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Articles

Radioactivity of ^{137}Cs in Papers and Migration of the Nuclide in the Environment

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The ^{137}Cs contents of papers such as magazines and newspapers printed in Japan were determined by gamma-ray spectrometry. The average ^{137}Cs content of weekly general interest magazines was 0.95 Bq kg^{-1} and far higher than those of other kinds of samples. The result of phloroglucinol color test showed that the main printed pages of the weekly general interest magazines were chiefly made of mechanical pulp. Possibly the pages held ^{137}Cs brought by wood, raw material of paper, and that accounted for the high ^{137}Cs content of the magazines. The experimental result suggested that ^{137}Cs was removed from wood into aqueous solution in the production of chemical pulp. Cesium-137 radioactivity removed during paper manufacturing in the year 2000 in Japan was estimated to be 15 GBq, and the influence of paper manufacturing on the migration of ^{137}Cs in the environment was discussed.

1. Introduction

In the previous researches[1-4] the author analyzed naturally occurring radionuclides (^{226}Ra, ^{228}Ra, ^{228}Th, and ^{40}K) and a fall-out nuclide ^{137}Cs (half-life 30.07 y) in papers such as books, magazines, and newspapers and elucidated the radioactivity levels and the sources of the nuclides in the samples.

A large amount of paper are produced, consumed, discarded, and recycled in developed countries. In Japan, 3.11×10^{10} kg of paper and paperboard was produced, and 1.88×10^{10} kg of waste paper was recycled in the year 2006.[5] Raw materials of paper, paper, and waste paper migrate in the environment with these processes. As a result, radionuclides contained in the raw materials, paper, and waste paper also migrate in the environment with the processes.

The author[1-3] has shown that the source of ^{137}Cs contained in papers is plants used as raw material of pulp. Cesium-137 is present in the environment mainly as a product of nuclear weapon tests. A part of the ^{137}Cs is taken in plants and then incorporated into paper making, discarding, and recycling processes. How the radionuclide behaves in the environment with the processes is not clear at all. The analysis of ^{137}Cs in various paper samples may give a clue to the elucidation of the behavior of the nuclide. In this article, the author reports the result of measurement of ^{137}Cs radioactivity in paper samples and discusses the influence of paper manufacturing on the migration of ^{137}Cs in the environment.

2. Experimental

2.1. Sample description. Pocket-sized books, weekly photo magazines, magazines full of color pages, weekly general interest magazines, weekly comic magazines, and newspapers 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 details of the samples are as follows:

(A) Pocket-sized book. The samples were pocket-sized books issued from five different large publishing companies. The jacket covers were removed, and so the samples were made up of printed pages and a cover. The weight percentage of printed pages ranged from 97 to 98%.

(B) Weekly photo magazine. The samples were weekly or biweekly magazines with a large circulation from different four publishing companies. They were composed of a cover and photo pages made of coated paper printed in a single color or many colors.

(C) Magazine full of color pages. The samples were monthly magazines published from different publishing companies. They were mainly made of coated paper printed in many colors.

(D) Weekly general interest magazine. The samples were weekly general interest magazines with a large circulation from four different large publishing companies. They consisted of a cover, pages printed in many colors, monochrome photo pages (coated paper was used for the above parts), and main printed pages made of low-grade paper. The proportions of the above four parts were similar among the four samples. The weight percentage of the main printed pages ranged between 55 and 63%.

(E) Weekly comic magazine. The samples were four weekly comic magazines for boys with a large circulation from different publishing companies. They consisted of a cover, pages printed in many colors (coated paper was used for the above two parts), and main printed pages made of low-grade paper. The proportions of the three parts were similar among the four samples. The weight percentage of the main printed pages ranged from 95 to 96%.

(F) Printing paper. The samples were two kinds of printing paper prepared by different paper manufacturers.

(G) Copying paper. The samples were three kinds of PPC paper prepared by different paper manufacturers.

(H) Notebook. The samples were four kinds of notebooks made by different companies. Three of them were ordinary notebooks with a thin cover whereas the other was a loose-leaf notebook without a cover.

