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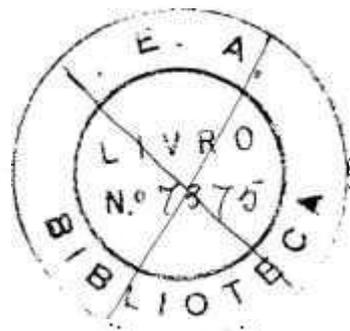
CIDADE UNIVERSITÁRIA "ARMANDO DE SALLES OLIVEIRA"

SÃO PAULO — BRASIL

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# DETERMINATION OF Sr-90 IN SÃO PAULO CITY FALLOUT<sup>(1)</sup>

A. Abrão and F. W. Lima

## PURPOSE

The purpose of this report is to register the extent of the contamination due to Sr-90 in São Paulo city.

It presents the results obtained by the Fallout Program carried out by the Radiochemistry Division of the Atomic Energy Institute (I.E.A.). The results of the Sr-90 precipitation in São Paulo are presented, covering a period beginning in June 1962 to February 1965. Although they were not determined quantitatively, other fission products were detected.

## SITES OF COLLECTION

At the beginning, two rainwater collection stations were installed at the swimming-pool reactor area, at the I.E.A., in the campus of the University City. They collected rainwater from June 1962 to August 1963. The first station was set up very close to the reactor building and the other one in front of the Administration building, having a distance of about 150 meters between both. In August 1963, one of these stations was transferred to the Astronomic Observatory of São Paulo, 20 kilometers away from the I.E.A..

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## RAINWATER COLLECTION

The rainwater at these sites is collected into polyethylene tanks of 220 liters capacity from an exposed area of 10.4 square meters (3.6 meters diameter) of sail-cloth (see Figure 1).

## Sr-90 CONCENTRATION

At the bottom of each polyethylene reservoir a column of cationic resin is attached and the rainwater is percolated through this column. The rainfall collections are made individually or combined in order to obtain a total volume of 200-600 liters per column of resin. The column is then transferred to the laboratory and processed.

## OTHER FISSION RADIOISOTOPES

Some of the rainwater samples were processed and analysed to identify other fission products. The fission product radioisotopes Zr-95, Nb-95, Ru-103, Ru-106 and Ce-144 were detected. These radionuclides were not determined quantitatively.

## RESULTS

The results are presented in Table 1 where only Sr-90 is reported; it was measured through its Y-90 daughter after radiochemical equilibrium had been attained. The results are expressed as pCi/l (picocuries per liter), mCi/km<sup>2</sup> (millicuries per square kilometer) and mCi/mi<sup>2</sup> (millicuries per square mile). The rainwater collections were controlled by the volume of water collected and were individual, covering a certain period. As it was mentioned, although other fission products were detected, they

were not measured quantitatively. The relation between Sr-89/Sr-90 was not measured either.

The Sr-90 specific activity of each rainwater collection was calculated using the relation: <sup>(1)</sup>

$$\text{specific activity (pCi/l)} = \frac{\text{Precipitation (mCi/km}^2\text{)}}{\text{Rainfall (cm)}} \times 100$$

### EXPERIMENTAL

Apparatus - Polyethylene tank of 220 liters capacity, having a cation resin column attached at the exit (photo).

Resin column - A column (glass) of 4 cm I.D. was set up containing 250 ml of cationic resin (Nalcite HCR, H-form, 30-50 mesh).

Filter - As the rainwater presents insoluble material (dust) and colloids (oil, etc), difficulties arose during the percolation. When the first column had the resin bed over a wad of glass wool, the flow rate gradually decreased due to obstruction. This was avoided by using a plastic tube having a thin nylon screen adapted by means of a rubber ring. This filter was placed at the bottom exit of the column (Figure 2).

The flow rate in the column was kept between 5-6 liters/hour.

The amount of resin required was previously determined by experiment with rainwater using one column of mixed bed resin (H-form and OH-form) and measuring the conductivity of the effluent.

Distilling apparatus - A glass distilling apparatus of 2 liters capacity was set up to distill at low pressure, and

provided with a heating mantle and a collector to retain the distilled HCl consisting of two filtering flasks connected in series and containing 500 and 200 ml of 8M NaOH, respectively.

Counting equipment - Throughout the work the following counting equipment was used: a Tracerlab counter with mica window Geiger tube of  $1.8 \text{ mg/cm}^2$ , with efficiency of  $0.20 \pm 0.03$  to Y-90 beta radiation; a Nuclear Chicago scaler model 181A with mica window Geiger tube of  $3.5 \text{ mg/cm}^2$  and  $0.13 \pm 0.03$  efficiency to Y-90 beta radiation; a Nuclear Chicago single channel pulse height analyser; a well type NaI scintillation crystal and a low background counting system, Nuclear Chicago, model C-110B.

