To be submitted to Science

RADIOACTIVE CERIUM ISOTOPES: THE FALLOUT FROM RECENT FRENCH AND CHINESE NUCLEAR WEAPONS TESTS

Abstract: A new method involving gamma-ray spectroscopy was employed to determine the Ce^{141}/Ce^{144} ratios in individual samples of rain and snow collected at Fayetteville, Arkansas (36°5'N, 94°9'W). By the use of a Ge(Li) detector, the counting period for the cerium samples can now be shortened from over 8 months to only one day. The presence was noted of fresh nuclear debris in the Northern tropospheric rain from the French nuclear weapons tests conducted in the Southern Hemisphere during the summer months of 1970, followed by the appearance of the fresh Chinese debris after October 1970.

The importance of bomb-produced radioactive cerium isotopes, Ce^{141} and Ce^{144}, as atmospheric radioactive tracers has long been well recognized (1). The half-lives of Ce^{141} (32.5 days) and Ce^{144} (285 days), are quite similar to the tropospheric and stratospheric mean residence times of particulate matter of about 30 days and one year, respectively. The fact that the cerium samples had to be counted over a period of 8 to 11 months to analyze the composite beta-decay curve was, however, a major handicap which tended to limit the wide use of this pair of isotopes as atmospheric radioactive tracers. During the early stage of this work, the Biller analysis method (2) was used, but this method still required a counting period of 4 to 5 months. We then counted the gamma-rays emitted by Ce^{141} (145 keV) and Ce^{144} (134 keV) in the remaining cerium samples using a Ge(Li) detector. These gamma-ray peaks were easily separated with high-gain amplification, and this method required the cerium samples be counted only once for a period of 10 to 20 hours. Fig. 1 shows a typical gamma spectrum.

The rain and snow samples were collected on the roof of the Chemistry Building of the University of Arkansas (36°5'N, 94°9'W) in five galvanized iron collectors, each having a surface area of 9×10^4 cm^2 and emptying into 30-gallon polyethylene containers. Each sample analyzed contained 10-20 liters of rain or melted snow. A radiochemical procedure, which was developed in this laboratory, was used for the separation of radioactive cerium from rain (1).

All cerium samples which were analyzed by the Biller method (2) were counted by a Tracerlab low-level beta-counting system (CE 14). A Nuclear Diode (LGCC 5.0-3.2) Germanium-Lithium detector with a relative peak efficiency of 6.65% at 1.33 MeV was used to count the remaining samples. The counting efficiencies for the 145 and 134 keV gamma-rays were calculated to be 17.2% and 19.6%, respectively.

The experimental results obtained are shown in Table 1. The Ce^{141}/Ce^{144} ratios in rain are plotted in Fig. 1. The Ce^{144} concentrations in rain were generally low during the fall months, but a marked increase was noted after the second half of January 1971, reaching a maximum in early May 1971. The marked increase in the Ce^{144} concentration in rain during the spring months is most likely due to a combination of the well-known spring maximum in fallout (3) and the atmospheric injections by the French and Chinese nuclear weapons tests of the previous year. The shorter lived

Ce^{141} was present in all of the rains since the 14 October Chinese test, prior to which it was absent in rain for about 5 weeks. We interpret the peaks of the Ce^{141}/Ce^{144} ratios observed in the August and September 1970 rains as an indication of the transport of fresh nuclear debris from the summer 1970 French test series conducted in the Southern Hemisphere. The pattern of variation of the Ce^{141}/Ce^{144} ratio in rain is in line with those of the Sr^{89}/Sr^{90} ratios observed at Fayetteville and also at Key West, Florida (24°33'N, 81°45'W). A similar variation of the Ce^{141}/Ce^{144} and Sr^{89}/Sr^{90} ratios in rain was observed in this laboratory following the summer 1968 French test series (1). A number of investigators have recently discussed the problems concerning the inter-hemispheric transport of nuclear debris across the equator (4).

A marked zig-zag pattern of variation of the Ce^{141}/Ce^{144} ratio in rain observed shortly after the 14 October 1970 is most likely due to the fallout particles traveling eastward and circling the globe a number of times (1). Unfortunately, no rainfall occurred between 19 November and 10 December 1970 at Fayetteville, Arkansas, and hence the exact dates of occurrence of the Ce^{141}/Ce^{144} peak values could not have been established.

