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

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

Luis Humberto Colmenero Sujo*, María de Lourdes Villaiba**, Héctor Rubio Arias**, María Montero Cabrera***, Humberto Silva Hidalgo**

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 y 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 40 K. 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.

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; Topcuoglu, Türer, Güngör & Kırbasoglu, 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; Dohaničzuk-Sródka, 2012; García-Talavera,

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 (underlined 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 (Dominguez 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 used a square steel structure, 50 cm length and 5 cm depth, which was placed on the ground to define the soil sample (approximately 12.5 l). 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 Marielli 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) $3' 	imes 3'$ 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>
<thead>
<tr>
<th>City</th>
<th>North latitude</th>
<th>West longitude</th>
<th>High (mosl)*</th>
<th>Inhabitants**</th>
<th>Type of rocks***</th>
<th>Samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aldama</td>
<td>28 50</td>
<td>105 55</td>
<td>1270</td>
<td>19 378</td>
<td>ird</td>
<td>15</td>
</tr>
<tr>
<td>Bocoyna</td>
<td>27 51</td>
<td>107 35</td>
<td>2240</td>
<td>27 907</td>
<td>ird</td>
<td>5</td>
</tr>
<tr>
<td>Camargo</td>
<td>27 41</td>
<td>105 10</td>
<td>1220</td>
<td>45 852</td>
<td>sll</td>
<td>5</td>
</tr>
<tr>
<td>Creel</td>
<td>27 45</td>
<td>107 38</td>
<td>2345</td>
<td>5026</td>
<td>ird</td>
<td>8</td>
</tr>
<tr>
<td>Cuauhtémoc</td>
<td>28 24</td>
<td>106 52</td>
<td>2060</td>
<td>124 378</td>
<td>ird</td>
<td>9</td>
</tr>
<tr>
<td>Chihuahua</td>
<td>28 38</td>
<td>106 04</td>
<td>1440</td>
<td>671 790</td>
<td>ird</td>
<td>30</td>
</tr>
<tr>
<td>Delicias</td>
<td>28 11</td>
<td>105 28</td>
<td>1170</td>
<td>116 426</td>
<td>ird</td>
<td>8</td>
</tr>
<tr>
<td>Jiménez</td>
<td>27 08</td>
<td>104 55</td>
<td>1380</td>
<td>38 323</td>
<td>ird</td>
<td>10</td>
</tr>
<tr>
<td>Juárez</td>
<td>31 44</td>
<td>106 29</td>
<td>1140</td>
<td>1 218 817</td>
<td>sll</td>
<td>20</td>
</tr>
<tr>
<td>Manuel Benavides</td>
<td>29 06</td>
<td>103 54</td>
<td>1060</td>
<td>1746</td>
<td>sll</td>
<td>5</td>
</tr>
<tr>
<td>Nuevo Casas Grandes</td>
<td>30 25</td>
<td>107 54</td>
<td>1460</td>
<td>54 390</td>
<td>ird</td>
<td>12</td>
</tr>
<tr>
<td>Ojinaga</td>
<td>29 34</td>
<td>104 24</td>
<td>800</td>
<td>24 307</td>
<td>sll</td>
<td>8</td>
</tr>
<tr>
<td>Parral</td>
<td>26 56</td>
<td>105 40</td>
<td>1720</td>
<td>100 821</td>
<td>ird</td>
<td>29</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td>2 449 161</td>
<td>164</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* 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 (α = 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; Dolhańczuk-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 dose (AED) in mSv y\(^{-1}\) was obtained:

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

**RESULTS**

Table 2 shows the mean values, standard deviation and minimum and maximum values for \( ^{238}\text{U}, ^{232}\text{Th} \) and \( ^{40}\text{K} \) found in the soils of the 13 cities evaluated. Concentration of \( ^{238}\text{U} \) 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 \( ^{232}\text{Th} \) 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 \( ^{238}\text{U} \), 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 \( ^{40}\text{K} \). 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 \( ^{40}\text{K} \).

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, 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 \( ^{238}\text{U}, ^{232}\text{Th} \) and \( ^{40}\text{K} \) (Bq kg\(^{-1}\)) in selected cities in Chihuahua State.

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 C, 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 (Dolhańczuk-Sródk, 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 other countries (Dolhańczuk-Sródk, 2012; Jabbar et al., 2010; Mehra et al., 2007; Quindos, Fernández, Rodenas, Gómez-Arozamena & Arteche, 2004; Tsourtzis, 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.

