Activity concentration of NORM and $^{137}$Cs radionuclide in soil samples from the Andes Cordillera at latitude $33^\circ 56'$ South

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Abstract. Activity concentrations of naturally occurring radioactive material ($^{238}$U, $^{232}$Th, $^{40}$K) and $^{137}$Cs were determined in surface soil samples collected from four sites at latitude $33^\circ 56'$ South and at an average altitude of 1650 msl in the Andes Cordillera, Chile. The handling of samples and the gamma spectroscopic analyses followed the recommended international protocols for this type of work. The presence of $^{137}$Cs due to fallout remains from atomic blasts produced in the Pacific atolls in the seventies was detected only in three sites analyzed in the current study. The activity concentration of this anthropogenic radionuclide showed significant fluctuations ranging from below minimum detection activity to 21.32 Bq/kg. The ranges of activity concentrations of $^{238}$U, $^{232}$Th, $^{40}$K were found to be around 15-30, 22-30 and 411-611 Bq/kg respectively. The weighted mean activities concentrations of NORM radionuclides are within those reported from literature and were then used to calculate the radiological hazard indices. Values of external absorbed dose rate ($D$), the annual outdoor effective dose ($AEDE$), the radium equivalent activity ($Ra_{eq}$), and the external radiation hazard ($H_{ex}$) index are within the accepted international ranges. These results point out that the radiation hazard due to NORM radionuclides found in soil samples from the sites studied in this work is not significant.

1. Introduction

The concern about the amount of environmental radioactivity to which the population is exposed is stimulating new research in this field. Today around the world there is broad interest in studying the radiological characterization of soils, not only to learn about concentration activity levels and associated health risks, but also to know more accurately the contribution of human activity to the environmental radioactivity [1-5]. However, in countries like Chile studies about soil inventories of naturally occurring radionuclide (NORM) and $^{137}$Cs are not only scarce but usually their objectives have been rather different. For instance, in Ref. [6] a radon and gamma monitoring program was presented which aimed to estimate population indoor exposure in dwellings and in some Chilean mines. P. Schuller et al., in Ref. [7] evaluated the applicability of a technique using $^{137}$Cs concentration to obtain spatial information on mean soil redistribution rates in Central-South Chile, obtaining similar results to those obtained by...
pedological observations. In 2010 a campaign for detection of $^{137}$Cs and $^{40}$K radionuclides was conducted along the Maipo River at altitudes from 900 to 1800 masl showing that after almost two half life since the nuclear tests were detonated in the atmosphere in the Pacific atolls, between 1966 and 1974, $^{137}$Cs is easily detected [8]. Finally, Chamizo et al. in Ref. [9] provides new data on the impact of nuclear tests performed in the Southern Hemisphere to South America soils through the study of $^{239}$Pu and $^{240}$Pu in different areas of Northern and Southern Chile. Their results showed the presence of debris from the French Tests with homogeneous $^{240}$Pu/$^{239}$Pu ratios ranging from 0.02-0.23. It is worth to mention that the geographical location of Chile, just in front of the Pacific Ocean, extending from 17° latitude South to Cape Horn at 56°, along the Andes Cordillera, favored the reception of fallout debris from those nuclear explosions occurred around 21.8° South. The results presented in this work are the first obtained within a cooperative research project between the University of Chile (UCH), the Metropolitan Technological University (UTEM), and the Chilean Nuclear Energy Commission (CCHEN). This project aims to determine activity concentrations of $^{137}$Cs and natural occurring radionuclides in surface soil samples collected in Central Chile along latitude 33° South, from the Andes to the coast of the Pacific Ocean. During the first stage of this project, soil samples from Andes Mountain, near Santiago, Chile, were collected in order to determine the activity concentration of NORM and that of the anthropogenic $^{137}$Cs radionuclide. Our long-range objective is to evaluate the contribution of these radionuclides to the radiation background in different zones of the country.

