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● *Environmental Considerations*

DISTRIBUTION OF ^{137}Cs IN SOILS DUE TO THE GOIÂNIA ACCIDENT AND DECISIONS FOR REMEDIAL ACTION DURING THE RECOVERY PHASE

E. C. S. Amaral, M. E. C. Vianna, J. M. Godoy, E. R. R. Rochedo, M. J. Campos, M. A. Pires do Rio, J. P. Oliveira, J. C. A. Pereira and W. G. Reis
Instituto de Radioproteção e Dosimetria/CNEN, Av. das Américas, Km 11,5, Rio de Janeiro, CEP 22602, Brazil

***Abstract*—In September 1987, a powder radioactive source was removed from a teletherapy machine in Goiânia, Brazil. Subsequently, it was ruptured in a residential garden causing the dissemination of ^{137}Cs throughout the city. Soil resuspension processes and burial of contaminated house waste in unused gardens were the major contributors to the Cs dissemination in soils at the secondary contaminated sites. Only locations within a radius of 50 m from the primary contaminated sites presented the need for remedial action. The radiation dose-rate measurements and the soil profiles were good indicators of the extent of the secondary contamination and were fundamental for the decisions taken regarding decontamination procedures. In cases of surface contamination, 60% on average of the total activity remained in the upper 1.5-cm layer over the first 5 mo after the accident, and topsoil removal proved to be an effective procedure for decontamination.**

INTRODUCTION

ON 13 SEPTEMBER 1987, a shielded radioactive ^{137}Cs source (50.9 TBq or 1375 Ci at the time) was removed from a teletherapy machine in an abandoned clinic in the city of Goiânia, Brazil. The source was ruptured in a residential garden, and the remnants of the source assembly were sold to a junkyard owner. It started a radiological accident (its seriousness only began to be appreciated on September 29) in a total urbanized suburb, resulting in large doses of radiation to about 30 people and contamination of residences and public places.

The primary contaminated sites were the yards where the source assembly was broken out of its shielding and where the source capsule was ruptured, and some residences of the most affected people. However, the source characteristics, together with the meteorological conditions, favored the dispersion of ^{137}Cs from the primary contaminated sites. The radioactive source was a cesium chloride salt powder, which is readily dispersible and highly soluble. Its dispersion from the broken source into the environment was increased by heavy rains. In addition, the high temperatures dried out the ground, and strong winds caused resuspension and dispersion. Contamination deposited on roofs was the major contributor to dose rates indoors, and for some houses the roof tiles had to be removed.

Values of ^{137}Cs radioactivity concentration in soils and leaves ranged from 10^2 to 10^5 Bq kg^{-1}, and fruits were one order of magnitude lower. The fruits were collected and the trees pruned according to a derived limit of 650 Bq kg^{-1} for fruits (IAEA 1988). In addition to these natural processes, there was also transport of radioactive material by people passing through the contaminated sites and by the deposition of contaminated house wastes in unused gardens.

Radiological surveys have shown that the most primary and secondary contaminated sites occurred at the urbanized suburb involving an area of about 1 km^2. Most of the region is constituted by small houses surrounded by big trees. At the residential gardens, there are common fruit trees and sometimes vegetable gardens and a few chicken coops.

Among several remedial actions, the measures to be taken were selected under strong social and public pressure. Therefore, a total dose criteria of 5 mSv in the first year was adopted, for the critical group, to establish the action levels for soil and house decontamination. In turn, this was subdivided into 3 mSv for outdoor external irradiation; 1 mSv for indoor external irradiation; and 1 mSv for the actual or projected internal dose, considering all the relevant pathways.

In the early phase following the accident, after the primary contaminated sites were identified, evacuation or isolation actions took place according to the radiation dose-rate levels set. Later, these primary sites were decontaminated using heavy machinery to remove large amounts of soil and demolish houses. The time period

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required for decontamination was the most important factor to be considered, since the radiation dose-rate levels were elevated (up to 1 Gy h^{-1}). The amount of waste generated, and the way decontamination measures were put into action, were not the primary concern at this point (IAEA 1988).

Three months later, decontamination was initiated in areas surrounding the primary contaminated sites, where the radiation dose rates were much lower (on the order of μGy h^{-1}). During this phase, soil was decontaminated to a level of ^{137}Cs surface activity, estimated to yield 3 mSv for outdoor external irradiation and 1 mSv for the projected internal radiation dose for the first year. Limited guidance was available on the unrestricted use of contaminated land. Thus, decisions whether to decontaminate the gardens and determinations of the amount of soil that must be removed were based on the potential dose to persons who might inhabit the area at some point after cleanup.

