h$^{-1}$. This value is about 7% of the dose equivalent rate produced by natural external sources in the normal background area (800 $\mu$Sv y$^{-1}$, i.e. 0.09 $\mu$Sv h$^{-1}$)\textsuperscript{27}.

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REFERENCES

1. Lalit, B. Y. and Shukla, V. K. (1984) Radioactivity in books produced during the last 50 years. Health Phys. 47: 447–451.
2. Osburn W. S. (1965) Primordial radionuclides: their distribution, movement, and possible effect within terrestrial ecosystems. Health Phys. 11: 1275–1295.
3. Kobashi, A. and Tominaga, T. (1985) $^{228}\text{Ra} - ^{228}\text{Th}$ dating of plant samples. Int. J. Appl. Radiat. Isotop. 36: 547–553.
4. Takahashi, H., Koizumi, Y., Sato, K. and Sato, J. (1989) Background of low-level gamma-ray spectrometer due to the atmospheric $^{220}\text{Rn}$ and $^{222}\text{Rn}$. Radioisotopes 38: 144–147 (in Japanese with English abstract).
5. Matsumoto, K. (1963) Filler. Japan Tappi J. 17: 80–87 (in Japanese).
6. Nakada, K. (1987) Clay for paper making: In "Handbook of clay (2nd edition)\textsuperscript{nd}, Ed. Japan Clay Society, pp. 887–897. Gihodo, Tokyo (in Japanese).
7. Kuroda, S. (1987) Filler and chemicals. Japan Tappi J. 41: 943–947 (in Japanese).
8. Eisenbud, M. (1973) Environmental radiation (2nd edition). Academic Press, New York.
9. Hirose, K., Aoyama, M., Katsuragi, Y. and Sugimura, Y. (1987) Annual deposition of Sr-90, Cs-137 and Pu-239, 240 from the 1961–1980 nuclear explosions: a simple model. J. Meteor. Soc. Japan 65: 259–277.
10. Chigira, M., Saito, Y. and Kimura, K. (1988) Distribution of strontium-90 and cesium-137 in annual tree rings of Japanese cedar, Cryptomeria japonica D. Don. J. Radiat. Res. 29: 152–160.
11. Kohno, M., Koizumi, Y., Okumura, K. and Mito, I. (1987) Distribution of environmental cesium-137 in tree rings. J. Environ. Radioact. 8: 15–19.
12. Momoshima, N. and Bondietti, E. A. (1994) The radial distribution of $^{89}\text{Sr}$ and $^{137}\text{Cs}$ in trees. J. Environ. Radioact. 22: 93–109.
13. Plummer, G. L. and Helseth, F. (1965) Movement and distribution of radionuclides on granitic outcrops within the Georgia Piedmont. Health Phys. 11: 1423–1428.
14. Russell, I. R. (1971) Distribution of fallout nuclides in a 26 year white pine, U. S. Atomic Energy Commission NYO-3756-4.
15. Russell, I. R. (1971) Radial distribution of fallout nuclides and radium isotopes in a 107 year oak trunk. U. S. Atomic Energy Commission NYO-3756-7.
16. Ichikawa, R., Eto, M. and Abe, M. (1962) Strontium-90 and cesium-137 absorbed by rice plants in Japan, 1960. Science 138: 1072–1072.
17. Nikitin, N. I. (1966) The chemistry of cellulose and wood. Oldbourne Press, London.
18. Ishizuka, Y. and Tanaka, A. (1969) Nutrition physiology of rice plants (revised and enlarged edition). Yokendo, Tokyo (in Japanese).
19. Smith, K. A. (1971) The comparative uptake and translocation by plants of calcium, strontium, barium and radium. II. *Triticum vulgare* (wheat). Plant and Soil 34: 643–651.
20. D'Souza, T. J. and Mistry, K. B. (1970) Comparative uptake of thorium-230, radium-226, lead-210 and polonium-210 by plants. Radiation Botany 10: 293–295.
21. Verkhovskaja, I. N., Vavilov, P. P. and Maslov, V. I. (1967) The migration of natural radioactive elements under natural conditions and their distribution according to biotic and abiotic environmental components: In “Proceedings of international symposium on radioecological concentration processes” pp. 313–328.
22. Beretka, J. and Mathew, P. J. (1985) Natural radioactivity of Australian building materials, industrial waste and by-products. Health Phys. 48: 87–95.
23. Doi, H., Tsuchimoto, M. and Ono, M. (1985) A chemical condensation technique for detection of uranium and thorium contained in LSI constituent materials. Denki Kagaku oyobi Kogyo Butsuri Kagaku 53: 282–286 (in Japanese with English abstract).
24. Rankama, K. and Sahama, T. G. (1950) Geochemistry. University of Chicago Press, Chicago.
25. Japan Radioisotope Association (1990) Isotope techo (Pocketbook of isotopes). Japan Radioisotope Association, Tokyo (in Japanese).
26. Ichimiyi, T. (1990) Transmission of 1 cm dose equivalent. Radioisotopes 39: 124–133 (in Japanese).
27. United Nations Scientific Committee on the Effects of Atomic Radiation (1988) Sources, effects and risks of ionizing radiation. United Nations, New York.