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QUARTERLY RESEARCH REPORT
TO THE NASA MANNED SPACECRAFT CENTER

THE MEASUREMENT OF RADIATION EXPOSURE OF ASTRONAUTS BY RADIOCHEMICAL TECHNIQUES

OCTOBER 6, 1969 THROUGH JANUARY 4, 1970

January 15, 1970

AEC RESEARCH & DEVELOPMENT REPORT

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UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

BNWL-1183 3

UC-41, Health and Safety

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by

R. L. Brodzinski, L. A. Rancitelli, and W. A. Haller

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Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.65

TABLE OF CONTENTS

List of Tables iii

Abstract 1

Task - Determination of the Radionuclide Content of Feces and Urine From Astronauts Engaged in Space Flight 2

Task - Neutron Activation Analysis of Feces and Urine From Astronauts Engaged in Space Flight 3

- Alkali Elements 4
- Transition Metals 6
- Miscellaneous Elements 7

Task - Glass Fibers in Astronaut Fecal Samples 9

Task - Induced Radionuclides in Spacecraft 10

Expenditures 12

References 13

LIST OF TABLES

I	Radioactivity in Feces From Apollo 11 Astronauts	14
II	Radioactivity in Urine From Apollo 11 Astronauts	15
III	Na, K, Rb, and Cs Concentrations in Astronaut Fecal Samples	16
IV	Fe, Co, Zn, Cr, and Sc Concentrations in Astronaut Fecal Samples	17
V	Br, Se, Hg, Ag, Sb, Au, and Sn Concentrations in Astronaut Fecal Samples	18
VI	Glass Fiber Concentrations in Apollo 11 Astronaut Fecal Samples	19

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ABSTRACT

The urine and feces specimens from the Apollo 11 mission were analyzed for their radionuclide content. Estimates of cosmic radiation dose received by the astronauts were difficult to determine due to decay of the short-lived radionuclides during quarantine. The concentrations of ^{7}Be, ^{22}Na, ^{40}K, ^{59}Fe, ^{60}Co, and ^{137}Cs were determined. No ^{147}Pm was observed in any of the samples.

The concentrations of 16 major, minor, and trace elements were determined in fecal samples from Apollos 8 and 10. Large discrepancies between the excretion rates and normal dietary intakes were noted for cobalt, iron, tin, and potassium. An interpretation of the hazards these deviations may produce requires the determination of the elemental concentrations of the foodstuffs used during these missions.

The fecal samples from the Apollo 11 mission were analyzed for glass fiber content. One anomalous sample was observed having a glass fiber content twofold greater than any previously measured specimen.

A piece of the outer thermal coating of the Apollo 12 spacecraft was analyzed for cosmic-ray-induced radioactivity. Beryllium-7 was observed.

TASK - DETERMINATION OF THE RADIONUCLIDE CONTENT OF FECES AND URINE FROM ASTRONAUTS ENGAGE3 IN SPACE FLIGHT

Astronauts engaged in space flight are subject to cosmic radiation which does biological damage to, arid induces radioactive isotopes in, their bodies. The radiation dose received from the cosmic particles can be determined from the quantities of induced radionuclides.(1) 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 arid urine. The latter approach was used for evaluation of radiation activation during the course of the Apollo 11 mission. The urine and feces specimens from the Apollo 11 astronauts were examined for their radionuclide content using previously developed procedures. (1, 2) Due to the quarantine period following splashdown, the samples were not immediately available for analyses, thus allowing the short-lived radionuclides to decay.

The radioactivity in the feces samples from the Apollo 11 astronauts is summarized in Table 1. These inflight samples were collected on the trip from the moon to the earth. Prior inflight specimens were jettisoned with the lunar excursion module. The measured ^{40}K, ^{59}Fe, ^{60}Co, and ^{137}Cs activities are normal in comparison with the results of previous missions although the source of the ^{60}Co ha; not been established. The absence of ^{59}Fe activity in samples 2, 3, and 4 indicates that tney are not from the same astronaut as sample 1. Although ^{7}Be was detected in one sample, it, value is uncertain. The loss of analytical precision was due, in part, to the ^{7}Be decay during sample quarantine. Since ^{7}Be was the only cosmogenic radionuclide detected, no accurate cosmic radiation dose estimate is available.

