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Investigation of the radiation risk due to environmental contamination by ^{241}Am from lightning rods disposed at uncontrolled garbage dumps

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Abstract Radioactive lightning rods were manufactured in Brazil until 1989, when the licenses for using radioactive sources in these products were lifted by the national nuclear authority. Since then, these rods have been replaced by the Franklin type and collected as radioactive waste. However, only 20% of the estimated total number of installed rods has been delivered to the Brazilian Nuclear Commission. This situation causes concern, since there is the possibility of the rods to be disposed as domestic waste. In Brazil, 64% of the municipal solid waste is disposed at garbage dumps without sufficient control. In addition, ^{241}Am, the radionuclide most commonly employed, is classified as a high-toxicity element, when incorporated. In the present study, ^{241}Am migration experiments were performed by means of a lysimeter system, in order to evaluate the risk of contamination caused by radioactive lightning rods disposed as common solid waste. ^{241}Am sources removed from lightning rods were placed inside lysimeters filled with organic waste that was collected at the restaurant of the Instituto de Pesquisas Energéticas e Nucleares. The generated leachate was periodically analyzed, and characteristics such as pH, redox potential, solid content and the concentration of the radioactive material were determined. The equivalent dose for members of the public was calculated considering ingestion of contaminated drinking water as the major path of exposure. Estimated doses were about 20-times below the effective dose limit of 1 mSv year^{-1} for members of the public as recommended by the International Commission on Radiological Protection. This suggests the radiation risk caused by lightning rods disposed at uncontrolled garbage dumps to be low. It should be noted, however, that the number of investigated lightning rods was quite small. The results of this study might therefore not be entirely representative and should be interpreted with care. They provide, however, a very first basis for characterizing the transfer of ^{241}Am from lightning rods to the human food chain.

Introduction

Radioactive lightning rods were manufactured in Brazil until 1989, when the licenses for using radioactive sources in these products were withdrawn by the national nuclear authority. Since then, radioactive rods have been replaced by those of the Franklin-type, and collected as radioactive waste.

Based on data obtained on americium sources imported between 1970 and 1986 the Brazilian Nuclear Commission, CNEN, estimated that about 75,000 lightning rods had been installed in Brazil, with an average individual activity of 55.5 MBq [1]. Since 1989, only 20% of the estimated total number of rods installed has been delivered to the CNEN. This situation causes some concern as some of the remaining rods might well be discarded as domestic waste, and because in Brazil about 64% of the municipal solid waste is disposed at garbage dumps which are uncontrolled or only minimally controlled. In addition, when incorporated, americium is classified as a high-radiotoxicity element and the critical organs are kidneys, lungs and bones.

In the present study, ^{241}Am migration experiments using a lysimeter system were performed, to evaluate the risk of

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contamination caused by radioactive lightning rods disposed as solid waste. ^{241}Am sources removed from lightning rods were placed inside lysimeters filled with organic waste collected at the restaurant of the Instituto de Pesquisas Energéticas e Nucleares (São Paulo, Brazil). The leachate was periodically analyzed and parameters such as pH, redox potential, solid content and concentration of radioactive material were measured.

Materials and methods

Leaching tests and lysimeter studies

Leaching tests with radioactive sources and acid solutions, and dynamic tests with lysimeters filled with organic waste and radioactive sources were performed.

In order to evaluate the properties of some of the investigated sources, leaching experiments were carried out based on an US Environmental Protection Agency procedure [2]. In addition to the pH value of 5.0 of the acetic acid solution recommended, the same solution at pH 4.0, a sulfuric–nitric acid mixture at pH 4.5, and distilled water were also used, since these are believed to reflect the environment conditions: In São Paulo City, sulfuric and nitric acids are responsible for the acidity of the rain, and pH values below 4.0 were observed until 1990. Nowadays, pH values are higher than 4.5. This is due to the enforced emission control program for automobile vehicles, reducing their SO_2 emissions [3]. Different sources, removed from lightning rods, were used for these experiments, being three replicate experiments for acetic and distilled water and six replicate for sulfuric–nitric acid mixture. After a 24-hour leaching period under agitation, the sources were removed and the solution analyzed with a Ge detector.

