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Occurrence of Radioactive Materials in Pyroclastic Flows Of Tungurahua Volcano Using Gamma Spectrometry

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Abstract. The Tungurahua volcano began its last eruptive process in 1999 and has not ceased its activity since then, as a result of this, tons of volcanic material has been deposited on its flanks in each episode of greatest activity. An exploratory study of radioactivity of natural origin has been carried out in the pyroclastic flows near the Palma Urcu ravine (Juive) of the Tungurahua volcano using gamma spectrometry. This pyroclastic material corresponds to the accumulation that occurred after the eruption of August 2006 in an area adjacent to this ravine after its overflow. This material is used in construction. At present, much emphasis is placed on the health risks associated with the radioactivity present in construction materials. In this context, concentrations of activity in several samples of material have been analyzed to determine their risk, finding average values of 27.58 ± 1.99 Bq / kg for Ra-226; 30.28 ± 2.48 Bq / kg for Th-232, and 411 ± 28.77 Bq / kg for K-40. Finding an activity index of 0.38 ± 0.03 It was also calculated the equivalent radio activity, external and internal risk indexes and effective annual equivalent doses found that is bordering the maximum permissible limit recommended by control organisms.

INTRODUCTION

The Tungurahua volcano is a young andesitic stratovolcano located in the Eastern Cordillera of Ecuadorean Andes. In addition, it is one of the most active in Ecuador. It is 33 km southeast of the city of Ambato and 120 km south of Quito. It has a conical shape that rises to a maximum height of 5023 meters above sea level, has flanks with slopes between 30º and 35º with a basal diameter of 12 km in the North-South direction. The volcano crater has an approximate size of 400 meters in diameter and 100 meters deep. The surrounding topography between 2000 and 3000 meters elevation.[1] It has experienced pyroclastic flow-forming eruption in 1773, 1886, 1916–18 and 2006–08, in mid-1998, the Tungurahua volcano enters a stage of reactivation. In October 1999 its intensity is accentuated with a considerable increase in micro-seismicity and the production of large amounts of water vapor, ash and pyroclastic flows, similarly in July and August 2006 a reactivation of the eruptive process, with eruptions that generated the release of large pyroclastic flows, one of the main pyroclastic flows caused the overflow of Palma Urcu ravine (Juive)[2], where there was a significant accumulation of material that buried several houses and structures such as the monument of the birds at the entrance to the city of Baños, at this time the deposits generated in the sector of Juive by the Tungurahua volcano are potential mines of building material that are exploited daily, it is important to carry out a study of the main radionuclides that may be present , when these materials are used for construction are potential health risks in closed places, this study presents a gamma spectrometry analysis of 5 points strategically located in the Juive sector as you can see in the Fig. 1.

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MATERIALS AND METHODS

Sampling

Five composite samples of 3kg (formed by 5 subsamples within a $1m^2$ of area) were collected in a superficial way of pyroclastic flow of 5 to 10 cm depth in the Juive sector located at S1°40`90`` of latitude and O78°47 `01`` of longitude on the slopes of the Tungurahua volcano. The samples were dried at 60 ° C and sieved (sieve of <850µm) as previous treatment before being placed in Marinelli plastic containers (1L volume). The samples were hermetically sealed in the Marinelli to reach the secular equilibrium. The samples were analyzed in the laboratory for the radioactive environmental monitoring of SCAN (Subsecretaría de Control y Aplicaciones Nucleares) by means of gamma spectrometry of coaxial detector of HyperPure germanium (HPGe).

