Reconstruction of Atmospheric Concentrations and Deposition of Uranium and Decay Products at Aldama, Mexico

Paper 2012-A-488-AWMA

Michel Y. Montelongo, Eduardo F. Herrera, Jorge I. Carrillo, Luis M. Rodríguez Román P. Balam, Balter Trujillo, María E. Montero, Elías Ramírez, Alfredo Campos and Ana C. Rodríguez.

Centro de Investigación en Materiales Avanzados, S. C. (CIMAV)
michel.montelongo@cimav.edu.mx, eduardo.herrera@cimav.edu.mx,
jorge.carrillo@cimav.edu.mx, miguel.rodriguez@cimav.edu.mx,
roman.balam@cimav.edu.mx, balter.trujillo@cimav.edu.mx,
elena.montero@cimav.edu.mx, elias.ramirez@cimav.edu.mx,
Alfredo.campos@cimav.edu.mx, ceccy_864@hotmail.com

ABSTRACT

In the city of Aldama, Chihuahua, Mexico a plant was installed for milling uranium ore. This plant called URAMEX, processed ore for a period of 4 years, starting in 1983 and ending in 1987. The concentrations of radionuclides of uranium and thorium released to air by the milling process were estimated. The milling process that produced air emissions of radionuclides included crushing and grinding of ores; conveyance of ore; ore roasting, drying, and packaging of the product (U3O8); and fugitive dust releases from ore piles, tailing's piles, and roads. A grid node spacing of 500 m was designed for the extraction of eighty-one silt sampling points to determine the concentration of radionuclides in the soil. During 2011, 3 HV Graseby-Andersen collectors were placed to capture the PM10 and calculate its concentration in air at the study area. In parallel, 81 samples of silts were collected to establish comparison between the concentration of studied isotopes in air and silt. Atmospheric transport modeling requires a complex terrain model. Because there was a lack of historical meteorological data required for modeling in complex terrain, it was necessary to install instruments that would measure it for a year. Monthly average dispersion and deposition factors were calculated using a complex terrain model: CALPUFF. The initial and boundary conditions required for calibrating the model are presented in this work as well.

INTRODUCTION

The world's population is constantly exposed to radiation through natural and anthropogenic sources. This includes radiation and radioactive substances from mining, energy production through combustion of coal, medicine, and environmental pollution from radioactive wastes resulting from large-scale nuclear weapons testing and nuclear accidents (UNSCEAR, 2001)[1].

It is interesting to study the behavior of radionuclide concentrations in air and surface soil in order to determine the current levels of radioactivity. The first exploration of radioactive minerals in Mexico, focused on calcareous rocks, the mineral that was first

extracted in the Sierra Gomez, Chihuahua, to separate molybdenum ore, and the Comisión de Fomento Minero (Mining Development Commission) would benefit from a pilot plant installed a few hundred meters from the city of Aldama, Chihuahua.

The exploration of radioactive minerals in Mexico began in 1955, with the Comisión Nacional de Energía Nuclear (National Nuclear Energy Commission) (CNEN) being the federal government agency in charge until 1972. Later in 1979, this was divided into the Instituto Nacional de Investigaciones Nucleares (National Institute for Nuclear Research) (ININ) and the Comisión Nacional de Seguridad Nuclear y Salvaguardias y Uranio Mexicano (National Commission of Nuclear Safety and Safeguards, Mexican Uranium) (URAMEX) to develop the mining stage of the nuclear cycle (exploration, exploitation and processing of radioactive minerals), as mentioned in Perea (1979)[2].

The uranium deposits located in the Sierra Pena Blanca, a municipality of Aldama, Chihuahua, are 85 km north of Chihuahua City. This range consists of a sequence of limestone, shale and sandstone of Paleozoic and Cretaceous limestones, covered by volcanic rocks of acid composition of the Tertiary.

URAMEX, inherited its functions of exploitation and profit from the Consejo de Recursos Minerales (Council of Mineral Resources) (CRM) in a political and labor conflict that was settled in accordance with the law in 1987 and is now called the Servicio Geológico Mexicano (Mexican Geological Service) (SGM).

Based on the foregoing, we have the following objectives: estimating the activities of ^{238}U radioisotopes, ^{232}Th, ^{226}Ra and ^{210}Pb, deposited in the lifetime of the firm URAMEX (1979-1987). with the goals of determining correlations between the concentrations of isotopes in PM_{10} and silts, establish equivalent doses of radiation to map of isopleths around the plant and the city of Aldama, and to obtain the concentrations of radioisotopes used in this work. These concentrations will also serve as boundary conditions in the initial CALPUFF program, in order to produce known dispersion and deposition factors of radioactive particles during the extraction of uranium (Rood, 2008)[3].