The approach used was to assume that gardens are available for unrestricted use over the first year after cleanup, and so the following pathways were applicable: surface exposure, inhalation of resuspended radioactive soil and ingestion of vegetables, fruits, pork, chickens, and eggs. An IAEA environmental pathway analysis model (IAEA 1982, 1986) was used with the introduction of some modifications to consider the radionuclide uptake by chickens due to soil ingestion (Boone et al. 1981) and the local soil profile data instead of homogeneous soil concentration usually adopted on assessment models. A resuspension factor of 10^{-7} was adopted according to preliminary local measurements. Based on all these assumptions and considering an occupancy factor of 0.5, it was found that the external dose was responsible for about 75% of the total estimated dose for individuals. Ingestion dose was roughly responsible for 25% of the total dose, with the chicken flesh consumption as the critical pathway.

From this analysis, 0.8 μGy h^{-1} and 22.5 kBq kg^{-1}, respectively, were established as action levels for the net radiation dose rate measured 1 m above the ground and for the ^{137}Cs surface radioactivity concentration in soil (1.5 cm deep). As a consequence, gardens of most homes in a region within a 50-m radius from the primary contaminated sites had to be decontaminated. However, initial efforts to accomplish this task were characterized by drastic manual removal of the soils, sometimes making big holes and even destroying most of the gardens. To the homeowners, this procedure seemed to be a terrible action. It was difficult for them to understand why their gardens had to be decontaminated at this late phase. They thought that they were being exposed to high doses of radiation from the beginning of the accident, and only at that time were the authorities taking adequate care of them.

In addition to this lack of knowledge of radioactivity and radiation protection, they also did not trust the nuclear authorities; thus, it was very difficult for them to understand the difference between the two distinct decontamination phases. Furthermore, a poorly detailed evaluation of the distribution of the soil contamination, together with a poorly designed procedure for removal of the contaminated soil, caused an unnecessary mixing of a large amount of "clean" soil with actual contaminated soil. The net result was not only a problem for waste management, but also an additional emotional distress to the population. In fact, the homeowners thought the number of containers of contaminated soil removed from their houses was proportional to the amount of radioactivity they were being exposed to. At this stage, we were conscious that minimization of the time taken for decontamination was no longer a primary requirement, and the problems mentioned above became our main concern.

Therefore, it was necessary to develop an improved and less severe procedure to remove the contaminated soil and also leave the gardens with a more acceptable appearance, while producing no more waste than necessary.

MATERIALS AND METHODS

Decisions on the remedial actions could not be made until comprehensive surveys were completed. Then it was decided to conduct a characterization survey within a radius of 100 m from the primary contaminated sites. During this second phase, the radiation dose rates were lower and without the radiometric influence of the hot spots. This region can be seen in Fig. 1. Sometimes the survey region had to be enlarged due to the insistence of the homeowners. The survey consisted of radiation dose-rate measurements 1 m above the ground, house by house, and at all the unused gardens as well.

Soil profiles were determined, based on the radiation dose-rate value, and the ^{137}Cs activity for each layer was measured. The soil profiles were determined after removing all stones and grass. A flat blade of a digger (20 cm long) was pressed into the soil perpendicularly to the surface. Soil layers of 0–1.5, 1.5–3.0, 3.0–6.0, and 6.0–15 cm thick were sampled. Each one of the soil layers was cut with the help of a ruler to select the thickness and taken with a flat blade digger. Care was taken to prevent, or at least to minimize, any soil particles from the previous layer from falling into the next one. This was done cutting smaller surface areas for each successive layer.

To develop a less-severe procedure to remove contaminated soil and also to leave the gardens with a more acceptable appearance while producing no more waste than necessary, some experiments were performed. The terrain of the experimental area was about 180 m^2 of a few trees and some wild grass. After pruning the trees, the terrain was divided into 23 rectangles of 2 × 4 m. A radiation dose-rate survey was performed within each rectangle before and after removing of the grass.