(I) Newspaper. The samples were three newspapers issued from major newspaper publishing companies.

2.2. Gamma-ray spectrometry. The size of each sample was adjusted to 210 mm long, 150 mm wide, and 30 mm thick by cutting and stacking the paper sheets forming the sample. In case of a sample which was composed of different kinds of paper, the paper sheets of different kinds were arranged to be uniformly distributed in the sample. The samples were sealed

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in two nylon bags to prevent contamination of the gamma-ray spectrometer used for the measurement.

Gamma-ray spectrometry was made by using a Canberra GC-series gamma-ray spectrometer equipped with a pure germanium detector of effective volume of 100 cm^3 and a 4096 channel pulse-height analyzer. The detector was placed in a shielding box made up of layers of 50-mm lead, 50-mm copper, and 2-mm acrylic resin. The spectrometer was placed in a well ventilated, air conditioned room to ensure low background counts. For each sample, measurement was made for 3 days. Cesium-137 was determined by the intensity of 662 keV gamma-rays. The details of the method were described in the previous paper.[3] The ^{137}Cs analysis was carried out simultaneously with the analysis of naturally occurring radionuclides. The results of analysis of naturally occurring radionuclides were reported in the author's other article.[4]

2.3. Phloroglucinol color test. Paper is mainly made of pulp. More than 99% of papermaking pulp has been prepared from wood for 50 years in Japan. The major components of wood are cellulose, hemicellulose, and lignin. Cellulose and hemicellulose are fibrous materials, whereas lignin not. Wood also contains inorganic constituents, resin and so on as minor components.

Papermaking pulps are classified into mechanical pulp and chemical pulp according to their preparation method. Mechanical pulp is prepared by grinding wood by a machine. On the other hand, chemical pulp is prepared by cooking wood in aqueous solution such as NaS-NaOH mixture solution and then by eliminating lignin and other useless constituents chemically to obtain fibrous component. However, there are intermediate pulps between mechanical pulp and chemical pulp, and there are papers which are prepared by mixing mechanical pulp and chemical pulp. Phloroglucinol reacts with lignin and dyes it reddish purple, and the reagent is used to detect lignin in paper and to estimate lignin content of paper.[6] The test using the reagent is not a quantitative method but makes it possible to know the extent of purification of pulp.

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. Each sample was divided after the gamma-ray spectrometry into parts based on the difference of paper used for the parts. The weight of each part was measured, and the weight percentage of each part was calculated. A piece of paper was cut from each part. Each part was assumed to be uniform in chemical composition, and the piece cut from it was assumed to be representative of the whole part. A drop of phloroglucinol solution was placed on the piece, and the degree of reddish purple coloration was checked. The details of the phloroglucinol color test were described in the previous paper.[1] The intensities of reddish purple color of the tested pieces were mutually compared by naked eyes. The intensity varied from dark reddish purple to colorless, and it was possible to classify the tested pieces into six groups depending on the color intensity. Based on the result of the comparison, the lignin content of each part was evaluated to be one of the six levels as follows: very high, high, medium, low, very low, and not detected.

3. Results and Discussion

3.1. Cesium-137 contents of the paper samples. The result of gamma-ray spectrometry is shown in Figure 1. The ^{137}Cs contents were corrected for the decay to January 1, 2000. The errors in the ^{137}Cs contents shown in the figure are standard deviations estimated on the counting statistics.

As shown in Figure 1, ^{137}Cs is detected in the weekly general interest magazines, weekly comic magazines, and newspapers, whereas the radionuclide is not detected in the other kinds of samples. The average concentrations of the weekly general interest magazines, weekly comic magazines, and newspapers are respectively 0.95, 0.06, and 0.11 Bq kg^{-1}. It is evident that the weekly general interest magazines contain far more ^{137}Cs than the other kinds of samples.