Other isotopes of anthropogenic, natural, and cosmogenic origin as ^{40}K, ^{137}Cs and ^{7}Be, respectively, were also included in the analysis.

BACKGROUND OF THE WORK

Materials and Methods

Three high-volume collectors Graseby-Andersen (2001)[4], with PM_{10} head were placed in specific locations in the city of Aldama in order to measure isotope concentrations in the air. The points were located in the upper part of the same plant URAMEX, on the roof of City Hall and on the roof of a private home to make the location an approximate isosceles triangle and thus, cover the entire city (see Figure 1). As shown in the same figure, the city area of 16 km^2 was divided into 64 squares of 500 m. Silt samples were taken between January and June 2011, before the rains that began in late June.

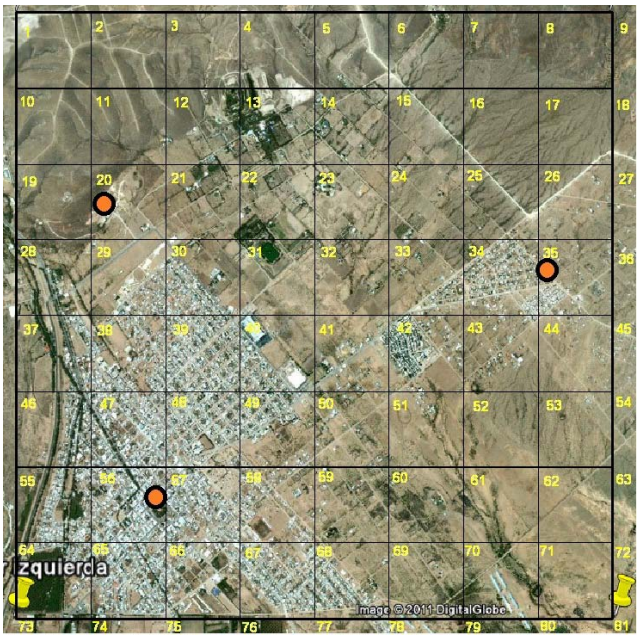

Figure 1. Aerosol collection points (red circles) and silts (numbers in yellow) within the city of Aldama.

Glass-fiber filters (WHATMAN[5] of 8 x 11 in.) were weighed and conditioned before and after the collection. The aerosol collection was performed following the timetable proposed by the EPA and the Mexican Official Standard NOM-035-ECOL-1993[6] in 2011 every six days. The mud was collected and sieved according to ASTM method C136[7,8,9,10], with grains of up to 75 microns and deposited in polyethylene containers 5 x 5 cm diameter.

The concentrations of uranium and thorium from anthropogenic, natural, and anthropogenic isotopes were calculated from PM_{10} collected in the silts. For the case of the filters, concentrations were determined from the following concentration method described in the Mexican Official Standard NOM-035-ECOL-1993[6]. The values of the concentrations from the particles collected on the filters were determined in $\mu g/m^3$ and were converted to miliBecquerel per cubic meter (mBq/m^3). For the case of mud, concentration was calculated in Becquerel per kilogram (Bq/kg), based on the weight of the collected sample.

Both filters and silt samples were measured in a gamma spectrometer CANBERRA with a 60 cm^3 active volume and processed using the Spectrum Analyzer (SA-1997)[11]. The spectra obtained in measurements of filters and polyethylene cylindrical containers must be corrected for efficiency. The filters were folded into four parts; however, the geometry was completely atypical and it was necessary to resort to Monte Carlo modeling, using the EGS4-2007 program[12]. The same procedure and program was used to measure the silt.

We selected those isotopes present in the spectrum based on the Natural Radioactive Series ^{238}U and ^{232}Th. To achieve the radioactive secular equilibrium, both samples were collected and deposited in PM_{10} filters , such as polyethylene containers mentioned for the case of the silt and thereafter were stored for at least one month.

Table 1 shows the isotope selected for the measurement of gamma lines. This table lists the daughter measured and its parent isotope, the energy of the gamma line that is measured, the period of semi-disintegration, and the relative intensity or (branshing ratio).

Table 1. In which the main characteristics of the isotopes measured was showed.

