Radon in Ground Waters of the South Texas Uranium District

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Abstract

Uranium and its radioactive daughter products are ubiquitous in the South Texas tertiary environment. The effects radon gas in groundwater upon the inhabitants of the counties where uranium mineralization occurs has not been sufficiently addressed in current scholarly literature. This work reviews the literature that is available for radon emanation in South Texas, the physics of the emanation of radon into ground waters, the distribution of public and private water wells relative to zones of uranium mineralization and the geologic controls upon uranium deposition. We present a GIS based model of the spatial factors controlling radon concentrations in ground waters in the proximity of the uranium district, the distribution and classification of the wells selected by the model and the preliminary results of model verification as determined by sampling and analyzing a subset of the wells determined to be at risk of radon contamination.

Introduction

Radon is a colorless, tasteless, and odorless gas that comes from the decay of uranium. It is the heaviest naturally occurring noble gas. Radon occurs in trace concentrations and frequently provides characteristic signatures identifying the nature of its source. Due to its radioactive nature it can be a health hazard by increasing the risk of lung cancer (Reimer and Tanner, 1992; USEPA, 2000a). The movement of radon in the geologic environment is of research interest to earth scientists because of the varied applications for which it can be used; earthquake prediction, tracing atmospheric and oceanic circulation (Reimer and Tanner, 1992), ground water flow (Gascoyne et al., 1993) to name a few.

Radon concentrations in ground waters represents a health concern as sixty-eight percent of our exposure to natural sources of ionizing radiation usually comes from radon (USEPA, 2000b). Radon gas usually moves from the ground up and migrates into homes and buildings through cracks and other holes in their foundations. The buildings trap radon inside, where it accumulates and may become a health hazard if the building is not properly ventilated. Elevated levels of radon in ground waters will contribute to the levels indoors as radon outgases during water use.

By breathing air containing a large amount of radon, the radiation can damage the lungs and eventually cause lung cancer should the radon decay while in the lungs. The USEPA cites radon as the second leading cause of lung cancer in the United States estimating 7,000 to 30,000 deaths a year from radon-induced lung cancer. Only smoking causes more lung cancer deaths and smokers exposed to radon are at higher risk than nonsmokers (USEPA, 2000a). If the radon gas

decays to a solid while in the lungs it becomes lodged there. This concentrates the energy from the ionizing radiation released by its daughter products in the surrounding cells leading to malignant cell growth.

On November 2, 1999 in the Federal Register (64 FR 59246) the EPA proposed a Maximum Contaminant Level (MCL) of 300 pico Curies /liter (pC/l) for radon in Public Drinking Water Supply Wells (those serving 25 people or more). As with many proposed MCLs, this standard has not yet been enforced. Nor does this MCL for radon apply to the thousands of private wells in South Texas that draw water from zones of uranium mineralization for domestic use. Regardless of the final MCL for the new EPA Radon standard a number of water wells of the South Texas Uranium district will likely far exceed the criteria. Initial measurements have confirmed this hypothesis.

The most regionally applicable study found to date is "Radon in the Coastal Plain of Texas, Alabama, and New Jersey" (Gundersen and Peake, 1992). This Geological Society of America (GSA) special publication addresses soil radon concentrations through transects across the Coastal Plains states relevant to the underlying geologic formations. Two transects were conducted in Texas. The first transect ran across the Cretaceous formations near Dallas and the second across Tertiary formations from Austin to Houston. This second transect however turns eastward before reaching the South Texas Uranium District and thereby avoids the known areas of uranium mineralization.

One study of indoor radon concentrations has been conducted in Texas by the Texas Dept. of Health. "The Texas Indoor Radon Survey" (Smith et al., 1994) sent 4000 activated charcoal canisters out to the entire state. Of the 4000 statistically distributed canisters over 2500 were returned and used in a County based analysis. While the purpose of this work was a state wide survey, many counties in the zone of uranium mineralization are absent in the final results. Review of the results does not particularly lend support for this study from its findings but rather highlights again the lack of radon data in the zone of uranium mineralization.

Radon Emanation

Before an atom of radon can migrate into the soil pore spaces it must escape from the site of its parent isotope radium (^{226}Ra). Under steady-state conditions the fraction of radon atoms formed in a solid that escape from the solid is defined as the emanating power of the solid by Tanner (1978). Other historical terms cited by Tanner include the coefficient of emanation, escape ratio, escape-to product ratio, and percent emanation.

Upon the decay of a radium atom most of the energy is carried of as ionizing α radiation. The remaining energy (still 10^4-10^5 times that of normal chemical bonds) carries of the resulting radon atom by recoil. The typical recoil distance

within the grain where decay takes place is 20-70 nm for minerals of common density. Similarly the radon recoil distance is greatest for air at 0.06 mm and an intermediate 0.1 µm in water (Reimer and Tanner, 1992). If the radon atom recoils in the direction of the mineral grain boundary and the site of decay was near enough to the boundary the radon may terminate in the pore space. It is also possible that the radon atom will terminate its recoil in an adjacent mineral grain and therefore stand very little chance of escaping. The direct recoil fraction is the amount of radon that actually makes it into the pore spaces (Tanner, 1978). Once recoil has terminated in the pore space the radon atom is subject to Fickian diffusion and advective flow for its half-life of 3.82 days.

Reimer and Tanner (1994) cite experiments with crystals in which radium is uniformly distributed confirm theoretical calculations that the emanating power of minerals should be on the order of 1% or less, but because of the tendency for radium to be sited near the interstitial space in weathered rocks and most soils they emanate a substantial amount of the radon generated in them. Since the radon atom's range in air is greater than the lengths of most trajectories in the pores of rocks and compacted soils it is unlikely to terminate its path in an air-filled pore. When the recoil path is through a liquid (water) in crack, joint, capillary, or pore, the radon atom is much more likely to come to rest in a place from which it can diffuse through the rock or soil. Consequently sorbed water and capillary water significantly increase the emanating power in porous materials. On average, the emanating power of rocks is less than 10% and soils typically emanate 15 -50% of their radon (Reimer and Tanner, 1992; Rogers and Nielson, 1991; Schery et al., 1984; Tanner, 1978).

The discrepancy between the 1% radon emanation of the homogenous crystal model of Reimer and Tanner (1994) and the average emanation values should be attributed to the propensity of radium to adsorb strongly onto mineral surfaces thereby essentially coating the mineral grains and providing additional sites of radon emanation not accounted for in the crystal model (Wanty, 1972).