As already mentioned, most of pulp have been prepared from wood for many years in Japan. It is reasonable to assume

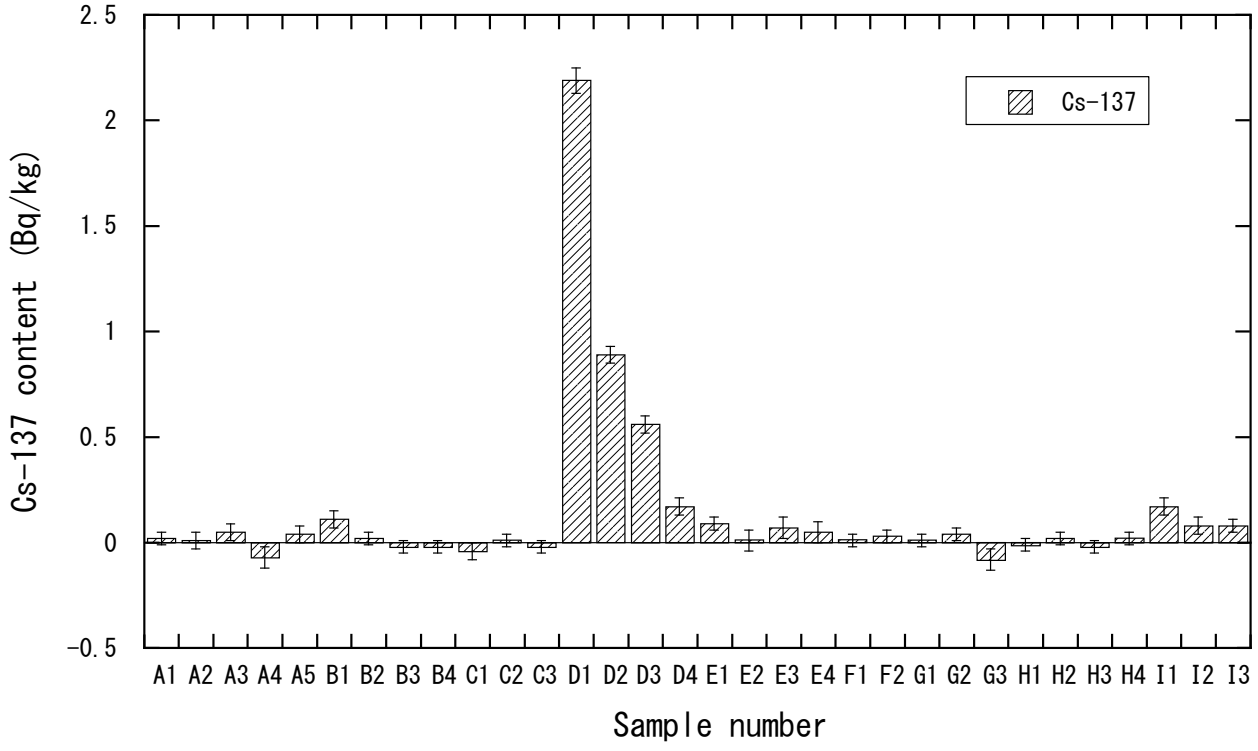

Figure 1. Concentration of ^{137}Cs in paper samples. Kinds of samples are as follows: A, pocket-sized book; B, weekly photo magazine; C, magazine full of color pages; D, weekly general interest magazine; E, weekly comic magazine; F, printing paper; G, copying paper; H, notebook; I, newspaper. The ^{137}Cs concentrations have been decay corrected to January 1, 2000.

that the raw material for pulp of the paper samples tested in this work is wood. Several works[7-10] were done to investigate ^{137}Cs concentration in rings of trees that experienced the time of fallout due to nuclear weapon tests. The data of the works are summarized in Table 1. As shown in Table 1, the ^{137}Cs content depends on the species of trees and the localities of trees. The ^{137}Cs content of whole wood of each tree was estimated by averaging the ^{137}Cs contents of all the tree rings, and the obtained value was corrected for the decay to January 1, 2000. The result of the estimation is also shown in Table 1. As can be seen from Table 1, the ^{137}Cs content of whole wood varies from 0.01 to 4.1 Bq kg^{-1}, and the average of all the ^{137}Cs contents of whole wood is 0.88 Bq kg^{-1}. The ^{137}Cs contents of the weekly general interest magazines examined in this work are comparable to the average ^{137}Cs content of whole wood thus obtained.