The venting of debris from a Nevada underground test has been reported by the news media in the second half of December 1970. The Ce^{141}/Ce^{144} ratio data shows no clear indication of a sharp increase at this time. Our tentative explanation for this is that the venting was probably limited to the so-called 'volatile' fission product chains with fairly long lived gaseous precursors. The fission product chains at mass numbers 141 and 144 do not have such precursors.

The Ce^{141}/Ce^{144} ratio in the atmosphere seems to have reached a saturation value shown by the line ab in Fig. 2 by the end of February 1971. We interpret this to mean that a bulk of the fresh nuclear debris injected into the stratosphere during the previous fall started to reach the lower level of the atmosphere during the early spring fallout peak period. The line ab in Fig. 2 thus should give us a rough idea about the Ce^{141}/Ce^{144} ratio in the Northern Stratosphere as a whole. The line ab intercepts the date of 14 October 1970 Chinese test at Ce^{141}/Ce^{144} ratio = 7. If one assumes that the production ratio of the cerium isotopes in fresh debris is about 12, it would seem that the Ce^{144} inventory in the Northern Stratosphere was increased by a factor of about 2 (5).

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(5) We thank the U. S. Atomic Energy Commission for the support of this research under contract No. At-(40-1)-2529; we also wish to thank D. L. Swindle, J. R. Noyce, and D. T. Moore for their invaluable assistance and advice.

Table 1. ^{141}Ce, ^{144}Ce, and ^{141}Ce/^{144}Ce Ratios in Rain.

Date	Rainfall (mm)	^{141}Ce (pc/l)	^{144}Ce (pc/l)	^{141}Ce/^{144}Ce (pc/pc)
1970				
Aug. 9	6.1	0.46 ± 0.07	2.36 ± 0.21	0.20 ± 0.01
Aug. 10	0.5	0.22 ± 0.04	4.09 ± 0.13	0.05 ± 0.01
Aug. 18	5.2	----	10.87 ± 0.83	----
Aug. 20	14.0	0.72 ± 0.06	11.12 ± 0.25	0.06 ± 0.01
Aug. 21	27.2	1.79 ± 0.05	4.42 ± 0.09	0.28 ± 0.08
Aug. 31	27.5	2.34 ± 0.18	4.63 ± 0.82	0.50 ± 0.09
Sep. 2	25.4	----	0.92 ± 0.02	----
Sep. 3	55.9	2.29 ± 0.07	5.17 ± 0.15	0.45 ± 0.03
Sep. 8	14.0	1.35 ± 0.05	4.94 ± 0.12	0.27 ± 0.01
Sep. 9	2.8	0.95 ± 0.10	10.62 ± 1.06	0.09 ± 0.01
Sep. 13	33.5	3.15 ± 0.31	2.42 ± 0.24	1.30 ± 0.18
Sep. 17	3.3	1.81 ± 0.08	3.49 ± 0.16	0.52 ± 0.08
Sep. 18	4.3	----	7.10 ± 0.19	----
Sep. 22	95.6	----	3.80 ± 0.17	----
Sep. 25	22.9	----	2.62 ± 0.14	----
Oct. 5	13.8	----	1.38 ± 0.09	----
Oct. 7	42.0	----	2.97 ± 0.38	----
Oct. 11	19.3	----	4.16 ± 0.26	----
Oct. 18	2.1	----	1.42 ± 0.12	----
Oct. 24	52.4	9.91 ± 0.39	5.92 ± 0.29	1.67 ± 0.14
Oct. 27	79.1	7.44 ± 0.90	5.71 ± 0.27	1.30 ± 0.29
Nov. 1	27.7	5.61 ± 0.42	8.33 ± 0.37	0.67 ± 0.07
Nov. 3	4.4	1.92 ± 0.16	2.67 ± 0.19	0.72 ± 0.07
Nov. 8	21.2	5.33 ± 0.34	4.65 ± 0.22	1.15 ± 0.09
Nov. 14	13.0	3.75 ± 0.16	4.98 ± 0.21	0.75 ± 0.04
Nov. 19	2.2	6.15 ± 0.44	30.58 ± 0.60	0.20 ± 0.01
Dec. 10	12.4	2.72 ± 0.20	7.82 ± 0.23	0.35 ± 0.02
Dec. 16	21.8	1.11 ± 0.11	5.06 ± 0.19	0.22 ± 0.02
Dec. 20	5.1	1.16 ± 0.09	4.81 ± 0.16	0.24 ± 0.01
1971				
Jan. 2	30.8	2.66 ± 0.19	8.21 ± 0.38	0.32 ± 0.02
Jan. 13	31.1	1.43 ± 0.12	4.81 ± 0.21	0.30 ± 0.02
Jan. 23	1.9	6.57 ± 0.36	16.60 ± 0.49	0.40 ± 0.03
Jan. 31	0.1	11.79 ± 0.93	23.62 ± 0.84	0.50 ± 0.04

