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# Statistical Comparison of Gross Alpha and Radium Measurements Between Background and Point of Compliance (POC) Wells at the SRS Sanitary Landfill (SLF)

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## Executive Summary

Statistical analyses were performed on groundwater monitoring data obtained for gross alpha, radium (Ra) 226, and 228 from the SRS Sanitary Landfill (SLF). Several inference tests were performed using the prescribed statistical methods of the Resource Conservation and Recovery Act (RCRA) Part B Permit regulations (US EPA 1992). Test results show that the LFW 29R, LFW 57B and LFW 62B wells have significantly elevated concentrations of all three radiological constituents above background. However, isotopic ratios of Ra228 to Ra226 are consistent with naturally occurring concentrations of each. In addition, corresponding pH data for these same three wells suggests that acidic conditions may have effected a mobilization of naturally occurring radium from the soils associated with the SLF. In fact, there is a statistically significant and negative correlation between gross alpha, Ra226, and Ra228 measurements vs pH.

## Introduction

Recent (2003) lab analyses of routine groundwater samples collected from the Sanitary Landfill (SLF) monitoring well network found that some radiological constituents of concern (COCs) were elevated above the regulatory groundwater protection standard (GWPS). These COCs are gross alpha, Ra226, and Ra228. A subsequent statistical analysis among all wells in the SLF network indicated that these elevated measurements are reliable. However, other evidence suggested that they are the consequence of naturally occurring subsurface conditions (see Tuckfield et al. 2004 (WSRC-TR-2004-00141)).

The report herein presents the results of a specific series of statistical inference tests as required by the RCRA 40 CFR 264 Subpart F regulations for post closure care of the SLF facility (US EPA 1992). These regulations require a detection monitoring program that periodically compares the concentration of COCs in Background monitoring wells to other individual monitoring wells along the point of compliance (POC) perimeter of the post closure care facility (e.g., SLF).

The purpose of this report is to document the statistical analysis results that determine whether or not gross alpha, Ra226, or Ra228 concentrations are significantly higher in the SLF POC wells compared to Background wells.

## Methods

### Data Retrieval and Qualification

Measurement data were retrieved from ERDMS, the Environmental Restoration Data Management System, for five Background and thirteen POC wells in the SLF

groundwater monitoring network. Version 8.2 of the SAS System for Windows from SAS Institute was utilized to retrieve and statistically qualify these data via various types of data displays.

Data were retrieved for gross alpha (ALPHAG), Ra226, and Ra228. The five Background wells are LFW 29, LFW 31, LFW 43B, LFW 43C, and LFW 43D. The thirteen POC wells are LFW 23R, LFW 36R, LFW 41R, LFW 48C, LFW 48D, LFW 57B, LFW 59C, LFW 59D, LFW 61C, LFW 61D, LFW 62B, LFW 62C, and LFW 62D. Table 1 below provides pertinent well construction information on each of these RCRA required wells in the SLF monitoring network.

**Table 1.** Sanitary Landfill (SLF) Background and POC wells

	Well	Screen Zone	Well Depth (ft)	Installation Date
<b>Background Wells:</b>	LFW 29	D	51.4	10/28/1986
	LFW 31	D	84.3	10/27/1986
	LFW 43B	B	112.6	6/21/1991
	LFW 43C	C	74.1	6/25/1991
	LFW 43D	D	52	6/24/1991
<b>POC Wells:</b>	LFW 23R	D	60	3/6/1996
	LFW 36R	D	52	3/8/1996
	LFW 41R	D	55	3/6/1996
	LFW 48C	C	61.1	5/28/1991
	LFW 48D	D	34.6	5/23/1991
	LFW 57B	B	97	6/11/1991
	LFW 59C	C	67	6/18/1991
	LFW 59D	D	38.3	6/18/1991
	LFW 61C	C	57.3	5/20/1991
	LFW 61D	D	38	5/20/1991
	LFW 62B	B	102.1	6/11/1991
	LFW 62C	C	57.1	6/11/1991
	LFW 62D	D	37.2	6/5/1991

The SLF monitoring well network consists of LFW series monitoring wells and LFP series piezometers that circumvent the SLF and was installed in the water table (Steed Pond Aquifer Unit, SPA) beginning in 1975.

The placing of well screens in the water table aquifer included screens to intercept the

- water table surface (designated D)
- water approximately 25-30 feet below the water table surface (designated C)

- water just above the Upper Clay Zone of the Crouch Branch Confining Unit, approximately 50-60 feet below the water table surface (designated B)

The following data were retrieved from ERDMS database tables

- the well name
- 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 (e.g., pCi/L)
- review and lab qualifiers
- qualification codes
- lab identifier
- analytical method
- sample quantitation limit (SQL) and units
- validation & verification status

The result qualifier was defined from the lab and review qualifiers, and the units were all converted to a consistent basis (pCi/L) 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 (or zero if the result was 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. 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.

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).

The analytical data were then averaged sequentially to obtain quarter averages for the statistical 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. A detection value was also determined for each quarter average.

Each of the wells was assigned a category based on the concentrations of Ra2628 and gross alpha by sample date. Concentrations of 5 pCi/L or above are considered elevated for Ra2628, and concentrations of 15 pCi/L or above of gross alpha are elevated. Sample dates from 2003 on were categorized as recent, while earlier dates are considered old. The Ra2628 categories are “RA2628 elev recent,” “RA2628 elev old”, and “RA2628 not elev.” Similar categories were defined for gross alpha using the analyte code ALPHAG. Each well was labeled with the levels of Ra2628 and gross alpha. For example, Background well LFW 29 has category “RA2628 elev recent and ALPHAG not elev.”

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.

### Data Displays

This report provides the following types of data displays.

- Time series plots – to illustrate, per well, the temporal contaminant concentration data relative to the concentration limit (constructed from individual qualified data)
- Box-and-whisker plots – to illustrate the comparison of gross alpha, Ra226, Ra228, Ra2628, and the isotopic ratio measurements in the individual SLF POC wells to those from the SLF Background wells (constructed from individual qualified data)
- Dunnett’s test plots – to illustrate the application of this statistical test of significance for each POC well against Background by analyte (constructed from quarter averages) and featured in the Results section below

For each of the eighteen wells, a time series plot displays the concentrations of analytes Ra226, Ra228, Ra2628, and gross alpha through time (Appendix A). Lab is distinguished by plot symbol, detection by plot symbol color, and analyte by colored lines. The limits for radium (5 pCi/L) and gross alpha (15 pCi/L) are shown as dotted lines. The plots are scaled identically to facilitate comparison between wells. The concentration (Y) axis ranges from 0 – 50 pCi/L, and the sample date (X) axis ranges from 1/1/1987 to 1/1/2004. The plot header indicates the classification level for Ra2628 and gross alpha.

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 plus sign for the average. A line called a whisker is drawn from the 25th quantile (bottom of box) to the smallest observation within 1.5 times the interquartile range from the bottom. Another whisker is drawn from the 75th quantile (top of box) to the largest observation within 1.5 times the interquartile range from the top. In other words, the whiskers are drawn from the box to the largest/smallest observations not considered outliers. Symbols are used to represent values past the whiskers, considered outliers. In these particular boxplots, the far outliers (beyond 3 times the interquartile range from the 25th and 75th percentiles) have been labeled with their values. Comparison of boxplots is a nonparametric approach to data analysis, meaning that assumptions about the distribution of the data population are not needed. Note that there are two graphs for each analyte since the thirteen POC wells cannot fit on a single graph and that the boxplot for Background appears first on each of the two. The data are presented on the common log scale to minimize skewness.

#### Isotopic Ratio Data

Isotopic ratios of Ra228 to Ra226 were constructed for comparison of POC wells to Background. Radium isotopes are an important indicator of the origin of elevated radium concentrations in groundwater. A daughter of Th232 with a half-life of 5.75 years, Ra228 will reach secular equilibrium with Th232 in less than 60 years. A daughter in the decay chain of U238 with a half-life of 1600 years, Ra226 will take over 2 million years to reach secular equilibrium with U238. At a site where processed Th232 was disposed, Ra228 concentrations may be elevated from decay of the Th232. In contrast, elevated concentrations of Ra226 cannot be derived from disposal of processed U238 within a reasonable time frame. Thus, an explanation for elevated radium concentrations must account for the Ra228/Ra226 ratio, as well as for the actual concentrations.

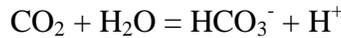
Radium emanating from a point source such as a disposal site should show a systematic variation in the Ra228/Ra226 ratio within the plume due to the different half-lives of the two isotopes. As radium moves with groundwater, the ratio decreases because of Ra228 decay relative to Ra226. Consistent Ra228/Ra226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

#### pH Data

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. 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. Radium is more mobile at acidic conditions because the solubility of minerals that contain radium increases and adsorption of radium to soil decreases. Benes (1990) illustrates decreasing adsorption with a figure showing that adsorption of radium onto ferric hydroxide decreases below a pH of 9, and is near zero by pH 6. Likewise, the adsorption onto kaolinite begins to decrease at a pH of about 7. Also, the solubility of radium-bearing phosphate minerals such as apatite and monazite increases below a pH of about 5. As these minerals dissolve, radium is released to groundwater. Thus, Ra226 and Ra228 concentrations should correlate with pH at pH values below 7. In turn, Ra226 contributes to gross alpha and this combined with an increase in uranium concentration under acidic conditions explains the correlation between pH and gross alpha.

The pH of groundwater in some of the SLF wells has been historically below typical SRS values. Acidic groundwater can result from direct dissolution of acid producing materials such as aluminum salts or oxidation of ferrous iron materials. Alternatively, acid may be produced from biodegradation of organic materials in the landfill. As organic materials degrade, CO<sub>2</sub> may be produced and will form acid by the reaction:



Thus, pH values of associated groundwater are a function of partial pressure of CO<sub>2</sub> (PCO<sub>2</sub>) in the aquifer. Atmospheric PCO<sub>2</sub> is 10<sup>-3.5</sup> atm., but typical values of SRS aquifers are on the order of 10<sup>-2</sup> atm. Degradation of organic materials can raise PCO<sub>2</sub> to values exceeding 10<sup>-1</sup> atm. Figure 1 shows the effect of PCO<sub>2</sub> on pH of unbuffered water. As PCO<sub>2</sub> increases to a value of 10<sup>-1</sup> atm., pH decreases to 4.4. Values of pH below 4 indicate an additional source of acid such as oxidation of sulfide or dissolution of aluminum or ferrous iron salts.

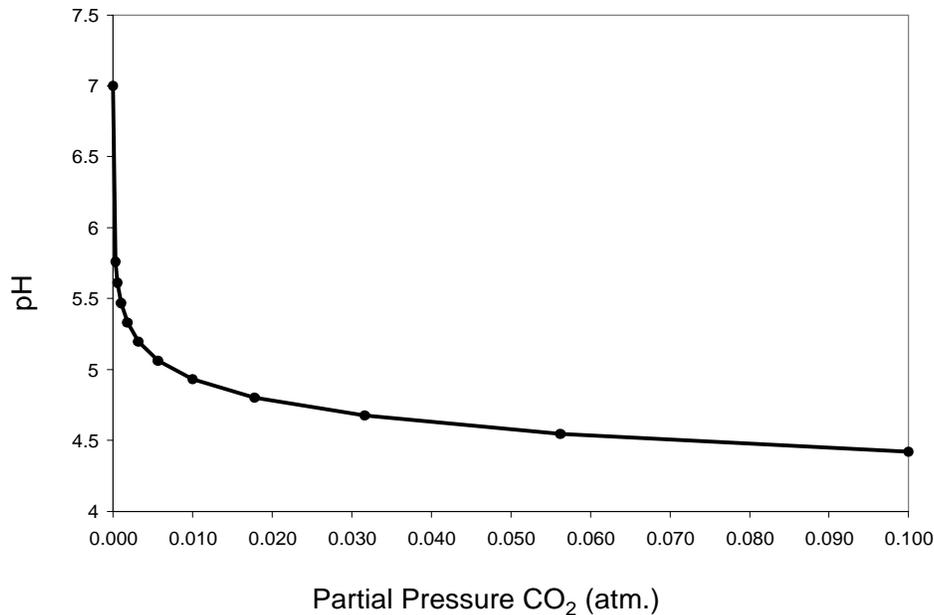


Figure 1. pH versus PCO<sub>2</sub> in unbuffered water.

### Statistical Inferences and Comparisons

In section 264.97 of the federal RCRA legislation referenced above, several statistical methods are allowed in order to test the null hypothesis that none of the POC wells show a significantly elevated COC concentration above Background. Each POC well must be compared separately to Background regardless of the statistical method selected.

The approved statistical method selected for the data analyses reported here is Analysis of Variance (ANOVA). In addition, the Dunnett's test (Steel and Torrie 1980) was used in conjunction with the ANOVA method as a means of making specific comparisons of data from each POC well to the combined data among all five Background wells. All ANOVAs and Dunnett's test comparisons were performed using the JMP 5.01 software from SAS Institute, Cary, NC.

Histograms of the data retrieved from the ERDMS database and the Ra226/228 ratios showed substantial skewness, and a subsequent Shapiro-Wilk W test (Gibbons 1994) for each of the four principle COCs (viz., gross alpha, Ra226, Ra228, and Ra2628) and the Ra228/226 ratio showed statistically significant ( $p < .05$ ) departures from the standard ANOVA assumption of normality. Each of these five response measures was then transformed to a common logarithm scale prior to the statistical analyses. Among these log transformed variables, log Ra226, log Ra228, and log Ra2628 still showed statistically significant ( $p < .05$ ) departures from normality, although the histograms of these variables showed the typical bell-shaped form of a normal probability distribution function. Finally, the original five variables were also rank transformed prior to statistical analysis as a nonparametric equivalent (Conover 1999) to an ANOVA using the log transformed data.

### Results

Time series plots (one for each well) are intended to show any temporal trends among the four principle radiological COCs. The data in these plots included all individual sample measurements by sampling date as a means of identifying the within quarter variability. These plots are presented in Appendix A and collectively show little evidence for time trends among these COCs. However, there is an indication (not shown here) that some recently high measurements and high detection limits were associated with one analytical laboratory, viz., the Mobile Lab.

The series of box-and-whisker (box) plots provided in Appendix B allow for an overall comparison of the quarterly averaged measurements for the same four COCs among wells. These plots of log transformed data identify several POC wells with consistently higher measurements than those for the combined Background well data. These POC wells are LFW 23R, LFW 57B, and LFW 62B. Interestingly, Appendix B box plots of pH measurements in the same groundwater samples show that pH is substantially lower

in the same three POC wells compared to Background.

Figure 2 shows the results of the ANOVA comparison of COC monitoring well means via the Dunnett’s test. Test results are reported for the log transformed measurements. Note that there are 13 comparisons, one for each POC well versus the combined data from Background wells. Each well comparison has the same error rate (5%), that is, the same probability of declaring a statistically significant difference when, in fact, there is none. Decision error rates • .05 are a sufficiently small risk. A significant ( $p < .05$ ) difference from Background is indicated by a “black” text color for the POC Well ID. The text for all other POC Well IDs is “red” indicating *no* significant difference from Background (whose Well ID text color is also “red”). To the right of the boxplot series for each COC in Figure 2 are Comparison Circles. The center of each circle is the mean COC value for the corresponding Well ID, and the diameter of each circle is the 95% confidence interval for the Well ID mean. If the POC well circle does not intersect with the Background circle or if the inside angle between the two circles is  $< 90^\circ$ , we may conclude that the POC well differs significantly ( $p < .05$ ) from Background. Otherwise, there is no significant difference between Background and POC well. Table 2 summarizes these results.

Table 2. POC wells that were statistically different ( $p < .05$ ) from Background for each of the COCs.

(Symbols: < significantly less than Background, > significantly more than Background.)

Well ID	Gross Alpha	Ra226	Ra228	Ra2628	Ra226/228 Ratio
LFW 23R	>	>	>	>	
LFW 36R			>		
LFW 41R				>	
LFW 48C	<			<	
LFW 48D			>	>	
LFW 57B	>	>	>	>	
LFW 59C					
LFW 59D					
LFW 61C	<			<	
LFW 61D					
LFW 62B	>	>		>	<
LFW 62C					
LFW 62D					

The same statistical analyses were performed on the rank transformed data and on data without the measurements from the Mobile Lab. In either case, the statistical test results were virtually identical.

