1
UNCLASSIFIED

UR-501

Health and Safety

THE UNIVERSITY OF ROCHESTER
Atomic Energy Project
P. O. Box 287, Station 3
Rochester 20, New York

* * *

Contract W-7401-eng-49 between the U. S. Atomic
Energy Commission and the University of Rochester,
administered by the Department of Radiation Biology
of the School of Medicine and Dentistry.

POLONIUM URINALYSIS

by

Carolyn A. Krebs
G. Hoyt Whipple

Division: Medical

Division Head: Joe W. Howland

Section: Health Physics

Section Head: G. Hoyt Whipple

Date Completed: 7/5/57

Date of Issue: 8/7/57

UNCLASSIFIED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

POLONIUM URINALYSIS

ABSTRACT

Po 7

A method is described for routine urinalysis for ~~polonium~~ by plating on copper at room temperature from acid solution. A discussion is presented based on experience gained by this laboratory in working with the method over a four-year period. (auth)

U

ACKNOWLEDGEMENTS

The authors are happy to acknowledge the work of Ruth Kennedy Scherberger who was chiefly responsible for the original adaptation of the Mound Laboratory procedure for use at the University of Rochester and who carried out the polonium urinalysis program here during its first years.

POLONIUM URINALYSIS

1. Introduction.

A program for the routine analysis of polonium in the urine of people potentially exposed to this material was started in January of 1953 at the University of Rochester, Atomic Energy Project. Since that time a number of minor modifications have been introduced, and a routine procedure has evolved which is satisfactory for detecting the accidental ingestion, inhalation, or incision of amounts of polonium in the range of the maximum permissible urinary excretion rate. This report describes the method now in use and summarizes our experience with it.

2. The Method.

The method described here is an adaptation of that in use at Mound Laboratory (1). The urine sample analyzed is the single specimen voided upon rising Monday morning. It is collected in a one quart waxed cardboard ice cream container and analyzed the same day. No preservative is added to the urine. However, if the sample is not to be analyzed until afternoon, it is refrigerated during the morning and then allowed to return to room temperature before analysis.

The volume of each urine sample is measured and recorded. If the volume is less than 200 ml, the sample is diluted to this volume with distilled water. If the sample contains more than 200 ml, the excess

is discarded. Each sample is divided equally between two waxed paper drinking cups. In addition to the 100 ml of urine, each cup receives 10 ml concentrated HCl to make the urine aliquot roughly 1 normal in acid. One cup from each pair also receives a known amount of polonium and the urine aliquot in this cup becomes known as the "spiked urine aliquot." Composition of the two aliquots derived from each urine sample is given in Table 1.

Table 1

Composition of the Aliquots Used in Polonium Urinalysis

<u>Designation</u>	<u>Volume of Urine</u>	<u>Volume of Conc. HCl</u>	<u>Volume of Po Spike Solution *</u>
unspiked urine aliquot	100 ml	10 ml	0
spiked urine aliquot	100 ml	10 ml	1 ml

* The polonium spike solution is made up to have an activity in the range of 10-20 dpm/ml. It represents a single dilution of a solution standardized by plating on silver at 90-95°C (3).

A copper disc, 1 inch in diameter and 0.01 inch thick, is prepared for each urine aliquot by immersion in the following series of baths:

Concentrated nitric acid,
 Distilled water,
 Concentrated nitric acid,
 Two distilled water baths,
 Concentrated hydrochloric acid,
 Two distilled water baths,
 Two 95% alcohol baths,
 Two anhydrous ether baths.

Each disc is put through this series and then immediately hung in its urine aliquot from a glass hook. Each cup is loosely covered with a cardboard disc. The contents of each cup are then stirred with a motor-driven glass propeller. Spontaneous displacement of polonium on the copper discs is allowed to proceed for two hours at room temperature. At the end of this time the copper discs are removed from the paper cups, rinsed in distilled water, patted dry with cleansing tissue, and placed in envelopes until counted.

Each side of each disc is counted for thirty minutes in an alpha scintillation counter. These counters consist of modified Keleket scalars, Models K-262 and K-263, and an RCA 5819 photomultiplier coupled optically by means of mineral oil to a lucite cup which carries a layer of zinc sulfide on its outer surface. The background counting rate for each counter is obtained daily from a thirty-minute count taken at the beginning of the working day. The background count usually falls in the range 0.1 to 0.4 cpm.