For the dynamic tests, three replicate small-scale lysimeters, Lis 5, 6 and 8, and one control, Lis7, were used. The lysimeters consisted of lucite tubes with a diameter of 10 cm and a height of 50 cm. At the bottom, 1.5 cm tubes and valves for leachate collection were installed. (Fig. 1). Each tube received 2 cm of intermediate gravel as a drainage layer. The organic waste was carefully transferred to the lysimeters, so that no void spaces were left. Three ^{241}Am sources were placed in the middle of each tube, except for the control, and then covered with the remaining waste. Thermocouples were also positioned at the center. The upper waste surface was covered with medium sand (grain size 0.3–1.2 mm), to allow good distribution of the leachant. After loading, the lysimeters were capped and sealed with silicon glue. Polyethylene tubes, with their outlet immersed in water, were fitted on the top of each lysimeter, in order to prevent a build-up of internal gas
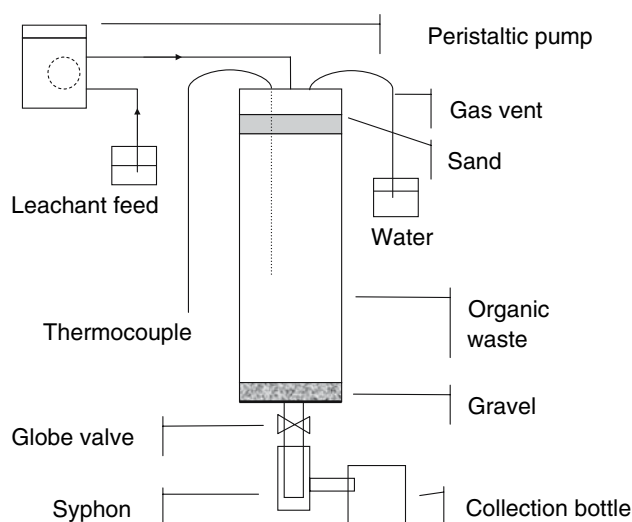
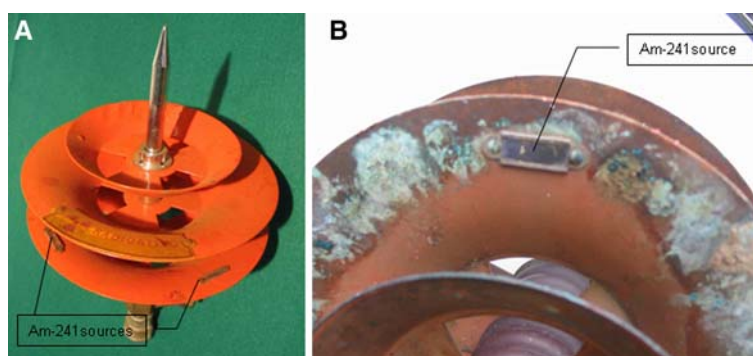**Fig. 1** Sketch of the experimental design

pressure. Thermal isolation was achieved by surrounding the lysimeters with expanded polystyrene strips.

As a leachant, distilled water was chosen. Because the organic waste used (see below) is a complex system, we decide not to use nitric or sulfuric solutions, because it would not be clear which agent would be responsible for any release observed. The irrigation rate was based on the pluviometry data of São Paulo City. According to the climatogram of the National Institute of Space Research [4], the average precipitation is 1,451 mm year^{-1} and the temperature 19°C. Considering the surface area of the lysimeters, the required volume of leachant to be added was 30 ml per day. Leachate samples were periodically collected to determine pH, redox potential, total dissolved solids, and ^{241}Am concentration. Visual aspects such as color and turbidity were also protocolled. Redox potential was determined with a combined Pt ORP electrode/reference electrode and a data-logger. This electrode was calibrated in Zobell's standard solution, every five samples analysed. ^{241}Am activity was determined by direct measurement with a Ge detector, and the results are expressed in becquerels per milliliter (Bq/ml). All lysimeters were operated for 21 months.