The remarkable difference in ^{137}Cs concentration between the weekly general interest magazines and the other kinds of samples can be interpreted as follows: the greater part of ^{137}Cs brought by material wood remained in the weekly general interest magazines while the nuclide was little left in the other kinds of samples.

3.2. The behavior of ^{137}Cs in the production of paper. It is necessary to verify the above interpretation. Cesium is an alkali metal element and has the property of dissolving easily in water. It is likely that ^{137}Cs contained in wood is dissolved in aqueous solution by chemical treatment done to wood in production of paper. Potassium is also an alkali metal element, and it is contained in wood because it is essential to plants. The author[4] already studied the ^{40}K contents of the paper samples tested in this work, and the result of the study suggested the dissolution of ^{40}K in the production of paper.

The most important factor affecting ^{137}Cs radioactivity left in paper is the intensity of chemical treatment in preparation of pulp. In preparation of pulp, a part of ^{137}Cs contained in wood is dissolved in aqueous solution while the rest of ^{137}Cs is left in pulp. Mechanical pulp will hold most of ^{137}Cs contained in wood because it is made simply by grounding wood. On the other hand, almost no ^{137}Cs will remain in chemical pulp, especially in high-grade chemical pulp, because chemical pulp is prepared by eliminating lignin and other useless constituents from wood in aqueous solution to obtain pure fibrous material. Therefore, the lignin content of paper is a good measure of ^{137}Cs left in pulp and paper.

Another important factor affecting ^{137}Cs radioactivity left in paper is deinking process in recycling paper. Deinked pulp is produced by taking waste paper apart in aqueous solution and removing ink from the waste paper chemically. Deinking is a kind of chemical treatment, and ^{137}Cs will be lost from the waste paper owing to the treatment. In the deinking process, a small proportion of ink inevitably remains in pulp as flakes adhered to pulp fibers. Therefore, flakes of residual ink are characteristic of deinked pulp.

The result of the phloroglucinol color test is shown in Figure 2. As seen from Figure 2, a part exhibiting high or very high lignin content is present in each of weekly general interest magazines (D1-D4). The part is the main printed pages of the magazines. Evidently the pages are chiefly made of mechanical pulp. The pages were observed by a microscope to examine the presence of flakes of residual ink. Such flakes were not found in the pages, showing that the pages are chiefly made of virgin mechanical pulp. Therefore, the main printed pages hold most of ^{137}Cs brought by wood. Because the proportions of the main printed pages are more than a half (55-63%), the ^{137}Cs contents of the weekly general interest magazines are high (Figure 1).

Figure 2 shows that the main printed pages of two weekly comic magazines (E3 and E4) contain high concentration of lignin. The figure also shows that all the newspaper samples (I1-I3) contain medium concentration of lignin. Cesium-137 was detected in the two weekly comic magazines and the newspapers (Figure 1). The result of the phloroglucinol color test indicates that considerable amount of mechanical pulp is present in the weekly comic magazines and newspapers, and that is the reason why ^{137}Cs remained enough to be detected in these

