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Natural and man-made radionuclides in the soil of a nuclear facility site located in a coniferous forest in central Mexico

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RESUMEN

Se determinaron ^{137}Cs, ^{226}Ra, ^{40}K y ^{235}U en muestras de suelo superficial y ^{222}Rn a distintas profundidades en el suelo del Centro Nuclear de México, localizado en un bosque de coníferas a 3000 m de altitud. Los emisores gamma se evaluaron por espectrometría gamma de bajo fondo y la concentración de radón en el suelo se midió utilizando detectores sólidos de trazas nucleares. Los resultados indican que las actividades específicas de ^{137}Cs, ^{226}Ra y ^{235}U fueron en general inferiores a los límites de referencia recomendados para suelos superficiales en campos y áreas boscosas, excepto en un área pequeña donde se encontró una contaminación moderada. Los valores de ^{222}Rn en suelo confirman que la contaminación de ^{226}Ra fue superficial.

PALABRAS CLAVE: ^{137}Cs, ^{226}Ra, ^{40}K y ^{235}U en muestras de suelo, radón en suelo, bosques, detectores sólidos de trazas nucleares, espectrometría gamma.

ABSTRACT

^{137}Cs, ^{226}Ra, ^{40}K and ^{235}U in surface soil samples and ^{222}Rn levels at different depths in the soil were studied at the Nuclear Centre of Mexico located in a forest at 3000 m altitude. The gamma emitters in the soil samples were measured with a low level counting gamma spectrometer and soil radon concentration was determined using Solid State Nuclear Track Detectors. The results indicate that the specific activities of ^{137}Cs, ^{226}Ra and ^{235}U were in general below the recommended screening limits for surface soils from open fields and forested sites, except at one small area where a moderate punctual contamination was found. The ^{222}Rn values confirm that ^{226}Ra contamination in the soil was of low level and superficial.

KEY WORDS: ^{137}Cs, ^{226}Ra, ^{40}K and ^{235}U in soil samples, soil radon, forest area, track detectors, gamma spectrometry.

INTRODUCTION

Atmospheric deposition of man-made radionuclides, such as ^{137}Cs, occurred as a consequence of the major series of nuclear weapons tests and the Chernobyl accident (Muck, 1997). The first estimations of environmental radioactivity in Mexico were performed in 1964 in order to obtain pre-operational data to license a research reactor to be located at the Nuclear Centre of Mexico (NCM). Since then, surveys have been systematically conducted at the site to determine the fluctuations over time of environmental radioactivity and to extract eventual variations due to a possible abnormal nuclear operation. It has been observed that the main source of environmental ^{137}Cs at the site and surrounding localities is the fallout from the nuclear weapons tests (Gaso *et al*., 2000). At present the environmental monitoring program includes samples of soil, local foodstuffs, bioindicators, rain water, air, etc. Soil and indoor radon surveys have also been eventually performed at the site (Segovia *et al*., 1989; Segovia *et al*., 2001).

Five years ago, a radiometric punctual contamination in the soil was discovered at the NCM, related to an unplanned release in the past, of ^{226}Ra, ^{235}U and ^{137}Cs from the drainage system of an uranium extraction pilot plant and associated radiochemical laboratories.

The main purpose of the present paper is to quantify the concentration values of ^{226}Ra, ^{235}U and ^{137}Cs in soil samples obtained at the site, with especial attention to the location where the punctual contamination was found. ^{40}K was also determined in the soil samples. ^{222}Rn levels were also monitored at different depths in the soil.