Radiation dose-rate measurements 1 m above the ground and at the surface were performed to identify the location that presented the highest radiation exposure value. Soil profile up to 15 cm deep was determined to indicate the soil layer to be removed. Finally, it was necessary to separate the contaminated soil, considered rad-

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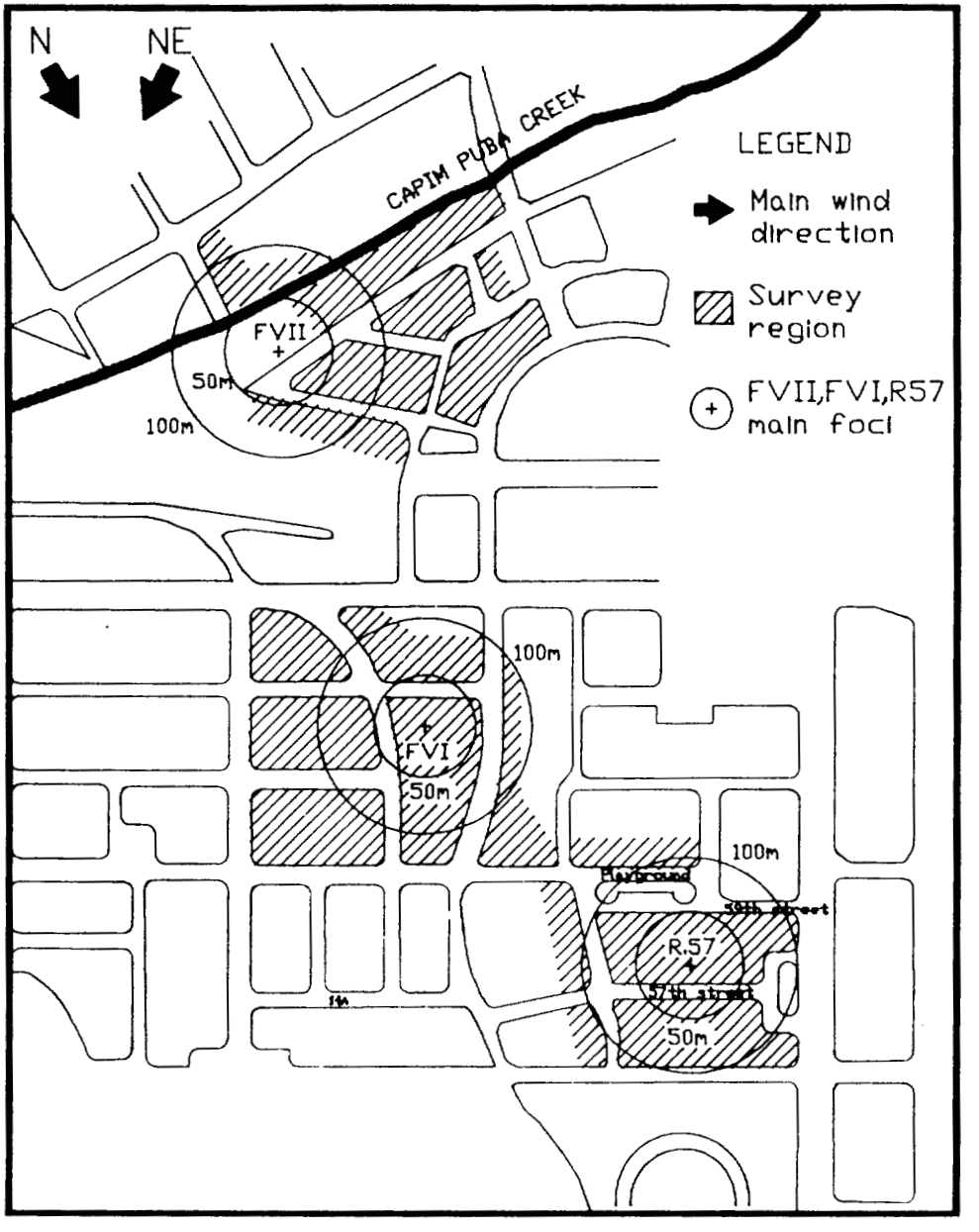

Fig. 1. Map of the primary contaminated sites and surrounding areas.

waste, from that considered normal or sanitary waste for each soil layer removed. Since the Brazilian National Nuclear Energy Commission set 75 kBq kg^{-1} for the radioactivity concentration to be considered radwaste (CNEN 1985), a value 30% lower was adopted as a safety factor.

The radwaste was placed in adequate containers and removed to the waste repository specially constructed in Goiânia (IAEA 1988). The soil considered sanitary waste was placed in adequate plastic bags and removed to the city sanitary waste disposal facility. Radiation dose-rate measurements were performed after each soil layer removal. At the end of the decontamination procedure, the ^{137}Cs activity at the soil surface was determined.

The screening survey was performed with a portable prospecting radiation meter* with a NaI(Tl) scintillation detector. The radiation dose-rate measurements were performed with a FAG radiometer,† a Geiger-Müller tube

* Model SPP-2-NF. Manufactured by Saphymo-Physiotechnie, 94623 Rungis Cedex, France.