Purpose

Due to the heterogeneity of South Texas Tertiary environment (Galloway, 1982) and the spatial nature of the input datasets this study would not be possible without the extensive use of GIS. In fact it is this author's penchant for the use of GIS in environmental studies that leads to the development of this work as a masters thesis in Environmental Science at Texas A&M University – Corpus Christi.

An extensive GIS database has been developed to geospatially correlate the areas of uranium mineralization and their relationship to public and private water supply wells (Beaman and McGee, 2002). Measurements of radon concentrations in a subset of these wells are ongoing and preliminary results are reported below. The measurements will become model verification and training

data for a GIS Based Decision Support System to evaluate the risk of excessive radon levels in private wells. Wells are sampled based on their proximity to zones of uranium mineralization, their locations within the channel sands of the aquifer and their accessibility for repeat measurements. These measurements will both assess the ability of the model to predict areas of potential high radon concentrations in ground waters and help evaluate the potential health impact to the region. Should the GIS based model prove valid funding will be sought to extend the sampling area.

Hypotheses

- Many private water wells of the South Texas Uranium district will likely far exceed the proposed criteria of 300 pC/l
- Wells located within the paleo-channel sands are more likely to contain elevated levels of radon
- Wells located outside of the paleo-channel sands are less likely to contain elevated levels of radon
- A GIS based model is applicable to predicting Radon concentrations in ground waters of the South Texas Tertiary Environment

Study Area

The Gulf Coast Uranium Province lies along the northwestern Gulf of Mexico in South Texas and adjacent Mexico. The Eocene Whitsett (Ej), Oligocene Frio (Of), Oligocene and Miocene Catahoula (Mc), Miocene Oakville (Mo), and Pliocene Goliad (Pg) Formations form the western boundary of the province. The northern boundary is based on the farthest northward extent of volcanic ash facies in Eocene and younger rocks. These host formations were formed by fluvial systems that prograded gulfward into near-shore and eventually true marine environments. A major source of volcanic ash was derived from an Oligocene volcanic arc, probably in the Big Bend country of West Texas. All the uranium deposits in the province are marginal marine roll-front sandstone deposits. Contemporaneous listric faults are most likely the source of the H_2S gas reductant for most of the uranium deposits (Eargle et al., 1977; Finch, 1996; Moxham, 1964).

Wells that are candidates for radon testing are determined in part using GIS and in part by practical consideration such as agreement from the well owner and accessibility. The combination of the well locations, this authors "Geodatabase of the South Texas Uranium District" (Beaman and McGee, 2002), and the initial radon measurements are the basis for the development of GIS model. The spatial analysis tools in ArcGIS will provide a ranking of the wells for the likelihood of finding high radon concentrations. Radon data gathered under the methods cited below are progressively added to the geodatabase. The final goal will be to determine a risk assessment for radon groundwater of South Texas.

While the above description makes the entire coastal plain of Texas a candidate for this study the data sets used in the GIS model are limited in extent. Further geographic limitations in the logistics of obtaining the samples for calibration and verification arise based upon travel cost. For this study the area of interest is limited to Gonzales, Karnes, Live Oak, Atascosa, San Patricio and Duval counties. This arbitrary delineation should be representative of the natural variability of the depositional systems examined. Figure 1 below presents a generalized map of the study area.
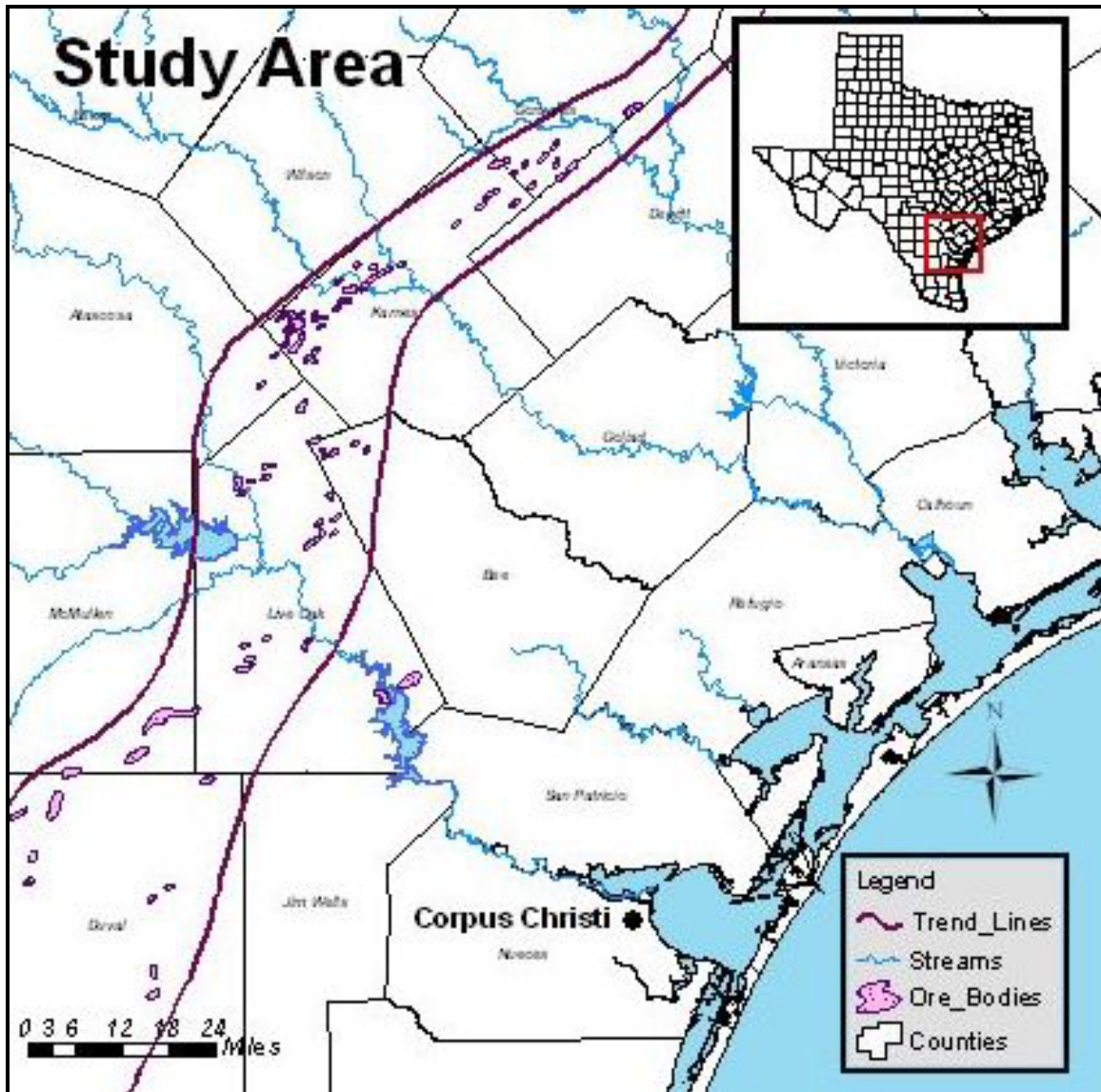

Figure 1. Study Area showing Mineralization Trend lines and Ore bodies after Adams and Smith (1981).