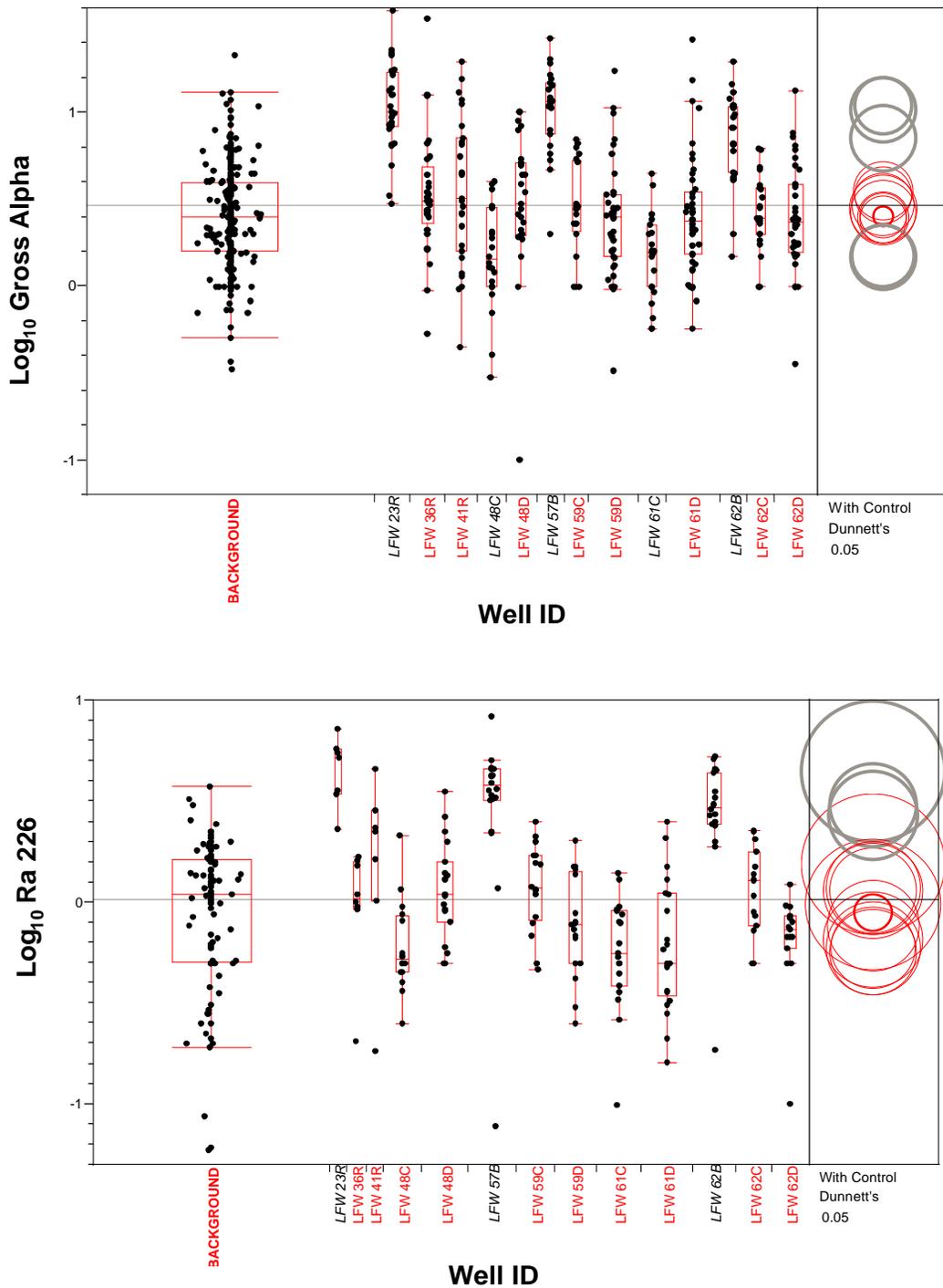


Figure 2. Quarterly measurement ANOVA results via Dunnett's Test. A comparison of each POC well versus the combined data from five Background wells is shown for each of five constituents of concern, viz., gross alpha, Ra226, Ra228, total Radium (Ra2628), and the Ra 228/226 Ratio. Note also that data were transformed to a common log scale. All Well IDs (names) in black are significantly different ( $p < .05$ ) than BACKGROUND. See text for explanation of comparison circles to the right of each boxplot.

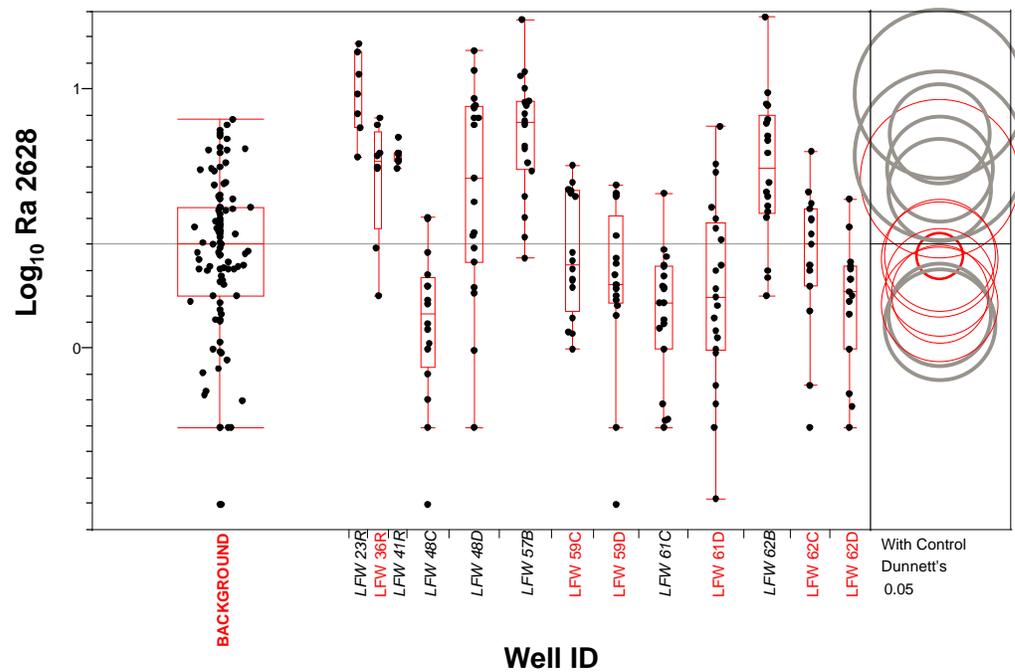
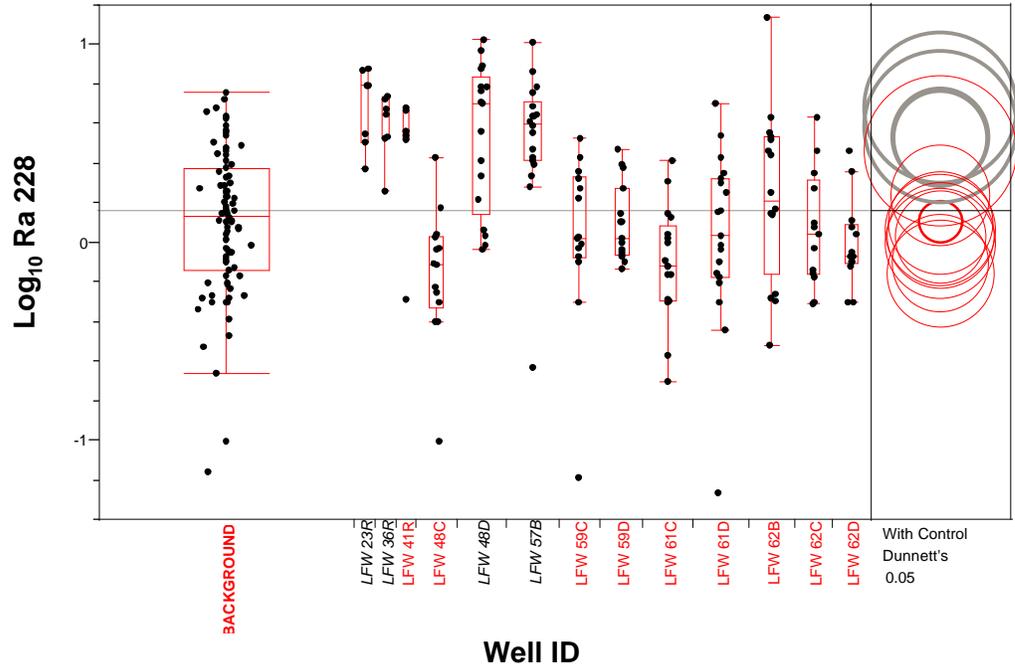


Figure 2. Continued.

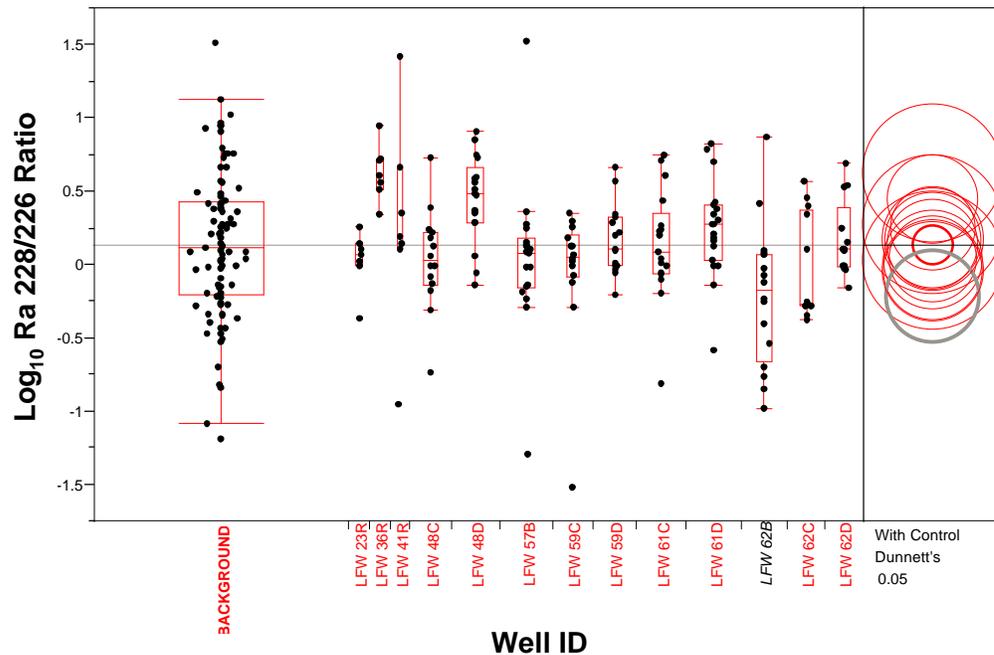


Figure 2. Continued.

## Discussion

It appears that gross alpha, Ra226, Ra228, and total radium (Ra2628) are significantly elevated above background in several POC wells (Fig. 2). However, only well LFW 62B showed a significantly lower Ra228/226 ratio than Background.

Recall, as stated earlier, that radium emanating from a suspected 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 (Tuckfield et al. 2004). Consistent Ra228/Ra226 ratios over a large area are suggestive of a native rock source rather than a point source for radium within a waste site. It would appear then that LFW 62B has a significantly smaller ratio compared to Background. However, we have also shown results that POC wells (LFW 23R, 41R, 57B, and 62B) with the highest concentrations of Ra226 (Figure 2), the principal alpha emitter, also have the lowest pH.

In addition, Figure 3 shows the spatial relation between the Background and these four POC wells at the SLF. The latter are located immediately adjacent to the SLF perimeter, a spatial arrangement in the SLF groundwater monitoring network that invites further investigation beyond the scope of this report.

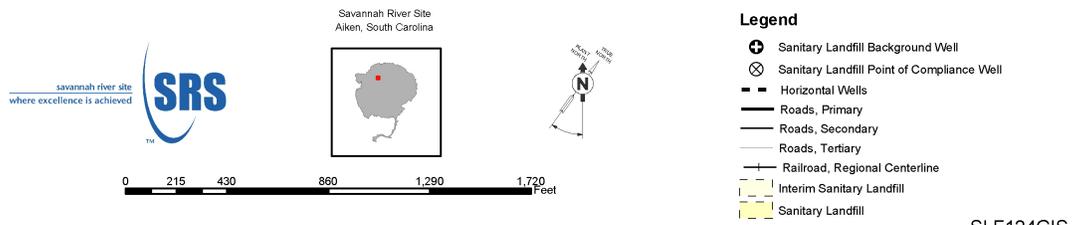
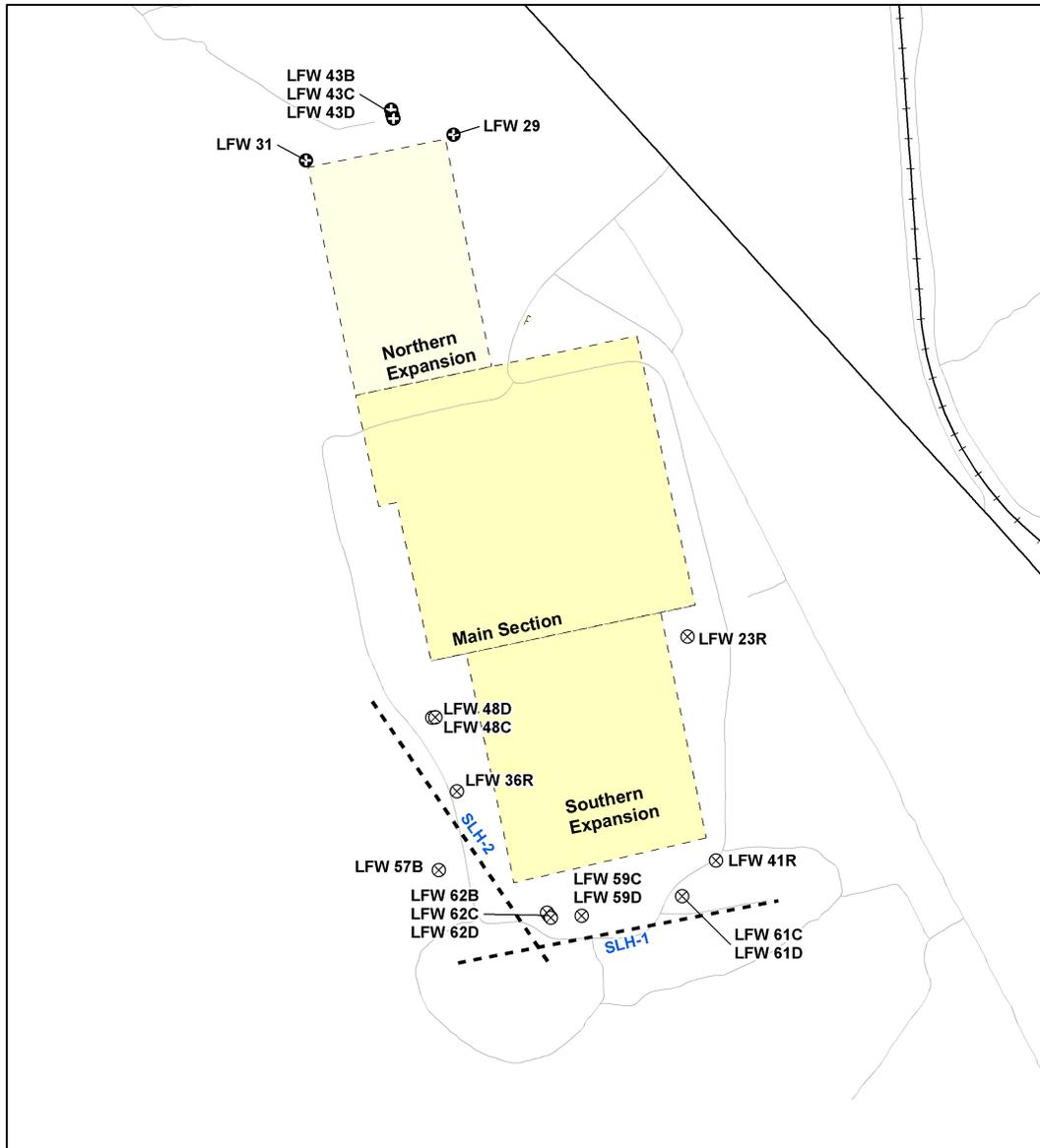


Figure 3. GIS map of Background and POC well locations for the SRS Sanitary Landfill. There are 5 Background wells and 13 POC wells. The 4 POC wells with the lowest average pH measurements are LFW 23R, LFW 41R, LFW 57B, and LFW 62B.