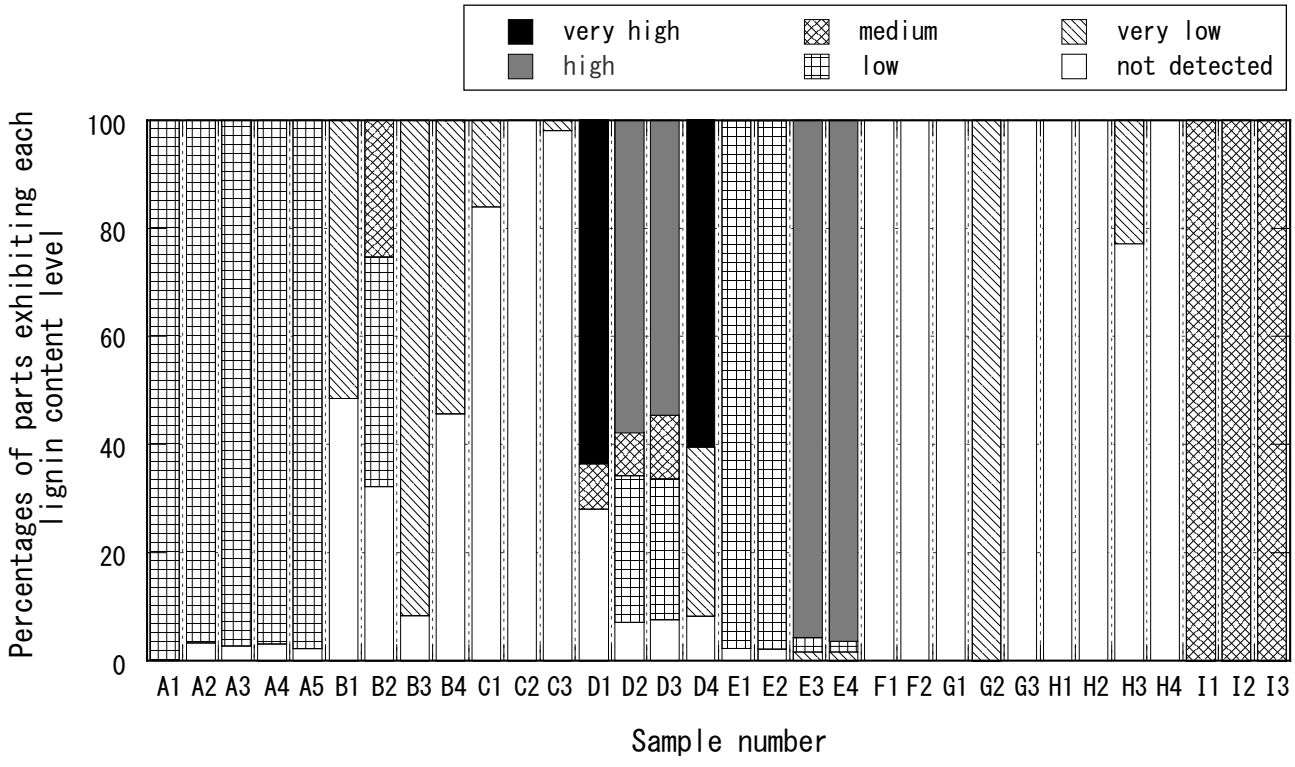

Figure 2. Lignin content and weight percentage of each part of paper samples. Kinds of samples are as in Figure 1. The lignin contents were evaluated into six levels on the basis of the result of phloroglucinol color test.

samples. However, the ^{137}Cs contents of the samples are much lower than those of weekly general interest magazines (Figure 1). Oji Paper Co. Ltd.[11] reported that main printed pages of Japanese weekly comic magazines were made of nearly 100% deinked pulp. Takeshita[12] reported that about 45% of pulp used for recent Japanese newspapers was deinked pulp. Possibly waste paper containing much mechanical pulp was the raw material of the deinked pulp used for the weekly comic magazines and newspapers tested in this work. The main printed pages of the weekly comic magazines and newspapers examined in this work were observed by a microscope. Many flakes of residual ink were found on the samples. Thus it is confirmed that deinked pulp is used for the samples, and that explains the reason for the lower ^{137}Cs contents of the weekly comic magazines and newspapers compared with the weekly general interest magazines. Another reason for the lower ^{137}Cs contents of the newspapers is the lower proportions of mechanical pulp in the newspapers as compared with the weekly general interest magazines.