## PROCEDURE

### 1. Elution of the cationic resin

1. Transfer the cationic resin to one liter beaker and wash with deionized water to remove suspended solids, stirring with a glass rod.
2. Transfer the resin to the same column and elute with 2 liters of 4M HCl at a flow rate of 200 ml/h.
3. Transfer the eluate (1 liter each time) to the distilling apparatus and distill until about 50 ml of solution in the distilling flask remains.
4. Transfer the residual solution to a 300 ml Kjeldahl flask, washing the distilling flask with small portions of  $\text{HNO}_3$ . Evaporate to dryness.
5. Add 5 ml of  $\text{H}_2\text{O}_2$  (130 vol.) and 20 ml of  $\text{HNO}_3$  and evaporate dryness again in order to destroy organic matter. Repeat if necessary.
6. Dissolve the residue with 100 ml of hot water and some drops of  $\text{HNO}_3$ . If a small residue results, filter it and wash with hot water.

7. Treat the residue with small excess of hot  $\text{Na}_2\text{CO}_3$  solution and filter. Discard the filtrate.
8. Dissolve the carbonate precipitate with 0.1M HCl and combine it with the filtrate of step 6.

## II. Strontium Precipitation

9. Add 50 mg of Sr carrier to this solution, warm it ( $80^\circ \text{C}$ ) and bubble  $\text{NH}_3$  gas slowly until the pH is 7-8 (pH paper). Decant the precipitate in a warm bath and filter. Wash the precipitate with dilute  $\text{NH}_4\text{OH}$  (1:400). This precipitate is named Fe-1 and contains iron, lead and manganese.
10. Acidify the filtrate until pH 2, add 5 mg of  $\text{Fe}^{*3}$  carrier and precipitate with  $\text{NH}_3$  as in step 9. This precipitate is named Fe-2.
11. Acidify the filtrate and repeat step 10, obtaining Fe-3.
12. Warm the Fe-3 filtrate containing the strontium, add 20 ml of 2M  $(\text{NH}_4)_2\text{CO}_3$  to precipitate Sr and Ca. Decant the precipitate keeping the beaker warm for 30 minutes. Filter and wash the precipitate with dilute  $\text{NH}_4\text{OH}$  (1:400). Concentrate the filtrate by evaporation to certify that the Sr was completely precipitated.
13. Dissolve the  $\text{SrCO}_3$  precipitate (in the same beaker where it was precipitated) with 3M HCl. After dissolution dilute with water to about 15 ml. Add 6 mg of yttrium carrier and store the solution for radiochemical equilibration, observing a minimum of 17 days.

## III. Yttrium Separation

14. Transfer the solution containing Sr and Y (step 13) to a 50 ml centrifuge tube.

15. Warm the tube in a steam bath ( $80^{\circ}$  C) and precipitate with 5 ml of concentrated  $\text{NH}_4\text{OH}$ . Decant keeping the mixture warm during 5 minutes and centrifuge. Discard the supernatant.
16. Wash the precipitate twice with 10 ml of dilute  $\text{NH}_4\text{OH}$  (1:400), centrifuge and discard the supernatant.
17. Dissolve the precipitate with 5-10 ml of 3M HCl, dilute with water to 10-15 ml, add 2-3 drops of thymol blue. Add 5M  $\text{NH}_4\text{OH}$  until a yellow-pink color. Precipitate with 10 ml of saturated oxalic acid, stirring with a glass rod. Raise the pH to a yellow-pink end point.
18. Keep the yttrium oxalate precipitate in the refrigerator for approximately 4 hours.
19. Centrifuge, discard the supernatant and add 10 ml of hot water to the precipitate and filter through a Whatman 42 paper on a sintered glass plate using vacuum. Filter again if necessary, using the same paper. Wash the precipitate with 10 ml of hot water and then 20 ml of alcohol and 20 ml of ether in small portions.
20. Mount the paper and precipitate on a ring and disc assembly for beta counting and follow its half-life.

Treatment of the Fe-precipitate - To identify other fission radioisotopes, the Fe-III hydroxide (Fe-1, step 9), which practically contains the whole activity except for Sr-90, Sr-89 and Cs-137, is processed. Other precipitates of ferric hydroxides (Fe-2, Fe-3) are discarded.