Methods

Concentrations of radon in ground water are now relatively simple and inexpensive to measure. The selected method for this study uses a passive environmental radon monitor, the E-PERM® electret ion chamber (EIC) (Kotrappa et al., 1981). A charged Teflon disk characterized by a measurable surface voltage is installed into an electrically conducting chamber, the combination becomes an EIC. Exposure of the EIC to air that emits ionizing radiation reduces the charge in the electret. In our case the air is emanating from a water sample located in a sealed jar below the EIC. The recorded voltage drop, the times of sampling, analysis and the radon decay constant allow for calculation of the radon concentration. The operational principles of these monitors are fully described elsewhere (Kotrappa et al., 1998). While originally designed to monitor airborne radon, the minimal effect of humidity upon these monitors makes them suitable for this application. E-PERMs measure radon gas concentrations as low as 10 pCi/L (0.37 Bq /L) in air with < l0% error when measured over 1 day. Electrets of different sensitivities and chambers of different volumes are used in conjunction with different measurement time periods to capture different concentrations. Currently six measuring set ups are available for this study. The procedure is outlined below.

Sampling Procedures

- Well waters are allowed to flow freely until the temperature is stable.
- Escape of Radon from water due to turbulent flow is minimized by running a hose from the wellhead faucet into a bucket
- Water samples are collected in 134-mL sample bottles with Teflon-lined screw caps.
- Two samples are taken at each sampling event for reproducibility
- Bottles are filled and capped underwater without any airspace left in the sample
- Time of sample, well depth (if known), owner contact information and GPS coordinates are logged.
- Samples are generally analyzed within 24 hours

Analytical Procedures

- A sealable glass analysis bottle (similar to a gallon pickle jar) of known volume is placed on its side
- The lid of the sample bottle is then removed and the sample bottle is quickly placed in an upright position in the clip on the bottom of the analysis bottle
- The glass analysis jar screw cap with an attached E-PERM electret (pre-measured and open) is screwed onto the bottle.

- The analysis bottle is then turned upright spilling the water out of the sample bottle
- The analysis bottle is sealed with a special rubber collar and shaken a few times to help release radon into the gas phase.
- Starting time of analysis and initial Electret voltage is logged.
- The steps taken from opening the sample bottle to closing the lid of the analysis bottle are accomplished quickly to minimize radon loss from the sample to the atmosphere
- After measuring for at least 24 hours the rubber collar and the screw cap are removed
- End time of analysis and the final Electret voltage are measured and recorded

The data on the initial electret voltage, final electret voltage, and the time period of exposure are used to calculate average radon concentration in the air phase during the exposure period. This result is then used to compute the radon concentration in water using the theory developed by (Kotrappa and Jester, 1993). The pertinent equations, variables and time constant tables can be found in the EPERM System Training Manual "Radon in Water" (Kotrappa, 1999). Photos of the set-up used for this work as well as additional information can be found at http://www.radelec.com/.

There are several potential sources of error in this radon analysis method. The most significant error is the accidental discharge of voltage from the electret due to water, fingers or particulate matter in the air contacting the electret surface. This discharge would lead to a misleading high value. This type of error may be the reason for the large difference between the two radon concentrations measured for Well TD01 in Table 2 below. At this stage both values were kept in the preliminary dataset and the average of the two values was used. All measurements will have at least one duplicate. In cases when discrepancies are observed between duplicates, additional measurements will be conducted. At this initial stage of the study all measurements are kept. False low values can occur when a sample bottle is not securely sealed allowing radon to escape before analysis. Another source of false lows is attributed to the elevated levels of methane gas in many of these samples. Even if the sample is successfully collected without an air pocket the methane content often produces up to centimeter sized bubbles that provide space for the radon to escape the water before analysis. This type of error has been minimized by conducting the replicate analysis simultaneously with the sample. Note that this procedure also decreases substantially the number of wells analyzed per field sampling trip. The repeatability of the measurements can be further assessed by considering the results of duplicate measurements in Table 2. The main discrepancy is observed for the previously discussed well TD01. Other smaller discrepancies are observed for wells RJ01 and FS02 however the magnitude of the difference in these duplicate measurements is not considered a problem for the purpose of this study.

GIS Analysis

The initial GIS methods will be very similar to the approach of (Brown et al., 1993) who mapped the radon potential as a calculated annual radon entry rate from underlying soils into a reference house. The approach consisted of:

- Creating map units and delineations involving a combination of soil map units and geologic map units
- Defining soil profile and geologic properties for each map unit
- Calculating a numerical radon potential for each map unit
- Grouping adjacent map units with similar potentials
- Collapsing the units into zones according to their radon potential

For this study the digitized maps of the channel sands and ore bodies from (Adams and Smith, 1981) will be geo-processed into radon potential. Adams and Smith (1981) present a model for successful exploration practices in south Texas in the Department of Energy report entitled "Geology and Recognition Criteria for Sandstone Uranium Deposits in Mixed Fluvial-Shallow Marine Sedimentary Sequences, South Texas". The rational of the Adams and Smith model can be summarized as a numerical flow chart solution that begins with regional geological information and then is refined by local exploration. While these maps were classic analog cartographic products, the exploration model is sound and should readily translate into a GIS environment. These maps have been georeferenced and digitized. The appropriate resulting geospatial information will be used to produce polygons of areas most likely to contain elevated radon levels in ground water. A summary of the GIS datasets complied for this work is presented as Table 1 below.

