Evaluation Of The Transport Of Norm Contaminants In The Air And Saturated Underground Zone Of The City of Aldama, Mexico.

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ABSTRACT

In this study, it was postulated that naturally occurring radioactive material (NORM) concentrations in air, soil, and Aldama City waters due to uranium and thorium must have some anthropogenic component. This suggests that it may have been a bad decision to locate the URAMEX plant, the current Majalca Processor, on the grounds, upstream of the hydrological basin of the Chuvíscar River. To achieve the objective of studying NORM contamination, the modeling provided by WRPLOT (Tee J., 2016), CALVIEW (Cire, J., CALPro Plus, 2011) and HYDRUS-1D (Simunek, J., 2008) were used to determine the following:

i) Dispersion of pollutants from NORM wastes that were stored and dispensed from piles of waste and the oxidation pond (sources) and that somehow were reaching the surface of the city soil, to the unsaturated zone and subsequently to the groundwater zone.

ii) Dispersion of anthropogenic radioisotopes without a defined source. This reference is made because of the vertical transport of ^{137}Cs from the total area of the city to the underlying layers of the unsaturated zone. The 1986 Chernobyl and 2011 Fukushima accidents are responsible of these events (Medvedev, A. 1990). (Carbonell, EF 2013).

iii) Drainage flow values obtained at different depths in a simulated way for the determination the possible contamination of the soil layers (root layer) until the termination of the vadose zone where the displacement of the groundwater flows begins.

INTRODUCTION

The world's population is constantly exposed to radiation through natural and anthropogenic sources, which involve the use of radiation and radioactive substances that cause additional exposure above natural background. Artificial sources such as mining, coal

combustion, medicine, environmental pollution due to radioactive waste from nuclear weapons and large-scale nuclear accidents, continue to be a global source of human exposure (UNSCEAR, 2016).

The city of Aldama in Mexico is located 30 km to the NNE of the capital of the State of Chihuahua, with coordinates of 28º 50 '19 "N latitude and 105º 54' 40 " W longitude and an altitude of 1 270 meter above sea level (masl). The average maximum temperature is 44º C and a minimum of 14º C. The average annual rainfall in the municipality is 305.2 millimeters, with a relative humidity of 45% and an average of 49 days of rain. The prevailing winds come from the south. The population of 22,302 inhabitants (INEGI, Censo 2015).

The occurrence of uranium, in the Aldama region, is natural and small amounts are found. It accumulated at the top of the lithosphere within siliceous and felsitic rocks. Mexico joined the countries that study atomic energy on January 29, 1979, when Mexican Uranium (URAMEX) was officially created to exploit, benefit and commercialize radioactive minerals. The fundamental deposits of the state of Chihuahua are found mainly in volcanic rocks of the Sierra de Peña Blanca and neighboring areas. The depths that have been investigated do not exceed 100 m, although some outcrops are detected.

It should be noted that in the state of Chihuahua there are other important nearby deposits such as El Nopal, Margaritas, La Domitila and the ports. There are also mines in Sierra de Gómez, parallel to that of Peña Blanca.

Uranium is found in nature in the form of compounds mainly oxygenated, as the concentrations of residual magmas. Many mineralogical species contain uranium and radioactive elements, but only three are formed from hydrothermal solutions that come from the deep layers of the earth's crust. These include Uranite or uranium oxide, Pechblenda or uranium amorphous oxide, and Davidite or rare earth titanium oxide and uranium.

In Aldama, the URAMEX there was started as a minerals processing plant, which, operated in the decade of the 80s as a processor of yellow cake. This plant was closed, but was opened again to process other minerals like gold and silver. However, the same devices and storage areas, including discharge towers, crushing mills, conveyor belts to the pre-wash tanks and the spray mill, are still being used. On the outside you can observe the laboratories, the mechanical workshops and the oxidation lagoon. The grounds of the plant lie sloped about 10 degrees northwest, at a distance of 100 m from a school and a recreation center. The city limits are just 300 m from the fenced edge of the plant. URAMEX plant is located immediately to the south of the city. (Perea, A. 1979)

AIR QUALITY MONITORING

In specific locations of Aldama City, three Graseby-Andersen / GMW high-volume 1200 model manifolds with PM_{10} particle heads, with operational expenditure between 1.1 and 2.2 $m^3.s^{-1}$, were installed in order to measure the isotopic concentrations in the air particles collected throughout the year 2011. The collectors were placed in the highest part of the

URAMEX plant, on the roof of the Municipal Presidency and on the roof of a private house forming a triangle, as shown in Figure 1, to cover as much of the City area as possible.

PM_{10} particle monitoring was performed every 6 days for the entire year 2011 following the schedule proposed by the EPA (EPA, AP-42, Appendix C.1, 1993). One the day before each measurement, the new filters were placed and the filters used in the previous measurement were collected.

For the particulate matter measurements, WHATMAN filters of 8 x 11 inch fiberglass were used. These filters were weighed on a Sartorious electronic scale, model BP 211D, Serial No. 71104196, with a maximum capacity of 210 grams and resolution of 0.1 milligrams and conditioned for 24 hours in a controlled environment at a temperature of 25 ° C and humidity of 50% before and after the collection. Following the guidelines established in Official Mexican Standard NOM-035-ECOL-1993, which specifies that the difference in weight of the filters before and after the collection corresponds to the concentration of PM_{10} mass collected on the filters. The concentrations of PM_{10} particles collected in the measured filters were determined in $\mu g/m^3$.

