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NORM determination in urban soils from selected cities in Chihuahua, Mexico

Determinación de NORM en suelos urbanos de ciudades seleccionadas en Chihuahua, México

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ABSTRACT

Soil radioactivity affects the human body. This study determinates the ^{238}U, ^{232}Th and ^{40}K activity (NORM) in 164 urban soils in 13 cities in the State of Chihuahua, Mexico. Absorbed Dose Rate and the Annual Effective Dose were also calculated. Activities were analyzed using a coaxial hyper-pure germanium detector (HPGe), Ge Model 2020, Thermo and Dewar 7500 SL (CANBERRA brand). A one-way variance analysis (ANOVA) was performed and a Tukey test was applied to identify mean differences. Activity averages were 36.8 Bq kg^{-1} for ^{238}U, 41.4 Bq kg^{-1} for ^{232}Th and 805.7 Bq kg^{-1} for ^{40}K. Calculated absorbed dose rate ranged from 36 nGy h^{-1} to 83 nGy h^{-1} while Annual Effective Dose varied from 1.02 mSv y^{-1} to 0.44 mSv y^{-1}. Results suggest that some levels exceed global averages and are among the highest levels of natural radioactivity in the world.

RESUMEN

La radiactividad del suelo afecta al cuerpo humano. Este estudio determinó la actividad del ^{238}U, ^{232}Th and ^{40}K (NORM) en 164 suelos urbanos de trece ciudades del estado de Chihuahua, México. Se calculó la tasa de dosis absorbida y de dosis efectiva anual. Se usó un detector de germanio hiperpuro (HPGe), modelo Ge 2020, Termo y Dewar 7500 SL marca CANBERRA. Análisis de Varianza (ANOVA) y una prueba de Turkey fueron corridas para detectar las diferencias estadísticas. Los promedios de actividad fueron de 36.8 Bq kg^{-1} para ^{238}U, 41.4 Bq kg^{-1} para ^{232}Th y 805.7 Bq kg^{-1} para 40K. La tasa de dosis absorbida estuvo en un intervalo entre 36 nGy h^{-1} a 83 nGy h^{-1}, mientras la dosis efectiva anual varió de 1.02 mSv y^{-1} a 0.44 mSv y^{-1}. Los resultados muestran que algunos valores exceden los promedios mundiales y que están entre los más altos valores mundiales.

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Radioactivity; soil; Chihuahua State; dose.

Palabras clave:
Radiactividad; suelo; estado de Chihuahua; dosis.

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INTRODUCTION

Soil contains radioactive elements such as uranium (U) and thorium (Th) series elements, and potassium-40 (^{40}K), which are known as naturally occurring radioactive materials (NORM) (Kathren, 1998).The most important radioactive isotopes in soils are Uranium-238 (^{238}U,) Thorium-232 (^{232}Th) and Potassium-40 (^{40}K). ^{238}U and ^{232}Th generate many other radioactive isotopes in soils because they are the beginners of radioactive series (Bassioni, Abdulla, Morsy & El-Faramawy, 2012; Topcuoğlu, Türer, Güngör & Kırbaşoğlu, 2003). Potassium is the 7th most abundant element in the earth's crust, representing approximately 2.4% of its total weight. Most of the K is not radioactive, only the mass 40 isotope is radioactive and it corresponds to 0.0117% of natural potassium (Jabbar *et al.*, 2010).

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has published the world average activity for ^{238}U, ^{232}Th and ^{40}K in soil: 35 Bq kg^{-1}, 25 Bq kg^{-1}, and 370 Bq kg^{-1}, respectively (UNSCEAR, 2000). Moreover, many countries have published their own averages (Bassioni *et al.*, 2012; Dołhańczuk-Śródka, 2012; García-Talavera,

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Matarraz, Martínez, Salas & Ramos, 2007; Kathren, 1998; Mehra, Singh, Sing & Sonkawade, 2007; Topcuoğlu *et al*, 2003; UNSCEAR, 2000).

This world average converted into dose values of 55 nGy y^{-1} for Absorbed Dose Rate (D) and 0.48 mSv y^{-1} for Annual Effective Dose (AED), where the AED (0.48 mSv y^{-1}) is about 16% of the total dose average that any human body receives from the soil (Cancio-Pérez, 2010; UNSCEAR, 2008). In addition, the International Commission on Radiological Protection (ICRP) has recommended an annual maximum effective dose of 1 mSv y^{-1} for individuals (Jabbar *et al*., 2010; ICRP, 1993; Mehra *et al*., 2007). This is important because radioactivity can affect human health depending on the level of exposure. It is well known that bone marrow and the blood system are the most radiosensitive parts of the human body and can even be affected by low doses. Genitals are also quite sensitive to radiation. For example, people can suffer temporary sterility if they receive sufficiently high radiation, while prolonged exposure can result in permanent sterility. The lens of the eye is also highly vulnerable and radiation can cause cataracts and opacities after a certain period of exposure (Cancio-Pérez, 2010; Kathren, 1998).

