

plied age of the event is 1.73 million years in profile EL19N (see Fig. 4).

There is a younger normal event, the Gilsa event, dated at about 1.65 million years (7). No particular magnetic anomaly has been associated with the Gilsa event, because no obvious anomaly consistently appears between the ones usually associated with the Olduvai event (1.96 million years) and the Jaramillo event (0.92 million years). The above implies that the Gilsa anomaly, undefined until now, may be the anomaly usually associated with the Olduvai event. Similar ideas have been expressed by Cox (8), whereas, on the basis of age data from deep-sea sediment cores, Opdyke and Foster (9) prefer the usual Olduvai event-anomaly association.

There is extensive evidence for the existence of the Olduvai normal event at 1.96 million years, and it is reasonable to expect an anomaly associated with the event. Heirtzler *et al.* (1) discuss a minor anomaly, called X, which occurs with some consistency between the anomalies associated with the Olduvai event and the beginning of the Matuyama epoch. This anomaly, X, and a neighboring anomaly, W, which also consistently appears, are labeled in Fig. 2. Anomalies X and W sometimes occur as a doublet, and sometimes only one or the other appears. In the latter case it cannot be determined by simple examination of the profiles which anomaly is present. In order to overcome this difficulty the age of each pertinent anomaly was determined for all the profiles from the assumed constant spreading rates from 3.32 to 0.9 million years. Using only the profiles containing both anomalies, the average age of W turns out to be 2 million years (standard deviation, 0.06 million years) and the average age of X is 2.3 million years (standard deviation, 0.1 million years). The ages for X and W in EL19N are shown in Fig. 4. When a single anomaly occurred, it was subsequently labeled W or X depending on its relation to the previously determined average ages for W and X. All singly appearing anomalies were found to correspond to W; no ambiguities were encountered. The resulting average age of W (1.99 million years) corresponds very closely to the measured age of the Olduvai event (1.96 million years), and we postulate that under the assumption of constant spreading from 3.32 to 0.9 million years, W is the true Olduvai anomaly.

If the variable-rate spreading interpretation of Fig. 3 is accepted, then there is no anomaly that can be easily related to the Gilsa event. If a constant spreading rate from 3.32 to 0.9 million years is accepted, then both the Gilsa and the Olduvai events can be associated with consistently appearing anomalies. We regard the latter as more acceptable than the former because of the way it allows all the well-known paleomagnetic events to be associated with consistently appearing magnetic anomalies and does not require complications of the basic spreading hypothesis.

DAVID A. EMILIA

Marathon Oil Company,
Denver Research Center,
Littleton, Colorado 80120

DONALD F. HEINRICH

Department of Oceanography,
Oregon State University, Corvallis

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Radionuclide Composition of the Allende Meteorite from Nondestructive Gamma-Ray Spectrometric Analysis

Abstract. *The concentrations of beryllium-7, sodium-22, aluminum-26, potassium-40, scandium-46, vanadium-48, chromium-51, manganese-54, cobalt-57, cobalt-60, and thorium-232 (thallium-208) have been measured in the Allende meteorite by nondestructive gamma-ray spectrometry. The high cobalt-60 content of the meteorite is indicative of a preatmospheric body with a minimum effective radius of 50 centimeters and a weight of 1650 kilograms; the aluminum-26 activity indicates a minimum exposure age of 3 million years.*

Carbonaceous chondrites represent a small fraction of all observed meteorite falls (1); however, these chondrites are of particular interest because it is suspected that they represent relatively undifferentiated primitive matter. The last recorded fall of a Type-III carbonaceous chondrite was that of the Kainsaz meteorite in the U.S.S.R. in 1937. When sample acquisition of any meteorite is possible very shortly after its fall, a rare opportunity is provided for measurement of the short-lived as well as the long-lived cosmogenic radionuclides. Measurements of the absolute and relative concentrations of radionuclides in meteorites provide information which is basic to the understanding of their extraterrestrial history, their preatmospheric size, and the spatial and temporal variations of the cosmic-ray flux (2).

