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Table 1. Results of Ce<sup>144</sup> and Sr<sup>90</sup> analysis of different environmental samples.

Sample	Date of sampling	$\mu\text{C}/\text{kg}$ (wet wt.)		$\mu\text{C}/\text{g}$ of ash	
		Ce <sup>144</sup>	Sr <sup>90</sup>	Ce <sup>144</sup>	Sr <sup>90</sup>
Soil (depth 0-10 cm)	Jan. 1960	(2670 ± 59)*	(130 ± 20)*	(2.76 ± 0.06)*	(0.13 ± 0.02)*
Spinach (leaves)	Mar. 1960	49.1 ± 3.4	21.2 ± 2.1	2.50 ± 0.17	1.1 ± 0.1
Radish (root)	Mar. 1960	9.9 ± 5.0	12.4 ± 1.0	1.5 ± 0.8	1.98 ± 0.16
Clams, muscle	June 1960	68.0 ± 3.6	2.8 ± 0.8	15.4 ± 0.8	0.65 ± 0.19
Cuttlefish (total)	Feb. 1960	77 ± 9.0	1.6 ± 0.6	4.67 ± 0.56	0.10 ± 0.038
Crucian carp bone	Apr. 1960	31 ± 10	705.8 ± 8.1	0.30 ± 0.10	6.937 ± 0.081
Mixed animal bone (cattle and horse)	Jan. 1960	932 ± 24	9293 ± 60	3.10 ± 0.80	30.98 ± 0.20

\*  $\mu\text{C}/\text{dried}$  sample.

ratory have revealed the presence of Ce<sup>144</sup> in a wide variety of substances, with prominent occurrence in animal bone and clams.

Environmental substances, including some food, obtained from Ibaraki, Japan, during January to June, 1960, have been analyzed for Sr<sup>90</sup> and Ce<sup>144</sup>. Strontium-90 concentration was determined by the "Method of Analysis for Radioactive Strontium," compiled by the Science and Technics Agency of Japan. This is a method of fuming nitric acid separation.

The method of Ce<sup>144</sup> determination is as follows: A sample solution was prepared from the ashes of about 1 kg of dried sample by hydrochloric acid extraction and alkaline fusion. After addition of a cerium carrier solution to the sample solution, rare earths were isolated from the sample solution as hydroxides. Oxalates were separated from the hydroxides and converted into oxides by ignition in an electric furnace at 600° to 700°C. The oxides thus obtained were dissolved in hydrochloric acid, and hydroxides were again precipitated from this solution. This procedure was repeated two or three times for removal of calcium, phosphate, and other cations except for rare earths. After being dissolved in concentrated hydrochloric acid, the hydroxides were passed through a column containing anion exchange resin equilibrated with concentrated hydrochloric acid. By this procedure iron, uranium, and plutonium are absorbed. The effluent was dissolved in nitric acid solution (7.5M), and again passed through a column containing anion exchange resin equilibrated with nitric acid (7.5M). By this step thorium is absorbed. From the effluent, rare earths were precipitated as hydroxides. Cerium was isolated from other rare earths by the iodate method and finally prepared as oxalate. The radioactivity of each sample was measured under standard conditions

with a Geiger-Müller counter. The concentration of Ce<sup>144</sup> was determined by the chemical yield of added carrier and by comparison with a standard sample of known concentration.

The cerium fractions were combined, and the gamma spectrum was determined by using a 1¾ by 2 inch NaI (Tl) well-type crystal and a 256-channel pulse height analyzer. The distinct peaks of the spectrum were consistent with that of the Ce<sup>144</sup> + Pr<sup>144</sup> standard sample.

The Ce<sup>144</sup> + Pr<sup>144</sup> spectrum would be expected to show peaks at 0.134, 0.100, and 0.071 Mev due to Ce<sup>144</sup>, and at 2.18, 1.48, and 0.700 Mev due to Pr<sup>144</sup>. The peaks due to Ce<sup>144</sup> are more than 60 percent of the total gamma radiation of Ce<sup>144</sup> + Pr<sup>144</sup>, but the peaks due to Pr<sup>144</sup> are only a few percent (3) of the total.

