Cosmic-Ray Production Rates of Be^7 in Oxygen, and P^{32}, P^{33}, S^{35} in Argon at Mountain Altitudes

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The production rates of radioisotopes P^{32}, P^{33}, and S^{35} in argon, and of Be^7 in oxygen have been measured by exposing argon and water to cosmic rays at mountain altitudes for periods of two to four months during 1959. The measured values at $\lambda = 51°$ N, atmospheric depth 685 g cm^{-2} are 7.6×10^{-6}, 6.2×10^{-6}, 1.4×10^{-5} atoms(g argon)$^{-1}$ sec^{-1} of P^{32}, P^{33}, and S^{35}, respectively, and 9.0×10^{-6} Be^7 atoms(g oxygen)$^{-1}$ sec^{-1}.

Isotope production rates for all regions in the atmosphere have recently been calculated by Lal et al. The measured production rates are higher than the calculated rates by factors of 1.1, 1.8, 1.4, and 1.8 in the case of radioisotopes Be^7, P^{32}, P^{33}, and S^{35}, respectively. In this comparison, account has been taken of the fact that cosmic-ray intensity has decreased by about 15% since 1948, the time period to which the calculations apply. The measured production rates in oxygen are assumed to apply to air since cross sections for Be^7 formation have been found to be the same in nitrogen and oxygen over most of the energy region of interest.

The procedure used by Lal et al. yields a fairly accurate picture of the variation in production rates with altitude and latitude in the atmosphere. Isotope production rates in all regions of the atmosphere can, therefore, be obtained by normalizing their calculations at the points where our measurements have been made. The calculated production rates of these isotopes in the troposphere, and in the stratosphere corresponding to observed cosmic-ray intensity during 1948-49, are given. The available data on the concentration of the isotopes in rain-water and their averaged yearly deposition rates are compared with their revised production rates. From such a comparison, more definite conclusions can be drawn than hitherto possible in view of the more accurate knowledge of isotope production rates.

I. INTRODUCTION

THE use of short-lived isotopes produced in the atmosphere by cosmic radiation in the study of meteorology requires a knowledge of their production rates as a function of position in the atmosphere. Lal et al.[1] have calculated the rate of production of four such isotopes (P^{32}, P^{33}, S^{35}, and Be^7) which are produced in the cosmic-ray induced spallation of atmospheric argon, and nitrogen and oxygen, respectively. The relative variations of production rates of isotopes in the atmosphere were based on the variation of stars of particular sizes in which they are produced. Fortunately, this information could be obtained fairly accurately since extensive experimental data on stars and neutrons are available at several latitudes and altitudes. However, relevant cross sections for the formation of isotopes in N, O, and A targets were not available, and the absolute production rates of the isotopes had to be partly based on the general features of nuclear spallation systematics. The absolute star production rates were obtained by normalizing at $\lambda = 50°$ (geomagnetic latitude) and atmospheric depth 680 g cm^{-2} where the rate of production of stars in nitrogen and argon has been measured.[2] These data had to be corrected by a factor of about 2 because of the inefficiency of the instrument used for the detection of small stars. In view of these uncertainties, it was therefore considered important to measure absolute isotope production rates at some point in the atmosphere.

We have now measured the natural production rate of the radioisotopes P^{32}, P^{33}, and S^{35} in argon and of Be^7 in oxygen by exposing argon and water at two mountain altitudes for periods of two to four months.

The new estimates for isotope production rates, which are believed to be correct within $\pm 25\%$, are compared with the available geochemical data on their fall-out in rain and snow

II. EXPERIMENTAL DETAILS

A. Exposure

Water was exposed in plastic swimming pools at Mt. Evans ($\lambda = 51°$, 610 g cm^{-2}) and at Echo Lake ($\lambda = 51°$, 685 g cm^{-2}), Colorado, U. S. A. Details are given in Table I. Water was first decontaminated of its natural Be^7 content by passing it through a 2-liter column of Dowex 50 in the HR form, or by a ferric hydroxide precipitation. The degree of decontamination was not checked in experiments 4W and 6W. In experiment 5W the water from the pool in experiment 4W was used after passing through a Dowex 50 column. The activity of Be^7 was measured in the column. In this experiment the amount of Be^7 present in the pool at the time of exposure was known to be less than 2% of that observed at the end of exposure. The amount of Be^7 activity (atoms/g of water) found in experiments 4W and 6W was about 2 and 6 times higher than that found in 5W, showing incomplete decontamination for our purposes.