Data Set	Type	Source	Date	Processing
Surface Uranium Mines	V	Tx. Railroad Comm.	2000	CAD to GIS
Uranium Location Data	V	USEPA	2003	Geodatabase
Wells	V	CWSS, TAMU-CC	2003	Geodatabase
Wells	V	TWDB	2003	Geodatabase
Aquifers	V	TWDB	2003	Geodatabase
Geology	V	R. Gomez, CWSS	2002	Digitization
Mineralization Trend	P	DOE	1981	Digitization
Channel Sands	P	DOE	1981	Digitization
Ore Bodies	P	DOE	1981	Digitization
Faults	P	DOE	1981	Digitization
Surface Radiation	P	USGS	1961	Digitization
Digital Ortho Photos	R	USGS	1995	None
Digital Elevation	R	USGS	2001	None
Geology	R	Tx. BEG	1998	None

Table 1. Summary of the GIS data sets collected for the model where Type: V = Vector, P = paper, R = Raster

Wells sampled for this preliminary research are generally "randomly" selected within the study area outlined above. Candidate wells are random in that currently the "Word of mouth" method is employed. Once a well is sampled, the results are reported to the owner and the owner is then asked for the contact information for relatives and friends that might be amenable to having their wells sampled. In other words the sampling locations are (at this time) not premeditated by the GIS. Ideally this will generate data points both inside and outside the radon potential polygons from the geo-processing steps outlined above. Choosing well locations for sampling from the Texas Water Development Boards (TWDB) and Center for Water Supply Studies (CWSS-TAMUCC) GIS data has been problematic for two reasons. Primarily the current well location files contain only approximately 30% of the wells that actually exist (Hay, 2003). Second, time and budget constraints do not allow for a "Door to Door" salesman approach to sampling. While the sampling itself may only take 30 minutes or so the delivery of the layman's description of problem and the need for the sampling can take a couple of hours. Combined with driving time to and from the location this could easily result in only one sample a day. There is also the time required to report the results to the owner, but a conscientious owner can accelerate the process by the "Word of Mouth" or "Grapevine" and line up a number of candidates for the next sampling run. Once a substantial number of radon measurements have been accumulated, the location of verification and validation samples can be addressed in a more scientific GIS based method.

Data collection and model development are currently underway. Water samples are collected and analyzed at every opportunity. The relative simplicity of the analytical method allows for analysis in the field. Sampling trips are being planned for this summer. The GIS model development is an ongoing effort begun in 2000. Population of the model with radon concentrations occurs as soon as the value is determined.

Results

Sampling efforts to date have confirmed the following:

- The hypothesis that private water wells in the South Texas Uranium District may exceed the proposed MCL for Radon of 300 pCi/L. 18 of 25 wells sampled have exceeded the MCL. Two by an order of magnitude (see results in Table 2 below).
- Wells exist that are not captured in the current GIS well location datasets, of the 25 wells sampled so far only one directly corresponds to a TWDB location.

The results to date are presented in Table 2 below. Most wells were sampled in duplicate and two wells sampled again at a different dates. The average value is used in the maps below. Well depth is recorded when known. Water samples are classified into three possible types.

- WHD = Well Head, the preferred location, minimal opportunity for loss of radon and most representative of the groundwater levels.
- FCT = Faucet, a faucet nearest to the well. This class of sample is used when no hose bib exists to drain the volume tank.
- CST = Cistern, often wells are driven by windmill and flow into an open cistern. The class least likely to contain elevated levels of radon.

Of the 25 wells sampled to date the range of radon in pCi/L is 42 – 4813. The mean is 678, better that double the proposed MCL of 300.

Well_ID	Depth	SampType	RN_1	RN_2	RN_3	Av.
JS01	450	WHD	4745	4688	5006	4813
HL01	220	WHD	755	507	496	586
CD01	125	WHD	415	532		474
WW01	120	FCT	429	442		436
TD01	240	WHD	5523	938		3231
RJ01		FCT	654	257		456
RC01	480	WHD	330	349		340
OC01		WHD	55	153		104
MS01	240	WHD	239	275		257
MM01		FCT	500	498		499
ME01		WHD	336	325		331
MD01	350	WHD	301	182		242
MB01	150	WHD	711	640		676
LO01	240	WHD	385	411		398
LM01	320	WHD	254	352		303
KH01	452	WHD	821	802		812
KA01	225	CST	204	86		145
JW01	250	WHD	608	574		591
HP01		WHD	290	205		248
GK01		WHD	564	629		597
FS02	305	WHD	415	750		583
FS01S	200	WHD	394	542		468
EH01	540	WHD	112			112
CL01		FCT	42			42
LC01	280	WHD	211			211

Table 2. Summary of the Radon concentrations measured so far for the study
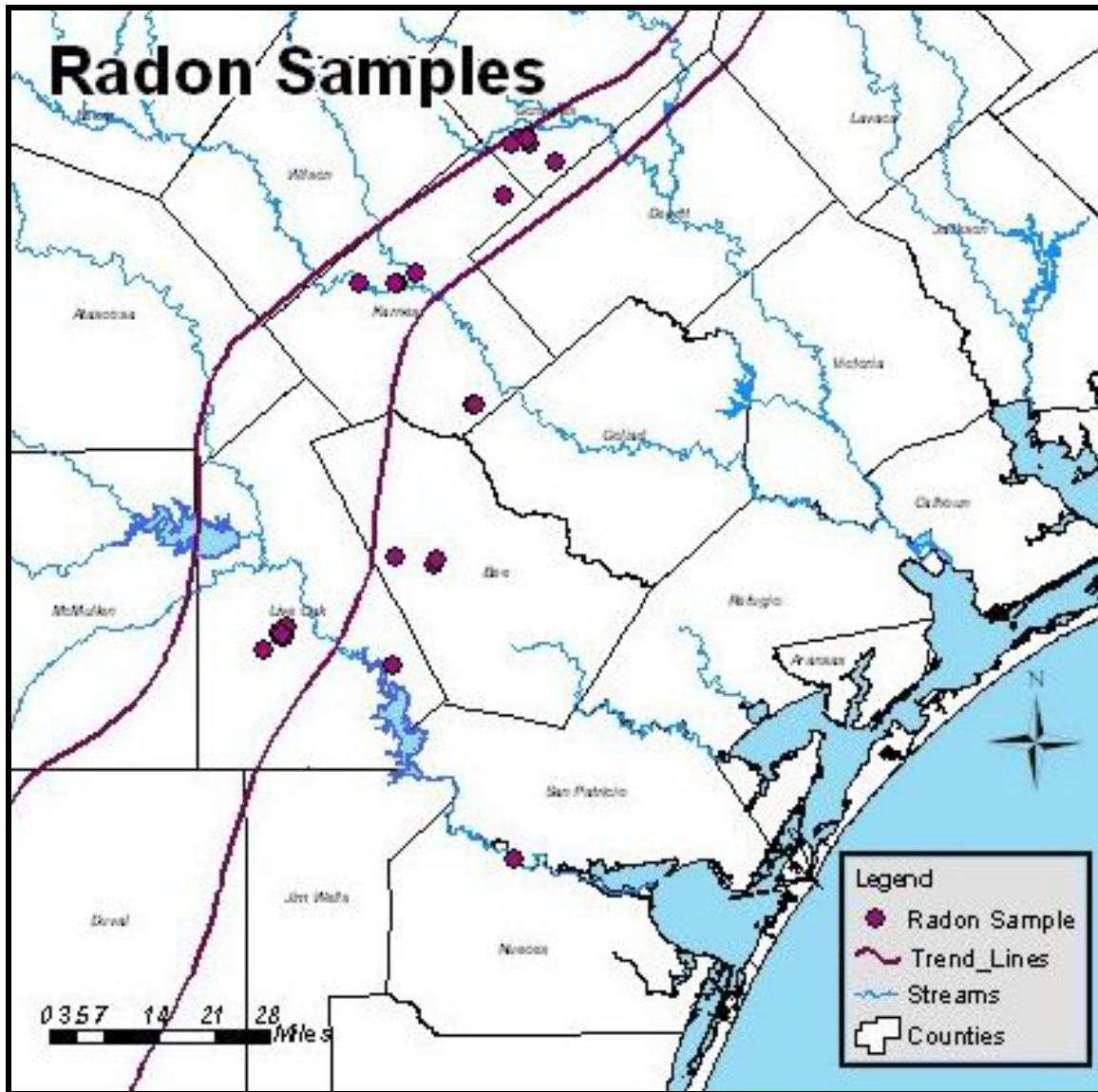
Figure 2. Radon Sample locations relative to the mineralization trend lines.

