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Statistical Analysis of Elevated Radium and Gross Alpha Measurements in the Sanitary Landfill

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1. INTRODUCTION

In 2002, radium (Ra) 226 and 228 measurements elevated above the 5 pCi/L groundwater protection standard (GWPS) and gross alpha measurements above the 15 pCi/L GWPS were noticed in several groundwater monitoring wells at the SRS Sanitary Landfill (SLF). An additional four quarters of confirmatory measurements for Ra in the SLF groundwater were taken during 2003 as directed by the SC Department of Health and Environmental Control (DHEC).

Elevated radium concentrations in groundwater of the Aiken County area are a common occurrence. Price and Michel (1990) compiled radium concentrations in drinking water wells of this area and showed several instances of the concentrations exceeding the regulatory limit. Ra226 is an alpha emitter and contributes much of the natural alpha radioactivity found in uncontaminated groundwater. Thus, the elevated radium concentrations are usually accompanied by elevated gross alpha concentrations. Appendix A2 indicates that this is the case at the SLF where Ra226 accounts for almost all elevated gross alpha.

1.1. Problem Statement

The Annual 2002 Sanitary Landfill Groundwater Monitoring Report (WSRC 2003) shows exceedences for radium and gross alpha in the groundwater from 4th quarter 2002 through 3rd quarter 2003. In addition, the 2003 data show that 17 monitoring wells in the SLF network are confirmed as having elevated Ra226, or 228, or a sum of these radioisotopes above the 5 pCi/L GWPS. The 17 are LFP series wells 5WP, 6WP, and 13WP and LFW series wells 8R, 18, 23R, 29, 36R, 41R, 45D, 47C, 57B, 62B, 64C, 66B, 67B, and 69C.

The problem presented to SRTC was whether these elevated data are consistent with groundwater impacted by SLF leachate or whether they are typical of natural conditions and therefore unlikely to have derived from anthropogenic inputs.

1.1.1. Focus of Solution

The solution to this problem will require the comparison of recent Ra measurements among these 17 monitoring wells to the past measurements in the same wells, and, if possible, comparison of the data from the SLF network to other groundwater monitoring well data from radiologically unimpacted SRS and non-SRS facilities.

1.1.2. Working Hypothesis

Because the facility in question is a sanitary landfill and therefore has no record of receiving radioactive waste, the working hypothesis is that the appearance of these radionuclides in recent groundwater samples is non-anthropogenic in origin.

1.1.3. Lines of Evidence

To accept the above hypothesis, it is requisite to demonstrate multiple lines of evidence as a convincing argument in favor of that hypothesis. Statistical data display methods will be used to compare recent and historical SLF radium data and to compare SLF radium data with that from other on- and off-Site wells, and correlation analysis will be used to examine relationships of SLF radium with different constituents and water quality parameters.

The principles of geochemistry suggest that elevated measurements of some radioisotopes should be correlated with elevated measurements in other analytes. Geoscience will be used to develop additional lines of evidence in support of the hypothesis by comparing Ra concentrations to other groundwater chemistry and water quality measurements as discussed in section 3.3 below.

2. MONITORING WELL NETWORK DESCRIPTION

The SLF groundwater monitoring network consists of LFW and LFP monitoring wells that are sampled regularly and often quarterly for a variety of constituents of concern (COCs) as specified in the 1992 SLF Postclosure RCRA Part B Permit Renewal Application (WSRC 1993). A more thorough description of the groundwater monitoring program for the SLF can be found in the Sanitary Landfill Groundwater Quality Assessment (GWQAP) (WSRC 1995).

3. DATA AND METHODS

This section describes the data used to generate the multiple lines of evidence in support of the working hypothesis and the statistical and geological science methodology used to test the working hypothesis.

3.1. Data Sources

Analytical, field, and depth to water data and well information for the Sanitary Landfill LFP and LFW series wells were retrieved from ERDMS, the Environmental Restoration Data Management System, for this analysis. Version 8.2 of the SAS System for Windows from SAS Institute was utilized to obtain and analyze the data and to produce various types of plots.

The analytes of concern are Ra226, Ra228, and gross alpha (ALPHAG). Analytes investigated for possible correlation with/ impact on the analytes of concern include other rads, barium (BA), and calcium (CA). Turbidity, pH, alkalinity (as CaCO₃), and purge volume were the field analytes studied. For the analytical data, the well, analyte name and testcode, sample date and time, analysis date, sample type (to exclude field QC samples), analysis code (to exclude lab QC results), analyte type (to exclude tentatively identified compounds-TICs and surrogates), sample matrix (groundwater only), result and units, review and lab qualifiers, qualification codes, lab, analytical method, filter code (for metals), sample quantitation limit (SQL) and units, and validation & verification status were pulled. The subset of variables applicable to field measurements was retrieved for the field and depth to water data.

For the analytical data, the result qualifier was defined from the lab and review qualifiers, and

the units were all converted to a consistent basis for the result and the SQL. Detection was determined from the result qualifier, and then an adjusted result for nondetects was set as follows. Half of the SQL was the preferred estimate for nondetects, as is standard with environmental data. If the SQL was missing or invalid, the result was used: half of the result for chemical analytes and the result itself for rad analytes (or zero for rad results less than zero). Data with missing sample dates, invalid or inconsistent units, rejected results (result qualifier of R), or invalid results (validation and verification status of VI) were deleted. The field and depth to water data were similarly screened. Hold times for Ra226, Ra228, and gross alpha were assessed as the time from sampling to analysis, and none of the hold times exceeded the current limit of 180 days. Water elevation was calculated from the well reference elevation less the measured depth to water.

The analyte testcode Ra2628 was created to represent the sum of Ra226 and Ra228. The highest result for each well, analyte, sample date, and lab was identified from detected data. If there were no detects, the highest adjusted result from the nondetects was chosen. The selected Ra226 and Ra228 results were added together, and a detection value was determined to indicate whether the Ra2628 value represented a below detect, detect, or combination (Ra226 detect and Ra228 nondetect or vice versa).

In addition to the concentration of the radium analytes, the ratio of Ra228 to Ra226 was examined. The max detected result for each well, analyte, sample date, and lab was identified, and then the isotopic ratio was calculated from these results. Detects only were included since ratios of differing detection limits do not have much bearing on concentration ratios.

The analytical data were then averaged sequentially to obtain quarter averages for the correlation analyses. Data for an analyte in a well were averaged over replicates to derive method averages, over methods to derive lab averages, over labs to derive sample date averages, and over sample dates to derive quarter averages. Note that adjusted results were substituted for nondetects in the calculations. Field data and water elevations were processed in the same way to return quarter averages.

ERDMS has information for 124 LFP/LFW wells. Each of the wells was assigned a category based on the concentrations of Ra2628 and the associated sample dates. Concentrations of 5 pCi/L or above are considered elevated. Wells with elevated detected Ra2628 results from 2003 to present were classified as "Ra recent," and wells with elevated detected Ra2628 results from prior to 2003 only were labeled as "Ra old." Wells with no elevated detected Ra2628 but with elevated detected gross alpha were classified as "ALPHAG only." Some wells had no elevated concentrations of Ra2628 or of gross alpha or had no data at all. Of the 124 wells, there were 17 "Ra recent" wells, 57 "Ra old" wells, 14 "ALPHAG only" wells, 23 wells with no elevated concentrations, and 13 wells with no data. This breakdown reveals that 2/3 of the wells with data exhibited a problem with radium at some point. The focus of the analysis was on the 17 wells with recent Ra2628 elevated values: LFP 5WP, 6WP, 13WP; LFW 8R, 18, 23R, 29, 36R, 41R, 45D, 47C, 57B, 62B, 64C, 66B, 67B, 69C.

Refer to Appendix A1 for a listing of all SLF wells with summary information. Appendix A2 compiles detection data for all radioactive analytes from the SLF wells.

Data from the SLF wells were compared to data from other well groups on Site and to data from a study of natural radioactivity conducted in the local area (Price and Michel 1990). The DCB 18, 19, 21, 22, and 24 well clusters; P series wells; and C series wells on Site are known to have natural radium only. Based on Ra228/Ra226 ratios, Denham et al. (1999) concluded that the primary source of radium in the DCB wells is the native soils. The evidence suggests that acidic groundwater from the nearby coal storage pile leaches radium from soils of the aquifer. This, combined with some radium from the coal itself, results in elevated radium concentrations. The P series wells were installed at the SRS to provide information on groundwater flow and quality in uncontaminated background areas. The C series wells are similar but were installed off-Site by the state of South Carolina. (The C series wells had no data in ERDMS.) The data from the study of natural radioactivity were divided into groups of on-Site and off-Site wells. Since some of the non-SLF data had extremely high detection limits, only detected data were involved in the comparison.

3.2. Statistical Methods

Perhaps the simplest and most illustrative statistical method for providing evidence for or against a hypothesis is in the use of data displays. This report provides

- Time series plots – to illustrate the contaminant concentration data over time relative to the GWPS
- Box-and-whisker plots – to illustrate the comparison of Ra measurements in the SLF groundwater to Ra measurements in other on- and off-Site groundwater monitoring wells
- Correlation analyses – to describe the tendency of Ra measurements to increase or decrease in relation to corresponding increases or decreases in other measured values such as water quality variables
- Simple linear regression (SLR) and Orthogonal regression models – to compare the slopes of the linear relationships between Ra and other groundwater variables suggested by the correlation analyses.

A box-and-whisker plot (or simply boxplot) consists of a box around the middle 50% of the data, with a center line representing the median and a dot for the average. Lines called whiskers are drawn from the 25th quantile (bottom of box) to the smallest observation and from the 75th quantile (top of box) to the largest observation. Comparison of boxplots is a nonparametric approach to data analysis, meaning that assumptions about the distribution of the data population are not needed.

Simple linear regression (SLR) is a method in which a predictor variable is used to model a response variable using a straight line. The best fit minimizes the differences between the predicted and observed values of the response variable. Orthogonal regression differs from SLR in that there is assumed to be measurement uncertainty (i.e. variation) in regressor (X) variable as well as the response (Y) variable. This method then finds the best estimate of the slope and intercept parameters for the fitted line by minimizing the sum of the squared perpendicular distances from the X,Y coordinates of each data point to that regression line and not to the X axis as is typical in SLR.

Common logarithms were used in many of the plots and analyses, because the data on the original scale cross more than one order of magnitude.

The time series plots and boxplots were generated using the SAS[®] 8.2 statistical computing software on a PC running Microsoft Windows 2000. All other statistical analyses were performed using the JMP[®] 5.01 software on the same computer platform. Both are products of the SAS Institute, Cary, NC. The statistical methods described above are explained in SAS (2000) and JMP (2000) reference documentation.

3.3. Geochemistry

Radium is a naturally occurring radionuclide in the alkaline earth group of elements. Its geochemical behavior is similar to other alkaline earths such as calcium, strontium, and barium. In uncontaminated aquifers, Ra226 is generated from decay of U238 while Ra228 is produced from decay of Th232. Most of the radium in an aquifer is bound in minerals that contain uranium and thorium but can be released to groundwater if the mineral dissolves. Alpha recoil can also eject radium from the lattice of a mineral directly into groundwater. Once in groundwater, radium is subject to adsorption on aquifer mineral surfaces. Adsorption/desorption is related to chemistry of the groundwater with adsorption being stronger with increasing pH. Thus, it is expected that radium concentrations in groundwater should correlate with certain other parameters.

Prior to developing geochemical arguments about observed radium behavior in groundwater, it must be determined whether the data are a true reflection of groundwater conditions. Sampling conditions or analytical error can lead to the appearance of elevated concentrations of radium. The sampling condition most likely to affect radium concentrations is collection of turbid samples. Radium, as with other metals, can adhere to clay particles in a turbid sample (depending on sample preparation methods), resulting in radium concentrations greater than the actual dissolved concentration. If this is the cause, radium should correlate with turbidity. Analytical errors or biases can also give the appearance of elevated radium concentrations. If this is the case, elevated radium isotope concentrations should not correlate with other parameters.

The factors that affect concentrations of other alkaline earths in groundwater should also affect radium concentrations. However, any relation between radium concentrations and concentrations of non-radioactive constituents may be obscured by the large difference in mass concentrations between radium and other constituents. When converted to mass concentrations, 10 pCi/L of Ra226 is 3.1×10^{-14} moles/L; 10 pCi/L of Ra228 is 1.1×10^{-16} moles/L. Most other natural constituents are present at concentrations that are several orders of magnitude greater than these. For example, groundwater with a barium concentration of 1 ug/L (7.3×10^{-9} moles/L) and a radium concentration of 10 pCi/L contains about 200,000 times more barium ions than Ra226 ions and about 66 million times more barium ions than Ra228 ions. Thus, correlative concentrations of radium and most non-radioactive constituents may not be detectable, because measurable changes in radium concentrations often correspond to undetectably small changes in other constituents.

Sulfate concentrations can also be correlative with radium concentrations if a sulfate phase such as barite (BaSO_4) is near saturation in the aquifer. Increases in sulfate concentration will cause the sulfate phase to precipitate, removing radium by co-precipitation. In this case, radium would be inversely correlated with sulfate. Conversely, positive correlations could result from dissolution of a sulfate mineral containing radium or could simply be coincidental. However, barium concentrations in SLF groundwater show no relation to sulfate concentrations and are well below saturation with barite. This indicates that another non-sulfate phase is controlling barium concentrations and probably radium concentrations. Thus, any trend of radium with sulfate may be a spatial rather than a geochemical relation or may reflect independent co-variation of radium and sulfate with another parameter. If the trend occurs only within the defined plume boundaries, then it is likely that some property of the landfill leachate is capable of mobilizing radium and sulfate. The property could be high ionic strength, acidity, high chloride concentrations, dissolved organic matter, or a combination of these.

This emphasizes the fact that elevated radium concentrations in groundwater associated with a waste site can be caused by three mechanisms:

- Leaching of radium from disposed materials in the waste site
- Leaching of natural aquifer radium by some component of the waste site leachate
- Natural variations in groundwater chemistry

If the radium is from natural variations, then the variations in concentrations should be consistent with variations in groundwater from uncontaminated areas of the same region.

Radium isotopes are an important indicator of the origin of elevated radium concentrations in groundwater. A daughter of $\text{Th}232$ with a half-life of 5.75 years, $\text{Ra}228$ will reach secular equilibrium with $\text{Th}232$ in less than 60 years. A daughter in the decay chain of $\text{U}238$ with a half-life of 1600 years, $\text{Ra}226$ will take over 2 million years to reach secular equilibrium with $\text{U}238$. At a site where processed $\text{Th}232$ was disposed, $\text{Ra}228$ concentrations may be elevated from decay of the $\text{Th}232$. In contrast, elevated concentrations of $\text{Ra}226$ cannot be derived from disposal of processed $\text{U}238$ within a reasonable time frame. Thus, an explanation for elevated radium concentrations

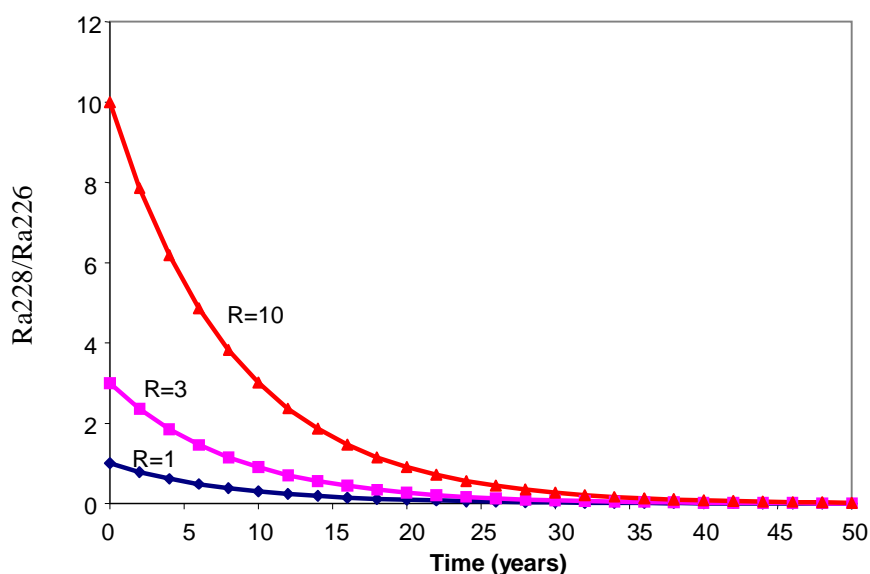


Figure 1. Calculated $\text{Ra}228/\text{Ra}226$ ratios with time. R is the initial ratio.

must account for the Ra228/Ra226 ratio, as well as for the actual concentrations.

