THE RADIOACTIVITY OF POTASSIUM FROM HUMAN SOURCES

By W. O. FENN, WILLIAM F. BALE, AND LORIN J. MULLINS

(From the Department of Physiology and the Department of Radiology of the School of Medicine and Dentistry of The University of Rochester, Rochester, New York)

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The purpose of this paper is to present data which have accumulated in our laboratory over several years tending to show that the relative abundance of the naturally radioactive isotope of potassium K^{40} in the human body is probably about 2 per cent less than in commercial or "shelf" potassium as supplied in the market. The work was begun on account of the statement of Ernst (1934) based on an inadequate photographic method that K^{40} is (relative to K^{39}) more abundant in living cells. Since the work was begun other papers by Pohlmann (1938), Pohlmann and Netter (1938), Lasnitzki and Oeser (1937), and by A. Lasnitzki (1939) have appeared, all concluding that if K^{40} is more abundant in biological potassium the difference is not over 5 per cent. Since more data are desirable to improve the precision of the result it seems worth while to publish our data. Moreover, since our experiments were completed, Lasnitzki and Brewer (1941) have investigated the abundance of K^{41} in potassium from various sources and have found it abnormally abundant in bone (including the marrow) and in plasma but normal in other tissues except tumors where it was low. Presumably similar but smaller differences should be found for the radioactive isotope K^{40}. In our measurements the K^{40} was directly determined but the results are not wholly consistent with these expectations.

Method

Human ashes were obtained from the crematorium of the dissecting room. It is probable that some ashes from dogs and occasional pieces of wood and wrappings were included, but all of the ash was biological and most of it was human. The ashes were extracted with hot water. Potassium was precipitated from the filtered extract by ammonium perchlorate. The precipitate was filtered off and washed in water and converted to KCl by heating in the oven. This was changed to K_2SO_4 by dissolving in H_2SO_4 and evaporating to dryness. This in turn was converted to potassium acetate by the addition of the calculated amount of barium acetate. The filtrate was evaporated until it was nearly saturated, the final volume being usually about 10 cc. This solution was sampled with a wash-out pipette for a potassium analysis by a slightly modified Shohl and Bennett method and its radioactivity was measured

by a Geiger-Müller counter described by Bale *et al.* (1939). Other samples of shelf potassium (Baker's analyzed and Kahlbaum's for last 2 samples) were treated in a similar way as controls. The actual number of impulses received by the counting circuit was four times as large as the figures given since only 1 out of 4 impulses was passed on to the counter through the scaling circuit. In some preparations the potassium was dissolved from the ashes in HCl and the calcium was subsequently removed by making the solution alkaline. Excess of barium in preparation No. 7 was removed by hydrogen sulfide and the KCl was recrystallized several times before it was finally prepared as the acetate. In this case a spectrographic analysis of the solution, kindly made for us by Dr. L. T. Steadman, indicated that the concentration of rubidium was not greater than one part in 5000. A trace of barium was present in both samples in apparently equal amounts.

RESULTS

Nine different comparisons were made between samples of shelf and of human potassium and the results are shown in Table I. Two to six independent chemical analyses for potassium were made on each sample. Lacking sufficient data to calculate a probable error we have quoted only the maximum deviation of these analyses from the average figure which is taken as the concentration of the solution. The solutions were counted in alternation in periods of 15 minutes (usually). The total time devoted to counting each solution is given in the table as well as the total number of counts recorded for each solution and the background count (using distilled water) calculated for the same length of time. The net counts are then divided by the concentration of the solution and by the dilution factor to give the corrected radioactivity in the next to last column. The relative radioactivity of the human K in terms of shelf K as 1.0 is given in the last column. In all cases (but one) the radioactivity of human K is slightly less than that of shelf K and has an average value of 0.98.

