RETENTION AND EXCRETION OF STRONTIUM-90 BY A FORMER DIAL PAINTER (CASE 01-576)

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Excretion rates of ^{90}Sr by a former dial painter, Case 01-576, were determined on excreta collected in 1968. She excreted 249 and 64 pCi/day in urine and feces, respectively, or about 7% of the body content per year. Analysis of the data on this subject indicated that the power function described the retention and excretion rates better than did the single exponential.

Introduction

In order to assess the radiation levels and accompanying effects experienced by persons exposed to internally-deposited radionuclides, we have been studying the body content and excretion rates of these nuclides. While this effort has been concentrated mainly on the radium isotopes, ^{226}Ra and ^{228}Ra, ^{90}Sr is also of interest because of its chemical and metabolic similarities to radium and because of the large population exposed (low level exposure from fallout from nuclear test explosions and acquisition from industrial exposure). The use of luminous paint containing ^{90}Sr in the watch industry in the late 1950's led to sizeable accumulations of this nuclide in dial painters.[1-3] Apparently ^{90}Sr is still a hazard since Swiss paints of recent origin still contain ^{90}Sr.[2]

This nuclide is also of interest in that its metabolic parameters may lead to a better understanding of the metabolism of other bone-seeking elements, such as radium and the transuranic elements, and of long-term skeletal parameters, such as calcium and bone turnover rates.

Reported here is a study of a dial painter who had been studied extensively by Wenger and Soucas[1,2] in Geneva. We believe our report to be of interest because our radiochemical procedures and our analyses of the data differ from theirs.

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This subject, CHR Case 01-576 (Geneva Case GE 03), had been exposed to luminous dial paints containing both ^{90}Sr and ^{226}Ra. Data on her case history are presented in Table 1. The excreta samples for this study were collected from August 4, 1968 to October 31, 1968 at the MIT Clinical Research Center, and whole-body radioactivity measurements were made on her at the MIT Radioactivity Center during the same period.[4]

Experimental

The ^{90}Sr activity in the samples was determined by extraction of the 64.0-hr ^{90}Y daughter of ^{90}Sr into a liquid scintillator containing bis-(2-ethylhexyl) hydrogen phosphate (HDEHP). This extractant was then placed in a liquid scintillation counter which detected the beta rays emitted by ^{90}Y. This procedure is based on a similar one used by Williams,[5] which in turn has been adapted from that of Kauffman and Matuszek.[6] Details of the extraction procedure for urinary and fecal samples are given in the Appendix.

Counting Procedure

Four spiked samples were measured to determine extraction and counting efficiency. These samples were also used to determine the proper gain and window settings for the liquid scintillation counter.* They were spiked with 1.0 ml of a standard ^{90}Sr solution, which had 4440 dis/min/ml on February 22, 1968 (Amersham**). ^{90}Y was extracted as outlined in steps 6–14 of the Appendix. Maximum counting efficiency of the 2.26-MeV beta ray emitted by ^{90}Y was obtained at a gain control setting of 0.90%. A window setting of 200 to 800 represented the optimum condition of minimizing background counts while retaining a high counting efficiency. Because of the relatively low activity in the urine samples which

TABLE 1. Data on CHR Case 01-576 (GE03)[a]

Sex	Female
Year of birth	1930
Weight	56 kg
Height	160 cm
Exposure periods:	
^{90}Sr	May 56 – May 59 (1100 d)
^{226}Ra	Oct 46 – Oct 63 (6200 d)
^{226}Ra (1968) content	165 nCi

[a] Taken from Ref. 2.

* Packard 3002 Tri-Carb Scintillation Spectrometer.

** The Radiochemical Centre, Amersham, England.

were to be measured, a small sacrifice in counting efficiency because of the window setting was justified by a greatly reduced background count. The combined extraction and counting efficiency was 0.572 ± 0.009 counts/dis of ^{90}Sr (S.E., n = 4).

Corrections were made for the decay of the 64.0-hr ^{90}Y in the time between extraction and counting. The time was taken from the second of the three extractions to the midpoint of the count.