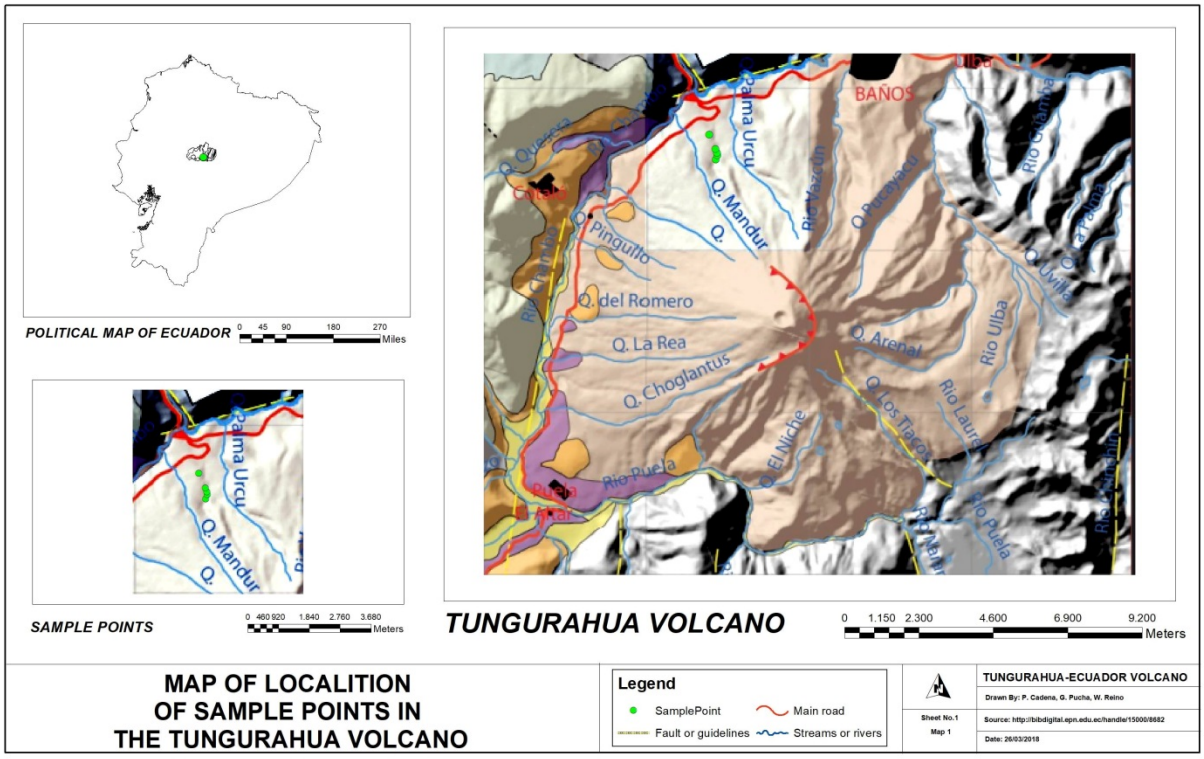
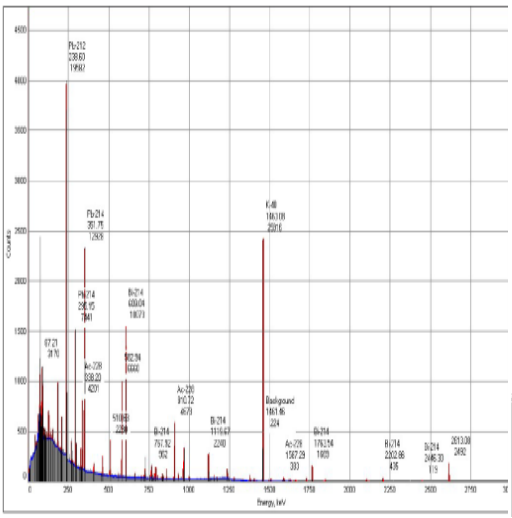


FIGURE 1. Localization of the piroclastic flow samples at Tungurahua volcano.

Determination of gamma emitters with hyperpure germanium spectrometer

To carry out this research, a GCD-40180X gamma spectrometer with lead shielding and cooled by liquid nitrogen was used, the main data of the equipment are detailed in the Fig. 2. The gamma spectrometer has calibrated in energy by means of a multinúclidos standard (provided by the Czech Institute of Metrology), the geometry used in this research is Marinelli type obtaining the same geometry used in the calibration, and therefore it is not necessary to make any geometry correction.



N	Parameter Name	TS Value	Actual Value
1.	Gamma – ray energy range, keV	5-10000	5-10000
2.	Energy resolution* for energies, eV, no more than: 122 keV 1332 keV ***total system resolution for a source at 1000 counts/s measured in accordance with ANSI/IEEE Std. 325-1996, using multichannel analyzer MCA-527, GBS Ltd.***	 <875 <1800	 814 1774
3.	Relative efficiency for energy 1,33 MeV to (NaI)Tl, % (Co-60 source mounted 25cm above the detector) Peak/Compton ratio, no less than Peak parameters: FW.1M/FWHM, no more than FW.02M/FWHM, no more than	40 63:1 1,90 2,65	41.2 64:1 1,87 2,50
4.	**Optimal operating voltage of SCD (HV), V, positive**	**1000-4000**	**2500**
5.	Conversion factor, no less than, mV/MeV	220	225
6.	Output signal polarity	negative	
7.	Output signal voltage rise time, μs, no more than	0.15	0,15

1.	Sensitive area: diameter, mm depth, mm thickness of front contacts, μm	 61.4 61.4 ~0.3
2.	Carbon fiber end cap thickness, mm	0,8
3.	Spacing between detector face and end cap window, mm	8
4.	Maximum energy rate, MeV/sec, no less than	200 000
5.	Conversion function integral nonlinearity, % not more than	0,04
6.	PA power supply	±12V
7.	Preamplifier output resistance, Ohm	50+/-1
8.	Connective cables length, m	3
9.	Cooling time, hours, not over	8
10.	Dewar vessel volume, l	30
11.	Period between liquid nitrogen refilling , days, no less than	15
12.	Overall dimension of the component part of the spectrometer, mm, no more than: - Detection unit with lead shield and support table - MCA-527	 629×539×1344 164x111x145
13.	Weight of the component parts of the spectrometer, kg, no more than: - Detection unit (at empty Dewar vessel) - MCA-527 - Lead shield with support table	 20.00 0.840 750.0

(a) (b)

FIGURE 2. (a) Laboratory of gamma spectrometry and characteristic spectrum, (b) technical parameters of the gamma spectrometer.