The negative correlation between pH vs gross alpha, Ra226, and Ra228 (see Appendix C) is consistent with the geochemical behavior of radium. As noted earlier, radium is more mobile at acidic conditions because the solubility of minerals that contain radium increases and adsorption of radium to soil decreases. Thus, Ra226 and Ra228 concentrations should correlate with pH at pH values below 7. In turn, Ra226 contributes to gross alpha and this combined with an increase in uranium concentration under acidic conditions would also explain the correlation between pH and gross alpha.

We cannot conclude, however, that the consistently elevated gross alpha and radium 226 above Background in three POC wells is caused by low pH. Causality is not the objective of this report. Therefore, if it is of interest to determine the cause of gross alpha or Ra226 elevation above background, other concomitant and supporting evidence will be required.

Since the intent of this study is a RCRA mandated comparison, we can conclude that gross alpha, Ra226, and Ra228 concentrations in some POC wells are significantly elevated above concentrations of the same among the permitted background wells, collectively, and therefore retained for compliance monitoring purposes.

## References

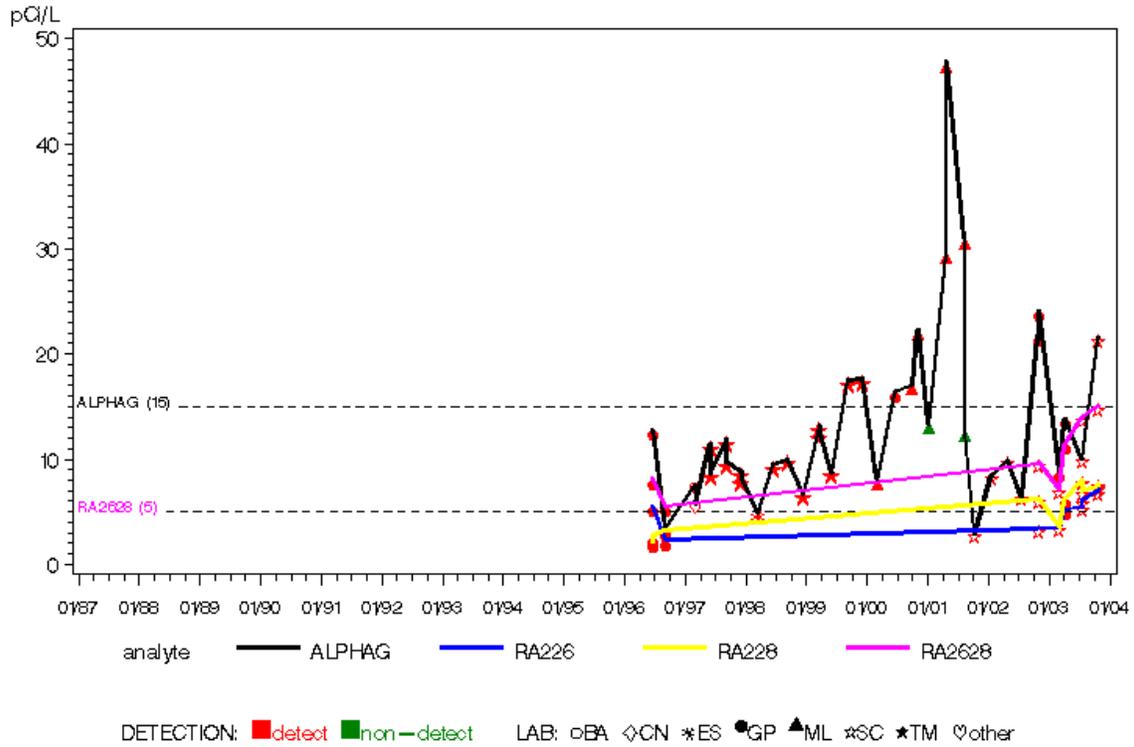
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[http://www.epa.gov/epaoswer/hazwaste/ca/resource/guidance/sitechar/gwmonitr/rcra\\_gw.pdf](http://www.epa.gov/epaoswer/hazwaste/ca/resource/guidance/sitechar/gwmonitr/rcra_gw.pdf)

## **Appendix A**

**Time series plots of gross alpha (ALPHAG), Ra226, Ra228, and Ra2628  
for every RCRA compliance monitoring well (Background and POC) of  
the study**