Daughter	Parent	• • • •	T(1/2)	I . (%)
^{210}Pb	^{238}U	46.53	22.3 y	4.25
^{226} Ra	^{238}U	186.21	1600 y	3.59
^{212} Pb	^{232}Th	238.6	10.6 (1.4 x10 ^{10} y)	43.3
^{214} Pb	^{238}U	241.9	28.6 m	7.43
^{214} Pb	^{238}U	351.9	28.6 m	37.6
^{7} Be	N/A	477.6	53.12 d	10.52
^{214} Bi	^{238}U	609.3	19.9 min (4.5 x 10^{3}y)	46.1
^{137}Cs	N/A	661.6	30.07 y	85.1
^{214} Bi	^{238}U	1120.3	19.9 min (4.5 x 10^{3}y)	15.1
^{40} K	N/A	1460.8	1.3 x10 ^{9} y	11

The results of the values of the specific activities (Bq/kg) in each of the 81 vertices sampled were processed using the Surfer V8 2002[13]. Using the values of the isotopic concentration isopleths, it can be shown that dispersions of the material were produced during the start-up of the plant in 1987 and throughout the period.

Dispersion of natural uranium in the city of Aldama

Although 10 lines were measured we only show the results of isotope of uranium for the emissions in the years when the plant was active. There was a surprising result of alterations of ^{137}Cs in air and soil, because measurements were made throughout the year 2011 and the results appear during the accident at the nuclear plant in Fukushima, Japan following the earthquake which occurred on March 11, 2011.

The high values of uranium concentrations in the plant indicate that there was a shift in the material, and radioactive contamination in the northwest section of the plant. Research indicates that once the plant was closed, cleansed tanks or storage hoppers, were used to carry out the separation process of the minerals of interest. This cleaning was done with pressure water hoses, so that all dissolved, contaminated material ran toward plant areas of lower height, resulting in stagnation, and later, resulting in the wind spreading throughout the city . Figure 2 shows the specific activity of uranium in the areas surrounding the plant. The specific activity values reached nearly 2000 Bq/kg northwest of the entrance of the plant. The city is only a few hundred meters from the plant. All dimensions are decreasing in height towards the direction of the Chuviscar River.

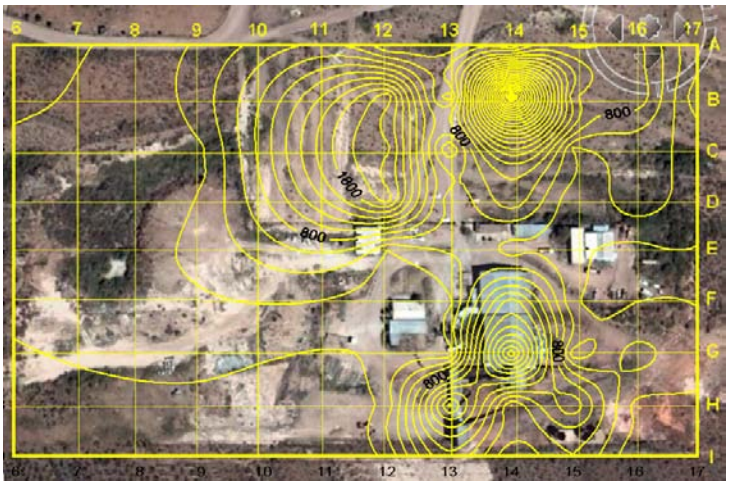

Figure 2. Dispersion of pollutants in the grounds of the plant today.

The overall analysis for the entire city of Aldama shows that higher concentrations of uranium eventually flowed into the region Chuviscar River at the entrance to the city, but the specific activity decreases rapidly in Becquerel per kilogram thousands of dozens. This has been proven in previous works (Colmenero, L. 2004)[14]. As shown in Figure 3, the concentration values can reach nearly 100 Bq/kg. In the upper right corner of the figure, some increases in the road leading to the mining areas of extraction of the material can be seen.

