An Overview of ^{226}Ra and ^{228}Ra in Drinking Water in Several Counties in Texas, USA

S. G. Landsberger and G. George
Enviroklean Product Development Inc. Houston, Texas, USA 77064
e-mail: shaunaL@epdi.biz

Abstract

Naturally Occurring Radioactive Material (NORM) or Technology Enhanced Naturally Occurring Radioactive Material (TENORM) can be a potential health risk. It is now well known that the underlying geology in many parts of Texas has given rise to levels of ^{226}Ra and ^{228}Ra that often exceed the limits set by the US Environmental Protection Agency. A detailed literature search was undertaken to assess the levels of ^{226}Ra and ^{228}Ra in all of the Texas counties. Several statistical evaluations of the data were performed. The Hickory aquifer in the Llano Uplift region of Texas has consistently had the highest number of ^{226}Ra and ^{228}Ra concentrations above the legal limit. As well many of the affected rural communities may not have the financial resources to rectify the problem.

Keywords: ^{226}Radium, ^{228}Radium, drinking water, Texas aquifers

Introduction

In 1979 the United States Environmental Protection Agency (EPA) set the current rules and guidelines for different radionuclides in drinking water. These guidelines were set so that all water systems could meet health standards without creating much financial burden to the towns and counties with the potentially contaminated water arising from naturally occurring radioactive material (NORM) or technology enhanced NORM known as TENORM (Hess, 1985). The Texas Commission on Environmental Quality (TCEQ, 2012) is in charge of monitoring the drinking water systems throughout the state. Both water testing and enforcement of any violations that may occur fall under their mandate. The radionuclides of most interest in the Texas drinking water are ^{226}Ra and ^{228}Ra which emit alpha and beta particles along with gamma-ray emissions, respectively. Radium is efficiently incorporated into the human body, with intake by way of food and water, and then as a chemical analogue of calcium, radium is incorporated into tissue. There it can potentially cause an array of health effects including bone sarcoma, leukemia, cancer of the mastoid and paranasal sinsuses, cancer of the upper digestive tract and orofacial cleft (Finkelstein, 1995, Fuortes, 1990, Littman, 1978, Hirunwatthanakul, 2006, Chech, 2008).

Drinking water supplies come from several sources including surface water and aquifers. The most common source of ^{226}Ra and ^{228}Ra in drinking water is from radiological decay of naturally occurring uranium and thorium deposits within the earth's crust. The ^{226}Ra and ^{228}Ra are relatively soluble and travel within the aquifer. Since ^{226}Ra has a very long half- life of about 1630 years it is able travel farther distance away from its parent than ^{228}Ra which only has a half-life of 5.75 years (Chech, 1987, Kim, 1999).

There are five aquifers in Texas that have a history of containing elevated amounts naturally occurring radionuclides. These include Hickory, Gulf Coast, Dockum, Edwards-Trinity and Ogallala aquifers (TWDB, 2012). All of these aquifers are used for multiple purposes including mining, manufacturing, irrigation, animal livestock, municipal and rural household water supply. Most of the population that uses the aquifers for a drinking water supply lives in small towns, with the bigger cities using surface water as their main source. The goal of this paper was to systematically identify the areas of Texas which have levels of the combined ^{226}Ra and ^{228}Ra exceeding the legal limits of the US EPA and to perform some statistical analyses including ^{226}Ra concentrations total alpha activity and correlation analysis of ^{226}Ra and ^{228}Ra.

Aquifer Description

The Hickory is a small aquifer that lies in central Texas and is primarily used for industry with a small portion distributed for drinking water (Hudak, 1999). The area of the Hickory Aquifer is known as the Llano Uplift region. The region is characterized by Precambrian igneous and metamorphic rocks which are exposed at the earth's surface. There are no significant uranium deposits in this area but the igneous rocks typically have elevated uranium (Chech, 1987).