### Table 2.

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

<table>
<thead>
<tr>
<th>City</th>
<th>(^{238})U</th>
<th>Min-Max</th>
<th>(^{232})Th</th>
<th>Min-Max</th>
<th>(^{40})K</th>
<th>Min-Max</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aldama</td>
<td>51.9 ± 0.3a*</td>
<td>44.6 - 58.2</td>
<td>61.1 ± 0.5a*</td>
<td>53.7 - 68.4</td>
<td>1014.5 ± 6.2a*</td>
<td>817.5 - 1106.3</td>
</tr>
<tr>
<td>Bocoyna</td>
<td>41.6 ± 0.3ab</td>
<td>34.0 - 47.2</td>
<td>48.1 ± 0.4abc</td>
<td>37.9 - 54.7</td>
<td>815.8 ± 5.8abcd</td>
<td>720.7 - 893.4</td>
</tr>
<tr>
<td>Camargo</td>
<td>34.1 ± 0.2b</td>
<td>30.7 - 40.1</td>
<td>38.0 ± 0.3c</td>
<td>33.1 - 44.6</td>
<td>827.0 ± 5.1abcd</td>
<td>766.4 - 909.8</td>
</tr>
<tr>
<td>Creel</td>
<td>35.3 ± 0.3b</td>
<td>29.7 - 55.1</td>
<td>38.5 ± 0.4c</td>
<td>33.3 - 55.6</td>
<td>663.7 ± 5.3de</td>
<td>552.9 - 823.4</td>
</tr>
<tr>
<td>Cuauhtémoc</td>
<td>37.7 ± 0.3b</td>
<td>32.0 - 43.3</td>
<td>43.9 ± 0.4bc</td>
<td>36.0 - 55.8</td>
<td>797.6 ± 5.5bcd</td>
<td>571.0 - 972.8</td>
</tr>
<tr>
<td>Chihuahua</td>
<td>39.8 ± 0.3b</td>
<td>28.9 - 54.6</td>
<td>55.2 ± 0.4ab</td>
<td>23.5 - 102</td>
<td>916.6 ± 6.0ab</td>
<td>653.1 - 1179.2</td>
</tr>
<tr>
<td>Delicias</td>
<td>36.5 ± 0.3b</td>
<td>31.3 - 43.6</td>
<td>33.4 ± 0.4c</td>
<td>28.5 - 38.2</td>
<td>772.0 ± 5.8bcde</td>
<td>685.6 - 848.5</td>
</tr>
<tr>
<td>Jiménez</td>
<td>40.4 ± 0.3b</td>
<td>38.1 - 45.9</td>
<td>42.3 ± 0.4c</td>
<td>38.2 - 49.5</td>
<td>930.5 ± 6.1ab</td>
<td>842.5 - 1002.8</td>
</tr>
<tr>
<td>Juárez</td>
<td>19.8 ± 0.2c</td>
<td>18.5 - 23.0</td>
<td>20.0 ± 0.3d</td>
<td>15.3 - 25.4</td>
<td>570.7 ± 2.9e</td>
<td>512.5 - 631.4</td>
</tr>
<tr>
<td>Manuel Benavides</td>
<td>34.8 ± 0.2b</td>
<td>27.3 - 41.1</td>
<td>32.9 ± 0.3cd</td>
<td>20.0 - 39.4</td>
<td>601.7 ± 4.3de</td>
<td>414.4 - 828.9</td>
</tr>
<tr>
<td>Nuevo Casas Grandes</td>
<td>41.1 ± 0.3b</td>
<td>37.7 - 44.9</td>
<td>55.9 ± 0.5ab</td>
<td>48.9 - 63.6</td>
<td>1013.7 ± 6.9a</td>
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</tr>
<tr>
<td>Ojinaga</td>
<td>31.8 ± 0.3b</td>
<td>27.6 - 34.1</td>
<td>32.0 ± 0.4cd</td>
<td>27.2 - 36.1</td>
<td>688.3 ± 4.7cde</td>
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<td>Parral</td>
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<td>18.3 - 63.3</td>
<td>36.6 ± 0.4c</td>
<td>10.3 - 63.1</td>
<td>859.0 ± 5.9abc</td>
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<td>Mean</td>
<td>36.8 ± 0.3</td>
<td>41.4 ± 0.4</td>
<td></td>
<td>805.7 ± 5.4</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Means in columns that do not share a letter are statistically different. 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 (Dolhańczuk-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.

REFERENCES