Figures 3 through 8 below depict the locations of the samples and their values against the channel sands and ore bodies digitized from Adams and Smith (1981) at a larger scale. These figures are presented as a qualitative preview of the preliminary model design to validate the hypothesis above.
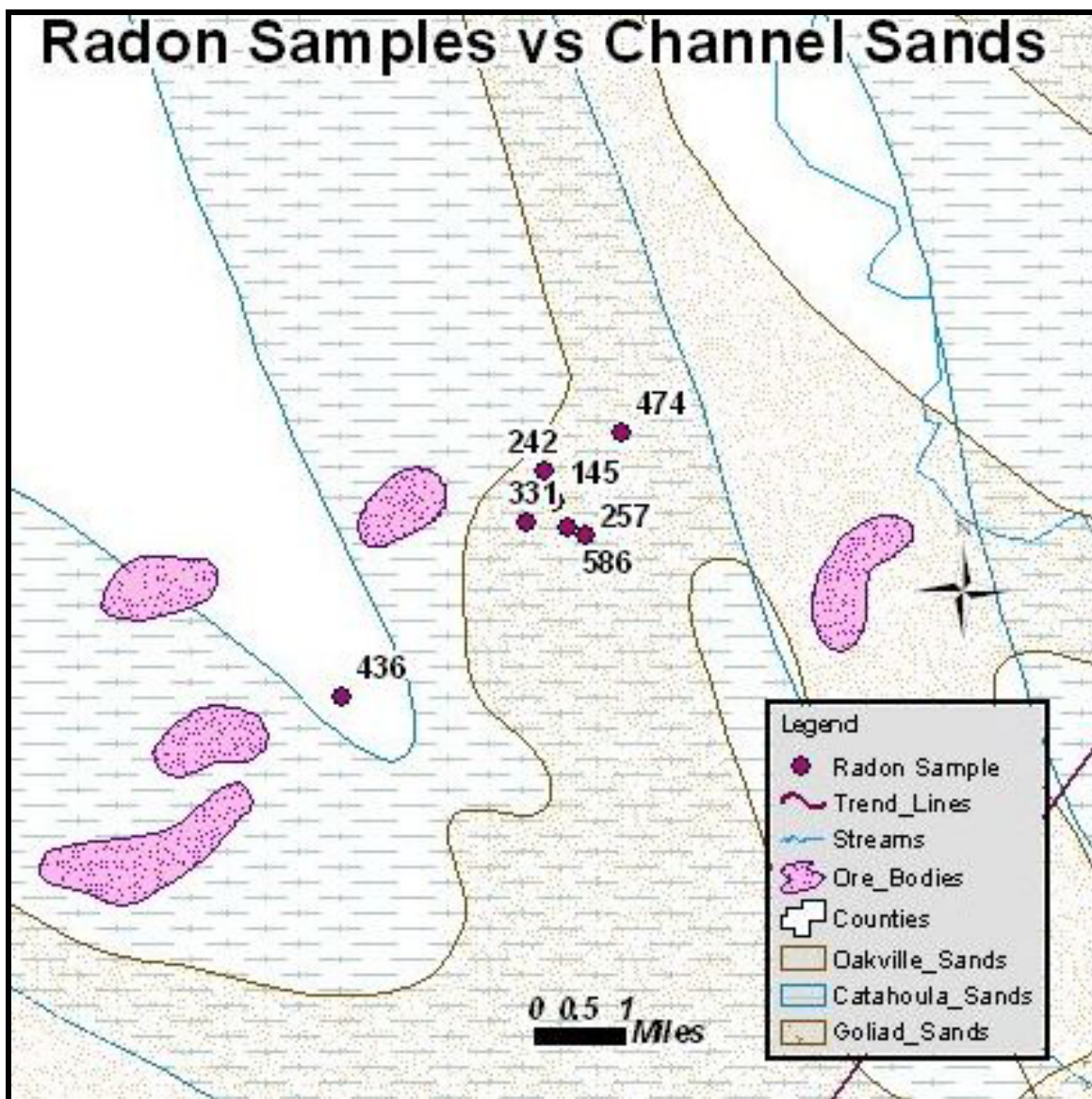
