Essays on Nuclear Energy & Radioactive Waste Management

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- 3 -
Opening the Goiânia Accident Unburied Waste Packages[3]

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Abstract: The year 2017 marks 30 years since the radiological accident in Goiânia, Brazil, which resulted from the leakage of Cs-137 from a teletherapy equipment. The contaminated material collected during the response to the accident was disposed of in Abadia de Goiás, about 20 km from Goiânia. However, in the initial 15-day period before the authorities were notified, contaminated paper bales and scrap metal were sold and transported to material recycling facilities in the State of São Paulo, one thousand kilometers away. These materials were later collected in steel boxes and drums, and stored in the intermediate waste storage facility of the Nuclear and Energy Research Institute - IPEN, in São Paulo. The objective of this paper is to describe the work performed to check the present condition of the paper bales waste boxes, reassess the reported Cs-137 activities, and evaluate possible treatment methods that can be applied to reduce the volume of waste. Prospective waste treatment methods are discussed.

Resumo: O ano de 2017 marca 30 anos desde o acidente radiológico em Goiânia, Brasil, que resultou do vazamento de Cs-137 de um equipamento de teleterapia. O material contaminado coletado durante a resposta ao acidente foi depositado em Abadia de Goiás, a cerca de 20 quilômetros de Goiânia. No entanto, nos 15 dias iniciais até a notificação das autoridades, fardos de papel e sucatas

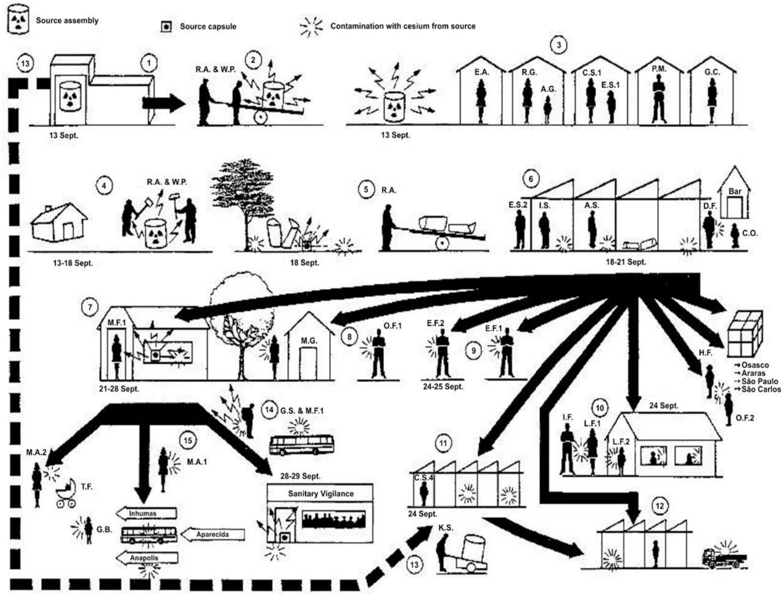
3 Lecture presented at the 2018 Waste Management Symposia (WMS) on March 18-22, 2018 in the city of Phoenix, AZ, United States. Available at: <https://www.xcdsystem.com/wmsym/2018/pdfs/FinalPaper_18422_0124110238.pdf>.

contaminados foram vendidos e transportados para empresas de reciclagem de materiais no Estado de São Paulo, a mil quilômetros de distância. Esses materiais foram posteriormente recolhidos em caixas de aço e tambores, e armazenados na unidade intermediária de armazenamento de rejeitos do Instituto de Pesquisas Energéticas e Nucleares - IPEN, em São Paulo. O objetivo deste artigo é descrever o trabalho realizado para verificar o estado atual das caixas de rejeitos de fardos de papel, reexaminar as atividades reportadas do Cs-137, e avaliar possíveis métodos de tratamento que possam ser aplicados para reduzir o volume de rejeitos. Potenciais métodos de tratamento de rejeitos são discutidos.

Introduction

The Goiânia accident was one of the most publicized radiological incidents and with the most serious consequences related to non-nuclear power. Approximately one Cs-137 half-life ago, a couple of scavengers removed equipment used for teletherapy from a derelict clinic, took it to a scrapyard, ruptured the sealed source capsule and divided a significant fraction of the about 50.9 TBq of Cs-137 among many individuals, who marveled at its bluish shine. The next day, many of them started having acute radiation syndrome, but only 15 days later a Sanitary Vigilance official identified the cause of the illness that affected all those who contacted the material, and alerted the radiation protection authorities.