Mexico uranium deposits are not commercial grade. The State of Chihuahua is considered the most important uraniferous site with 50 natural zones with about 30% of all the uranium of Mexico (Bazán-Barrón, 1978; Burciaga-Valencia *et al*., 2010; Colmenero-Sujo *et al*., 2004; Domínguez, Hernández, Arango & Medina, 2006; Rentería-Villalobos *et al*., 2007; Villalba *et al*., 2006; Villalba, Colmenero-Sujo & Montero, 2012).

Figure 1 shows the 12 most important uraniferous zones; some of them are near (underled uraniferous zones) large cities. There is little information about radioactivity and absorbed dose rates in Mexico. Therefore, there is no baseline for possible human risk and the potential effects of specific doses on human health have not been calculated (Bazán-Barrón, 1978; Domínguez *et al*., 2006).

One objective of this study was to determine the specific activity of ^{238}U, ^{232}Th series and ^{40}K (NORM) in urban soils in 13 important large cities of Chihuahua State, Mexico. A second objective was to calculate the Dosis Range (D) and the Annual Effective Dosis (AED) that might affect Chihuahua's inhabitants. This information should be helpful for establishing preventive health measures.

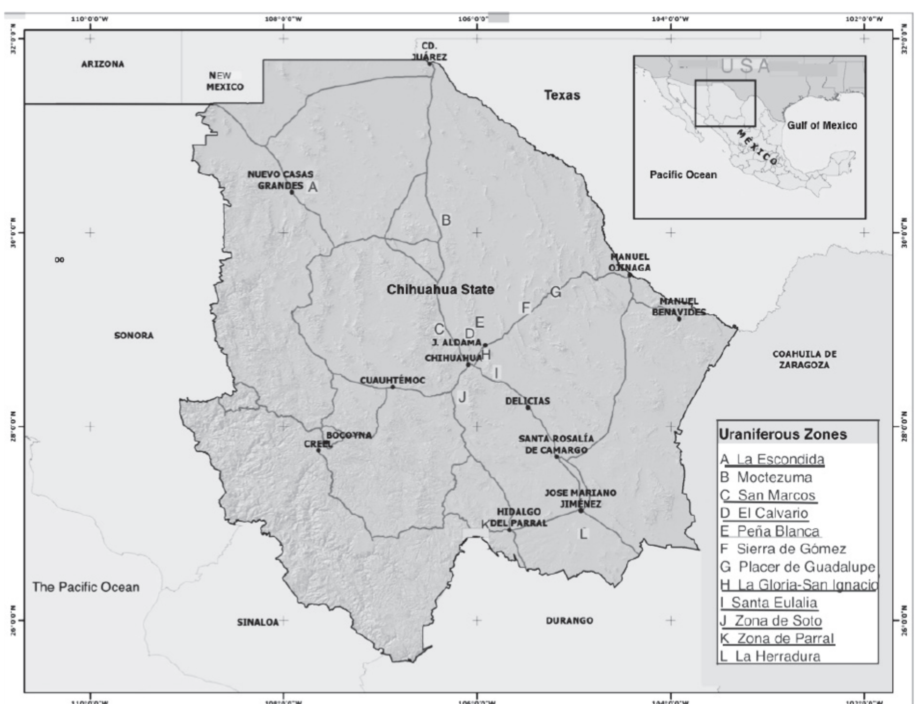

Figure 1. Main cities of Chihuahua State and its major uraniferous zones.
Source: Villalba *et al*. (2006).

MATERIALS AND METHODS

Study area and Activity

The State of Chihuahua is located in northern Mexico (figure 1), and has a total population of 3 406 465 (*Instituto Nacional de Estadística, Geografía e Informática* [INEGI], 2010). Geologically, Northern Mexico is composed mainly of Mesozoic and Cenozoic sedimentary rock, as well as recent continental deposits. Natural uranium is found in Chihuahua in acidic extrusive igneous and rhyolitic and andesitic rock. Some cities are close to known uraniferous zones (Domínguez *et al.*, 2006; INEGI, 2010).