† Model FH 40 F. Manufactured by System Frieseke, D-85200 Erlangen, Federal Republic of Germany.

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with a measuring range between 5×10^{-7} and 10^{-2} Gy h^{-1}. Both devices were calibrated at the Secondary Standard Dosimetry Laboratory (SSDL), Rio de Janeiro.

The ^{137}Cs radioactivity concentration in the soil samples was determined with a NaI(Tl) 76 × 76 mm crystal‡ coupled to a single-channel analyzer. The soil samples were set in plastic pots of 250 mL volume at a standardized geometry with an efficiency of 3.6%.

The quick field estimate of ^{137}Cs radioactivity concentration in the removed soil was taken from the surface radiation dose-rate measurements. This procedure was used to quickly discriminate between soils to be considered radwaste and those to be discharged at the city sanitary waste disposal facility. The correlation between the two quantities was established in a laboratory, using a fixed source-detector geometry and soil samples with different magnitudes of known ^{137}Cs radioactivity concentrations.

RESULTS AND DISCUSSION

Generally, within a radius of about 50 m from the primary contaminated sites, radiation dose-rate values higher than 0.1 μGy h^{-1} were found. A heterogeneity in the contamination occurred in the same terrain, and hot spots up to 30 Gy h^{-1} were identified. Most of the results, in the region of a radius between 50 and 100 m from the primary contaminated sites, ranged from 0.02 to 0.05 Gy h^{-1}. The exception was a playground on 59th Street (Fig. 1), where results of up to 0.1 Gy h^{-1} were obtained and were mainly due to the influence of toys and seats not totally decontaminated during the initial phase. In addition to the deposition of resuspended radioactive material from the garden on 57th Street, where the source was opened, people directly involved in the accident used to spend time at the playground.

In the residential gardens, the soil radioactivity concentrations showed the ^{137}Cs to be randomly distributed at the soil surface (Table 1). The most hot spots, however, occurred around the trees or near the roofs. This was probably due to the higher ^{137}Cs activity deposited on roofs and canopies of trees that later reached the grassland as a result of weathering by rain and wind. Deposition of Cs in forest and in nearby grassland as a function of time after the beginning of the Chernobyl radioactive fallout confirms this assumption for trees (Bunzl et al. 1989).

Soil profiles were determined and all presented the same shape irrespective of the specific activity at the surface. On the average, the profile results showed that 60% of the total radioactivity remained in the upper 1.5-cm layer over the first 5 mo after the accident occurred. This result is in accordance with the observations of natural ^{137}Cs fallout (UNSCEAR 1977). However, it is important to emphasize that soil in residential gardens was not disturbed after the accident. Some examples of the ^{137}Cs vertical distribution in soils obtained at residential gardens are presented in Fig. 2.

‡ Model 76 SP 76. Manufactured by Quartz & Silice—"Le Miroirs"—Cedex 27—92 096, Paris, La Defense, France.

Table 1. Distribution of ^{137}Cs radioactivity concentration in surface soil at a residential garden.

Location[a]	^{137}Cs radioactivity concentration[b] (kBq kg^{-1})	Location[a]	^{137}Cs radioactivity concentration[b] (kBq kg^{-1})
1	20 ± 1.0	9	7 ± 0.3
2	14 ± 0.8	10	7 ± 0.3
3	17 ± 0.8	11	12 ± 0.5
4	17 ± 0.8	12	4 ± 0.2
5	7 ± 0.3	13	17 ± 0.9
6	38 ± 1.8	14	115 ± 5.8
7	14 ± 0.7	15	14 ± 0.7
8	6 ± 0.3	16	41 ± 2.0

[a] Rectangles of 2 × 4 m, one near the other.
[b] Error with 68% of confidence.

The high solubility of Cs together with precipitation up to 25 mm d^{-1} during the period after the accident can explain the radioactivity concentrations found at deeper soil layers. Rains were preceded by winds and very high temperatures. In the 15 d prior to the accident discovery, the mean temperature was 26.4°C. These conditions not only contributed to the rapid spreading of ^{137}Cs in the urban area, but also the temperature was responsible for the very dry soil conditions. Therefore, it facilitated the ^{137}Cs migration in soil during the rainy period. These weather conditions are common in tropical countries since short-duration heavy showers occur during summer and are always preceded by strong winds following a period of high temperatures.