During this period, the cesium chloride from the sealed source was being dispersed between people and their homes, contaminating buildings and every object inside them, in their yards and among the domestic animals, and the materials they collected for recycling and stored in the scrapyards. A diagram based on a drawing made shortly after the discovery of the accident, trying to explain what happened, is presented in Figure 1 [1].



Key: (1) the derelict clinic of the IGR; (2) removal of the rotating source assembly from an abandoned teletherapy machine by R.A. and W.P.; (3) source assembly placed in R.A.'s yard near houses rented out by R.A.'s mother E.A.; (4) R.A. and W.P. break up source wheel and puncture source capsule; (5) R.A. sells pieces of the source assembly to Junkyard I; (6) Junkyard I: the cesium chloride is fragmented and dispersed by I.S. and A.S. via public places; (7) D.F.'s house: contamination is further dispersed; (8) visitors and neighbors, e.g. O.F.1 are contaminated; (9) E.F.1 and E.F.2 contaminated; (10) I.F.'s house; other arrows indicate dispersion via visitors and contaminated scrap paper sent to other towns; (11) contamination is spread to Junkyard II; (12) contamination is spread to Junkyard III; (13) K.S. returns to the IGR clinic to remove the rest of the teletherapy machine to Junkyard II; (14) M.F.1 and G.S. take the source remnants by city bus to the Sanitary Vigilance; (15) contamination transferred to other towns by M.A.1.

Figure 1 - Diagram of the dispersion of Cs-137 in the Goiânia accident [1].

In the response to the accident, over 112,000 people had to be screened for radiation and 249 of them were found to have significant levels of contamination in or on their bodies. Twenty-four needed specialized medical care and four of the most exposed victims died within a month after the accident [2].

Three months were necessary for the complete cleanup of the contaminated sites, a work that involved about 600 professionals who took care of the victims, identified contaminated sites, decontaminated them, as well as managed the waste generated during these procedures.

During the cleanup operation, topsoil had to be removed from several sites and many houses were demolished. All the objects that were inside most houses were removed and examined for radiation, and in a number of cases, almost everything was beyond on-site decontamination capability. In the end, contaminated material amounting to 4.5 thousand tons was conditioned in packages as radioactive waste [3].

A repository with the same concept of the repository of L'Aube in France, or El Cabril in Spain, was built in the nearby municipality of Abadia de Goiás, about 20km from the initial contamination site, for disposal of this radioactive waste [4].

One important aspect of the decontamination and waste management work was the assessment of the collected radioactivity. Just after the response initiated, the rainy season in the Goiânia region was at the beginning and a copious amount of rain accompanied the process for recovery of the contaminated material. Approximately 10% of the initial activity is estimated to have been lost by dilution beyond the detection capacity during the response. Later work detected Cs-137 in water, sediment and other media, but no estimates of the total activity in each medium were calculated.

Another aspect that stands out in the Goiânia accident from other accidents involving sealed sources is that some of the contaminated material had been transported to locations up to 1,000 kilometers away from the initial incident, before the accident was recognized by the authorities. Besides Goiânia, the material was also taken to three nearby towns in the State of Goiás (Inhumas, Aparecida and Anápolis), as external and internal contamination of the bodies of the involved individuals or in their belongings.

Figure 2 - Contaminated paper bales collected and stored in the waste boxes in August 1988.

In the same way, recycling materials contaminated locations in four cities in the State of São Paulo. Scrap metal and paper bales were sold by the scavengers to recycling factories in the cities of São Paulo, Osasco, Araras and São Carlos. Approximately 8,000 kg of metal pieces, collected in the operations of decontamination of those factories (Figure 2), resulted in forty-three 200-liter drums, and 39,000 kg of discarded paper resulted in fifty 1.6 m3 steel boxes. The option of sending these waste packages back to Goiânia was discarded because of the anxiety and disturbance throughout the country after the accident. These drums and boxes containing the recovered wastes are currently stored in the intermediate radioactive waste storage facility of the Nuclear and Energy Research Institute, in the city of São Paulo. Figure 3 shows part of the packages in the storage room.

Final disposal of this waste is being evaluated under a technical and economic feasibility assessment for an alternative management. The purpose is to apply some sort of treatment to reduce the volume that will be transported to the final disposal facility, which is being planned for construction in Brazil in the near future. According to the Brazilian National Commission of Nuclear Energy (CNEN), this facility has a reference disposal cost of R$ 10,000 per cubic meter (US$ 3,000 or EU 2,600 per cubic meter approximately, by December 2017 exchange rates) [5], not including transportation by an estimated distance of 300 miles (about 500 kilometers) and handling costs. The total volume of the paper bales is around 80 m^3.