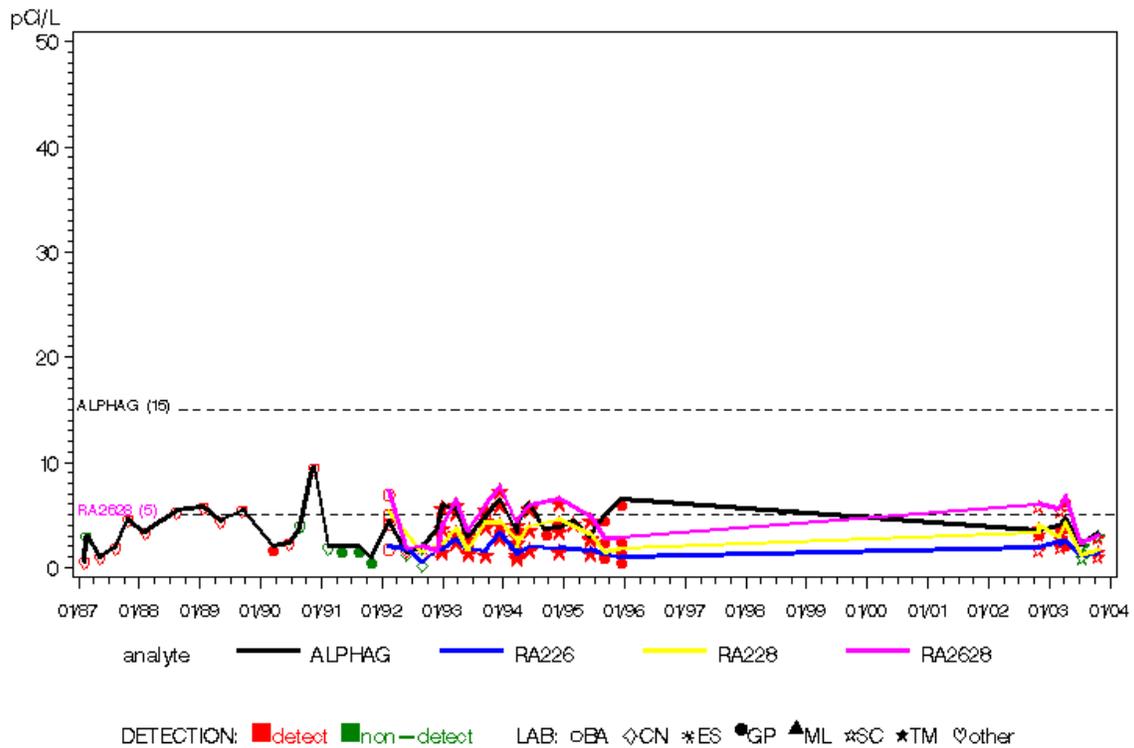
SLF: POC Wells

LFW 23R — RA2628 elev recent & ALPHAG elev recent



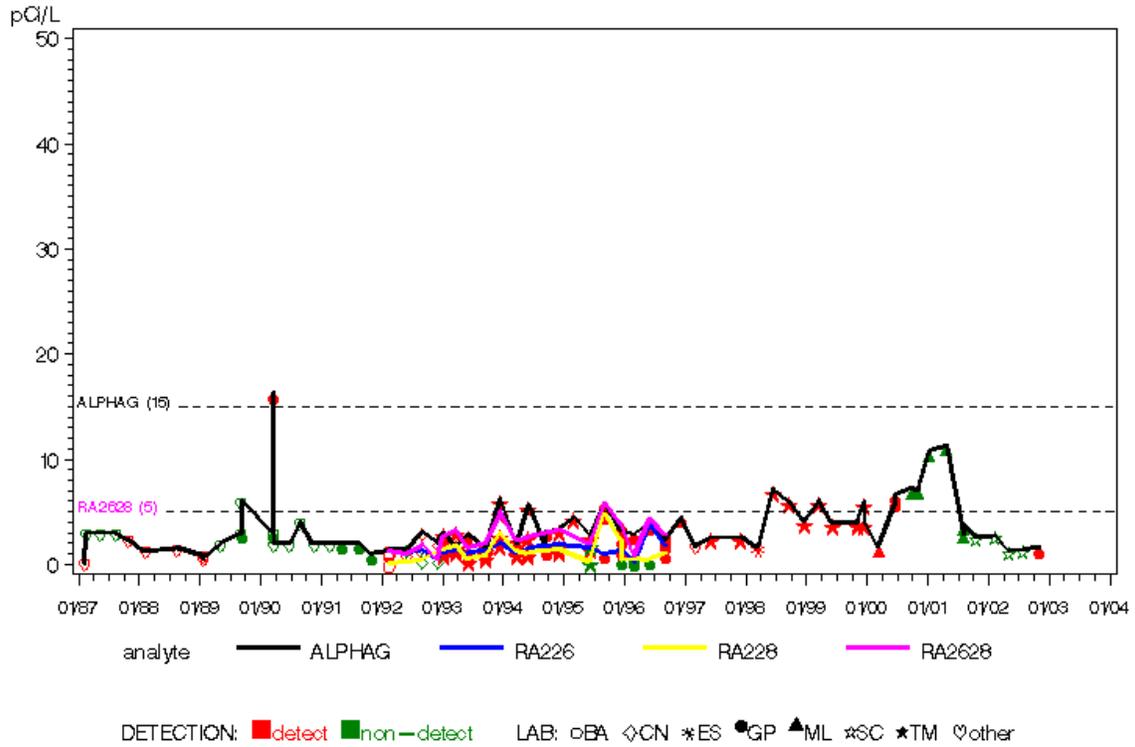
SLF: Background Wells

LFW 29 — RA2628 elev recent & ALPHAG not elev



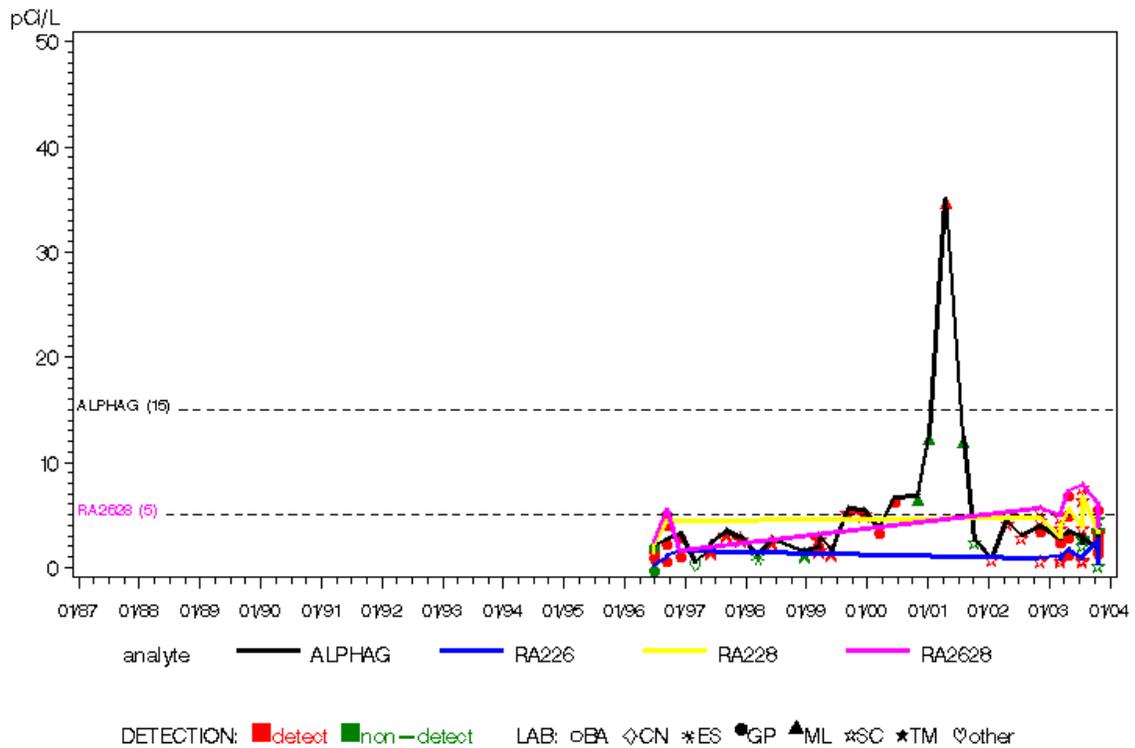
### SLF: Background Wells

LFW 31 — RA2628 elev old & ALPHAG elev old



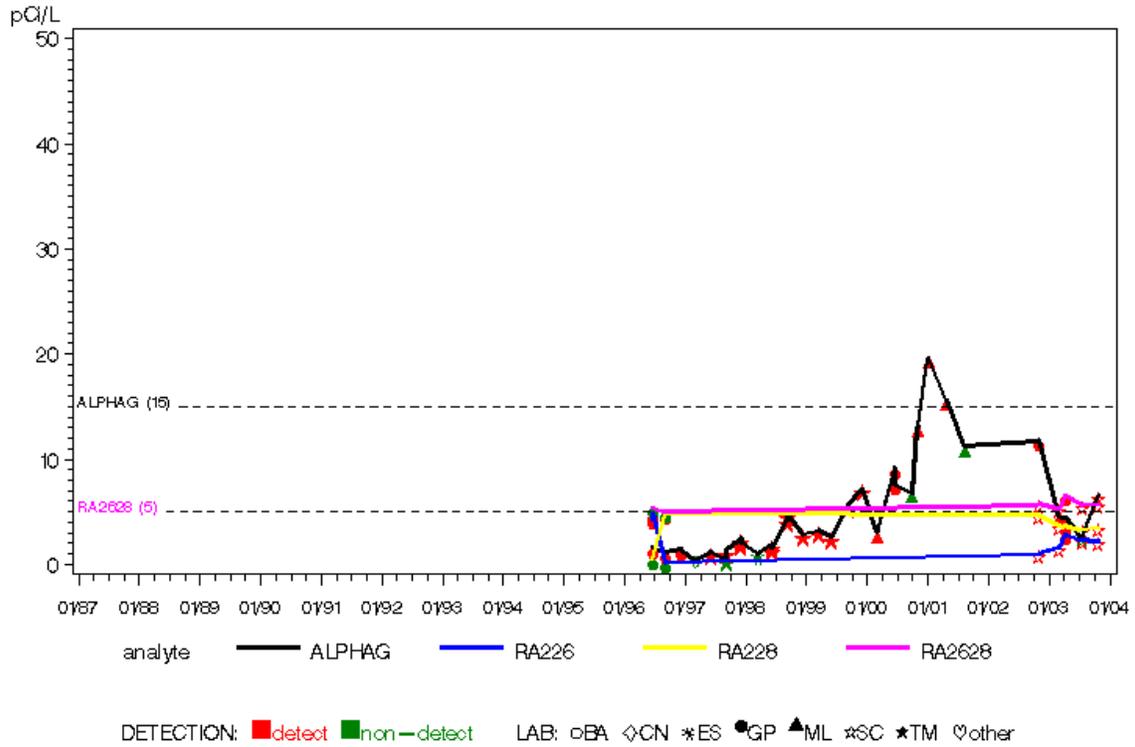
### SLF: POC Wells

LFW 36R — RA2628 elev recent & ALPHAG elev old



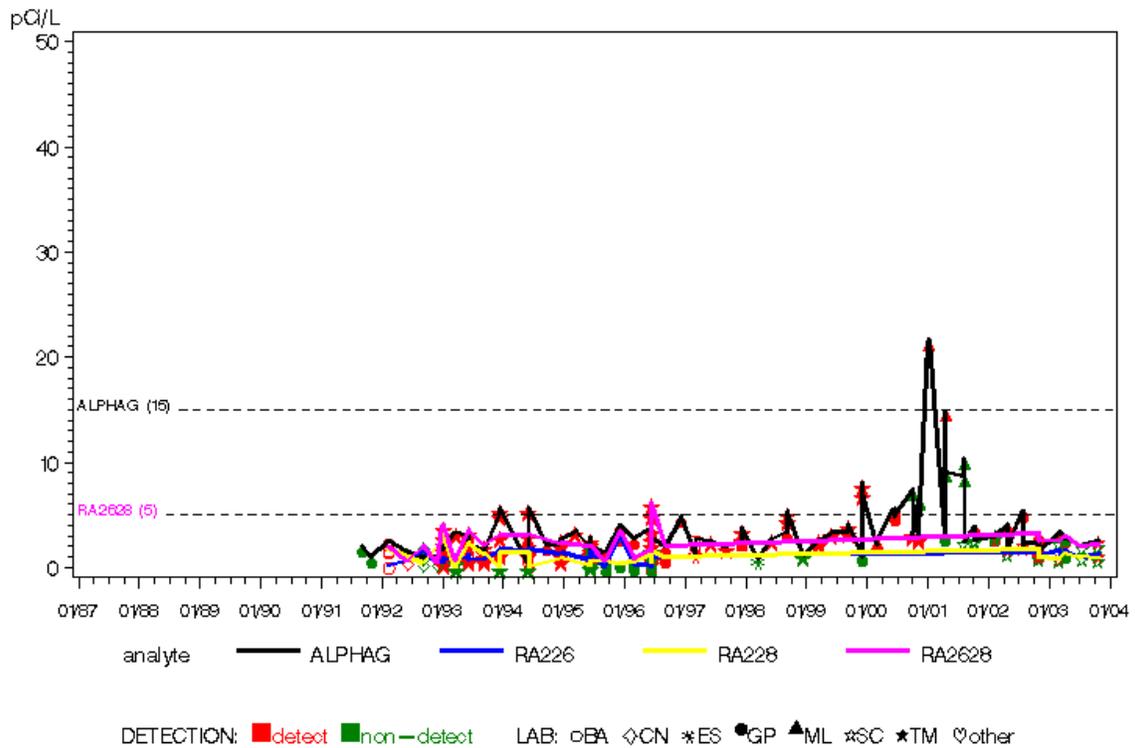
SLF: POC Wells

LFW 41R — RA2628 elev recent & ALPHAG elev old



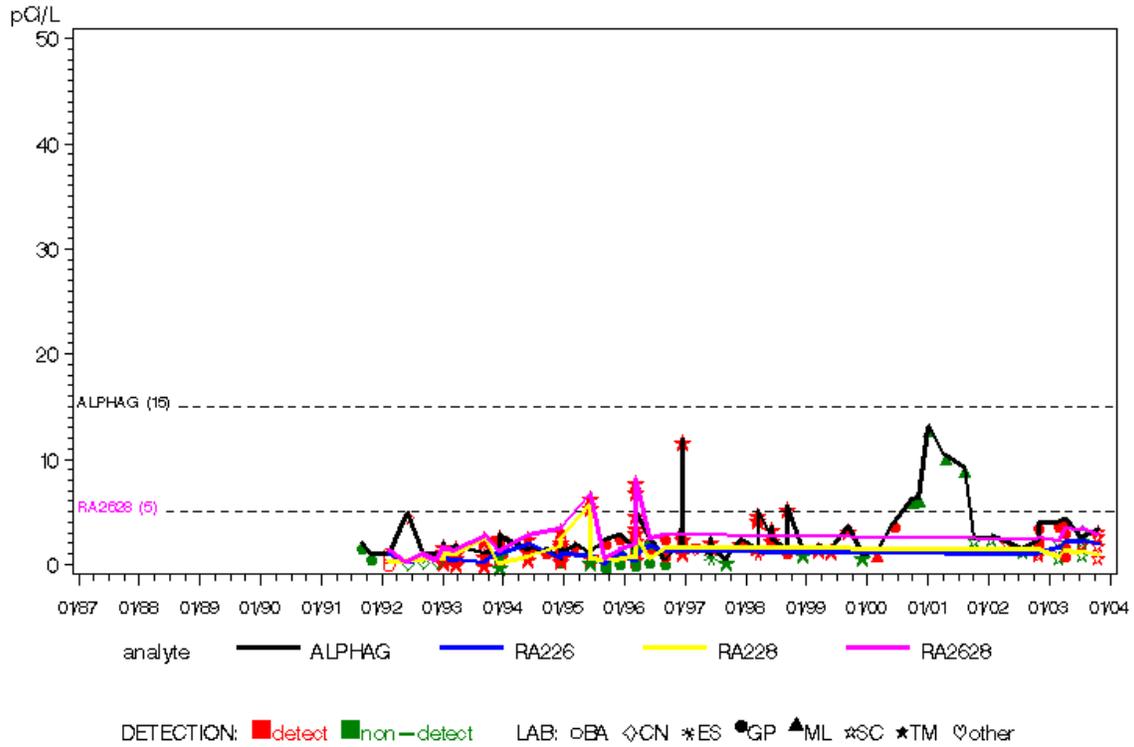
SLF: Background Wells

LFW 43B — RA2628 elev old & ALPHAG elev old



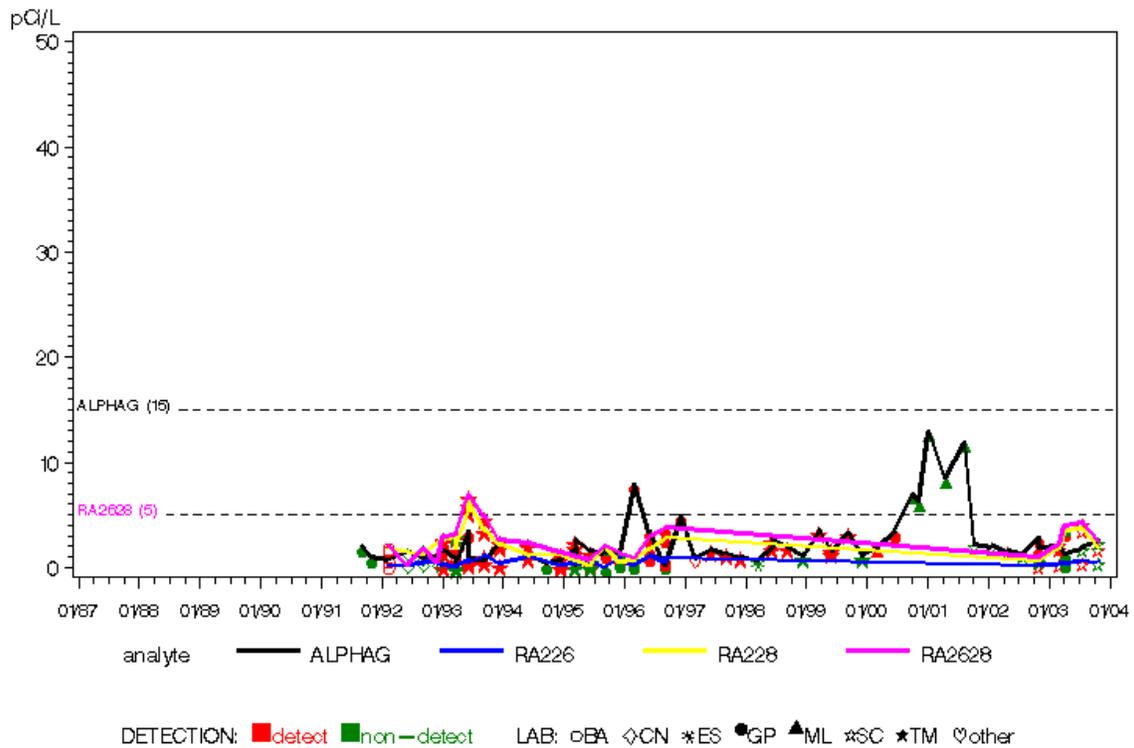