EXPERIMENTAL

The study was performed at the Nuclear Centre of Mexico (NCM) (19° 17’19’’N; 99° 22’43’’ W) and two reference sites (Ajusco and Coatepec hills, 15 km away) (Figure 1). The three sites are located at an average altitude of

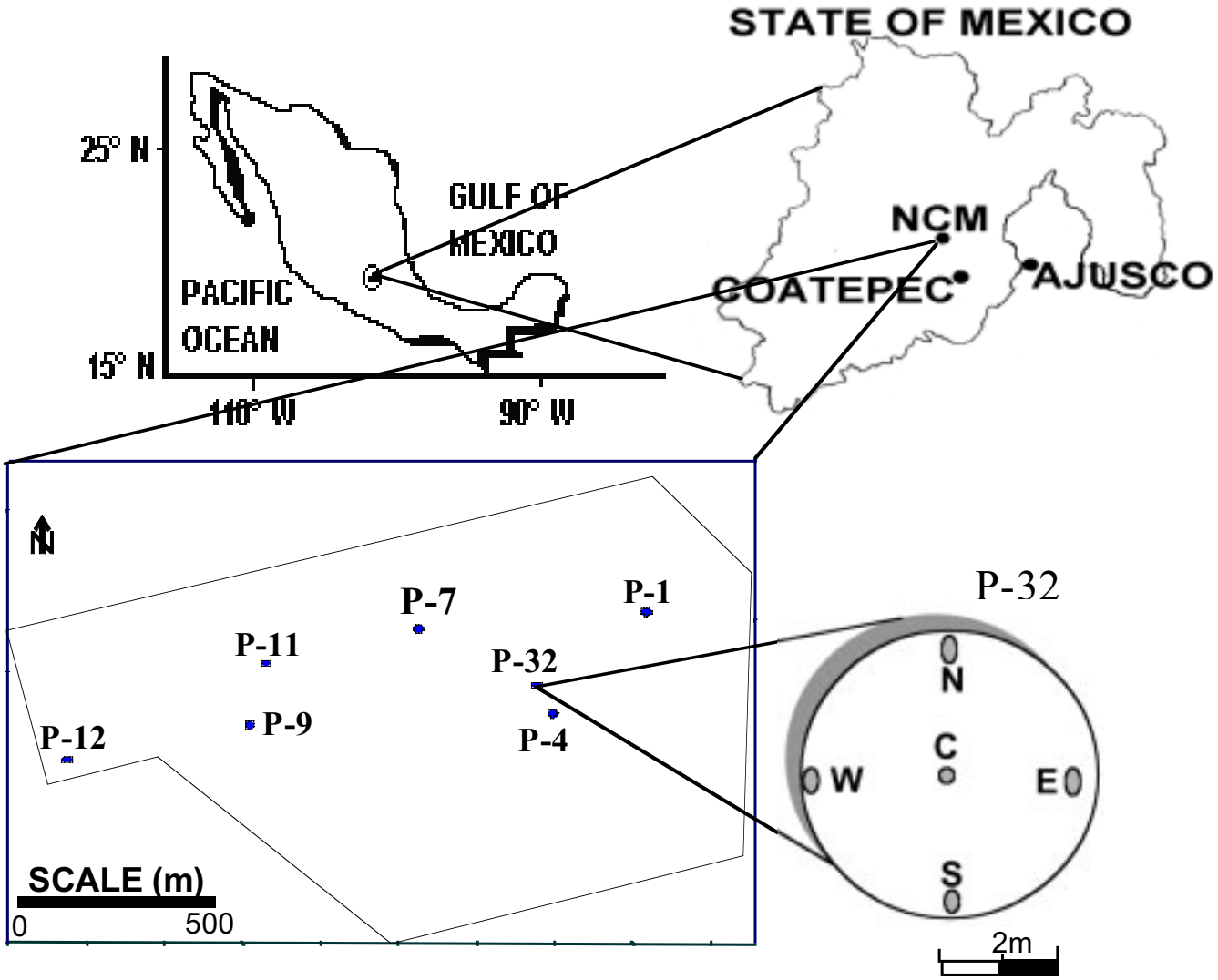

Fig. 1. Location of the sampling sites.