Standard size (1A) argon tanks were filled with argon to about 150 atmospheres and then exposed at Mt. Evans and Echo Lake. All relevant details of the exposures are given in Table II. The inside walls of the tanks were coated with polystyrene ($\sim$3 mg cm^{-2}) after a mechanical cleaning by sand blasting. About 500 cc of polystyrene solution in toluene was first poured into the tanks and the excess solution was

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[1] D. Lal, P. K. Malhotra, and B. Peters, J. Atmospheric and Terrest. Phys. **12**, 306 (1958).
[2] W. W. Brown, Phys. Rev., **93**, 528 (1954).

TABLE I. Swimming pool exposure details. (Diameter of pool ~1.5 meters.)

Expt. No.	Location of exposure	Initial average depth of water (cm)	Initial wt. of water (kg)	Final wt. of water (kg)	Period of exposure (1959)	Wt. of carrier added (mg BeO)
4W	Echo Lake	23	420	395	March 19–June 22	150.2
5W	Echo Lake	23	395	370	June 23–August 25	150.2
6W	Mt. Evans	25	460	245	June 24–August 26	150.2

drained off. To ensure uniform coating the cylinders were rotated slowly while they were dried by a stream of dry air. A violet dye in the polystyrene solution permitted visual inspection of the completeness of coating. Two coats were usually necessary. One atmosphere of air[3] was left in the cylinders, to ensure that the phosphorus and sulfur atoms formed oxides rather than complex organic compounds on chemical interactions with the coating of the wall.

In order to determine the amount of radioisotopes produced in the interactions of cosmic rays with the wall of the cylinder, an empty cylinder was also exposed (Table II).

B. Chemical Extraction of Beryllium, Phosphorus and Sulphur from Targets

(1) Beryllium

Water was passed through two cation exchange columns (Dowex 50W-X8) in parallel. The columns were eluted by 3 liters of $3M$ HCl. Hydrogen peroxide was added to the effluents and excess HCl was neutralized by bubbling NH_3 gas. Iron and beryllium hydroxides were precipitated at $pH = 7-8$. The hydroxides were separated and dissolved in $8M$ HCl. Iron was extracted by isopropyl ether. Radiochemical purification of beryllium was carried out using the solvent extraction method developed by Merrill et al.[4]

(2) Phosphorus and Sulfur

Argon gas was passed through two bubblers (Pyrex filter sticks) containing approximately $1M$ NaOH and

0.1% $KMnO_4$ solution in water and one bubbler containing approximately $1.5M$ HNO_3 and 0.1% $KMnO_4$ solution in water. Hereafter we will denote these bubblers as alkaline and acidic bubblers, respectively. Carriers of phosphorus (KH_2PO_4) and sulfur (H_2SO_4) were added to the solutions. The bubblers were arranged in series. The bubbling rate was approximately 5–10 liters a minute. At this rate the size of the bubbles was the order of 1 mm.

After the gas had been passed, the wall of the cylinders was washed first with an alkaline solution ($NaOH + KMnO_4$) containing phosphorus and sulfur carriers (alkaline wash). The polystyrene film was then stripped off by dissolving it in a solution of 0.5% "Hyamine" (p-diisobutyl-cresoxyethoxyethyl dimethylbenzylammonium hydroxide, a quaternary ammonium base, Packard Instrument Company) in toluene. This precaution was taken in order to avoid any chance of reaction of phosphorus and sulfur oxides with the steel walls. The carriers were extracted from the toluene wash solutions by shaking with $0.05M$ NaOH solution and some bromine. (Bromine was added to improve the separation of organic and aqueous layers and for oxidation of trace species.)