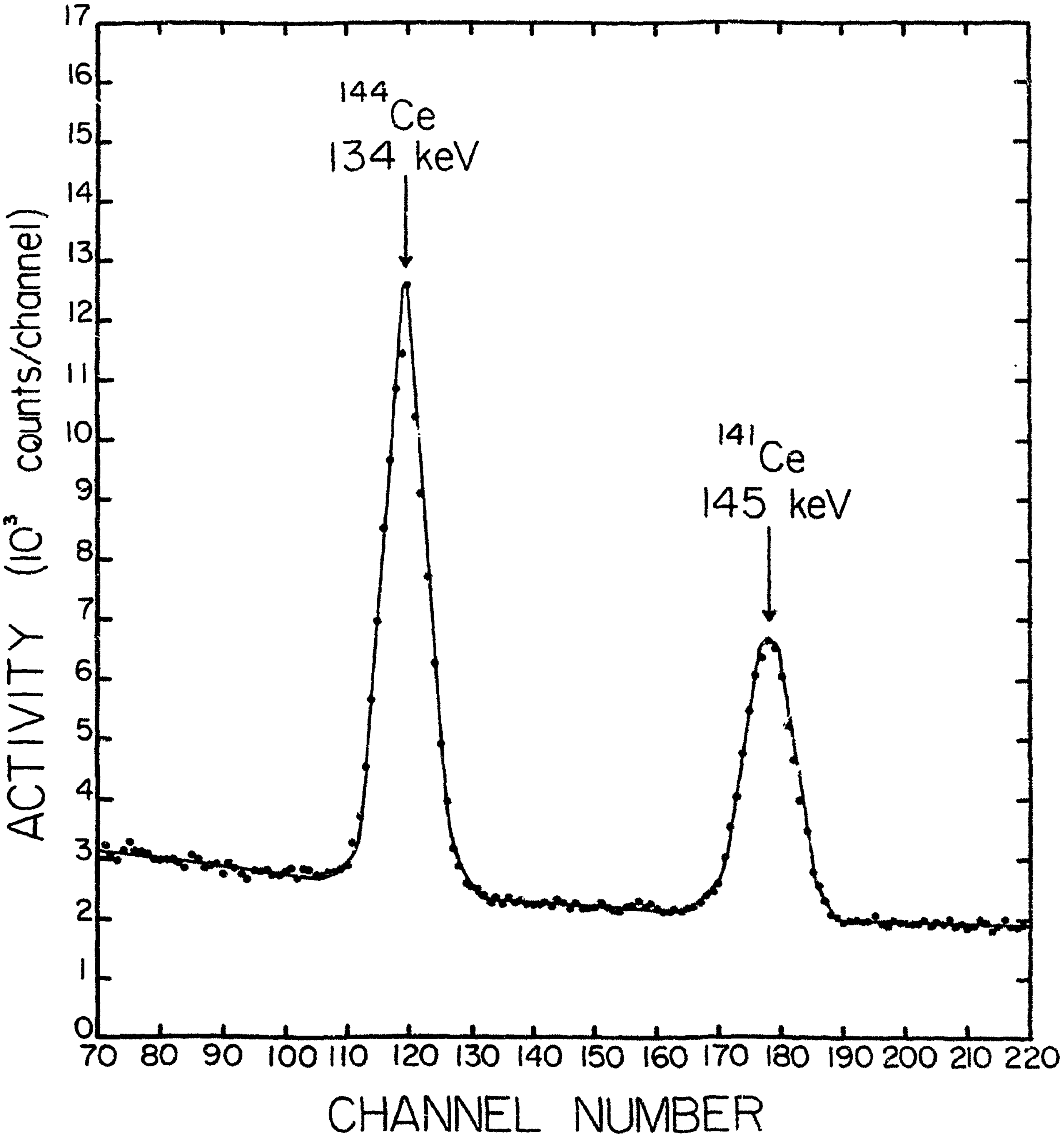
Table 1 (Continued)