The radioactivity concentrations in the urine samples, which are summarized in Table II, are similar to those determined in samples from previous Apollo flights. The standard deviations in ^{22}Na concentrations do not allow a radiation dose calculation to be made. In fact, the ^{7}Be activities observed in the preflight urine specimens are higher than the ^{7}Be activities observed in the postflight specimens.

A luminous material composed of ^{147}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 possibility of the presence of ^{147}Pm in the weightless environment in the space capsule, No ^{147}Pm (< 16 pCi per defecation) was observed in any of the specimens (see Table I) indicating that ^{147}Pm in the Apollo 11 space capsule environment was minimal.

TASK - NEUTRON ACTIVATION ANALYSIS OF FECES AND URINE FROM ASTRONAUTS ENGAGED IN SPACE FLIGHT

This program has been instituted in an attempt to foresee any possible metabolic changes in astronauts caused by conditions of weightlessness and prolonged physical inactivity which are manifested by an uptake or loss of an element or elements by their bodies. While the primary concern is the terrestrially observed phenomenon of osteoporosis (loss of skeletal calcium), changes in the uptake and excretion rates of other essential microconstituents of the body, such as cobalt, iron, selenium, and the alkali metals which act as electrolytes, are also important. The concentrations of 17 major, minor, and trace elements are being measured in the feces and urine from astronauts engaged in space flight by a previously described technique[1, 2] of instrumental neutron activation analysis.

Neutron activation analyses of the fecal samples from Apollo 11 and the urine specimens from Apollos 9, 10, and 11 have been initiated. The concentrations of Na, K, Rb, Cs, Fe, Co, Zn, Cr, Sc, Br, Se, Hg, Ag, Sb, Au, and Sn In the Apollo 8 and 10 fecal samples are summarized in Tables III through . The calcium concentrations for the two missions have been reported previously.(2) All samples from the Apollo 8 mission are from the same astronaut. Two aliquots from sample 3 were analyzed to demonstrate the homogeneity of the specimen and the reproducibility of the neutron activation technique. Although the three Apollo 10 samples are undocumented, they were all collected after transearth injection.

Since all the samples summarized in Tables III through V are undocumented as to elapsed time into the mission, only an integrated excretion rate can be determined. This can be accomplished by dividing the total weight of each element excreted in a mission by the length of the mission and the number of participating astronauts. Although the uncertainty of the collection period for Apollo 10 refutes the use of this procedure, it has been utilized to compare the fecal excretion rates from Ppollos 7, 8, and 9 with the normal rates on earth.

ALKALI ELEMENTS

The alkali metals occupy a unique position in the chemistry of the metabolic processes and are generally thought to act as electrolytes essential to the proper functioning of the nervous system, as well as establishing the osmotic balances necessary for the transfer of essential material across cellular membranes. Although the chemistry of the alkali metals is dominated by the monovalent state, they apparently function in different ways throughout a biological system.

Four of six samples analyzed show extremely high concentrations of sodium. Only samples S/N 3513 and S/N 3527 from the Apollo 10 mission exhibit a normal sodium excretion concentration. The high values of the other four samples are probably due to the addition of the preservative-deodorant sodium orthophenylphenol, which is in the fecal collection kits. This sodium salt addition precludes making meaningful observations on the electrolytic balance of the astronauts.

The average fecal excretion rate of potassium for the Apollo 8 mission astronaut is 0.499 g K/day. This is a slightly higher fecal elimination rate than the 0.311 g K/man day observed in the Apollo 9 mission,[2] and when corrected for total body excretion[3] yields a value of 2.49 g K/day. Although this is above the excretion rate detected in Apollo 9 samples, it is not equal to the normal dietary intake of 2.85 to 3.0 g/day.[3, 4] This combination of loss and dietary intake would result in a 5 gram increase in potassium in the body for an eleven-day mission or a change of 1 part in 36.

Very little is known of the chemistry of rubidium and cesium in the human body. Only 23% of the eliminated rubidium is found in the feces,[5] which would give a total body excretion rate of 4.57 mg Rb/day for the Apollo 8 astronaut from the data in Table III. This is slightly higher than the values of 3.79 and 2.86 mg Rb/man day observed for Apollo 7 and Apollo 9[2] respectively and almost twice as high as the normal intake of 2.53 mg Rb/day.[5]

The fecal excretion rate of 3.57 μg Cs/day observed for the Apollo 8 mission astronaut is considerably higher than the values of 1.4 μg Cs/man day and 2.0 μg Cs/man day observed in the Apollo 7 end 9 missions respectively.[2]

TRANSITION METALS

Of the transition metals, iron is an integral part cf the heme molecule, cobalt plays a role in the chemistry of vitamins and stimulates the hematopoietic system, while the body functions of zinc and chromium are poorly understood. All transition elements, with the exception of scandium and titanium, are biologically active.