Ferric hydroxide precipitations were carried out in the bubbler, alkaline wash and the aqueous extract of toluene wash solutions. Radio-chemically pure magnesium-ammonium phosphate was extracted from the hydroxides using the technique of Marquez and Costa,[5] and then ignited to $Mg_2P_2O_7$, and deposited for counting. Barium sulfate was extracted from the filtrate in the hydroxide step, and converted to sulfur

TABLE II. Argon tank exposure details. (Approximate weight of an empty tank = 49 kg. Average wall thickness ca 5 g/cm^2.)

Expt. No.	No. of tanks	Location of exposure	Period of exposure (1959)	Initial wt. of argon (kg)	Final wt. of argon (kg)
1A	1	Echo Lake	March 19–June 24	12.8	11.4
2A	2	Echo Lake	March 19–July 7	25.6	25.0
3A	2[a]	Echo Lake and Mt. Evans	March 19–June 23 plus June 23–August 25	25.6	24.2
4B	1	Echo Lake	July 7–August 25	Empty	

[a] One of the tanks was not coated with the polystyrene film.

[3] We are indebted to Dr. W. F. Libby for this suggestion.
[4] J. R. Merrill, M. Honda, and J. R. Arnold, Anal. Chem. (to be published).
[5] L. Marquez and N. L. Costa, Nuovo cimento 2, 1038 (1955).

TABLE III. ($P^{32}+P^{33}$) counting data.

Expt. No.	Sample	Carrier added $Mg_2P_2O_7$eq. (mg)	Carrier recovered $Mg_2P_2O_7$ (mg)	Area of deposit (cm^2)	Net counting rate (counts/min) and date (1959)
1A	1st alkaline bubbler[a]	102	69.0	8	0.05±0.04 June 27
	2nd alkaline bubbler	102	75.2	8	0.06±0.04 June 28
	acid bubbler[b]	102	75.0	8	0.01±0.05 July 1
	alkaline wash[c]	102	84.0	10	1.56±0.13 June 27
	toluene wash[d]	102			Sample lost
2A	(1–4) alkaline bubbler	408	265	15	0.14±0.05 July 13
	alkaline wash	30.6	10.3	17	1.38±0.06 July 9
			14.3	8.4	2.13±0.08 July 10
	toluene wash	102	46.4	10	0.17±0.04 July 12
3A	alkaline bubbler	204	173.8	7	0.02±0.03 September 13
	alkaline wash	15.0[e]	38.0	20	1.06±0.06 September 3[f]
			21.6	21	0.43±0.03 September 15[f]
	toluene wash	102	83.4	7	0.08±0.03 September 13
	acid wash of uncoated tank	102	117	20	0.03±0.02 September 21
4B	alkaline wash	204	175.8	7	0.10±0.03 September 13[g]
	toluene wash	102	83.0	7	0.02±0.03 September 13

[a] Alkaline bubbler refers to bubbler containing NaOH ($1M$) and KMnO$_4$ (0.1%) solution. Numbers refer to first and second bubbler.
[b] Acid bubbler refers to bubbler containing HNO$_3$ ($1.5M$) and KMnO$_4$ (0.1%) solution.
[c] Alkaline wash refers to washing of inside tank surface with solution of NaOH ($1M$).
[d] Toluene wash refers to washing of inside tank surface with solution of Hyamine in toluene.
[e] The amount of Mg$_2$P$_2$O$_7$ recovered was 114.1 mg. Extra phosphorus arose from contamination.
[f] Deposited on Lucite sample holder; the rest were mounted on copper holders.
[g] Counted on counter B; other samples were counted on counter A.

which was deposited for counting by sublimation (Goel et al.[6]).

C. Counting of Samples

The 479-kev gamma radiation of Be^7 (11%) was measured in a mercury shielded gamma spectrometer employing a NaI(Tl) (2-in. diameter) well crystal. The background in a channel comprising about 75% of the photopeak was 6 counts/min. The counting was carried out on a 100 channel analyzer (R.I.D.L.). The counting efficiency in the photopeak was experimentally determined to be 17.8±0.9% by counting a Be^7 source calibrated by two independent methods.[7]

The beta counting of phosphorus and sulfur activities was carried out on a low-level beta-assembly[8] employing a cylindrical thin-wall (window thickness ~ 1 mg cm^{-2}) counter. Two counters were used. Their background counting rates were 0.25 (counter A) and 0.35 (counter B) counts per min, respectively, when used inside an 8-in. steel shielding and operated in anticoincidence with cosmic-ray guard counters. The samples were mounted on either Lucite or copper split cylindrical halves. Background counting rates were always taken with the supports before samples were deposited.