1. Dissolve the Fe-1 precipitate with hot 5M  $\text{HNO}_3$  and dilute to 40 ml with water. Filter if necessary.
2. Add 10 mg of europium carrier, adjust the pH to 1,

warm (30° C) and precipitate with 15 ml of saturated oxalic acid, stirring with a glass rod until the precipitation begins.

3. Keep the precipitate in the refrigerator for 2-3 hours and filter. Wash with 0.5% oxalic acid solution.
4. Evaporate the filtrate and destroy the oxalic acid with aqua regia. Transfer the solution to a vial and determine the gamma-ray spectrum.
5. Transfer the europium oxalate precipitate to a vial and determine its gamma-ray spectrum. This precipitate contains the rare earths, mainly Ce-141 and Ce-144.

#### Yttrium-90

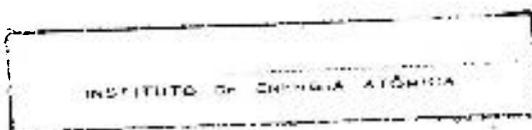
The Y-90 activity is determined by extrapolating the decay straight line to  $t_0$ , i.e., the time of its separation from Sr-90 (step 15), and compared with the activity of Y-90 separated from a Sr-90 solution of known disintegration rate, using the same procedure applied to the analysis.

#### Purity

The half-life of the separated Y-90 was followed for various periods, and shown to be radiochemically pure.

#### Chemical Yield

The chemical yield of the retention and elution of Sr-90 in the cationic resin column was determined and it was concluded that its elution is complete. It was observed that in each one of the rainwater collection experiments, a small residual activity remains in the resin after its elution with hydrochloric



acid. This activity was identified after thoroughly ashing the resin and following chemical treatment as being Zr-95, Nb-95 and Ru-100, Ru-106.

The chemical yield for Sr-90 throughout the operations of loading the cationic resin column, hydrochloric acid elution, acid distillations and strontium carbonate precipitation is 96.4% ± 2.1%.

The chemical yield for the yttrium separation is 99.7% ± 1.5%.

The total chemical yield, including the strontium separation in the mixture adsorbed by the resin and yttrium separation in equilibrium with radiocesium is 95.4% ± 2.1%.

#### REFERENCE

- (1) Health and Safety Laboratory, HASL-144, April 1, 1964.

#### Part III - Interpretive Reports and Notes

"Worldwide Deposition of Sr-90 Through 1964" by H.L. Volchok, Health and Safety Laboratory, USAEC.

"Estimated Worldwide Deposition of Sr-90 in 1965" by H.L. Volchok, Health and Safety Laboratory, USAEC.

"Sr-90 Fallout in Antarctica" by H.L. Volchok, Health and Safety Laboratory, USAEC.

"Fallout from the Second Chinese Nuclear Test and Comparison with the First" by P.W. Krey and W.C. Rosa, Jr., Health and Safety Laboratory, USAEC.

"Sr-90 in Human Vertebrae - 1964 - Results" by J. Rivers, Health and Safety Laboratory, USAEC.

