not rule out the possibility that some of the Al^{26} we find reached the ice in cosmic dust; nonetheless, we find a much lower precipitation rate of Al^{26} than do Wasson *et al.* and Lal *et al.*

In the work of Lal *et al.* (*3*) and Wasson *et al.* (*4*), positron activity, interpreted as that of Al^{26}, is measured in aluminum separated from pelagic red clays. This activity and the measured or estimated accumulation rate of the sediment yield an Al^{26} precipitation rate five (*4*) or ten (*3*) times that expected from atmospheric production; the "excess" is assumed to be Al^{26} from cosmic dust. We believe that the difference between these results and ours can be explained entirely by the uncertainties in their measurements. The measurements by Lal *et al.* are borderline; the positron peak they found was little above noise level and the same peak appeared in their background. Wasson *et al.* did not recycle their aluminum to see whether the positron activity followed aluminum chemistry, and the sedimentation rate they used in their calculations was an estimate. In our measurements we cannot think of any likely mechanism by which Al^{26} could have been lost, and further measurements are more likely to cause us to lower our estimate of the rate of Al^{26} precipitation than to raise it.

We can set an upper limit to the influx rate of cosmic dust bearing Al^{26} by combining our upper limit for Al^{26} precipitation with the most likely value for its fallout rate from atmospheric production. This gives $(1.6 - 0.8)10^{-8} = (0.8)10^{-8}$ dpm/cm^2 · year for Al^{26} in cosmic dust. From this figure, the assumptions of Lal *et al.* give an upper limit for the influx rate of zodiacal dust over Earth of 3.2×10^5 ton/year, and the assumptions of Wasson *et al.* give an upper limit of 1.0×10^5 ton/year. However, we emphasize that these numbers have little intrinsic value: not only are they merely upper limits; a great many very weak assumptions have been needed to turn an observed Al^{26} precipitation rate into a cosmic-dust accretion rate.

Our Be^{10} measurements are in good agreement both with the calculated atmospheric production rates and with those found from measurements of Be^{10} in sea sediments. However, to derive an average production rate of Be^{10} from our measurements, we would need to know the effect of latitude and of the nature of the precipitation (rain or snow) on the precipitation rate of isotopes from the atmosphere. This point is in some doubt, and we therefore prefer to let our results stand without further interpretation.

R. McCorkell
E. L. Fireman
Smithsonian Astrophysical Observatory, Cambridge, Massachusetts 02138
C. C. Langway, Jr.
U.S. Army Cold Regions Research and Engineering Laboratory, Hanover, New Hampshire 03755

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Fallout from the Chinese Nuclear Explosion of 17 June 1967

Abstract. *Low-level activities of iodine-131, barium-140, and strontium-89 were found in a series of rain samples collected at Fayetteville, Arkansas, during the period from late June through August 1967. The ratios of these short-lived isotopes to strontium-90 were determined as accurately as possible. The data indicate that the debris from the Chinese nuclear explosion was injected primarily into the stratosphere.*

It was announced that a thermonuclear weapon was tested in China on 17 June 1967. Heavy fallout was expected soon after, but the preliminary indications were that the fallout was almost negligible. This indication suggested the possibility that the debris was injected primarily into the stratosphere, and that it would be some time before the fallout reached ground level. We studied the fallout of I^{131}, Ba^{140}, and Sr^{89} from this explosion at Fayetteville (94°W, 36°N), Arkansas, using large volumes of rain samples and attempting to measure the ratios of a number of fission products to Sr^{90} as accurately as possible. We now report the data obtained so far.

The rain samples were collected on the roof of the Chemistry Building of the University of Arkansas. Strontium and Ba carriers were added to 8 to 16 liters of rain samples, and the Sr and Ba fractions were radiochemically purified by a reported method (*1*, *2*). Carrier of I^- was added to 12 liters of rain samples, and the iodine fraction was radiochemically purified by a reported method (*3*). The procedure used is briefly described: the I^- was oxidized to IO_4^- with NaClO in alkaline solution; IO_4^- was then reduced to I_2 with hydroxylamine hydrochloride, and the iodine was extracted with CCl_4; the iodine was removed from the CCl_4 by shaking with $NaHSO_3$, and then was purified by another CCl_4 extraction cycle in which $NaNO_2$ was used for oxidation of I^- to I_2; this extraction cycle was repeated three times; the iodine was finally precipitated as AgI and counted. Tracerlab Omni Guard low-level background system was used for the radioactive measurements, with a background of about 0.5 count/min.