The amount of ^{90}Sr excreted per day at the time of collection, A^0, in each sample was calculated from the equation

$$A^0 = \frac{Ae^{\lambda t}}{2.22\ Efn} \text{ pCi/day},$$

where

A = activity of ^{90}Sr in the aliquot of sample on the day ^{90}Y is extracted,

E = counting efficiency,

2.22 = number of dis/min in 1 pCi,

f = fraction (aliquot) of total sample measured,

λ = decay constant of ^{90}Sr (0.02390 yr^{-1}, $T_{\frac{1}{2}}$ = 29.0 yr),

t = time (yr) from measurement of sample to the midpoint day of collection of the sample, and

n = length of collection period (days).

The analytical errors based on replicate measurements and calibration (systematic) errors were about 8% for the urine and 10% for the fecal samples.

Results

The results of our analyses are shown in Table 2 for eight urine and two fecal samples, each shown with its respective midtime of collection and the number of days of collection in each sample. The mean urinary excretion rate was 249 ± 66 pCi/d (S.E., n = 8) and the mean fecal excretion rate was 64 ± 13 pCi/d. For the year 1968, Wenger and Soucas reported 271 ± 16 pCi/d (n = 7) for urine and 93 ± 2.9 pCi/d (n = 2) for feces. The agreement between our values and theirs is quite good when we consider that the results were from two sets of samples collected at different times and places. As seen in Table 2,

TABLE 2. ^{90}Sr Urinary and Fecal Excretion Rates

Sample No.	Collection period, days	Midpoint of collection	^{90}Sr activity, pCi/day
Urine			
1	3	8/7/68	227
2	5	8/11/68	251
3	5	8/16/68	190
4	5	8/21/68	283
5	5	8/26/68	208
6	5	9/16/68	204
7	5	9/28/68	318
8	2	10/31/68	380
Mean ± S.E.			249 ± 66
Feces			
1	6	8/24/68	51
2	6	10/2/68	77
Mean ± S.E.			64 ± 13

the daily urinary excretion rate fluctuates and appears to rise significantly in the last two samples, which had been taken about a month after the others. This rise may be due to a seasonal variation. This variability is similar to that reported by Müller et al.[3] for 24-hr samples. The fecal-to-urinary ratio (F/U) of the mean values is about 0.25 (0.24 for each fecal value relative to the urinary value in the nearest time period). If correction is made for a dietary intake of about 10 pCi/day in the U.S. in 1968,[7] of which 15% is excreted in the urine and 85% in the feces, the excretion of the ^{90}Sr due to occupational exposure was 247 and 56 pCi/day in urine and feces, respectively. The value for F/U of 0.23 is within the wide range of ratios found by others (Table 3).

Retention and excretion parameters of ^{90}Sr metabolism for this subject may be estimated from the whole-body content and excreta data reported by Wenger and Soucas[2] (Table 4). In an earlier report their whole-body results were low in comparison to those from six other European laboratories.[8] Their value of 1.7 for the bremsstrahlung ratio (the ratio of bremsstrahlung production in the human to that in a ^{90}Sr standard source mounted in a Lucite holder) appeared to be high relative to the 1.45 used by other laboratories.

TABLE 3. Fecal-to-Urinary Ratios for Sr in Man

Typo of exposure	Ratio	Number of subjects	References
Dial painter	0.26	9	Müller and Thomas[9]
^{85}Sr injected	0.24	2	Harrison et al.[10]
^{85}Sr injected	0.17 – 0.50	10	Spencer et al.[7]
^{85}Sr injected	0.50	1	Fujita et al.[11]

TABLE 4. ^{90}Sr Body Content and Excretion Rates[a]

Year	Days since exposure (midexposure to midyear)	^{90}Sr burden, nCi	Dietary intake, pCi/day	Excretion rate, pCi/day	
				Urinary	Fecal
1962	1705	—	50	880	—
1963	2070	1561	100	672	—
1964	2436	1351	100	625	—
1965	2801	1259	72	443	—
1966	3166	1125	49	308	152
1967	3531	1210	25	308	102
1968	3897	1035	20	271	93
1969	4262	940	(15)-20	226	48
1970	4627	849	(15)-20	199	68
1971	4992	799	(15)-20	—	—

[a]From Ref. 4.

The coefficients of excretion (fraction of body burden excreted per unit time) in urine appeared to be high, viz., 15% of the body burden excreted annually, four years after her last industrial exposure to ^{90}Sr. This high coefficient may be due to their estimates of the body burden being too low.