Radium emanating from a point source such as a disposal site should show a systematic variation in the Ra228/Ra226 ratio within the plume due to the different half-lives of the two isotopes. As radium moves with groundwater, the ratio decreases because of Ra228 decay relative to Ra226 (Figure 1). Consistent Ra228/Ra226 ratios over a large area are suggestive of a native rock rather than a point source for radium within a waste site.

4. RESULTS

The data results will be in two formats, viz., within individual wells and across all wells. Ra isotopic and gross alpha (ALPHAG) data were used to generate data displays for each well in the SLF; however it is sufficient to portray these results for only the 17 recently identified wells with elevated Ra measurements.

4.1. Data Reliability

Prior to performing statistical analyses for testing the working hypothesis, concerns relative to data reliability were resolved first. This was accomplished by comparing the relationships between Ra226 and Ra228 versus ALPHAG. Although Ra228 is not an alpha emitter, it is often related geochemically (see section 3.3) to Ra226 and therefore should show a positive correlation with ALPHAG. Also, each of these variables is usually analyzed by different methods, and laboratory measurement error should not propagate between the methods.

Figure 2 is a plot of quarterly measurement data for all SLF wells and shows that both Ra226 and Ra228 are, in fact, positively correlated with ALPHAG. This supports the expected relationship based on geochemistry, and therefore the data obtained from the Site environmental database (ERDMS) are considered to be reliable.

4.2. Spatial Evidence

Figure 3 is a spatial map of the SLF monitoring well network that also includes special symbols (black stars) lying on top of the well locations to show the source of the recently elevated

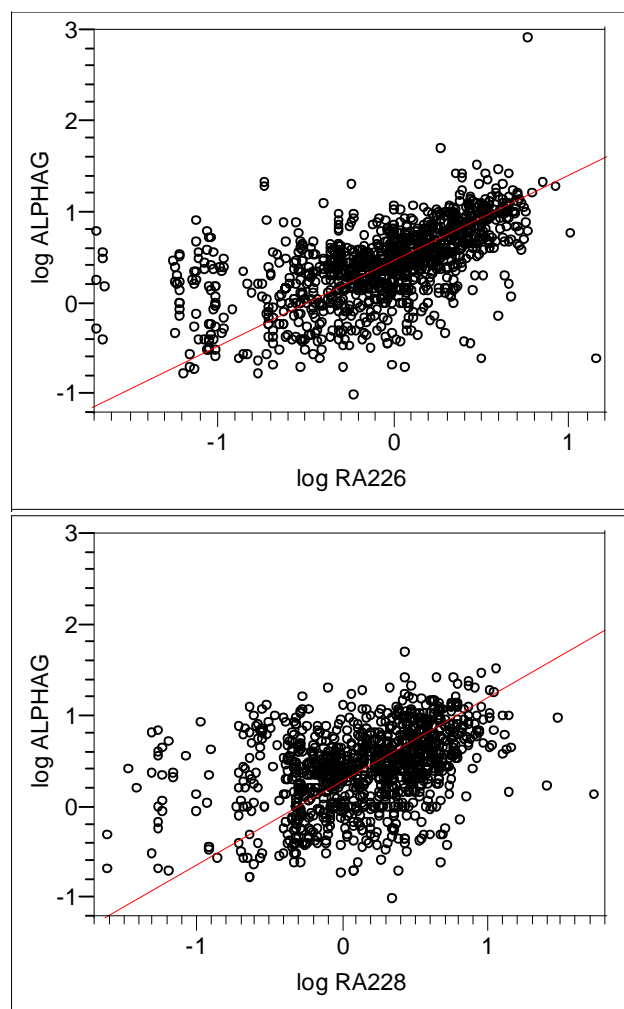


Figure 2. Plots of Ra226 and Ra228 versus Gross Alpha on common log scale showing strong positive ($p < .05$) correlation between both pairs of variables.

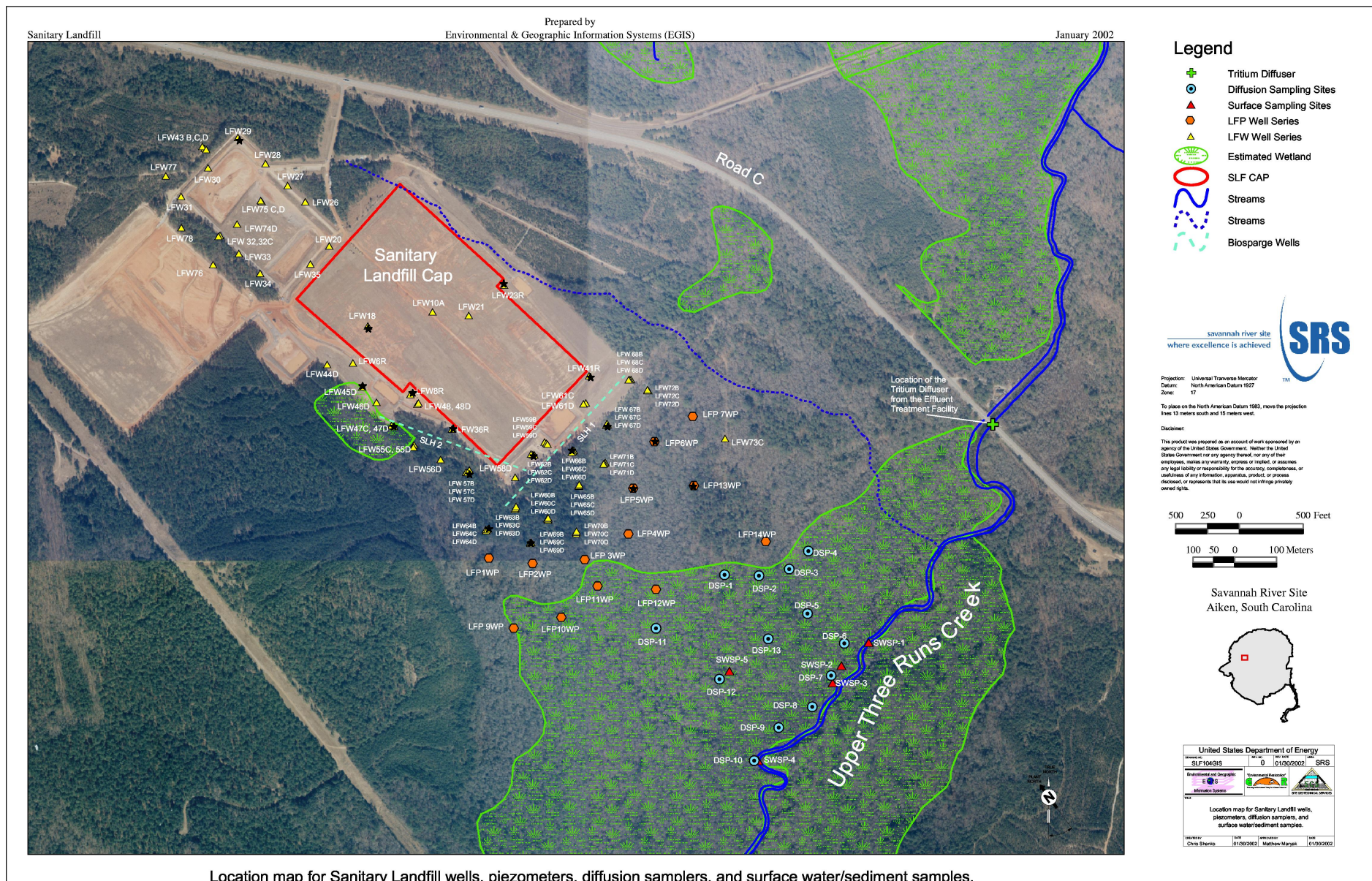


Figure 3: Spatial map of SLF with black stars indicating wells with recently elevated radium.

measurements of Ra isotopes. Note that even though most of the elevated measurements are located downgradient to the SLF facility there are also elevated measurements in at least one upgradient well (LFW 29). In addition, there is no consistent spatial pattern for Ra contamination in the downgradient wells. Ra exceedences in the LFW 62 cluster, for instance, would be expected to appear in other downgradient well clusters in close proximity to LFW 62, but do not.

4.3. Total Ra Analysis

The GWPS for radium does not allow concentrations of Ra226, Ra228, or sum of these two isotopes (Ra2628) to exceed 5 pCi/L.

4.3.1. Wells with Recent Exceedences

The time series plots for the 17 wells with recent (2003) elevated Ra measurements above the GWPS are shown in Appendix B1. For each well, the concentration is depicted through time for Ra226, Ra228, Ra2628, and gross alpha on a single plot. The plot symbol color indicates detection; the plot symbol denotes the lab; and colored lines distinguish analytes. Most of the plots have the same sample date scale (January 1987 – January 2004) and result scale (0-70 pCi/L) for easy comparison of wells. The only exception is the result scale for LFP 5WP, which has a gross alpha result of 839 pCi/L (and an SQL of 633 pCi/L) on 6/28/2002. Since the next highest gross alpha result from all of the SLF wells through all time is 69.1 pCi/L, this is obviously an extreme (and suspect) outlier. The three LFP wells have only recent data, but the plots for most of the LFW wells portray analyte concentrations that are relatively consistent through time.

These results show not only the recently elevated Ra results, but that also Ra has shown historically elevated concentrations. For instance, the Appendix B1 plot for LFW 18 shows that Ra2628 was elevated above the GWPS during the mid 1990's as well as in 2003 primarily due to Ra228.

4.3.2. Historical Ra and Gross Alpha Concentrations

Appendix B2 displays time series plots by analyte for all SLF wells in this study. These plots were constructed for Ra226, Ra228, Ra2628 (sum), and gross alpha and for the ratio of detected Ra228 to Ra226. All of the SLF data were included in the plots for the purpose of examining the radium or gross alpha concentrations through time for all SLF wells. The plot symbol color indicates detection, and the plot symbol distinguishes the lab. Note that some of the gross alpha values, both detects and nondetects, in the 2000 to 2001 time frame seem high relative to the surrounding data, and that only the Mobile Lab performed those analyses. The levels of each analyte appear to be consistent through time, with the exception of the afore-mentioned gross alpha values from the Mobile Lab. Note that of the 111 SLF wells with data in this study, 42 of them showed concentrations of gross alpha elevated above the 15 pCi/L GWPS. This fact has also been presented in a similar data display in the 2004 Noffsinger report ERD-EN-2004-0087.

4.4. Radium Isotopic Ratio Analysis – On-Site versus Off-Site Ra Comparisons

The ratio of the two Ra isotopes (228 to 226) was constructed to assess the weight of evidence for an anthropogenic source of radium in the SLF. Ratios that are relatively constant in the SLF wells by comparison to ratios in the wells of other on- and off-Site facilities would be regarded as evidence in favor of the working hypothesis. Data from the study “Radioactivity in Groundwater near the Savannah River Site” (Price and Michel 1990) were utilized in this analysis.

Sets of boxplots were drawn for detected Ra226, detected Ra228, detected gross alpha, and detected isotopic ratios for the SLF wells, selected DCB series wells, P series wells, study wells from on-Site, and study wells from off-Site. Only detects were involved in this analysis since some of the non-SLF data had overly large detection limits, which made the plots misleading. Common logarithms of the data were used to better show the variability of these ratios among the on- and off-Site groups of wells, because the measurement ratios among wells often crossed one or more orders of magnitude. For each set of plots, reference lines were built from the 25th and 75th quantiles of the SLF boxplot. The overlap of the boxes on every plot indicates that the SLF values are not significantly different from those of the other groups.

Figure 4 is a set of boxplots depicting the results of the comparison for the isotopic ratio. The Figure shows that the SLF radium ratios are not unlike those among wells in other facilities known to have natural radium only. The DCB ratios are slightly higher than others because the radium is a mixture of radium from native soils and radium from coal (ratio of about 1.3). This suggests that the SLF ratios are in line with what would be expected from naturally occurring radium, and, therefore, that natural variations in radium concentration could account for the radium exceedences at the SLF.

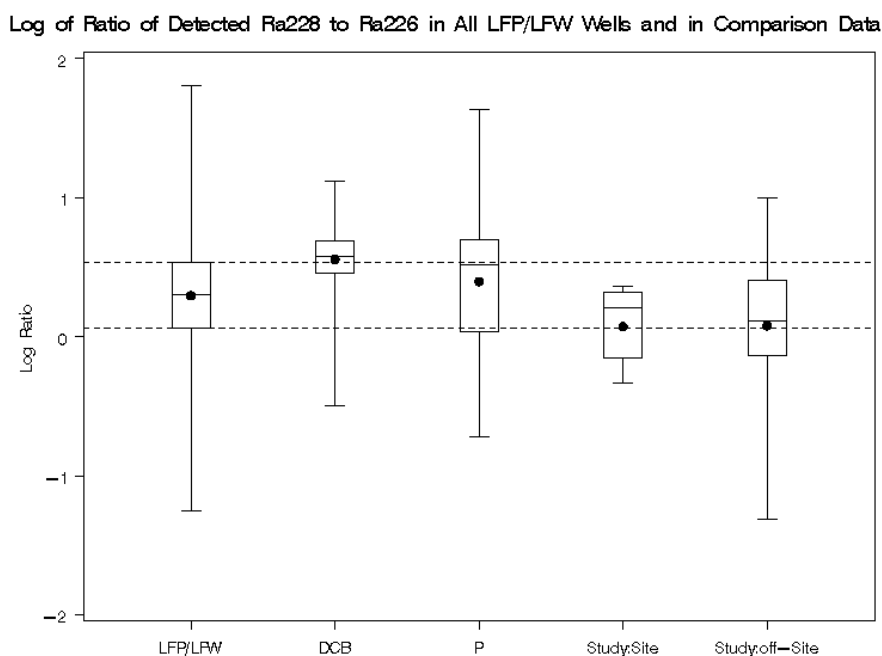


Figure 4: Comparison of the isotopic ratio of Ra228 to Ra226 concentrations in the SLF wells to these same ratios in the monitoring wells of four other facilities: three of which are on-Site (DCB, P, Study:Site) and one off-Site (Study:off-Site).

Appendix B3 contains boxplots showing the relation of Ra226, Ra228, and gross alpha concentrations among the same five on- and off-Site locations. Again, the common logarithms are used, because the data on the original scale cross more than one order of magnitude. These plots also show reasonably equivalent concentrations of both radium isotopes and gross alpha in the SLF monitoring wells compared to well concentrations among the other on- and off-Site facilities known to be uncontaminated.