The study was carried out in 13 cities (figure 1). A total of 164 samples were collected (table 1). The sampling employed was the one used by the Radiation Monitoring Network in Spain according to the ReViRa (for its acronym in spanish) Manual from Spain (ReViRa, 1994), and mentioned in other works (Blanco, Vera-Tomé & Lozano, 2005; Colmenero-Sujo *et al.*, 2004; Rentería-Villalobos *et al.*, 2007). This method use a square steel structure, 50 cm length and 5 cm depth, which was placed on the ground to define the soil sample (approximately 12.5l). Large stones and other objects were removed. Two liters of soil were taken to the laboratory, where they were powdered and sieved through a 2 mm mesh. Finally, one liter was packed into a Marinelli beaker and sealed, and then allowed to stand for at least 4 weeks, so that ^{238}U series was able to reach radioactive secular equilibrium.

The activity of different isotopes was analyzed by a gamma spectroscopy using a coaxial hyperpure germanium detector (HPGe), Ge Model 2020, Thermo and Dewar 7500 SL (CANBERRA brand), which belongs to the Centro de Investigaciones en *Materiales Avanzados* (CIMAV-Chihuahua). The detector is cylindrical, 45-mm high and 44 mm in diameter, with an active volume of approximately 80 cm^3. The efficiency ratio of the NaI (TI) 3' × 3' detector is 20%. As a measure of quality control, the resolution was checked by determining the full-width at half-maximum (FWHM) for the 1332.47 keV peak of ^{60}Co, it was did daily with a source SRM4203D-65 of National Institute of Standards and Technology (NIST) whose value was 2.0 keV.

To calculate the series of ^{238}U activity, we used the lines of 351 keV of ^{214}Pb and 609 keV of ^{214}Bi. The series of ^{232}Th activity was obtained from lines ^{238}keV of ^{212}Pb and 912 keV of ^{228}Ac. For the particular case of ^{40}K, we used its unique line of 1460 keV. Every sample was read for 22 h (Bassioni *et al.*, 2012; Burciaga-Valencia *et al.*, 2010; Mehra *et al.*, 2007).

Table 1.
Characteristics and samples for the selected cities.

City	North latitude	West longitude	High (mosl)*	Inhabitants**	Type of rocks***	Samples
Aldama	28 50	105 55	1270	19 378	ird	15
Bocoyna	27 51	107 35	2240	27 907	ird	5
Camargo	27 41	105 10	1220	45 852	sll	5
Creel	27 45	107 38	2345	5026	ird	8
Cuauhtémoc	28 24	106 52	2060	124 378	ird	9
Chihuahua	28 38	106 04	1440	671 790	ird	30
Delicias	28 11	105 28	1170	116 426	ird	8
Jiménez	27 08	104 55	1380	38 323	ird	10
Juárez	31 44	106 29	1140	1 218 817	sll	20
Manuel Benavides	29 06	103 54	1060	1746	sll	5
Nuevo Casas Grandes	30 25	107 54	1460	54 390	ird	12
Ojinaga	29 34	104 24	800	24 307	sll	8
Parral	26 56	105 40	1720	100 821	ird	29
Total				2 449 161		164

* meters on the sea level; ** INEGI (2010); *** description Uraniferous rocks; ird = acid igneous/rhyolite/dacite; Little uraniferous rocks; sll = sandstone/ limestone/limonite
Source: Author's own elaboration.

A rigorous process of gamma ray testing was carried out using standards to calibrate the HPGe equipment with the objective of assuring consistency. These samples were analyzed, once more by other institutions such as the *Centro Regional de Estudios Nucleares* of the *Universidad Autónoma de Zacatecas*, Mexico (CREN-UAZ), and *El Colegio de Física Atómica y Nuclear* of the *Universidad de Sevilla* in Spain, corroborating that the values obtained in this study were reliable.

Statistical Analysis

A one-way Analysis of Variance (ANOVA) was performed for each variable and if statistical differences were obtained a Tukey test was used to separate means that using 0.01 as a level of significance ($\alpha = 0.01$). MINITAB 15 software was used to analyze the data (Rubio & Jiménez, 2012).

Absorbed Dose rate (D) and Annual Effective Dose (AED) calculations

The next equations were used to calculate dose rate (d) from terrestrial radiation. It is measured at 1 m above the ground (Bassioni *et al.*, 2012; Dołhańczuk-Śródka, 2012):

$$D\ (\text{nGy h}^{-1}) = (0.462\ C_U + 0.604\ C_{Th} + 0.0417\ C_K).$$

Where C_U, C_{Th} and C_K represent the specific activities of each isotope in Bq kg^{-1}.