The iron concentrations summarized in Table IV yield an Apollo 8 average fecal excretion rate of 13.3 mg Fe/day. Since iron is primarily eliminated in the feces,(3) this approximates the total iron excretion rate end compares well with the valuer of 16 and 14 mg Fe/man day observed for Apollos 7 and 9 respectively.(2) However, all iron excretion rates observed for Apollos 7, 8, and 9 ere lower than the lowest quoted value for the normal dietary intake, which is 27 mg Fe/day.(4) Two previous explanations exist: (1) The astronauts are concentrating iron or (2) the foodstuffs contain a lower iron concentration. The latter possibility can be easily evaluated by a trace element analysis for iron in the astronaut's food supply.

The fecal excretion rate of cobalt for the Apollo 8 astronaut is 10.5 μg Co/day, which compares well with values of 10.2 and 13.3 μg Co/man day observed for the Apollo 7 and 9 missions.(2) The total cobalt excretion rate, 77.2 μg Co/day, was calculated assuming 13.6% of the cobalt is eliminated in the feces.(3) Since the normal dietary intake of cobalt is only 5.67 μg(3) to 7 μg(4) per day, either the foodstuffs contain high cobalt concentrations or the astronauts are losing cobalt from their systems. This apparent loss of cobalt could become extremely important considering the role it plays in stimulating the hematopoietic system.

Although the exact functions of chromium and zinc in the body ere not yet fully understood, it is felt they are essential elements. The zinc fecal excretion rate of the Apollo 8 astronaut, 12.0 mg Zn/day, compares well with the values from other missions.[2] Assuming 85% of the zinc is eliminated in the feces,[3] a total excretion rate of 14.1 mg Zn/day is calculated, which is in the range of the normal dietary intake of 8.3 to 17 mg/day.[3, 4]

Fecal excretion rates of 100 and 78 μg Cr/man day were observed for Apollos 7 and 9,[2] while the chromium fecal excretion rate in the Apollo 8 astronaut was 117 μg Cr/day. These values are fairly similar to the normal daily intake of chromium, which is approximately 150 μg/day.[4] These concentrations could be combined with data obtained from a trace element study of the food to provide the as yet unknown chromium excretion percentage in the feces.

Of the transition metals, nothing is known of the metabolic function of scandium. The fecal excretion rates from Apollos 7 and 9 are 0.85 and 1.0 μg Sc/man day respectively[2] and are comparable to the fecal excretion rate of 1.09 μg Sc/day obtained for the Apollo 8 astronaut.

[illegible]

A number of elements with as yet unknown functions are present in biological systems at concentrations comparable to biologically essential elements. Of the miscellaneous elements whose concentrations are summarized in Table V, only tin varied significantly from a normal intake.

A fecal excretion rate of 209 μg Br/day was calculated from the bromine concentrations in Table V for the Apollo 8 astronaut. This value is higher than the 151 and 113 μg Br/man day observed for the Apollo 7 and 9 missions[2] but is still a very small fraction of the normal daily intake of 17,000 μg,[4] most of which is excreted in the urine.[3]

A very large difference is noted between the observed tin fecal excretion rate of 3.74 μg Sn/day for the Apollo 8 astronaut and the normal dietary intake of 17,000 to 22,000 μg/day.[3, 4] Values of 5.3 and 1.9 μg Sn/man day were observed in the Apollo 7 and 9 missions respectively.[2] Almost all tin is excreted in the feces,[3] and although the function of this element in the body is not well known, the quantities of this element ingested by the astronauts should be investigated. It is quite pcssible that the reported normal dietary intakes are in error.

Fecal excretion rates of 38 and 44 μg Se/man day for Apollos 7 and 9[2] can be compared to the calculated rate of 35.3 μg Se/day for the Apollo 8 astronaut based on the data in Table V. Selenium is a biologically important element in mammals, and although little is known of its intake or retention, an excess may cause chronic poisoning.