The results appear in Table 1. Presence of I^{131} and Ba^{140} was first detected in the rain sample collected on 27

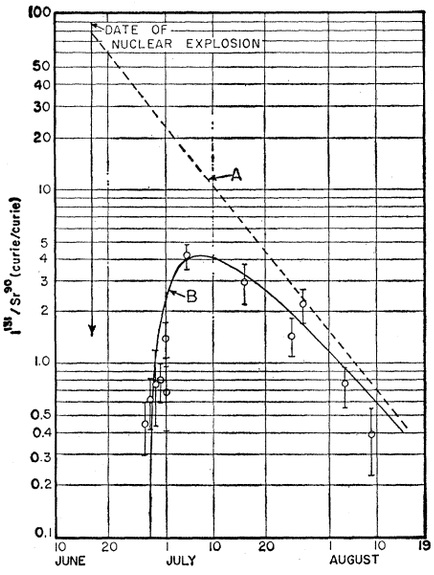

Fig. 1. Variation of the I^{131} : Sr^{90} ratio in rain at Fayetteville, Arkansas, after the Chinese nuclear explosion of 17 June 1967.

June 1967, approximately 10 days after the explosion. After previous Chinese nuclear tests, the first "wave" of fresh debris usually arrived within about 10 days (*4*, *5*); it arrived in Tokyo, Japan, about 5 days after the nuclear explosion of 14 May 1965 (*6*). The presence of Sr^{89} in rain samples collected on 24 and 25 June 1967 can be attributed to old debris from the Chinese nuclear explosion of 28 December 1966.

Variation of the I^{131} : Sr^{90} ratio in rain is shown in Fig. 1; a sharp increase was observed at the beginning of July; after reaching a maximum value around 5 July, it appeared to approach asymptotically the straight line *A*, with a slope corresponding to the 8.05-day half-life of I^{131}. Curve *B* (Fig. 1) is given by an empirical equation:

$$(B/A)_{T,t} = 30 \cdot e^{-0.0862t}(1 - e^{-0.04t}) \quad (1)$$

where $(B/A)_{T,t}$ is the I^{131} : Sr^{90} ratio in rain falling t days after 28 June 1967 ($t = 0$).

The straight line *A* yields a value for I^{131} : Sr^{90} of about 80 on 17 June 1967, while the production ratio (I^{131} : Sr^{90}) in fresh debris should be roughly 10^3 (the half-life ratio). This finding indicates that the increase in Sr^{90} inventory in the stratosphere, by this explosion, was only a few percent. If this interpretation is correct, the straight line *A* (Fig. 1) shows the I^{131} : Sr^{90} ratio in the northern stratosphere. Needless to say, I^{131} and Sr^{90} are not expected to be evenly mixed. The scatter of the experimental points (Fig. 1) may indicate the magnitude of nonuniform distribution of the fission products in the stratosphere. Nevertheless the fact that the I^{131} : Sr^{90} ratio data can be expressed by a simple empirical equation, such as Eq. 1, seems to support the view that the atmosphere can be treated as a two-compartment system consisting of stratosphere and troposphere.

The empirical Eq. 1 can be derived, from the general equation given by Kuroda (*7*) in 1958, as follows:

$$(B/A)_{T,t} = \frac{\frac{k_T - k_s}{k_s} \cdot \frac{A_{T,0}}{A_{S,0}}}{(e^{(k_T - k_S)t} - 1) + \frac{k_T - k_S}{k_S} \cdot \frac{A_{T,0}}{A_{S,0}}} \times [(B/A)_{T'0} - (B/A)_{S'0}] \times e^{-(\lambda B - \lambda A)t} + (B/A)_{S,0} \times e^{-(\lambda B - \lambda A)t} \quad (2)$$

where $(B/A)_{T,t}$ is the ratio of nuclides *A* and *B* in the troposphere at time *t*; A_{T0} and A_{S0} are the amounts of *A* in the troposphere and in the stratosphere at $t = 0$; k_T and k_S are the reciprocals of the mean storage times of the fission products in the troposphere and in the stratosphere, respectively; $(B/A)_{T,0}$ is the *B*:*A* ratio in the troposphere at $t = 0$; and $(B/A)_{S,0}$ is the *B* : *A* ratio in the stratosphere at $t = 0$.