Dilution Correction

In certain of the experiments the potassium concentrations were not the same in the two solutions which were being compared. In such cases it was necessary to make a correction for dilution because the number of counts recorded is not proportional to the concentration but falls off slightly on account of the increasing absorption of the β rays by the solution. To determine the magnitude of this self absorption a strong solution of potassium acetate (80 to 90 gm. per 100 cc.) was diluted to various fractions (0.5–0.8) of its original strength and counts were made in the original and in the diluted solution.

A complete theoretical treatment of the self absorption of β rays is probably too complex to be attempted here but the following approximate formulation will be sufficiently accurate for present purposes. The number of β rays absorbed should be proportional to the number present or to the concentration of K^{40} and the fraction of those present which are absorbed should be proportional

TABLE I

Relative Radioactivity of Shelf Potassium and Human Potassium

No.	Source of K	Concentration of K acetate	Extreme variation	No. of analyses	Dilution correction	Duration of counting	Total counts	Background counts	Net counts	Counts per unit of K corrected	Relative radioactivity
		per cent				*min.*					
1	Shelf	94.5	±1.1	3	1.0	52	2184	133	2051	21.7	1.0
	Human	71.4	±0.5	3	1.04	52	1727	133	1594	21.47	0.989
2	Shelf	94.5	±1.1	3	1.0	61	1744	176	1568	16.59	1.0
	Human	66.2	±0.2	3	1.05	61	1314	176	1138	16.37	0.987
3	Shelf	93.1	±0.6	2	1.0	75	1393	168	1225	13.16	1.0
	Human	59.5	±0.8	2	1.06	75	935	168	767	12.16	0.924
4	Shelf	80.7	±0.6	5	1.0	60	1675	167	1508	18.69	1.0
	Human	67.2	±0.9	6	1.027	60	1448	167	1281	18.56	0.993
5	Shelf	67.2	Diluted from No. 4		1.0	125	3056	353	2703	40.23	1.0
	Human	67.2	Same as No. 4		1.0	125	3004	353	2651	30.45	0.981
6	Shelf	86.8	±1.0	8	1.0	140	4275	543	3732	43.00	1.0
	Human	61.1	±1.6	8	1.05	140	3328	543	2785	43.41	1.01
7	Shelf	51.0	±0.2	3	1.0	50	890	110	780	15.29	1.0
	Human	51.0	±0.3	3	1.0	50	870	110	760	14.90	0.974
8	Shelf	32.2	±.02	3	1.0	200	2414	422	1992	61.86	1.0
	Human	29.6	±.13	3	1.015	200	2214	422	1792	59.65	0.964
9	Shelf	85.2	±0.5	3	1.0	100	2653	200	2453	28.79	1.0
	Human	82.1	±0.2	3	1.006	100	2524	200	2324	28.14	0.977

The next to last column is obtained by dividing the net counts by the dilution correction and by the concentration. In No. 8 the densities of the solution were 1.162 and 1.147 whence the concentrations as determined from density tables were 35.8 per cent and 32.5 per cent. Using these values the radioactivity of the human K was 97.7 per cent of that of the shelf K. These solutions were made from No. 7 after the latter had been partially converted to sulfate for analysis. In No. 7 the human potassium had a slightly higher density (1.245) than the shelf potassium (1.235) although the chemical analyses came out exactly equal. The corresponding concentrations are 56 per cent and 53.5 per cent respectively. Using these values the radioactivity of the human K is 93.7 per cent of that of the shelf K. In No. 9 the densities were 1.342 and 1.335 respectively in the shelf and human samples respectively. Using concentrations read from these values the relative radioactivity of the human sample would have been 0.96.