Radioactive sources

The ^{241}Am sources placed in the lysimeters were similar to those used in the leaching experiments, and were also removed from used lightning rods. They consisted of a metal support with a fixed layer of AmO_2 and a gold covering. The initial inventories of ^{241}Am in each lysimeter were 19.1, 18.0 and 19.2 MBq for Lis5, Lis6 and Lis8, respectively. Figure 2 shows ^{241}Am sources fixed onto the

Fig. 2 Americium sources fixed onto lighting rods' plates. **a** new device, **b** weathered device

lightning rods' plates. The sources were not identical since they were removed from used lightning rods taken at different regions of the country and exposed to different environmental conditions. They thus represent realistic surface conditions. However, it might well be that individual sources could show higher leaching rates, depending on the degree of damage of the gold covering.

Organic waste

Organic waste samples were collected from the restaurant in plastic bags, separating the processed from non-processed food. Fruit pulps and vegetables were chopped before being mixed with processed food. This waste stayed untouched for 5 days, to allow the release of gases. The amount of carbohydrates was controlled, to prevent blockage and excess of gas release. Organic material was used as garbage, as it is generally known that waste degradation is controlled mainly by microorganisms in organic matter.

Results and discussion

Results from the leaching experiments performed with distilled water, acetic and sulfuric–nitric acid mixture acid solutions are shown in Table 1. In all cases, ^{241}Am was detected in the solution, indicating that the sources were not sealed. These results may be attributed to the deterioration of the gold covering, caused by atmospheric agents or by the release of the material from the strip surface, exposed during the manufacturing of the lightning rods [5].

Lysimeter leachate pH, redox and solid content profiles are shown in Figs. 3–5, respectively. Each point represents average over a period of 30 days. The pH value increased for 8 months in all lysimeters, reaching a maximum of around 7.5. Samples collected from Lis7 (control) showed a faster increase (Fig. 3).

Figure 4 shows that reducing conditions were achieved after 2 months in all lysimeters, and a gradual increase of the redox is observed after 7 months. Lower redox values coincided with the higher average temperatures observed in this period.

Table 1 Results from the 24-hour leaching experiments performed with distilled water, acetic and sulfuric-nitric acid mixture

Solution	Source activity (Bq)	Released activity (Bq)	Weighted means of fraction released (%)
Acetic acid pH 5.0	$(74.9 \pm 6.5) \times 10^5$	349 ± 21	$(53.1 \pm 4.9) \times 10^{-4}$
	$(59.0 \pm 5.1) \times 10^5$	3800 ± 200	
	$(13.0 \pm 1.2) \times 10^5$	800 ± 43	
Acetic acid pH 4.0	$(56.4 \pm 4.9) \times 10^5$	2900 ± 148	$(35.8 \pm 2.8) \times 10^{-4}$
	$(70.0 \pm 6.1) \times 10^5$	445 ± 25	
	$(56.0 \pm 4.8) \times 10^5$	157 ± 11	
Distilled water	$(42.5 \pm 3.7) \times 10^5$	792 ± 43	$(52.0 \pm 4.2) \times 10^{-4}$
	$(24.3 \pm 2.1) \times 10^5$	131 ± 92	
	$(45.0 \pm 3.9) \times 10^5$	175 ± 12	
Sulfuric–nitric acid pH 4.5	$(69.8 \pm 6.5) \times 10^5$	430 ± 8	$(708 \pm 9.3) \times 10^{-5}$
	$(83.2 \pm 6.5) \times 10^5$	575 ± 11	
	$(76.4 \pm 6.5) \times 10^5$	1200 ± 21	
	$(83.3 \pm 1.9) \times 10^5$	1269 ± 22	
	$(80.9 \pm 1.9) \times 10^5$	406 ± 8	
	$(83.6 \pm 6.5) \times 10^5$	1033 ± 18	