The counting efficiencies under the conditions of sample mounting were determined for S^{35}, Co^{60}, Cs^{137}, Tl^{204} standard solutions supplied by the National Bureau of Standards (specified accuracy, ±2–3%) and

[6] P. S. Goel, N. Narsappaya, C. Prabhakara, Rama, and P. K. Zutshi, Tellus 11, 91 (1959).
[7] M. Honda and D. Lal, preceding paper [Phys. Rev. 118, 1618 (1960)].
[8] T. T. Sugihara, R. L. Wolfgang, and W. F. Libby, Rev. Sci. Instr. 24, 511 (1953).

natural K^{40} (KCl). The counting efficiencies for P^{32} and P^{33} were obtained by an interpolation.

Several reagent blanks of phosphorus and sulfur were prepared to check on the contaminations from natural and fission activities. The sulfur and phosphorus blanks showed a positive but small activity of 0.02 net counts/min. The individual rates ranged from 0 to 0.05 counts/min.

III. RESULTS AND DISCUSSIONS

The chemical yield of beryllium in experiment 5W was found to be 76% (Table I). The background and the sample were counted alternately for a period of about 70 hours. The pulse-height distribution of the net activity agreed well with that obtained for an artificial Be^7 sample. The total counting rate in the photopeak was found to be 1.1±0.12 counts/min.

The details of the chemical yields, and net counting rates of phosphorus and sulfur activities are summarized in Tables III and IV, respectively. The chemical yield of phosphorus is not known in the case of experiment 3A because of contamination of stable phosphorus which is believed to have come from the sand (and wall) in the tank which was not coated with polystyrene. Due to an oversight, the tank was not cleaned in this case. The chemical yield in this case was assumed to be 85%. This uncertainty, of course, does not affect the measurements of the ratio P^{33}/P^{32}.

An examination of Tables III and IV shows that most of the phosphorus and sulfur activity appears in alkaline washings of the wall of the tanks. The amount of activity in bubblers and in the toluene wash of the tank ranges between 0 and 10% of the activity found

TABLE IV. S^{35} counting data.

Expt. No.	Sample	Carrier added S eq. (mg)	Wt. of S deposited (mg)	Area of deposit (cm²)	Net counting rate (counts/min) and date (1959)
1A	acidic bubbler	15.5	3.5	4.9	0.03±0.02 July 17
	alkaline wash	15.5	4.6	4.9	0.01±0.04 July 18
	Other samples were lost				
2A	alkaline bubbler	31.0	14.2	4.9	0.06±0.04 July 19
	alkaline wash	62.0	33.0	9.8	0.39±0.04 July 19
	toluene wash	31.0	23.8	4.9	0.10±0.04 July 20
3A	alkaline bubbler	62.0	18.1	4.9	0.01±0.02 September 16ª
	alkaline wash	62.0	30.2	9.8	0.48±0.03 September 17
	toluene wash	62.0	20.7	4.9	0.06±0.03 September 17ª
	acid wash of uncoated tank	31.0	7.2	4.9	0.02±0.03 September 21
4B	alkaline wash	62.0	29.6	4.9	0.06±0.03 September 13ª
	toluene wash	62.0	24.7	4.9	0.00±0.03 September 17

ª Counter B.

in the alkaline wash of the tank walls. Within the errors of counting, only the activity in the toluene wash seems to be significant. We have, therefore, neglected the activity in the bubblers in our calculations; this does not introduce an error of more than 5%.

A very small activity in the blank cylinder is found in the case of S^{35} and P^{32} activities. The observed activities are of the order found in blank samples, except in the case of phosphorus activity in the alkaline wash. If this counting rate is taken seriously, the contribution to $P^{32,33}$ production from the wall amounts to less than 2% of that found in argon.