3000 m in the southern part of the Mexican Neovolcanic Belt. The geology of the region shows igneous rocks, basically andesite and basalt including associated pyroclastic material. The NCM is found in a small valley with a regional N-W inclination, it is a seminatural ecosystem in a temperate zone where the dominant vegetation type is coniferous mixed forest with pines, junipers spruces and oaks (Gaso *et al*., 1998). At the NCM several nuclear research facilities are located in a 1.5 km^2 forest area.

Surface soil samples were obtained at the NCM from the location where the punctual contamination was found, P-32, at 5 fixed monitoring stations (North (N), South (S), East (E) West (W) and Central (C)), and at 6 additional stations distributed over the site. Samples were also taken at the two reference sites (Ajusco and Coatepec hills) (Figure 1). The soil samples were measured for ^{137}Cs, ^{226}Ra, ^{40}K and ^{235}U specific activities (Bq kg^{-1} dry weight), by low background spectrometry with a 29.7% relative efficiency HPGe detector, Princeton Gamma Tech., Model N-IGC 29. Appropriate standard mixtures of γ-ray emitting isotopes were used to calibrate the detector (Quintero *et al*., 1996). The geometry used was a 500-ml Marinelli beaker. The analysed gamma spectrometry data correspond to a period of 5 years sampling (N=160). The natural background range of the zone was obtained from 10 years measurements performed at 10 localities surrounding the NCM (Gaso *et al*., 2000).

Soil ^{222}Rn was monitored at the NCM, from February to July 2001, at 70 cm depth at the same stations where soil samples were obtained. At P-32, the monitoring was performed at 70, 40 and 20 cm depth. Solid State Nuclear Track Detectors (SSNTD) LR 115 Type II (Dosirad Co., France) were used for ^{222}Rn determination. The detectors were exposed in the field for 30 days periods. They were then etched and spark counted according to a routine procedure previously described (Segovia *et al*., 1997).

RESULTS AND DISCUSSION

The ^{40}K, ^{137}Cs, ^{226}Ra and ^{235}U average values in the soil samples from NCM and reference sites are shown in Figure 2. Each measurement is the average value of all available samples and the minimum and maximum error values associated, are also shown. In this figure, the recommended screening limits (RSL) (NCRP, 1999) are also displayed. As indicated in this last cited report, the RSL in the soil for a scenario that includes open fields and forested sites are 150 Bq kg^{-1} for ^{137}Cs; 4.1 Bq kg^{-1} above the local background for ^{226}Ra and 280 Bq kg^{-1} for ^{235}U. The RSL are intended to insure that, if the exposure is from a single site, the dose to the maximally exposed individual, or for a critical group, should not exceed 0.25 mSv y^{-1}.

Figure 2 indicates that ^{40}K was within the natural background in all monitoring stations. On the other hand, ^{137}Cs, ^{226}Ra and ^{235}U had values within the range of the normal background of the zone in all stations except P-32.