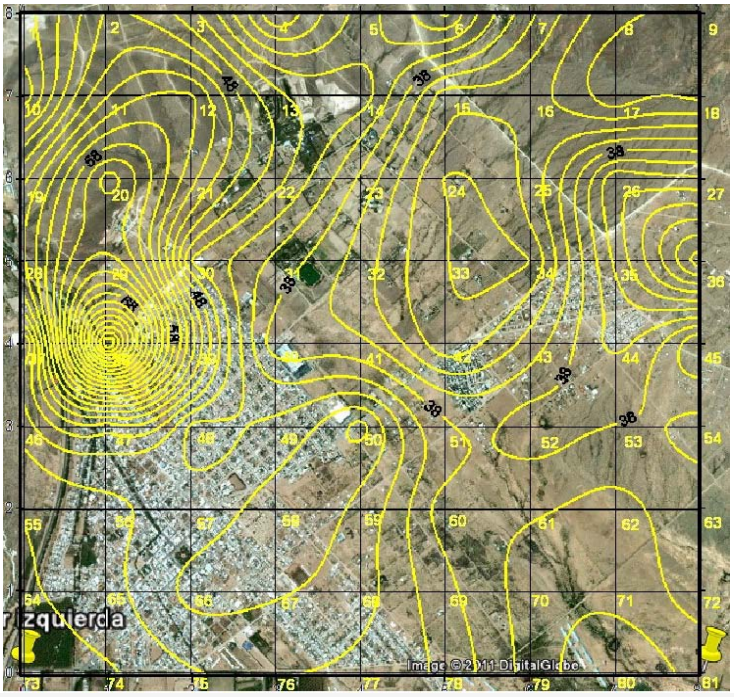

Figure 3. Dispersion of natural uranium on the city of Aldama.

Figure 4 shows more representative values of corresponding surface of the material to higher activities. Meteorological data recorded at stations of the Municipal Board of Water and Sanitation indicate that the annual average wind in the area southwest. This

coincides with the great mass of pollution that has been moving toward the northwest of the city where the greatest population density exists.

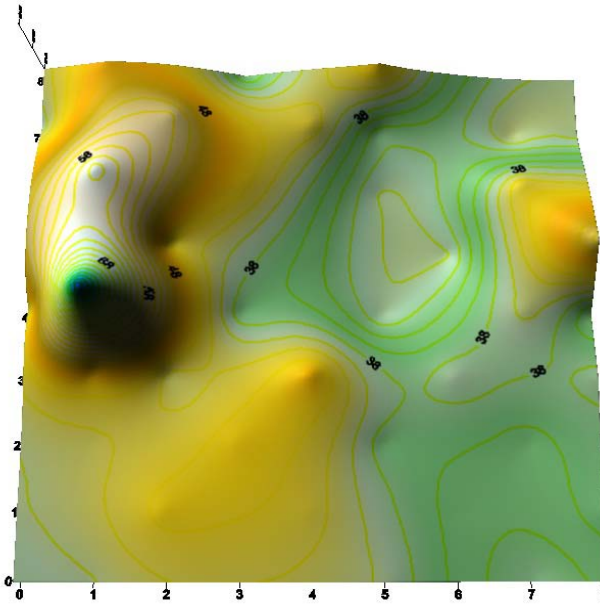

Figure 4. Surface graphic of the dispersion of natural uranium.

^{137}Cs in atmospheric air and on the floor of Aldama

^{137}Cs is a completely anthropogenic isotope found throughout the Earth as a result of nuclear explosions at Hiroshima and Nagasaki. Since 1945, high amounts of this isotope is mainly due to a phenomenon known as fallout and resulted in ^{137}Cs in the air around the 60's because of nuclear tests in the United States, Russia, France and China. This can be seen in Figure 5 (Escudero and 2007)[15].

Normally, the ^{137}Cs is found in higher or lower concentrations in both air and ground, because once it is captured by the general circulation of the atmosphere from its original source, it spreads across the surface of the Earth, and returns to the air by wind drag. Because ^{137}Cs, has a half-life period of 30 years, it may be present in any of the samples of PM_{10} or silts. The latest data showed that our laboratory with concentrations of ^{137}Cs, before 2011, were due to accident at Chernobyl, where the activity in Chihuahua was 2.3 mBq/m^3 in air.

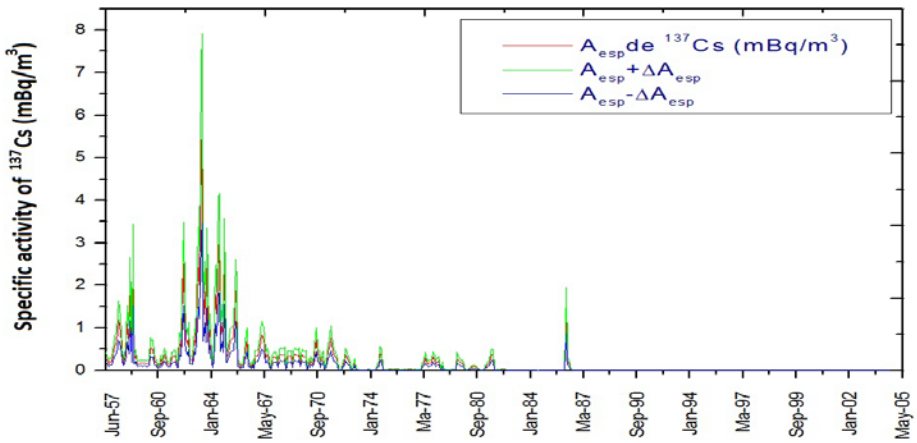

Figure 5. Specific activity of ^{137}Cs in the atmospheric air of Chihuahua since 1957.