The Gulf Coast aquifer is one of five major ones which reside underneath the states of Louisiana and Texas stretching into Mexico. There are deposits of high uranium concentrations within the geology surrounding the aquifer, primarily present in the Eocene and young geological formations (Chech, 1987). The region of the Gulf Coast aquifer has a history of uranium mining and there are currently several active mines including Kleberg, Duval and Brooks County with permits pending in Goliad county and an inactive processing facility in Karnes County. Currently all the active mines obtain the uranium by in situ methods whereby they inject fluids into the ground to dissolve the minerals which are then brought to the surface in the fluid and sent for processing (Texas Groundwater Protection Committee, 2012).

Both the Ogallala and Dockum aquifers are located in west Texas and the Panhandle region. The Ogallala aquifer is part of large system that spreads through other parts of western United States. The water supplies many different municipal systems in the area. The underlying geology of the aquifer contains calcrete, silcrete and lacustrine sediments which include high concentrations of uranium. The Dockum aquifer is composed of formations from the Triassic period which includes deposits of uranium within the sand and shale. There are uranium deposits throughout the area of both aquifers (Hudak, 2004). It should be noted that geology of the aquifers overlap in many parts of the Texas panhandle and water in this region can be pulled from either aquifer.

The Edwards-Trinity aquifer underlying the Texas borders are composed of three interconnecting bodies of water including the Edwards-Trinity, Trinity and Edwards aquifers. The aquifer is composed of carbonate and clastic rocks from the Cretaceous time period. There are no indications of uranium deposits in the aquifer's geology (Hopkins, 1995).

Methodology

Data were collected from the Texas Commission of Environmental Quality (TCEQ) database and the New York Times Toxic Water System Report (TCEQ, 2012). The data were evaluated in detail for each active water system within the county. The TCEQ has data as far back as 1980, which is one year after the US EPA regulations were set forth. Whenever a violation for combined $^{226, 228}$Ra was found, it was noted in the report. When a specific water system could not be accessed, the TCEQ Consumer Confidence Report (CCR) was used to ascertain any violations. The CCR is a detailed breakdown of contaminants for a shorter period of time. The TCEQ data were combined with data for Texas from the New York Times Toxic Water System Report which took water samples from 2004-2007 (New York Times Toxic Water Report, 2012). The New York Times reported ^{226}Ra and ^{228}Ra as separate entities; however, the combined concentrations of the $^{226, 228}$Ra were often above the US EPA 0.185 Bq/L (5 pCi/L) limit. In the New York Times study, water samples were collected by the Environmental Working Group from one to several times a year from municipal wells over a 4 year period. A detailed tabulation of the average and maximum concentrations of $^{226, 228}$Ra was reported. The TCEQ collected water samples since 1980 several times a year until present also from municipal wells. A times series evaluation of $^{226, 228}$Ra in all the counties of Texas was beyond the scope of this current study. Results for the TCEQ and New York Times data did not always coincide since the sampling was done by a different group at different times.

Results

A visual representation of the five major and minor Texas aquifers with a regulatory history of elevated concentrations of radionuclides is shown in Figures 1 and 2, respectively, while the results for combined and separate concentrations of $^{226, 228}$Ra are shown in Figures 3-5. The areas mapped in dark gray represent at least one violation of the US EPA regulatory limit. Exceedances varied from 1 to 195 times for all water systems within the county. In particular seven counties had fifty or more exceedances. The results showed that out of the 254 Texas counties, 62 (24.4%) of them have had concentrations above the US EPA regulatory limits for combined $^{226, 228}$Ra of 0.185 Bq/L (5 pCi/L) or greater. Even though the radionuclides are natural occurring they are considered legal violations.

The data showed that the majority of Texas counties have low levels of ^{228}Ra with the exception of six counties in the Llano Uplift region over the Hickory Aquifer including Burnet, Gillespie, Kendall, Mason, McCulloch and San Saba. In these Counties, many thousands of inhabitants have drunk water exceeding the combined $^{226, 228}Ra$ regulations.

