Uncertainty and Trend Analysis for Radium in Groundwater and Drinking Water

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Abstract

Radium activity measurements in water samples have a high uncertainty, especially at values near the combined MCL of 5 pCi/L. This poses problems for determining regulatory compliance and evaluating whether concentrations are increasing or decreasing with time at a given location. This study looks at 409 samples with radium measurements from 139 locations in Escambia County, Florida. Median concentrations range between non-detect and 20.5 pCi/L for total radium. Observed variability – calculated as Relative Percent Difference (RPD = range/mean) for multiple measurements of the same sample or split samples – ranges from 0% to 385% for Ra-226 (130 samples), and from 0% to 183% for Ra-228 (129 samples). The median RPD is 37% for Ra-226 and 42% for Ra-228. The acceptability of this level of variability is evaluated using established criteria.

The observed uncertainty in radium measurements also highlights a general problem of applying trend analysis techniques to data with large error bars. The meaning of error bars depends on how the reported value and the uncertainty are derived. Typically, radium measurements are reported with a 2-sigma uncertainty based on analytical counting error, giving a 95% confidence interval around the reported value. The error bar, then, depicts a normal distribution with the reported value as the mean. Trend analysis is often performed on the reported values without considering the error bar. This procedure ignores the uncertainty in the individual reported values. For radium data that often carry large error bars relative to the reported values, the procedure can result in the reporting of trends that are not truly supported by the data. In this study, Mann-Kendall's test for the presence of a trend is applied to the radium data at 35 locations with a series of at least four radium measurements (249 samples in total). At the 90% confidence level for a two-tailed test, there are 2 locations with positive trends and 8 with negative trends. A trend analysis that accounts for the error bars is then applied to the same data set and the results are compared. This secondary analysis assesses the robustness of the Mann-Kendall result obtained in the first test by recalculating the result using maximum and minimum values alternately along the time series. After applying the secondary test on the data set, 8 of the 10 trends detected by the first test are shown to no longer be significant at the 90% confidence level.

Introduction

The presence of radium in groundwater can be a major concern for the drinking water supply, particularly in areas where the combined radium-226/228 activity in groundwater approaches or encompasses the drinking water Maximum Concentration Level (MCL) of 5 pCi/L. Radium occurs naturally in groundwater but can also result from anthropogenic activities. Dissolved radium can also vary significantly in response to changes in groundwater chemistry (pH, ionic strength, nitrate, etc.). To monitor radium, water samples are periodically analyzed and the data is used to protect and manage the pumping of groundwater for consumption. Changes in radium activity can be determinant for the development or abandonment of supply wells and for the overall management of a water supply system. It is therefore important to understand the variation of radium activity over time, whether as trends or fluctuations. The main problem with radium data is that analytical results are impaired with relatively large error bars relative to the regulatory MCL. For water supply wells that have radium activities close to the MCL, the interpretation of the data has serious regulatory implications that can result in major changes in the operation of a water supply system.

This paper examines the interpretation of publicly available radium data for groundwater and drinking water samples in Escambia County, Florida. Most of the drinking water supply in Escambia County is derived from the pumping of the Sand-and-Gravel aquifer. Radium activity in the aquifer is naturally close to the MCL with exceedances reported in several wells and locations in the water supply system. The data set is derived from 409 water samples collected from 139 locations that include municipal supply wells, distribution points, irrigation wells and monitoring wells in the main production zone of the Sand-and-Gravel aquifer. Samples were collected between 1992 and 2003. The data consist of 592 analyses for Ra-226 and 573 analyses for Ra-228. Three additional results were reported only as total radium. Median total radium values by sample location range between non-detect and 20.5 pCi/L with an overall median of 3.0 pCi/L. The radium analyses were performed at 8 different laboratories. Ra-226 was analyzed by EPA Methods 900.0 and 903.1; and Ra-228 by EPA methods 904 and RA-05.