4.5. Geochemical Analysis

The following results were obtained based on expected geochemical relationships between inorganic constituents and water quality parameters versus isotopic radium and gross alpha.

4.5.1. pH versus Radium and Gross Alpha

Figure 5 shows plots of radium and gross alpha versus the field measurement of pH for quarterly samples among all SLF wells in this study. The plots suggest a quadratic (polynomial) relationship between Ra226 and pH, between gross alpha and pH, and possibly between Ra228 and pH. In each plot, the quadratic relationship is statistically significant ($p < .001$). Note however, that there is no geochemical explanation for the polynomial fit. At the low (<5) pH range, an inverse correlation between radium level and pH, i.e., increased radium mobility, is expected. Natural pH values at the SRS are rarely below 4.5. The low pH values may be from landfill leachate, and such leachate could mobilize natural aquifer radium. However, there is no geochemical explanation for reverse in this relationship at higher pH ranges. Other plots indicated no relationship between pH and a given well or set of well, nor is there any trend in pH with sampling date (quarters) across years. The polynomial could be a statistical artifact of the large number ($> 2,700$) of measurements in 16 years of sampling.

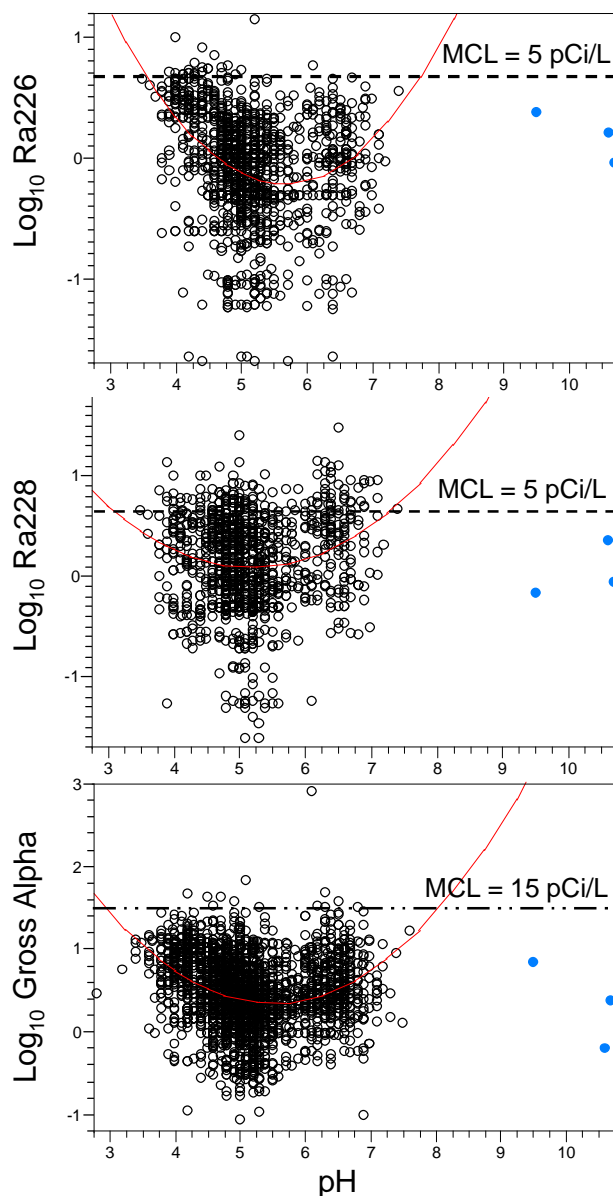


Figure 5. Scatter plots and quadratic linear regression fits of Ra226, Ra228, and gross alpha measurements vs pH from all wells and all years of the study – blue pH outliers excluded from regressions.

The relatively small parameter estimation variances can yield statistically significant relationships with pH, although the practical significance is in question as illustrated by the weak bivariate correlations seen here ($r = .34$, $.20$, and $.31$, respectively).

4.5.2. Total Barium (BATOT) and Total Calcium (CATOT) versus Radium and Gross Alpha

As indicated earlier, both Ca and Ba may behave geochemically as replacements for the Ra isotopes in the subsurface geology. Figures 6A and 6B display plots of the BATOT and CATOT concentrations, respectively, versus Ra226 and ALPHAG for quarterly samples among all SLF wells. The patterns of Ra228 with these same metals are similar.

Note that the simple linear regression (SLR) fits (in red) in Figure 6A have statistically significant ($p < .05$) and positive slope, which means that as the concentration of Ba increases, Ra226 and gross alpha measurements also increase. The Orthogonal regression fits (in green) likewise show a positive relationship between Ba and Ra226 or gross alpha. Although the statistical relation for either radiological measurements is significant, the very low correlation ($.10$ and $.14$, respectively) indicates there is substantial unexplained variability in the SLR relationships.

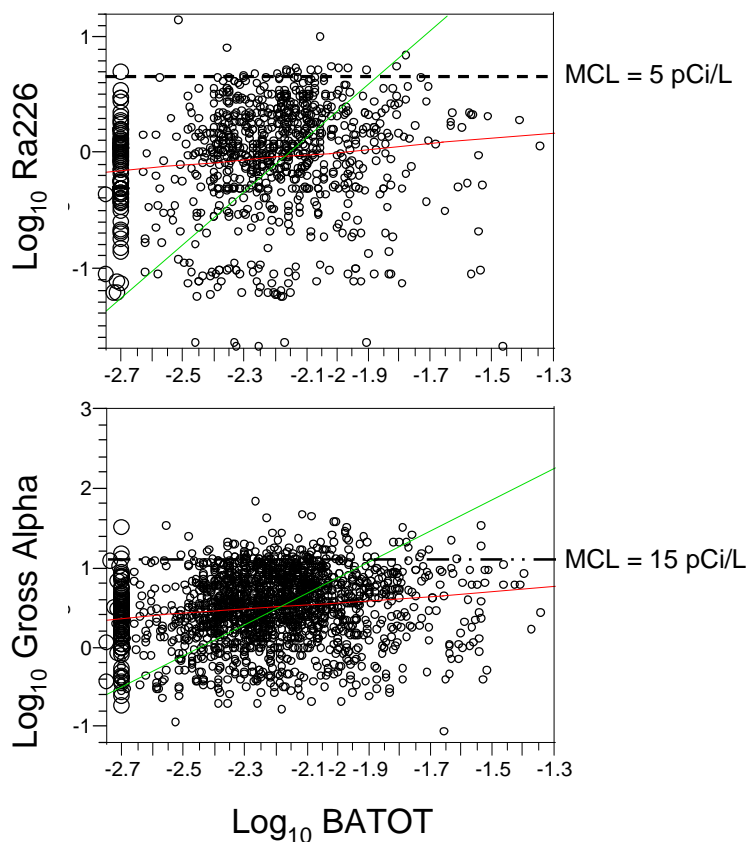


Figure 6A. Scatter plots and regressions fits for Ra226 and gross alpha concentrations (pCi/L) versus total Barium (BATOT) for all wells and all years, on the common log scale. The large symbols indicate BATOT values below detection that were excluded from regression fits.

Again, this is not unexpected geochemically because of the mass balance issues discussed in section 3.3.

Note also that vertical line of sample points at the -2.7 value of log BATOT (Figure 6A). This occurs because these data were recorded as laboratory measurements below detection. So regardless of the measured value for log Ra226, the measured value of log BATOT was the same.

Similarly, Figure 6B shows the SLR and Orthogonal regression fits to the same Ra226 and gross alpha data versus CATOT. The SLR has a statistically significant ($p < .0001$) and positive slope indicating that as total calcium concentration increases, so do Ra226 and gross alpha by a factor of .12 and .19 to 1, respectively. There is a stronger positive correlation between CATOT and Ra226 or gross alpha (.19 and .26, respectively) than exists for BATOT. As noted before, the large amount of scatter or variability in these data is not unexpected because of accuracy and precision issues in mass balance calculations.

4.5.3. Turbidity, Water Elevation, and Hold Times

The scatter plots and linear regression analyses results for Ra226 or Ra228 versus turbidity and water elevation showed no statistically significant patterns or relationships. In addition, a review of the hold times for all samples prior to analysis indicated no excessive values. This suggests that elevated radium concentrations were not the result of variation in field sampling conditions or violations of laboratory analysis protocol.

5. CONCLUSIONS

The conclusion from this study is that there is insufficient evidence to reject the working hypothesis, viz., that the recent elevated measurements of Ra226, Ra228, or gross alpha are not from anthropogenic sources. In fact the evidence presented here suggests that radium in the SLF groundwater is not substantially different from the natural radium in the surrounding area. This does not eliminate the possibility that some natural radium was mobilized by the interaction of native soils with landfill leachate.

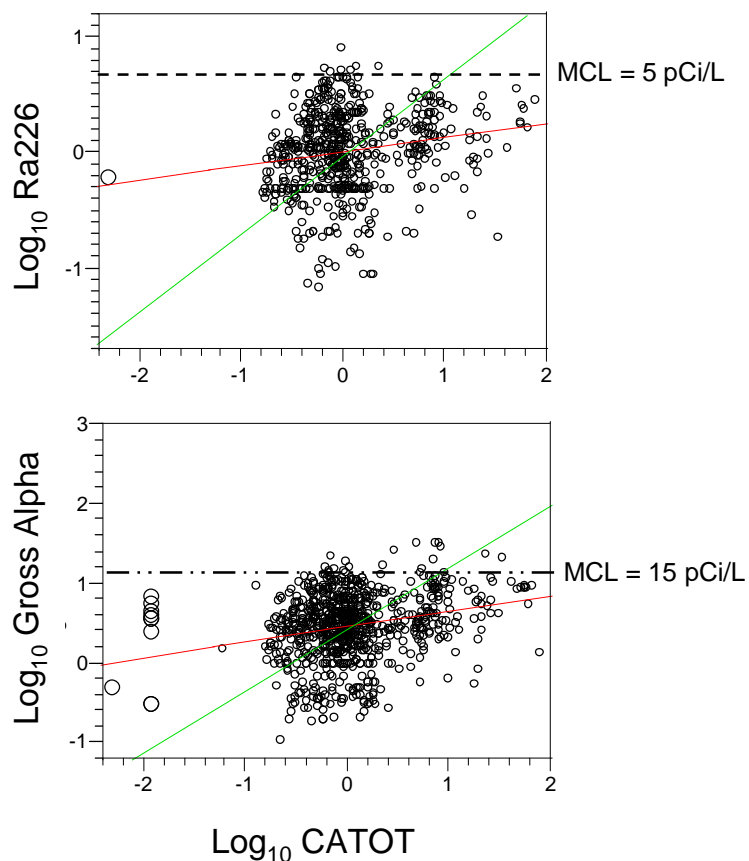


Figure 6B. Scatter plots and regressions fits for Ra226 and gross alpha concentrations (pCi/L) versus total Calcium (CATOT) for all wells and all years, on the common log scale. The large symbols indicate CATOT values below detection that were excluded from regression fits.

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APPENDIX A

Appendix A contains summary information for the SLF wells used in this study and for the analyses performed for radioactive analytes in these wells.

Appendix A1- Summary data for LFP/LFW wells used in this study of the SLF groundwater monitoring network

Sanitary Landfill Wells: LFP and LFW Well Series

(Note: Ra2628 represents the sum of Ra226 and Ra228, and the level of concern is 5 pCi/L for radium and 15 pCi/L for gross alpha)

Well	Well Type	Install Date	First Sampled	Last Sampled	Abandoned	Elevation Level	maxRA2628 (pCi/L)	maxRA226 (pCi/L)	maxRA228 (pCi/L)	maxALPHAG (pCi/L)
LFP 1WP	MONITORING WELL	04/05/1994	06/27/2001	07/01/2002		no elevation	2.35	2.35		1.55
LFP 2WP	MONITORING WELL	04/06/1994	06/28/2001	07/01/2002		no elevation	0.2335	0.2335		1.27
LFP 3WP	MONITORING WELL	04/06/1994	05/01/2002	05/01/2002		no data				
LFP 4WP	MONITORING WELL	04/06/1994	06/28/2001	11/21/2003		no elevation	2.555	1.52	1.035	2.865
LFP 5WP	MONITORING WELL	04/08/1994	06/25/2001	12/02/2003		RA recent	9.19	5.92	5.82	839
LFP 6WP	MONITORING WELL	04/08/1994	06/26/2001	11/21/2003		RA recent	14.352	2.31	14	26.3
LFP 7WP	MONITORING WELL	04/08/1994	05/02/2002	05/02/2002		no data				
LFP 9WP	MONITORING WELL	04/05/1994	05/14/2002	05/14/2002		no data				
LFP 10WP	MONITORING WELL	04/04/1994	06/21/2001	06/28/2002		no elevation	0.851	0.851		1.75
LFP 11WP	MONITORING WELL	04/06/1994	06/20/2001	11/21/2003		no elevation	1.71	0.78	0.966	2.38
LFP 12WP	MONITORING WELL	04/07/1994	06/12/2001	11/21/2003		no elevation	2.6	2.01	0.936	3.33
LFP 13WP	MONITORING WELL	04/07/1994	06/28/2001	11/21/2003		RA recent	55.56	1.67	54.7	2.44
LFP 14WP	MONITORING WELL	04/07/1994	06/26/2001	12/01/2003		no elevation	1.665	0.585	1.08	3.445
LFW 1					11/24/1986	no data				
LFW 2					12/05/1986	no data				
LFW 3					12/04/1986	no data				
LFW 4					12/02/1986	no data				
LFW 5					12/01/1986	no data				
LFW 6	MONITORING WELL	02/02/1981	07/17/1984	02/07/1996	02/26/1996	RA old	12.6	4.26	10.9	18
LFW 6R	MONITORING WELL	03/12/1996	06/25/1996	10/27/2003		ALPHAG only	4.21	1.62	3.17	34.5
LFW 7	MONITORING WELL	01/30/1981	07/17/1984	09/06/1995	02/26/1996	RA old	33	2.1	30.9	28.6
LFW 8	MONITORING WELL	01/29/1981	07/17/1984	02/08/1996	02/26/1996	RA old	15.74	4	11.9	34.6
LFW 8R	MONITORING WELL	03/12/1996	06/25/1996	10/30/2003		RA recent	15.62	4.52	11.1	27.3
LFW 9	MONITORING WELL	01/28/1981	07/18/1984	10/02/1986	11/26/1986	no data				
LFW 10					01/01/1984	no data				
LFW 10A	MONITORING WELL	01/13/1984	07/19/1984	10/27/2003		RA old	8.3	4.7	6.6	25