The body burden values of Table 4 are shown in Table 5, modified by being normalized to the 1968 body content measurements made by Evans et al.[4]

TABLE 5. Modified ^{90}Sr Body Content and Excretion Rates[a]

Year	Time to end of exposure (to midyear), days	^{90}Sr burden, (normalized), nCi	^{90}Sr burden corrected for decay ($T_{\frac{1}{2}}$ = 29.0 yr), nCi	Urinary excretion, pCi/day	Coefficient of excretion (urine), % body content/yr	Fecal excretion, pCi/day	Coefficient of excretion (total), % body content/yr
1962	1157	—		942	—	—	—
1963	1522	2341	2586	726	10.2	—	—
1964	1888	2025	2291	690	11.0	—	—
1965	2253	1888	2188	501	8.4	—	—
1966	2618	1687	2002	356	6.5	131	8.0
1967	2983	1815	2206	370	6.1	98	7.7
1968	3349	1550	1930	334	6.3	95	8.1
1969	3714	1419	1809	284	5.7	43	6.6
1970	4079	1274	1663	257	5.6	71	7.1
1971	4444	1198	1602	—	—	—	—

[a]Body content data of Table 4 were normalized to Keane's measurement of 1550 nCi in 1968 and corrected for radioactive decay of ^{90}Sr to end of exposure in May 1959 ($T_{\frac{1}{2}}$ = 29.0 yr).

Urinary and fecal excretion rates (15% in urine and 85% in feces) were corrected for dietary intake (Table 4). These were then corrected for radioactive decay of ^{90}Sr.

and corrected for radioactive decay of the ^{90}Sr ($T_{\frac{1}{2}}$ = 29.0 yr). The excretion values of Table 4 are also given, compensated for dietary levels from Switzerland (to give the excretion rates of the endogenous ^{90}Sr) and corrected for radioactive decay. We assume that 15% of the dietary ^{90}Sr is excreted in the urine and 85% in the feces. Also given are the coefficients of excretion in units of % body content per year.

The ratios of fecal-to-urinary excretion rates from Table 5 are shown in Table 6. The mean value is 0.30, if the ratio from 1969, which appears to be very low, is neglected. We assume this ratio to have been constant over the measurement period so that the total excretion is then about 30% greater than the urinary excretion alone.

TABLE 6. Ratios of Fecal-to-Urinary Excretion Rates (F/U) from Table 5[a]

	Excretion, pCi/day		
Year	Urinary, (U)	Fecal, (F)	F/U
1966	356	131	0.368
1967	370	98	0.265
1968	334	95	0.284
1969	284	43	(0.151)[b]
1970	257	71	0.276
		Mean ± S.E.	0.298 ± 0.023

[a] See Ref. 2.

[b] Not used in calculation of mean

Discussion

These data may be used to estimate some of the metabolic parameters of ^{90}Sr in this subject, such as the consistency of changes in retention and excretion rates. The applicability of a model, such as the single exponential or the power function, which describes the ^{90}Sr retention, may then be inferred. It may also be possible to check the accuracy of the body content values which were based on bremsstrahlung measurements.

The parameters of the two simple models are shown in Table 7. The loss of ^{90}Sr from radioactive decay is shown in the retention equations, but since we compensate for this effect, it is not used subsequently. The power function model implies a single injection at t = 0. This assumption will produce little error at long times after exposure, and it does not affect the estimation of biological half-life, λ_b.

TABLE 7. Equations for Two Models Describing ^{90}Sr Retention and Excretion

	Single exponential[(a)]	Power function[(a)]
Retention	(1a) $R = R_0 e^{-\lambda_b t} e^{-\lambda_p t}$	(1b) $R = R_1 t^{-b} e^{-\lambda_p t}$
Excretion (radioactive decay neglected)	(2a) $dR/dt = -\lambda_b R_0 e^{-\lambda_b t}$	(2b) $dR/dt = -bR_1 t^{-b-1}$
Coefficient of excretion	(3a) $CE = \frac{1}{R} dR/dt = -\lambda_b$	(3b) $CE = \frac{1}{R} dR/dt = -b/t$
		(3c) $b = CE \cdot t$