As can be seen from Figure 2, all the samples other than weekly general interest magazines, weekly comic magazines, and newspapers are composed of parts of lignin content lower than medium level except for B2 sample. Although a part of B2 sample exhibits medium level of lignin content, the weight percentage of the part is 25%. Consequently, the lignin content of the whole B2 sample is low level. Judging from the result of the phloroglucinol color test, all the samples other than general interest magazines, weekly comic magazines, and newspapers are essentially made of chemical pulp. This is in agreement with the result that ^{137}Cs was not detected in these samples (Figure 1).

The above discussion clearly accounts for the difference in ^{137}Cs concentration between the weekly general interest magazines and the other kinds of samples.

3.3. The radioactivity of ^{137}Cs removed from wood into aqueous solution during paper manufacturing in Japan. The above discussion has revealed that most of ^{137}Cs contained in material wood dissolves into aqueous solution in preparation of chemical pulp. This result makes it possible to estimate ^{137}Cs radioactivity removed from wood to aqueous solution in paper manufacturing in Japan.

The weight percentages of pulps prepared in 2000 in Japan were as follows: kraft pulp, 85.9%; half-chemical pulp, 0.9%; mechanical pulp, 12.3%; others, 0.9%.[13] Thus most of pulp prepared in Japan is kraft pulp, a kind of chemical pulp. The radioactivity A of ^{137}Cs removed from wood into aqueous solution during paper manufacturing in Japan is calculated by the following equation:

$$A = V\,r\,d\,C \tag{1}$$

where V is the volume of wood used for preparation of pulp, r the proportion of wood used to prepare chemical pulp, d the density of the wood, and C the average ^{137}Cs content of the wood. The consumption of pulp wood in 2000 in Japan[13] (3.76×10^7 m^3) is adopted as the value of V. The value of r is estimated to be 0.93 based on the above pulp production data and the data on yields of kraft pulp and mechanical pulp from wood (45% and 91%, respectively[14]). On the basis of the data on the density of typical woods used in Japan[15], 500 kg m^{-3} is adopted as the value of d. The average ^{137}Cs content of wood estimated in Table 1 (0.88 Bq kg^{-1}) is adopted as the value of C. The radioactivity A of ^{137}Cs removed from wood to aqueous solution during paper manufacturing in the year 2000 in Japan is calculated to be 1.5×10^{10} Bq (15 GBq) by eq 1.

3.4. Migration of ^{137}Cs in the environment. The environmental contamination with ^{137}Cs began with the first nuclear test in July 1945.[16] It became global because many atmospheric tests of nuclear weapons were done after the World War II.

TABLE 1: ^{137}Cs concentration in wood

Tree	Locality	Growth period	Range of ^{137}Cs concentration in tree rings* (Bq kg^{-1})	Reference	^{137}Cs concentration in whole wood** (Bq kg^{-1})	Corrected ^{137}Cs concentration in whole wood*** (Bq kg^{-1})
White oak	Massachusetts, USA	1860-1967	0.94-9.3	7)	3.8	1.8
Red spruce	Tennessee, USA	1813-1987	4.4-7.2	8)	5.5	4.1
Eastern hemlock	Tennessee, USA	1887-1986	1.4-4.7	8)	2.9	2.1
Eastern white pine	Tennessee, USA	1916-1987	0.07-0.36	8)	0.22	0.16
Hickory	Tennessee, USA	1800-1987	0.16-0.67	8)	0.27	0.2
Elm	Tennessee, USA	1934-1987	0.08-0.27	8)	0.15	0.11
American beech	Tennessee, USA	1870-1987	0.8-2.8	8)	1.8	1.3
Sugar maple	Tennessee, USA	1920-1987	0.0-0.35	8)	0.09	0.07
Yellow poplar	Tennessee, USA	1907-1987	0.0-0.05	8)	0.02	0.01
Japanese cedar	Chiba Pref., Japan	1914-1979	0.07-1.48	9)	0.78	0.49
Cypress	Ehime Pref., Japan	1929-1979	0.44-1.73	9)	0.96	0.6
Japanese cedar	Tokyo, Japan	1914-1982	0.09-0.63	10)	0.33	0.22
Japanese cedar	Kanagawa Pref., Japan	1930-1982	0.26-1.07	10)	0.48	0.32
Average ^{137}Cs concentration in whole wood						0.88

* The original ^{137}Cs data in the literatures were given in ^{137}Cs concentration variation in tree rings at the time of felling down the tree.