The material samples were hermetically stored for four weeks so that ^{226}Ra reaches secular equilibrium with its progeny. The concentration of activity of ^{226}Ra has been determined through the photopeaks of their daughters, such as ^{214}Bi and ^{214}Pb. From the ^{214}Bi the 609 keV photopeak was used with a gamma intensity of 45.49%, the photopeak of 1120 keV with an intensity of 14.92% and that of 1764.5 keV with an intensity of 15.30%. The photopeak of 295 keV with a gamma intensity of 18.42% and the photopeak of 352 keV with an intensity of 35.60% were used for the ^{214}Pb .[3]

For the ^{232}Th the photopeaks of his daughters were used: ^{228}Ac and ^{212}Pb. From the ^{228}Ac, the 338 keV photopeak was used with a gamma intensity of 11.27%, the photopeak of 911 keV with an intensity of 25.8% and that of 969 keV with an intensity of 15.8%. From the ^{212}Pb, the 238.6 keV photopeak with a gamma intensity of 43.6% was used. The ^{40}K was measured directly using its gamma emission photopeak of 1461 keV with an intensity of 10.66%.[4]

The activity concentrations of each emission line were obtained from equation (1). Where N is the accounts of each photopeak subtracted the respective radioactive background, T is the live time of measurement in seconds, which in this case for each sample was 14399 s. ε_γ is the detection efficiency of each photopeak. I_γ is the gamma intensity of each photopeak and W is the mass of the sample[5].

$$A = \frac{N}{T * \varepsilon_\gamma * I_\gamma * W}(Bq/kg) \quad (1)$$

RESULTS AND DISCUSSION

Determination of activity concentration indexes

The European Commission Guidance proposes an activity concentration index that considers the dose generated by exposure to building materials, which is expressed in equation (2). Where $A_{Ra,}$ $A_{Th\ y}$ A_K are the activity concentrations (Bq / kg) that are derived from the equation (1).

$$I = \frac{A_{Ra}}{300(Bq/kg)} + \frac{A_{Th}}{200(Bq/kg)} + \frac{A_K}{3000(Bq/kg)} \quad (2)$$

TABLE 1. Concentration of activity for each radioisotope and its respective activity concentration index

Sample ID	A	B	C	D	E	Average
Weight(g)	1282.67	1279.99	1268.96	1268.96	1245.25	1269.17
226Ra(Bq/kg)	24.25±1.69	33.25±2.66	27.69±1.94	26.45±1.85	26.25±1.84	27.58±1.99
232Th (Bq/kg)	29.15±2.3	30.6±2.4	30.95±2.79	29.4±2.35	31.3±2.5	30.28±2.48
40K (Bq/kg)	393±27.5	410± 28.7	440±30.8	402±28.14	410±28.7	411±28.77
Activity Index(I)	0.35±0.03	0.41±0.03	0.39±0.029	0.36±0.026	0.38±0.028	0.38±0.028

The average value of radioactivity in the pyroclastic flows of the Tungurahua volcano are 27.58±1.99 (Bq/kg) for ^{226}Ra; 30.28±2.48 (Bq/kg) for ^{232}Th and 411±28.77 (Bq/kg) for ^{40}K.
In any case, the control criterion should be based on the dose range of 0.3 to 1mSv / year shown in Table 2. This indicates that the activity index of pyroclastic flow samples exceeds 0.3 mSv / year and should be monitored regularly. On the other hand, our activity index complies with the I≤1 parameter, so it can be used without any regulation since it does not exceed the maximum dose of 1 mSv /year.[6]

TABLE 2. Correlation between the activity index and the annual dose rate.