A counter is not used on any day that its background is higher than 0.4 cpm. The efficiency of each counter is checked daily by counting for one minute the alpha particles from an aged RaD+E standard calibrated by the National Bureau of Standards. The alpha particles are emitted by Radium F (Po-210) which is essentially in equilibrium with RaD+E after three years. The counter efficiency is:

$$E_c = \frac{\text{observed cpm from } \alpha \text{ standard}}{\text{certified dpm of same standard}}, \quad (1)$$

and for these counters is about 42%.

The recovery efficiency for the method is obtained from the relation:

$$E_r = \frac{1}{P} \cdot \frac{1}{E_c} \cdot \frac{S_a + S_b - 2B}{30} \quad (2)$$

where P = polonium activity added to the spiked urine aliquot in dpm.

E_c = the counter efficiency given by equation (1).

S_a = total count obtained in 30 minutes from one side of the spiked urine aliquot disc.

S_b = total count obtained in 30 minutes from the other side of the spiked urine aliquot disc.

B = counter background for a 30-minute period.

In this recovery efficiency, E_r , is determined for each individual urine sample. Fig. 1 is a plot of recovery efficiency vs. polonium concentration and shows that the recovery is 50 to 60% for concentrations of significance.

The activity present in the unspiked urine aliquot is:

$$\frac{1}{E_r} \cdot \frac{1}{E_c} \cdot \frac{C_a + C_b - 2B}{30} \text{ dpm in 100 ml,}$$

where C_a = total count obtained in 30 minutes from one side of the unspiked urine disc.

C_b = total count obtained in 30 minutes from the other side of the unspiked urine disc.

Then, on the assumption that the standard urine excretion is 1500 ml in a 24-hour period (10), the urinary excretion rate is:

$$U = \frac{15}{E_r E_c} \cdot \frac{C_a + C_b - 213}{30} \cdot \text{dpm in 24 hours.}$$

If the aliquot contained less than 100 ml of urine, i.e., if the sample submitted was less than 200 ml, an appropriate dilution factor is included in the formula.

A urine sample is considered positive if it is found to contain more than 0.2 cpm/100 ml, i.e., if:

$$\frac{C_a + C_b + 2B}{30} > 0.2 \text{ cpm.}$$

With this figure as the lower limit, the probability of calling a sample negative which contains the maximum permissible urinary excretion rate is 0.01. The probability of calling a sample positive which contains less than the maximum permissible urinary excretion rate is 0.13. Thus, the chance of obtaining a false positive is one in eight. This may be inconvenient, but is necessary in order to keep the probability of a false negative at one chance in a hundred.

3. Discussion.

It is proper to point out that an analytical method which is good for precise quantitative determinations may not be a good method for urinalysis and that the converse may also be true. High precision and even efficient recovery are not always essential to a satisfactory method of urinalysis.

This is true of urinalysis for polonium. Polonium will plate spontaneously on copper, silver, nickel, and nickel-containing alloys such as stainless steel and monel metal (2). Plating on silver gives higher recovery of polonium than plating on copper (about 95% vs. 65% recovery) (3). However, spontaneous plating of polonium on silver

occurs with this high recovery only at an elevated temperature. This necessitates more equipment and time, and the increased temperature precludes use of the disposable paper cups which have proved so convenient in the method reported here.

Plating of polonium on nickel has not been attempted in this laboratory. However, several samples were analyzed for polonium by plating at room temperature on stainless steel discs, and use of this material was found to give a recovery not significantly higher than that with copper.

General analytical technique calls for the establishment of recovery standards by analysis of samples containing known amounts of the material in question. Therefore, a solution of polonium must be available whose polonium content is accurately known. This solution should be relatively low in activity (of the order of 5,000 dpm/ml), so that it can be brought to the desired spiking level in one, or possibly two, dilutions. It is preferable that this solution be standardized by a method different from the one whose recovery it is to be used to check.

The standard polonium solution used in the work reported here is obtained from the Radioactive Inhalation Laboratory of this installation, where it is standardized by plating in duplicate on silver foils (3). We receive it containing about 3,000 dpm/ml and use it as a standard until the activity has decayed to about 800 dpm/ml.

All polonium solutions are made up to be approximately 1 N in HCl because it has been found that the tendency of polonium to plate out on the walls of the containing vessel is negligible at this hydrogen ion concentration.