On 8 February 1969, a Type-III carbonaceous chondrite fell in south-central Mexico near the village of Pueblito de Allende, scattering fragments over

a large area (3). The Allende meteorite, which was estimated to have weighed several tons (3), is by far the largest carbonaceous chondrite ever observed; several hundred kilograms have been recovered. Fragments are presently being studied by several groups throughout the world. The results should provide detailed information on the radiation exposure of the meteorite and on numerous other processes in which it has been involved. Samples of the Allende meteorite were obtained by the NASA Manned Spacecraft Center in Houston, Texas, within a few days after the fall, and a qualitative radionuclide analysis was made (4). On 21 February 1969, we obtained from R. S. Clarke of the National Museum of Natural History (NMNH) two halves of an individual fragment which had broken apart upon impact with the earth. The concentrations of several cosmogenic and primordial radionuclides were measured by sophisticated gamma-ray spectrometric techniques in

Table 1. Radionuclide concentrations in the Allende meteorite at the time of fall.

Radionuclide	Half-life	Activity (dpm/kg)			
		NMNH No. 3515 (526 g)	NMNH No. 3499 (314 g)	NMNH No. 3525	
				Fragment weighing 852 g	Fragment weighing 905 g
⁷ Be	53 days			< 32*	
²² Na	2.6 years	69 ± 2	76 ± 3	81 ± 2	79 ± 2
²⁶ Al	7.4 × 10 ⁵ years	57 ± 2	58 ± 2	61 ± 2	60 ± 2
⁴⁰ K	1.3 × 10 ⁹ years	430 ± 30	440 ± 40	520 ± 30	480 ± 30
⁴⁶ Sc	83.8 days	5.9 ± 1.0	6.8 ± 1.3	9.2 ± 1.5	7.5 ± 1.3
⁴⁸ V	16.1 days			23 ± 7	18 ± 6
⁵¹ Cr	27.8 days			54 ± 15	
⁵⁴ Mn	303 days	99 ± 4	117 ± 4	129 ± 4	128 ± 4
⁵⁷ Co	267 days				32 ± 4
⁶⁰ Co	5.26 days	41 ± 2	73 ± 3	127 ± 3	128 ± 4
²³² Th (²⁰⁸ Tl)	1.39 × 10 ¹⁰ years			11 ± 3	10 ± 3

* Two standard deviations (2σ) of the gross counting rate at the ⁷Be photopeak area (477 keV) during a 6000-minute count interval with an anticoincidence-shielded Ge(Li) spectrometer.

these two specimens (NMNH No. 3525) weighing 852 and 905 g, and in two additional irregular fragments of other individual specimens (obtained somewhat later) weighing 526 and 314 g (NMNH Nos. 3515 and 3499).

The radionuclides in the meteorite fragments that emit gamma rays were measured by direct counting on multidimensional and both anticoincidence-shielded and normal Ge(Li) gamma-ray spectrometers (5). The counting techniques used have recently been developed for the nondestructive analysis of lunar surface materials (6). The multidimensional spectrometer, which employs two NaI(Tl) detectors (28 cm in diameter by 15 cm thick), offers very high sensitivity for the measurements of those radionuclides which decay by cascade emission of several gamma rays or by emission of a positron and a gamma ray (for example, ²²Na, ²⁶Al, ⁴⁶Sc, ⁴⁸V, and ⁶⁰Co). The anticoincidence-shielded and normal Ge(Li) spectrometers with effective volumes of 30 and 40 cm³, respectively, have a much lower efficiency than the NaI(Tl) detection system but, because of their high resolution, offer a higher sensitivity for radionuclides which decay with the emission of a single gamma ray (for example, ⁷Be, ⁴⁰K, ⁵¹Cr, and ⁵⁴Mn).