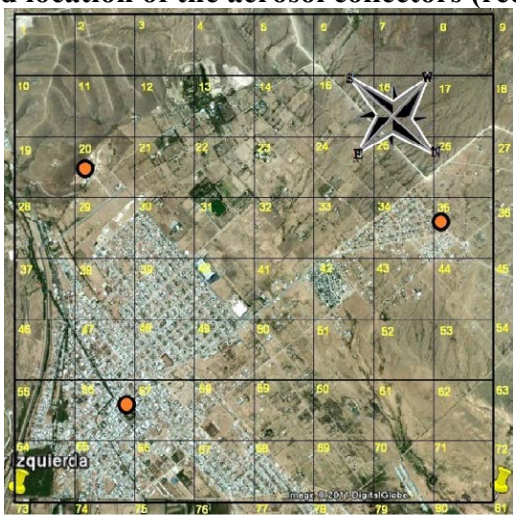
The measured concentration of PM_{10} particles in ambient air were compared with the maximum concentration of 120 $\mu g/m^3$ allowed in Official Mexican Standard NOM-025-SSA1-1993 to determine the population exposure.

The meteorological data generated by the station located in the city of Aldama, administered by the Central Board of Water and Sanitation of the State of Chihuahua was requested which provided the speed and direction of the wind and the environmental conditions during the collection days.

SILT MONITORING ON SOIL

The 16 km^2 of the city area were divided into 64 squares of 2500 m^2 and in the 81 corners of the formed grid where the samples of silt were taken between January and June of 2011, before the rainy season began at the end of June. See Figure 1.

Figure 1 Location of the collection points of the silt samples in the city of Aldama (vertices of the grids) and location of the aerosol collectors (red dots).



The collection of the samples is based on the procedure established by the EPA for obtaining the sediment load referred to in AP-42 Appendix C.1 (EPA, 1993). This procedure was modified for the accomplishment of the objectives of this work and was taken for reference only.

ANALYSIS OF SEDIMENTS

The silt content in a sediment sample is particulate matter less than 75 micrometers. The procedure that was performed is based on the one established by the EPA for the analysis of Sediment Load Samples through the ASTM Method C-136 contained in the AP-42 Appendix C.2 (EPA, 1993). For this work it is not necessary to know the sediment load, which procedure is also included in the method. The only interest was to guarantee the separation of the silts from the initial material that is collected.

OBTAINING THE RADIOISOTOPE CONCENTRATIONS

Both the filters and the silt samples were measured for 24 hours on a CANBERRA model 7500 SL pure germanium detector spectrometer with coaxial geometry of 80 cm3 active volume and a 2.5 cm radius with a spectrometric track of the same brand: Rack Model 2100, HV Power Supply Model 3106D, Amplifier Model 2022 and a Model 2004 preamplifier that are connected to the GENNIE 2000 with 8192 channels card.

All spectra were processed using the Spectrum Analyzer-1997 program (Perdomo et al., 1997). The spectra obtained in the filters measurements and cylindrical polyethylene containers had to be corrected for a higher efficiency. The filters were folded in four portions, so the geometry was completely atypical and the Monte Carlo method had to be used with the DOSRZ program (a variant of EGS4-2007). In this method 10 million events were played to minimize the error introduced. Using a calibrated point source of ^{125}Sb-^{154}Eu-^{155}Eu (SRM4275C-129 from NIST), the double log curve of the gamma ray energy efficiency was obtained. By means of DOSRZ, the geometric solid angle rates were calculated to correct the efficiency values of the geometry measured in the filters and the cylindrical containers of silts. The corrected value of the mentioned efficiency is obtained by multiplying the average of the punctual efficiency geometry of the photo-peak and the solid angle of the geometry for each energy detected.

The isotopes present in the spectrum were selected because of the ^{238}U and ^{232}Th Natural Radioactive Series. To achieve the radioactive secular equilibrium of the aerosols, the samples were collected and deposited in the filters. For the case of the silts, polyethylene bags were used and from that moment they were stored, for at least one month.

The selected isotopes for the measurement of gamma lines are shown in Table 1. The table specifies the measurements of daughters, the parent isotope, energy of the gamma line being measured, the half-disintegration period, and the relative intensity or branching ratio.

The concentration values of the particles collected in the filters were determined in $\mu g/m^3$ and to obtain the isotopic values a conversion to milliBecquerel per cubic meter (mBq/m^3) was required. As for slime, the concentration was obtained in (unit of Bq/kg), based on the weight of the sample collected.

The results of the values of the specific activities in Bq/kg were converted to Bq/m^2, at each of the 81 vertices sampled in the area of the city of Aldama and 62 of gird the areas of the URAMEX plant were processed by the Surfer V8 program (SURFER, 2002). By means of the Kriging method the isopleths of specific activity of each isotope were traced. This gives us an idea of how the dispersions of the material were produced during the plant's uptime throughout the period between 1980 and 1985.

Table 1 Main features of measured isotopes.

Hijos	Padre	Eγ (KeV)	T(1/2)	I γ (%)
^{210}Pb	^{238}U	46.53	22.3 y	4.25
^{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
^{214}Bi	^{238}U	609.3	19.9 min (4.5×10^3y)	46.1
^{137}Cs	N/A	661.6	30.07 y	85.1
^{214}Bi	^{238}U	1120.3	19.9 min (4.5×10^3y)	15.1

DIRECTION OF WINDS IN THE PERIOD ANALYZED

The WRPLOT program was used to determine the direction of prevailing winds in each of the years the plant remained open. This program was used in the condition "Flow Vector (blowing to)" instead of "Direction (blowing from)". Figure 2 shows the results of the wind rose.