The results obtained from this experiment were fundamental to establish the measurements necessary for site decontamination. Based on the use of the soil profile data as an indicator of the amount of soil to be removed, an improved and less-severe procedure was developed. The radiation dose-rate values at 1 m above the ground at the experimental garden ranged from 0.8 to 3.1 μGy h^{-1}. It is important to mention that no significant differences were observed between the radiation dose-rate values, before and after the removal of the grass. The grass was measured at a different location, and it was not considered radwaste.

A soil profile was performed at the "rectangle" that presented the highest dose-rate value. Again, the results showed most radioactivity to be within the upper 1.5 cm soil layer and, according to the action limits set, they indicated no need to remove more than that. Then, this first layer of soil was removed from the rectangle, and any significant difference in the radiation dose-rate values was observed. Since the dose rate was still above the action level, an additional layer was removed, and the same result was found. This condition was not in accordance to the profile data indication. It was not possible to go ahead, otherwise a large and deep hole would be created in the garden, as was done during the primary site decontamination phase.

The surface measurement of the second layer of removed material did not present any significant radiation

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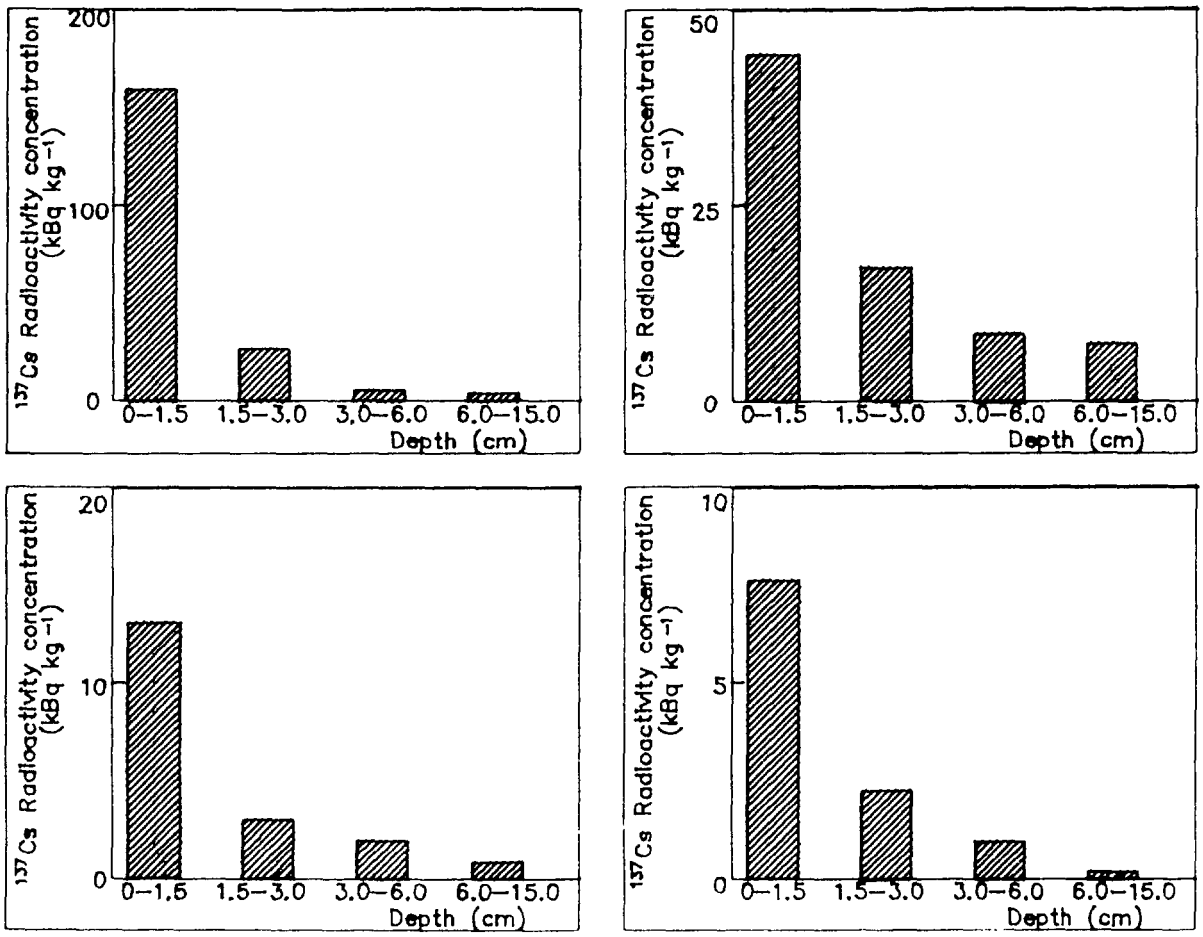