Observed peaks on this spectrum were 0.134, 0.100, and 0.069, Mev. With our instrument, the ordinates lower than 0.100 Mev are not in correct proportion to the energy. The gamma radioactivity of the cerium fraction was so low that the peaks due to Pr<sup>144</sup> could not be observed. From our data, the individual peaks due to Pr<sup>144</sup> cannot be distinguished from the Compton background. The maximum energy of beta rays, calculated from the beta-ray absorption curve of the cerium by the method of Bleuler and Zunti (4), was 2.92 Mev. The decay curve of the cerium fraction showed that the half-life was 282 days.

The radioactivity of the cerium fraction seems, therefore, to owe its origin to Ce<sup>144</sup> + Pr<sup>144</sup>. The Ce<sup>144</sup> concentrations of representative samples are summarized in Table 1. This radionuclide was present in every sample tested. The higher levels of Ce<sup>144</sup> in clams, as compared with levels in the other biological samples, are not unexpected, in view of the findings of Goldberg *et al.* (5), who demonstrated that some marine organ-

## Cerium-144 in Food

**Abstract.** Small amounts of cerium-144 have been found in samples of food and animal bone obtained from Ibaraki, Japan. The highest level of radioactivity was detected in clams (*Schizimi, Corbicura* sp.) harvested from Hinuma Marsh. In view of the widespread occurrence of cerium-144 in tested samples, it is assumed that the presence of this radionuclide is due to fallout.

The predominant fission product contributor to total radioactivity 0.85 to 3 years after detonation is Ce<sup>144</sup> (Pr<sup>144</sup>) (1). Because Ce<sup>144</sup> is abundant in fallout, it is reasonable to expect that food will be contaminated by this radionuclide.

Recently Van Dilla (2) reported that although grazing animals ingest large amounts of Ce<sup>144</sup> and other radionuclides as foliar contamination, very little Ce<sup>144</sup> is absorbed. Investigations at our labo-

isms such as mollusks concentrate metallic ions at levels many times higher than the levels found in the surrounding water. The concentration of  $Ce^{144}$  in clams during April 1959 was about  $7 \times 10^2 \mu\mu c$  per kilogram of clam muscle (wet weight). It had decreased to  $7 \times 10 \mu\mu c$  by July 1959.

The gross radioactivity of surrounding water during the time from August 1958 to February 1959 was about  $3 \pm 3 \mu\mu c$  per liter, but it increased to  $21 \pm 3.2 \mu\mu c$  per liter in March 1959. The radioactivity of the water has not been measured since April 1959, but radioactivity in rain and fallout in this region increased from March 1959 and reached a maximum in April 1959. Since August 1959, there has been less radioactivity in rain and fallout.

It has been reported that the concentration of  $Ce^{144}$  in the sediment of the sea in the year 1960 was  $1 \mu\mu c$  per gram of dried sample. This amount corresponded to that found in 5 liters of surrounding sea water. The concentration of  $Ce^{144}$  in sediment was about 100 times higher than that of  $Sr^{90}$  (6).

A clam is a marine organism that makes its home in brackish water, and thus can be found in marsh sediment. Since radioactive contamination in a marsh bed has its origin in fallout, it may safely be assumed that  $Ce^{144}$  was deposited in the marsh bed by fallout and then absorbed by the clams.

The concentration of  $Sr^{90}$  was generally at a higher level in bone than in other biological samples, whereas  $Ce^{144}$  was more concentrated in clams. Cattle and horse bone contained large quanti-

ties of both radionuclides. The presence of  $Ce^{144}$  in food was at a quite trivial level compared to the maximum permissible concentration of  $Ce^{144}$  ( $1 \times 10^5 \mu\mu c$  per liter of water).

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