The concentrations of radionuclides in the meteorite fragments were measured in the multidimensional and in the Ge(Li) spectrometers for periods of up to 6000 minutes. Standard mock-ups with the same size, shape, physical density, and electron density as each fragment and containing known amounts of each radionuclide of interest were prepared from a mixture of plaster of Paris, iron filings, and water

The concentrations of ⁷Be, ²²Na, ²⁶Al, ⁴⁰K, ⁴⁶Sc, ⁴⁸V, ⁵¹Cr, ⁵⁴Mn, ⁵⁷Co, ⁶⁰Co, and ²³²Th (²⁰⁸Tl) in the meteorite fragments were calculated by direct comparison of their count rates with those of the mock-up standards after application of the appropriate background and Compton corrections.

The concentrations of ²²Na, ²⁶Al, ⁴⁰K, ⁴⁶Sc, ⁵⁴Mn, and ⁶⁰Co were measured in all four of the meteorite fragments. The ⁴⁰K, ⁵⁴Mn, and ⁶⁰Co concentrations, which were measured with both Ge(Li) and multidimensional gamma-ray spectrometers, were well within the expected limits of variation attributed to counting statistics and thus support the internal consistency of the results. The larger size and earlier acquisition of the two halves of specimen NMNH No. 3525 allowed measurements of the additional radionuclides ⁴⁸V, ⁵¹Cr, and ⁵⁷Co, together with an upper limit estimate of the ⁷Be activity. The observed radionuclide concentrations in the four fragments, with the decay corrected to the time of meteorite fall, are presented in Table 1 in the order of increasing ⁶⁰Co content. The uncertainties listed with each concentration are the errors associated with counting statistics in the sample, background, and standard measurements. The radionuclide concentrations observed in the two halves of the specimen NMNH No. 3525 are identical within the limits of error of counting statistics. This excellent agreement, particularly for ²²Na, ²⁶Al, ⁵⁴Mn, and ⁶⁰Co, which show a 2 to 3 percent uncertainty, further attests to the high precision of these nondestructive measuring techniques.

In addition to the usual errors associated with counting statistics, there are

always some uncertainties in the accuracy of radioactive standards. The radionuclides used for mock-up preparation were calibrated by counting portions in a standard geometry on a Ge(Li) diode with an effective volume of 20 cm³ for which a calibration for energy efficiency accurate to 3 percent had been obtained. Since the mock-ups can be prepared so that they are homogeneous in radionuclide content (6), the 3 percent error in the standards should represent the only significant additional uncertainty other than the counting errors listed in Table 1.

The two primordial radionuclides measured in this study were ⁴⁰K and ²³²Th. On the basis of the terrestrial abundances of potassium isotopes, the ⁴⁰K concentrations of 430 to 500 disintegrations per minute (dpm) per kilogram would be equivalent to potassium concentrations of 0.023 to 0.028 percent (by weight). This range is in very good agreement with a concentration of 0.025 percent derived from chemical analysis (3). The ²³²Th concentration of 11 dpm/kg or 0.045 part per million of thorium (as measured from its ²⁰⁸Tl daughter) is in general agreement with most values for thorium that have been reported for Type-I and Type-II carbonaceous chondrites (7).

There was a wide range of ⁶⁰Co concentrations. Cobalt-60 results almost entirely from the neutron capture by stable ⁵⁹Co in large meteorites, and its concentration is therefore very sensitive to position. The very high ⁶⁰Co content of specimen NMNH No. 3525 (128 dpm/kg) is comparable to the reported ⁶⁰Co content in a fragment of the Type-II carbonaceous chondrite Murray (8), but it is approximately an order of magnitude higher than that of most other stony meteorites. The fact that the centers of the two halves of specimen NMNH No. 3525 were about 8 cm apart in the meteorite and that their concentrations of ⁶⁰Co, which are sensitive to position, were the same indicates that this specimen must have been located at a position in the meteorite which provided a uniform neutron flux. This could occur if the specimen were located at the center of the meteorite or at an intermediate depth with its long axis parallel to the closest surface.