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LFW 16	MONITORING WELL	08/26/1981	07/18/1984	03/01/1995	02/28/1996	RA old	6.3	1.5	5	12
LFW 17	MONITORING WELL	08/28/1981	07/18/1984	12/15/1995	02/28/1996	RA old	13.23	2.94	12.6	4.49
LFW 18	MONITORING WELL	08/27/1981	07/18/1984	10/27/2003		RA recent	17.62	4.34	14.6	39.5
LFW 19	MONITORING WELL	08/31/1981	07/18/1984	12/16/1995	02/26/1996	RA old	9.3	2.7	8	10
LFW 20	MONITORING WELL	08/25/1981	07/19/1984	12/08/1995		RA old	7.6	2.71	6	8.8
LFW 21	MONITORING WELL	01/11/1984	07/19/1984	10/27/2003		RA old	10.7	4.2	6.5	35
LFW 22	MONITORING WELL	01/12/1984	07/19/1984	12/18/1995	02/27/1996	RA old	5.4	2.5	3	22.2
LFW 23	MONITORING WELL	01/12/1984	07/19/1984	02/06/1996	02/28/1996	RA old	5.15	1.8	3.48	7.5
LFW 23R	MONITORING WELL	03/06/1996	06/17/1996	10/22/2003		RA recent	15.03	7.4	7.96	47.8
LFW 24	MONITORING WELL	01/13/1984	07/23/1984	12/13/1995	02/28/1996	RA old	5.7	1.84	4.4	7.6
LFW 25	MONITORING WELL	01/13/1984	07/23/1984	12/13/1995	02/28/1996	RA old	11.2	2.2	10	14
LFW 26	MONITORING WELL	10/29/1986	02/05/1987	12/08/1995		RA old	5.62	1.56	5	9.58
LFW 27	MONITORING WELL	10/30/1986	02/05/1987	12/08/1995		no elevation	4.29	1.19	3.7	4
LFW 28	MONITORING WELL	10/28/1986	02/05/1987	11/18/2003		RA old	6.9	2.2	5.7	6.68
LFW 29	MONITORING WELL	10/28/1986	02/05/1987	10/23/2003		RA recent	7.67	3.27	5.3	9.5
LFW 30	MONITORING WELL	12/31/1986	02/06/1987	11/18/2003		RA old	6.43	0.98	5.5	7.5
LFW 31	MONITORING WELL	10/27/1986	02/05/1987	11/18/2003		RA old	5.83	3.75	4.8	16.3
LFW 32	MONITORING WELL	10/24/1986	02/05/1987	12/02/2003		RA old	11.5	3.3	8.2	14.3
LFW 32C	MONITORING WELL	12/05/1997	02/06/1998	11/18/2003		no elevation				3.015
LFW 33	MONITORING WELL	10/23/1986	02/05/1987	04/22/1996		RA old	27	3.27	26	16.2
LFW 34	MONITORING WELL	10/22/1986	02/05/1987	11/18/2003		RA old	6.39	1.8	4.99	18.3
LFW 35	MONITORING WELL	10/21/1986	02/05/1987	12/08/1995		RA old	6.1	1.8	4.4	7.3
LFW 36	MONITORING WELL	11/05/1986	02/05/1987	12/16/1995	02/26/1996	RA old	14.49	2.99	11.5	32.9
LFW 36R	MONITORING WELL	03/08/1996	06/25/1996	10/24/2003		RA recent	7.8	2.7	6.71	35.1
LFW 37	MONITORING WELL	11/04/1986	02/06/1987	12/16/1995	02/26/1996	RA old	10.5	4.22	8.5	24.2
LFW 38	MONITORING WELL	11/03/1986	02/06/1987	03/03/1995	02/26/1996	ALPHAG only	4.8	1.5	3.7	7
LFW 39	MONITORING WELL	11/03/1986	02/06/1987	03/03/1995	02/27/1996	RA old	8.1	3.4	4.7	32.8
LFW 40	MONITORING WELL	10/31/1986	02/06/1987	03/03/1995	02/27/1996	RA old	7.85	3.2	6.9	21.7
LFW 41	MONITORING WELL	10/31/1986	02/06/1987	02/06/1996	02/27/1996	RA old	8.16	1.58	7.2	4
LFW 41R	MONITORING WELL	03/06/1996	06/17/1996	10/22/2003		RA recent	6.52	4.84	4.82	19.6
LFW 42	MONITORING WELL	10/30/1986	02/06/1987	03/03/1995	02/27/1996	ALPHAG only	3.9	1.2	2.8	8.7
LFW 43B	MONITORING WELL	06/21/1991	09/02/1991	11/18/2003		RA old	6.13	3.01	5.18	21.6
LFW 43C	MONITORING WELL	06/25/1991	09/02/1991	11/18/2003		RA old	8.09	2.23	7.26	13.13

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LFW 43D	MONITORING WELL	06/24/1991	09/02/1991	11/18/2003		RA old	6.94	1.23	6.2	12.93
LFW 44D	MONITORING WELL	05/29/1991	09/02/1991	11/18/2003		RA old	6.83	3.03	5.65	9.2
LFW 45D	MONITORING WELL	05/16/1991	09/02/1991	10/30/2003		RA recent	8.65	3.35	5.3	32.2
LFW 46D	MONITORING WELL	05/29/1991	09/02/1991	12/04/1995		RA old	6.5	2.7	5	11.6
LFW 47C	MONITORING WELL	05/29/1991	09/02/1991	10/30/2003		RA recent	12.44	6.39	6.05	28.5
LFW 47D	MONITORING WELL	05/23/1991	09/02/1991	10/30/2003		ALPHAG only	4.935	1.73	4.8	22.5
LFW 48C	MONITORING WELL	05/28/1991	09/02/1991	10/23/2003		no elevation	3.2315	2.16	2.7	4.035
LFW 48D	MONITORING WELL	05/23/1991	09/02/1991	10/23/2003		RA old	14.14	3.54	10.6	13.2
LFW 55C	MONITORING WELL	05/14/1991	09/03/1991	12/05/1995		RA old	6	2.05	5	5.45
LFW 55D	MONITORING WELL	05/15/1991	09/03/1991	12/05/1995		RA old	6.24	2.68	3.6	4.48
LFW 56D	MONITORING WELL	05/22/1991	09/03/1991	10/29/2003		ALPHAG only	4.814	4.36	2.8	44.7
LFW 57B	MONITORING WELL	06/11/1991	09/03/1991	01/23/2004		RA recent	18.7	8.4	10.3	27.1
LFW 57C	MONITORING WELL	05/21/1991	09/03/1991	10/24/2003		RA old	6.05	4.77	4.3	7.1
LFW 57D	MONITORING WELL	05/21/1991	09/03/1991	10/24/2003		ALPHAG only	4.58	1.05	3.89	5.3
LFW 58D	MONITORING WELL	05/21/1991	09/03/1991	10/29/2003		RA old	8.41	2.39	7.7	11.41
LFW 59B	MONITORING WELL	06/13/1991	09/04/1991	07/17/2001		RA old	10.6	4.2	6.4	13.4
LFW 59C	MONITORING WELL	06/18/1991	09/04/1991	10/24/2003		RA old	5.2	2.5	3.9	7
LFW 59D	MONITORING WELL	06/18/1991	09/04/1991	10/27/2003		ALPHAG only	4.29	2.03	3.5	22.9
LFW 60B	MONITORING WELL	12/10/1993	03/27/1994	12/08/1995		RA old	6.92	4.54	2.5	8.68
LFW 60C	MONITORING WELL	12/14/1993	03/21/1994	10/24/2003		ALPHAG only	4.4135	3.96	1.6	25.4
LFW 60D	MONITORING WELL	06/18/1991	09/05/1991	10/24/2003		ALPHAG only	2.35	0.95	2.1	24.8
LFW 61C	MONITORING WELL	05/20/1991	09/04/1991	10/27/2003		no elevation	4.19	1.4	3.6	4.49
LFW 61D	MONITORING WELL	05/20/1991	09/04/1991	10/29/2003		RA old	7.2	2.5	5.1	31
LFW 62B	MONITORING WELL	06/11/1991	09/04/1991	01/23/2004		RA recent	19.18	5.28	13.9	19.5
LFW 62C	MONITORING WELL	06/11/1991	09/04/1991	01/28/2004		RA old	5.8	2.27	4.3	6.18
LFW 62D	MONITORING WELL	06/05/1991	09/05/1991	01/23/2004		ALPHAG only	3.777	1.22	2.94	13.4
LFW 63B	MONITORING WELL	01/11/1994	03/29/1994	10/29/2002		RA old	8.04	5.39	4.29	20
LFW 63C	MONITORING WELL	01/11/1994	03/29/1994	07/17/2003		RA old	8.01	2.24	7.5	12.1
LFW 63D	MONITORING WELL	01/11/1994	03/29/1994	10/24/2003		no elevation	3.701	2.79	3.6	13.11
LFW 64B	MONITORING WELL	02/01/1994	04/05/1994	11/02/2002		RA old	8.8	3.8	6.4	13.3
LFW 64C	MONITORING WELL	02/02/1994	03/27/1994	10/30/2003		RA recent	13.7	10.6	5.5	47.1
LFW 64D	MONITORING WELL	02/02/1994	03/27/1994	10/29/2003		no elevation	3.84	1.31	2.54	11.61
LFW 65B	MONITORING WELL	12/23/1993	03/25/1994	10/29/2002		RA old	6.63	5.6	3.31	14.13

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LFW 65C	MONITORING WELL	01/03/1994	03/27/1994	10/30/2003		ALPHAG only	4.16	2.19	2.7	12.48
LFW 65D	MONITORING WELL	01/04/1994	04/05/1994	10/30/2003		ALPHAG only	4.495	0.92	3.93	17.1
LFW 66B	MONITORING WELL	12/21/1993	03/26/1994	04/08/2003	11/20/2003	RA recent	5.1	2.73	2.73	50.4
LFW 66C	MONITORING WELL	12/21/1993	03/26/1994	07/22/2003	08/20/2003	ALPHAG only	3.99	2.45	2.37	9.1
LFW 66CR	MONITORING WELL	08/19/2003	11/14/2003	11/14/2003		no elevation	2.095	1.3	0.795	2.575
LFW 66D	MONITORING WELL	12/21/1993	03/26/1994	11/01/2003		no elevation	3.22	0.914	2.5	3.905
LFW 67B	MONITORING WELL	01/24/1994	03/28/1994	10/30/2003		RA recent	7.78	4.11	4.68	44.1
LFW 67C	MONITORING WELL	01/26/1994	03/28/1994	10/30/2003		RA old	6.12	4.77	2.71	13.56
LFW 67D	MONITORING WELL	01/26/1994	03/28/1994	10/30/2003		RA old	7.97	5.52	3.41	69.1
LFW 68B	MONITORING WELL	01/18/1994	03/28/1994	12/08/1995		RA old	5.9	4.61	3.1	16.9
LFW 68C	MONITORING WELL	01/14/1994	03/28/1994	11/01/2003		RA old	8.73	5.01	3.72	13.43
LFW 68D	MONITORING WELL	01/18/1994	03/28/1994	11/01/2003		RA old	28.7835	28.3	6.51	12.81
LFW 69B	MONITORING WELL	02/07/1994	03/26/1994	12/07/1995		RA old	15.2	3.8	11.7	11.9
LFW 69C	MONITORING WELL	02/08/1994	03/26/1994	10/30/2003		RA recent	8.53	5.89	4.52	16
LFW 69D	MONITORING WELL	02/07/1994	03/26/1994	10/30/2003		no elevation	3.94	3.21	3.6	12.39
LFW 70B	MONITORING WELL	02/17/1994	03/20/1994	12/07/1995		RA old	6.06	5.8	2.8	10.2
LFW 70C	MONITORING WELL	02/17/1994	03/20/1994	10/31/2002		RA old	5.8	3.6	2.2	6.83
LFW 70D	MONITORING WELL	02/16/1994	03/20/1994	10/31/2002		no elevation	1.38	0.38	1	1.17
LFW 71B	MONITORING WELL	02/11/1994	03/29/1994	10/31/2002		RA old	7.6	3.6	4	20.5
LFW 71C	MONITORING WELL	02/11/1994	04/05/1994	10/30/2003		RA old	6	3.33	3.1	13.11
LFW 71D	MONITORING WELL	02/11/1994	03/29/1994	10/30/2003		ALPHAG only	2.621	0.952	2.05	11.68
LFW 72B	MONITORING WELL	02/22/1994	03/29/1994	12/06/1995		RA old	8.46	5.17	3.54	18.5
LFW 72C	MONITORING WELL	02/22/1994	03/29/1994	12/06/1995		RA old	7.84	2.54	5.3	2.9
LFW 72D	MONITORING WELL	02/21/1994	03/29/1994	12/06/1995		no elevation	3.77	0.87	2.9	3.6
LFW 73C	MONITORING WELL	04/29/1994	12/12/2001	11/01/2002		no data				
LFW 74C	MONITORING WELL	11/18/1994	03/17/1995	12/02/2003		no elevation				2.59
LFW 74D	MONITORING WELL	11/18/1994	03/17/1995	11/18/2003		no elevation				2.465
LFW 75C	MONITORING WELL	11/28/1994	03/17/1995	11/18/2003		no elevation				3.46
LFW 75D	MONITORING WELL	11/21/1994	03/17/1995	12/02/2003		no elevation				4.4
LFW 76	MONITORING WELL	12/04/1997	02/17/1998	04/14/2003		no elevation				2.43
LFW 77	MONITORING WELL	12/07/1997	02/06/1998	08/10/1998		no data				
LFW 78	MONITORING WELL	12/03/1997	02/17/1998	04/14/2003		no data				

	recent (2003 on) elevated Ra2628
	older (prior to 2003) elevated Ra2628
	elevated gross alpha only

Appendix A2- Analyses performed for radioactive analytes in the SLF wells

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analyte	# analyses	# detects	% detect	max detect	min detect	units
ALPHAG	3555	2431	68%	839	0.1	pCi/L
RA226	1581	1255	79%	28.3	0.09	pCi/L
RA228	1462	999	68%	54.7	0.1	pCi/L
BETAG	2302	1727	75%	140.6	0.33	pCi/L
NP237	10	1	10%	0.317	0.317	pCi/L
TH228	10	2	20%	1.33	0.493	pCi/L
TH232	10	4	40%	0.328	0.116	pCi/L
TOTACT	135	94	70%	66.6	1.03	pCi/L
TOTRAD	840	658	78%	63.7	0.2	pCi/L
TRITIUM	3799	3288	87%	1120.83	0.00084	pCi/mL
U238	11	2	18%	0.455	0.128	pCi/L
U3334	11	1	9%	0.369	0.369	pCi/L
AC228	1	0				
AM241	10	0				
BA133	1	0				
CE144	1	0				
CM242	10	0				
CM4344	10	0				
CM4546	6	0				
CO57	1	0				
CO58	1	0				
CO60	1	0				
CS134	1	0				
CS137	1	0				
EU152	1	0				
EU154	1	0				
EU155	1	0				
K40	1	0				
MN54	1	0				
NA22	1	0				
NP239	1	0				
PB212	1	0				
PM144	1	0				
PM146	1	0				
PU238	10	0				
PU3940	10	0				
RU106	1	0				
SB124	1	0				
SB125	1	0				
SN113	1	0				
TH230	2	0				
TH234	1	0				
U235	11	0				
Y88	1	0				
ZN65	1	0				
ZR95	1	0				

APPENDIX B

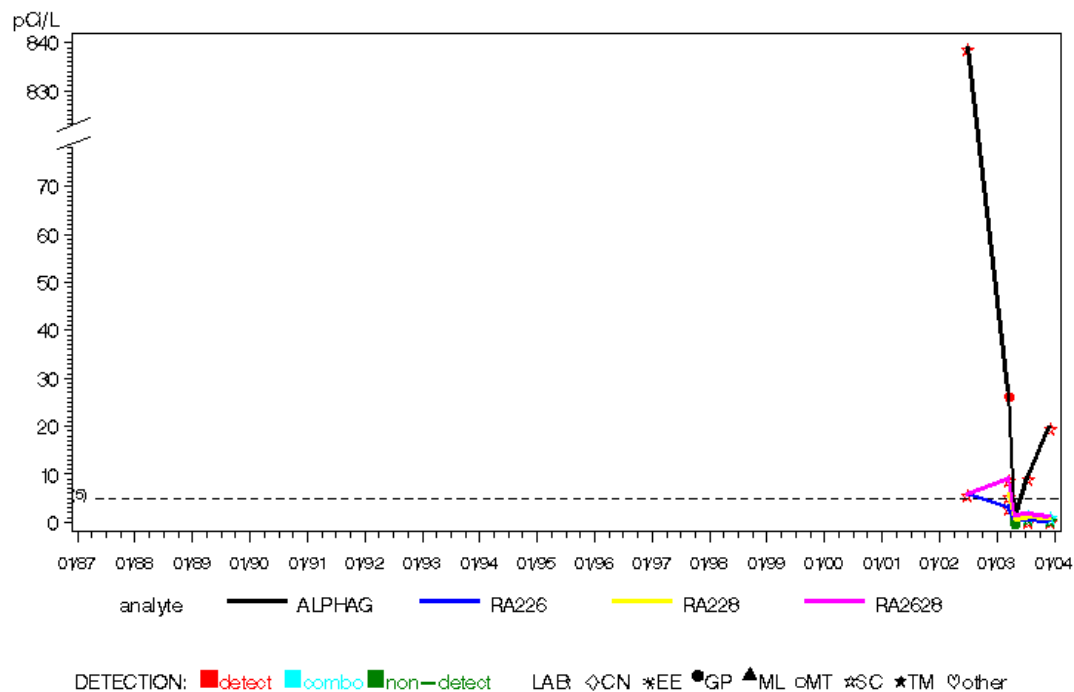
Appendix B contains three sets of plots: time plots by well, time plots by analyte, and comparative boxplots.