The counting rates given in Table III are due to both the phosphorus activities P^{32} and P^{33}. Their individual contributions were estimated by counting the sample with and without absorbers (10, 25, and 50 mg cm⁻²) over periods of 2–4 half-lives of P^{32}. The activity of P^{32} has been obtained from counting rates with external absorbers. (The soft beta radiation of P^{33} is strongly absorbed). The counting rate due to P^{33} has been estimated in two ways: (a) by subtracting the estimated counting rate of P^{32} from the total counting rate with no absorber, and (b) by following the gross decay curve

of the composite activities with no absorber. The analysis was carried out using Biller's plot method.[9] The counting rates assigned to P^{32} and P^{33} are summarized in Table V. In these calculations, the absorption coefficients for these activities were calculated using the relation, $0.693/\mu = t_{\frac{1}{2}} = 38E^{1.5}$, given by Libby[10] for cylindrical geometry; μ and $t^{\frac{1}{2}}$ are the absorption coefficient (cm² mg⁻¹) and half thickness (mg cm⁻²) for Al absorbers. The validity of this relation was confirmed by us for Tl^{204} and Cs^{137} beta radiations. The half thickness for vinyl acetate absorbers was found to be 10% higher than that for Al absorbers.

The calculated ratio of the production rates of P^{32} and P^{33} taking into account the counting efficiencies, self absorption in the sample and the duration of exposure, are given in the last column of Table V. The agreement between different determinations is good considering that the counting rate due to P^{33} is small compared to that due to P^{32}. The mean value of the ratio is found to be 0.82 with a measured standard deviation of 0.17.

The calculated rates of production of the various isotopes at Echo Lake, $\lambda = 51°$ N, atmospheric depth

TABLE V. The ratio P^{33}/P^{32} at production.

Sample No. (wt. of $Mg_2P_2O_7$)	Estimated individual counting rates from absorption measurements (counts/min) P^{32}	P^{33}	Estimated counting rate of P^{33} using Biller's plot method (counts/min)	Date (1959)	Ratio P^{33}/P^{32} at production
1A (84 mg)	1.32	0.24	⋯	June 27	0.90
2A (14.3 mg)	1.8	0.33	0.30	July 10	0.60
2A (10.3 mg)	1.12	0.26	0.25	July 9	0.72
2A (combined) (21.6 mg)	0.2	0.09	⋯	August 27	0.77
3A (38 mg)	0.80	0.26	0.35	September 3	1.09
3A (21.6 mg)	0.31	0.12	0.12	September 15	0.81
				Mean value	0.82±0.17

[9] W. Biller, thesis, University of California Radiation Laboratory Report UCRL-2067 (unpublished); A. C. Wahl, Phys. Rev. **99**, 730 (1955).
[10] W. F. Libby, Phys. Rev. **103**, 1900 (1956).

TABLE VI. Measured rate of production of isotopes in oxygen and argon at $\lambda = 51°$, 685 g cm^{-2}.
Production rate, atoms[g target (O or A)]$^{-1}$ sec.$^{-1}$

Nuclide	Expt. No.	Measured rate	Mean	Calculations of Lal et al.[1]
Be[7]	5W	0.90×10^{-5}	0.90×10^{-5}	0.98×10^{-5}
S[35]	2A 3A	1.37×10^{-5} 1.40×10^{-5}	1.4×10^{-5}	0.88×10^{-5}
P[32]	1A 2A 3A	0.74×10^{-5} 0.79×10^{-5} 0.74×10^{-5}	0.76×10^{-5}	0.48×10^{-5}
P[33]	(1–3)A	(see Table 6)	0.62×10^{-5}	0.52×10^{-5}

685 g cm^{-2} are summarized in Table VI. The ratio of isotope production rates at Echo Lake and Mt. Evans was taken to be 1.62 (Lal et al.[1]). According to this ratio, we find that 66% of the observed S[35] activity was produced in the exposure at Mt. Evans. The close agreement between the results from experiments 2A and 3A (Table VI) in fact support the value used for the relative isotope production rates at the two locations.

The production rates have been corrected for the small absorption of the isotope-producing component in the target, and the covering materials, including the roof (1-in. wood$+\frac{1}{16}$-in. Al) below which the argon tanks and the swimming pool were stored. The absorption of the star producing radiation in condensed matter has been reported by several authors (see Rossi[11]). In particular the absorption half-thicknesses in carbon, ice, aluminum, and lead have been reported. We have obtained the absorption coefficients in argon and steel (tanks) by assuming an $A^{\frac{1}{3}}$ dependence. The corrections amount to +6% and +4% in the case of water and argon exposures, respectively.