The annual effective dosis (AED) in mSv y^{-1} was obtained:

$$AED = D\ (\text{nGy h}^{-1}) \times 8760\ \text{h} \times 0.2 \times 0.7\ (\text{Sv Gy}^{-1}\ \text{y}^{-1}) \times 10^{-6}.$$

RESULTS

Table 2 shows the mean values, standard deviation and minimum and maximum values for ^{238}U, ^{232}Th and ^{40}K found in the soils of the 13 cities evaluated. Concentration of ^{238}U exceeded the global average (35 Bq kg^{-1}) in eight of the cities: Aldama, Bocoyna, Creel, Cuauhtémoc, Chihuahua, Delicias, Jiménez y Nuevo Casas Grandes. This agrees to the type of rock around these cities that have acid igneous, rhyolite and dacite rocks. The highest concentration was found in Aldama with 51.9 Bq kg^{-1}, while the minimum value was in Juárez with 19.8 Bq kg^{-1}. This last city is according to the type of rock it has around, sedimentary rocks like sandstone, limestone and limonite (table 2 and figure 2).

The ANOVA analysis for this isotope found statistical differences among the cities ($P < 0.01$). Besides, table 2 shows the results of the Tukey's test, which found three groups of cities. With respect to ^{232}Th ANOVA analysis found statistical differences among the cities ($P < 0.01$). It is noted that the concentration of this isotope exceeded the world average (35 Bq kg^{-1}) in nine of the evaluated cities. The highest value was in Aldama with 61.1 Bq kg^{-1}, while the lowest value was in Juárez with 20.0 Bq kg^{-1} (lowest again). This is the same case of ^{238}U, the difference founded between cities are the type of rocks around these cities. The mean and all values in the 13 cities were high in ^{40}K. The cities of Aldama and Nuevo Casas Grandes had the highest values with 1014.5 Bq kg^{-1} and 1013.7 Bq kg^{-1}, respectively, while the lowest value was in Juárez with 570.7 Bq kg^{-1}. Moreover, the ANOVA analysis detected significant differences among cities concerning the isotope ^{40}K.

In Chihuahua's cities, in general, there are two kinds of cities according of kind of rocks, ten cities have uraniferous rocks around; Aldama, Bocoyna, Camargo, Creel, Cuauhtémoc, Chihuahua, Delicias, Jiménez, Nuevo Casas Grandes and Parral. Three cities have sedimentary rocks (no uraniferous); Juárez, Manuel Benavides and Ojinaga.

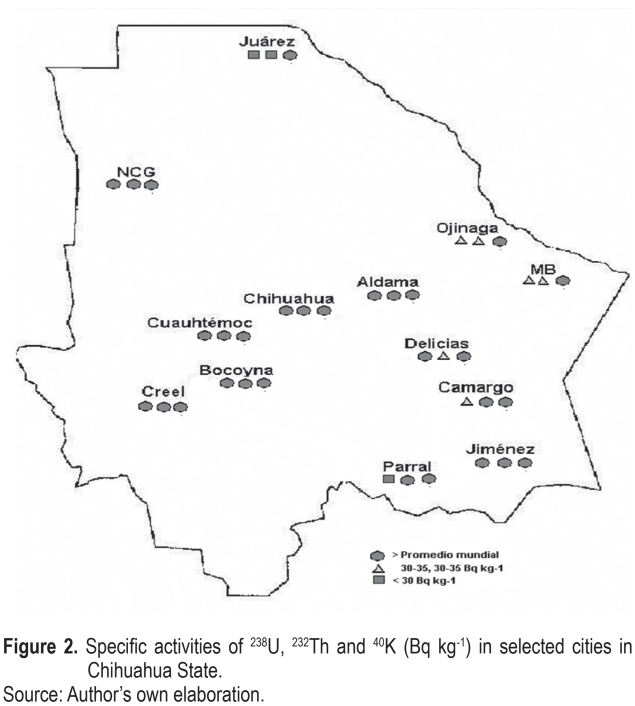

Figure 2. Specific activities of ^{238}U, ^{232}Th and ^{40}K (Bq kg^{-1}) in selected cities in Chihuahua State.
Source: Author's own elaboration.

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Table 2.
Specific activities of ^{238}U, ^{232}Th and ^{40}K (Bq kg^{-1}) in selected cities in Chihuahua State.