| Date of Collection     | Vol (l) | pd <sub>1</sub> /3 | cd <sub>1</sub> /cm <sup>2</sup> | sd <sub>1</sub> /eq<br>lit | mm  | inches | Site of<br>Collection |
|------------------------|---------|--------------------|----------------------------------|----------------------------|-----|--------|-----------------------|
| <u>1962</u>            |         |                    |                                  |                            |     |        |                       |
| June 1 to Sept. 1      | 475     | 0.09               | 0.004                            | 0.010                      | 48  | 1.89   | R                     |
| Sept. 8 Sept. 9        | 200     | 0.09               | 0.002                            | 0.005                      | 20  | 0.79   | R                     |
| Sept. 20 Oct. 19       | 595     | 0.18               | 0.010                            | 0.026                      | 56  | 2.20   | R                     |
| Oct. 24 Nov. 11        | 445     | 0.59               | 0.027                            | 0.069                      | 45  | 1.77   | A                     |
| Oct. 24 Nov. 11        | 450     | 0.23               | 0.010                            | 0.026                      | 45  | 1.77   | R                     |
| Nov. 20 Dec. 6         | 380     | 1.19               | 0.045                            | 0.116                      | 38  | 1.50   | A                     |
| Nov. 20 Dec. 6         | 380     | 0.87               | 0.033                            | 0.086                      | 38  | 1.50   | R                     |
| Dec. 11 Dec. 24        | 430     | 0.40               | 0.017                            | 0.044                      | 43  | 1.69   | A                     |
| Dec. 11 Dec. 24        | 440     | 0.42               | 0.018                            | 0.046                      | 44  | 1.73   | R                     |
| <u>1963</u>            |         |                    |                                  |                            |     |        |                       |
| Jan. 8 to Jan. 26      | 600     | 0.35               | 0.024                            | 0.062                      | 60  | 2.36   | A                     |
| Feb. 20 Mar. 25        | 660     | 0.21               | 0.014                            | 0.036                      | 66  | 2.60   | R                     |
| Mar. 26 April 16       | 435     | 0.11               | 0.005                            | 0.013                      | 44  | 1.73   | A + R                 |
| May 16 Aug 17          | 370     | 0.11               | 0.004                            | 0.010                      | 37  | 1.46   | R                     |
| May 16 July 22         | 370     | 0.18               | 0.007                            | 0.018                      | 37  | 1.46   | O                     |
| Sept. 28 Oct. 17       | 390     | 1.57               | 0.061                            | 0.158                      | 39  | 1.54   | R                     |
| Aug. 16 Oct. 10        | 220     | 3.48               | 0.077                            | 0.199                      | 22  | 0.87   | O                     |
| Oct. 21 Nov. 27        | 570     | 0.56               | 0.032                            | 0.083                      | 57  | 2.24   | R                     |
| Oct. 15 Nov. 10        | 660     | 0.50               | 0.033                            | 0.085                      | 66  | 2.60   | O                     |
| Nov. 20 Dec. 5         | 330     | 0.75               | 0.025                            | 0.065                      | 33  | 1.30   | R                     |
| <u>1964</u>            |         |                    |                                  |                            |     |        |                       |
| Nov. 13(63) to Jan. 27 | 660     | 1.31               | 0.086                            | 0.221                      | 66  | 2.60   | O                     |
| Jan. 1 Feb. 12         | 610     | 0.20               | 0.012                            | 0.031                      | 61  | 2.40   | R                     |
| Feb. 3 Feb. 14         | 440     | 0.41               | 0.018                            | 0.047                      | 44  | 1.73   | O                     |
| Feb. 17 Mar. 23        | 555     | 0.14               | 0.008                            | 0.021                      | 56  | 2.20   | R                     |
| Feb. 17 April 31       | 440     | 0.50               | 0.022                            | 0.057                      | 44  | 1.73   | O                     |
| April 18 Jun. 2        | 480     | 0.16               | 0.008                            | 0.021                      | 48  | 1.89   | R                     |
| Jun. 10 July 29        | 475     | 0.47               | 0.046                            | 0.119                      | 48  | 1.89   | R                     |
| July 29 Aug. 28        | 280     | 0.60               | 0.011                            | 0.029                      | 19  | 0.75   | R                     |
| July 10 Aug. 31        | 570     | 0.98               | 0.055                            | 0.142                      | 57  | 2.24   | O                     |
| Aug. 31 Sept. 23       | 220     | 1.33               | 0.056                            | 0.145                      | 42  | 1.65   | O                     |
| Sept. 24 Oct. 1        | 190     | 0.94               | 0.050                            | 0.130                      | 53  | 2.09   | O                     |
| Aug. 22 Sept. 22       | 225     | 1.67               | 0.054                            | 0.140                      | 33  | 1.28   | R                     |
| Sept. 23 Sept. 29      | 215     | 1.46               | 0.061                            | 0.158                      | 42  | 1.63   | R                     |
| Sept. 30 Oct. 5        | 200     | 0.68               | 0.027                            | 0.070                      | 25  | 0.98   | R                     |
| Sept. 30 Oct. 10       | 160     | 0.72               | 0.019                            | 0.049                      | 31  | 1.22   | O                     |
| Oct. 6 Nov. 3          | 400     | 1.44               | 0.121                            | 0.313                      | 84  | 3.31   | R                     |
| Oct. 11 Oct. 26        | 175     | 2.02               | 0.084                            | 0.218                      | 32  | 1.26   | O                     |
| Nov. 1 Nov. 10         | 165     | 2.99               | 0.070                            | 0.181                      | 23  | 0.91   | R                     |
| Nov. 3 Nov. 9          | 200     | 2.27               | 0.073                            | 0.189                      | 32  | 1.26   | O                     |
| Nov. 10 Nov. 20        | 215     | 2.48               | 0.082                            | 0.212                      | 33  | 1.30   | R                     |
| Nov. 12 Nov. 18        | 155     | 3.84               | 0.088                            | 0.228                      | 23  | 0.91   | O                     |
| Nov. 20 Nov. 23        | 195     | 1.75               | 0.075                            | 0.194                      | 42  | 1.65   | R                     |
| Nov. 30 Dec. 1         | 155     | LOST -             |                                  |                            | 21  | 0.83   | R                     |
| Nov. 19 Dec. 10        | 145     | 3.25               | 0.169                            | 0.438                      | 52  | 2.05   | O                     |
| Dec. 13 Dec. 15        | 165     | LOST -             |                                  |                            | 23  | 0.91   | R                     |
| Dec. 15 Dec. 31        | 145     | 3.97               | 0.143                            | 0.370                      | 36  | 1.42   | O                     |
| Dec. 16 Dec. 21        | 200     | 0.84               | 0.037                            | 0.096                      | 44  | 1.73   | R                     |
| Dec. 22-Jan. 5 (65)    | 210     | 1.89               | 0.250                            | 0.627                      | 105 | 7.28   | R                     |
| <u>1965</u>            |         |                    |                                  |                            |     |        |                       |
| Jan. 4 to Jan. 11      | 210     | 1.20               | 0.053                            | 0.137                      | 44  | 1.73   | O*                    |
| Jan. 6 Feb. 3          | 560     | 1.42               | 0.303                            | 0.785                      | 214 | 8.43   | P                     |