### SLF: Background Wells

LFW 43C — RA2628 elev old & ALPHAG not elev



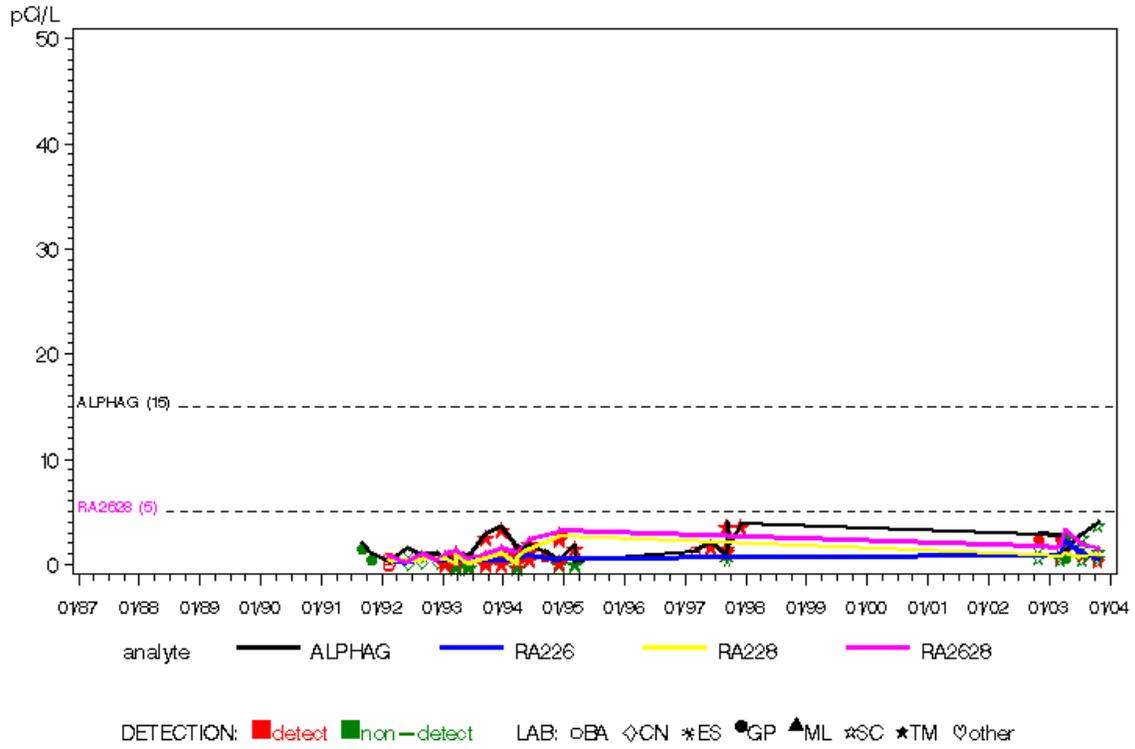
### SLF: Background Wells

LFW 43D — RA2628 elev old & ALPHAG not elev



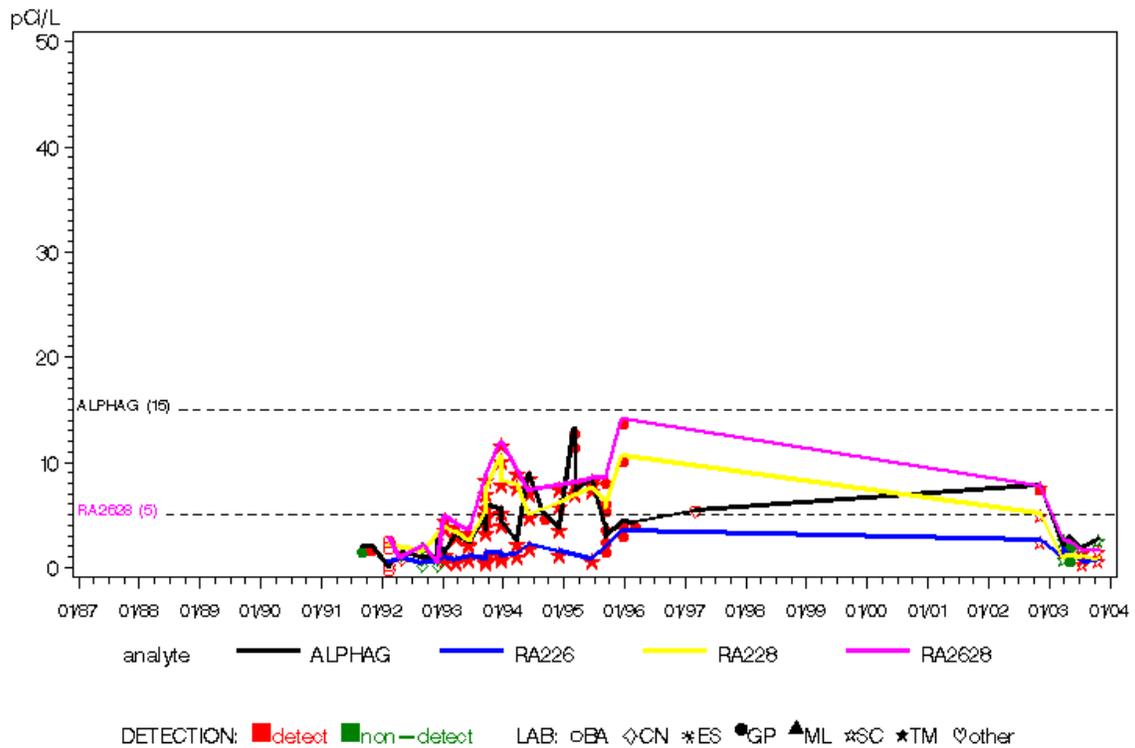
SLF: POC Wells

LFW 48C — RA2628 not elev & ALPHAG not elev



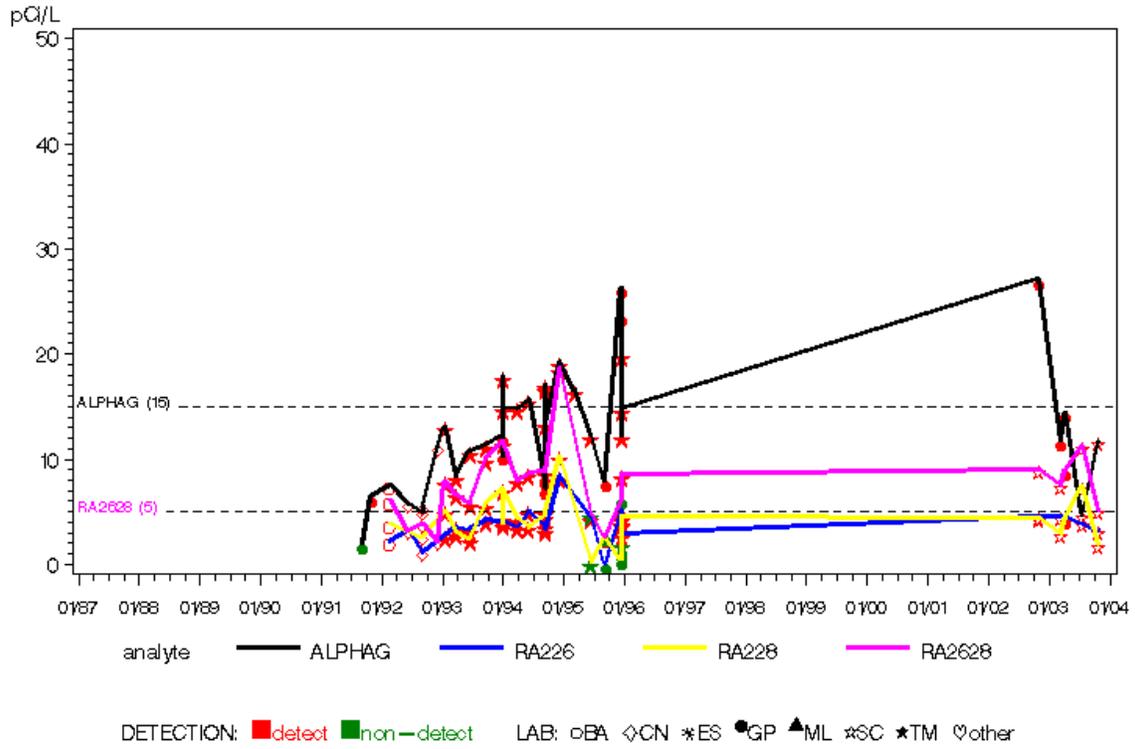
SLF: POC Wells

LFW 48D — RA2628 elev old & ALPHAG not elev



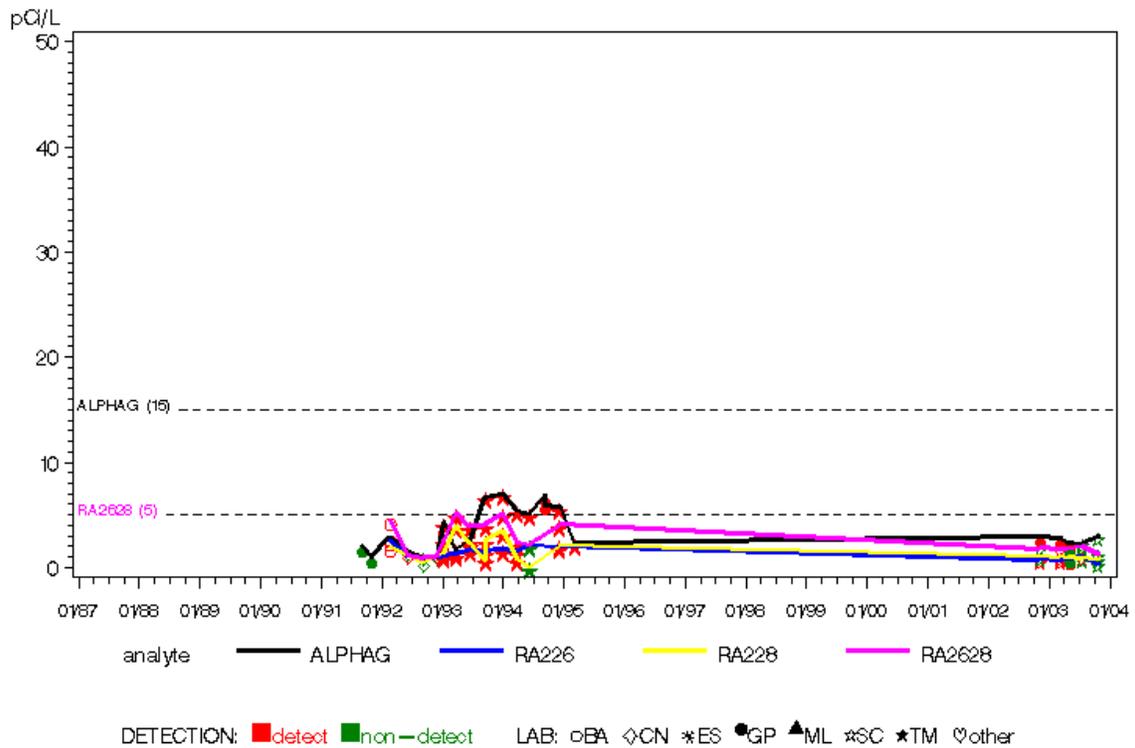
SLF: POC Wells

LFW 57B — RA2628 elev recent & ALPHAG elev old



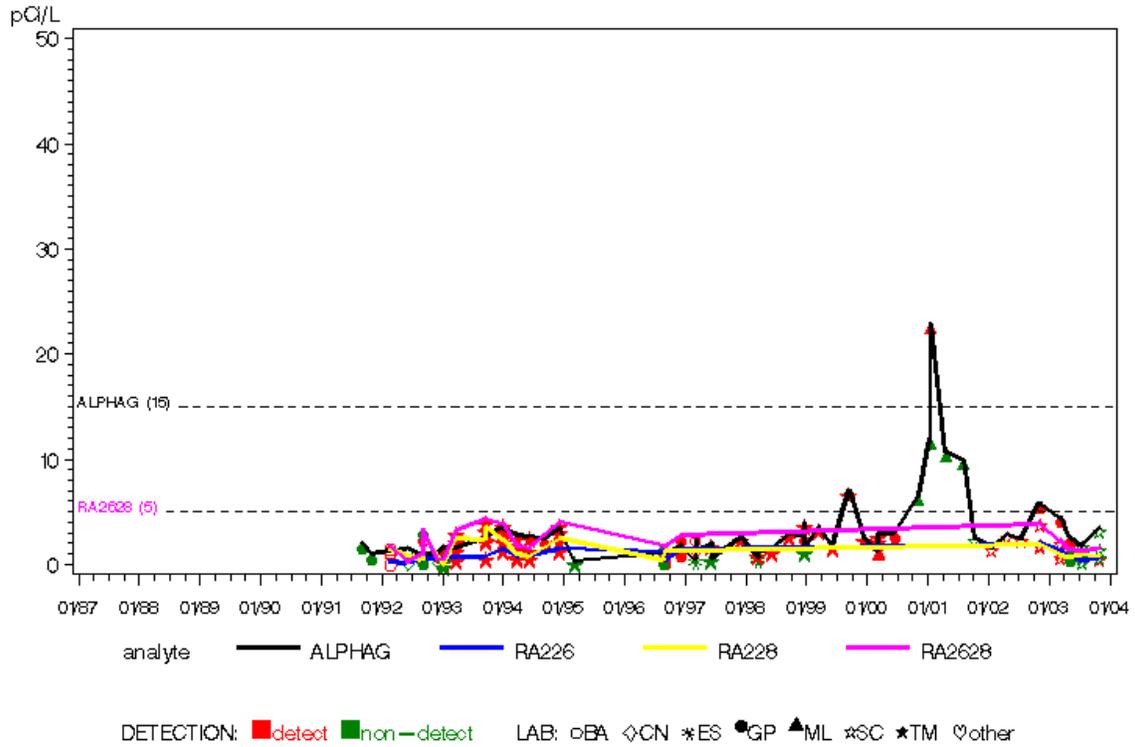
SLF: POC Wells

LFW 59C — RA2628 elev old & ALPHAG not elev



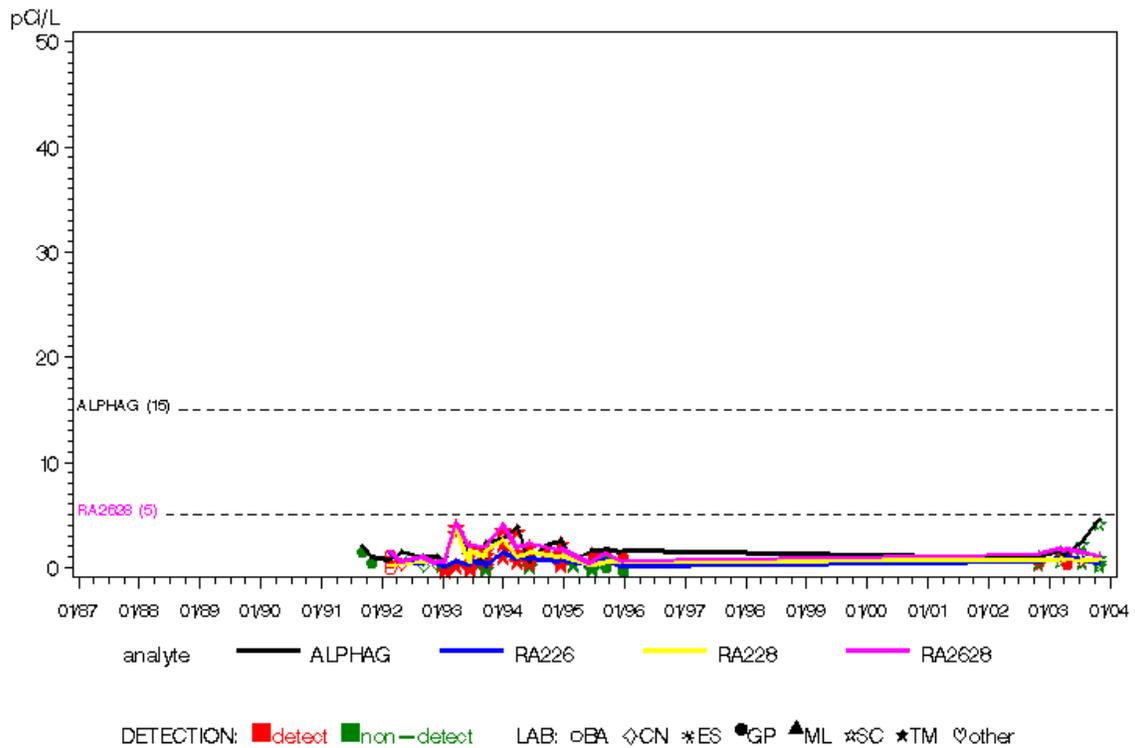
SLF: POC Wells

LFW 59D — RA2628 not elev & ALPHAG elev dd



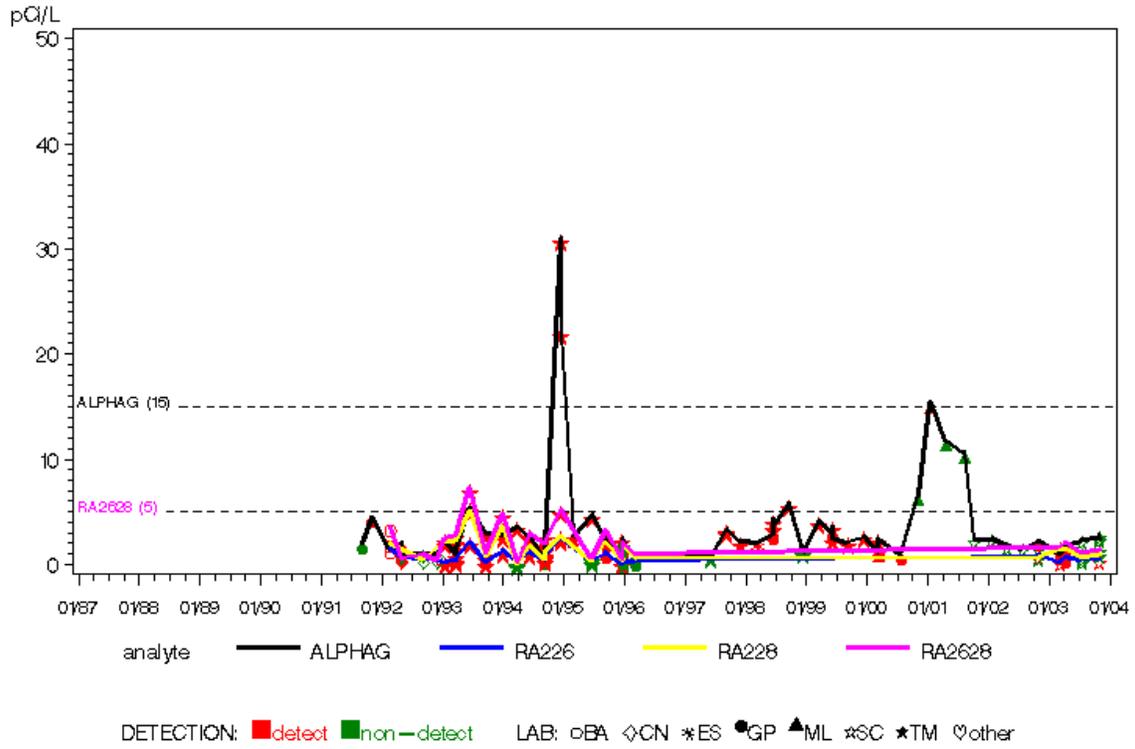
SLF: POC Wells