Figure 3.   Radon Samples vs. Channel Sands and Ore bodies. Note the clustering effect due to the "Word of Mouth" sampling scheme. This effect will be minimized as further sampling is conducted.
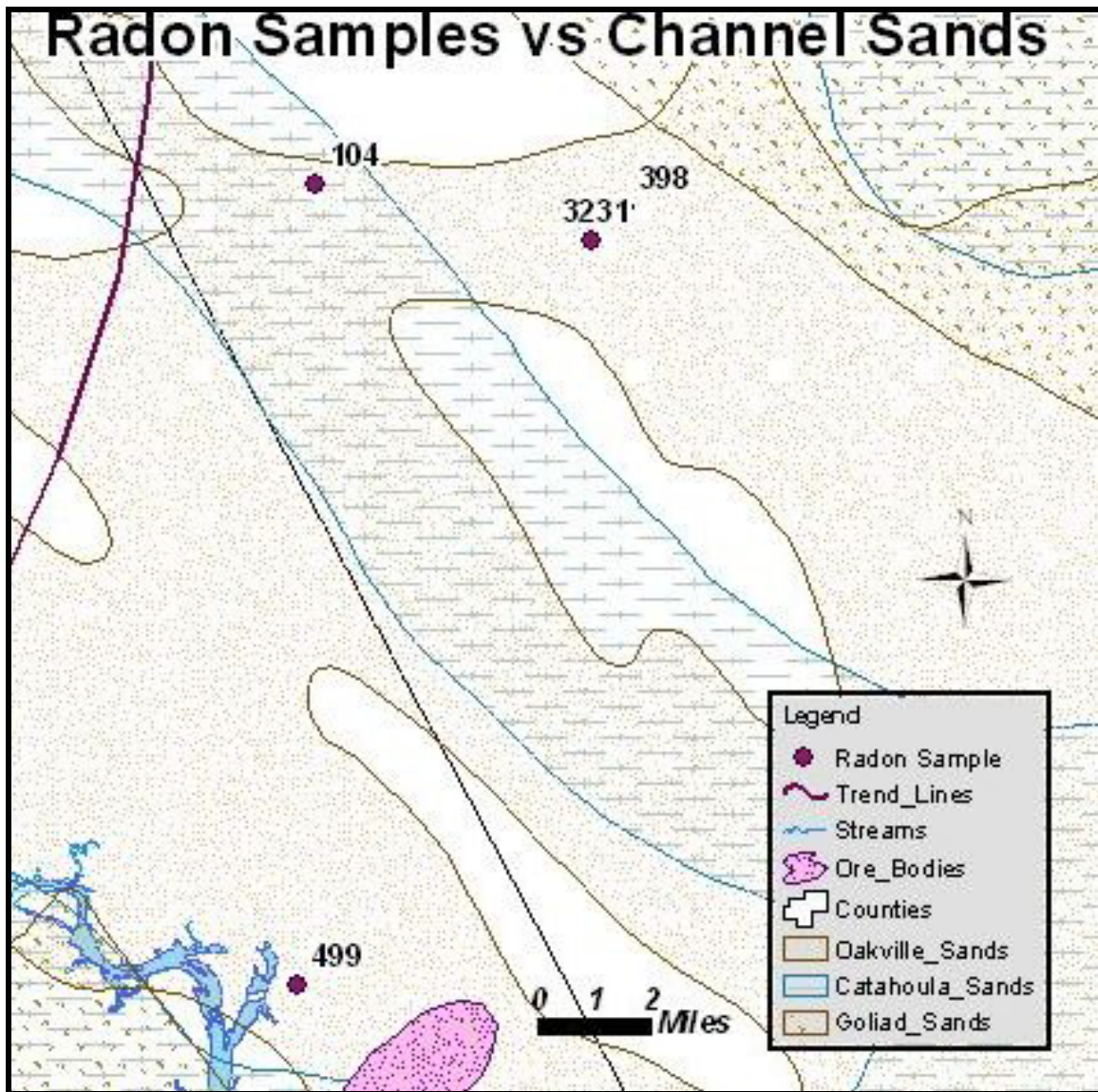
Figure 4. Radon Samples vs. Channel Sands and Ore bodies.
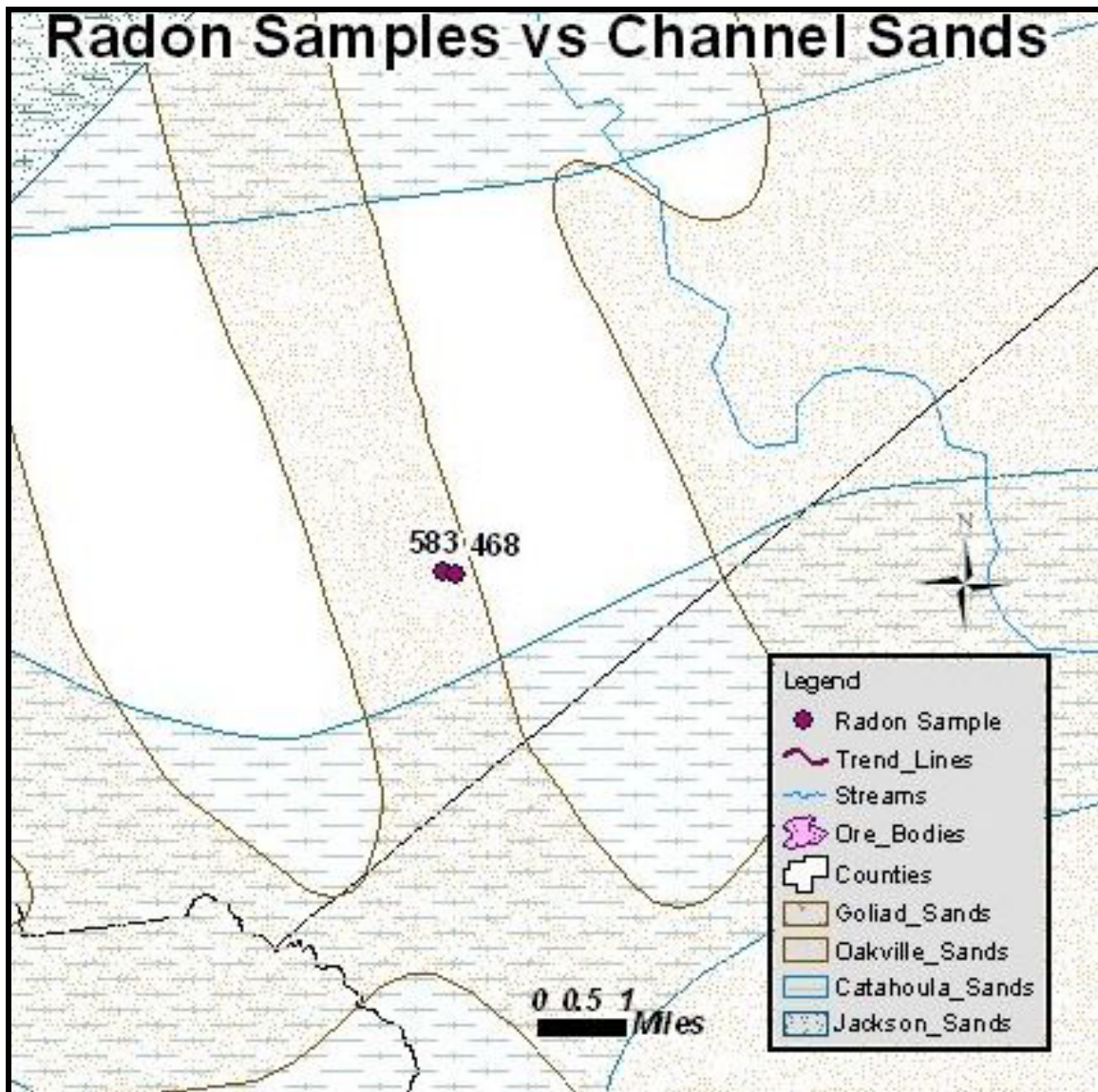