An additional toxic element, mercury, has no known function in the body and is usually eliminated in the feces.[3] Although the average daily intake ranges from 10 to 20 μg/day,[3, 4] fecal excretion rates of 49 and 43 μg Hg/man day were observed for Apollo 7 and Apollo 9, which indicate a higher than normal intake of mercury. From the mercury concentrations listed in Table V, a fecal excretion rate of 168 μg Hg/day is calculated for the Apollo 8 astronaut, which is almost fourfold higher than the other missions and eight- to sixteenfold higher than a normal daily intake. The intake concentration which corresponds to this excretion rate is unknown.

Silver, antimony, and gold are also moderately toxic elements. Although silver seems to be essential to the life process, little is known about antimony and gold. A fecal excretion rate of 43.4 μg Ag/day was calculated for the Apollo 8 astronaut from the silver concentrations observed in the fecal specimens. This value is only slightly higher than the values of 32 and 22 μg Ag/man day calculated for the Apollo 7 and Apollo 9 mission astronauts respectively.[2] These values ere in reasonable agreement with a reported normal intake of 56 μg Ag/day, most of which is eliminated in the feces.[3]

The fecal excretion rates of 7.03 μg Sb/man day for the Apollo 7 mission and 8.45 μg Sb/man day for the Apollo 9 mission are comparable to the fecal excretion rate of 6.66 μg Sb/day calculated for the Apollo 8 mission astronaut.

The gold fecal excretion rate of 8.55 μg Au/day calculated for the Apollo 8 astronaut from the data in Table V is approximately sixfold higher than the 1.4 to 1.5 μg Au/man day observed for the Apollo 7 and 9 mission astronauts.[2] However, since there is a probability that the gold intake is related to each astronaut individually, such as from gold in teeth or rings, this high value may not be unusual.

In summary, the fecal excretion rates of most elements observed for the Apollo 8 mission astronaut are similar to those measured for the Apollo 7 and 9 mission astronauts. The only notable exceptions are mercury and gold, both of which are known to be moderately toxic. Large discrepancies between the excretion rates and normal dietary intakes are observed for the elements cobalt, iron, potassium, and tin. It is recommended that the elemental concentrations of the foodstuffs used during these missions be determined so that a complete material balance can be performed. In this way it should be possible to precisely establish the uptake or loss of elements by the astronauts.

TASK - GLASS FIBERS IN ASTRONAUT FECAL SAMPLES

During the course of the Apollo 7 mission, it was observed by the astronauts that significant amounts of glass fibers were present within the atmosphere of the spacecraft. In order to determine the amount and origin of the fibers ingested by the astronauts, the fecal samples which were collected and stored an board the spacecraft during the flight were analyzed for glass fiber content.

Aliquots of the five returned fecal samples from the Apollo 11 mission were also analyzed for glass fiber content according to a procedure described in an earlier report.[1] There were no identification marks on any of the collection bags; hence, the samples were arbitrarily numbered one through five. For sample 5 there were two clusters of 20 to 50 beta fibers still held together by the bonding agent. No attempt was made to distinguish between alpha and beta fibers in these determinations although a significant fraction of the fibers listed in the table were certainly alpha fibers. The only anomaly is sample 1, which has a twofold higher glass concentration and content than any previously observed specimen.

TASK - INDUCED RADIONUCLIDES IN SPACECRAFT

A piece of the outer "skin" from Apollo 12 (3.3868 g) was counted for induced radionuclides 19 days after splashdown of the mission. A ^{7}Be concentration of 2.88 ± 34.6% dis/min/g "skin" decay corrected to the day of splashdown was observed. The ^{7}Be can be produced by the cosmic-ray spallation of carbon, nitrogen, oxygen, aluminum, and silicon. The "skin" is a silicon monoxide-coated, aluminized Capton with a silicone base adhesive which was found by means of fast neutron activation analysis to be composed of 7.7% aluminum and 51% silicon by weight.[••] From information supplied by E. I. du Pont de Nemours & Co. concerning the composition of the plastic base (Capton), the skin contains 8.637 oxygen, 28.5% carbon, and 3.0% nitrogen by weight, with the balance being hydrogen.

