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Cerium-144 and Cesium-137 Measurements in the 1963 United States Wheat Crop and Milling Products

Abstract. *The distribution of an abundant short-lived (285 days) nuclide, cerium-144, and a long-lived (30.5 years) nuclide, cesium-137, has been measured in wheat samples and milling products by means of radiochemical methods. The patent flour fraction contained approximately 10 and 15 percent of the measured cerium and cesium.*

Cerium-144 and cesium-137 distributions have not been studied as frequently in environmental samples as strontium-90 since neither of the first two radionuclides are deemed as hazardous to man. They are significant, however, because of their major contribution to world-wide fallout. Cerium-144 and its daughter praseodymium-144 are the most abundant radionuclides in the atmosphere 1 to 3 years after nuclear detonations (*1*). While Cs^{137} is initially a lesser contributor to fallout during this interval, its longer physical half-life, chemical similarity to potassium, and potential as a hazard make necessary more detailed studies. In the study reported here, we measured the amounts of Ce^{144} and Cs^{137} in the 1963 U.S. wheat crop and milling products by means of radiochemical methods.

Relatively little has been reported on the distribution of natural cerium and its family, the rare earth elements, in the environment. Late in the 19th century it was reported that "small quantities of the rare earth elements were detected in the ashes of many plants, rice, tobacco and also in human bones" (*2*).

Recently, Nezu *et al.* (*3*) found that the amount of Ce^{144} was one-tenth that of Sr^{90} in mixed animal bone, approximately equivalent to the amount in a root vegetable, and twice as much as the amount of Sr^{90} in a leafy vegetable. It is generally agreed that the rare earth radionuclides are assimilated by the plant root in negligible concentrations and that the association of these radionuclides with plants is due to physical adherence resulting from their particulate origin. However, solubility measurements of fallout debris in New York City indicated that 42 percent of the Ce^{144} was soluble at the time of deposition on the ground (*4*). In studies of the penetration of radionuclides in New Jersey soils it was found that Ce^{144} was measurable down to 30 cm (*5*). Therefore, it seems reasonable to assume that a soluble and exchangeable, as well as a particulate form of cerium, exists in radioactive fallout.

Most of the Cs^{137} is soluble in fallout debris and is strongly adsorbed within the top 5 cm of soil. The removal of Cs^{137} from soils and its availability to plants is retarded by the strong absorptive forces of soil. Two of the factors which tend to reduce these retentive forces, thus making cesium more available to plants, are increases in the concentration of hydrogen ions in the soil (*6*), and increases in the amount of available potassium (*7*).

The degree of radionuclide solubility and the distribution of radioactive elements in soil are only two of the physical parameters of the mechanism of radionuclide transfer from the atmosphere to plants. Another consideration is translocation of radionuclides by plants. Gulyakin and Yudintseva (*8*) observed approximately 2 and 15 percent Ce^{144} and Cs^{137} in wheat grain from plants grown in nutrient solution. The amount was less for soil grown plants. From autoradiographic studies of wheat plants sprayed with Cs^{137}, Middleton (*9*) found dispersion throughout the plant with approximately 10 percent concentrated in the grain.

The amounts of Ce^{144} and Cs^{137} found in samples of the 1963 U.S. wheat crop from eleven states are shown in Table 1. The values are reported as of the date considered to be about the middle of harvest. The states listed produced about 75 percent of all the wheat grown in the United States during 1962. Kansas produced 20 percent of the 1962 wheat crop. Two composite samples were made from eight varieties of Kansas wheat (Table 1). One of the composites (Kansas II) was separated into milling fractions (Table 2). As shown, the patent flour fraction, which is used for human consumption, contained approximately 10 and 15 percent, respectively, of the Ce^{144} and Cs^{137} measured in the grain. This amount is consistent with

Table 1. Results of analysis of Ce^{144} and Cs^{137} in samples of wheat grown in the United States during 1963.

Location	Harvest mid-point*	Content of original material (pc/kg)	
		Ce^{144}	Cs^{137}
Indiana	15 July	234	483
Kansas (I)	15 June	474	405
Kansas (II)†	15 June	488	410
Missouri	30 June	210	645
Montana	31 July	411	524
Nebraska	30 June	308	576
North Dakota	14 July	334	466
Ohio	15 July	177	225
Oklahoma	25 May	780	477
South Dakota	31 July	90	309
Texas	15 May	471	322
Washington	31 July	132	342

* The date considered to be about the middle of harvest, for the year 1963. † Duplicate sample of Kansas (II), which is a composite of eight varieties of wheat.

Table 2. Cerium-144 and cesium-137 in Kansas wheat and milling products.

Fraction	Percent of total	Content of original material (pc/kg)		Content of fraction (pc/kg)		Percentage in fraction	
		Ce^{144}	Cs^{137}	Ce^{144}	Cs^{137}	Ce^{144}	Cs^{137}
Wheat (Kansas II)	100	488	410	488	410		
Patent flour	52	84	124	44	64	9	16
1st Clear flour	17	162	166	28	28	6	7
2nd Clear flour	6	184	230	11	14	2	3
Shorts	14	1384	1021	194	143	39	35
Bran	11	1945	1420	214	156	44	39
Total				491	405	100	100

Table 3. Cerium-144, cesium-137, and strontium-90 in samples of patent flour and whole wheat bread, obtained from different locations in 1963.