![Map of Central Chile](image1.jpg)

**Figure 1.** Area of Andes Cordillera where the samples were collected (33° South) during sampling campaign performed in March 2015

2. **Experimental**

2.1. **Study Area**

The map of Central Chile shown in figure 1 gives a general view of the locations where the soils samples were collected during a campaign performed in March 2015. In the south western extreme of South America, Chile occupies a narrow (177 km on average) and long (4,270 km) zone that extends roughly from latitude 17° South to Cape Horn at 56°. Other important feature about the geomorphology of continental Chile is the East-West topographical profile. For instance, at 33° it presents four different zones, namely, Marine Terraces, Coastal Cordillera, Longitudinal Central Valley and the Andes Cordillera. In Ref. [10] it has been established that the local relief in the Andes is due to a balance between extended fluvial erosion and glacial erosion acting as opposite agents. On the other hand, and according to the distribution of Soil
Taxonomy system (USDA) orders observed in Chile, the most probable classification of soil at the Central Andes sector would belong to two soil orders, namely Andisols and Inceptisols (see Fig. 2.88 in Ref.[11]). The sampling locations studied in this work are characterized as very thin soils (∼15 cm thick) over rock with poor vegetation and influenced by running water, thermal and gravitational processes, with an average annual precipitation in the range from 1500 mm to 3500 mm, mostly snow.

2.2. Sample collection and preparation
In this study soil samples were collected at four sites in the Andes Mountain near Santiago, Chile, at a distance of about 100 km to South-east, and extracted from holes having a surface of 30×30 cm$^2$ and depth of 5 cm at an average altitude of 1650 m above sea level. Sites were selected according to their accessibility and by showing no evidence of surface modification by human action. In each case, organic residues and stone fragments were removed from the surface. Soil samples from each site were packed in plastic bags and labeled accordingly. At the laboratory, preparation of samples for gamma spectroscopy measurements followed procedures recommended in [12], i.e., the samples were weighted and dried for 24 hours in an oven at 105 °C. Next, they were sieved with a 0.6 mm mesh screen and transferred to Petri Dishes and finally sealed for one month in order to reach secular equilibrium among the daughter products of $^{222}\text{Ra}$, $^{220}\text{Ra}$ and their short lived decay products. An average of 0.100 kg of soil was used per sample.

Figure 2. Gamma spectra corresponding to soil samples collected in two different locations at the Andes Cordillera. In the upper part, the gamma spectrum shows that anthropogenic radionuclide $^{137}\text{Cs}$ is not present in site M2 while in the lower part the gamma energy peak at 662 keV is easily detected in sample M1.
2.3. Calibration and Gamma spectrometry measurements

Gamma spectrometric measurements were carried out in the Gamma Spectrometry Laboratory at the University of Chile using a HPGe detector ORTEC (GEM-10195) with an energy resolution of 1.95 keV at 1330 keV and with a relative efficiency of 10% reported by the manufacturer. In order to reduce background radiation, the detector is placed inside a 10 cm cylindrical lead shielding with internal covers of cadmium and copper, 1 mm thickness each. Detector efficiency curve was obtained by using Certified Reference Material IAEA 447 [13]. Energy calibration of the acquisition system was frequently checked by using standard reference radionuclide sources. For quality control and quality assurance, three samples were prepared and analyzed for each one of the four sites (M1, M2, M3, and M4). After analyzing these three samples a very good agreement was found since their activity concentrations agreed within 2%. As an example, gamma spectra corresponding to soil samples collected in sites M1 and M2 at the Andes Cordillera are shown in figure 2. The gamma spectrum of M2 shows that anthropogenic radionuclide $^{137}$Cs is below the detection limit in this site while gamma energy peak at 662 keV is easily detected in sample M1. Collection times of 48 hours were set for each spectrum. The activity concentration of each radionuclide of our interest, $A_n$, was calculated from the respective full-energy peak area $C_n$ (background subtracted), after applying appropriate factors such as peak efficiency $\varepsilon(E_\gamma)$ and gamma intensity $I_\gamma$. Hence, using the dry weight ($M$) of the samples, their respective activity concentrations of NORM and $^{137}$Cs radionuclide could be expressed in activity per unit weight (Bq/kg) [14]:

$$A_n = \frac{C_n}{M \varepsilon(E_\gamma) I_\gamma \Delta t}$$  \hspace{1cm} (1)