Fig. 2. Vertical distribution of ^{137}Cs in soils from residential gardens.

dose-rate value. Therefore, it would not have been necessary to remove the second layer. Then it was assumed the dose-rate value measured 1 m above the ground at this location was greatly influenced by other contaminated areas, most likely due to the skyshine effect and ground contributions from other locations (Jacob and Paretzke 1986). Hence, it became evident that it would not make sense to try to decontaminate a small area, since the contributions from the surroundings, even in lower concentrations, influenced the radiation dose-rate measurements and thus would contribute to the external exposure of a person located there.

As a consequence, it was decided to remove the soil from the entire area. At this step, the objective was not to remove the 1.5-cm layer, but rather to scrape the soil surface with a rake. After each scraping, radiation dose-rate measurements were performed 1 m above ground. As can be observed from Table 2, it was necessary to conduct this procedure twice in order to reach the action limits set. It was also observed that the radiation dose rate decreased by a factor of two and three, respectively, after the first and second removal. It is important to mention that the majority of the soil removed in the last step was not considered radwaste.

After that, the radioactivity concentration of a composite sample of surface soil (1.5 cm) from each rectangle was 18 kBq kg^{-1}, with a standard deviation of 7 kBq kg^{-1}. The radiation dose-rate mean value of 0.56 μGy h^{-1} (Table 2) may correspond to a net value of 0.41 μGy h^{-1} if it is considered as background 0.15 μGy h^{-1}. The background was measured at locations far from the area, and values ranging from 0.12 to 0.17 μGy h^{-1} were found. Even considering the uncertainty of the background dose-rate value, the final soil surface dose rate was about one-half that predicted for the ^{137}Cs radioactivity at the surface

Table 2. Summary of the dose-rate measurements 1 m above ground at the experimental garden (μGy h^{-1}).

Situation	Mean	Median	Std. Dev.	Min.	Max
Initial	1.74	1.70	0.61	0.80	3.1
First soil removal	1.01	1.00	0.21	0.60	1.5
Second soil removal	0.56	0.58	0.20	0.23	1.1

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using the conversion factors for infinite plane sources (IAEA 1982, 1986).

The aim of this experiment was to establish a less-severe, yet effective, decontamination procedure. Accuracy and precision in some steps of the practical work were not requirements. The profile results were only used to be sure the amount of contaminated soil to be removed was within a specific layer. However, thinner layers were removed at each step, and the radiation dose-rate results at 1 m above the ground were the deciding factor. This procedure was successfully applied to all other residential gardens, since the top layers were responsible for ^{137}Cs retention. It was an improved and less-severe procedure that resulted in the generation of less radwaste than the primary decontamination procedure; the gardens were returned to the owners without holes and with a more acceptable appearance.

There were some unused gardens near the residences, where people used to place domestic waste. In these gardens, the contamination was randomly distributed, and the radiation dose-rate measurements usually showed the presence of significant hot spots at very specific locations. In these gardens, the soil profiles presented very irregular shapes. At these unused gardens, the results showed the presence of contaminated domestic waste, mostly already buried, as the main contributor to both the soil contamination and the high dose-rate values. As an example, the results obtained in a survey administered at an unused garden will be described. This garden presented two different types of soil: the original soil, which was similar to those from the residential gardens, and one area of 15 × 8 m covered with about 20 cm of red clay. In the original soil, the radiation dose rate ranged from 0.1 to 0.2 μGy h^{-1}, and in the other one, it presented a value from 0.1 μGy h^{-1}, increasing with the proximity to a specific location with 20 μGy h^{-1}.

The profiles obtained at the two different regions of the garden are shown in Fig. 3. In a general way, the profiles presented similar shapes according to those obtained at the residential gardens. Therefore, the same decontamination process adopted for the residential gardens was adopted for this unused garden. However, once the 20-cm layer of clay had been removed, the radiation dose rate started to increase. Other soil profiles were performed