Discussion

The data presented here only represents those counties having $^{226,228}Ra$ concentrations above the legal limit and also reported by the New Times report. Although this may be considered to be skewed it nevertheless provides a good basis for evaluation of US EPA exceedences. The range of ^{226}Ra was from 0.044-0.42 Bq/L (1.2-11.4 pCi/L) with a geometric mean of 0.22 Bq/L (5.84 pCi/L) and geometric standard deviation of 0.068 Bq/L (1.84 pCi/L). The range for ^{228}Ra was 0.0074- 0.44 Bq/L (0.2-11.9 pCi/L) with a geometric mean of 0.092 Bq/L (2.50 pCi/L) and a geometric standard deviation of 0.107 Bq/L (2.91pCi/L). The combined maximum $^{226,228}Ra$ concentration (which is the interpreted legal limit by US EPA), had an average of 0.39 Bq/L (10.5 pCi/L) with a geometric mean of 0.334 Bq/L (9.04 pCi/L) and geometric standard 0.038 Bq/L (1.03 pCi/L) with a range from 0.19-1.84 Bq/L (5.0-49.6 pCi/L). An analysis between ^{226}Ra and ^{228}Ra showed no correlation for sixty-seven results above the US EPA limits. The r-factor was 0.03 with a probability also 0.03 signifying that there were individual geological sources for these two radionuclides. The concentration of ^{226}Ra above the legal US EPA limits as compared to the concentration of the total alpha particle activity (also reported in the New York Times) had an average ratio of 0.29 with a standard deviation of 0.15. The correlation between ^{226}Ra and the total alpha particle activity for 73 samples gave an r-value of 0.69.

These aquifers are often the only source for drinking water in many of the rural areas due to topography and current drought conditions. If the severe drought situation in Texas continues the reservoir water (surface water) will be depleted thus forcing more communities to use ground water.

With the exception of a few scattered counties and those in north Texas, the majority of the cases of elevated ^{226}Ra and ^{228}Ra in drinking water are limited to the Gulf Coast, Hickory, Dockum, Ogallala, and Edwards-Trinity aquifers. The high levels of ^{228}Ra over the Hickory aquifer suggest elevated thorium in the bedrock.

A nationwide study from 1968 showed five separate Texas water systems with elevated ^{226}Ra (Hickey, 1968). This indicates that the problem with $^{226,\,228}Ra$ has been known for over 40 years and it would appear there have been few improvements in many of the smaller communities. The Llano Uplift region (Hickory aquifer) appears to have had consistently the highest levels of ^{226}Ra and ^{228}Ra.

Reducing the levels of $^{226,\,228}Ra$ can be very expensive. While larger cities can absorb the cost, smaller communities generally do not have the resources. This is particularly true in many parts of rural Texas where levels of $^{226,\,228}Ra$ levels regularly exceed US EPA standards. For instance in the City of Brady, $9 million was recently spent on a new water filtration system yet the population served is only 5,500 people (The Brady Standard, 2012). This project was specifically allocated to address the radium issue. An alternative solution is various house-hold water systems such as reverse osmosis, ion exchange, or nanofiltration (Water Research Foundation, 2012). These systems typically cost several hundred US dollars.

In 2006 the State of California (California Public Health Goals, 2006) estimated the calculated risk of cancer from drinking water with elevated $^{226, 228}Ra$ and suggested that concentrations should be much lower than the current US EPA regulations. The report concluded levels of ^{226}Ra and ^{228}Ra needed of be at the level of 1.85 mBq/L (0.05 pCi/L) and 0.703 mBq/L (0.019 pCi/L) respectively, when a person consumes an average 2 L of water per day and a child consumes an average 1 L of water per day. This level for ^{226}Ra is one hundred times less than the current US EPA standards of 0.185 Bq/L (5 pCi/L) which equates to a 99.9% reduction. Given the current state of technology for the $^{226, 228}Ra$ removal achieving this level would prohibitively expensive if not impossible to implement.

Conclusion

The ^{226}Ra and ^{228}Ra concentrations in the Texas drinking water coming from naturally occurring uranium and thorium deposits throughout the state have been a problem for many years. The areas over the Hickory aquifer have the longest documented occurrence of $^{226, 228}Ra$ in the water as well as some of the highest levels within the state. A greater than 99% reduction these levels to a proposed California report may be impossible to implement. There may be a severe economic impact to smaller communities to adhere to the US EPA regulations.