to the density of the solution or to the total concentration of potassium acetate in grams per 100 cc. In other words the number of counts not recorded be-

cause of self absorption should be proportional to the square of the concentration. Consider two concentrations (grams per 100 cc.) $C_1 < C_0$ giving total counts $T_1 < T_0$. Then

$$\frac{T_0 + kC_0^2}{C_0} = \frac{T_1 + kC_1^2}{C_1} \tag{1}$$

This may be transposed to

$$\frac{T_1/C_1}{T_0/C_0} - 1 = \frac{k(C_0 - C_1)}{T_0/C_0} = K\left(1 - \frac{C_1}{C_0}\right) \tag{2}$$

where the constant $K = \frac{kC_0^2}{T_0}$ and C_0/T_0 is assumed constant. Now T/C may be termed the specific radioactivity of the sample of potassium (number of counts per unit of K) so that in words this means that the specific radioactivity of a dilute solution is higher than that of a concentrated solution by a *factor which, for a given value of* C_0, is proportional to the difference in concentration.

To determine the dilution correction we have made six comparisons between a concentrated solution of K acetate and a solution obtained by diluting that solution to 0.5 or less of its original strength. The results are shown in Table II.

In the first two of these comparisons the dilutions were made by pipettes in the usual way. This, however, introduced some possible error because of the high viscosity of the potassium acetate solution and because of the shrinkage of volume when it is mixed with water. In all the later dilutions the shrinkage of volume was measured and corrected for by measurements of the density of the solutions and in the last three comparisons the dilutions were made altogether on a basis of weight and density rather than by direct volumetric measurement. In Fig. 1 the values of $\frac{T_1/C_1}{T_0/C_0}$ are plotted against the dilution fraction C_1/C_0, the results of the last three comparisons being indicated by open circles. With the exception of one point, which seems to be too high all the points lie fairly well along a straight line through the origin of slope $= K$ according to equation 2. We can find no reason for the high value obtained in this one case; the dilution was checked by density determinations and the solutions were counted with unusual thoroughness on two different counters (3 *a* and 3 *b* of Table II) with almost identical results. We have drawn the curve without regard to this aberrant value and have used it for correcting values obtained in Table I. However, if the higher value had been used it would have made the radioactivity of the human potassium still lower in relation to the shelf potassium.

In Table I all the solutions except Nos. 5, 7, and 9 were prepared directly from human ashes as already described. In all these cases some correction for difference in concentration on the basis of Fig. 1 was required. To avoid this

correction Nos. 5 and 7 were prepared from previous solutions of human potassium to have the same concentration as the shelf potassium used for comparison. No. 9 represented a pooled sample of human potassium prepared from all that remained of all the solutions previously used. Before use it was reprecipitated and purified. Since the correction for dilution can be regarded as having only empirical validity and since such a correction introduces many

TABLE II

Data for Dilution Correction

No.	Concentration of K acetate	Dilution	Density	Duration of counting	Total counts	Background counts	Net counts	Counts per unit of K corrected	Relative radioactivity
	per cent			*min.*					
1	94.5	0.7		40	1141	120	1021	10.8	1.0
	66.2			40	874	120	754	11.4	1.054
2	101.0	0.6		70	1531	145	1386	13.82	1.0
	60.6			70	1040	145	895	14.77	1.069
3*a*	78.0	0.715	1.328	90	2585	332	2253	28.9	1.0
	55.8		1.245	90	2072	332	1740	31.2	1.080
3*b*	78.0	0.715	1.328	90	2374	215	2159	27.7	1.0
	55.8		1.245	90	1877	215	1662	29.8	1.075
4	99.2	0.724	1.395	60	1896	120	1776	17.90	1.0
	71.8		1.302	60	1564	120	1344	18.72	1.045
5	99.2	0.510	1.392	60	1872	132	1740	17.54	1.0
	50.53		1.218	60	1092	132	960	19.00	1.083
6	99.2	0.633	1.395	90	2887	213	2674	26.95	1.0
	62.8		1.267	90	1981	213	1768	28.15	1.044

