

THE MEASUREMENT OF RADIATION EXPOSURE OF  
ASTRONAUTS BY RADIOCHEMICAL TECHNIQUES (a)

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ABSTRACT

The principal gamma-ray-emitting radioisotopes produced in the body of astronauts by cosmic-ray bombardment which have half-lives long enough to be useful for radiation dose evaluation are  $^7\text{Be}$ ,  $^{22}\text{Na}$ , and  $^{24}\text{Na}$ . The sodium isotopes were measured in the preflight and postflight urine and feces, and those feces specimens collected during the manned Apollo missions, by analysis of the urine salts and the raw feces in large crystal multidimensional gamma-ray spectrometers. The  $^7\text{Be}$  was chemically separated, and its concentration measured in an all NaI(Tl), anticoincidence shielded, scintillation well crystal.

The overall sensitivity of the experiment was reduced by almost all variables such as low concentrations of excreted cosmogenic radionuclides, high concentrations of injected radionuclides, low sample sizes, long delay periods before analysis, and uncertain excretion rates. The astronaut radiation dose in millirads, as determined by this technique, for the Apollo 7, 8, 9, 10, 11, 12, and 13 missions was 330, 160, <315, 870 + 550, 31, 110, and <250 respectively. In view of these limitations this technique would be best applied to cases of unusually high exposures, such as that encountered from solar flares.

INTRODUCTION

With the advent of space flight, it has become necessary to determine the radiation dose to man from exposure to the galactic, Van Allen, and solar flare particles. The high-energy galactic portion of the spectrum is fairly constant and has a relatively low intensity. The high intensity Van Allen radiation is of medium energy and localized in space. However, the solar radiation is not so predictable, and the flux and energy of particles from the sun can vary tremendously depending on solar activity. Since high levels of radiation exposure are possible, radiation dosimetry which will properly define radiation exposures is essential in space research programs. Dosimetry methods employed thus far, such as nuclear emulsion films, thermoluminescent dosimeters, and ionization gauges provide very useful indirect methods for estimating radiation dose but are subject to limitations. They measure only a surface exposure at a specific point(s) in the spacecraft or on the astronaut's body rather than an integral whole body exposure, and they have a limited sensitivity to large variations in particle energy. Some of the inherent limitations of these external dosimeters are avoided by using the induced radio-

activity in the body of an astronaut as a measure of his radiation exposure. During a space flight, radionuclides are produced throughout the entire body of an astronaut, and the production rates are related directly to the cosmic particle flux within the body. The absolute and relative amounts of the various radionuclides bear a direct relationship to the intensity and energy spectrum of the particles which are doing the biological damage.

The radiation dose received from the cosmic particles can be determined from the quantities of induced radionuclides<sup>(1-4)</sup>. The amounts of these induced activities can be determined by direct measurement, i.e., whole body counting of the astronaut, or by indirect measurement, such as counting the radionuclides excreted in the feces and urine. The latter approach was used for evaluation of radiation activation during the course of the manned Apollo missions.

The principal gamma-ray-emitting radioisotopes produced in the body by cosmic-ray bombardment are  $^7\text{Be}$  ( $t_{1/2}=53$  day),  $^{11}\text{C}$  ( $t_{1/2}=20.5$  min),  $^{13}\text{N}$  ( $t_{1/2}=9.96$  min),  $^{22}\text{Na}$  ( $t_{1/2}=2.60$  yr), and  $^{24}\text{Na}$  ( $t_{1/2}=15.0$  hr). The primary mode of production of  $^7\text{Be}$  and  $^{11}\text{C}$  is the spallation of carbon, nitrogen, and oxygen in the body. The  $^{13}\text{N}$  comes

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principally from the spallation of nitrogen and oxygen, the  $^{22}\text{Na}$  from the spallation of sodium, phosphorus, and calcium, and the  $^{24}\text{Na}$  from the neutron activation of natural sodium. Of these,  $^{11}\text{C}$  and  $^{13}\text{N}$  are too short-lived to be measured by any method other than a direct determination, and this direct counting would have to be done as soon as possible after recovery. This is unfortunate, since these radioisotopes are produced in the largest abundance. The radionuclides  $^7\text{Be}$ ,  $^{22}\text{Na}$ , and  $^{24}\text{Na}$  are, however, sufficiently long-lived to facilitate their use in making dose estimates from measurement of their quantities in urine and fecal samples.