On Figure 2 it is clear that the prevailing winds go to north, except in the years 1981 and 1982, where the components of the winds are maximum and have some composition from the SSE direction. This trend allows the average wind of the six years to lose the direction towards the north.

Figure 3 shows the average wind rose that is used at the URAMEX station to show that the strongest mean winds are directed towards NW with some tendency toward NNE.

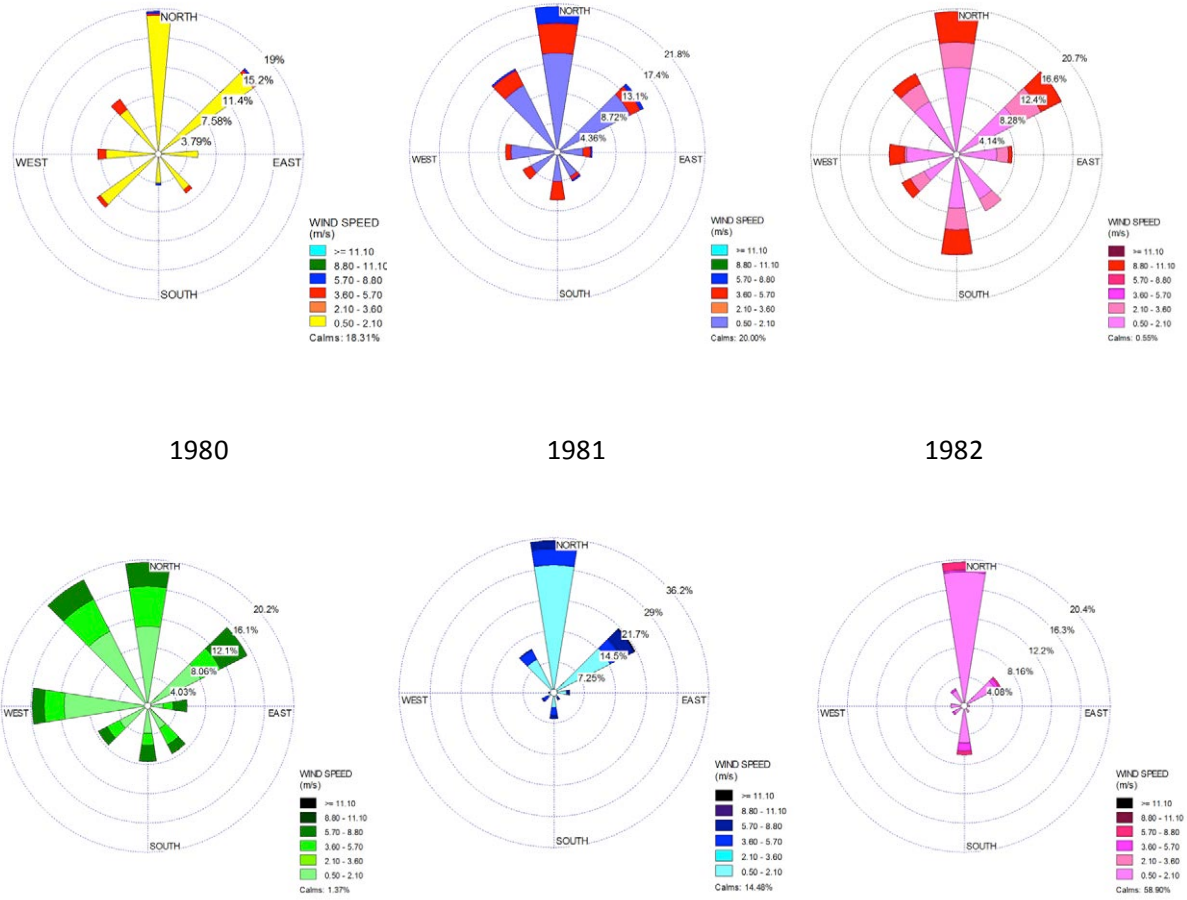
INFLUENCE OF THE DIRECTION OF WINDS IN THE DISPERSION OF RADIOISOTOPES.

The use of CALVIEW (CALPRO Plus program) yielded the average area of contamination during the years when the plant was operating, which coincides with the data obtained experimentally by direct measurements of soil silts. This graph is shown in Figure 4.

In addition, by combining the SURFER v8.02 and Google Earth v 2016 programs, contours, isopleths or lines of equal concentration of isotopes were designed for the processed and measured collected silts. This type of chart is shown in two dimensions superimposed on different layers of other maps taken from Google Earth.

The ^{238}U was measured at the 609.3 keV line, 214Bi with a bifurcation ratio of I_γ = 46.1%. The general analysis for the entire city of Aldama shows that the highest concentrations of ^{238}U finally flowed into the Chuviscar River at the entrance to the city, but the specific activity rapidly declines from thousands to tens of Bq/m2. This has been demonstrated in previous studies (Colmenero L. et al., 2004, Montelongo M. et al., 2015).

Figure 2 Rose of the winds for each of the years that the URAMEX plant was in operation.