Fig. 3 Lysimeter leachate pH as a function of time; *open diamond* Lis5, *opens square* Lis 6, *open triangle* Lis7, × Lis8
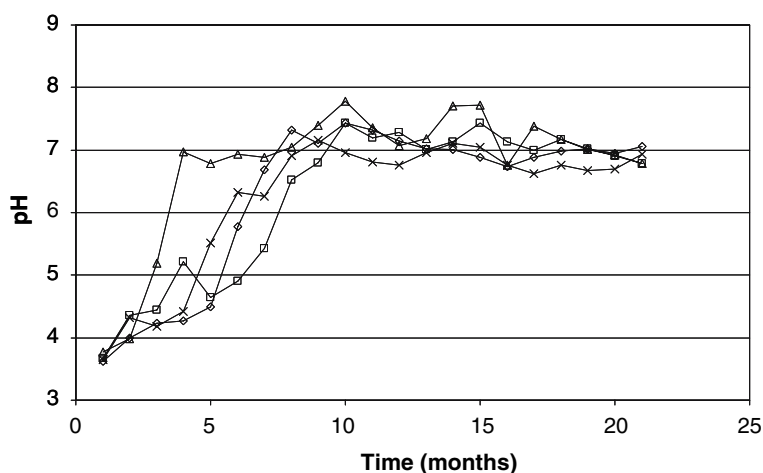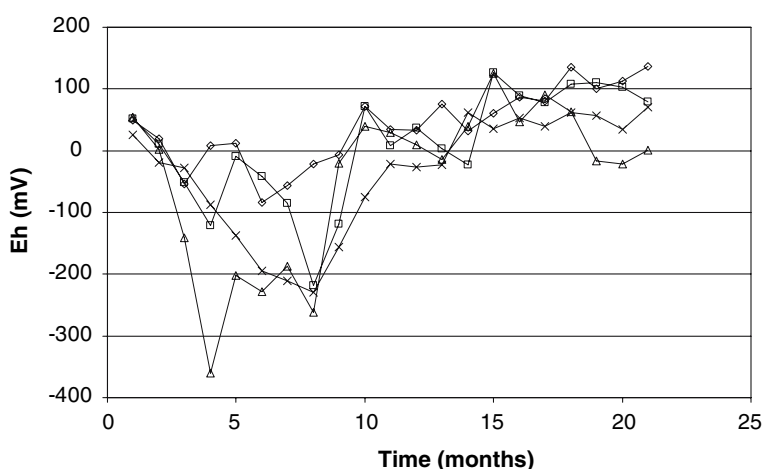

Fig. 4 Lysimeter leachate: redox as a function of time; *open diamond* Lis5, *open square* Lis 6, *open triangle* Lis7, × Lis8

In all collected samples, total solid content ranged from 4.5 to 0% (Fig. 5). Higher values were observed during the first period; after 6 months, the values were below 0.5%. Apparent color of the leachant varied much more during the first 4 months, showing colors like yellow, orange, brown and sometimes green. Afterward, all lysimeters became well behaved, displaying a beige or colorless appearance. Turbidity of the leachant samples changed from high, at the beginning, to low or none, during the last months.

No significant difference was observed between external and internal lysimeter temperatures, indicating that the waste degradation process did not produce heat. Although aerobic and anaerobic degradation of waste yields heat, this behavior would indicate that degradation was governed by anaerobic microorganisms. According to Christensen and Kjeldsen [6] aerobic and anaerobic heat generation is often neglected as its low generation rate does not very often result in temperature rise.

Figure 6 shows cumulative ^{241}Am releases over time for each lysimeter, expressed in Becquerels (Bq). The release pattern was bimodal for Lis5 and Lis6 as observed by Kelly et al. [7], with relatively large initial leaching followed by a more constant long-term release. According to these authors, the initial release is attributed to the washout of the easily accessible sources of contamination. Lis8 showed the lowest cumulative release, being restricted to a period of about 120 days; later ^{241}Am was observed only occasionally in very low concentrations. This process is largely controlled by physical phenomena, and is dependent on preferential pathways and on the location of the sources. Although the initial ^{241}Am activity in each lysimeter was approximately the same, the amount released differed in the three lysimeters. This may have been due to different degrees of deterioration of the sources, which were randomly chosen. In fact, other used sources were analyzed by optical microscopy, and they revealed irregular surfaces, with many dark marks and scratches, probably of exposed americium oxide (Fig. 7).