Other radioisotopes were also expected to be present in the bioassay samples. In addition to the aforementioned cosmogenic radionuclides, measurements of naturally present  $^{40}\text{K}$ ; normally occurring  $^7\text{Be}$ ,  $^{22}\text{Na}$ , and  $^{137}\text{Cs}$ ; and  $^{51}\text{Cr}$  and  $^{59}\text{Fe}$  which were injected for medical studies were also made. Another radioisotope,  $^{60}\text{Co}$ , was detected and quantitatively measured in some of the specimens. Corrections to the cosmogenic  $^7\text{Be}$  and  $^{22}\text{Na}$  must be made to account for the quantities of these radioisotopes normally occurring in the body because of fallout, food intake, and other ingestion processes. The quantities of the naturally occurring  $^{40}\text{K}$  and the injected  $^{51}\text{Cr}$  and  $^{59}\text{Fe}$  in the bioassay samples could serve as biological tracers of various changes of metabolic processes during the course of a mission.

In previous studies, induced radioactivity to radiation dose relationships have been established for the radionuclides  $^7\text{Be}$ ,  $^{22}\text{Na}$ , and  $^{24}\text{Na}$  as a function of energy for proton bombardment of muscle tissue<sup>(2)</sup>. From these relationships and from the ratios in which these radionuclides are produced, the "effective proton energy" of cosmic radiation incident on an astronaut can be determined. This allows the direct estimation of the whole body radiation exposure received by astronauts from measurements of the radionuclides produced in their bodies.

#### EXPERIMENTAL

Preflight and postflight urine and feces and those feces specimens collected in flight were analyzed. Due to the quarantine period following lunar landing missions, all samples were not immediately available for analyses,

thus allowing the short-lived radionuclides to decay. The urine specimens which were of small volume were solidified prior to analysis by the addition of  $\text{CaSO}_4$  to 25 ml or less of the raw urine in order to form a standard counting geometry. Any samples of initial volume greater than 25 ml were treated by repeatedly boiling to dryness with nitric acid to destroy the organic matter present. The remaining salts were counted in large crystal multidimensional gamma-ray spectrometers<sup>(5-7)</sup> for determination of  $^{22}\text{Na}$ ,  $^{24}\text{Na}$ ,  $^{40}\text{K}$ ,  $^{51}\text{Cr}$ ,  $^{59}\text{Fe}$ ,  $^{60}\text{Co}$ , and  $^{137}\text{Cs}$ . The salts were then redissolved in a weak HCl solution and diluted to known volume. An aliquot of this solution

was taken for neutron activation analysis to determine the concentrations of stable elements in the sample. The remainder of the solution was reduced in volume to approximately 15 ml and transferred to a 100 ml polyethylene centrifuge tube. Approximately 5 mg of  $\text{Be}^{++}$  carrier and 20 mg of  $\text{Fe}^{+++}$  carrier were added, and the solution was neutralized with concentrated  $\text{NH}_4\text{OH}$ . After centrifugation the supernatant solution was discarded. Thirty-five ml of 3 N NaOH were added to the remaining precipitate and stirred vigorously until well mixed. After centrifugation the supernatant liquid was transferred to a clear centrifuge tube, saturated with  $\text{NH}_4\text{Cl}$ , and heated in a water bath. If necessary, additional  $\text{NH}_4\text{Cl}$  was added until a  $\text{Be}(\text{OH})_2$  precipitate settled from the solution. The solution was then centrifuged, and the supernatant fraction was discarded. The resulting quantitative precipitate containing the  $^7\text{Be}$  activity was counted in an all NaI(Tl) anticoincidence shielded, 7-inch diameter scintillation well crystal in the absence of all interfering activities. This was necessary in order to measure the relatively small quantities of  $^7\text{Be}$  present.

Fecal samples were thoroughly mixed in their collection bags to ensure homogeneity of the specimens. A small corner was cut off each bag and aliquots were extruded into standard counting geometry containers for measurements on multidimensional gamma-ray spectrometers to measure the radioisotopes  $^{22}\text{Na}$ ,  $^{40}\text{K}$ ,  $^{51}\text{Cr}$ ,  $^{59}\text{Fe}$ ,  $^{60}\text{Co}$ , and  $^{137}\text{Cs}$ . Separate aliquots were wet ashed with nitric acid and hydrogen peroxide to destroy the organic matter present. The resulting salts were dissolved in dilute nitric acid, and the same procedure as above was followed for separation of the  $^7\text{Be}$  activity.