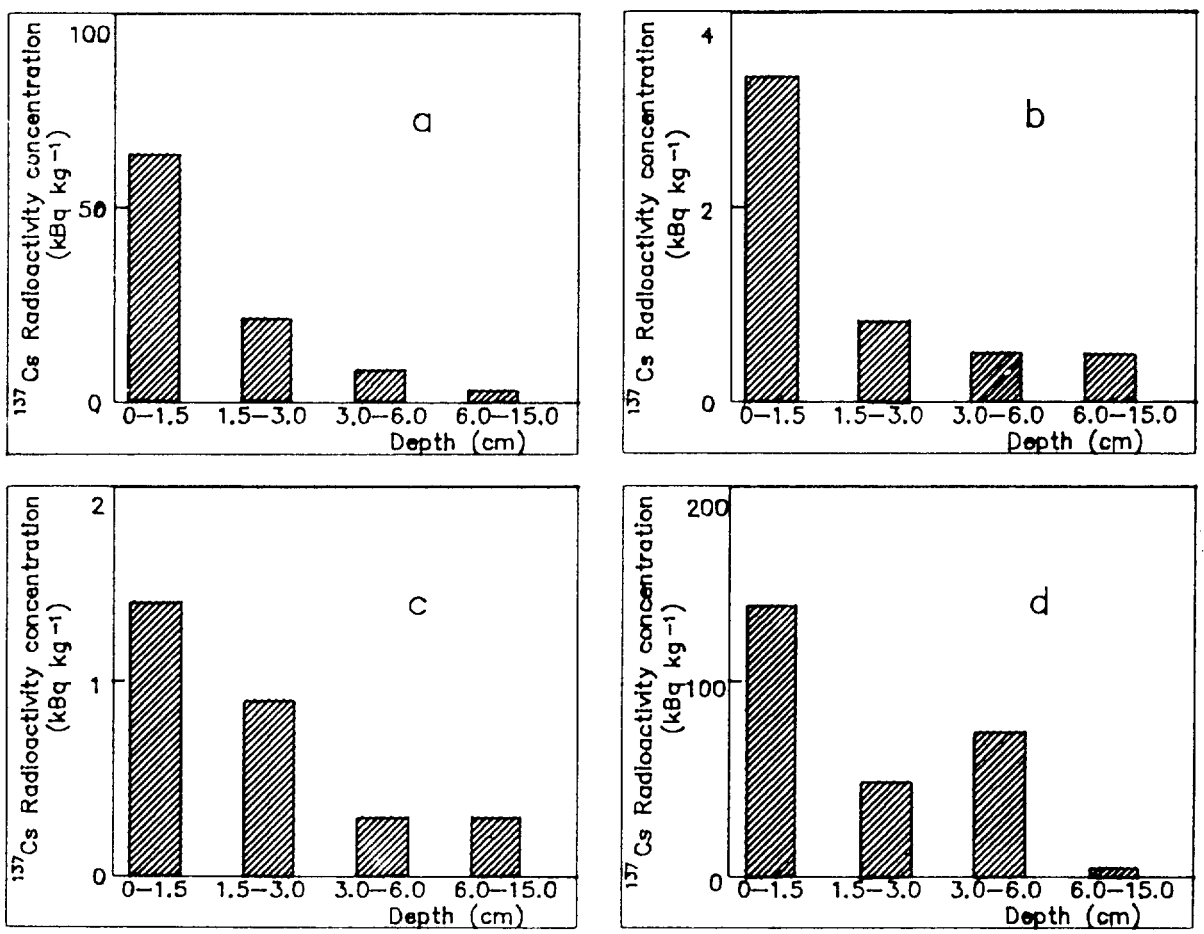

Fig. 3. Vertical distribution of ^{137}Cs in soils at an unused garden: a and b, original soil: c and d, red clay.

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and demonstrated the existence of true sanitary waste under the clay layer.

Different types of debris from contaminated houses (mainly from one primary contamination site nearby) were placed on the unused garden before the National Nuclear Energy Commission intervention and were probably covered later. In Fig. 4, the different and irregular shapes of some soil profiles, performed on adjacent sites, can be observed. Cesium migration was probably facilitated by rainfall and by the presence of ammonium hydroxide due to the waste decomposition. The presence of several types of debris can explain the different profile shapes.

The clay layer contamination probably occurred due to Cs leaching, from the uncovered waste by rainfall, or the occurrence of a partition process between the clay and the waste. Although other unused gardens presented the same type of contamination, most of them were covered with new uncontaminated waste. At these gardens, the decontamination efforts were difficult since manual techniques were employed to avoid mixing of the soils. Nevertheless, the soil profile data were useful as a good indicator of the extent and depth of the contamination before starting the decontamination.

CONCLUSIONS

1. In addition to the physical displacement and personal contact of the people involved in the accident, the ^{137}Cs dissemination in the urban area surrounding the primary contaminated sites was due to:

- Dispersion of the resuspended contaminated material, mainly radioactive soil, from the primary contaminated sites to the surrounding areas.
- Buried contaminated house waste in unused gardens near the residences.