The excitation functions for the production of ^{7}Be from the five possible elements were normalized to a typical shape cosmic particle spectrum. The weighted average cross section for energies exceeding 25 MeV were 11.4, 5.65, 6.53, 2.31, and 1.26 mb from carbon, nitrogen, oxygen, aluminum, and silicon respectively. Using these cross sections, the above chemical composition and mass of skin, and the above measured activity of ^{7}Be, a value of 1720 particles/cm^{2}/sec has been estimated for the incident cosmic-ray flux above 25 MeV. This value seems unreasonably high when compared to previously measured values of the cosmic flux of 130 protons cm^{-2} sec^{-1} above 40 MeV,[1] and 280 protons cm^{-2} sec^{-1} above 21 MeV and 320 protons cm^{-2} sec^{-1} above 16 MeV.[6] There are several possible sources for this discrepancy. The "typical" cosmic particle spectrum chosen for normalization of excitation functions may not be representative of the cosmic spectrum at the time of the Apollo 12 mission. The excitation functions used may have been inaccurate (accurate versions are presently being determined), and a small error in the shape of the function at low particle energies can result in a large error in the overall cross section. Finally, the measured ^{7}Be activity is quite uncertain (34.6% at one standard deviation). It is felt that a combination of these error sources is responsible for the seemingly high value for the particle flux estimated during the Apollo 12 mission.

EXPENDITURES

The following table documents the expenditures according to task and total costs incurred from October 6, 1969, through January 4, 1970, for the work reported herein.

TASK	EXPENDITURES
Determination of the Radionuclide Content of Feces and Urine From Astronauts Engaged in Space Flight	$ 3,218
Neutron Activation Analysis of Feces and Urine From Astronauts Engaged in Space Flight	7,326
Glass Fibers in Astronaut Fecal Samples	978
Induced Radionuclides in Spacecraft	322
Promethium-147 in Space Capsule Environment	250
TOTAL COSTS	$ 12,094

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RADIOACTIVITY IN FECES FROM APOLLO II ASTRONAUTS *

Sample #	dis/min/g on 7-24-69					
	^{7}Be	^{40}K	^{59}Fe	^{60}Co	^{137}Cs	^{147}Pm
1		7.70 ± 0.23	1.33 ± 0.24	0.0070 ± 0.0037	0.330 ± 0.031	< 0.038
2		8.59 ± 0.22		0.0067 ± 0.0036	0.196 ± 0.030	< 0.047
3	0.41 ± 0.33	4.45 ± 0.22				< 0.094
4		7.94 ± 0.38				< 0.33

The radioactivity has been normalized by dividing the disintegration rate by the appropriate fecal weight or urine volume and decay correcting to the splashdown date, 7-24-69.

TABLE II

RADIOACTIVITY IN URINE FROM APOLLO 11 ASTRONAUTS*

Astronaut	Flight Period	dis/min/ml on 7-24-69					
		^{7}Be	^{22}Na	^{40}K	^{59}Fe	^{60}Co	^{137}Cs
Aldrin	Pre	0.440 ± 0.091		2.52 ± 0.07			0.0137 ± 0.0087
Armstrong	Pre	0.208 ± 0.069		2.62 ± 0.05			0.0429 ± 0.0068
Collins	Pre	0.144 ± 0.071	0.00080 ± 0.00063	1.55 ± 0.05	0.067 ± 0.026		0.0601 ± 0.0069
Aldrin	Post	0.154 ± 0.074	0.0010 ± 0.0006	2.04 ± 0.05		0.0009 ± 0.0008	0.0072 ± 0.0068
Armstrong	Post		0.0014 ± 0.0007	0.768 ± 0.06			0.0196 ± 0.0077
Collins	Post			1.73 ± 0.05			0.0112 ± 0.0066

The radioactivity has been normalized by dividing the disintegration rate by the appropriate fecal weight or urine volume and decay correcting to the splashdown date, 7-24-69.