Appendix B1 – Time series plots by well

SLF Wells (LFW, LFP Series): Recent Elevated Radium

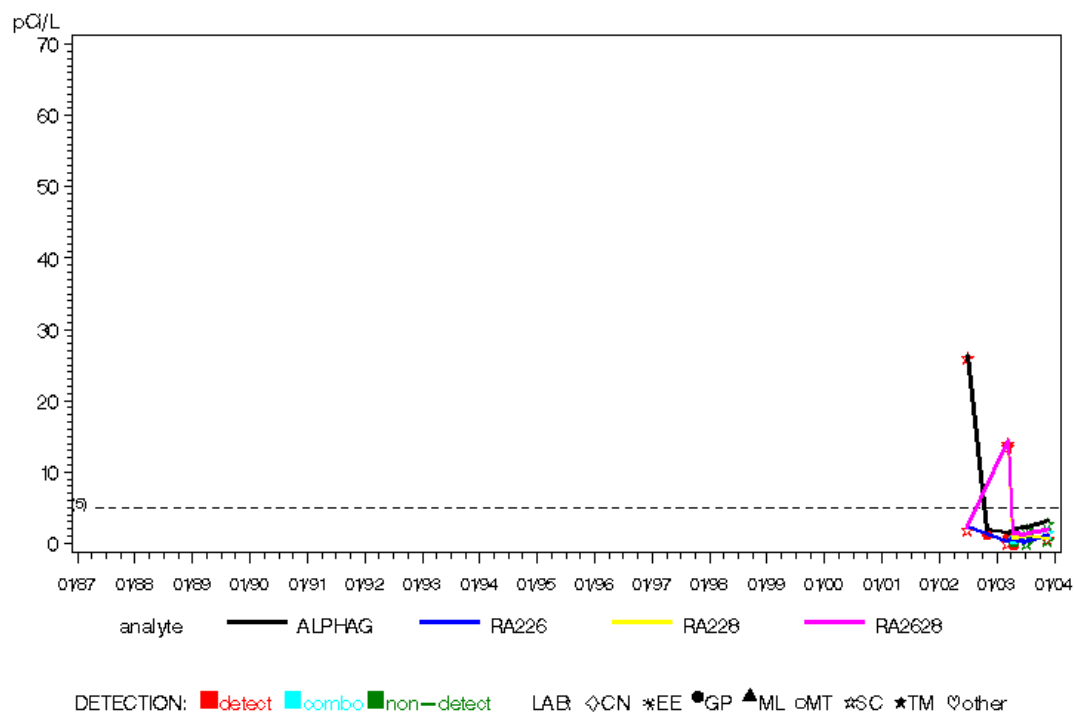
(Note: Gross Alpha Outlier Triggers Different Scale)

WELL= LFP 5WP



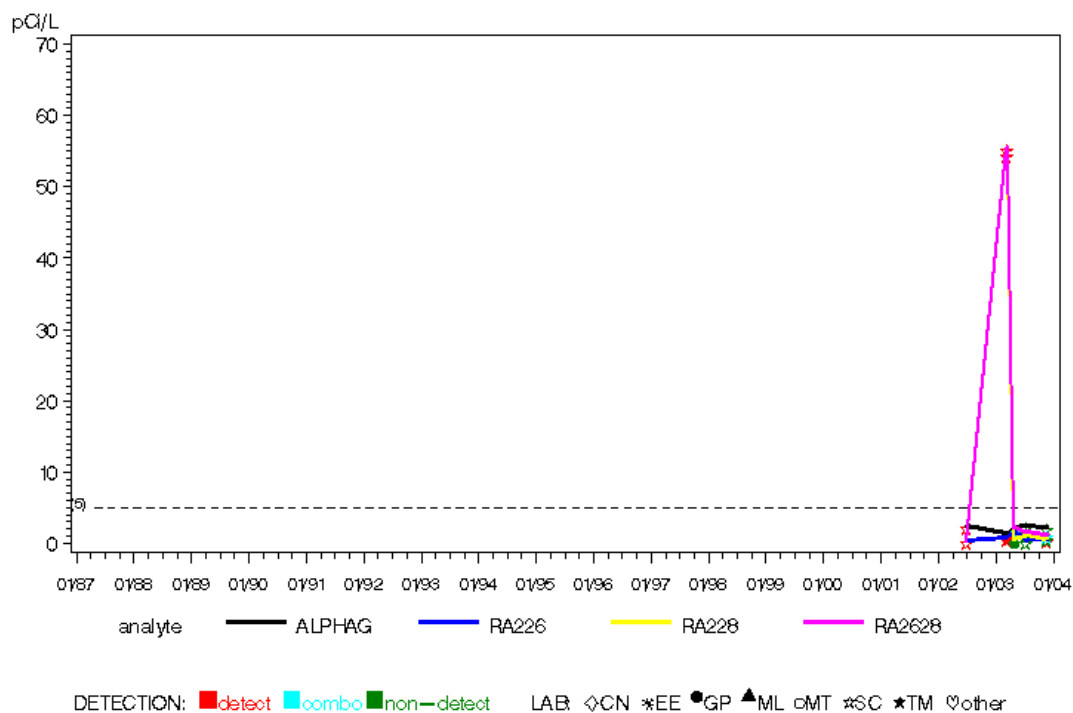
SLF Wells (LFW, LFP Series): Recent Elevated Radium

WELL= LFP 6WP



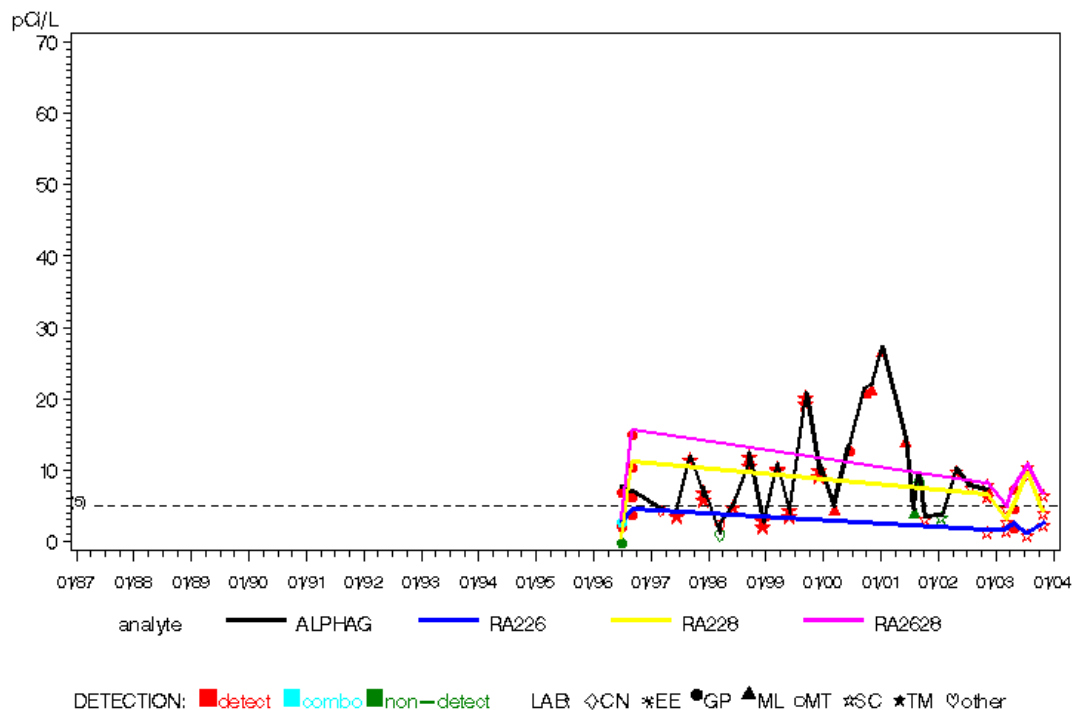
SLF Wells (LFW, LFP Series): Recent Elevated Radium

WELL= LFP 13WP



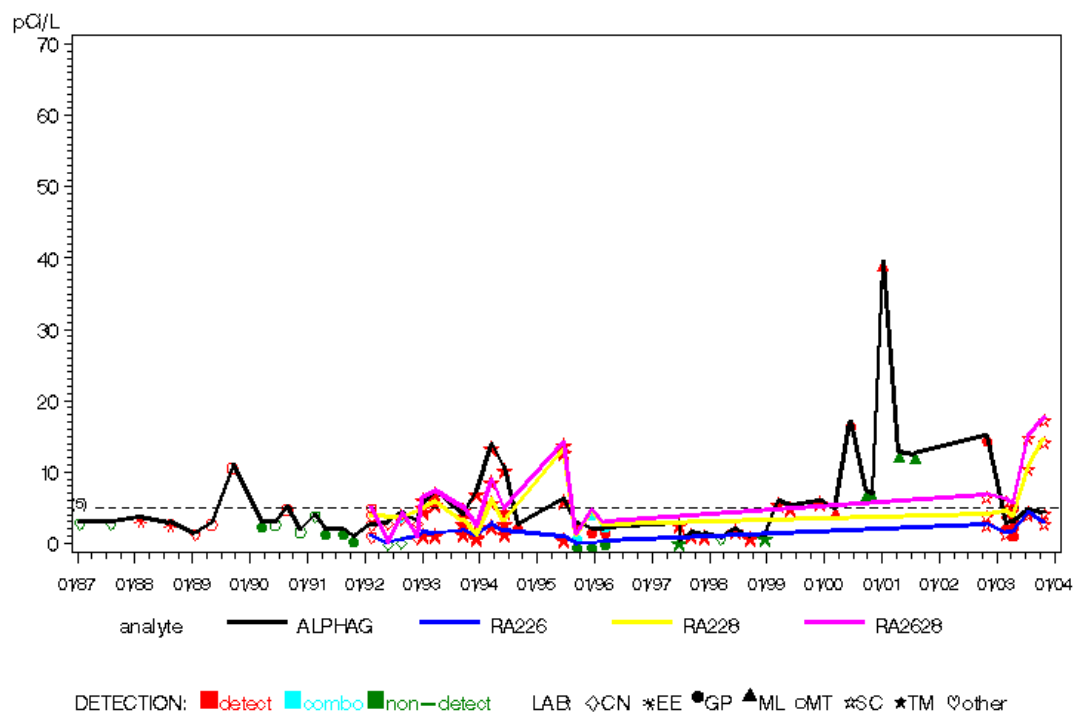
SLF Wells (LFW, LFP Series): Recent Elevated Radium

WELL= LFW 8R



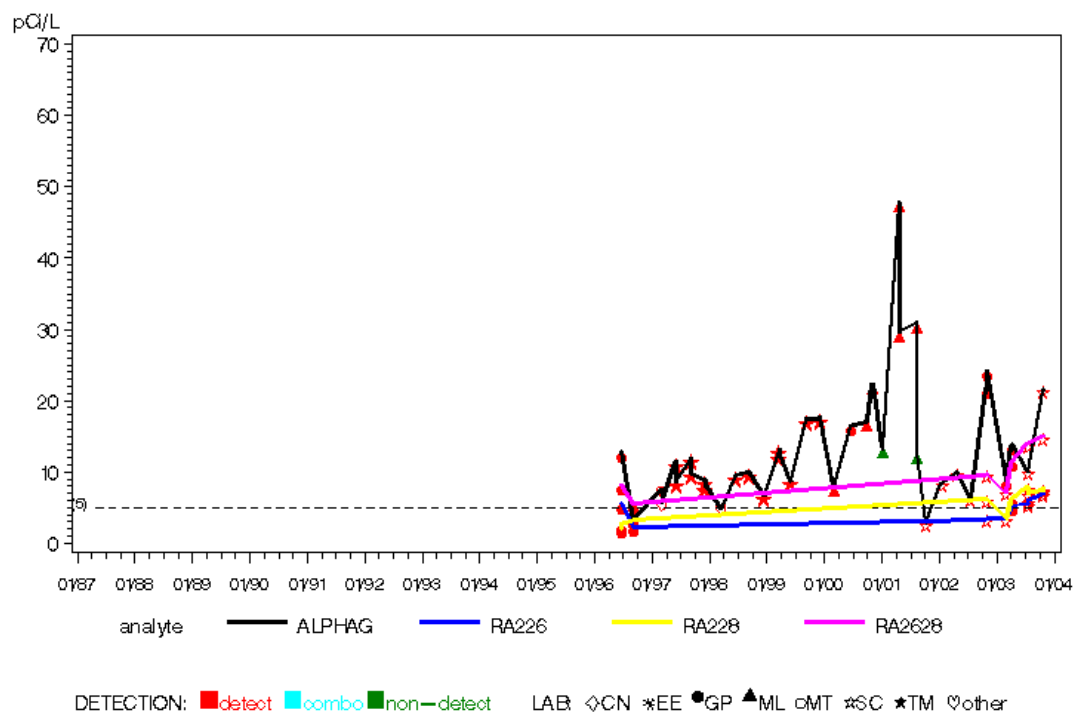
SLF Wells (LFW, LFP Series): Recent Elevated Radium