A luminous material composed of  $^{147}\text{Pm}$  microspheres mixed with a scintillator is used extensively in the spacecraft in acrylic switch tips and sighting figures used in docking maneuvers. Because of the high rejection rate of switch tips caused by promethium leaks, there is some concern about the possible presence of  $^{147}\text{Pm}$  in the weightless space capsule environment. For the later missions, approximately 10 mg of mixed rare earths were added to the feces prior to wet ashing. These were to serve as carriers for  $^{147}\text{Pm}$ , which could possibly have been ingested by the crew members. This rare earth fraction was separated from the beryllium fraction after the initial  $\text{NH}_4\text{OH}$  precipitation by dissolving the precipitate in approximately 8 ml of 3M HCl and adding 2 ml of 49 percent HF. Centrifugation separated the rare earth precipitate from the beryllium in the supernatant solution. The rare earth fraction was then dissolved in two parts concentrated  $\text{HNO}_3$  and three parts saturated boric acid solution and reprecipitated with  $\text{NH}_4\text{OH}$ . After centrifugation and decantation, the precipitate was dissolved in dilute HCl; and saturated oxalic acid solution was added to precipitate the rare earth oxalates. The solution was centrifuged; the supernatant solution was decanted; and the quantitative precipitate was washed with alcohol, transferred to a 1-inch diameter stainless steel dish and counted in an end window, gas flow beta counter for the measurement of  $^{147}\text{Pm}$ .

RESULTS

The results of the individual determinations are given in Tables I through IV. All data have been normalized to a gram of feces, a milliliter of urine, or a gram of the respective stable element as determined by a technique of instrumental neutron activation analysis(8) All data have been decay corrected to the time of splash-down of each respective mission. The results of all the radionuclide measurements in the excreta are given in the tables although only the concentrations of the cosmogenic radionuclides 7Be, 22Na, and 24Na are of importance for the subject matter of this communication. The various samples in the tables are listed by the letters A, B, and C or LMP, CMP, and CDR to identify an individual astronaut. Those samples listed by numbers are unidentified and arbitrarily coded. The collection time for each specimen is given as Pre-, In-, or Post-flight unless more detail is known, in which case a number refers to elapsed time into the mission in hours, the letter F followed by a number indicates that number of days prior to flight, post+0 refers to the first voiding after splashdown, post+1 is the first 24 hour collection after splashdown and day 2 is the following day after splashdown.

TABLE II
RADIONUCLIDES IN FECES FROM APOLLO ASTRONAUTS

Table with columns: MISSION, SAMPLE IDENTIFICATION, FLIGHT PERIOD, 7Be, 22Na, 24Na, 40K, 51Cr, 59Fe, 60Co, 137Cs, 147Pm. Data rows include missions 7, 8, 9, 10, 11, 12.

TABLE I
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TABLE III
RADIONUCLIDES IN URINE FROM APOLLO ASTRONAUTS

Table with columns: MISSION, SAMPLE IDENTIFICATION, FLIGHT PERIOD, 7Be, 22Na, 24Na, 40K, 51Cr, 59Fe, 60Co, 137Cs. Data rows include missions 7, 8, 9, 10, 11, 12.