LFW 61C — RA2628 not elev & ALPHAG not elev



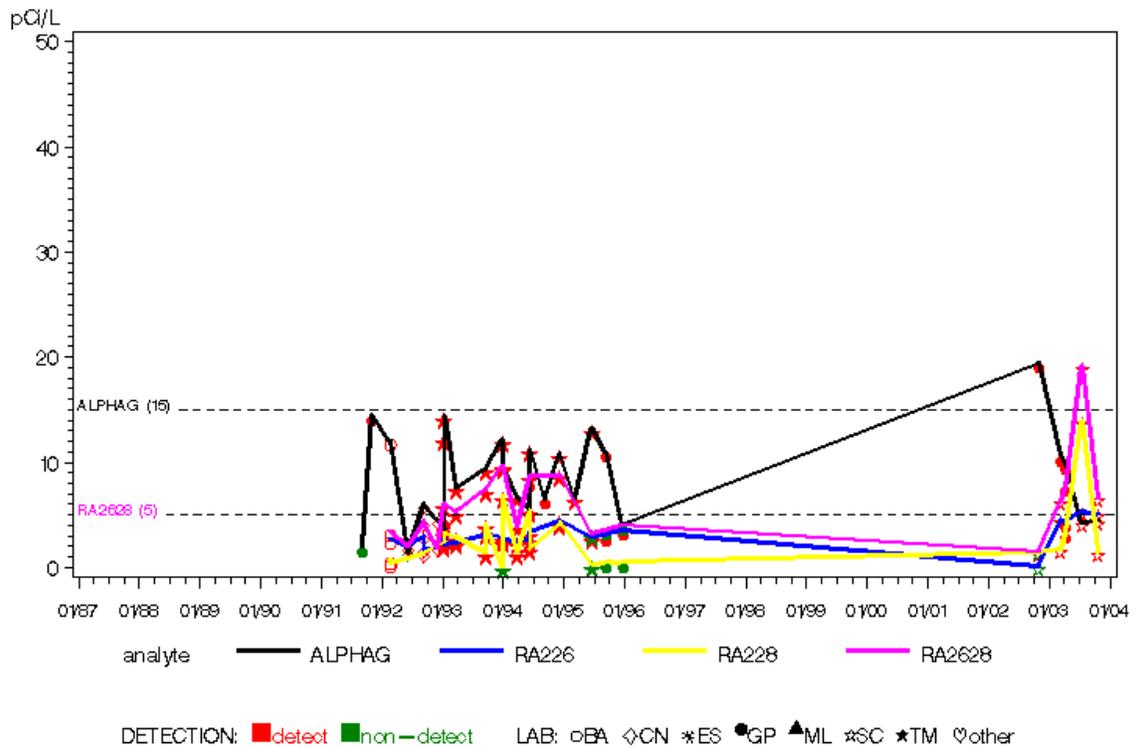
SLF: POC Wells

LFW 61D — RA2628 elev old & ALPHAG elev old



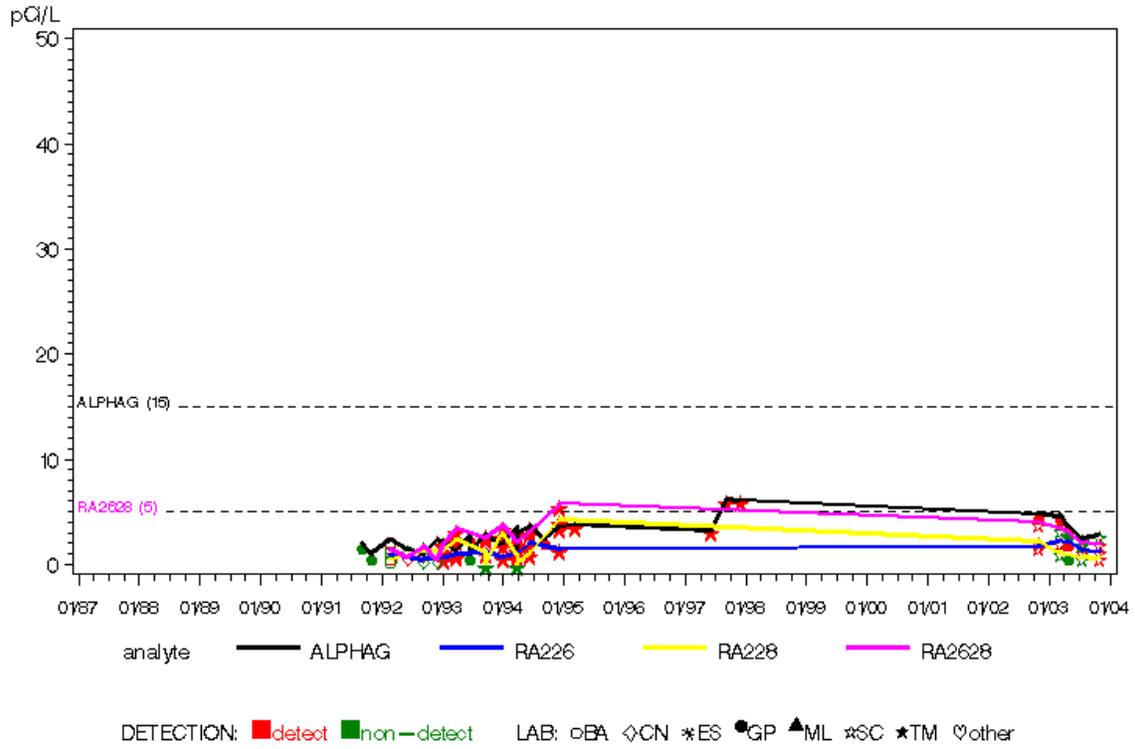
SLF: POC Wells

LFW 62B — RA2628 elev recent & ALPHAG elev old



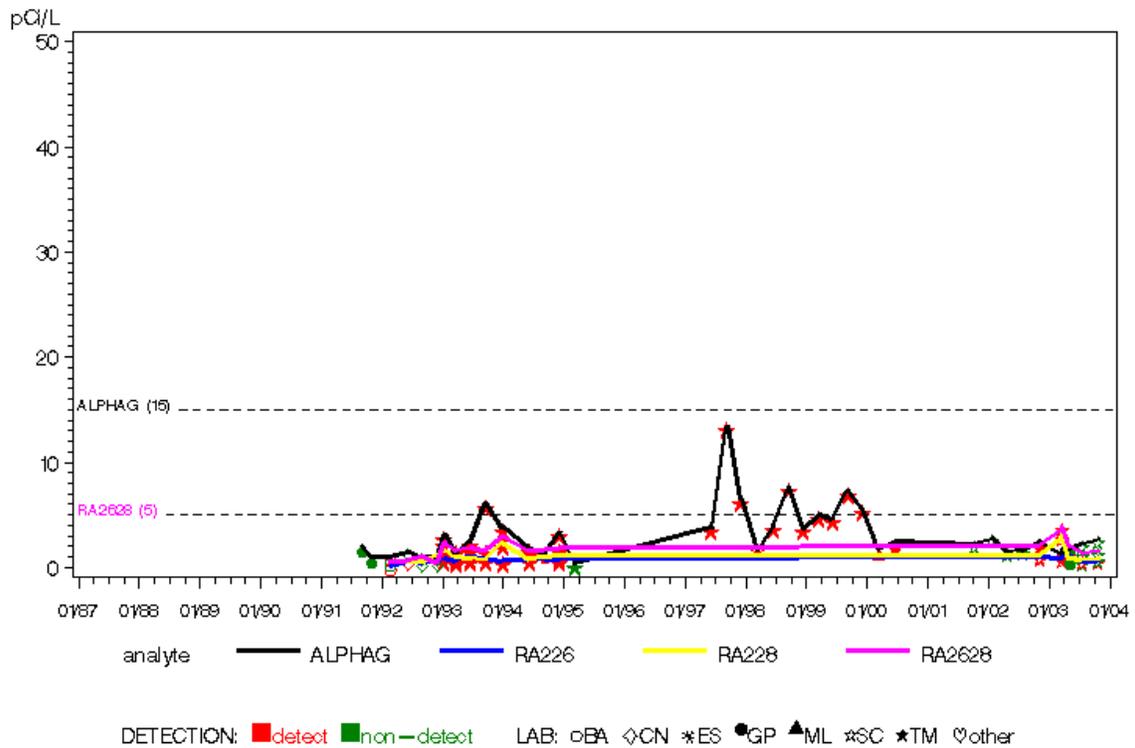
SLF: POC Wells

LFW 62C — RA2628 elev old & ALPHAG not elev



SLF: POC Wells

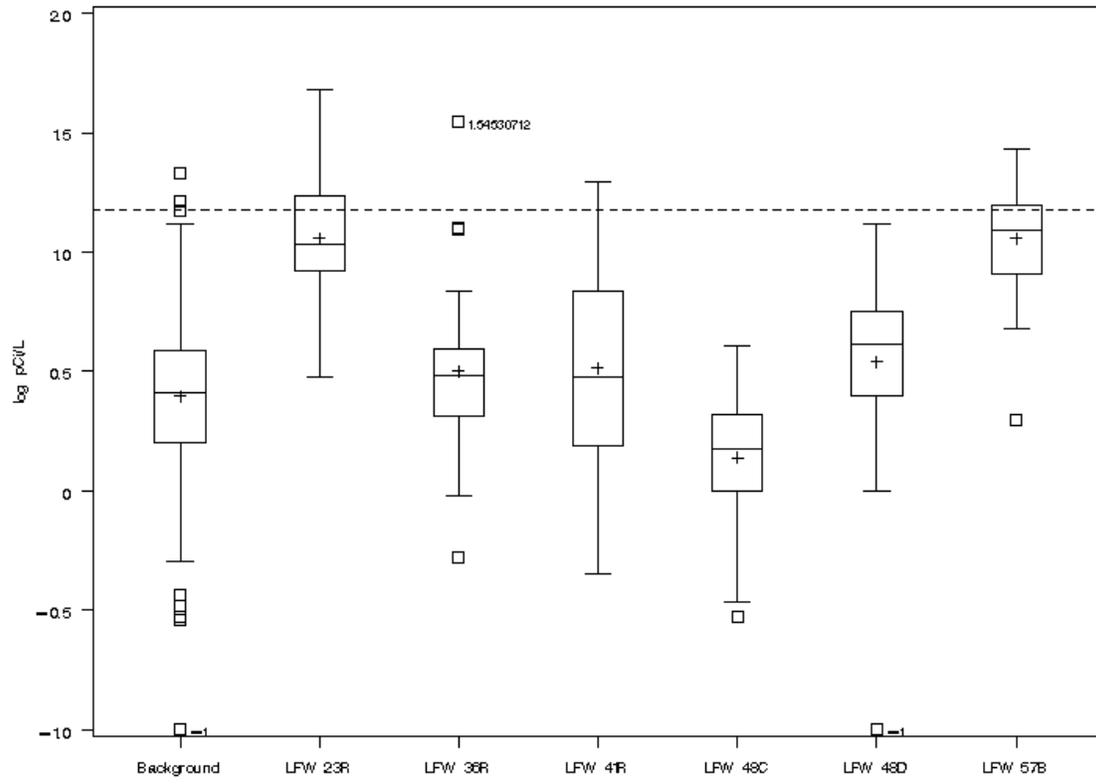
LFW 62D — RA2628 not elev & ALPHAG not elev



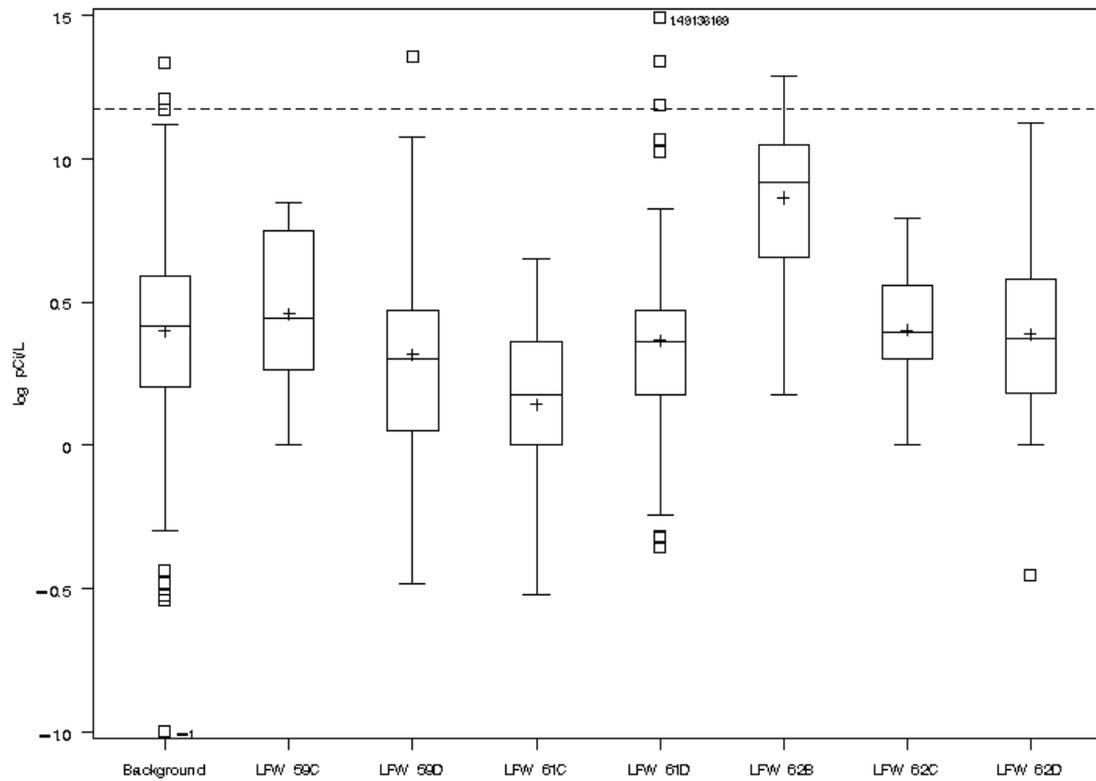
## **Appendix B**

**Box-and-whisker plots for gross alpha (ALPHAG), Ra226, Ra228, Ra2628, Ratio of Ra228 to Ra226, and pH for every RCRA Point of Compliance (POC) monitoring well vs Background wells (combined) in the study**