WELL= LFW 18



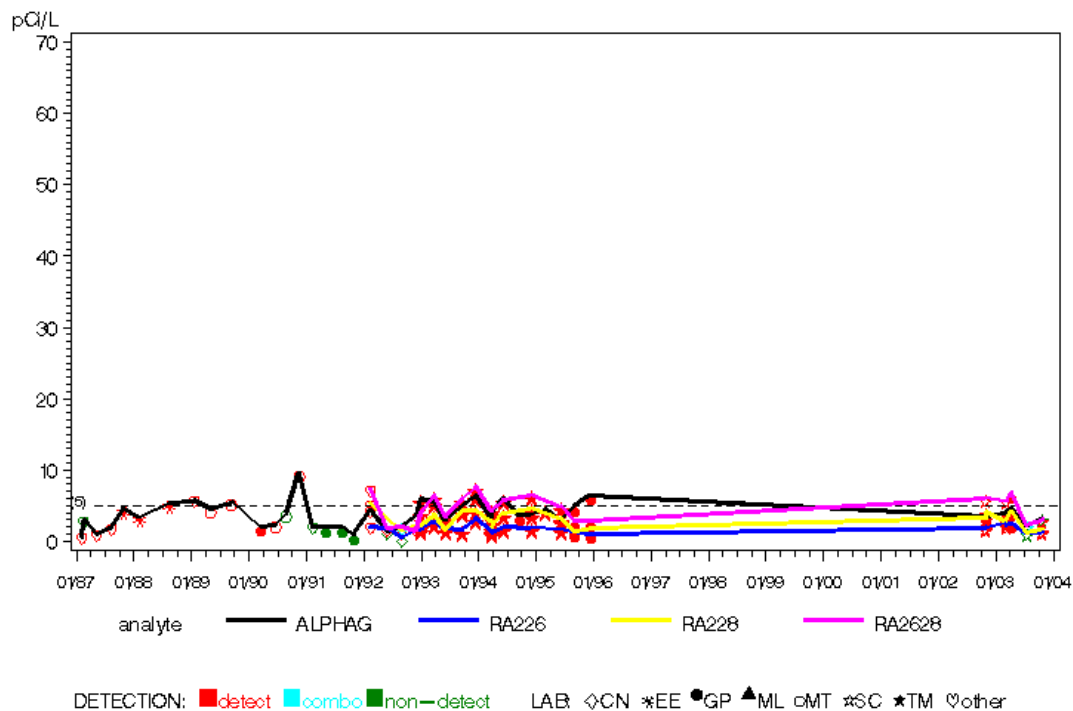
SLF Wells (LFW, LFP Series): Recent Elevated Radium

WELL= LFW 23R



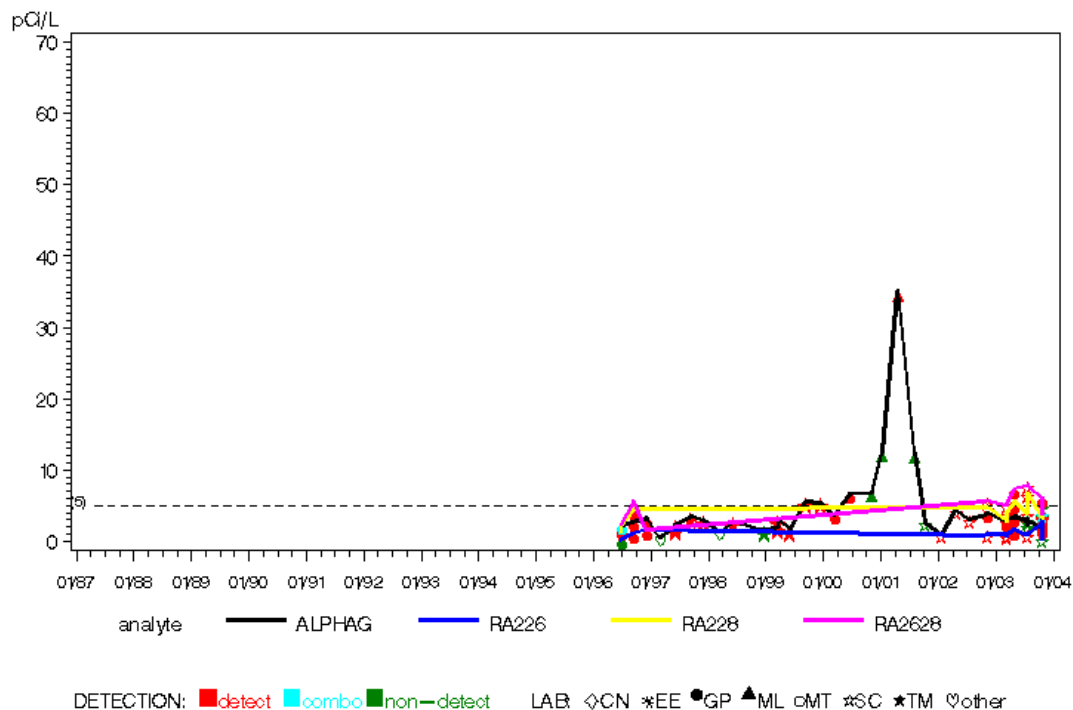
SLF Wells (LFW, LFP Series): Recent Elevated Radium

WELL= LFW 29

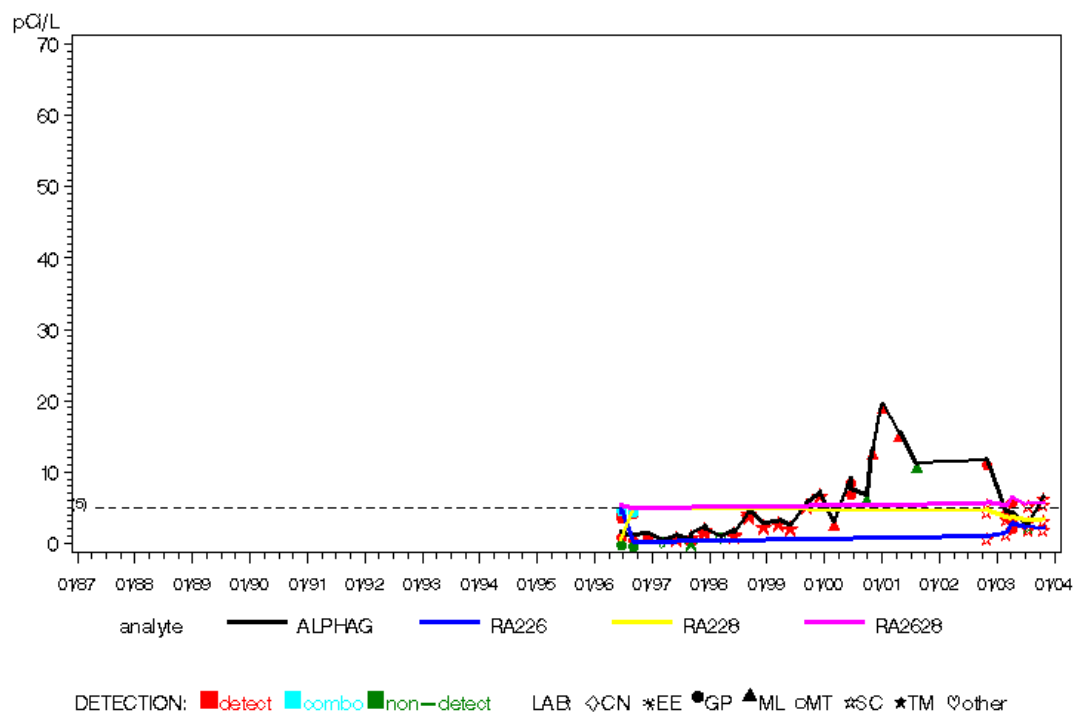


SLF Wells (LFW, LFP Series): Recent Elevated Radium

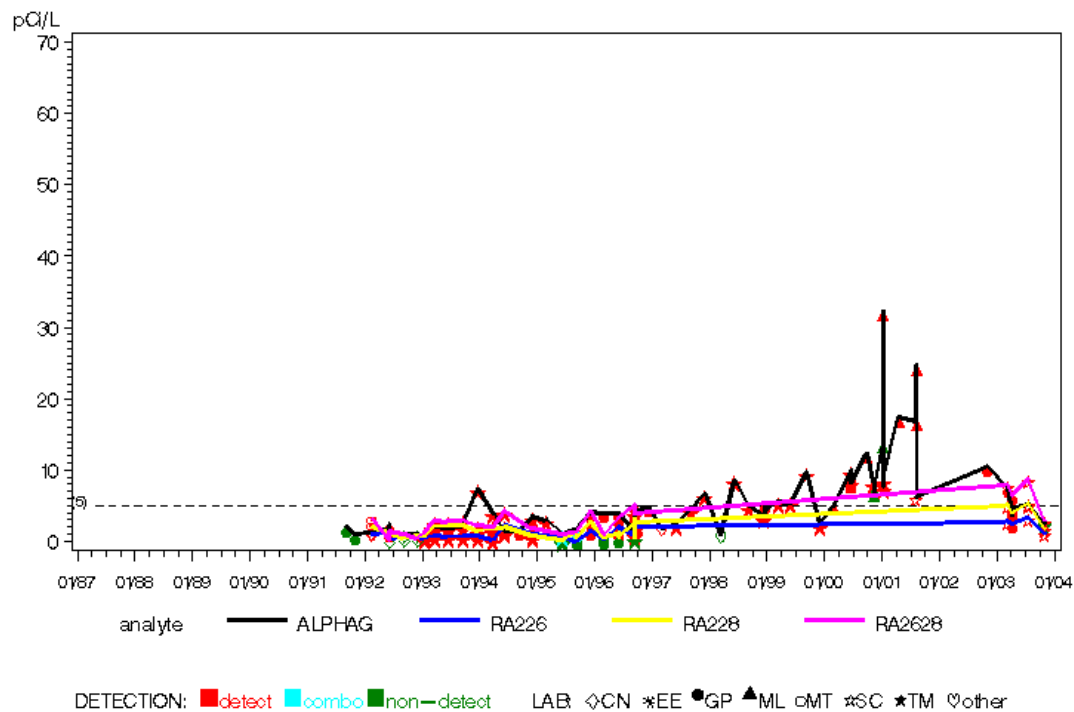
WELL= LFW 36R



SLF Wells (LFW, LFP Series): Recent Elevated Radium
WELL= LFW 41R

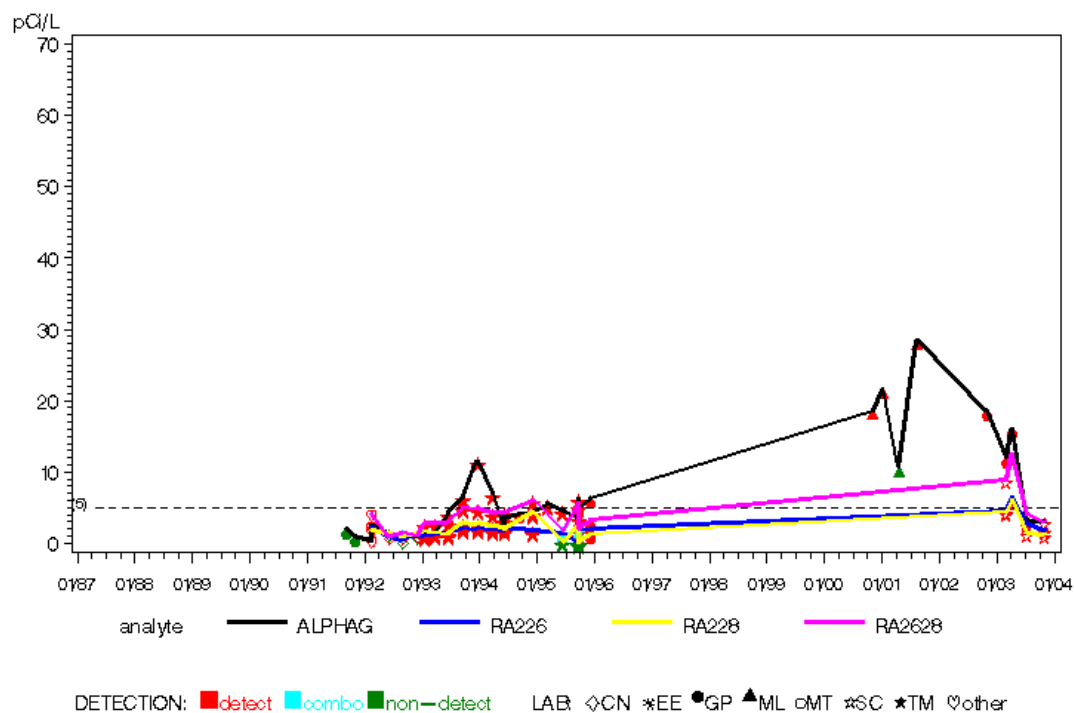


SLF Wells (LFW, LFP Series): Recent Elevated Radium
WELL= LFW 45D



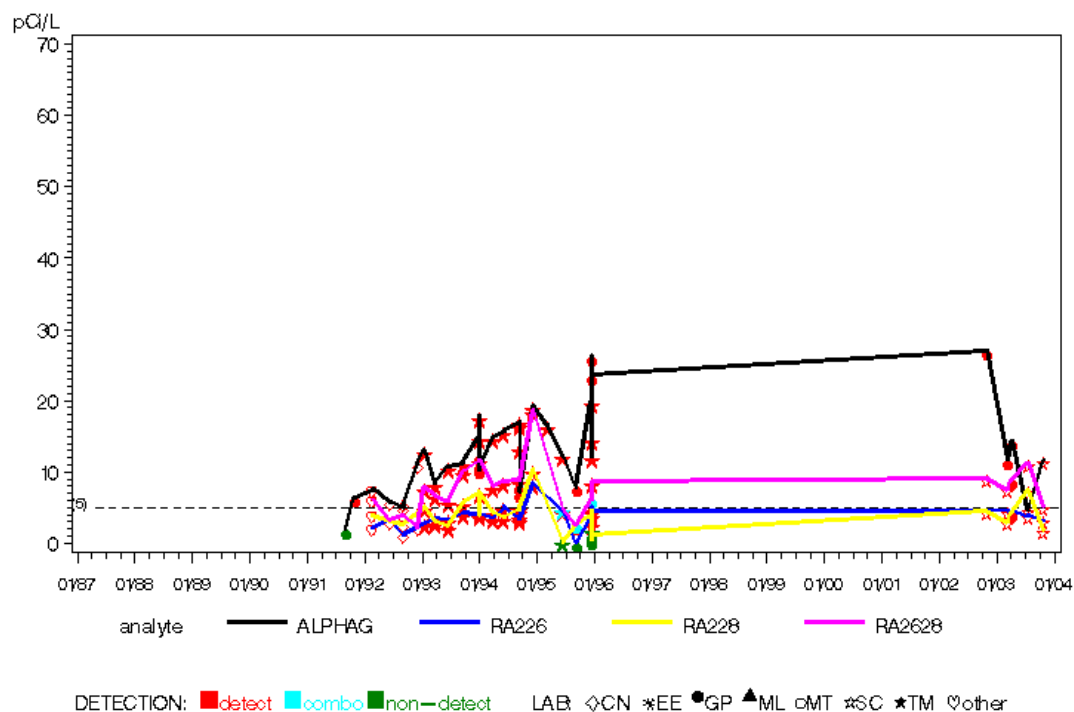
SLF Wells (LFW, LFP Series): Recent Elevated Radium