Eberhardt *et al.* (9) have related the ⁶⁰Co content at each location in a spherical body of average chondritic composition to the meteorite radius. This makes possible the establishment of a minimum size of the original meteorite body on the assumption that

the sample containing the highest observed ^{60}Co concentration was originally located at the center of the body. The Eberhardt relationships would be applicable to the Allende meteorite since it has a chemical composition very similar to that of average chondritic material (3, 10). The ^{60}Co content in specimen NMNH No. 3525 would require that the meteorite have a minimum radius of approximately 50 cm and thus a total weight of at least 1650 kg. This minimum size is certainly in agreement with the large amount of Allende material which has been collected. Fireman has suggested (11) that the very low hydrogen content of the Allende meteorite might significantly decrease neutron moderation within the body. This would cause our minimum size estimate to be somewhat conservative. To a first approximation, it is assumed that this effect, as well as the fact that the content of stable cobalt in the Allende meteorite is lower than the value used in the Eberhardt model, will not produce a large change relative to other uncertainties inherent in the model. The ^{60}Co concentration observed in this Allende fragment could also occur at regions of intermediate depth in a much larger meteorite. For example, in a spherical chondritic meteorite with a radius greater than 75 cm (some 5500 kg), the ^{60}Co concentration would reach a maximum at a point between the surface and the center; however, even in a meteorite with a radius of 150 cm, the concentration at the center would be only approximately 30 percent below that in the maximum spherical shell. From these considerations, it appears that the meteorite fragment NMNH No. 3515, with a ^{60}Co content of 41 dpm/kg, must have been relatively close to the surface, probably at a depth of 10 to 12 cm in a body with a radius of 50 to 100 cm. The fragment with a ^{60}Co content of 73 dpm/kg must have been located at an intermediate depth.

The proposed preatmospheric locations of the meteorite fragments are also supported by the relative concentrations of the other cosmogenic radionuclides. The concentrations of the four radionuclides ^{22}Na , ^{26}Al , ^{46}Sc , and ^{54}Mn show a positive correlation with the ^{60}Co content and, hence, with the depth of the samples within the preatmospheric body. Since all of these are produced in stony meteorites by both high- and low-energy reactions, it would be expected that their concentrations should increase with depth dur-

ing the growth of a well-developed, low-energy secondary flux. The ^{26}Al concentration seems to be the least sensitive to this depth dependence, possibly because it has no precursors formed by low-energy secondary reactions, as does ^{22}Na . Neutron reactions may also contribute significantly to the production of ^{46}Sc and ^{54}Mn . For example, on the basis of the neutron flux calculated from the observed ^{60}Co concentration in the Allende meteorite and an average concentration of 8 parts per million of Sc in chondrites (12), approximately 15 percent of the ^{46}Sc in specimen NMNH No. 3525 could have been produced by $^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$ and $^{46}\text{Ti}(n,p)^{46}\text{Sc}$ reactions.

The ^{26}Al concentrations observed in the Allende fragments are in good agreement with published values and with the calculated production rate of 57.5 atom/min per kilogram for a Type-III carbonaceous chondrite (13). This indicates that the exposure of the chondrite to cosmic rays was sufficient to produce an equilibrium concentration of ^{26}Al (half-life, 7.4×10^5 years). Thus, the Allende meteorite must have had a cosmic-ray exposure age at its recent preatmospheric size of at least a few mean lives of ^{26}Al , or approximately 3×10^6 years. This is in agreement with the exposure age of 5×10^6 years determined from $^{26}\text{Al}:^{21}\text{Ne}$ ratios in Allende fragments (14).

The ^{57}Co concentration (32 dpm/kg), which was measured only in the most centrally located sample, NMNH No. 3525, was higher than concentrations reported for the Peace River, Harleton, and Bruderheim meteorites (15). This high value for a radionuclide produced by low-energy reactions further substantiates the proposed central location for this fragment.