1980 1981 1982

Figure 3 Direction of the average winds of the analyzed years

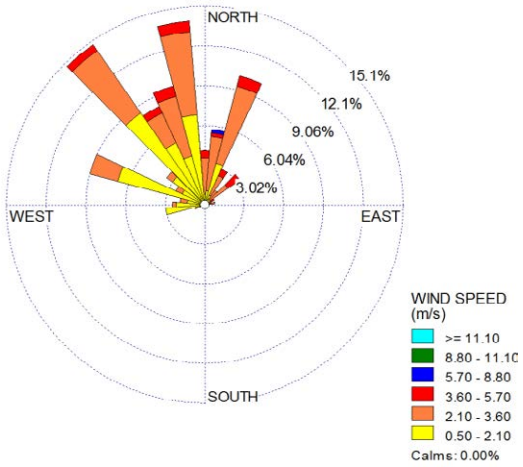
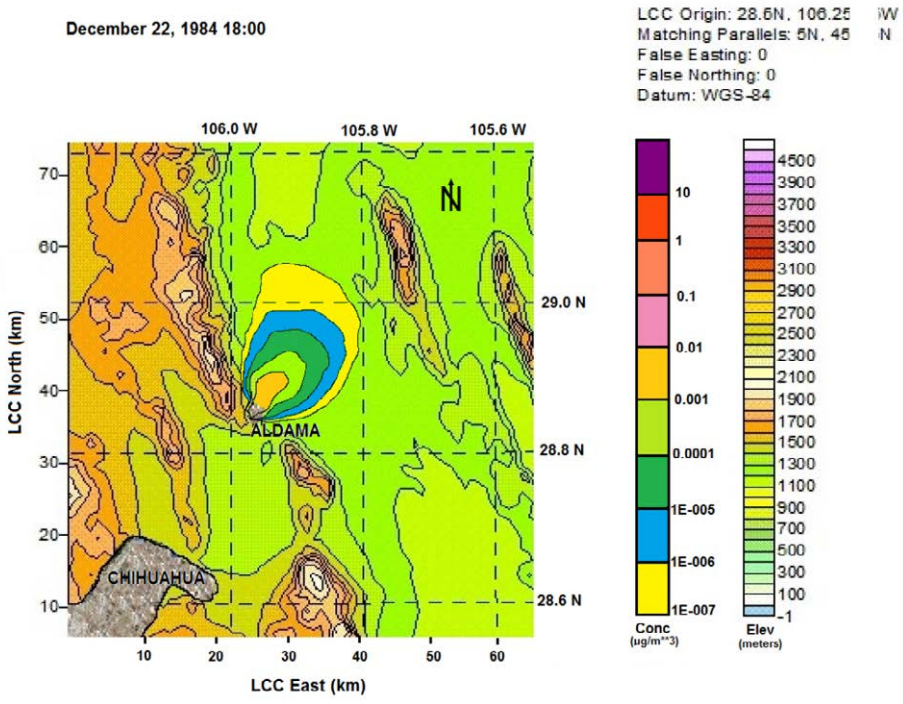


Figure 4 Average displacement of the contamination plume of the URAMEX plant on the city of Aldama.



As shown in Figure 5 (a), the concentration can reach almost 80 Bq/m^2 in the study area. In the center left of the figure, you can see some increases in specific activity along the road leading to the mining areas, whose maximum value can reach up to 58 Bq/m^2. The position of this maximum coincides with the meteorological data of the average wind direction from the SE. However, there is another large nucleus of contamination in the central right part of the figure. We assume that it is due to the cleaning of the plant with strong jets of water at the end of its working period. This concentration is not only due to the aerial deposition of

the radioactive powders produced during the operation of the plant but to the displacement of the waters with which it was cleaned. It can be verified that the height dimension decreases towards the zone of the Chuviscar River, place to where the polluted waters moved.

The ^{232}Th was determined by the 238.6 keV line of ^{212}Pb, with a bifurcation ratio of I_γ = 43.3%. In Figure 5 (b) it is observed that the values of the concentrations of ^{232}Th are distributed completely throughout the area of the city of Aldama. No point was found in the area where the concentration of this radioisotope shows an abnormal accumulation point or because of anthropogenic radioisotope concentration. Their dispersion is due only to the wind regime. A maximum of 64 Bq/m^2 is observed to the right of Figure 5 (b), which is to the west of the City of Aldama and a minimum of 42 Bq/m^2 in the upper right of the figure, which is to the east of the city.

The ^{210}Pb was determined by the 46.53 keV line, with a bifurcation ratio of I_γ = 43.3%., Considered in equilibrium with the parent nucleus ^{238}U, shows a somewhat different distribution, a maximum of 45 Bq/m^2 at the top Left of Figure 4 (c), that is, in the northwest of the city.

Finally, ^{137}Cs (anthropogenic isotope) are found in higher or lower concentrations, both in air and soil, because once it is captured by the general circulation of the atmosphere from its original source, it spreads throughout The surface of the Earth, and returns to the air by means of the wind trawling. Because the ^{137}Cs, has a half-disintegration period of 30 years, it can be present in any of the aerosol or silt samples. The latest data that our laboratory registered with concentrations of ^{137}Cs, before 2011, were due to the Chernobyl accident, whose activity in Chihuahua was 2.3 mBq/m^3 in air. It was measured on the gamma line of 661.64 keV, with a branching ratio of I_γ = 85.1%. By happenstance, the measurements of PM_{10} and silts of this work were realized throughout the year 2011, coinciding with the accident of Fukushima, Japan the 11 March. For this reason, it was possible to appreciate the increases in the concentrations of this isotope both in air and in the soil of this isotope. The contents of ^{137}Cs in the soil vary according to the weather conditions of the place. Figure 5(d) shows the values detected in the soil in the area of the city of Aldama. In this figure it can be observed that the ^{137}Cs accumulate in the places to the outskirts of the city, near the saws that the. Also its high concentration coincides approximately with the point of high concentration of ^{238}U of figure 5(a).