** The ^{137}Cs concentrations in this column are the ^{137}Cs concentrations in whole wood obtained by averaging ^{137}Cs cocentrations in all the tree rings.

*** The ^{137}Cs concentrations in this column are obtained by decay-correcting the ^{137}Cs concentrations in the left column to January 1, 2000.

The ^{137}Cs contamination of the surface of the earth was caused by the deposition of the radionuclide from the air where the nuclide was produced by nuclear weapon testing or released from nuclear facilities such as nuclear reactors. The deposition of ^{137}Cs fallout on the surface of the earth had a peak in the first half of the 1960s and then has been decreasing except for the temporary increase due to the Chernobyl accident.[17] Fallout ^{137}Cs is adsorbed on the surface of plants and is gradually absorbed into the plant bodies. Plants have a tendency to absorb more ^{137}Cs from their parts in contact with the air such as leaves than from their root.[16] Therefore, the ^{137}Cs content of annual plants shows a time-dependent change similar to that of ^{137}Cs fallout deposition on the earth surface.

As for annual plants, Komamura et al[18,19] studied the ^{137}Cs contents of rice and wheat collected at a number of localities in Japan. Their result indicated that the ^{137}Cs contents of rice and wheat were the highest for the samples collected in 1963. The average ^{137}Cs contents of rice and wheat samples collected in 1995 were respectively 0.046 and 0.018 Bq kg^{-1}, and the values were two or three orders of magnitude below the corresponding values of samples collected in 1963.

Trees are perennial and live long lives. Therefore, trees which grew after July 1945 hold ^{137}Cs in their bodies. Man uses wood, xylem part of trees, and ^{137}Cs contained in wood migrates in the environment with the activity of man. Wood has been used as fuel, construction material, raw material for pulp, etc. The volume of wood consumed as raw material for pulp in the year 2000 in Japan[13] was 3.76×10^7 m^3. Thirty per cent of it was domestic wood, while 70 per cent imported wood. Other main uses of wood are lumber and plywood in Japan, and the volumes of wood used for lumber and plywood in 2000 were 2.65×10^7 m^3 and 5.4×10^6 m^3, respectively.[20]

As a result, the volume of wood consumed in 2000 in Japan is estimated to be about 7×10^7 m^3 (about 3.5×10^{10} kg).

Food is also contaminated with ^{137}Cs. The amount of food consumed in Japan[21] was 1.21×10^{11} kg in 2000. This value includes the amount of feed for domestic animals and fish. Japan Chemical Analysis Center[22] analyzed ^{137}Cs in a wide variety of food samples purchased between 1989 and 1996 in Japan. The number of samples tested was 337. The result showed that 83% of the samples had concentrations below 0.2 Bq kg^{-1}. The average ^{137}Cs concentration of twenty four mushroom samples was 4.8 Bq kg^{-1} and was far higher than those of the other kinds of samples. However, mushrooms comprised only 0.4% of total food consumed in 2000 in Japan.[21] Therefore, to average the ^{137}Cs contents of all the samples except the mushrooms is adequate for the estimation of the average ^{137}Cs content of foods consumed in Japan. The average ^{137}Cs content of foods estimated by this method is 0.15 Bq kg^{-1}.

The amount of wood consumed in 2000 in Japan is about one third of that of food. However, the estimated average ^{137}Cs content of woods (Table 1) is about six times as high as that of foods. Therefore, the ^{137}Cs radioactivity contained in wood consumed in recent Japan may be higher than that contained in food consumed in recent Japan. Paper manufacturing accounts for a half of the use of wood in Japan, and it causes transfer of high degree of ^{137}Cs radioactivity from wood to aqueous solution as already explained. Consequently, paper manufacturing may play an important role in the migration of ^{137}Cs in and near Japan.

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