Uncertainties were estimated by combining counting statistics, mass and calibration uncertainties. The activity concentration of the single decaying nuclei, $^{137}$Cs and $^{40}$K were determined directly by the measurement of the gamma-ray transitions at 662 keV and 1461 keV, respectively. On the other hand, the activity concentration of $^{238}$U was determined from the 609.3, 1120.3 and 1764.5 keV gamma-ray peaks of $^{214}$Bi and from the 295.2 and 351.9 keV gamma-ray peaks of $^{214}$Pb. Finally, the activity concentration of $^{232}$Th was evaluated by taking into account gamma-ray line of $^{212}$Pb at 238.6 keV, gamma-ray lines of $^{228}$Ac at 338.2, 911.2 and 969.0 keV, and gamma-ray line of $^{208}$Tl at 583.2 keV. The activity concentration obtained from each line was then combined to determine a weighted mean and uncertainty of the weighted mean ($k=1$) for the corresponding decay chain [14].

3. Results and Discussion

Typical gamma spectra corresponding to soil samples collected at sites M1 and M2 are shown in figure 2. As shown in this figure, the identified radionuclides detected in all samples mostly belong to the $^{232}$Th and $^{238}$U decay series, though the most prominent gamma-ray energy peak observed in log scale corresponds to $^{40}$K. In addition, a significant $^{137}$Cs peak to background ratio for gamma emission is clearly observed only in samples M1, M3 and M4, indicating that their $^{137}$Cs activity concentrations are greater than the detection limit of 0.1 Bq/kg.

3.1. Natural radionuclides in soil samples

Using gamma spectrometry, radioactivity of materials commonly found in the environment, like water and soil, is frequently studied to determine background levels of radiation or to assess the level of contamination as a consequence of human activity [14]. In this work the latter goal does not apply since in the Andes Cordillera human activities are rather scarce. Therefore, the results presented in this work can be used as a baseline in order to evaluate the contribution of NORM and anthropogenic radionuclides to the radiation background in different zones of the country.
Table 1. Comparison between mean activity concentrations and radiation hazard indices of NORM and $^{137}$Cs from soil samples collected at the Andes Cordillera and the mean value worldwide.

| Sample | $A_U$ | $A_{Th}$ | $A_K$ | $A_{Cs}$ | $D$ | $AEDE$ | $R_{eq}$ | $H_{ex}$ |
|--------|-------|---------|-------|----------|-----|--------|---------|---------|
|        | Bq/kg | Bq/kg   | Bq/kg | Bq/kg    | nGy/h | μSv/y  | Bq/kg   |         |
| M1     | 30.0±1.6 | 28.71±0.93 | 572±26 | 11.83±0.53 | 55.1±1.0 | 67.6±1.3 | 115.0±2.9 | 0.3108±0.0078 |
| M2     | 25.2±1.4 | 29.95±0.93 | 611±27 | MDA      | 55.25±0.94 | 67.8±1.2 | 115.1±2.8 | 0.3110±0.0076 |
| M3     | 17.33±0.96 | 21.67±0.72 | 411±19 | 21.32±0.56 | 38.22±0.89 | 46.9±1.1 | 79.9±2.0 | 0.2159±0.0055 |
| M4     | 14.53±0.79 | 23.23±0.72 | 429±22 | 0.97±0.16 | 38.62±0.79 | 47.4±1.0 | 80.7±2.1 | 0.2181±0.0057 |

Table 1 shows that in general, the activity concentration of soil samples collected from four sites at the Andes Mountain are within world range values [15]. Solid lines in figure 3(a) and 3(b) represent the weighted mean activity concentrations of $^{232}$Th and $^{238}$U respectively as measured for soil sample M3 while the dashed lines correspond to the uncertainties associated with these mean values. In the case of $^{232}$Th and $^{238}$U and $^{137}$Cs our results are generally lower than world average values while for $^{40}$K measured activity concentration data are higher. It is worth noting that, regarding NORM radionuclides, samples M1 and M2 show higher values compared to samples M3 and M4. Fallout radionuclide $^{137}$Cs does not follow this trend since its activity concentration shows significant fluctuations ranging from below MDA to 21.32 Bq/kg.