By chance, measurements of PM_{10} and silt in this work were done during the year 2011, coinciding with the accident Fukushima, Japan on March 11. Therefore, we were able to see the increases in concentrations of this isotope in both air and soil. The contents of ^{137}Cs in soil vary with the weather conditions of the place. Figure 6 shows the values detected in the soil.

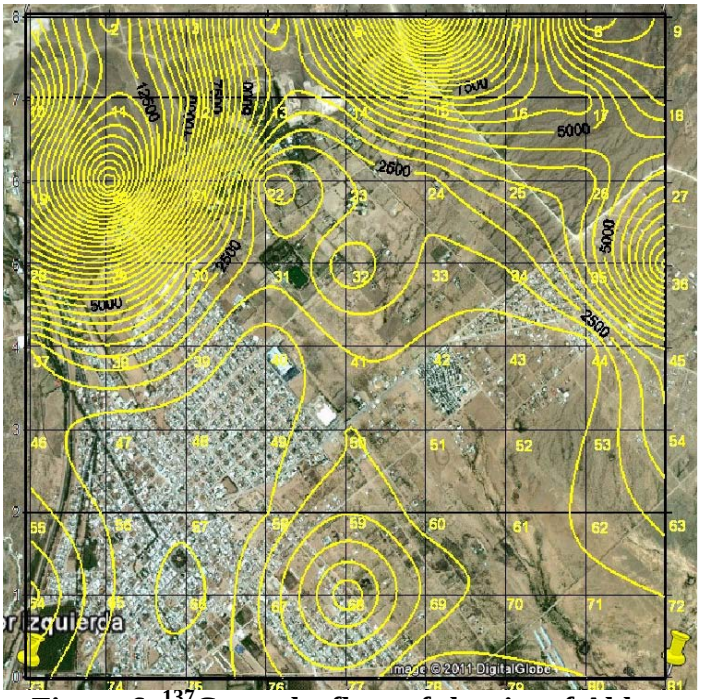

Figure 6. ^{137}Cs on the floor of the city of Aldama.

The values reported in the graph are given in mBq/kg. In addition to this spatial representation, it was possible to make comparisons of the concentrations of ^{137}Cs detected in PM_{10} temporarily, showing an increase between April and July.

Figure 7 shows through a bar chart the values of ^{137}Cs concentration in PM_{10}. It can be seen that the concentration increased leading into April and decreased just after July, with the arrival of the rains.

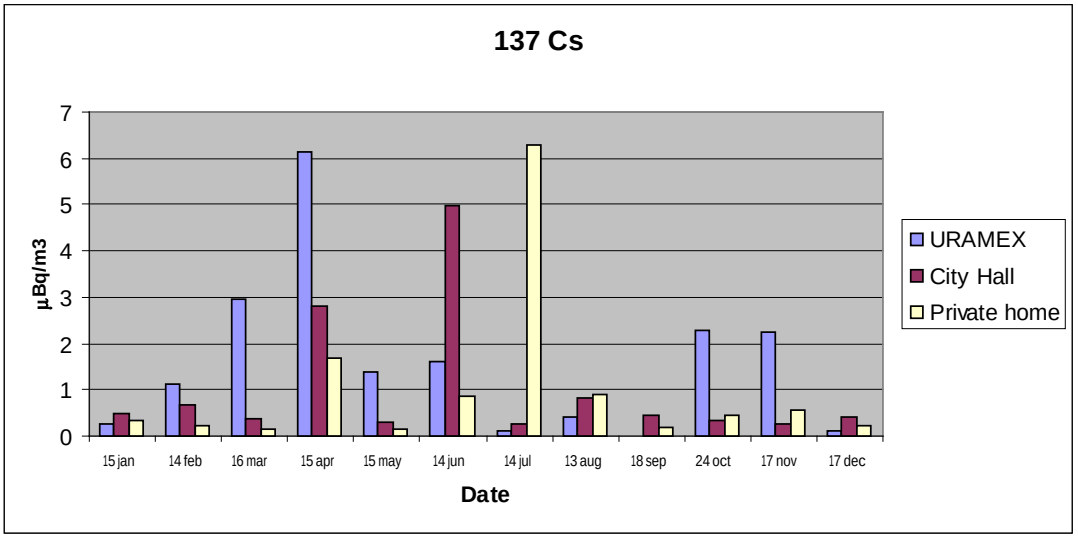

Figure 7. Increments and decrements in the contents of ^{137}Cs in PM_{10}.