The total released fraction, over 16 months, represents 0.03, 0.28 and 0.001% of the initial inventory of Lis5, Lis6 and Lis8, respectively. The average activities released were 17 Bq, 84 Bq and 0.4 Bq per day, respectively, which were significantly lower than those observed in the leaching tests. No correlation was found between the lysimeters and

Fig. 5 Lysimeter leachate: solid content as a function of time; *open diamond* Lis5, *open square* Lis 6, *open triangle* Lis7, × Lis8
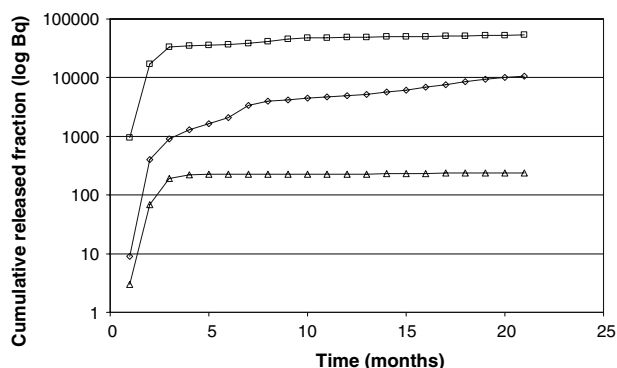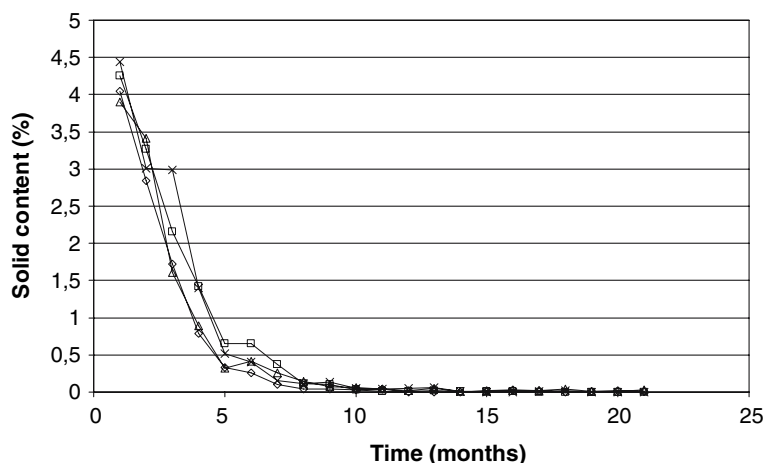

Fig. 6 Lysimeter leachate: ^{241}Am cumulative release fractions as a function of time; *open diamond* Lis5, *open square* Lis 6, *open triangle* Lis8
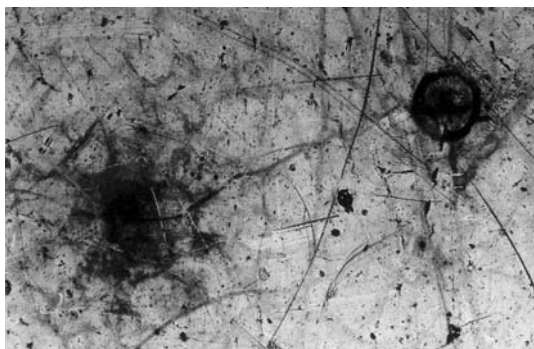

Fig. 7 Source surface observed with an optical microscope

the leaching assays. In the leaching experiments, the weighted means were 0.005% (acetic acid pH 5.0), 0.004% (acetic acid pH 4.0), 0.005% (distilled water) and 0.007% (sulfuric and nitric acid pH 4.5), for 24 h-leaching test. These results, associated with the gradual release of ^{241}Am suggest a relatively complex process, in which more parameters are involved. There are two reasons that might explain the differences observed between the two methods. The first is the mechanical agitation applied during the

leaching experiments that could have accelerated and intensified the ^{241}Am release. The second is the interaction between ^{241}Am and humic substances present in the lysimeters, which could affect the transportation of the element. Such interactions have already been studied [8, 9] and it was shown that humic substances sorbs many toxic metals and retard their release.