WELL= LFW 47C

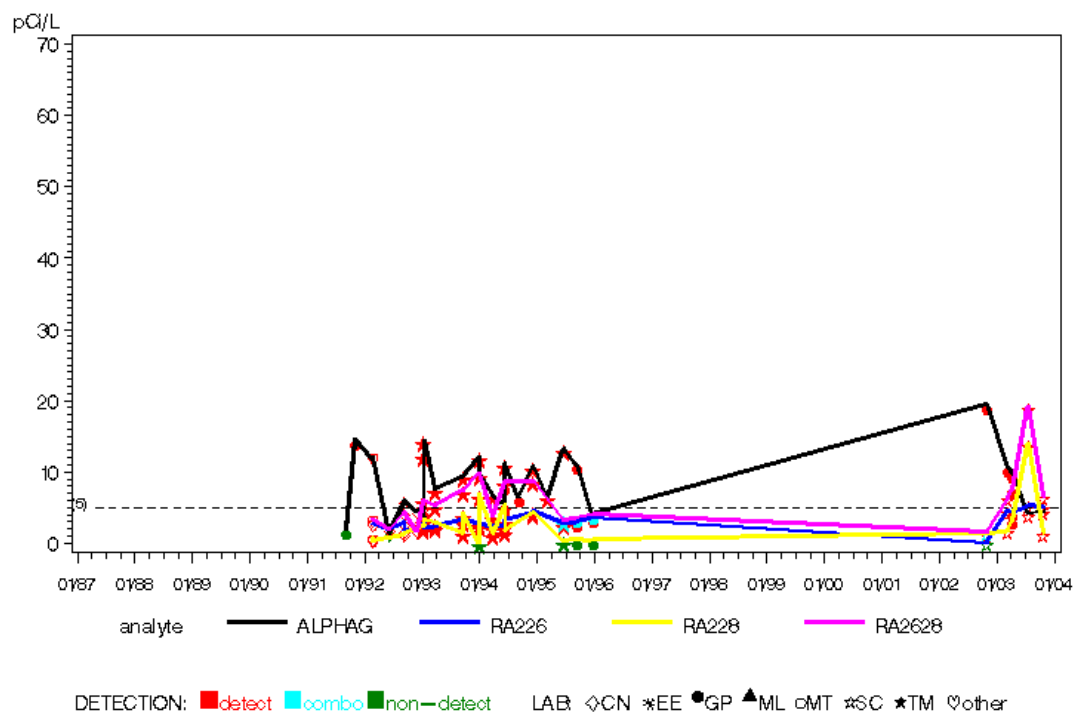


SLF Wells (LFW, LFP Series): Recent Elevated Radium

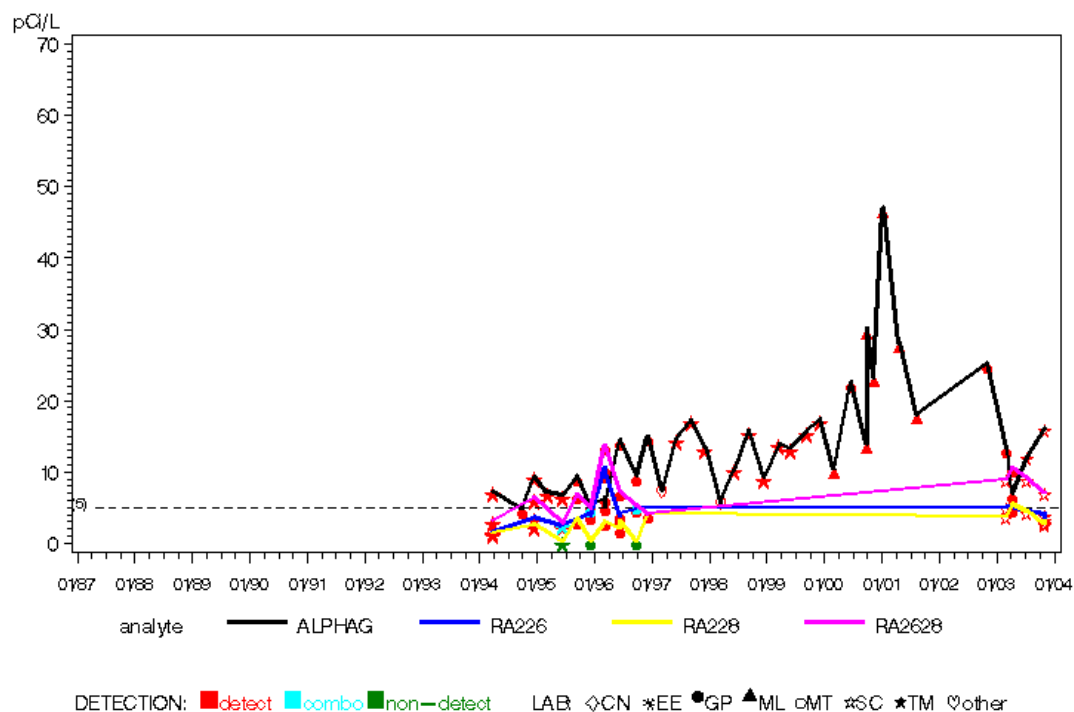
WELL= LFW 57B



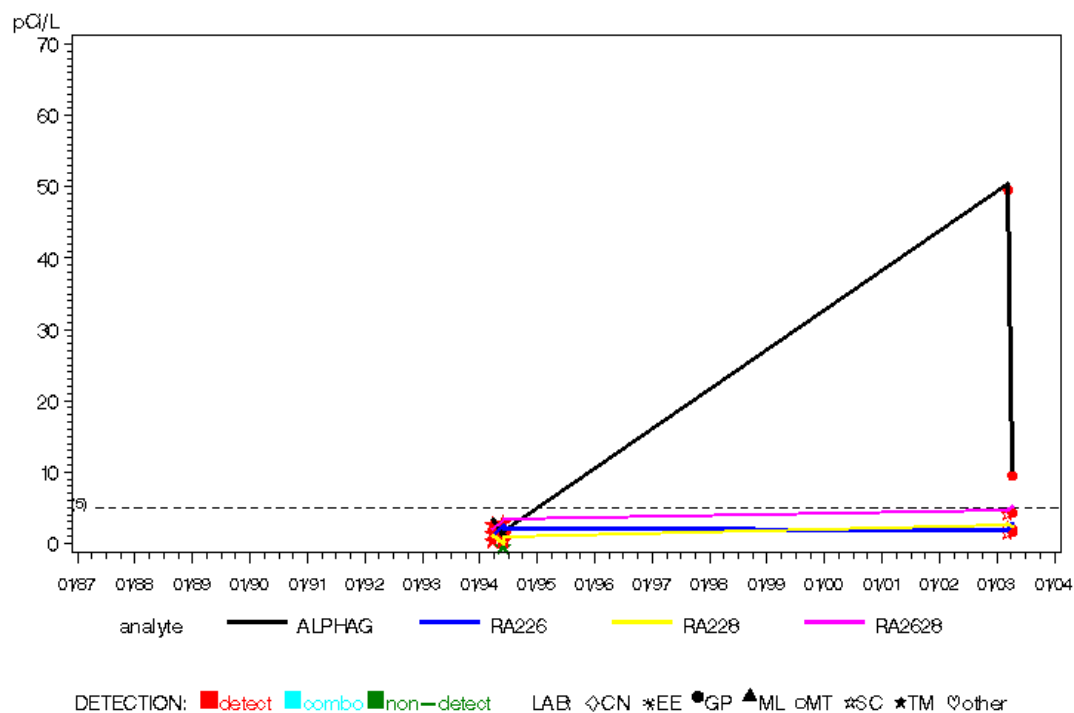
SLF Wells (LFW, LFP Series): Recent Elevated Radium
WELL= LFW 62B



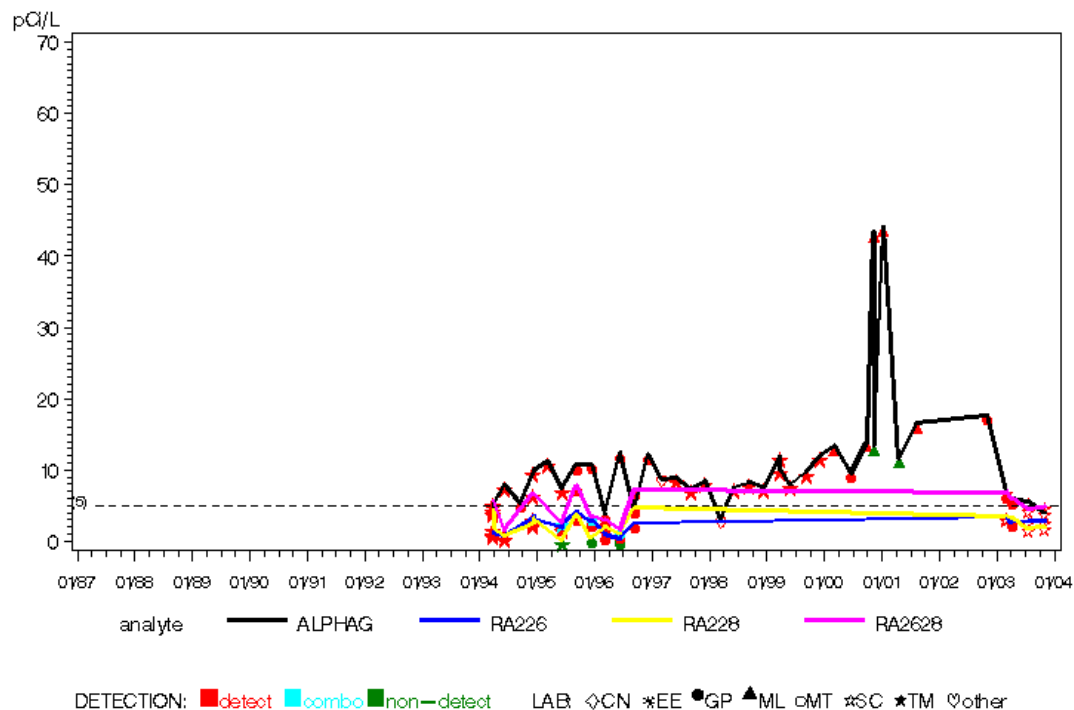
SLF Wells (LFW, LFP Series): Recent Elevated Radium
WELL= LFW 64C



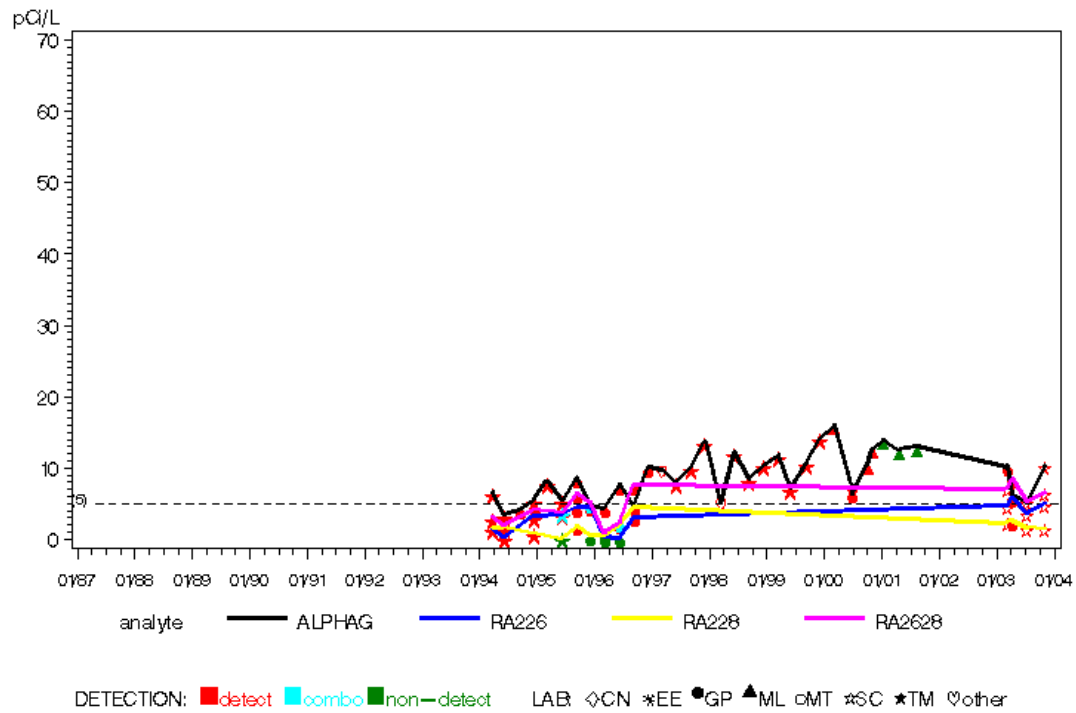
SLF Wells (LFW, LFP Series): Recent Elevated Radium
WELL= LFW 66B