Interestingly, the report made by the CNSNC on the content of ^{137}Cs in the air showed 2.5 µBq/m^3 to La Paz, while our measurements reached 3 µBq/m^3, around the same date. In contrast, in the months of April through July, values up to 6.3 µBq/m^3 are reported, indicating likely influence of the Fukushima accident, and possibly implying release of more radioactive material in Fukushima accident than Chernobyl.

ACKNOWLEDGMENTS

We determined the content of natural uranium with other elements of less interest. That leaves the foundations remaining to perform back-testing using the CALPUFF program, which will be the next step in this work. There was also a brief discussion of variations in the concentrations of ^{137}Cs in PM_{10} and the content of the silt soil. In the case of PM_{10} it was found that increased levels of this element in the month of April were due to the accident at the Fukushima plant and started to decrease from the month of July when the rainy season began. The reports agree with the values reported by the CNSNC officers.

REFERENCES

1. ONCEAR "Report of the United Nations Scientific Committee on the Effect of Atomic Radiation to the General Assembly, Exposure from Natural Radiation Sources"., NY, 2008.

2. Perea Saenz, A. Explotación del yacimiento uranífero el "Nopal 3" Sierra Peña Blanca Mpio. de Villa Aldama, Estado de Chihuahua. Tesis de Ingenieria. Facultad de Ingenieria. Universidad Autonoma de Chihuahua. Chihuahua, Mex.1979.

3. Arthur S. Rood, Paul G. Voilleque, Susan K. Rope, Helen A. Grogan, John E. Till. Reconstruction of atmospheric concentrations and deposition of uranium and decay products released from the former uranium mill at Uravan, Colorado. Journal of Environmental Radioactivity 99 (2008) 1258–1278.

4. USEPA RFPS-1287-063 Graseby Andersen/GMW Model 1200 High-Volume Air Sampler.2001.

5. WHATMAN. Leadership in separation technology for the life sciences. http://www.whatman.com/products/?pageID=7.30, 2005.

6. NOM-035-ECOL-1993 que Establece los métodos de medición para determinar la concentración de partículas suspendidas totales en el aire ambiente y el procedimiento para la calibración de los equipos de medición.

7. ASTM C136 - 06 Standard Test Method for Sieve Analysis of Fine and Coarse Aggregates, 2012.

8. EPA. Environmental Protection Agency. Method IO-2.2: Sampling of Ambient Air for PM10 Using an Andersen Dichotomous Sampler. Compendium of Methods for the Determination of Inorganic Compounds in Ambient Air. Office of Research and Development. US, 1999.

9. EPA Environmental Protection Agency. 1993. Procedures for Laboratory Analysis of Surface/Bulk Dust Loading Samples. AP-42, Appendix C.2.E.U.

10. EPA Environmental Protection Agency. 1993. Procedures for Sampling Surface/Bulk Dust Loading. AP-42, Appendix C.1. E.U.

11. J. Perdomo, C. Trápaga. Programa Spectrum Analyzer. Tesis de Licenciatura. ISCTN, 1997.

12. **EGS4** Source **Code** Availability and Distribution. 2007.

13. SURFER Version 8, Surface Mapping System. Golden Software Inc. 2002.

14. L. Colmenero Sujo, M.E. Montero Cabrera , L. Villalba ,M. Renterıa Villalobos, E. Torres Moye a, M. Garcıa Leon c, R. Garcıa-Tenorio, F. Mireles Garcıa E.F. Herrera Peraza, D. Sanchez Aroche. Uranium-238 and thorium-232 series concentrations in soil, radon-222 indoor and drinking water concentrations and dose assessment in the city of Aldama, Chihuahua, Mexico. Journal of Environmental Radioactivity 77 (2004) 205–219.

15. Escudero Y. Obtención de las concentraciones históricas de ^{137}Cs en la ciudad de Chihuahua utilizando el ^{7}Be como trazador. Tesis de Maestría, CIMAV, 2007.