2. The physical and chemical properties of a radioactive source are important factors in an accident. The local characteristics largely determine the extension of the contamination as well.

3. The radiation dose-rate measurements, together with the soil profile results, were good indicators of the

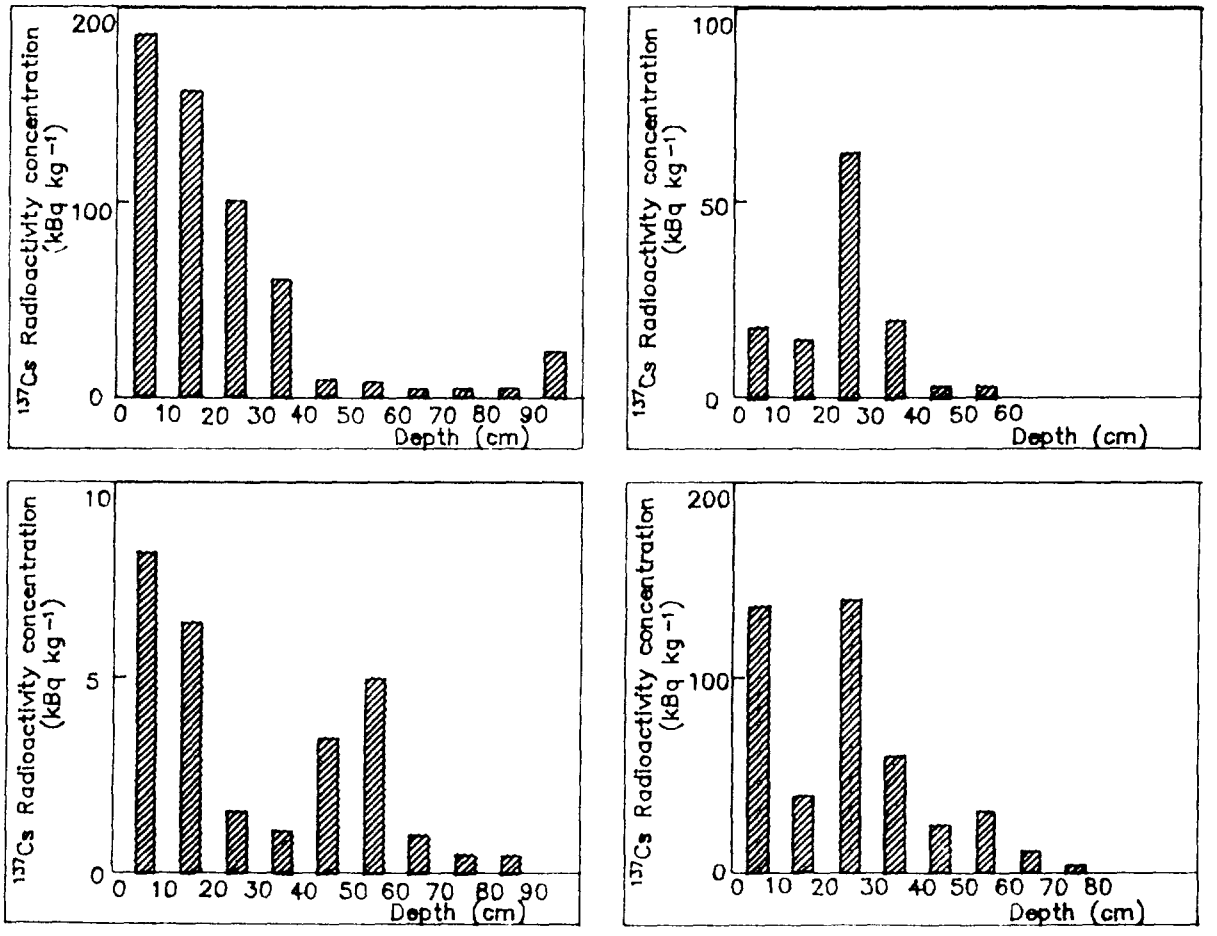

Fig. 4. Vertical distribution of ^{137}Cs in sanitary waste at an unused garden.

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contamination mechanisms and were the basis for decisions concerning decontamination measures.

4. In cases of surface contamination, 60% on average of the total soil activity remained in the upper 1.5-cm layer over the first 5 mo after the accident; topsoil removal proved to be an effective procedure for decontamination.

5. The public should be made aware of what radiation is and the benefits it has. In addition, an adequate system of information is fundamental to avert panic during the different phases of emergency response.

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