(a) R = retention at time t

R_0 = retention at t = 0

R_1 = retention at t = 1 or 1 day

λ_b = biological elimination constant of ^{90}Sr or: $\lambda_b = \ln 2/T_B$, where T_B is the biological half life of ^{90}Sr

λ_p = physical decay constant ^{90}Sr

b = parameter describing loss of ^{90}Sr from the body

The consistencies among the various parameters may be used to choose the best model for these data. The data from Table 5 are shown in Fig. 1 in which the logarithms of retention and excretion values are plotted against time in order to illustrate the exponential model. The linear fits are quite good, as shown by the correlation coefficients, $r > 0.97$ (d.f. = 7, $p < 0.01$). The value for the point in parentheses was excluded from the calculations. The biological half-life of 4740 days for the retention confirms the effective half-life of 3210 days (biological half-life 4610 days) reported by Wenger and Soucas.[(2)] The excretion rates decrease with the much shorter biological half-lives of 1560 and 1790 days for urine and feces, respectively. This difference in half-lives between retention and excretion implies that the exponential model does not describe these data properly, as is seen from Table 7. The urinary data also appear to be better fitted by a curve than by a straight line. The poor fit of this model is also confirmed in the plot of coefficients of excretion (urinary and total) vs. time in Fig. 2, which shows the coefficients to decrease with time, rather than remain constant as predicted by this model.

The retention and excretion data are shown in Fig. 3 on logarithmic scales. The quality of these linear fits is comparable to that on the semilog scales ($r > 0.95$, $P < 0.01$). However, because in the power function model,

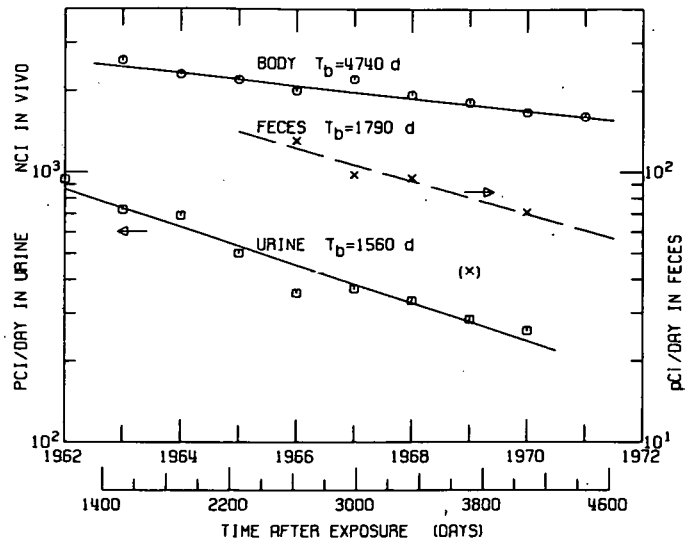
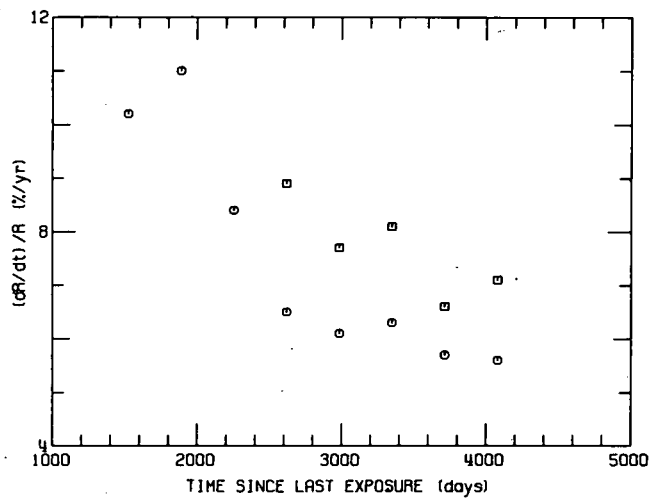


FIG. 1.--Semilog plot of ^{90}Sr body content and excretion rates as a function of time after exposure.

FIG. 2.--Coefficients of excretion of ^{90}Sr as a function of time after last exposure. Circles, urinary excretion; squares, total excretion.