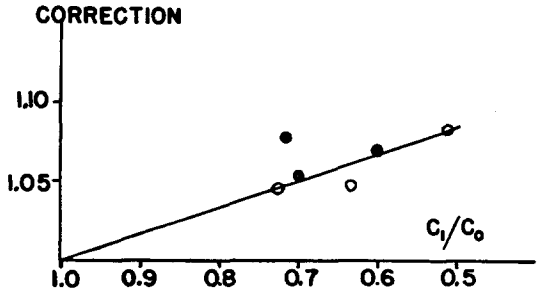

FIG. 1. Correction for dilution of potassium acetate in range where C_0 = about 90 gm. of salt in 100 cc. of solution. The correction $= \frac{T_1/C_1}{T_0/C_0}$ in equation 2.

additional sources of error in the final result most emphasis must be laid upon those comparisons where both solutions were nearly equal in concentration; *i.e.*, Nos. 5, 7, 8, and 9. All these four values agree very closely the average value being 0.975. The average of all the other values where a dilution correction was used was 0.98.

DISCUSSION

It is difficult to estimate accurately the reliability of these measurements. The average error of the K analyses, however, is about 2 per cent. Taking all the comparisons together there were 31 analyses of each type of potassium which gives an error of analysis of about 0.36 per cent. The total number of counts recorded for the human K was 15,092 × 4. Since the random error of counting is proportional to the square root of the number of counts the percentage error becomes 0.41 per cent. The error of a ratio of counts to concentration is then about 0.55 per cent. Taking the average radioactivity of human K as 98 per cent of that of shelf K, it may be stated that a difference of 2 per cent with an error of 0.55 per cent could occur by accident less than once in 1000 times. The error of the final result may also be estimated directly from the results of the nine different comparisons which gave an average value of 0.978 with a probable error of the mean of 0.005. On this basis it may be stated that a difference of 2.2 per cent could occur by chance once in many thousand times. Analysis of our counts on solution 6 for example shows that the ratio of the net counts in the two solutions was determined with a probable error of 0.6 per cent. Thus in any single determination of the relative radioactivity the chief error is in the chemical analyses and no one figure by itself is significantly different from 1.0. Unless, however, there is some consistent error in all the determinations which we have not detected it must be concluded that all nine determinations together indicate a significant deficiency of K^{40} in human potassium.

This deficiency in radioactivity of human potassium cannot be due to radium salts which are stated to be a common impurity in commercial KCl (Bramley and Brewer, 1938) because all of these preparations were sulfated during the procedure. Thus the radium would have been precipitated as sulfate and removed along with the barium sulfate. Spectrographic analysis shows that the rubidium present was too small to influence the count. A separation of isotopes of the magnitude observed is by no means impossible. Even greater separations have been made by distillation of metallic K (Hevesy, Seith, and Pahl, 1931) and by the use of a zeolite tower (Taylor, 1939). In the latter method the heavy potassium exchanges somewhat less readily with other cations combined with the zeolite. It might be mentioned also that the heavy potassium would have a slightly lower diffusion rate. Taking the diffusion rates of the two isotopes as inversely proportional to their atomic weights, their ratio would be $\sqrt{39}/\sqrt{40} = 0.987$ which is not far from the difference observed.

In most cases the shelf potassium used for comparison was reprecipitated, etc. in the same way as the human potassium so that the presence of radium emanation which is reported in commercial potassium[1] could not have vitiated the results. In a few cases, however, the commercial K acetate was used as purchased. To test for radium emanation in such a solution we have compared a boiled (15 minutes) and an unboiled sample of shelf K acetate and have found no change due to the boiling except a possible slight (1 per cent) *increase* in radioactivity without appreciable (0.2 per cent decrease) change in concentration as shown by density.