Exposure in condensed matter on a mountain differs somewhat from the actual case of isotope production in free atmosphere. There exists a possibility of overestimating the production rate due to disintegrations produced by fast π mesons, and capture of negative π mesons at rest. π mesons are more numerous than in the case of free atmosphere for two reasons: (a) fast π mesons produced in the condensed target and surrounding materials produce disintegrations; in the atmosphere they mostly decay since the interaction mean free path in air (in cm) is very large; (b) slow π^- mesons produced in the surface rocks and surrounding materials may lead to isotope production. The production frequency of fast π mesons in nuclear disintegrations and the capture rate of slow π^- mesons at mountain altitudes have been studied (Camerini et al.[12]; Barton et al.[13]). From these data we estimate that the upper limit of nuclear disintegrations induced

by π mesons is 6% of total; the contribution to isotope production seems to be less than 1% under the conditions of the experiments.

IV. ISOTOPE PRODUCTION RATES IN THE ATMOSPHERE

Isotope production rates in different regions of the atmosphere can now be obtained by normalizing the calculations of Lal et al.[1] with the measured production rates at $\lambda = 51°$ and atmospheric depth 685 g cm^{-2}. Our measurements have been made during a period of high sunspot activity. The cosmic-ray intensity exhibits a long term periodic variation opposite in phase to the solar sunspot activity. During the period of exposure the sunspot activity was unusually high, and the cosmic-ray intensity was therefore very low. Changes in the isotope production rates with time can be estimated from the observed variations of the slow neutron component. The neutron intensity at Climax, Colorado ($\lambda = 48°$, 620 g cm^{-2}), decreased by 25% from 1954 to mid-1959.[14] The calculations of Lal et al. refer to the period 1948 since they are based on the measurements of latitude effect of neutron intensity at 312 g cm^{-2} measured by Simpson[15] during 1948. By comparing the sunspot activities during these periods (1948–1959) we estimate that isotope production rates during 1948 should have been higher than during 1959 by about 15%. Taking this into account, we find that in order to agree with our experimental observations the calculated values of Lal et al.[1] should be multiplied by 1.06, 1.82, 1.37, and 1.83 in the case of isotopes Be[7], P[32], P[33], and S[35], respectively.

TABLE VII. Global average isotope production rates in the atmosphere.

Isotope	Production rates (atoms/cm^2 yr)		
	Stratosphere	Troposphere	Total
Be[7]	1.6×10^6	8.0×10^5	2.4×10^6
P[32]	1.6×10^4	8.0×10^3	2.4×10^4
P[33]	1.3×10^4	6.6×10^3	2.0×10^4
S[35]	2.7×10^4	1.5×10^4	4.2×10^4

Average production ratios in the troposphere:
Be[7]/P[32] = 100; S[35]/P[32] = 1.87; P[33]/P[32] = 0.82

[11] B. Rossi, *High-Energy Particles* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1956).
[12] U. Camerini, W. O. Lock, and D. H. Perkins, *Progress in Cosmic-Ray Physics*, edited by J. G. Wilson (North Holland Publishing Company, Amsterdam, 1957), Vol. 1.
[13] J. C. Barton, E. P. George, and A. C. Jason, Proc. Phys. Soc. (London) A64, 175 (1951).
[14] J. A. Simpson (private communication).
[15] J. A. Simpson, Phys. Rev. 83, 1175 (1951).