TABLE III

Na, K, Rb, AND Cs CONCENTRATIONS IN ASTRONAUT FECAL SAMPLES[π]

	Na		K		Rb		Cs	
Apollo 8	ppm*	g**	ppm*	g**	ppm*	mg**	ppm*	μg**
#1	8,760	1.63	3,690	0.688	15.4	2.87	0.0544	10.2
#2	10,500	0.895	10,300	0.878	9.15	0.783	0.0381	3.26
#3A +	5,780	1.15			14.2	2.83	0.0421	8.36
#3B +	5,380	1.07	7,480	1.49	13.4	2.67	0.0424	8.42
Apollo 10								
S/N 3512	10,700	0.817	9,030	0.689	17.8	1.36	0.0511	3.90
S/N 3513	2,440	0.0975	6,480	0.259	18.9	0.755	0.0491	1.97
S/N 3527	1 510	0.0619	6,140	0.251	20.6	0.842	0.0714	2.92

* Wet weight basis
** Total weight per defecation
\+ Different aliquots of the same sample
π The precision of all analyses is ± 10%

TABLE IV

Fe, Co, Zn, Cr, AND Sc CONCENTRATIONS IN ASTRONAUT FECAL SAMPLES $^{\pi}$

	Fe		Co		Zn		Cr		Sc	
Apollo 8	ppm*	mg**	ppm*	μg**	ppm*	mg**	ppm*	μg**	ppm*	μg**
#1	195	36.4	0.156	29.0	168	31.3	1.76	328	0.0177	3.31
#2	207	17.7	0.124	10.6	186	15.9	0.574	49.2	0.00782	0.669
#3A +	139	27.7	0.130	25.8	120	23.7	1.68	334	0.0138	2.73
#3B +	135	26.8	0.122	24.2	147	29.2	1.72	341	0.0133	2.65
Apollo 10										
S/N 3512	197	13 0	0.169	12.9	210	16.0	1.26	96.1	0 0124	0.945
S/N 35 13	268	[illegible] 7	0.193	7.71	282	11.3	1.00	40.1	0 0135	0.538
S/N 3527	491	20 1	0.382	15.6	412	16.8	1.59	65.0	0 0173	0.706

* Wet weight basis
** Total weight per defecation
\+ Different aliquots of the same sample
π The precision of all analyses is ± 10%

TABLE V

Br, Se, Hg, Ag, Sb, Au, AND Sn CONCENTRATIONS IN ASTRONAUT ECAL SAMPLES[++]

	Br		Se		Hg		Ag		Sb		Au		Sn	
	ppm*	µg**	ppm*	µg**	ppm*	µg**	ppm*	µg**	ppm*	µg**	ppm*	µg**	ppm*	µg**
APOLLO 8														
#1	2.74	511	0.544	102	2.10	392	0.973	181	0.102	18.9	0.156	29.2	0.0237	4.42
#2	4.01	343	0.344	29.4	0.494	42.3	0.139	11.9	0.0295	2.52	0.0107	0. 18	0.198	17.0
#3 A[+]	2.63	523	0.441	87.6	1.10	219	0.337	67.0	0.0989	19.6	0.0969	19.2	0.00735	1.46
#3 B[+]	1.68	334	0.414	82.3	1.06	210	0.394	78.3	0.0969	19.2	0.128	25.3	0.00735	1.46
APOLLO 10														
S/N 3512	3.37	257	0.487	37.1	0.861	≤3.7	0.462	35.3	0.119	9.11	0.00378	0.288	0.0323	2.46
S/N 3513	3.25	130	0.542	21.7	0.820	≤2.8	0.373	14.9	0.117	4.68	0.00307	0.123	0.0561	2.25
S/N 3527	4.25	174	1.16	47.4	0.787	≤2.2	0.188	7.70	0.104	4.26	0.00790	0.323	0.485	19.8

* Wet weight basis

** Total weight per defecation

[+] Different aliquots of the same sample

[++] The precision of all analyses is ± 10%

TABLE VI

GLASS FIBER CONCENTRATIONS IN APOLLO II ASTRONAUT FECAL SAMPLES

Sample	Aliquot (Grams)	Observed Beta	Fibers De	Defecation Weight (Grams)	Fibers/g Feces Beta	Fibers/g Feces De	Fibers/Defecation Beta	Fibers/Defecation De	µg Glass/ g Feces	µg Glass/ Defecation
1	3.6462	3753	23	208.1	1029	6.3	214,200	1300	7.516	1564
2	4 1720	556	1	230.6	133	0.2	30,700	60	0.948	219
3	3 9739	57	0	129.0	14	0	1,900	0	0.099	13
4	1 5196	270	1	35.1	178	1	6,240	20	1.30	45.5
5*	3 0544	733	16	10.0	240	5.2	2,400	52	1.90	19.0

Does not include two clusters of 20 to 50 beta fibers still held together with the bonding agent.

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