The concentrations of the other radionuclides in the Allende meteorite are generally in agreement with those reported for other meteorites and would not, of themselves, provide definite information on position or recent cosmic-ray exposure. The observed ^{51}Cr concentration (54 dpm/kg) in specimen NMNH No. 3525 was approximately one-half the reported values for the Peace River and Bruderheim meteorites but comparable to that in the Harleton meteorite (15). The ^{54}Mn concentration (99 to 129 dpm/kg) was higher by as much as a factor of 2 than values for the Harleton meteorite but comparable to values for the Peace River, Bruderheim, and Ehole meteorites (15). The ratio of ^{51}Cr to ^{54}Mn

(0.42) is the lowest yet reported in a chondrite (15) and is in agreement with values obtained from bombardment of thick iron targets (16). The ^{48}V concentration (23 dpm/kg) falls within the range reported for the Peace River, Harleton, and Bruderheim meteorites (15). The ^7Be measurement, which was made only on the more centrally located fragment, NMNH No. 3525, shows a concentration of less than 32 dpm/kg. The production rate of ^{10}Be relative to ^7Be has been estimated to be 0.5 (17). This would indicate a ^{10}Be content in the Allende meteorite of less than 16 dpm/kg, in agreement with the ^{10}Be values measured in other chondrites (15).

Based upon the nine cosmogenic radionuclides observed in this study, the following model of the Allende meteorite is indicated. The preatmospheric body of this meteorite must have had an effective radius of at least 50 cm and a minimum weight of 1650 kg. These dimensions are required to produce the observed ^{60}Co concentrations. A large size is also required to explain the substantial variations in the ^{22}Na , ^{46}Sc , and ^{54}Mn concentrations in the interior fragments of the meteorite. The last major collision to which the Allende meteorite was subjected must have occurred at least 3 million years ago in order that the meteorite have the apparent equilibrium concentrations of ^{26}Al .

The excellent correlations between the ^{60}Co concentration in the Allende meteorite and those of other radionuclides produced by low-energy reactions suggest that an analysis of a large number of Allende fragments for descriptive radionuclides such as ^{22}Na , ^{26}Al , ^{46}Sc , ^{54}Mn , and ^{60}Co would allow a fair reconstruction of their preatmospheric location and possibly of the preatmospheric shape of this meteorite. Of particular interest in such an analysis would be samples of low ^{60}Co content, which should be plentiful and which would indicate a near-surface preatmospheric position. Such surface samples with low ablation could contain relatively large amounts of low-energy spallation products; these values could be used to determine the amounts of low-energy spallation products arising from primary solar protons.

L. A. RANCITELLI

R. W. PERKINS, J. A. COOPER

J. H. KAYE, N. A. WOGMAN

*Battelle Memorial Institute,
Pacific Northwest Laboratories,
Richland, Washington 99352*

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Cytokinin of Wheat Germ Transfer RNA: 6-(4-Hydroxy-3-methyl-2-butenylamino)-2-methylthio-9- β -D-ribofuranosylpurine

Abstract. *A new modified nucleoside is responsible, in part, for the cytokinin activity of transfer RNA from wheat germ. The structure as judged by mass spectrometry is 6-(4-hydroxy-3-methyl-2-butenylamino)-2-methylthio-9- β -D-ribofuranosylpurine. Unequivocal synthesis afforded material having ultraviolet, mass spectral, and chromatographic properties identical with those of the natural product.*

Hydrolyzates of wheat germ tRNA (1) exhibit cytokinin activity in the tobacco callus bioassay. The activity was due, in part, to a compound which was different from the cytokinins known to be naturally occurring, and the identification of this compound is of considerable interest. The new component was isolated by following the fractionation of wheat germ tRNA by means of the tobacco bioassay.