TABLE VIII. Observed and expected deposition rates of Be^7 and P^{32}; and the minimum ratio Be^7/P^{32} at tropical latitudes.

| | Period of measurement | | | Expected |
	1956	1957	1958	values
Be^7 deposition rate (atoms/cm^2 yr)	4.8×10^5	3.4×10^5	4.3×10^5	4.9×10^5
P^{32} deposition rate (atoms/cm^2 yr)	3.1×10^3	2.6×10^3	4.3×10^3	2.6×10^3
Minimum ratio, Be^7/P^{32} in rains	$\cdots$	100 ± 10	60 ± 5	100

The measured isotope production rates agree well with those calculated by Lal et al. in the case of Be^7 and P^{33}, but are appreciably higher in the case of P^{32} and S^{35}. The agreement in the case of Be^7 was expected as its yield per star was estimated to be 4.2% based mainly on the then available Be^7 production cross sections in carbon. The Be^7 yield/star is probably known more accurately now as 4.9%.[16] The agreement in the case of Be^7 in fact leads to a confirmation of the star production rates in air calculated by Lal et al.[1] No cross sections for the production of phosphorus or sulfur isotopes in spallation of argon are as yet available. The disagreement in these cases can, therefore, not be definitely attributed to either an underestimation of the star production rates in argon or to isotope formation cross sections.

The average global isotope production rates in the troposphere[17] and in the stratosphere are given in Table VII. These values correspond to the cosmic-ray intensities observed during 1948–49, since at atmospheric depths less than 312 g cm^{-2} the relevant cosmic-ray data used refer to this period. A detailed discussion of the time variation of isotope production rates in the atmosphere will be presented elsewhere.

V. COMPARISON OF CALCULATED ISOTOPE PRODUCTION RATES WITH GEOCHEMICAL DATA

The fall-out of the various isotopes has been determined in tropical and mid-temperate latitudes from the measurements of isotope concentration in rains. The ratios of the isotope concentrations, Be^7/P^{32} and and P^{33}/P^{32}, have been measured in individual rain samples.[5,18,19] Observed fall-out rates and the isotope

concentration ratios were compared with those expected on the basis of calculated production rates of Lal et al.[1] in the troposphere. The downward mixing of air from the stratosphere is known to be slow compared to the isotope half-lives so that most of the activity produced in the stratosphere decays *in situ*. Experimental support for this argument was found from the observed fall-out rates of Be^7 at widely different latitudes (Peters[18]). The absolute fall-out rate of Be^7 was found to be close to the amount produced in the troposphere, which is nearly independent of latitude. The measured production rate of Be^7 as determined in the present work checks with the calculations of Lal et al.[1] and, therefore, confirms the interpretation that the observed fall-out of Be^7 is due to its production in the troposphere alone. A similar comparison made for the case of P^{32} showed that its fall-out was higher than that expected from the calculations of Lal et al.[1] by a factor of ~2. The agreement is now good, since the measured production rate of P^{32} is found to be higher by 80% than the calculated value of Lal et al.[1] The observed fall-out rates of Be^7 and P^{32} during 1956 and 1957 in tropical latitudes are compared with those expected using their production rates in the troposphere (Table VII) and a mean residence time of 40 days[5,19] in the troposphere, in Table VIII. The results agree within the uncertainties of fall-out estimates.

The observed value of the minimum ratio of the concentrations of Be^7 and P^{32} in individual rains is found to be in good agreement with the measured ratio at production (Table VIII).

The fall-out of S^{35} at Bombay has varied between 3×10^4 and 1.5×10^5 atoms/cm^2 yr during 1956–1958. The value of 1.5×10^5 is higher than the total production rate in the atmosphere (Table VII) by a factor of ~3.5. This supports the conclusion[20] that an appreciable fall-out of S^{35} is due to nuclear weapons. The fall-out of P^{33} and the ratios of the concentrations of P^{33} and P^{32} in rain samples are found to be higher by about a factor of two than the values expected on the basis of present measurements. It seems difficult to explain the discrepancy by assuming that appreciable amounts of P^{33} have been contributed by bombs, since all possible nuclear reactions favor the production of P^{32}. It seems possible that the disagreement is due to the difficulties in the measurement of P^{33} activity in rain samples.[6]

[16] The cross section for star production in a mixture of nitrogen and oxygen corresponding to the composition of air is 225 mb [D. Lal, thesis, 1958 (unpublished)]. The cross section for the production of Be^7 in bombardments of carbon, nitrogen or oxygen by protons of energies, 225 to 730 Mev has recently been determined.[7] The cross sections are found to be the same for the three targets at any given energy, within ± 10%. Assuming that this is also true for neutrons at lower energies, we have calculated the *average* cross section of Be^7 in air for the energy spectrum of cosmic ray neutrons at $\lambda = 50°$, 680 g cm^{-2}. The average cross section is found to be 11 mb which gives for the yield of Be^7/star, 11/225 $= 4.9$%.