References

1. Aksoy, A., Al-Jarallah, M., Al-Haddad, M.N., 2002. Natural Radioactivity in the Scale of Water Well Pipes. Journal of Environmental Radioactivity. 61, 33-40.
2. California Environmental Protection Agency 2006. Public Health Goals for Chemicals i4n Drinking Water-Radium-226 and -228. Office of Environmental Health Hazard Assessment.
3. Chech, I.M., Prichard, H.M., Mayerson, A., Lemma M.,1987. Pattern of Distribution of Radium 226 in Drinking Water of Texas. Water Resources Research. 23 1987-1995.
4. Chech, I., Patnaik, A., Burau, K.D., Smolensky, M.H., 2008. Spatial Distribution of Orofacial Cleft Defect Births in Harris County Texas, and Radium in the Public Water Supplies: A Persistent Problem? Texas Medical Association. 104, 55-63.
5. Finkelstein, M.M., Kreiger, N., 1995. Radium in Drinking Water and Risk of Bone Cancer in Ontario Youths: A Second Study and Combined Analysis. Occupational Environmental Radiation. 53, 305-311.
6. Fuortes, L., McNutt, L.A., Lynch, C., 1990. Leukemia Incidence and Radioactivity in Drinking Water in 59 Iowa Towns. American Journal of Public Health. 80, 1261-1262.
7. Hess, C.T., Michel, J., Horton, T.R., Prichard, H.M., Conglio, W.A.,1985. The Occurrence of Radioactivity in Public Water Supplies in the United States. Health Physics. 48, 553-586.
8. Hickey, J. L.S., Campbell, S. D., 1968. High Radium-226 Concentrations in Public Water Supplies. Public Health Reports. 83, 551-557.
9. Hirunwatthanakul, P., Sriplung, H., Geater, A., 2006. Radium-Contaminated Water: a Risk Factor for Cancer of the Upper Digestive Tract. Asian Pacific Journal of Cancer Prevention. 7, 295-298.
10. Hopkins, Janie, 1985. Water Quality in the Edwards-Trinity (Plateau) Aquifer, Edwards Plateau and Trans-Pecos, Texas. TWDB (Texas Water Development Board).
11. Hudak P. F., 1999. Chloride and Nitrate Distributions in the Hickory Aquifer, Central Texas, USA. Environment International. 25, 393-401.
12. Hudak P. F., 2004. Radioactivity in the Ogallala and Dockum Aquifers, northwest Texas, USA. Environmental Geology. 47, 283-289.
13. Kim, Y, 1999.The Concentrations and Distributions of U and Th in Paleozoic Aquifers Surrounding the Llano Uplift Area, Central Texas, U.S.A.: Applications to the Sources of Ra and Rn in Groundwater. Geosciences Journal. 3, 201-211.
14. Littman, M.S., Kirsh, I.E., Keane, A.T., 1978. Radium-Induced Malignant Tumors of the Mastoid and Paranasal Sinuses. American Journal of Roentgenology. 131, 773-785.
15. New York Times-Toxic Water Report http://projects.nytimes.com/toxic-waters
16. Texas Commission of Environmental Quality www.tceq.state.tx.us/ 2012
17. Texas Ground Water Protection Committee (TGPC) www.tgpc.state.tx.us 2012

18. Texas Water Development Board (TWDB) www.twdb.state.tx.us/ 2012
19. The Brady Standard-Herald Newspaper and Heart of Texas News 2012. How City Water Wells, Brady Lake and Water Treatment Plant Work.
20. Water Research Foundation www.waterrf.org 2012

Acknowledgments: We gratefully thank Kristina Tolman for her assistance in the ARC GIS software.