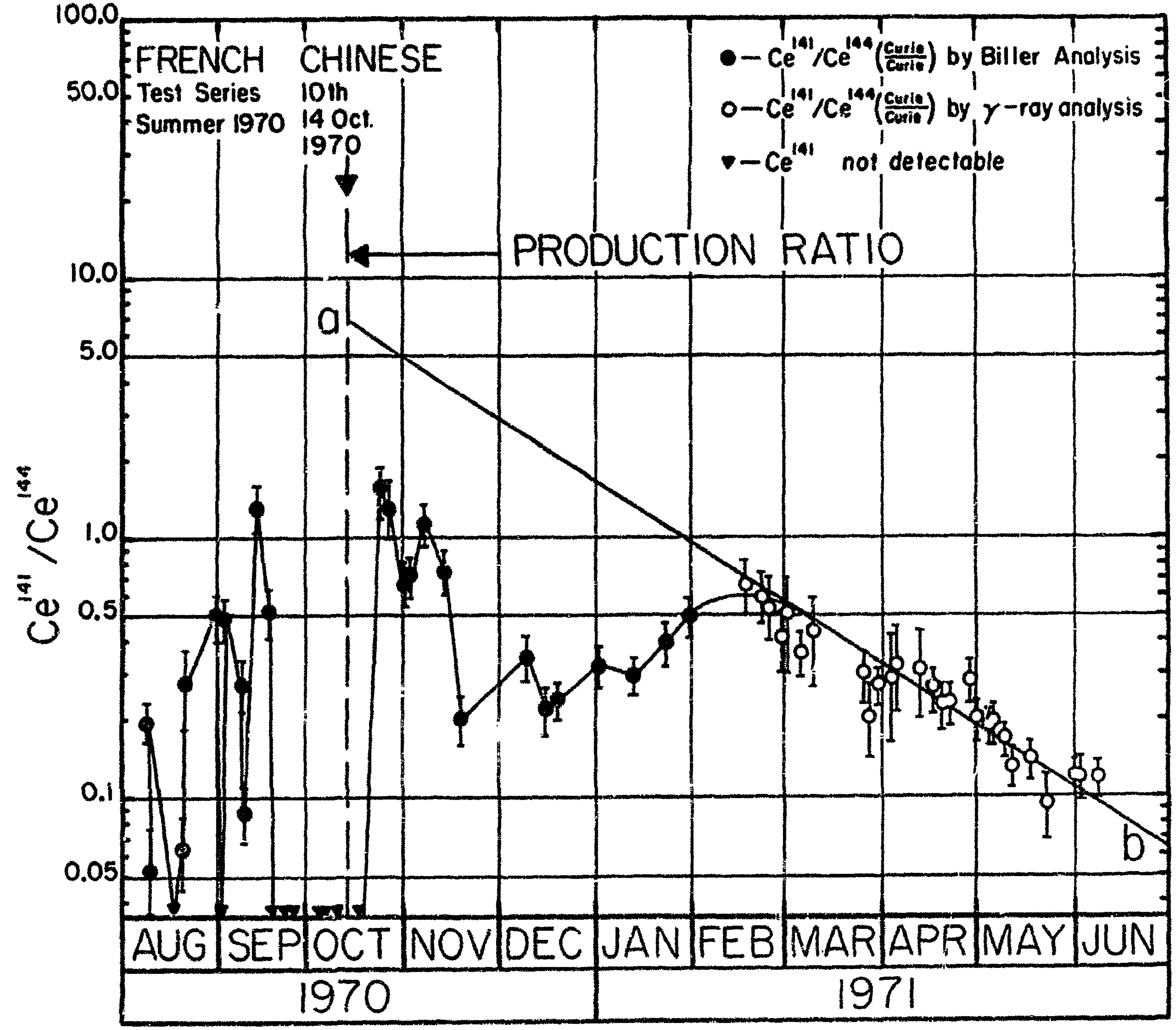
Date	Rainfall (mm)	^{141}Ce (pc/l)	^{144}Ce (pc/l)	$^{141}Ce/^{144}Ce$ (pc/pc)
Feb. 17	8.4	8.15 ± 1.53	12.33 ± 1.52	0.66 ± 0.15
Feb. 22	20.3	4.83 ± 0.77	8.21 ± 1.05	0.59 ± 0.12
Feb. 24	6.3	9.92 ± 2.32	18.22 ± 1.62	0.54 ± 0.14
Feb. 28	0.4	10.19 ± 2.62	25.19 ± 2.37	0.41 ± 0.11
Mar. 2	5.0	6.30 ± 2.32	12.44 ± 1.79	0.51 ± 0.20
Mar. 6	10.5	11.58 ± 2.19	32.24 ± 3.44	0.36 ± 0.07
Mar. 10	3.8	4.45 ± 0.62	10.21 ± 3.09	0.44 ± 0.14
Mar. 26	0.4	15.34 ± 2.78	50.48 ± 3.16	0.30 ± 0.05
Mar. 28	1.0	4.05 ± 1.05	19.71 ± 2.87	0.21 ± 0.06
Mar. 31	0.7	9.51 ± 0.98	34.88 ± 1.67	0.27 ± 0.30
Apr. 4	17.8	4.22 ± 1.83	14.57 ± 3.21	0.29 ± 0.14
Apr. 6	1.2	3.45 ± 1.10	10.61 ± 1.18	0.33 ± 0.11
Apr. 13	1.9	6.20 ± 1.87	19.63 ± 4.04	0.32 ± 0.11
Apr. 17	1.2	16.85 ± 2.21	62.30 ± 1.58	0.27 ± 0.03
Apr. 20	21.9	3.94 ± 0.66	17.09 ± 1.80	0.23 ± 0.04
Apr. 23	27.6	6.22 ± 0.59	26.13 ± 2.67	0.24 ± 0.03
Apr. 29	23.0	10.42 ± 1.42	36.16 ± 3.19	0.27 ± 0.04
May 1	4.1	17.67 ± 1.00	84.82 ± 2.34	0.21 ± 0.01
May 5	0.9	25.88 ± 1.41	136.16 ± 3.38	0.19 ± 0.01
May 6	31.5	7.35 ± 0.55	37.37 ± 1.37	0.20 ± 0.01
May 10	27.2	6.26 ± 0.52	36.92 ± 1.37	0.17 ± 0.01
May 12	0.9	3.39 ± 0.42	25.89 ± 1.37	0.13 ± 0.01
May 18	11.2	4.09 ± 0.39	28.68 ± 2.08	0.14 ± 0.01
May 23	62.0	2.64 ± 0.69	27.51 ± 2.00	0.10 ± 0.02
June 1	6.4	7.07 0.34	57.98 1.60	0.12 ± 0.00
June 2	5.0	5.17 0.79	41.60 2.57	0.12 ± 0.02
June 9	3.1	3.88 0.33	31.24 1.60	0.12 ± 0.01