Dose criterion	0.3 mSv/year	1 mSv/year
Materials used in bulk amounts, e.g. concrete	I≤0.5	I≤1
Superficial and other materials with restricted use: tiles, boards, etc.	I≤2	I≤6

Determination of radio equivalent activity, dose rate and risk

Radio equivalent activity is used to calculate the radiological risk of building materials in terms of gamma radiation. According to this, it is estimated that 1 Bq/kg de ^{226}Ra; 0.7 Bq/kg of ^{232}Th and 13 Bq/kg of ^{40}K they produce the same dose of gamma radiation. The equivalent radio activity is given by the equation (3).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \leq 370 \quad (3)$$

Where, A_{Ra}, A_{Th} y A_K are the activity concentrations in Bq/kg for ^{226}Ra; ^{232}Th and ^{40}K respectively. The average radio equivalent activity obtained was 387.35±27.7 Bq/kg; which is over the maximum limit of 370 Bq/kg.[7]
Similarly, to calculate the dose rate the equation (4) was used. Where A_X (Bq/kg) corresponds to the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K respectively and C_X (nGy/hour per Bq/kq) is the conversion factor for each radioisotope (^{226}Ra=0.427, ^{232}Th=0.662 and ^{40}K=0.043).

$$\dot{D} = \sum_X A_X * C_X \quad (4)$$

Also, the equivalent effective annual dose received by the public due to exposure to building materials can be calculated using equation (5). Where the term Q corresponds to the environmental exposure of moderate energy gamma rays. T is the time of a year expressed in hours (8760 h), Q_f is the occupancy factor (0.8) and $\dot{D}$ is the dose rate obtained from the equation (4).

$$\dot{E} = T * Q * \dot{D} * Q_f * 10^{-6} \quad (5)$$

The average dose rate in air for the pyroclastic flows analyzed is given in Table 3. It corresponds to 49.49±3.73 (nGy/hour). Similarly, the average effective annual equivalent dose is 0.24±0.02 (mSv/year) which is within the maximum allowable limit given by the ICRP that is 1mSv/year.
It is also possible to evaluate the risk indexes for internal and external exposure. The external risk index (H_{ex}) is given by equation (6) and the internal risk index (H_{in}) is given by equation (7); where A_{Ra} A_{Th} and A_K (Bq/kg) corresponds to the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K respectively. The internal risk index considers exposure to radon and its short-lived progeny, which can cause a risk mainly in the respiratory organs.

$$H_{Ex} = \frac{A_{Ra}}{370(Bq/kg)} + \frac{A_{Th}}{259(Bq/kg)} + \frac{A_K}{4810(Bq/kg)} \quad (6)$$

$$H_{In} = \frac{A_{Ra}}{185(Bq/kg)} + \frac{A_{Th}}{259(Bq/kg)} + \frac{A_K}{4810(Bq/kg)} \quad (7)$$

The average values for internal and external risk indices are given in Table 3. The average external risk index (H_{ex}=0.28±0.02) and the average internal risk index (H_{in}=0.35±0.03) they are within the regulations since they must not exceed the unit[8].

TABLE 3. Radio equivalent activity, air dose rate, annual effective dose and their respective external and internal risk indices for the pyroclastic flows of the Tungurahua volcano.

Sample ID	Ra eq. (Bq/kg)	$\dot{D}_{air}$ (nGy/hour)	Annual effective dose (mSv/year)	Hazard index	
				Hex	Hin
A	368.54±26.21	46.55±3.45	0.23±0.02	0.26±0.02	0.33±0.02
B	392.71±28.26	52.08±3.99	0.26±0.02	0.29±0.02	0.38±0.03
C	410.75±29.64	51.23±4.00	0.25±0.02	0.29±0.02	0.36±0.03
D	378.03±26.88	48.04±3.56	0.24±0.02	0.27±0.02	0.34±0.02
E	386.71±27.52	49.56±3.68	0.24±0.02	0.28±0,02	0.35±0.03
Average	487.35±27.7	49.49±3.73	0.24±0.02	0.28±0.02	0.35±0.03
Requirement	370	-	1	1	

CONCLUSIONS

Analyzes of this investigation were carried out to determine the radiological environmental impact of the volcanic material coming from the pyroclastic flows of the Tungurahua volcano. One of the main points to take into account was the reuse of this volcanic material as a construction material, evaluating if there is any risk to health. The average values of radioactivity in the pyroclastic flows of the Tungurahua volcano for primordial radionuclides are: 27.58 ± 1.99 (Bq/kg) for ^{226}Ra; 30.28 ± 2.48 (Bq/kg) for ^{232}Th and 411 ± 28.77 (Bq/kg) for ^{40}K. The global average for construction materials is 50 (Bq/kg) for ^{226}Ra; 50 (Bq/kg) for ^{232}Th and 500 (Bq/kg) for ^{40}K. This indicates that our calculation of concentration of activity for radionuclides gives us values lower than the world average. For the activity index, $I \leq 1$ but $I \geq 0.5$, which generates a dose rate lower than 1 mSv/year but values of 0.3 mSv/year can be reached. The average radio equivalent activity found was 387.35 ± 27.70 Bq/kg, which exceeds the maximum limit that is 370 Bq/kg. As for the internal risk (0.35 ± 0.03) and external (0.28 ± 0.02), they are below the maximum.

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