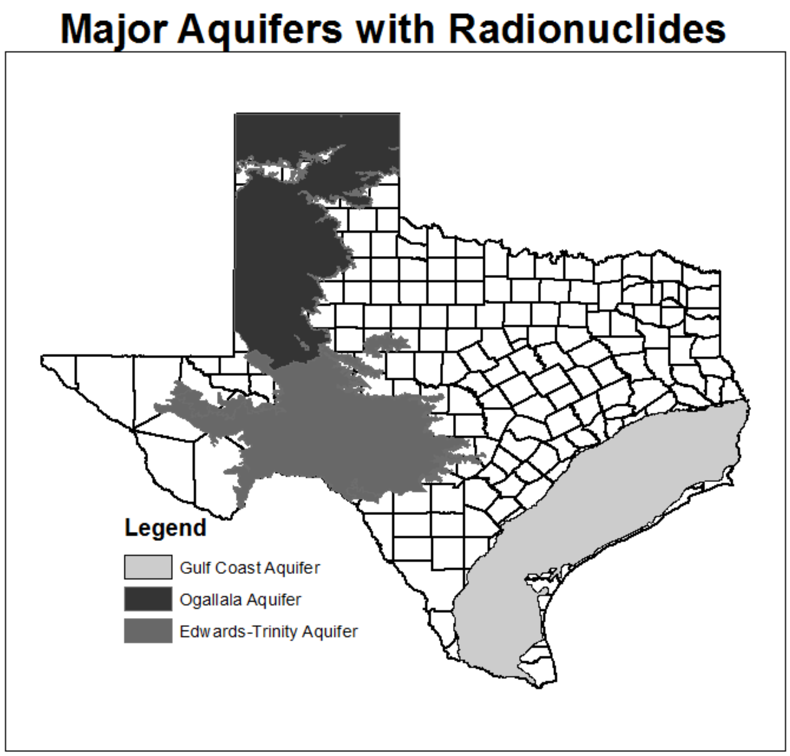

Figure 1: Major Texas Aquifers with a Regulatory History of Elevated Concentrations of Radionuclides

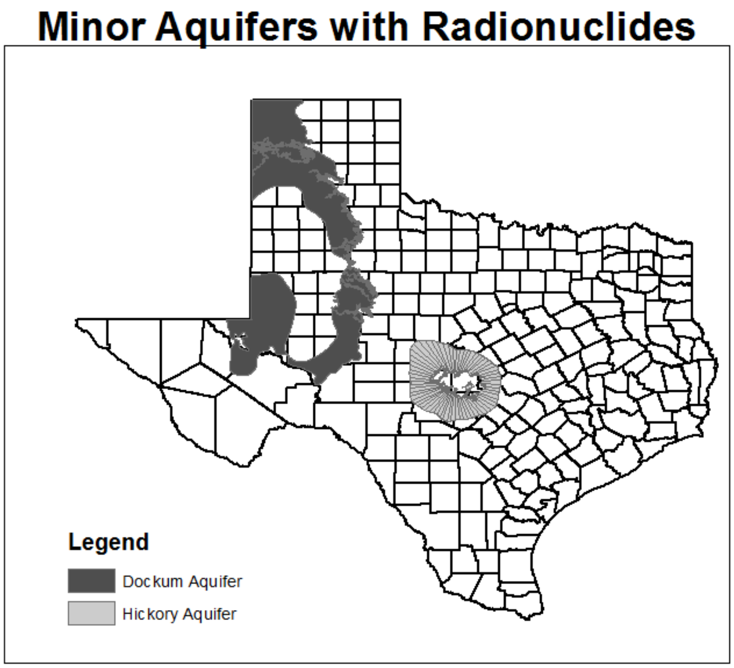

Figure 2: Minor Texas Aquifers with a Regulatory History of Elevated Concentrations of Radionuclides