SLF Wells (LFW, LFP Series): Recent Elevated Radium
WELL= LFW 67B

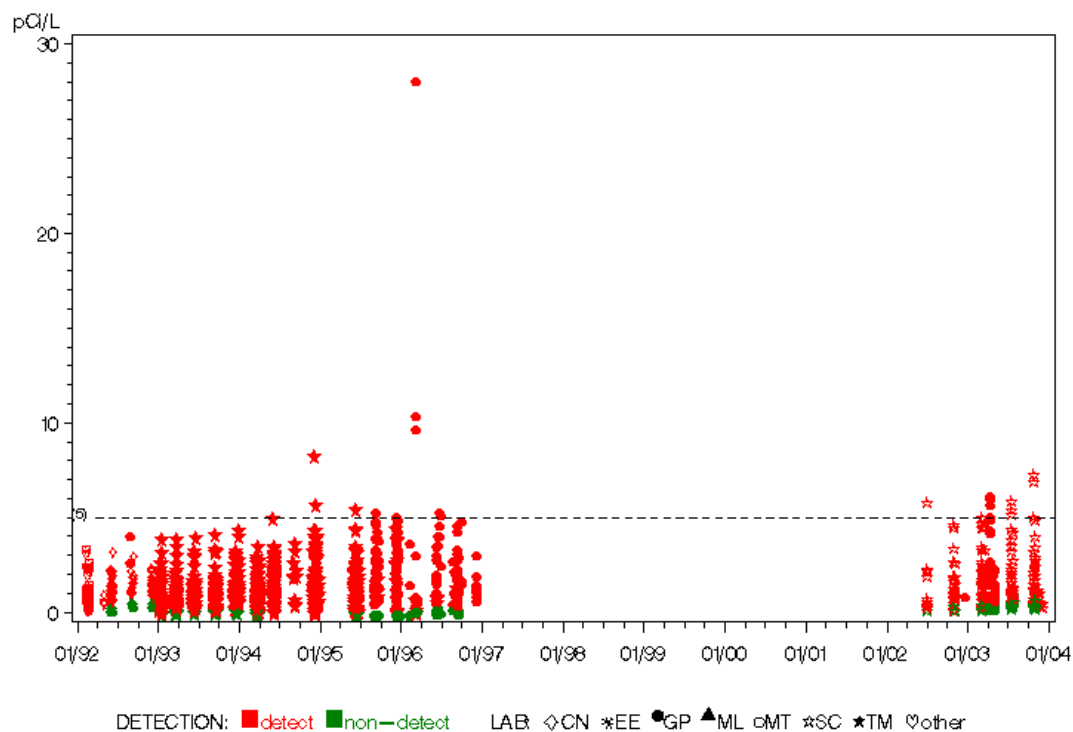


SLF Wells (LFW, LFP Series): Recent Elevated Radium
WELL= LFW 69C

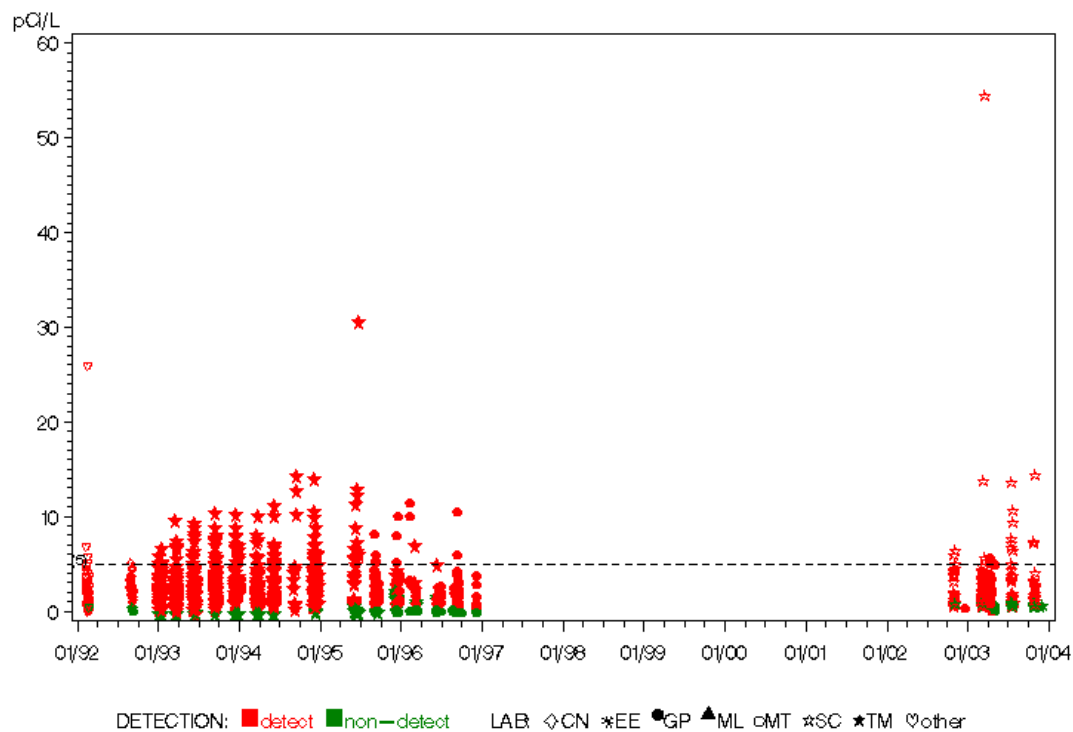


Appendix B2 – Time series plots by analyte

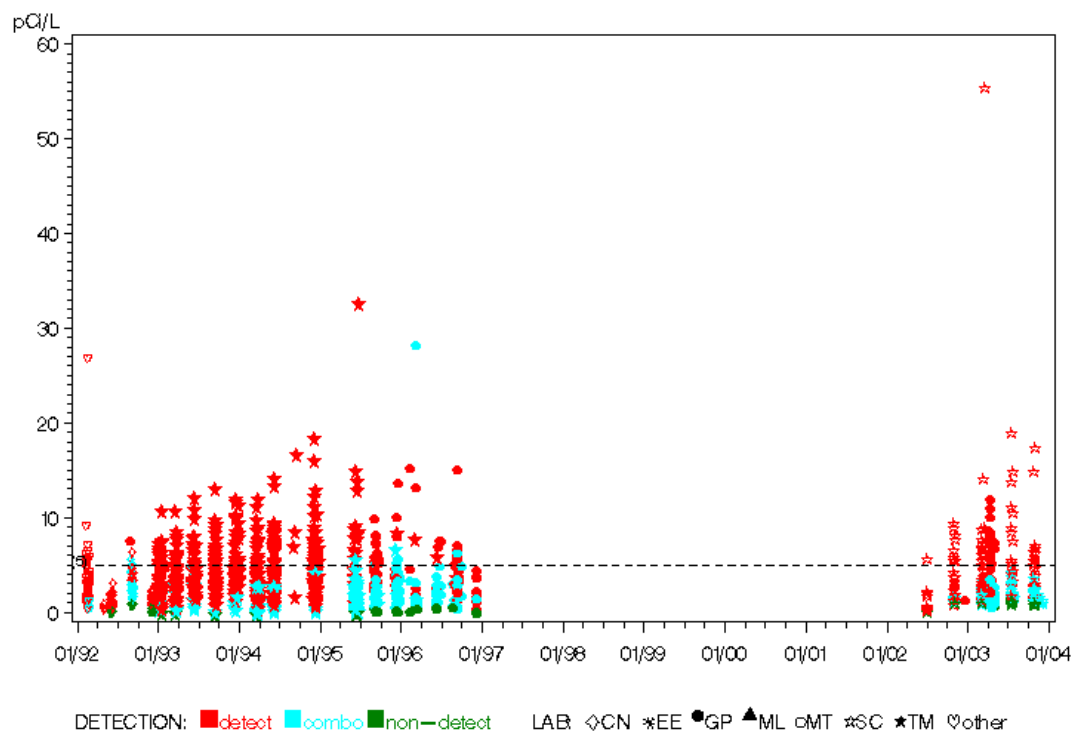
Ra226 in All LFP/LFW Wells



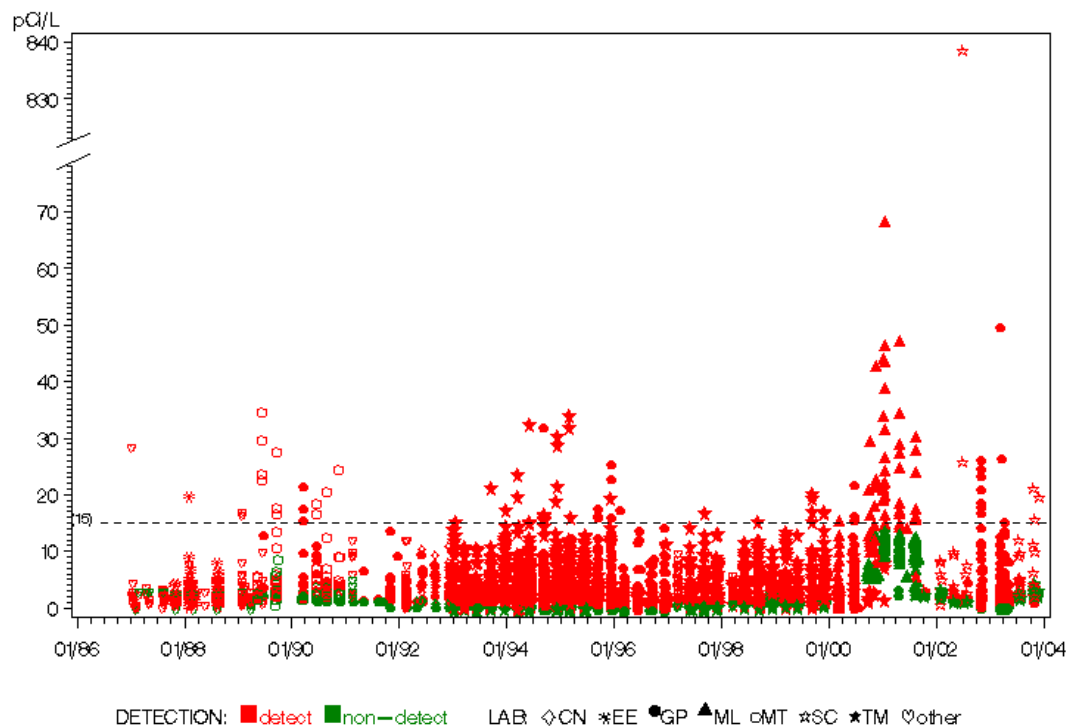
Ra228 in All LFP/LFW Wells



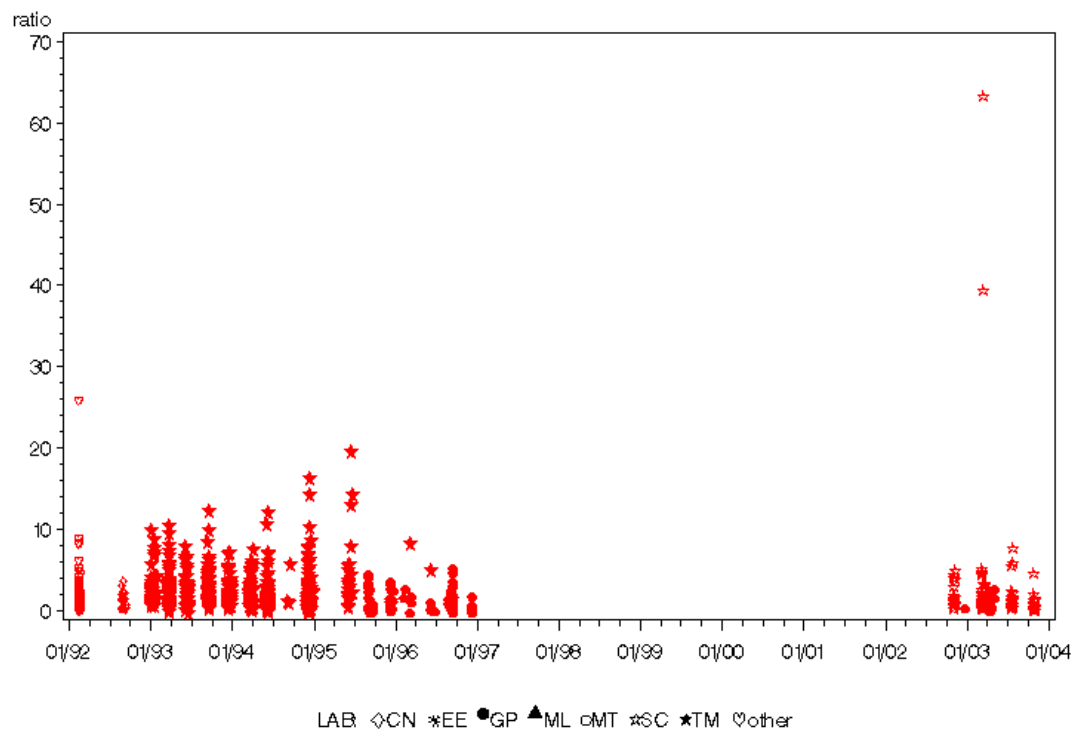
Ra226 + Ra228 in All LFP/LFW Wells



Gross Alpha in All LFP/LFW Wells

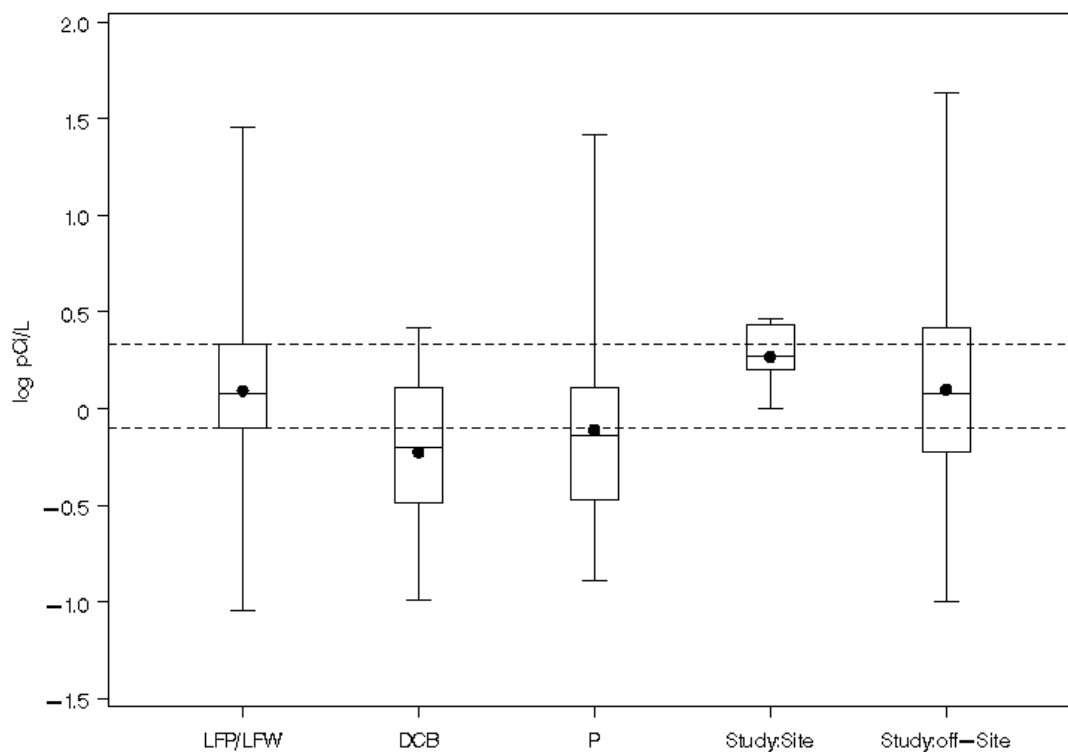


Ratio of Detected Ra228 to Ra226 in All LFP/LFW Wells

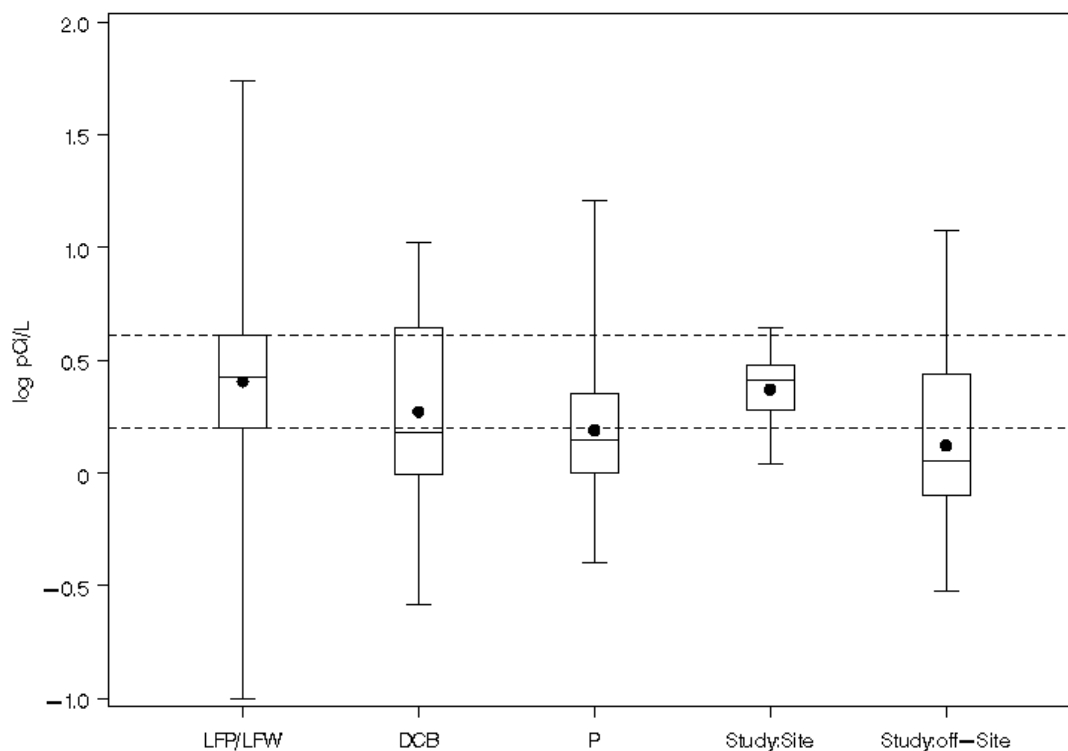


Appendix B3 –Comparative Boxplots for Ra226, Ra228, and Gross Alpha

Log of Detected Ra226 in All LFP/LFW Wells and in Comparison Data



Log of Detected Ra228 in All LFP/LFW Wells and in Comparison Data



Log of Detected Gross Alpha in All LFP/LFW Wells and in Comparison Data