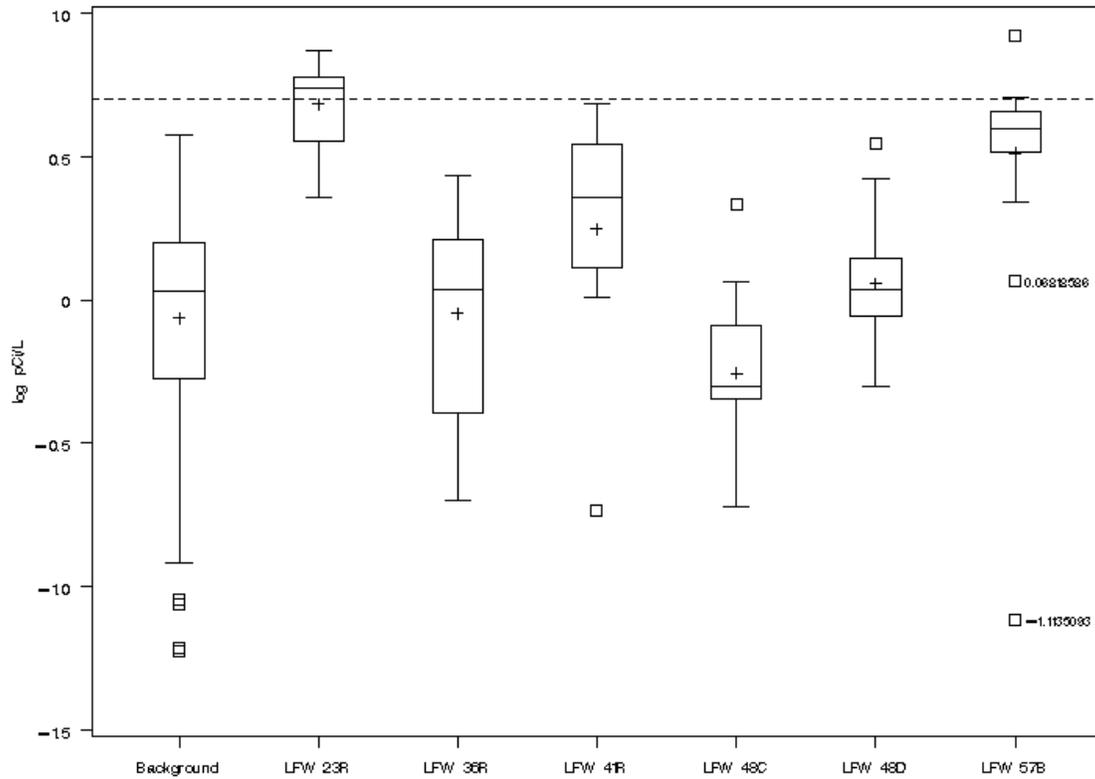
### Gross Alpha in SLF



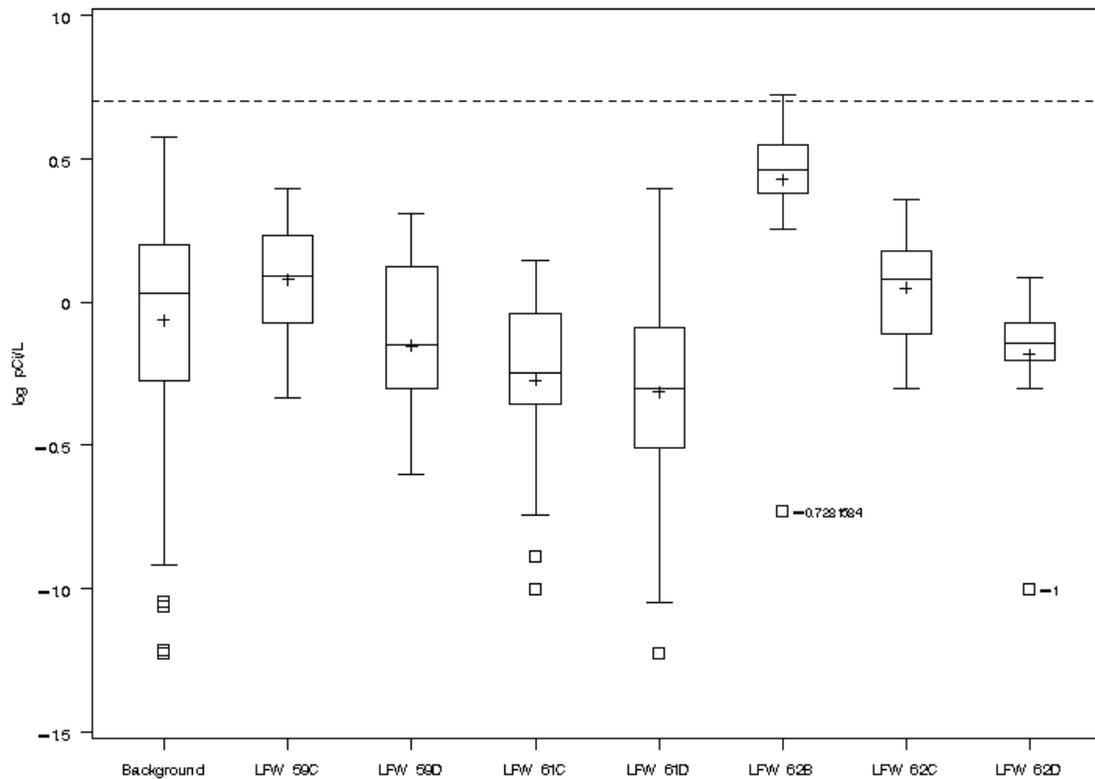
### Gross Alpha in SLF



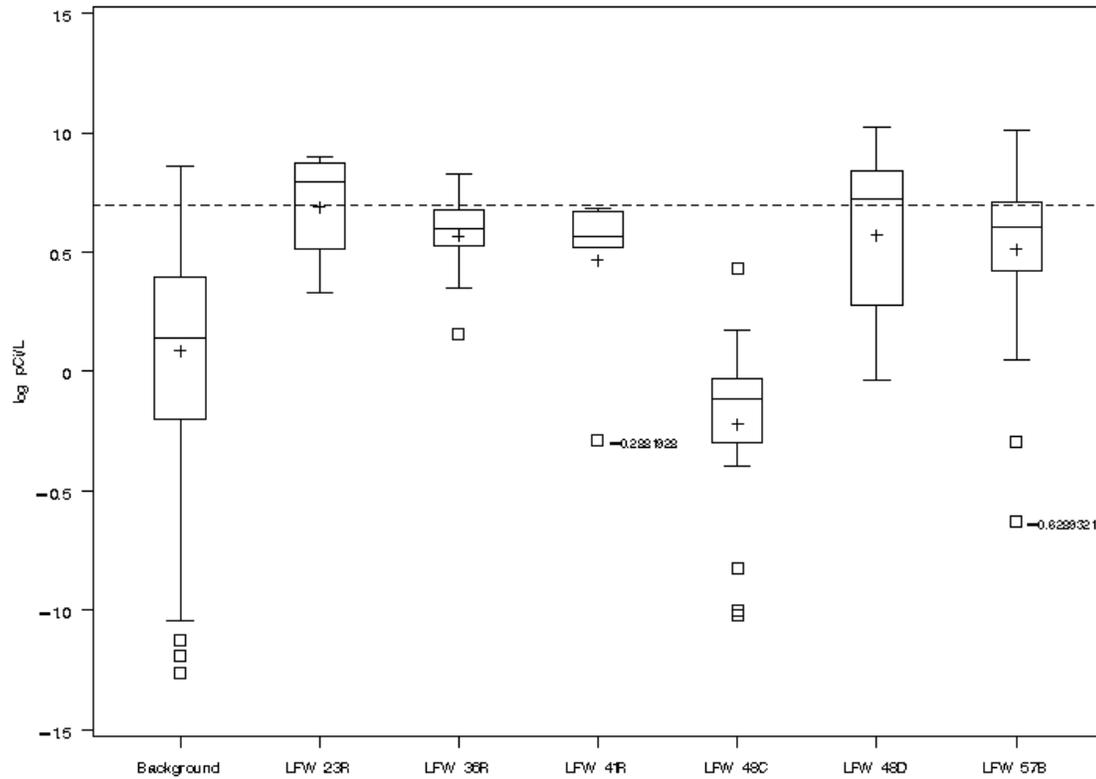
Radium-226 in SLF



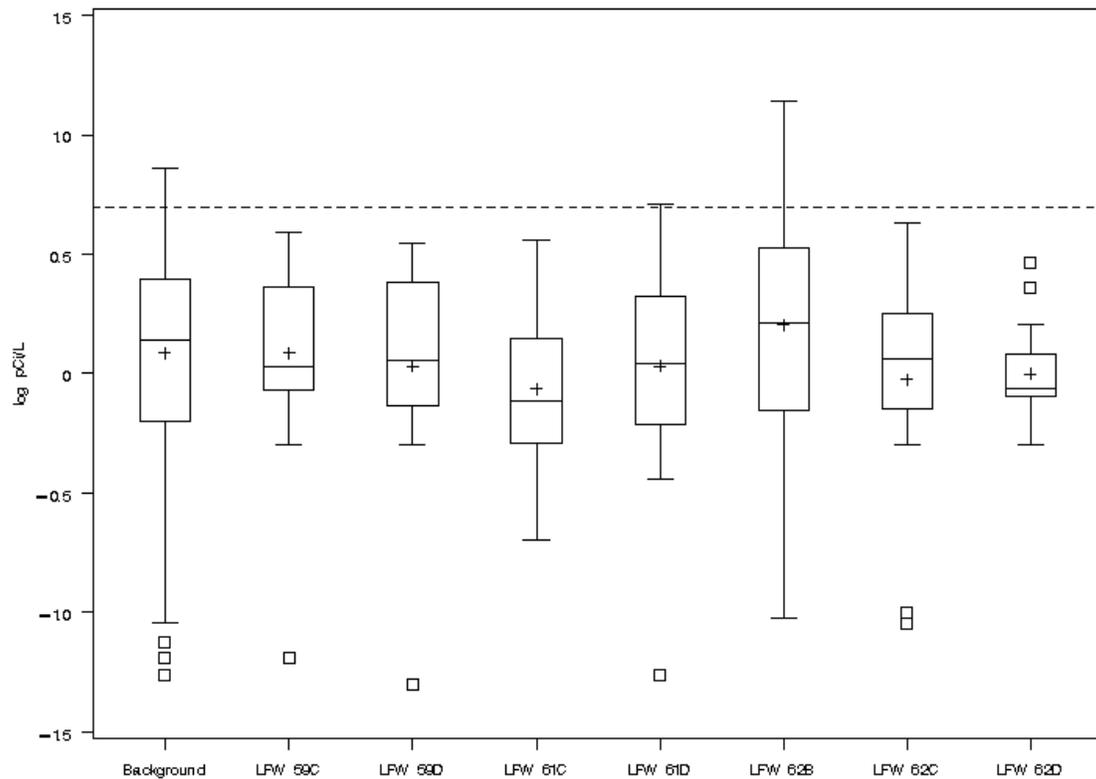
Radium-226 in SLF



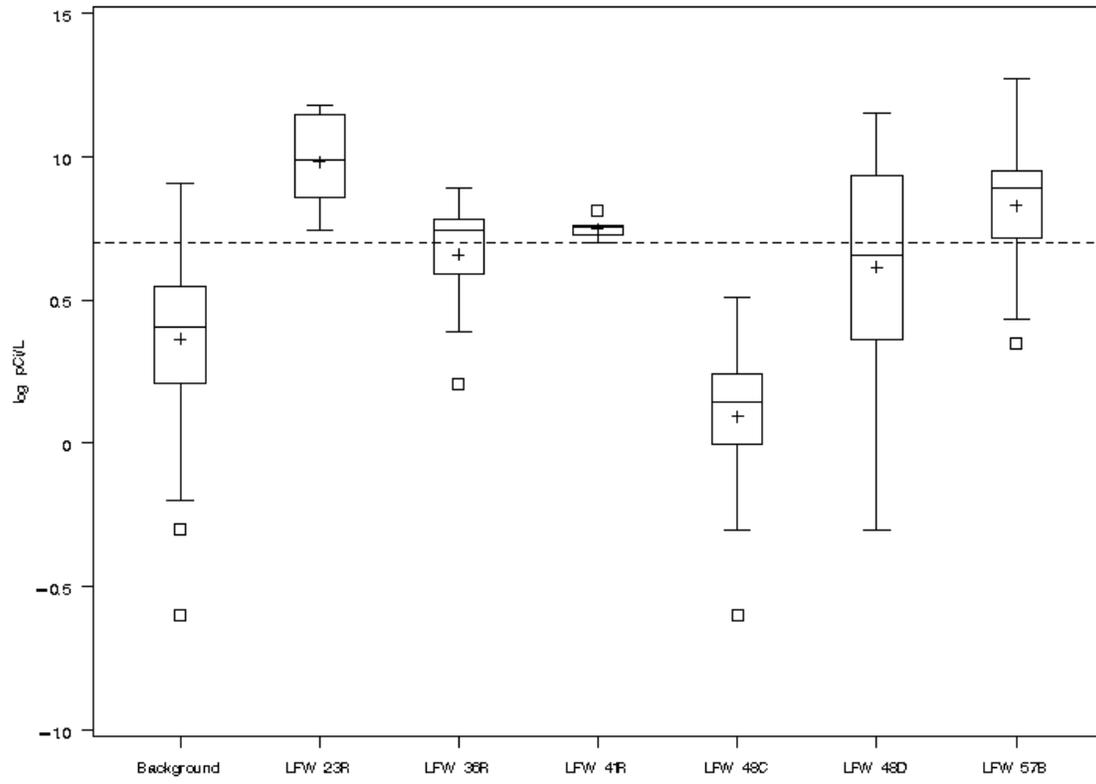
### Radium-228 in SLF



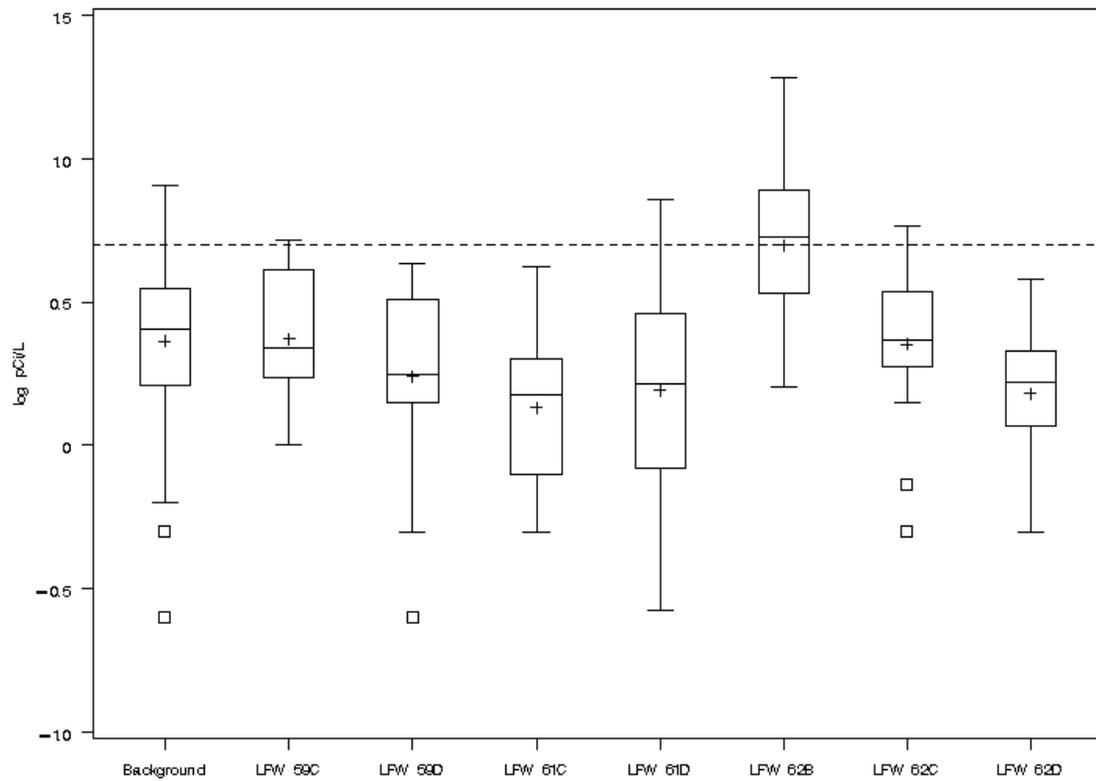
### Radium-228 in SLF



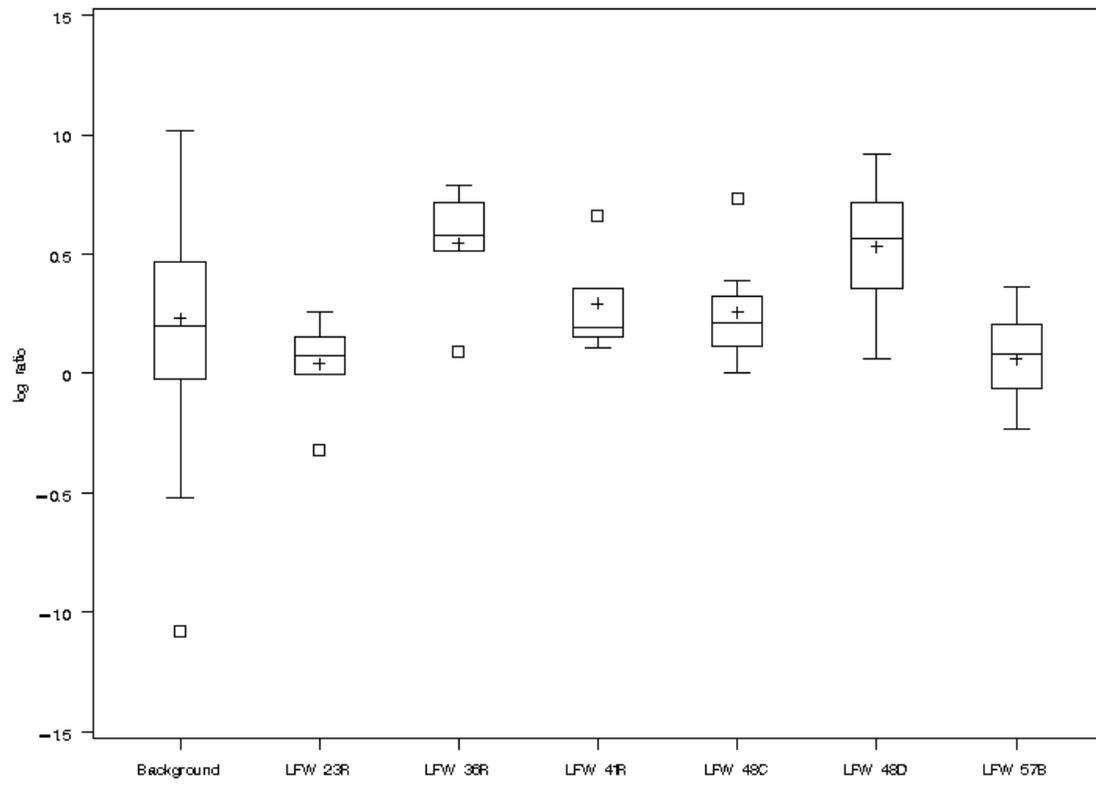
### Radium-226/228 in SLF



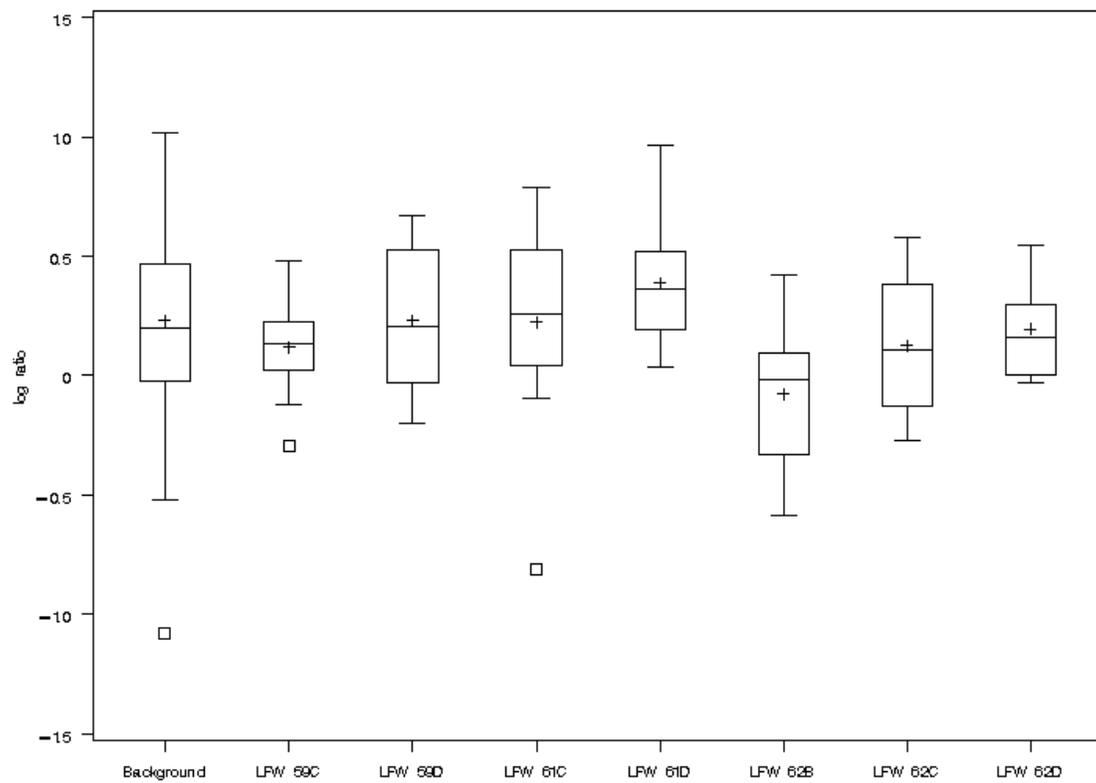
### Radium-226/228 in SLF



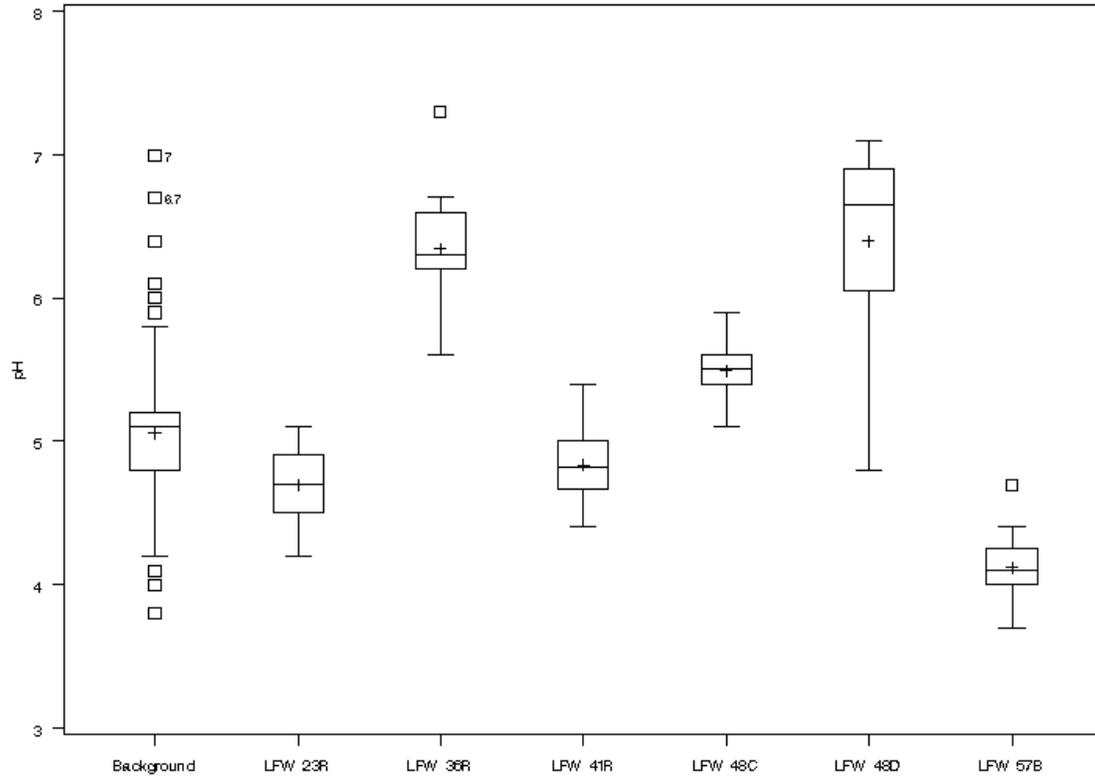
Ratio of Detected Ra228/Ra226 in SLF



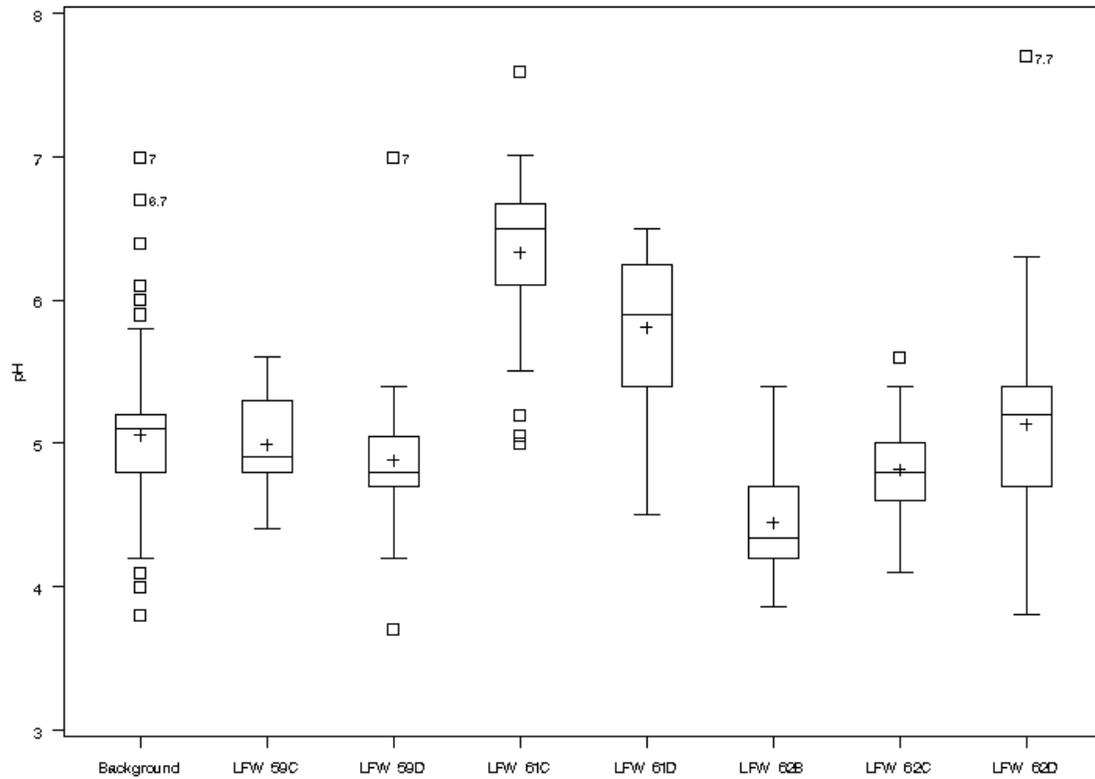
Ratio of Detected Ra228/Ra226 in SLF



pH in SLF



pH in SLF

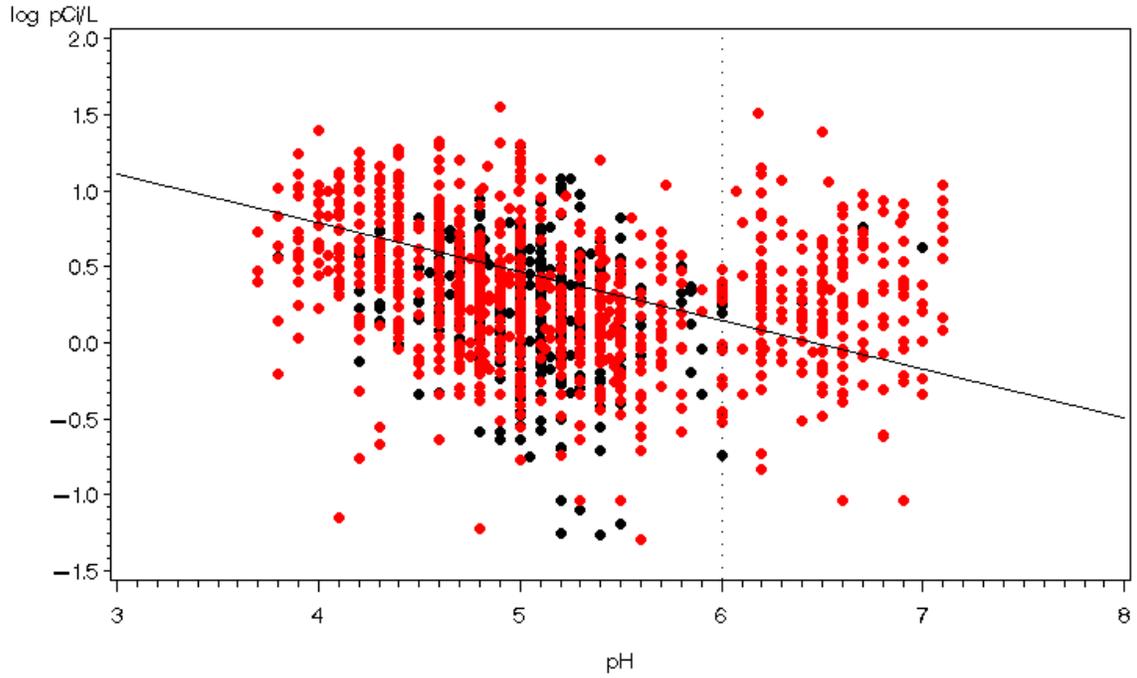