TABLE IV  
RADIONUCLIDES IN URINE FROM APOLLO ASTRONAUTS

MISSION	SAMPLE IDENTIFICATION	FLIGHT PERIOD	ACTIVITY IN DISINTEGRATIONS/MINUTE/GRAM INERT ELEMENT ON DAY OF SPLASHDOWN			
			<sup>22</sup> Na/g Na	<sup>24</sup> Na/g Na	<sup>60</sup> Co/g Co	<sup>137</sup> Cs/g Cs
7	A	POST	1.4±0.8	20±13		
7	B	POST	4.9±1.6			
8	B	POST	1.6±1.2			
11	CMP	PRE				(2.2±1.4) · 10 <sup>6</sup>
11	LMP	PRE	0.51±0.40			(1.07±0.12) · 10 <sup>7</sup>
11	CDR	PRE				(5.17±0.82) · 10 <sup>6</sup>
11	CMP	POST	0.48±0.30		(1.3±1.1) · 10 <sup>6</sup>	(7.2±6.8) · 10 <sup>5</sup>
11	LMP	POST				(2.4±1.4) · 10 <sup>6</sup>
11	CDR	POST	0.43±0.23			(2.0±0.80) · 10 <sup>6</sup>
12	CMP	F-30				(2.3±0.27) · 10 <sup>7</sup>
12	LMP	F-30				(1.80±0.17) · 10 <sup>7</sup>
12	CDR	F-30	0.49±0.19			(5.78±0.99) · 10 <sup>6</sup>
12	CMP	F-15				(5.22±0.31) · 10 <sup>7</sup>
12	LMP	F-15				(1.77±0.16) · 10 <sup>7</sup>
12	CDR	F-15				(1.15±0.17) · 10 <sup>7</sup>
12	CMP	POST+0				(1.45±0.43) · 10 <sup>7</sup>
12	LMP	POST+0				(1.98±0.12) · 10 <sup>8</sup>
12	CDR	POST+0				(4.51±0.41) · 10 <sup>7</sup>
12	CMP	DAY 2	4.4±3.6			
12	LMP	DAY 2			(5.7±3.0) · 10 <sup>6</sup>	(1.97±0.35) · 10 <sup>7</sup>
12	CDR	DAY 2	2.2±1.1			

The average values of the cosmogenic radionuclide concentrations in each basic flight period are summarized in Table V according to the various methods of normalization. The increase in the activities from preflight to inflight and postflight periods should be indicative of the exposure to cosmic radiation. The concentrations of each radionuclide increase rather regularly for the Apollo 7 mission regardless of the method of normalization. However, the fecal data for the Apollo 8 mission are quite irregular, with only the urine data demonstrating increases in the cosmogenic radionuclides. The Apollo 9 and 13 missions show increases in the <sup>7</sup>Be concentration in the urine but demonstrate decreases in the <sup>22</sup>Na concentrations while the reverse is true for Apollo 11. Regular increases are shown for Apollo 10 and 12.

The increases in cosmogenic radioactivity from preflight levels to those after exposure to the space environment are almost certainly due to cosmic particle activation. Equating the magnitude of the increase with the radiation dose delivered by the particles is still fairly difficult, particularly when the dose is quite small as has been the case on all manned Apollo missions thus far. Concentrations normalized to the unit mass or volume of excreta are subject to variation in the biological dilution of the specimen. Concentrations normalized to the unit mass of stable element in the feces are also subject to variations in the quantities of unmetabolized elements passing through the gastrointestinal tract. Only the quantities of radionuclides in the urine normalized to the amount of stable element present can be expected to be reasonably representative of the specific activity in the whole body since the urine contains only metabolized material.

Indeed, it is necessary to make some assumptions regarding the percentages of the body burden of an element excreted in the feces or urine, the relative dilution factors of feces and urine, and the "contamination" of feces by unmetabolized elements in order to compare the data with the experimental results for proton irradiated muscle tissue<sup>(1, 2)</sup>, proton irradiated radiotherapy patients<sup>(3)</sup>, and neutron irradiated radiotherapy patients<sup>(4)</sup>. In this manner, the average effective proton energy incident on the astronauts and the radiation dose received by them can be estimated. The details of these calculations will be omitted here since they are given elsewhere<sup>(9-12)</sup>. The results indicate an average effective proton energy of 38-40 MeV incident on the Apollo 7 mission astronauts and

<38 MeV on the Apollo 8 mission astronauts. Radiation doses of 480 + 310, <315, 870 + 550, <480, and <250 millirads for the Apollo 7, 9, 10, 12, and 13 missions respectively are calculated.

Since the specific activity of the cosmogenic radionuclides in the urine should be a more accurate representation of the whole body burden of induced radioactivity, the specific activity of the <sup>22</sup>Na in the postflight urine of astronauts is compared to the specific activity of <sup>22</sup>Na in the urine of radiotherapy patients who have received a known radiation dose. This comparison leads to estimated cosmic radiation doses received by the astronauts on the Apollo 7, 8, 11, and 12 missions of 330, 160, 31, and 110 millirads respectively. It should be pointed out here that the uncertainty of the data given in Table V, and hence of these results, is quite large in some instances.

#### DISCUSSION

In principle the relationships between induced activity and radiation dose are straightforward. The probability for production of a certain isotope in the body of an astronaut is basically a function of the energy of the proton. Similarly, the radiation dose from a cosmic proton is also a function of its energy, and therefore, the induced activity is logically related to the radiation dose. Such relationships have been empirically determined for several different situations<sup>(1-4)</sup>, and it remains only to measure the quantities of induced radionuclides in a particle irradiated person to determine the dose he received.