Figure 5. Radon Samples vs. Channel Sands and Ore bodies. This figure suggests sampling near but outside of the channel sands for model validation.
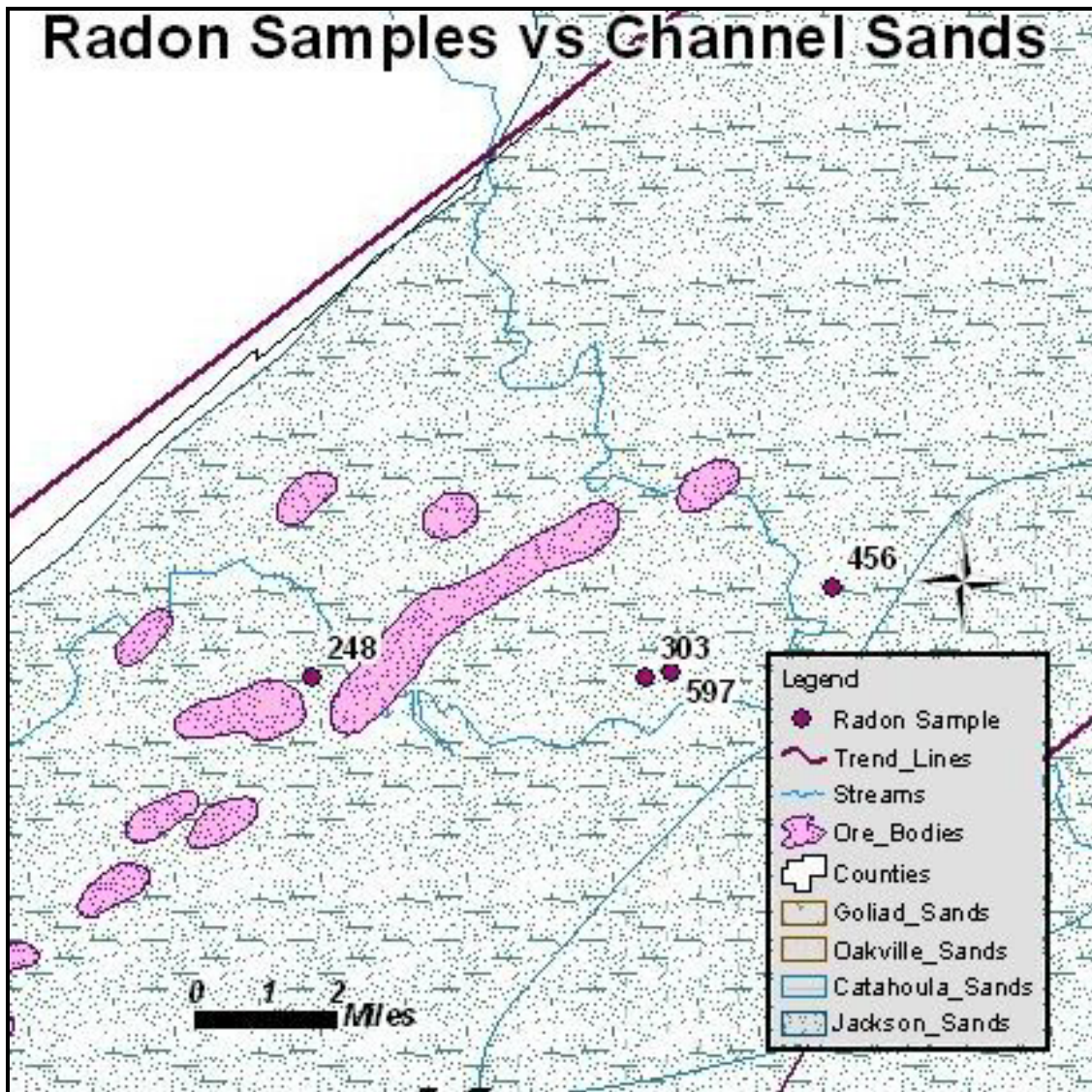
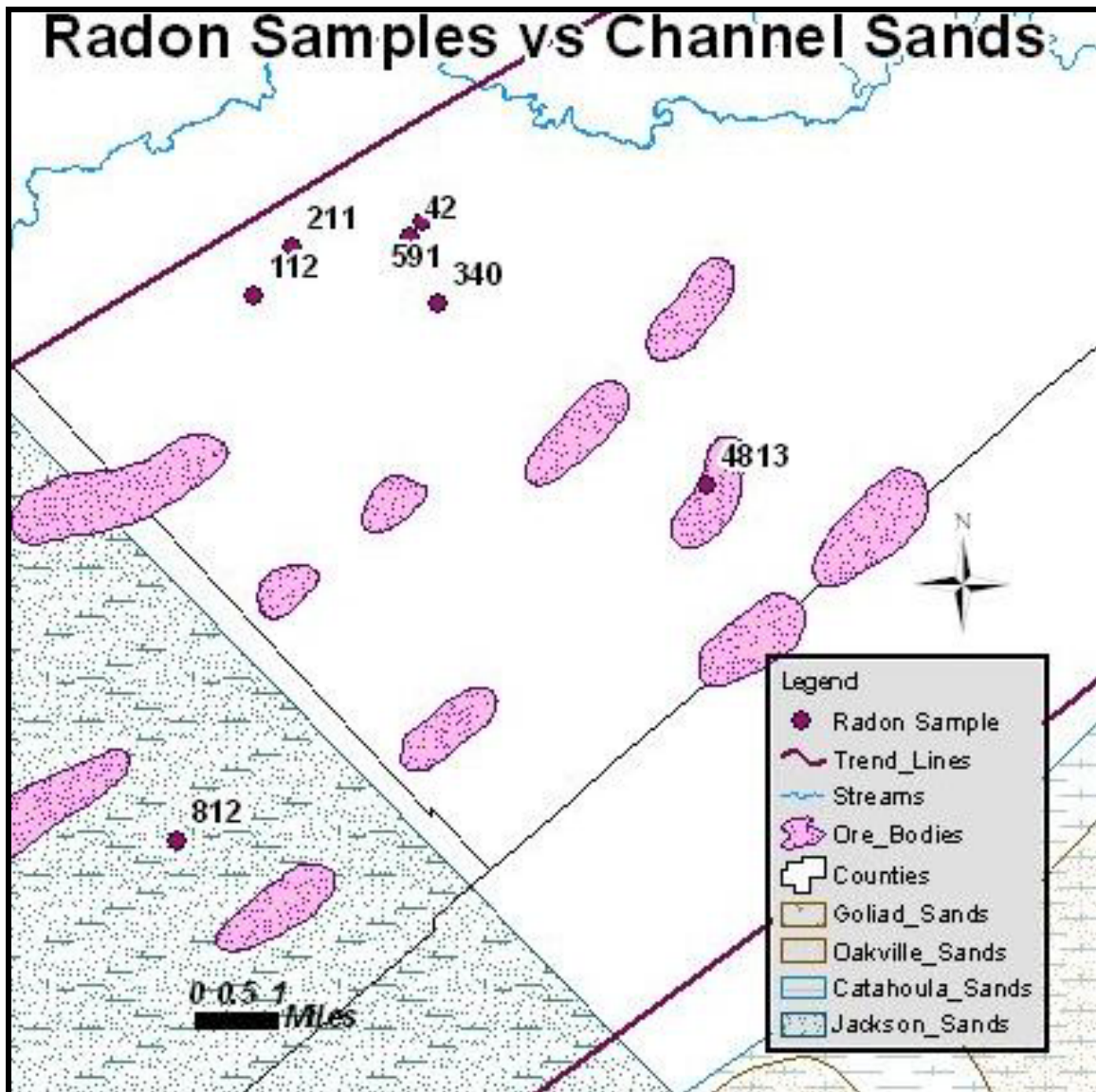Figure 6. Radon Samples vs. Channel Sands and Ore bodies. Note the low value near the ore body.