FIGURE CAPTIONS

Fig. 1. A typical spectrum of the gamma-rays emitted from ^{144}Ce and ^{141}Ce in a rain sample.

Fig. 2. The variation of ^{141}Ce/^{144}Ce ratios in rain at Fayetteville, Arkansas, from August 9, 1970, to June 9, 1971. The slope of the line ab corresponds to the difference of the decay constants of ^{141}Ce (0.0213 d^{-1}) and ^{144}Ce (0.00244 d^{-1}). This line shows the radioactive decay of the ^{141}Ce/^{144}Ce ratio in the stratosphere.


^{144}Ce
134 keV
^{141}Ce
145 keV
ACTIVITY (10^3 counts/channel)
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
CHANNEL NUMBER
70
80
90
100
110
120
130
140
150
160
170
180
190
200
210
220


100.0
50.0
10.0
5.0
1.0
0.5
0.1
0.05
Ce^{141}/Ce^{144}
FRENCH
Test Series
Summer 1970
CHINESE
10th
14 Oct.
1970
● – Ce^{141}/Ce^{144} ($\frac{Curie}{Curie}$) by Biller Analysis
○ – Ce^{141}/Ce^{144} ($\frac{Curie}{Curie}$) by γ-ray analysis
▼ – Ce^{141} not detectable
PRODUCTION RATIO
a
b
AUG
SEP
OCT
NOV
DEC
JAN
FEB
MAR
APR
MAY
JUN
1970
1971