Assuming that the human potassium does contain only about 98 per cent of the normal radioactivity found in shelf potassium the reasons for this difference may now be considered. One suggestion is that in extracting potassium from the human ashes the lighter potassium was more completely dissolved, especially from the bones. It seems hardly possible, however, that more than a very small fraction of the total potassium of the soft tissues could have remained behind because of the large amounts of fluid used for extraction. The fraction of the total potassium which could be found in the bones may be estimated at less than 5 per cent of the total. If only 10 per cent remained undissolved in the total ash the radioactivity would have to be 20 per cent greater than normal in this residue to explain the observed difference in the fraction extracted.

A second possibility is that the observed difference indicates that the shelf potassium is abnormally high in radioactivity rather than the reverse. We have been unable to obtain information from the manufacturer, however, concerning the origin of the shelf potassium and without this information it is difficult to discuss this point. A comparison between potassium extracted from very old unweathered Canadian granite and commercial potassium has been made by Smythe (1939) but the error of the comparison was of the order of 10 per cent so that the failure to find a difference is of no help in the consideration of our results. Some justification for the use of commercial KCl as a standard may perhaps be derived from the work of Brewer (1939) who used the mass spectrograph to establish the constancy of the K^{40} isotope in potassium derived from various mineral sources.

One much more probable explanation for our results is that the food which we ingest, coming as it does chiefly from plants, may contain relatively too little of the radioactive isotope (although the reverse seems to be the case in the sap of *Valonia* (Jacques, 1940)). The separation could easily be made by the root hairs which could permit a more ready diffusion of the lighter atoms and could somewhat intensify the difference which would be calculated for water diffusion. Against this suggestion is the finding of Brewer (1937) that certain plants (kelp) have an abnormally great abundance of the other heavy

[1] Personal communication from Dr. Brewer.

isotope K^{41}. In potato vines, however, he found a slight deficiency of K^{41} so that at present no general rule seems to hold for plants.

If the plants do not cause the observed deficit in K^{40} then the separation must be made by the animal body. The intestinal mucosa could hardly be responsible for this effect because it absorbs all but about 2 per cent of the total potassium of the food. It seems more likely that the tubular epithelium of the kidney might be responsible if it reabsorbed the light isotopes more rapidly than the heavy ones. (The glomerular membrane would have the opposite effect, if any.) As the excess of light potassium in the body accumulates, however, the chance of losing this excess in the urine also increases until finally a steady state would be reached in which the isotopic composition of the body potassium would become constant. In this condition the excess of light potassium in the glomerular filtrate would be such that the differential absorbing capacity of the tubules for the lighter atoms would just suffice to give the urinary potassium a normal isotopic composition. At this point the isotopic composition of the body would have become fixed and thereafter the isotopic composition of the urine would necessarily be identical with that of the food. One cannot expect to find, therefore, an excess of K^{40} in the urine corresponding to its deficit in the body; nor an increasing deficiency of K^{40} in the urine corresponding to its deficit in the body; nor an increasing deficiency of K^{40} in the body with increasing age. According to the tubule theory the plasma K should have a low radioactivity similar to the average of the body as a whole, since free exchange of K between plasma and tissues has been established (Noonan *et al.*, 1941). We have, however, some tentative evidence (unpublished) that plasma K is not abnormally deficient in K^{40} although there is such a deficiency in the K of the erythrocytes. If this is confirmed, the tubule theory must be discarded.

Finally it is possible to suppose that the cell membranes in the body are responsible for a selection of the lighter isotopes. Lasnitzki and Brewer have suggested an ingenious theory which predicts such an effect where the mobilities of the potassium isotopes are considerably diminished by association with constituents of the tissues. Our results seem to be consistent with this theory but it appears that Lasnitzki and Brewer should have found an abnormally low content of heavy isotopes in the tissue instead of the predominantly normal values which they reported. It is not, therefore altogether easy to reconcile our findings with all of those of Lasnitzki and Brewer (1941) although strictly there is no direct factual conflict.

SUMMARY

A comparison of the radioactivity of potassium from human and commercial sources indicates that the radioactive isotope K^{40} is probably 1 or 2 per cent less abundant in human potassium.

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