Figure 3 - Boxes with waste from the Goiânia accident today. The stains are scratches on the painting and corrosion points that were fixed.

One of the questions raised during the discussion about this work was the reliability of reported activity data, because at the time of conditioning, no significant effort was done to calculate the activity content of the boxes with a satisfactory degree of accuracy. In actuality, the activity values for the boxes were estimated based on calculations that assumed a homogeneous distribution of activity in the waste material and that used the highest exposure rate measured in the surface of the waste boxes; the model was quite simple and ignored the fact that the dose rates in each side of the box varied widely because of the hot spots in the waste. The calculations used the point-kernel method described by Rockwell [6].

The intended estimation of activity content for the waste boxes can take into consideration the exposure rates measured in each side and at different distances from the package surface. The calculations of activity content can make use today of the Microshield®[4] v.9.03 software package.

Therefore, the objective of this paper is to describe the work performed to check the present condition of the paper bales related to the dose rates, reassess the reported Cs-137 activities in waste boxes, and evaluate possible treatment methods that can be applied to reduce the volume of the waste.

Methods

A sample of 14 boxes was randomly selected from the 50 boxes in the storage. The boxes were weighed using the forklift scale, transported individually to a low background radiation area, out of the storage facility, and had their dose rates measured.

The measurement of the dose rates was used to estimate the activity by the dose-to-becquerel method, using the Microshield® v.9.03 software. Results of dose rate measurements at the distances of zero, 0.5 and 1.0 meter from

4 MicroShield® is a registered trademark of Grove Software, Inc.

each of the four lateral sides of the package surfaces were used to reduce the uncertainties of the estimates, as well as to model the distribution activity in each container as to better correlate with the measured dose rates.

The results of the measurements were used as input to calculate the estimated activities. To take into account the large inhomogeneity of the radioactive content, the measurement of each side was attributed to 1/9 fraction of the waste mass, as the modelling considers a 3x3 matrix of homogeneous regions, and used the MicroShield® to refine the initial estimates. The procedure was repeated until an acceptable distribution of activity was obtained, which correlates with the measurements (Figure 4).

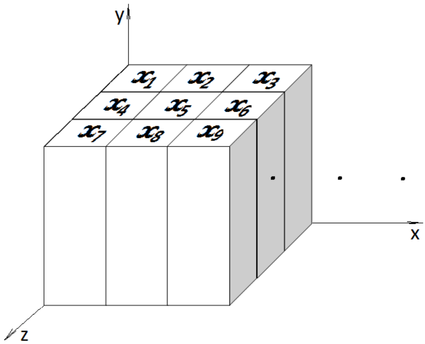


Figure 4 - Modelling of the MicroShield geometry (the dots are measurement points).

The dosimeters used for the measurements were the following (Figure 5):

- Kromek RayMon10®[5] radiation monitor;
- Eberline FH 40F2 radiation monitor.

Prospective waste treatment methods were discussed, such as wet combustion, incineration, biological degradation, among others.

5 Kromek RayMon10® is a registered trademark of Kromek Limited.

Figure 5 - Kromek RayMon10® & Eberline FH 40F2 radiation monitors.

Results & Discussion

Table I presents the results of the evaluation of dose rates differences between the measured and the calculated values that were based on the recorded activities of a sample of 14 waste boxes out of the fifty. The columns headed by 'old' and 'current' activities show the recorded activities for each box at the time of the conditioning of the waste and the calculated decayed present activity. The columns headed by 'old' and 'current', 'measured' and 'calculated' dose rates present the values obtained empirically in this work and those calculated with the recorded activities. It is clearly visible that the differences between values of corresponding points are not negligible, confirming that the recorded activities may be different from the actual values.

Table II presents the variations obtained between the calculated and measured dose rates. The difference between these values was expected, since the method used in the initial measurement in 1988 did not verify the four sides of the box in search of an average dose rate value.