Processes related to the mass transport such as advection, diffusion and dispersion are important to investigate the movement of contaminants in groundwater. In our study, however, they were not considered because our evaluation focused on the estimate of the equivalent dose for members of the public. Ingestion through drinking water was considered the most probable path of exposure and, therefore, ^{241}Am concentration was the most important parameter. Other parameters involved were number of sources, released activity, annual water consumption, and dose factor for members of the public. Factors like soil porosity and distribution coefficients were also considered. Except for the activity and the number of sources, all parameters adopted were retrieved from the literature. In general, the annual dose from any radionuclide incorporated through drinking water can be calculated with Eq. (1) [10, 11]:

$$D = D_{\text{coeff}} \cdot W \cdot C_{\text{w}} \tag{1}$$

Where D is the annual dose (mSv year^{-1}), D_{coeff} is the dose coefficient (mSv Bq^{-1}), W is the annual water consumption (l year^{-1}), and C_{w} is the radionuclide concentration in the water phase (Bq l^{-1}).

C_{w} is calculated by the following Eq. (2) [12]:

$$C_{\text{w}} = \frac{C}{\theta_{\text{w}} + K_{\text{d}} \cdot \rho_{\text{b}}} \tag{2}$$

Where C is the radionuclide concentration in porous media (Bq cm^{-3}), θ_{w} is the water-filled porosity, ρ_{b} is the bulk density (g cm^{-3}), and K_{d} is the distribution coefficient (cm^3 g^{-1}).

To give a simple conservative estimate, one lightning rod disposed at a dump is considered, containing six sources with a total activity of 49,089 Bq (which is almost twice the activity released in 1 year from Lis6), a drinking water intake of 730 l year^{-1} [12], a dose coefficient of 2.0×10^{-4} mSv Bq^{-1} [13], a distribution coefficient of 1,900 cm^3 g^{-1} (the lowest value reported by Yu et al. [14]; for sand), a porosity of 0.40 and a bulk density of 1.30 g cm^{-3} [15]. The equivalent dose obtained from Eqs. (1) and (2) must be divided by an attenuation factor of 10 [15], considering the radionuclide dilution in groundwater. As a result, an annual dose of 5.3×10^{-2} mSv year^{-1} is estimated, which lies well below the effective dose limit of 1 mSv year^{-1} for members of the public, as recommended by the International Commission on Radiological Protection (ICRP) [13] even though conservative parameters have been chosen. In other words, about 20 lightning rods, disposed at the same place, would be necessary to exceed the effective dose limit.

It should be noted that the number of investigated lightning rods is quite small. The results of this study should therefore be interpreted with care. For example, it cannot be excluded that leaching rates could be considerably higher for lightning rods which were more heavily affected by weathering or whose surface was more damaged than those investigated here. It should also be noted that many studies have been performed in the past on the behavior of heavy metals in garbage dumps [16, 17, 18]. The results obtained in these studies, however, might not be representative for our specific scenario in which ^{241}Am sources were covered by gold layers. Finally, the choice of distilled water for the lysimeter studies might not be entirely representative for the real situation where acid rain might occur. Future experiments could improve this evaluation by using sulfuric–nitric acid solutions as a leachant.

Conclusion

The study described here should not be seen as a large-scale test, as only a limited number of lysimeters could be used. However, it may provide a first basis for characterizing the transfer of ^{241}Am from lightning rods to the human food chain. This process is slow, and the amount of ^{241}Am released was lower than that observed in the 24 h-leaching test. The results obtained suggest the risk of groundwater contamination by lightning rods disposed at uncontrolled garbage dumps to be low, although there is considerable ^{241}Am release from the sources. Nevertheless, authorities should intensify their public information efforts concerning the possible health hazard and the safe handling of lightning rods.

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