Sample type	Sampling month	Content of original material (pc/kg)		
		Ce^{144}	Cs^{137}	Sr^{90}
	New York			
Flour	May	15	87	24
Flour	August	10	86	18
Bread	August	17	116	28
	Chicago			
Flour	April	18	35	11
Flour	July	17	45	28
Bread	July	4	122	47
	San Francisco			
Flour	June	9	40	7
Flour	Sept.	8	40	23
Bread	Sept.	19	36	28

values previously found in the patent flour fraction for each of these nuclides for a 1962 Kansas wheat sample (*10*).

Measurements of Ce^{144} and Cs^{137} in patent flour and whole wheat bread samples taken from the open market in three U.S. cities are reported in Table 3. The results for Sr^{90} are listed for comparison. All values are extrapolated to the middle of the month during which the sample was obtained. These samples, though collected in 1963, are not considered representative of the 1963 wheat crop. However, if the milling products on the open market each year are the result of the previous year's harvest, it may be assumed that these products are from the 1962 wheat crop. If this assumption is valid, samples of patent flour and milling products reaching the open market in 1964 will contain more Ce^{144} and Cs^{137} than is reported in Table 3. This prediction seems valid since there is more Ce^{144} and Cs^{137} in the 1963 wheat crop than was reported for wheat samples from the 1962 crop (*10*).

The measurements of Ce^{144} and Cs^{137} reported here were determined by means of radiochemical methods. The procedure, which was reported in detail elsewhere (*11*), was as follows. Cerium and cesium carriers were added to 10 g of dry-ashed sample and equilibrated with Ce^{144} and Cs^{137} by extraction with hydrochloric acid and hydrogen peroxide. Cesium was separated by batch adsorption on ammonium phosphomolybdate and isolated from the alkali metals by selective elution from a phenolsulfonic acid cation exchange resin. After precipitation as cesium tetraphenylborate, Cs^{137} was measured by beta counting under standard conditions with a thin-window Geiger-Muller counter.

After removal of cesium phosphomolybdate, the rare earth and alkaline earth groups were collected as oxalates from the filtrate and ignited to oxides. The two groups were separated by precipitation of the rare earths as fluorides in hydrochloric acid (8*M*). Cerium-144 was isolated from the trivalent rare earths after oxidation with sodium bromate, and was precipitated as iodate. The disintegration rate of Ce^{144} was determined from the counting rate (through 232 mg/cm² aluminum absorber) of Pr^{144} under standard conditions with a Geiger-Muller counter. The purity of each nuclide was established by beta absorption measurements. Average chemical recoveries were 82 and 90 percent, respectively, for cerium and cesium. The error due to counting for the tabulated values is within 5 percent.

Material balances of a Kansas wheat sample with its milling fractions show good agreement for both Ce^{144} and Cs^{137} (Table 2). The patent flour fraction contained approximately 10 and 15 percent of the measured Ce^{144} and Cs^{137}. Larger amounts, approximately 45 and 40 percent, respectively, of Ce^{144} and Cs^{137} were found in the bran fraction, which contains the outer layers of the wheat grain. Sampled patent flour and whole wheat bread from three cities contained more Cs^{137} than Sr^{90}, the average Cs^{137}/Sr^{90} ratio for flour and bread being 3.4 and 2.7, respectively. These ratios are not consistent with average ratios reported for rainfall or soil deposition, about 1.7, so that factors other than direct deposition are indicated.

DORIS C. SUTTON
KENNETH R. DWYER
Health and Safety Laboratory,
U.S. Atomic Energy Commission,
New York 10014

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Magnesium Compounds: New Dense Phases

Abstract. *Magnesium silicide (Mg_2Si) undergoes a transformation from a cubic (fluorite) type lattice to a hexagonal lattice at pressures above about 25 kilobars and temperatures above 900°C. There is an increase of density of about 15 percent associated with this structure change. Both the parent and product substances are semiconductors, but of different resistivities. The high pressure phase may be recovered and is indefinitely metastable at normal ambient conditions.*

That substances crystallizing in the fluorite and antifluorite (F_m3_m) lattices might exhibit polymorphic transformations has not previously attracted attention. It is possible that such substances, and particularly intermetallic compounds in this structure class, could undergo any or all of the following types of transformation, but it is not possible to predict which one: (i) a martensitic type transformation to a hexagonal phase, (ii) a polymorphic transformation based on either component, A or B, in such AB_2 compounds, and (iii) a second transformation based on the other of the two components. This set of possibilities has been investigated for the interesting semiconducting materials Mg_2Si, Mg_2Ge, and Mg_2Sn, the physical variables employed being the application of high pressures (> 20 kb) and high temperatures (up to ~ 1500°C). New phases of all three substances have been found. All are indefinitely metastable (for more than a year) at ambient conditions after release of the experimental pressures.

The compounds used were vacuum-cast in graphite molds from chemically pure starting materials by J. Kiesler, W. Moore, and W. Baxter of this laboratory. All samples showed a bright, silvery fracture surface when new, but on exposure to summer air, they more or less rapidly became covered with a blue (oxide) film, and foul-smelling