[17] Based on mean summer and winter tropopause heights, as given by Byers in *The Earth as a Planet*, edited by G. P. Kuiper (University of Chicago Press, Chicago, Illinois, 1954), Chap. 7.

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X-Ray Yields from μ-Mesonic Atoms*

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The interesting suggestion has been made that the rapid drop in the yield of mesonic K x rays in the light elements may be associated with the capture of μ mesons into the metastable $2s$ state. The mechanisms for making transitions from the $2s$ to the $1s$ state and from various p states into the $2s$ state have been investigated in detail for Li, Be, and B. It is found that the paradoxical reduction of K x rays remains unexplained. (1) Stark mixing of the mesonic $2s$ and $2p$ states by the electric fields of the atomic electrons allows "mixed" Auger-radiative transitions to the $1s$ state to compete favorably with radiationless transitions. These mixed transitions give a high-energy x ray and a relatively negligible (10–50 ev) electronic excitation and so contribute to the observed K_α yield. (2) Even if the above "mixed" transitions are ignored, there is no mechanism which gets a large fraction of μ mesons into the $2s$ state that at the same time does not violently contradict both theoretical estimates and observed K x-ray yields from light π-mesonic atoms.

I. INTRODUCTION

IN condensed matter the long lifetime and weak nuclear interaction of a μ meson presumably ensures its reaching a K orbit before it decays or is captured by a nucleus. But precisely how such mesons are captured into atomic orbits and the details of their subsequent cascade down into the K state has been understood only imperfectly and without certainty. In fact in the case of the lighter elements (Li, Be, B, and C) there is a striking and paradoxical discrepancy between theoretical calculations and various reported data.[1]

These data concern the yields of those x rays which are radiated when a μ meson makes a transition into a K state or into an L state. In addition to such radiation there exists the possibility of Auger transitions in which the energy difference is carried away by a single high-energy electron. For transitions of mesons into the $1s$ state, Auger transitions are not expected to compete appreciably with radiation and a yield of close to 100% should be observed for the K x-ray yield. The measured yields[2] are given in Table I, where only the relative yields have the small quoted error. (Only those x rays

are counted which are emitted within about 5×10^{-8} sec of the stopping of the μ meson, but this is very long compared to even the most conservative estimate of the time it could take to reach the $1s$ state.) It is apparent that there is a sharp drop in the K x-ray yield for the very light elements beginning with C. But the argument that radiation should proceed enormously faster than electronic Auger transitions is not a delicate one and is quite convincing. The mesonic orbits in the $n=2$ state have about 1/50 the radius of the surrounding K-state electrons. When a meson makes a $2p \rightarrow 1s$ transition the radiated dipole electric field is, as far as the electrons are concerned, identical to that of a point dipole located at the nucleus. An electronic Auger transition can begin to compete with a radiative transition only when the electron is much closer to the dipole than the wavelength of the emitted radiation, i.e., when $kR_e \ll 1$ where $k = 2\pi/\lambda$ and R_e is the expected radius of the electronic K state. With a μ meson in the $n=2$ state around a nucleus of charge Z, the K electrons see an effective nuclear charge $Z-1$ and

$$kR_e \approx \frac{3}{8}\left(\frac{\alpha\mu}{m}\right)\frac{Z^2}{Z-1} = 0.6\frac{Z^2}{Z-1}. \tag{1}$$

Here μ/m is the ratio of μ meson to electron mass and α is the fine structure constant. Even for Li, $kR \sim 3$ and we are enormously far from satisfying the criterion that Auger transitions reduce the K x-ray yield. A more detailed calculation confirms that the theoretical Auger

[1] A recent and comprehensive review of experimental and theoretical work on mesonic atoms is that of D. West, *Reports on Progress in Physics* (The Physical Society, London, 1958), Vol. 21, p. 271. Further references to work before 1958 are contained here.

[2] M. B. and M. Stearns, Phys. Rev. **105**, 1573 (1957).