Whole wheat germ (Viobin Corp.) was extracted twice with a mixture of ethanol and ether (1:1, by volume), and the solvent was removed by filtration. The defatted wheat germ was dried overnight at room temperature, ground to a powder, and extracted three times with cold phenol-tris buffer (0.025M, pH 7.3). Precipitation of the RNA from the aqueous phase was effected with cold 95 percent ethanol. The precipitate was triturated with 0.5 percent CTAB in 0.45M sodium chloride (2), and the tRNA was precipitated from the supernatant by the addition of two volumes of 0.5 percent CTAB in distilled water at room temperature. The

tRNA was converted to its sodium form by repeated treatment with 0.4M sodium acetate followed by reprecipitation with three volumes of cold 95 percent ethanol (3). The final precipitate was dissolved in cold 0.1M tris-HCl buffer (pH 7.3) and applied in the cold to a DEAE-cellulose column that had been equilibrated with the same buffer. The column was washed with 0.1M tris buffer containing 0.2M sodium chloride, and the tRNA was eluted with the same buffer containing 1.0M sodium chloride (4). The tRNA was treated twice with phenol-tris buffer; the supernatant was washed with ether and precipitated with cold 95 percent ethanol. The precipitate was washed with ether and dried in a vacuum at room temperature.

The tRNA (2.16 g, 19.7 O.D.₂₆₀ unit/mg) was dialyzed for 48 hours at 4°C before enzymatic hydrolysis. The hydrolysis procedure (5) was similar to that of Hall (6). The lyophilized riboside mixture was extracted with water-saturated ethyl acetate, and the soluble material was dissolved in 35

percent ethanol and purified by chromatography on Sephadex LH-20 (152 g; 52 by 3.65 cm) which had been equilibrated with the same solvent (7); the elution profile was similar to that observed for the fractionation of ribosides resulting from the hydrolysis of *E. coli* tRNA (8). A portion of the cytokinin activity was associated with a region near that in which 6-(3-methyl-2-butenylamino)-9- β -D-ribofuranosylpurine (1) was known to be eluted. Rechromatography on Sephadex LH-20 (20 g, 15 by 2.4 cm)—elution with water—afforded a symmetrical, ultraviolet-absorbing peak. The material in this fraction was dissolved in 95 percent ethanol and further purified by ascending chromatography on Whatman No. 1 paper in 20 percent ethanol. The ultraviolet-absorbing band (R_F 0.60 to 0.75) was eluted with 95 percent ethanol.

Although the chromatographic values of the compound on Sephadex LH-20 were similar to those of 1, the ultraviolet spectrum bore greatest resemblance to that of 6-(3-methyl-2-butenylamino)-2-methylthio-9- β -D-ribofuranosylpurine (3) (5, 8). Since it has been shown (5, 9) that the ultraviolet spectra of ring-substituted *N*⁶-isopentenyladenosines were dependent on the nature and position of the ring substituent, but not particularly on the nature of the *N*⁶-alkyl group, it seemed reasonable to conclude that this modified nucleoside might well be an *N*⁶-alkyl-2-methylthioadenosine.

The natural occurrence of 1 (10), ribosyl zeatin [6-(4-hydroxy-3-methyl-2-butenylamino)-9- β -D-ribofuranosylpurine] (2) (11), and 3 (5, 8) emphasized the possibility of the natural occurrence of a ribosyl derivative of 2-methylthiozeatin (12), [6-(4-hydroxy-3-methyl-2-butenylamino)-2-methylthio-9- β -D-ribofuranosylpurine] (4). If the postulated biochemical conversion of 1 to 3 is correct (9, 13), as shown for the conversion of 1 to 2 (14), then enzymes necessary to produce 4 from 1 are probably available for its biochemical synthesis. In support of the possibility that structure 4 may be the new cytokinin, the chromatographic behavior of this compound indicated that it was indeed more polar than 3. Also, it was anticipated that the ultraviolet spectra of 3 and 4 would be the same, on the basis of the spectra of the respective purines (9).

The high-resolution mass spectrum of the new cytokinin was indeed con-