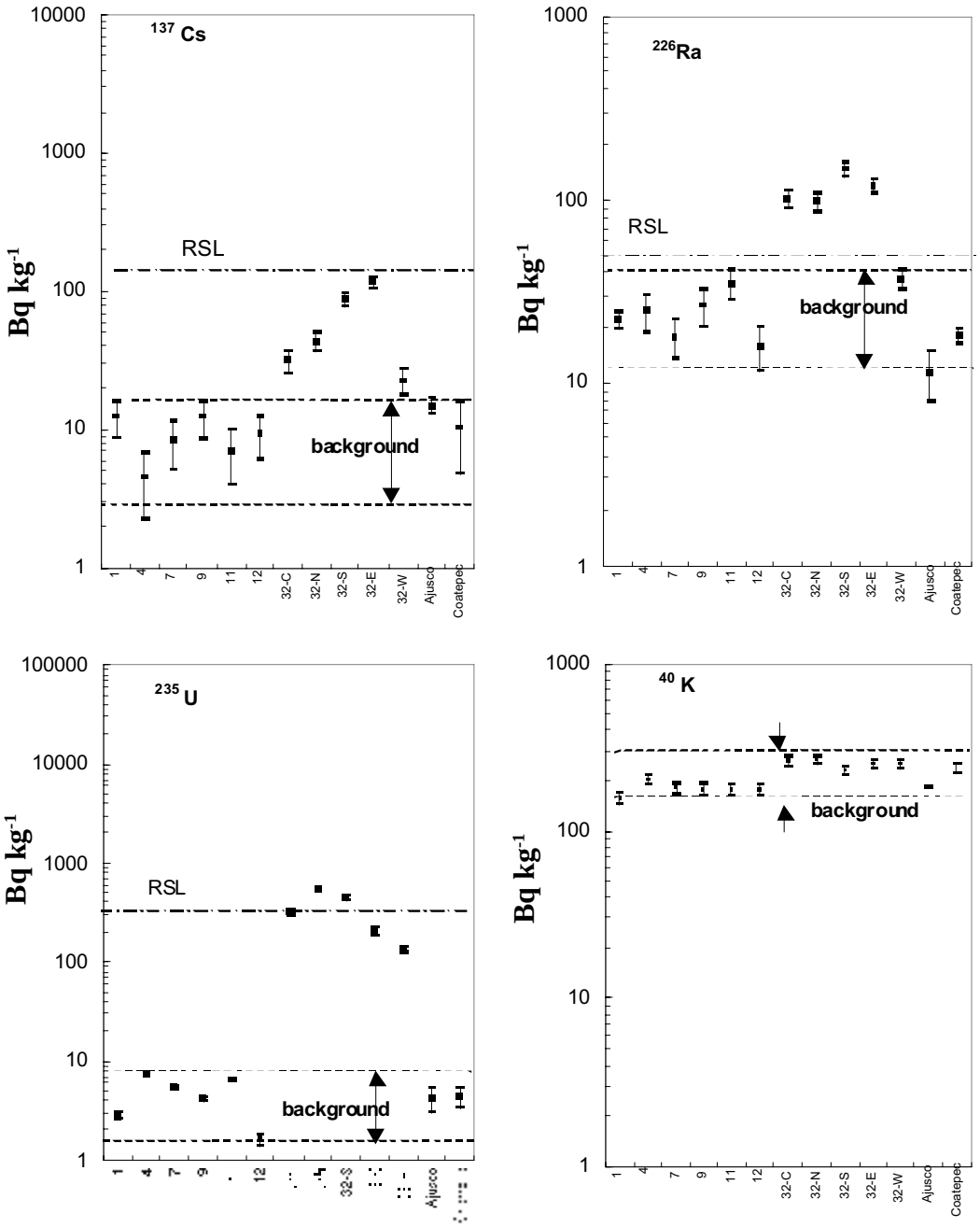

Fig. 2. ^{137}Cs, ^{226}Ra, ^{235}U and ^{40}K concentration values at NCM, Ajusco and Coatepec. The recommended screening limits (RSL) for each radionuclide are also shown. The background range and the errors for each measurement (see text) are included.

At this particular location (P-32) the surface soil ^{137}Cs values were below the RSL, but higher than the normal background. However, the ^{226}Ra and ^{235}U values were higher than the RSL for the corresponding radionuclides. These last values confirm the mentioned moderate punctual contamination of the local P-32 area soil generated from an unplanned release.

The data obtained in the last five years at the two reference sites, Ajusco and Coatepec (Figure 2), were within the natural background range of the zone for all the gamma emitters studied.

Several chemical studies of surface soil samples (6 cm) have shown (Gaso *et al*., 2000) that the average concentration value for stable Cs content at the NCM was 0.96 ± 0.05 mg kg^{-1} (dry weight) and 1.13 ± 0.06 mg kg^{-1} (dry weight) at the reference stations, Ajusco and Coatepec. Recent measurements of stable Cs at 30 cm depth in the soil from several NCM monitoring stations showed values slightly higher, 1.8 ± 0.09 mg kg^{-1} (dry weight), confirming a natural origin of stable Cs from the local geology.