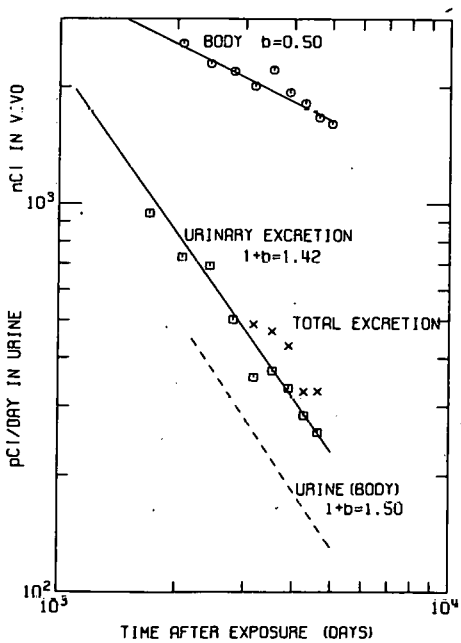


FIG. 3.--Logarithmic plot of ^{90}Sr body content and excretion rates vs. time from midpoint of exposure. Curve "urine (body)" is that derived from the retention curve, $dR/dt = -bR_1 t^{-b-1}$.

recent deposits would be losing activity more rapidly than older deposits, the midtime, rather than the end of the exposure, has been taken as t = 0. In this case the slope of the retention function, -b, is the exponent of the power function, and b = 0.50.

The slope of the urinary curve is -1.42 if the first two data points are omitted from the least squares calculation. From Eq. (2b) of Table 7 we then find b = 0.42. This value is consistent with the value of 0.50 from the retention data. Total excretion was not used directly because of the few fecal data and the large corrections used in arriving at a final value. Hence, use of the ratio appeared to be more appropriate. The retention equation then becomes

$$R = 1.2 \times 10^{5} t^{-0.50} ,$$

where R is the ^{90}Sr body content (in nCi) at a time, t days, after midtime of the exposure. The total excretion is then the derivative of R with respect to time [Eq. (2b), Table 7] and that for urinary excretion alone is $\frac{1}{1.3}$ of this value, namely

$$dR/dt = -4.6 \times 10^{7} t^{-1.50} \text{ pCi/day} .$$

A least squares fit of the urinary excretion data gives

$$dR/dt = -4.1 \times 10^{7} t^{-1.42} \text{ pCi/day.}$$

Thus, the exponents are essentially identical, as seen in Fig. 3. Curves "urine" and "urine (body)" are essentially parallel. However, the coefficients appear to differ significantly in that curve "urine (body)" derived from retention gives values of urinary excretion rates only about 60% of those measured, i.e., the "urine" curve. This difference is confirmed in the values of b derived from the coefficients of excretion [Eq. (3c), Table 7], which are fairly constant, but very large, at 0.83 ± 0.03 (S.E.). This effect may be due to one or more factors. The excretion rates may be too high, although this does not appear to be likely since our results agree well with those of Wenger and Soucas. The fecal-to-urinary excretion ratio may be in error (possibly 10% too high) because of few fecal data points, and the fecal-to-urinary ratio may have differed at earlier times. The body content measurements may be systematically low; they depend on a bremsstrahlung ratio, the estimation of which contains many

approximations, and they were measured in the presence of large amounts of ^{137}Cs and ^{226}Ra. This latter factor, however, cannot account for the large discrepancy in the values of b. Finally, this simple power function model may not be suitable for these data for a variety of reasons. For example, the uptake period was long relative to the study period, the uptake rate during exposure may have been very nonuniform, or the methods of analysis (especially choosing a starting time) may have been unsuitable. All of these factors need further study.

Preliminary calculations were also done with the more complex ICRP model of Marshall et al.[12] to check its fit to these data. The slope of the body retention curve was equivalent to that of the power function, about -0.57, when the ICRP values of the parameters were used. However, the predicted excretion curve [Eq. (15), page 16, Ref. 12] with a slope of -1.0 to -1.1 was significantly less negative than the slope -1.42 from the data (Fig. 3). The predicted coefficients of excretion did decrease with time after exposure, but they were lower and the rate of decrease was less than that of the observed values.

It should be noted that, given the power function model, the value of b should be derived from the retention curve, which is less sensitive to variations in the data than are the excretion curves. Then, given the value of b from the retention curve, the excretion curves may be used to derive the coefficient, R_1. This coefficient, in turn, may then be used to estimate a value of body content which is independent of bremsstrahlung ratios and the matching of calibration phantoms to the person being measured.