\* OBS: R = Reactor AREA

A = Administration Building

O = Astronomic Observatory

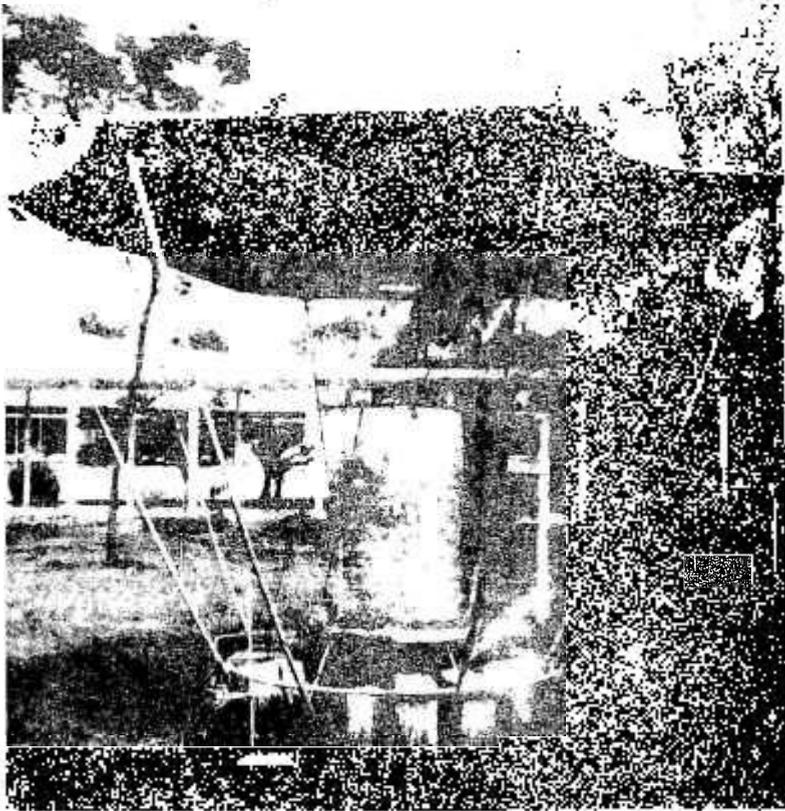


FIGURE 1

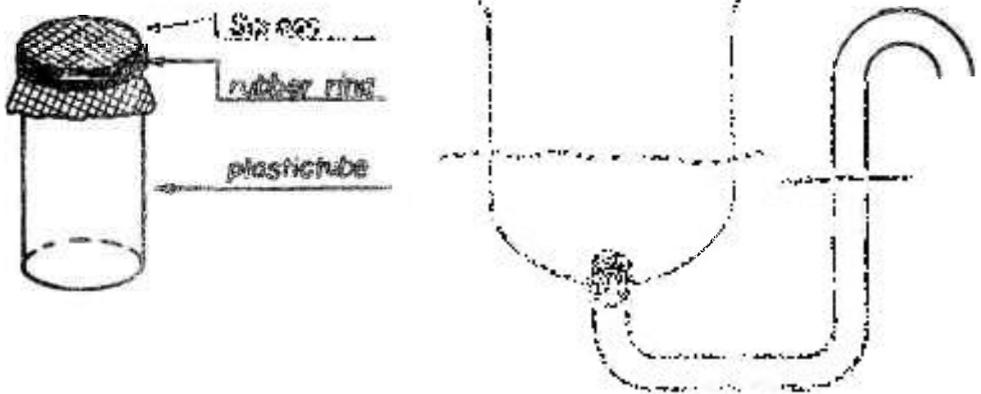


FIGURE 2