The correlation ^{238}U vs. ^{137}Cs shows a not so high Pearson coefficient, 39.6%. However, we found the low scatter probability values interesting (0.002). This indicates to us that there is some present phenomenon that shows a certain ordering between the values of the 137Cs concentrations with the ^{238}U.

If we look closely at Figure 5, when we correlate the ^{137}Cs with the ^{238}U, the residue values show interesting results in the settings (MINITAB v 17, 2013).

Two families of V-shaped pairs are opened, almost symmetrical with the zero of graph, as shown in Figure 6 (a). This indicates the ^{137}Cs reacts in two different ways with respect to the ^{238}U. This behavior is manifested even more in the regression graph of ^{137}Cs vs. ^{238}U, taking cesium as a regressor or predictor as shown in Figure 6 (b).

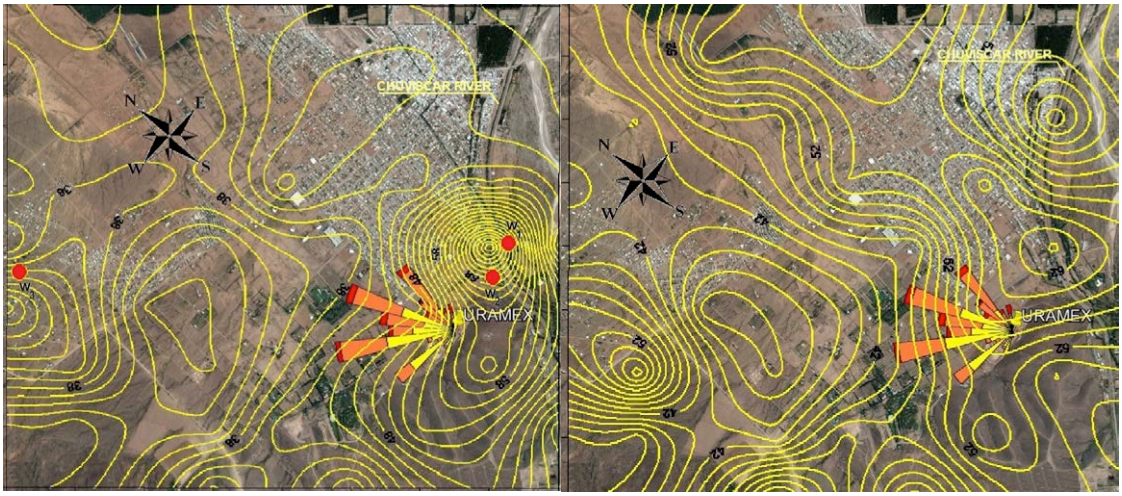
If we eliminate the lower family we obtain a regression of 78.4% with a scatter probability of p <0.0001, Figure 6 (b). This regression value corresponds to a positive correlation of 88.5% and a probability of practically zero point dispersion. This is indicating that there is a component of the concentrations of ^{137}Cs that cannot be dissociated from the origin of high concentrations of uranium in the plant.

This behavior is manifested even more in the regression graph of ^{137}Cs vs. ^{238}U, taking cesium as a regressor or predictor as shown in Figure 6 (b).

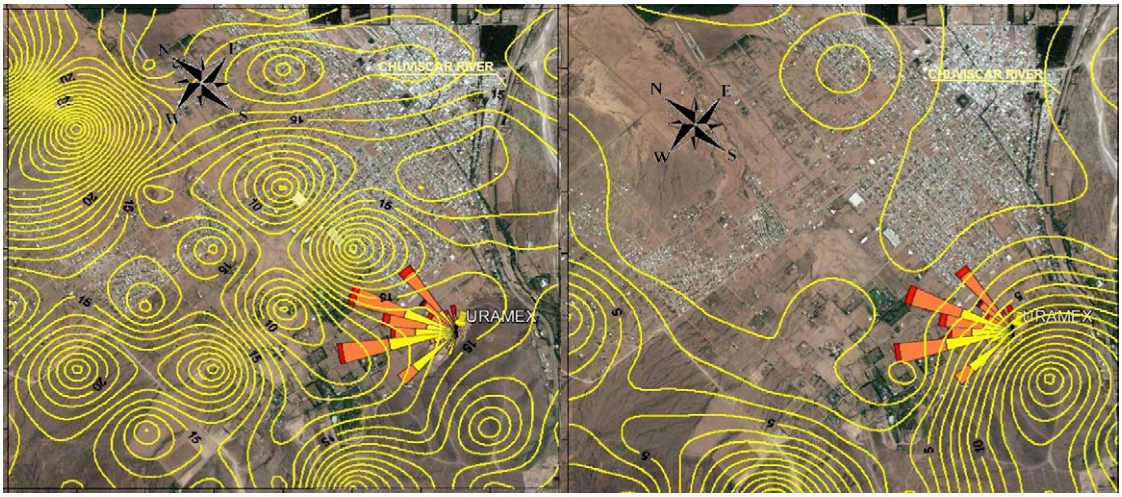
ANALYSIS OF THE RESULTS OF ^{238}U vs.^{137}Cs BEHAVIOR

The knowledge reported in the literature (Jiménez B. 2005) indicates that the existence of ^{137}Cs in the investigated regions of URAMEX silver and its specific activity has nothing to do with the evolution of the ^{238}U isotope since its origins are well defined and different as already indicated above. This would explain the regression component or the Pearson coefficient close to zero, but not the correlation of 88.5% of the upper branch.