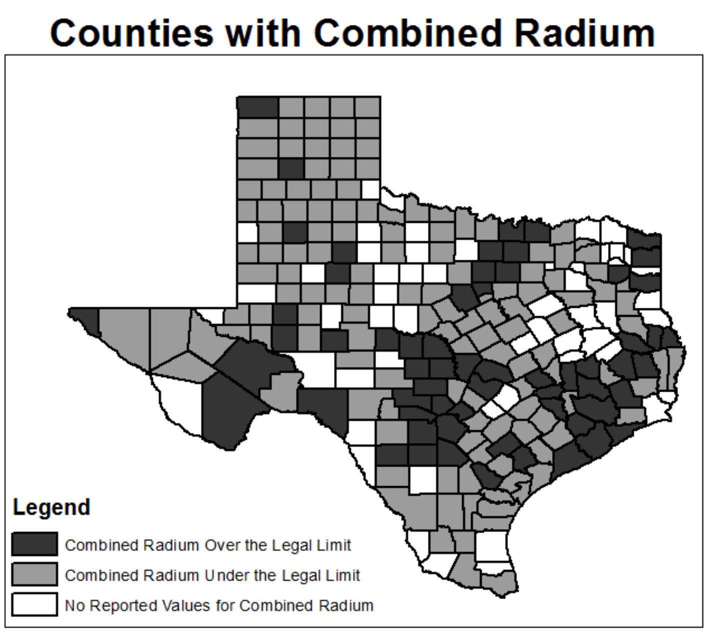

Figure 3: Map of the counties considered: those in dark gray counties had combined ^{226}Ra and ^{228}Ra over the legal limit and those in light gray counties consistently had combined ^{226}Ra and ^{228}Ra under the legal limit set by the USEPA, and those in white had no reported values.

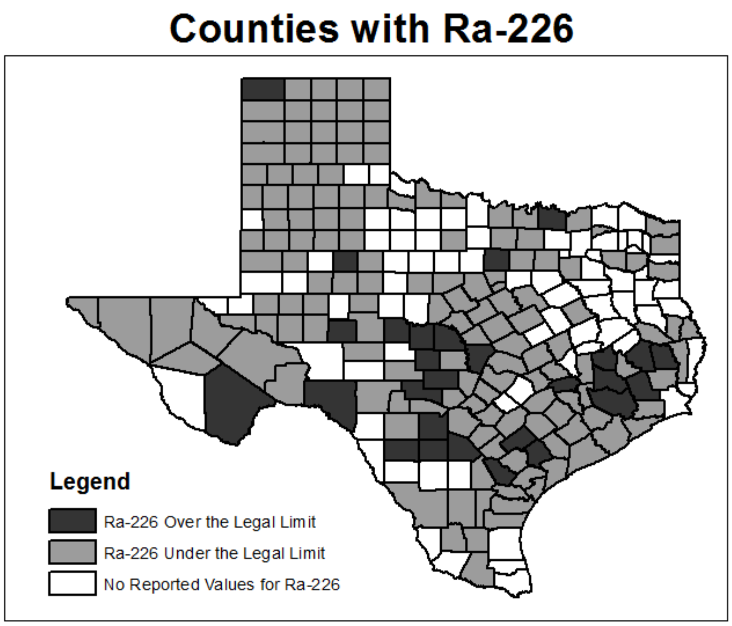

Figure 4: Map of the counties considered: those in dark gray counties had ^{226}Ra over the legal limit and those in light gray counties consistently had ^{226}Ra under the legal limit set by the USEPA, and those in white had no reported values.

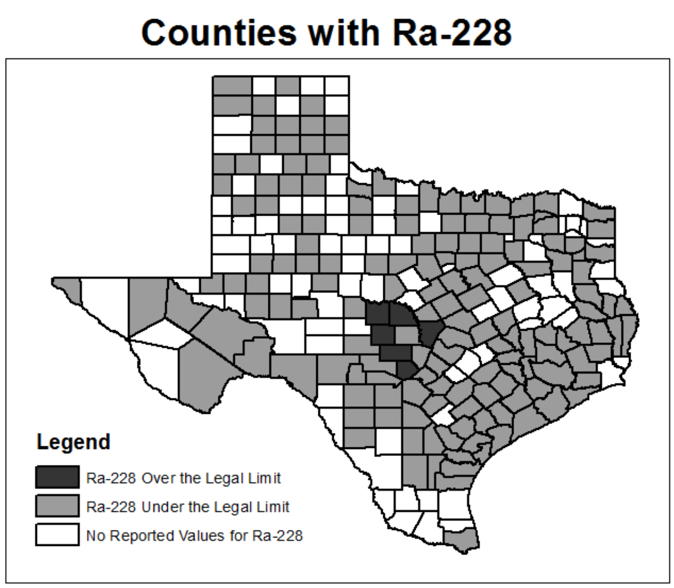

Figure 5: Map of counties considered: those in dark gray counties had ^{228}Ra over the legal limit and those in light gray counties consistently had ^{228}Ra under the legal limit set by the USEPA and those in white had no reported values.