If the Sr^{90} inventory in the stratosphere changed little, as we suggested earlier, a steady-state condition should have existed between the inventories of Sr^{90} in the stratosphere and in the troposphere. If so,

$$[(k_T - k_S)/k_S] \cdot (A_{T,0}/A_{S,0}) \cong 1$$

and if the nuclide *B* (I^{131} in this instance) was injected mainly into the stratosphere,

$$(B/A)_{T,0} \cong 0$$

Moreover, $k_T >> k_S$, and $\lambda_{131} >> \lambda_{90}$. Thus the general Eq. 2 is converted to the form

$$(B/A)_{T,t} \cong (B/A)_{S,0} \cdot e^{-\lambda 131 t} \times (1 - e^{-k_T \cdot t}) \quad (3)$$

As shown in Fig. 1, the best fit to the data seems to be obtained if k_T is taken to be 0.04, $(B/A)_{S,0}$ is taken to be 30, and $t = 0$ is taken to be 28 June 1967 (these values correspond to I^{131} : Sr^{90} ≅ 80 on 17 June 1967). The fact that a shift of about 10 days has to be applied to $t = 0$ is reasonable in view of the fact that it takes at least 10 days for the first wave of fresh debris to reach Fayetteville from Lop Nor.

The value $k_T = 0.04$ corresponds to a mean tropospheric-residence time of $1/k_T = 25$ days; this value is in reasonable agreement with the earlier estimates by Beck *et al.* (*4*) ($1/k_T = 30$ days), for example.

Although the data are not plotted here, the Ba^{140} : Sr^{90} ratio and the Sr^{89} : Sr^{90} ratio also show a similar trend, except that the points tend to scatter somewhat more irregularly than do the data for the I^{131} : Sr ratio.

Table 1. Iodine-131, barium-140, strontium-89, and strontium-90 in rain at Fayetteville, Ark.

Date (1967)	Rainfall (inches)	Content ($\times 10^{-12}$ c/liter)			
		I^{131}	Ba^{140}	Sr^{89}	Sr^{90}
24 June	0.92	< 0.1	< 0.05	0.15 ± 0.03	0.16 ± 0.03
25 June	.17	< .1	< .05	.40 ± .04	.40 ± .04
27 June	.26	.5 ± 0.2	.65 ± 0.07	.7 ± .1	1.1 ± .1
28 June	1.40	.4 ± .1	.23 ± .03	.42 ± .04	0.65 ± .06
29 June	1.04	.4 ± .2	.57 ± .06	.35 ± .04	.53 ± .05
30 June	0.06	.9 ± .2	1.64 ± .17	.97 ± .10	1.1 ± .1
1 July	.33	1.0 ± .5	3.41 ± .34	.7 ± .1	1.5 ± .1
1 July	.57	0.4 ± .1	0.70 ± .07	.28 ± .06	.29 ± .06
5 July	.09	.8 ± .1	1.87 ± .19	.45 ± .11	.19 ± .02
12 July	.77	*	1.46 ± .15	.43 ± .04	.33 ± .03
16 July	.18	.4 ± .1	1.14 ± .12	.28 ± .06	.14 ± .02
25 July	.08	1.6 ± .2	2.37 ± .24	1.7 ± .2	1.1 ± .1
27 July	.12	0.6 ± .1	1.61 ± .16	0.5 ± .1	0.28 ± .03
2–4 Aug.	.56	.4 ± .1	2.53 ± .26	1.0 ± .1	.53 ± .05
9 Aug.	1.35	.8 ± .3	2.37 ± .24	2.3 ± .2	2.1 ± .2

* Sample lost.

M. A. Reynolds, D. L. Swindle
R. J. Wright, P. K. Kuroda
Department of Chemistry,
University of Arkansas, Fayetteville

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