City	^{238}U	Min-Max	^{232}Th	Min-Max	^{40}K	Min-Max
Aldama	51.9 ± 0.3a*	44.6 - 58.2	61.1 ± 0.5a*	53.7 - 68.4	1014.5 ± 6.2a*	817.5 - 1106.3
Bocoyna	41.6 ± 0.3ab	34.0 - 47.2	48.1 ± 0.4abc	37.9 - 54.7	818.5 ± 5.8abcd	720.7 - 893.4
Camargo	34.1 ± 0.2b	30.7 - 40.1	38.0 ± 0.3c	33.1 - 44.6	827.0 ± 5.1abcd	766.4 - 909.8
Creel	35.3 ± 0.3b	29.7 - 55.1	38.5 ± 0.4c	33.3 - 55.6	663.7 ± 5.3de	552.9 - 823.4
Cuauhtémoc	37.7 ± 0.3b	32.0 - 43.3	43.9 ± 0.4bc	36.0 - 55.8	797.6 ± 5.5bcd	571.0 - 972.8
Chihuahua	39.8 ± 0.3b	28.9 - 54.6	55.2 ± 0.4ab	23.5 - 102	916.6 ± 6.0ab	653.1 - 1179.2
Delicias	36.5 ± 0.3b	31.3 - 43.6	33.4 ± 0.4c	28.5 - 38.2	772.0 ± 5.8bcde	685.6 - 848.5
Jiménez	40.4 ± 0.3b	38.1 - 45.9	42.3 ± 0.4c	38.2 - 49.5	930.5 ± 6.1ab	842.5 - 1002.8
Juárez	19.8 ± 0.2c	18.5 - 23.0	20.0 ± 0.3d	15.3 - 25.4	570.7 ± 2.9e	512.5 - 631.4
Manuel Benavides	34.8 ± 0.2b	27.3 - 41.1	32.9 ± 0.3cd	20.0 - 39.4	601.7 ± 4.3de	414.4 - 828.9
Nuevo Casas Grandes	41.1 ± 0.3b	37.7 - 44.9	55.9 ± 0.5ab	48.9 - 63.6	1013.7 ± 6.9a	954.0 - 1069.1
Ojinaga	31.8 ± 0.3b	27.6 - 34.1	32.0 ± 0.4cd	27.2 - 36.1	688.3 ± 4.7cde	577.9 - 742.2
Parral	34.8 ± 0.3b	18.3 - 63.3	36.6 ± 0.4c	10.3 - 63.1	859.0 ± 5.9abc	534.2 - 1796.8
Mean	36.8 ± 0.3		41.4 ± 0.4		805.7 ± 5.4	

* Means in columns that do not share a letter are statistically different.
Source: Author's own elaboration.

Absorbed Dose Rate and Annual Effective Dose

The values for D determined range from 36 nGy h^{-1} to 83 nGy h^{-1}. Nine of the 13 cities had D values equal to or higher than the global average, which is 55 nGy h^{-1} (UNSCEAR, 2008). The average D for Chihuahua State was 60.6 nGy h^{-1}.

Figure 3 shows the AED calculated for NORM for each sampled city. Twelve of the thirteen cities had levels equal to or higher than the world average (0.48 mSv y^{-1}) (UNSCEAR, 2008). Ciudad Aldama had the highest value with 1.02 mSv C while Ciudad Juarez the lowest value with 0.44 mSv y^{-1}, lower than the world average. The average AED for Chihuahua State was 0.74 mSv y^{-1}.

DISCUSSION

The three NORM means were greater than the value by United Nations Scientific Committee of the Effects of Atomic Radiation (UNSCEAR) published (UNSCEAR, 2000). Aldama had the highest values and Ciudad Juarez the lowest values. According to Jabbar *et al.* (2010) the abundance of potassium in the soil depends on its characteristics; artificially fertilized soil has high amounts of potassium because of its additive property from potassium in the fertilizers.

It is important to point out that Ciudad Aldama had the highest values of the three natural isotopes: 67% higher than the world average for ^{238}U, 48% higher for ^{232}Th and 174% higher for ^{40}K. Comparing the averages in Chihuahua to those in other countries we can see that it has higher NORM levels than Luxemburg, Hungary, Japan, Bangladesh, China, Romania and Spain, and similar values to regions like Punjab, India (Dołhańczuk-Śródka, 2012; Jabbar *et al.*, 2010; Mehra *et al.*, 2007). Chihuahua city and Chihuahua State have been mentioned in others papers for its high radioactive levels (Colmenero-Sujo *et al.*, 2004; Colmenero-Sujo & Villalba, 2010; Villalba *et al.*, 2012).