Figure 5 Distributions of isotopes (a) ^{238}U, (b) ^{232}Th, (c) ^{210}Pb and (d) ^{137}Cs on the grounds of the URAMEX plant in the city of Aldama, Mexico.



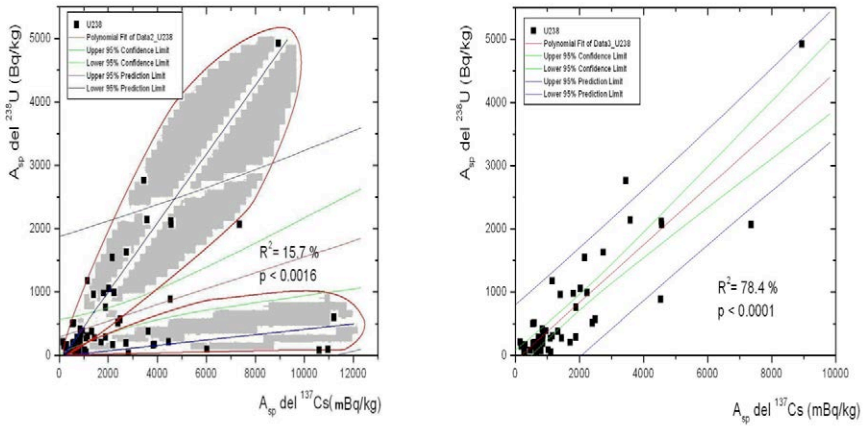
(a) (b)



(c) (d)

All concentrations of ^{137}Cs that might have come from the tropopause were due to the fallout since 1945 with the atomic bombs of Hiroshima and Nagasaki and other nuclear tests and nuclear-energy accidents).

Figure. 6(a) Regression graph of specific activity of the ^{238}U vs.^{137}Cs.The existence of two families of pairs is clearly observed. (B) Graph corresponding to the upper branch of the ^{238}U family vs.^{137}Cs, with R2 = 78.4% and r = 88.5%.

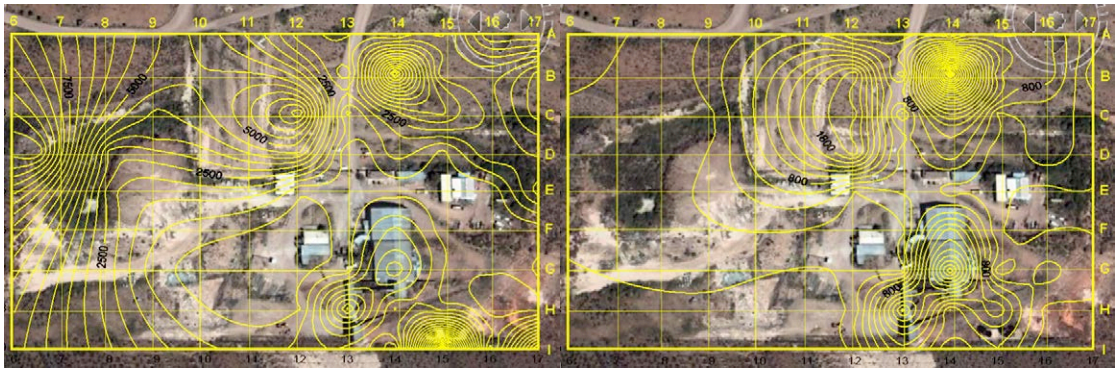


(a) (b)

It is assumed that at this time, the ^{137}Cs that is dispersed, only shows the mechanisms of resuspension from a certain threshold of wind, which allows to lift the heavier particles or greater aerodynamic radius.

Previous measurements carried out at the Center for Advanced Materials Research in Chihuahua (Rodríguez, L. 2010) have shown that ^{137}Cs exist with higher concentration, preferably in total suspended particles (PST), but not in PM_{10} or $PM_{2.5}$. This result allows us to think that ^{137}Cs are only transported in high speed wind regimes. In addition, despite the high solubility of the cesium-containing compounds, run-off and leaching cannot remove the cesium content in soil, remaining on the surface until a burst causes its re-loading. Cesium adheres firmly to most soils and does not mobilize much below the surface. As a consequence, cesium is not easily incorporated into plants through the roots. However, radioactive cesium could be absorbed by plants if it falls on if it falls on leaves. (ATSDR, 2004).

Figure. 7(a) Distribution of ^{137}Cs concentrations in the plant area and (b) Distribution of ^{238}U concentrations in the plant area.



(a) (b)

If we look closely at Figure 7 (a), where we show the distribution of ^{137}Cs in the plant area, it can be seen that there is a pattern of isopleths very similar to those shown for the ^{238}U in Figure 7 (b). The isopleths to the lower right of the graph of cesium concentrations no longer appear in the distribution graphs of the ^{238}U. For the latter case, the values of ^{137}Cs are due to the natural air cesium scattering cycle. For the first case, the only possible conclusion is that ^{137}Cs was collected from the mines of El Nopal, Las Margaritas and Puerto III and wine mixed with uranium compounds. So, that component has the same cycle as the ^{238}U.