3.2. Dose Rates and Hazard Indices

In order to assess radiation hazards associated with soil samples, several quantities have been defined [15]. These quantities are calculated from $^{238}$U, $^{232}$Th and $^{40}$K activity concentration values ($A_U$, $A_{Th}$ and $A_K$) and by assuming that in each sample parent radionuclides and their decay progeny are in secular equilibrium.

The external absorbed dose rate ($D$) in air is defined for terrestrial gamma-radiation at a height of about 1 m above the ground, and it is calculated according to the following relation [16]:

$$D \left[ \text{nGy/h} \right] = 0.462A_U + 0.604A_{Th} + 0.0417A_K \quad (2)$$

In the present study, the absorbed dose for the four different locations of soil samples varied from 38.22 to 55.25 nGy/h with a mean value of 47 nGy/h, which is within the expected range, and it is lower than the worldwide mean value of 57 nGy/h. From the above dose definition ($D$), the annual outdoor effective dose ($AEDE$) is obtained by applying a conversion coefficient of 0.7 Sv/Gy. Additionally, an outdoor occupancy factor of 0.2 must be taken into account. Therefore, $AEDE$ values are calculated from [17]:

$$AEDE \left[ \text{μSv/y} \right] = D \times 0.7 \times 0.2 \times 8.766 \quad (3)$$

The world average annual effective dose equivalent ($AEDE$) from outdoor terrestrial gamma radiation is 70 μSv/y [15]. From Table 1, the effective dose range from 47.4±1.0 to 67.6±1.3 μSv/y with a mean value of around 58 μSv/y.
Since the distribution of $^{238}$U, $^{232}$Th and $^{40}$K in soil is not uniform, a common index called Radium Equivalent Activity ($Ra_{eq}$) is defined in [18] under the assumption that 370 Bq/kg of $^{238}$U produces the same gamma dose rate as 259 Bq/kg of $^{232}$Th and 4810 Bq/kg of $^{40}$K giving rise to the next equation:

$$Ra_{eq} \left[ \frac{Bq}{kg} \right] = A_U + \frac{370}{259} A_{Th} + \frac{370}{4810} A_K$$

(4)

It is observed in Table 1 that the average radium equivalent activity for soil samples studied in this work is lower than the maximum value allowed (370 Bq/kg), ranging from around 80 (M1 and M2) to 115 Bq/kg (M3 and M4), with a mean value of around 98 Bq/kg.

Every human being is continuously exposed to an external radiation field produced by natural radionuclide coming from soil, sediment and rocks. In order to assess this issue, the External Radiation Hazard ($H_{ex}$) is defined on the basis that the radiation exposure due to natural radionuclides should not surpass the permissible dose equivalent limit of 1 mSv/yr [18]:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} < 1$$

(5)

As can be seen, this value should not exceed unity in order to consider the radiation hazard as insignificant. As listed in Table 1, the $H_{ex}$ values for all soil samples studied ranged from 0.216 to 0.311 with an average value of 0.265, which is well below the limit of unity.

4. Conclusions

In this work gamma radiation from soil samples collected in the Andes Mountains near Santiago, Chile, were analyzed. Activity concentration of natural radioactive isotopes originated in the
decay series of $^{232}$Th and $^{238}$U, together with $^{40}$K, were determined. Measured values are below world average values. Average dose rates and calculated hazard index were lower than world recommended values. These results point out that the radiation hazard due to NORM radionuclides found in soil samples from the sites studied in this work is not significant. On the other hand, activity concentration values for $^{137}$Cs show significant fluctuations. $^{137}$Cs was detected in samples collected in locations M1, M3 and M4 while the activity concentration of $^{137}$Cs in samples M2 is below MDA. The intrinsic soil chemical properties and the erosion by water draining at the sites where samples were collected can explain the observed differences in $^{137}$Cs activities in samples M1, M2, M3 and M4. It was observed that soils with significant erosion present about 10-20 times less $^{137}$Cs than those with minimal erosion. These preliminary results can be used as a baseline for information in the area under study and motivates further research.

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