The six months average soil radon concentration, at 70 cm depth, measured at the monitoring stations of the NCM, are shown in Figure 3a. The radon levels were generally low. The values were relatively stable for each station during the monitoring period. The highest radon values correspond to station 12 that is located out of the tree region in a grass land, in a small valley having the lower altitude of the zone and a local geology formed by high porosity alluvial layers, probably favouring a higher radon circulation in the soil. The ^{222}Rn values in the soil obtained in the present study agree with measurements previously reported at the NCM (Segovia *et al*., 1989), and do not show any anomalous behaviour.

Average values of the soil radon from P-32 as measured at 20, 40 and 70 cm depth, are shown in Figure 3b. The pattern for each one of the five monitoring spots was similar having higher values at 70 cm depth, as expected in a normal soil radon diffusion and transport pattern where the radon concentration decreases when approaching the surface. When compared with the other radon monitoring stations measured at 70 cm depth (Figure 3a), P-32 had radon values among the lowers. It is worth mentioning that this result indicates that the ^{226}Ra punctual contamination of the local P-32 soil was quite superficial and low level, since no effect was observed in the radon values behaviour at depth, nor in the average radon level at stations P-32 as compared with all the other radon monitoring stations at the site.

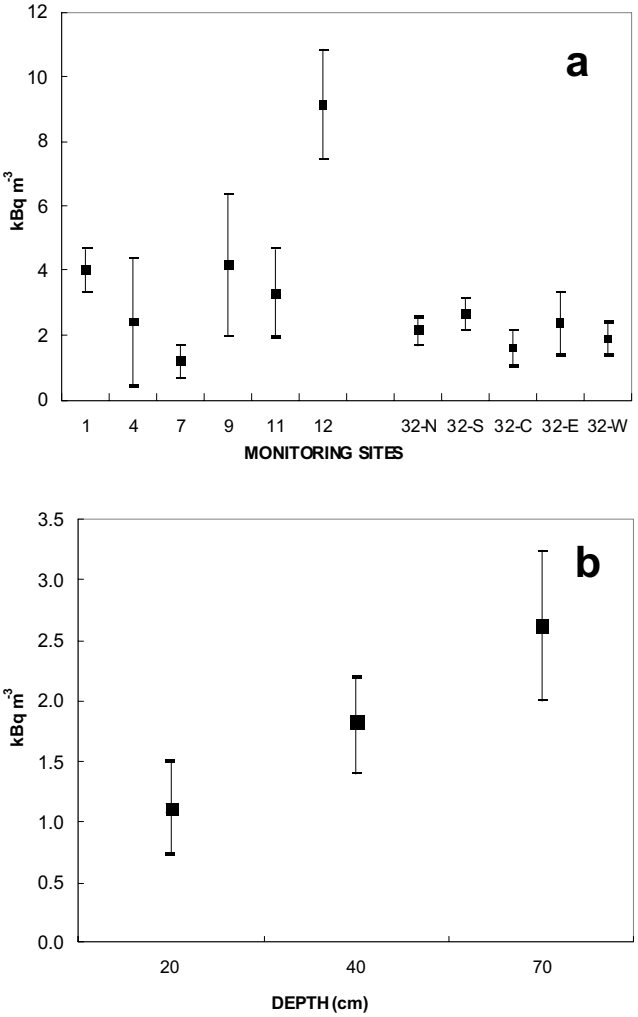

Fig. 3. a) Six months average soil radon concentration (70 cm depth) at the NCM monitoring stations. b) Average soil radon concentration values at 20, 40 and 70 cm depth at P-32.

CONCLUSIONS

The specific activities of ^{40}K, ^{137}Cs, ^{226}Ra and ^{235}U in the surface soil samples were in general below the recommended screening levels for contaminated surface soil in open fields and forest sites. However ^{226}Ra and ^{235}U at the contaminated area of the NCM, showed values higher than the recommended screening limits. The ^{222}Rn values in the soil had no anomalous behaviour.

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