Table I - Original and current waste boxes dose rates

Box	Packaging date	Old Activity (MBq)	Current Activity (MBq)	Old dose rate 0.0m (µSv/h)	Old dose rate 1.0m (µSv/h)	Current dose rate (measured) 0.0m (µSv/h)	Current dose rate (measured) 0.5m (µSv/h)	Current dose rate (measured) 1.0m (µSv/h)	Current dose rate (calculated) 0.0m (µSv/h)	Current dose rate (calculated) 0.5m (µSv/h)	Current dose rate (calculated) 1.0m (µSv/h)
261	01/mar/88	3245	1619	200.0	13.0	43.1	16.8	8.3	93.4	38.8	18.0
339	02/mar/88	810	404	50.0	3.0	9.7	7.4	6.9	22.7	9.5	4.4
348	01/mar/88	1624	810	100.0	6.0	5.2	1.7	1.0	43.4	18.2	8.4
350	03/mar/88	2272	1134	140.0	9.0	0.4	0.3	0.4	42.6	18.9	8.6
352	03/mar/88	260	130	16.0	1.0	1.4	0.8	0.6	6.4	2.7	1.3
354	03/mar/88	1624	810	100.0	6.0	4.0	1.5	1.0	43.6	18.3	8.5
1334	01/mar/88	3245	1619	200.0	13.0	71.5	32.1	14.4	79.9	34.0	15.7
1336	03/mar/88	714	356	44.0	3.0	0.4	0.4	0.5	18.8	7.9	3.7
1339	01/mar/88	19462	9711	1200.0	75.0	215.4	92.9	42.7	521.0	218.8	101.2
1340	01/mar/88	648	323	40.0	3.0	1037.4	745.5	413.0	18.1	7.6	3.5
1346	01/mar/88	1624	810	100.0	6.0	24.9	7.7	3.7	45.2	18.9	8.7
1356	02/mar/88	1624	810	100.0	6.0	43.7	22.2	11.1	47.6	19.7	9.2
1357	01/mar/88	324	162	20.0	1.0	2.5	1.9	1.5	9.5	4.0	1.8
1377	03/mar/88	455	227	28.0	2.0	1.0	0.9	0.9	12.2	5.1	2.4

Note: The current measurements were performed on November 29 and 30, 2017.

Table II - Percent variation of measured and calculated dose rates and estimated activity concentration

Box	Recorded net weight (kg)	Measured net weight (kg)	Dose rate variations (%) On contact Measured	Dose rate variations (%) On contact Calculated	Dose rate variations (%) At 1 meter Measured	Dose rate variations (%) At 1 meter Calculated	Current estimated activity concentration (kBq/kg)
261	475	333	78	53	36	(-38)	4858
339	419	349	81	55	(-130)	(-47)	1157
348	460	378	95	57	83	(-40)	2142
350	315	627	100	70	96	4	1807
352	311	435	91	60	40	(-30)	298
354	349	375	96	56	83	(-42)	2159
1334	374	430	64	60	(-11)	(-21)	3763
1336	350	388	99	57	83	(-23)	918
1339	321	377	82	57	43	(-35)	25740
1340	372	349	(-2494)	55	(-13667)	(-17)	926
1346	365	353	75	55	38	(-45)	2294
1356	430	322	56	52	(-85)	(-53)	2514
1357	352	321	88	53	(-50)	(-80)	503
1377	300	377	96	56	55	(-20)	602

Note: the figures in captions are the negative values related to the comparison between the old and new numbers.

Some alternative approaches were considered for the reduction of radioactive waste volume in the stored boxes. The evaluation suggested as the one with the greatest potential would be the wet combustion, which consists in the use of an oxidizing reagent, a chemical reactor operating at room temperature and using a filtering system appropriate to the gases generated in the process.

The contaminated paper could also be transformed into pulp by inserting it in a recipient with hot water under agitation. The Cs-137 is very soluble and will be retained in the water, for later treatment. The expected result is of an extensive volume reduction of the paper pulp, possibly even reaching the unconditional clearance limit.

Other methods have been considered, such as incineration and biological degradation. However, due to the difficulty in obtaining the required equipment, as well as the licensing, these methods were disregarded. The method of biological degradation may already have started inside some boxes, by bacteria or fungi, but at the time it was not possible to evaluate the current state of the material. A visual inspection of the interior of the boxes requires a fume hood with insulation from the atmosphere to prevent contamination and dispersion of the material during opening, which is still under planning.

Conclusions

The current results indicate that none of the boxes checked are close to the clearance limit, which is 10 kBq/kg [7] – box 352 presented the lowest estimated value of 298 kBq/kg, almost 30 times over the limit. Without any sort of treatment, these boxes will not reach the clearance level in less than 150 years, at least.

The current values measured are more accurate than the previous ones measured 30 years ago, allowing a better analysis of its contents. Therefore, future works are being planned, including visual inspection, taking samples and exploring options to identify the best treatment method of volume reduction for final disposal.

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