Figure 7. Radon Samples vs. Channel Sands and Ore bodies. These samples were taken within the mineralized zone but outside of the extent of the channel sands dataset. Note the highest value to date lies within an un-mined ore body.
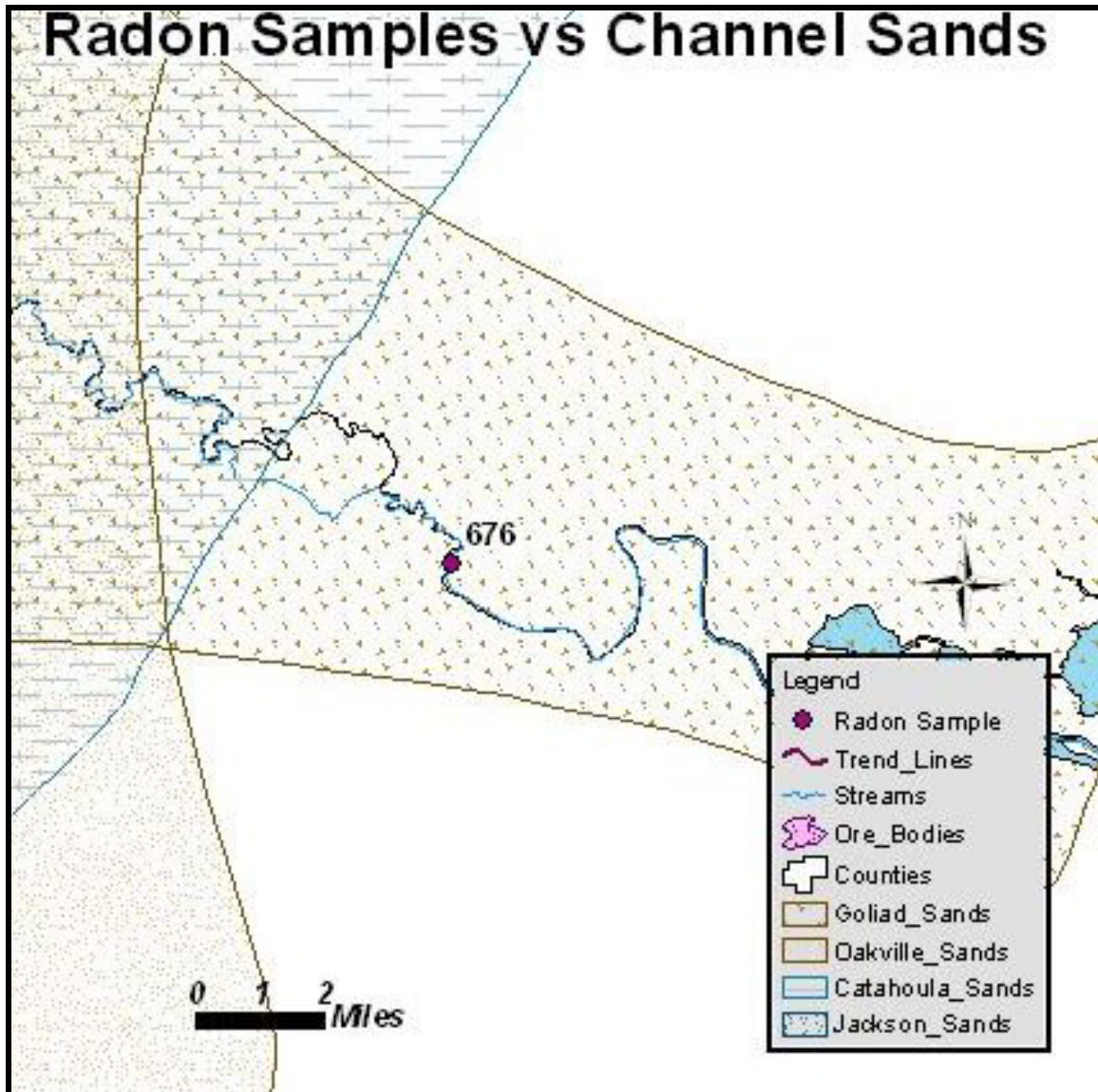
Figure 8. Radon Samples vs. Channel Sands and Ore bodies. This sample is in the Goliad channel sands and well out of the mineralized zone.

Conclusions

A GIS based model is being developed to evaluate the likelihood of measuring high, above EPA standard, Radon concentrations in the private water wells of the South Texas Uranium district.   Relevant information including water wells locations, Geological information, ore body locations has been scanned, rectified, digitized and imported into a geodatabase. Radon measurements in water wells are presently being conducted and included in the geodatabase. Based on the preliminary results the hypothesis that private water wells in the South Texas Uranium District may exceed the proposed MCL for Radon of 300 pCi/L will likely be accepted. So far 18 of 25 wells sampled have exceeded the MCL and two

wells have exceed the standard by an order of magnitude. These preliminary results reemphasize the need for such studies in the South Texas Uranium district. As the number of wells sampled is still relatively small, it is not yet possible to assess the potential of the GIS tool to accurately predict the occurrence of high Radon readings in water wells.

Acknowledgements

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