In Absorbed dose rate (D), it is important to point out that the value obtained is higher than the world average and higher than levels reported in the United States, Switzerland, Luxemburg, Japan, Romania, Taiwan and others countries (Dołhańczuk-Śródka, 2012; Jabbar *et al.*, 2010; Mehra *et al.*, 2007; Quindos, Fernández, Rodenas, Gómez-Arozamena & Arteche, 2004; Tzortzis, Tsertos, Christofides & Chistodoclides, 2003). Similar studies carried out in other regions of Mexico have reported lower values than those in Chihuahua. For example, the average D level in the State of Zacatecas is 44.9 nGy h^{-1} (Mireles *et al.*, 2003).

The *Comisión Nacional de Seguridad Nuclear y Salvaguardias* (CNSNS, 2012) of Mexico reported results in some regions of Mexico, however, the CNSNS did not refer to studies in Chihuahua State.

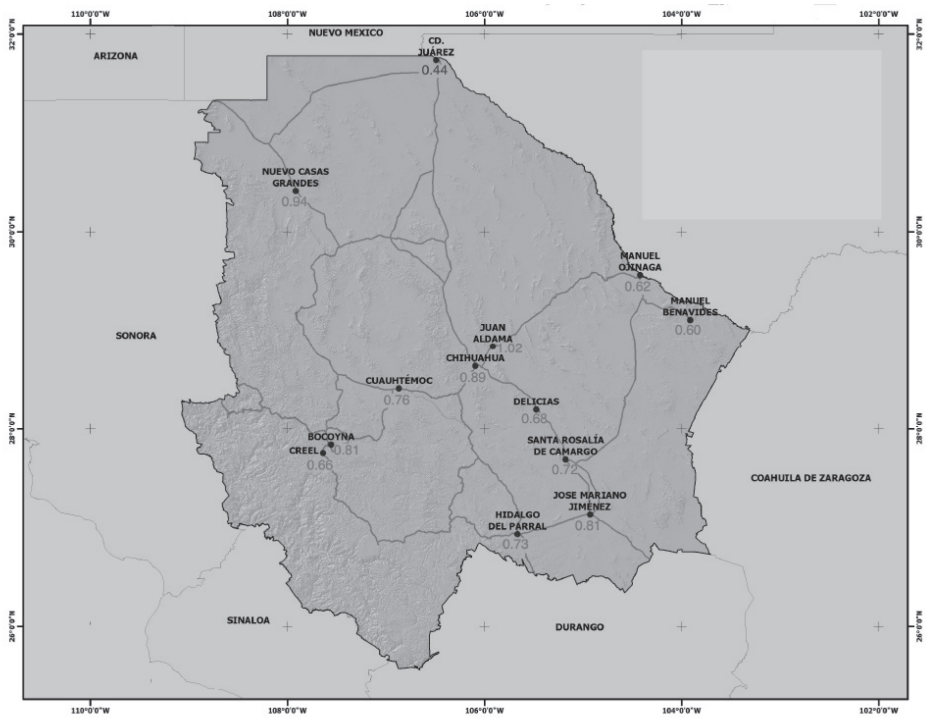

Figure 3. NORM doses in urban soils of selected cities of Chihuahua State (mSv y^{-1}).
Source: Author's own elaboration.

Some studies like Punjab, India, reported by Mehra *et al.* (2007) indicated that AED ranges from 0.28 mSv y^{-1} to 0.64 mSv y^{-1}, In some papers report that Pakistan and Czech Republic average values of 0.43 mSv y^{-1} and 0.44 mSv y^{-1} respectively have been reported (Dołhań czuk-Śródka, 2012; Jabbar *et al.*, 2010).

The ICRP (1993) should declare to Ciudad Aldama a radioactive risk zone according at this work and others.

CONCLUSIONS

Results reported here represent the radioactivity from the geology of selected cities in the Chihuahua State. Eight cities in Chihuahua had higher levels of natural radionuclides than the world average. Nine cities had dose rates equal to or higher than national and international averages, while 12 cities had annual effective doses equal to or higher than national and international averages. These cities have 72% of the inhabitants of Chihuahua State.

We recommend that the Mexican Nuclear Safety Agency consider adopting the averages reported here as they are the results of a rigorous scientific study.

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