CHARACTERISTICS OF UNDERGROUND WATER IN THE ALDAMA-SAN DIEGO AQUIFER (0836)

The ALDAMA-SANDIEGO aquifer (0836) has average water availability as shown in Table 2, according to the Public Register of Water Rights, published on June 30, 2014 (CONAGUA 2015).

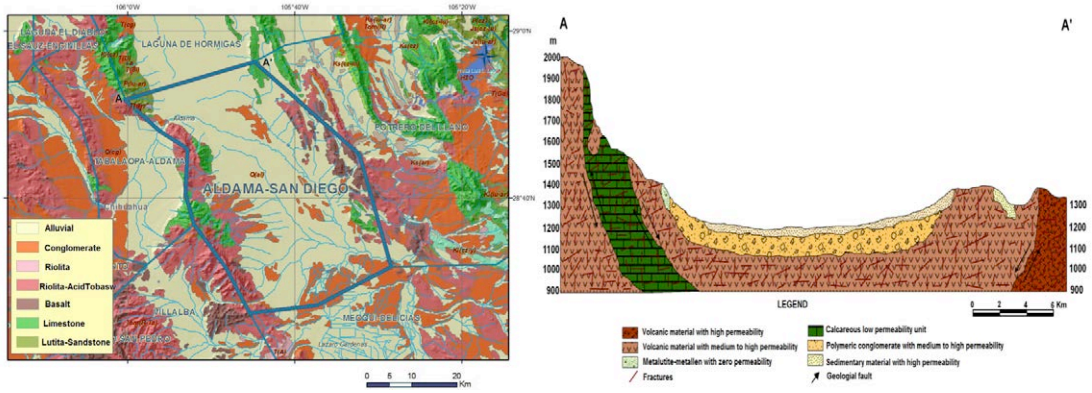
Table 2 Data of the ALDAMA-SAN DIEGO aquifer (836).

CCCXI	HYDROLOGICAL-ADMINISTRATIVE REGION "RÍO BRAVO"						
KEY	AQUIFER	R	DNCOM	VCAS	VEXTET	DAS	DÉFICIT
		VALUES IN MILLIONS OF ANNUAL CUBIC METERS					
CHIHUAHUA STATE							
0836	ALDAMA-SAN DIEGO	62.5	1.5	41.573119	63.6	19.426881	0.000000

Where R: annual average recharge, DNCOM annual discharge; VCAS volume of groundwater, VEXTET: volume of groundwater abstraction recorded in technical studies, DAS: average annual availability of groundwater. The definitions of these terms are those contained in the numerals "3" and "4" of Official Mexican Standard NOM-011-CONAGUA-2015.

Figures 8(a) and (b) show, in general, the hydrogeological section AA '. Also, the geometry of the aquifer system is a valley covered by recent sediments (alluvium) and conglomerate material with medium to high permeability. It is flanked to the east and west by volcanic material whose permeability varies from medium to high. Emerging to the west and underlining the volcanic material, are tertiary limestone intercalated with shales.

Figure 8(a) Map of surface geology of the Aldama-San Diego aquifer (836) and (b) The hydrogeological cutting section AA '.



For the analysis of the water quality of the aquifer 0836, three wells, with denomination for this work of W1, W2 and W3, were represented with red dots in Figure 5(a). The well classified as W1 is domestic. It is located in the town of "El Paso de las Vacas", with a depth of 7 m and a concessioned volume (allowed by Central Water and Sanitation Board of Chihuahua) of 1000 m^3. The well W2 is of multiple use and it is in the locality "The Chinampa", its depth is of 11 m and a concessioned volume of 1000 m^3. A third well of public use W3, located in the town of Aldama, has a depth of 180 m and a concessioned volume of 1,246,000 m^3. The values of specific volumetric activity are presented in Table 3. All were determined with a relative error of 1.6% (Colmenero, L, 2004).

It should be noted that two of the sampled wells (W1 and W2) are quite close to the maximum concentration of uranium. Unfortunately, only gamma spectrometry was available for measurements and measurements of the concentrations of ^{234}U, ^{230}Th, ^{226}Ra among other isotopes of interest could not be made. That is why the values presented are for total uranium.

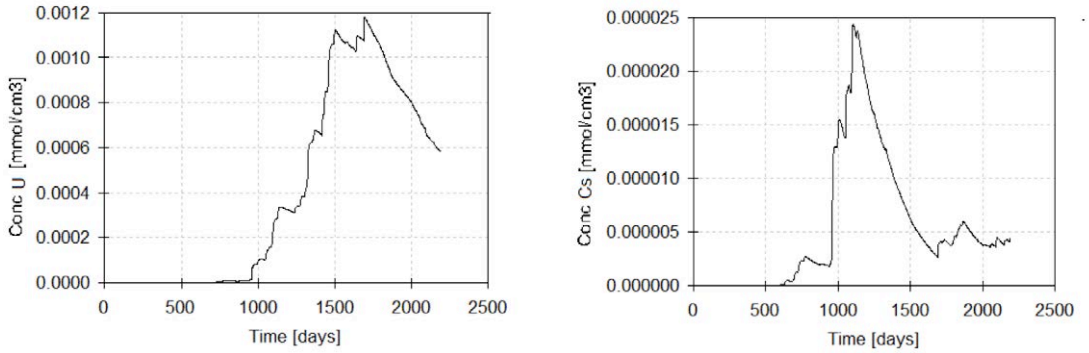
Table 3 Specific volumetric activities in three wells sampled in Aldama.