In practice, however, the procedure is not quite as simple as that just described. A calibrated whole-body counter is required to determine the quantities of induced radionuclides, and a high sensitivity-low background instrument would be required to measure the small quantities of radionuclides induced by the low levels of cosmic radiation encountered on a normal space flight. In lieu of the availability of a suitable whole-body counter, an indirect approach such as that used in this work can be applied. The principal limitations to this method have already been touched upon above. Only a small and uncertain fraction of the induced activity is eliminated in the excreta. Thus only the

TABLE V

## AVERAGE RADIONUCLIDE CONCENTRATIONS IN EXCRETA FROM APOLLO ASTRONAUTS

ACTIVITY	FLIGHT PERIOD	NORMALIZED RADIOACTIVITY IN DISINTEGRATIONS PER MINUTE						
		APOLLO 7	APOLLO 8	APOLLO 9	APOLLO 10	APOLLO 11	APOLLO 12	APOLLO 13
<sup>7</sup> Be/g Feces	Pre	1.12	3.25					
<sup>7</sup> Be/g Feces	In	1.59	1.78	1.25	0.36	0.10	2.79	0.98
<sup>7</sup> Be/g Feces	Post	2.94	2.33					
<sup>22</sup> Na/g Feces	Pre		0.025					
<sup>22</sup> Na/g Feces	In	0.0014	0.0014		0.015		0.0007	0.0039
<sup>22</sup> Na/g Feces	Post	0.0026	0.040					
<sup>22</sup> Na/g Na in Feces	In	0.26	0.26		7.8			
<sup>22</sup> Na/g Na in Feces	Post	2.8						
<sup>7</sup> Be/ml Urine	Pre	0.159		0.014		0.264		0.233
<sup>7</sup> Be/ml Urine	Post	0.755	1.20	0.055	0.077	0.051		0.68
<sup>22</sup> Na/ml Urine	Pre			0.0005	0.0004	0.0003	0.0003	0.0013
<sup>22</sup> Na/ml Urine	Post	0.0038	0.0009	0.0002	0.0009	0.0008	0.0016	
<sup>24</sup> Na/ml Urine	Post				0.04			
<sup>22</sup> Na/g Na in Urine	Pre					0.17	0.08	
<sup>22</sup> Na/g Na in Urine	Post	3.2	1.6			0.30	1.1	
<sup>24</sup> Na/g Na in Urine	Post	20						

specific activity of an induced radioisotope in the urine can be extrapolated to the whole body burden with a reasonable degree of accuracy.

While the efficiency of low-level sample counters is routinely several orders of magnitude higher than whole-body counters, the small fraction of the total body activity in any bioassay sample reduces the sensitivity of a specimen measurement to the point where it is little better than that of a whole-body count. To complicate the situation in this work even further, there is a large demand for aliquots of post-flight urine specimens from the astronauts and typically only 10% or less of a 24-hour collection has been available for radionuclide concentration measurements. An additional complication in the case of non-lunar-landing missions (Apollo 13 excepted) was the injection of radioisotopes into the astronauts for medical studies. Although these isotopes were not the same as the cosmogenic radionuclides measured, their presence in the excreta lowered the overall accuracy of the measurements. Finally, the quarantine requirements of the lunar landing missions caused a delay in the analysis of postflight specimens which allowed substantial decay of the radionuclides present. These factors all contribute to the reduced accuracy and sensitivity of the measurements reported herein. In an effort to improve the situation, a high sensitivity combination whole-body counter and sample counter has been proposed which could be rapidly utilized after a mission (even onboard the recovery vessel) to make accurate measurements of the whole body burden of radionuclides in the astronauts. The combination of direct measurement of whole body burdens of radionuclides and the early measurement of relatively large quantities of excreta should make much more accurate dose estimates possible.

This technique for measurement of radiation dose should be perfected during routine space missions so that in the event of an unusually high exposure, such as might be expected from a solar flare, an accurate determination of the radiation dose can be obtained. This situation would be analogous to those nuclear criticality accidents<sup>(13-15)</sup> where conventional dosimetry techniques were saturated and induced radioactivity was measured to interpret the radiation dose received by the exposed individuals.

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