The values of the exponent b derived from the retention and excretion data are consistent with those of Müller et al.[3] (0.39) and of Müller and Thomas[9] (0.41), but they are greater than the 0.20 reported by Bishop et al.[13] and by Cohn et al.[14] for ^{85}Sr (0.16 to 0.36). However, if the interpretation of Marshall et al. (Ref. 12, p. 42) is used, the study times of less than 500 days for ^{85}Sr may have been too short to observe the long-term excretion parameters. The present study started 1100 to 1500 days after the last exposure.

In conclusion, it appears that the single exponential function, while it fits the data well in a mathematical sense, does not give parameters that are consistent with both whole-body retention and excretion rate parameters, namely a constant coefficient of excretion. The power function, on the other hand, fits the data well and gives more consistent parameters. The exponents are high compared to those for ^{85}Sr but are similar to those from other long-term ^{90}Sr studies of dial painters. While the value of b derived from whole-body retention is similar to that from excretion measurements, that derived from the coefficients of excretion is appreciably greater.

These data need further study to resolve the discrepancies between the parameters: comparison with those from other persons studied similarly (such as this subject's husband), comparison with ^{226}Ra parameters, and further analysis as to the suitability of the power-function model in describing retention for this type of exposure.

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APPENDIX. Detailed Procedure for the Separation and Determination of ^{90}Sr in Urine and Fecal Samples

Preparation of Extractant (Scintillator)

The scintillator solution is prepared by dissolving 5.0 g of PPO (2,5-diphenyloxazole) and 0.10 g of POPOP [1,4-bis-2-4(4-methyl-5-diphenyloxazolyl) benzene] in 500 to 700 ml of scintillation grade toluene. Then 50 ml of HDEHP are dissolved in the above scintillation solution and sufficient toluene is added to bring the volume to 1.0 liter.

The ^{90}Sr was determined by separation of its ^{90}Y daughter in the samples according to the following methods.

Extraction Procedure: Urine

1. The sample is first dry- or wet-ashed and dissolved in dilute nitric acid.
2. An aliquot (5 or 10 ml) of the sample to be measured is pipetted into a Teflon beaker.
3. 20 mg Sr (II) carrier are added.
4. To remove silica 10 – 15 ml 48% HF are added, and the beaker is heated to evaporate all liquid. 10 ml of HF are added twice more and evaporated to dryness and until fuming ceases.
5. 5 to 10 ml 12 N HCl are added and evaporated. Then another 60 ml of HCl are added. The beaker is heated to dissolve the solid material, and heating is continued until the volume is approximately 30 ml.
6. The pH is adjusted to 1.9 by the addition of 6 N NH_4OH.
7. The solution is transferred to a 50-ml centrifuge tube. If the volume is over 40 ml, a definite fraction of the solution is used.
8. 7 ml of the scintillation mixture, previously equilibrated with 0.01 N HCl, are pipetted into the tube and the organic and aqueous phases are contacted for 2 min using an electric stirrer.
9. The phases are separated by centrifuging for 2 min.
10. The organic (upper) layer is pipetted into a second centrifuge tube.
11. Steps 6–9 are repeated twice. The end of the stirring of the two phases for the second of the three extractions is taken as the time for the start of the decay of the extracted ^{90}Y.
12. The scintillation mixture in the tube is washed with 6 ml of 0.01 N HCl by vigorous stirring. Again the phases are separated by centrifuging.
13. The scintillation mixture is pipetted into a glass scintillation vial.
14. The vial is counted on a liquid scintillation counter.
15. Steps 6–13 are repeated on the same sample. The wash from step 11 is reused and then combined with the original sample. (This keeps the total volume at a minimum.) Although the activity found

in the vial containing the second set of extractions is usually negligible and is not used in calculating the activity in the sample, it serves both as an assurance that very nearly all the ^{90}Y has been extracted in the first 3 extractions and as a check against external contamination from glassware.

Modifications to Extraction Procedure: Feces

The presence of copper in the samples (from having been stored in copper cans) inhibited the extraction procedure because copper compounds (probably copper phosphates) precipitated at a low pH. However, two fecal samples were analyzed; one was extracted at a pH of 1.0, and the other at a pH of 1.9 after most of the copper was precipitated by addition of finely divided magnesium.