WELL	^{222}Rn ($Bq.m^{-3}$)	U_{Total} ($Bq.m^{-3}$)
W_1	39,750	1,247
W_2	35,580	1,116
W_3	34,200	1,073

The reported transmissivity values for the wells vary from 1.62 x 10^{-7} m^2/s to 0.038 m^2/s (14 m^2/day to 3196 m^2/day); while for hydraulic conductivity values ranging from 5.9 x 10^{-7} m/s to 9.46 m/s (0.050 and 8.00 m/day) are reported. The highest values of these hydraulic parameters are registered in the harvests near the Chuvíscar River.

The HYDRUS-1D program was used to predict the transport of radionuclides vertically across polluted urban land and the underlying unsaturated zone assuming a one-dimensional model of groundwater transport. Calculations were performed for a hypothetical scenario involving a transport balance. The performance evaluation was carried out using a leach and small off-site farm (Pontedeiro et al., 2010).

Figure 9(a) Concentration of total U in equilibrium and (b) Cs in groundwater.



(a) (b)

It was assumed that the radionuclides transported in the groundwater are intercepted by a well of average depth of 60 m, with downward slope from the URAMEX plant. For the safety analysis we used aquifer concentrations from wells W1 and W2. Figure 9 (a) shows the concentration of total uranium and 9 (b) of the Cs, both in the groundwater zone and their variation in mmol/cm^3 during the six years that the URAMEX plant was functioning.

Figure 10 (a) and 10 (b), respectively, show the changes in concentration of total uranium and cesium in a node located about 20 cm deep, below the root layer and during the six years that the plant was running. Green corresponds to 1985, pink to 1984, red to 1983, blue to 1982, magenta to 1981 and light blue to 1980.

The cumulative fluxes of the solutes analyzed for natural uranium and cesium through the bottom of the soil profile located at a depth of 1 m are shown in Figure 11, assuming that the capillary barrier has a thickness of approximately 20 cm.

Figure 10(a) Concentration of total uranium and (b) Concentration of Cs in the root layer for the six cutting cycles from 1980 to 1985.

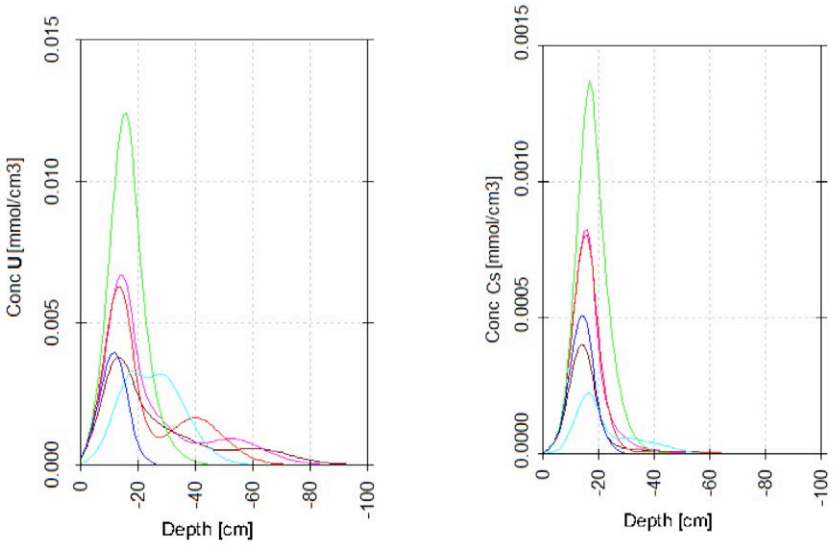
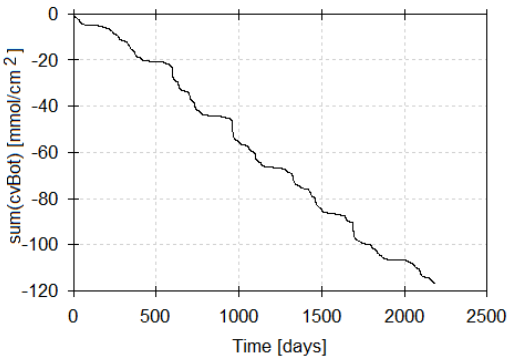


Figure 11 Cumulative fluxes of solutes analyzed for natural uranium and cesium.



SUMMARY

In this work the dispersion and accumulation of some NORM isotopes and the places with the greatest surface storage in Aldama City, Chihuahua, were studied. ^{137}Cs was included, considering that it is an anthropogenic source whose concentration varies greatly depending on the wind regimes. The experimental results were complemented with theoretical calculations based on the modeling programs WRPLOT, CALPro Plus and HYDRUS 1D, which allowed determination of the location showing increases in surface contamination and in the water table. In other words, through the modeling programs, the wind regimes were correlated with the isotopic accumulations of the NORM. Given the scarce precipitation, the cesium combined with the water discharged to clean the plant, and arrived little by little at the Region of the water table. Currently, no equivalent doses have been reported that endanger the physical integrity of the population.

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