## **Appendix C**

### **Scatter Plots of gross alpha (ALPHAG), Ra226, Ra228, and RA2628 vs ph**

### SLF Background & POC Wells: Gross Alpha versus pH

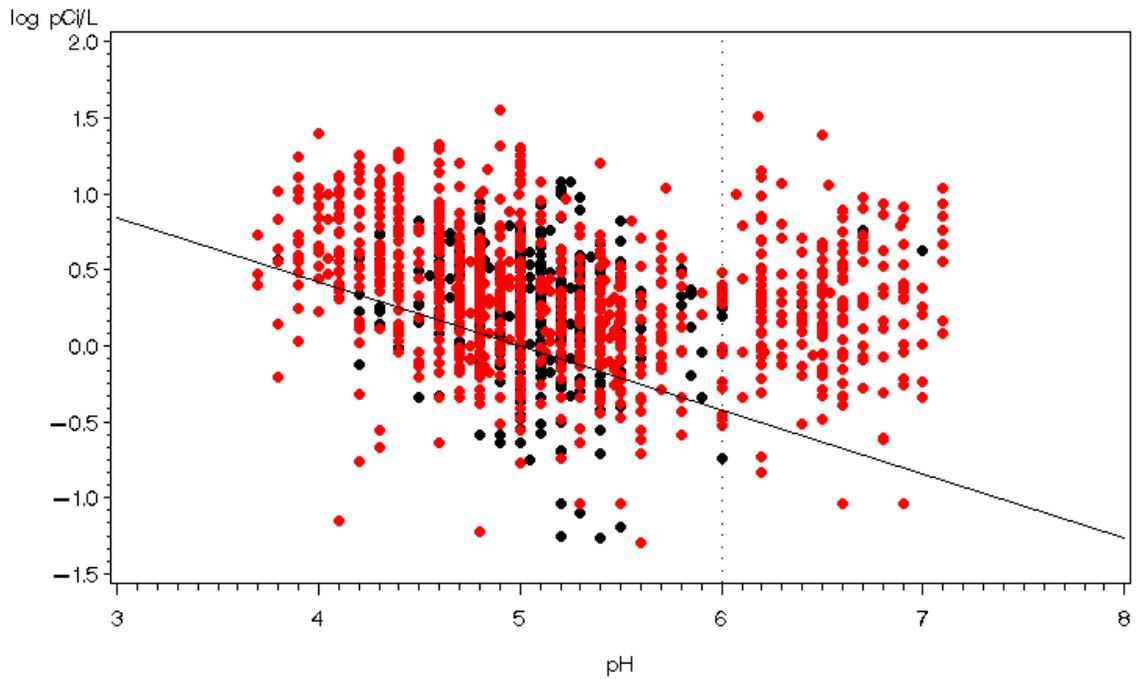
(Note: Fit for pH values of 6 and below only)



Type of Well: ● Background ● POC

### SLF Background & POC Wells: Radium-226 versus pH

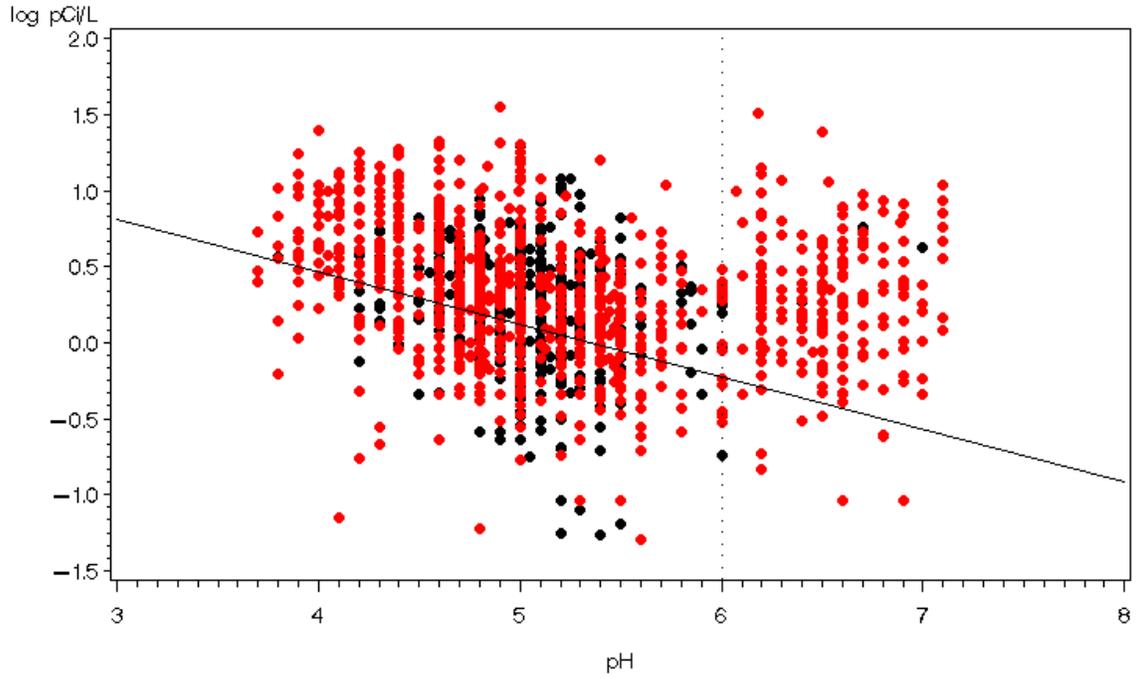
(Note: Fit for pH values of 6 and below only)



Type of Well: ● Background ● POC

### SLF Background & POC Wells: Radium-228 versus pH

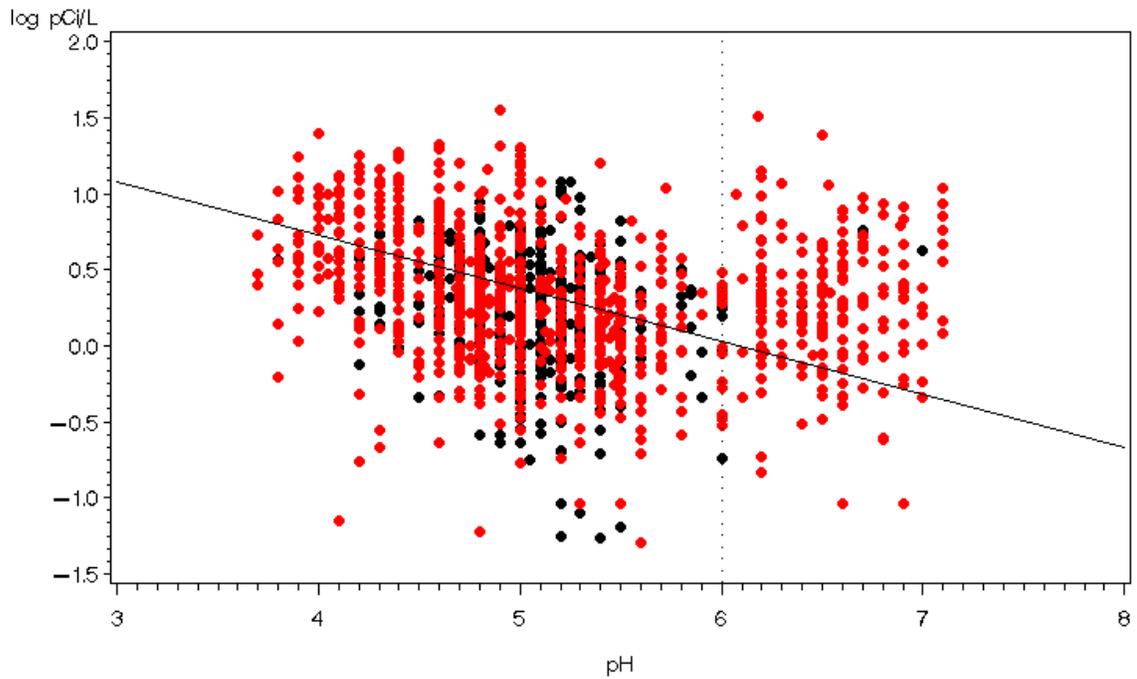
(Note: Fit for pH values of 6 and below only)



Type of Well: ● Background ● POC

### SLF Background & POC Wells: Radium-226/228 versus pH

(Note: Fit for pH values of 6 and below only)



Type of Well: ● Background ● POC