Fig. 1 is the recovery curve for the urinalysis method discussed here, obtained by spiking acidified aliquots of pooled urine with varying amounts of polonium. A recovery curve established in this way is not entirely satisfactory since it assumes that polonium placed in urine by the kidney is free to act in the same way as polonium which is placed in urine as PoCl_3 at the time of analysis. The possibility is ignored that at least part of the polonium excreted by the kidney is bound in chemical complexes which inhibit its action.

If polonium urinalysis results are to have any value, a rate must be set which will correspond to the amount of polonium excreted in the urine per unit time when the body has reached its total permissible burden of polonium. This maximum permissible urinary excretion rate (MPUER) will necessarily be a crude estimate, for it must be obtained by several extrapolations of the accepted figures for maximum permissible exposure to external radiation.

When the polonium urinalysis program was inaugurated at this laboratory, the figure commonly held in the United States as the MPUER for polonium was 500 dpm per liter of urine (4), and at first we adopted this figure. However, the value of 0.02 μc for the maximum

Fig. 1. Polonium Urinalysis Recovery Curve

permissible body burden of polonium (5) used with the value of 0.1% excretion of the body burden per day (6) gives an MPUER for polonium of about 44 dpm per twenty-four hour sample of urine:

$$0.001 \times 0.02 \mu\text{c} \times 2.2 \times 10^6 \text{ dpm}/\mu\text{c} = 44 \text{ dpm}/24 \text{ hours.}$$

Recently the trend in laboratories in the United States has been toward this lower figure. For example, Oak Ridge National Laboratory is now using 66 dpm/24 hour sample as the MPUER for polonium (7), and Los Alamos Scientific Laboratory uses 44 dpm/24 hour sample (8). In accordance with this trend we are now using 50 dpm/24 hour sample as the MPUER for polonium (9). It is assumed that a 24-hour sample of urine contains 1500 ml (10).

4. Results.

Personnel potentially exposed to polonium at this laboratory are separated into four categories.

- I. Individuals who spend a considerable portion of their work week in rooms containing polonium, or who handle large amounts (millicuries) of polonium.
- II. Individuals who work sporadically in rooms containing polonium for short periods of time and who handle only small amounts (microcuries) of polonium.
- III. Individuals who work sporadically in rooms containing polonium for short periods of time but who do not usually handle radioactive materials.
- IV. Individuals who are infrequent visitors to rooms containing polonium.

Personnel in category I are sampled monthly. Members of categories II and III are sampled at three-month intervals. Individuals who come under category IV are sampled only if a film badge or pocket chamber reading is positive, or in the event that they are present when an accident occurs. In the case of an accident or incident all persons connected in any way are sampled. Members of categories I and II are sampled more frequently when a new technique is being initiated or when larger than normal amounts of polonium are being handled.

Table 2

Summary of Polonium Urinalyses for the Period

January 1 through December 31, 1956

Sample Activity				Number of samples	%
cpm/100 ml	dpm/24 hrs.	cpm/100 ml	dpm/24 hrs.		
---	--	0.2	15	86	93.4
0.2	15	0.5	20	2	2.2
0.4	20	0.8	30	2	2.2
0.7	30	1.0	50	2	2.2
1.0	50	---	--	0	0.0
total				92	100.0

5. Bibliography.

1. Spoerl, E. S. Urine assay procedure at the Mound Laboratory. Final Report. AECD-3811, April 1950 (declassified December 1955).
2. Robbins, M. C. The determination of polonium in urine. LA-1904, April 1955.
3. Smith, F. A., F. J. DellaRosa and L. J. Casarett. Analytical and autoradiographic methods for polonium-210. UR-305, November 1955.
4. Langham, W. H. Report on Harwell Conference on human monitoring in the control of occupational health hazards in atomic energy, September 8-9, 1954. WASH-490, October 1, 1954.
5. National Bureau of Standards Handbook 52. Maximum permissible amounts of radioisotopes in the human body and maximum permissible concentrations in air and water. March 1953.
6. Fink, R. M. (ed.). Biological Studies with Polonium, Radium and Plutonium. McGraw-Hill, New York, 1950.
7. Anderson, E. E. Private communication.
8. Meyer, D. D. Private communication.
9. Stannard, J. N. Private communication.
10. Schubert, J. Estimating radioelements in exposed individuals--II. Nucleonics 8: 66-78, March 1951.