The data set is adequate for statistical interpretation and is representative of many real-life data sets that are relied upon by regulators or water supply managers. The long period of time encompassed by the data set and the fact that different laboratories and analytical and sampling methods were used introduce additional uncertainty that is not accounted for in this paper. However, the paper focuses on the comparison of two levels of approach to interpret radium activity trends and the results of the comparison are not directly affected by that additional uncertainty. Figure 1 is a representation of Ra-226 and Ra-228 activity values in the data set for 570 pairs of values at 139 locations. The figure illustrates the spread of the data and the fact that a significant portion of the radium results (27%) are above the MCL. Figure 2 shows time series at three locations from the data set. The three time series illustrate the type of information that is typically available and relied upon by regulators and water supply managers.

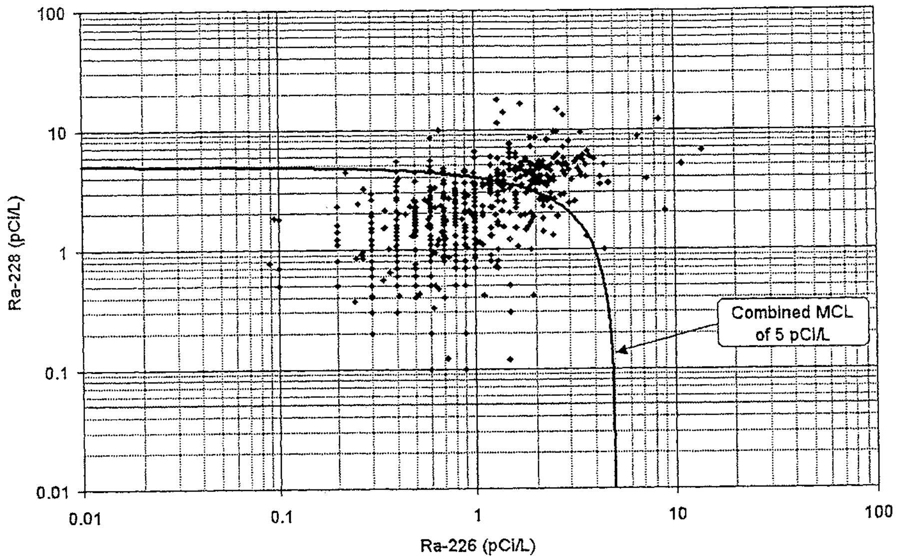
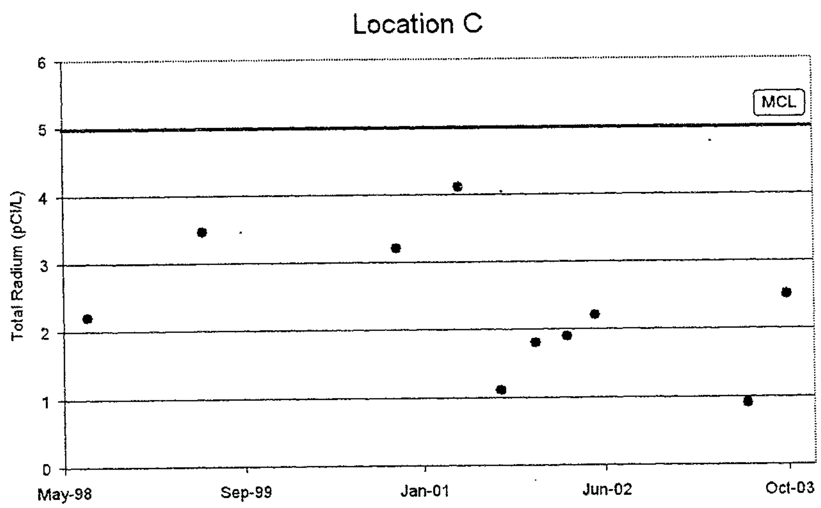


Figure 1: Ra-228 vs Ra-226 for 570 pairs of measurements. Non-detects are set as ½ of the detection limit.



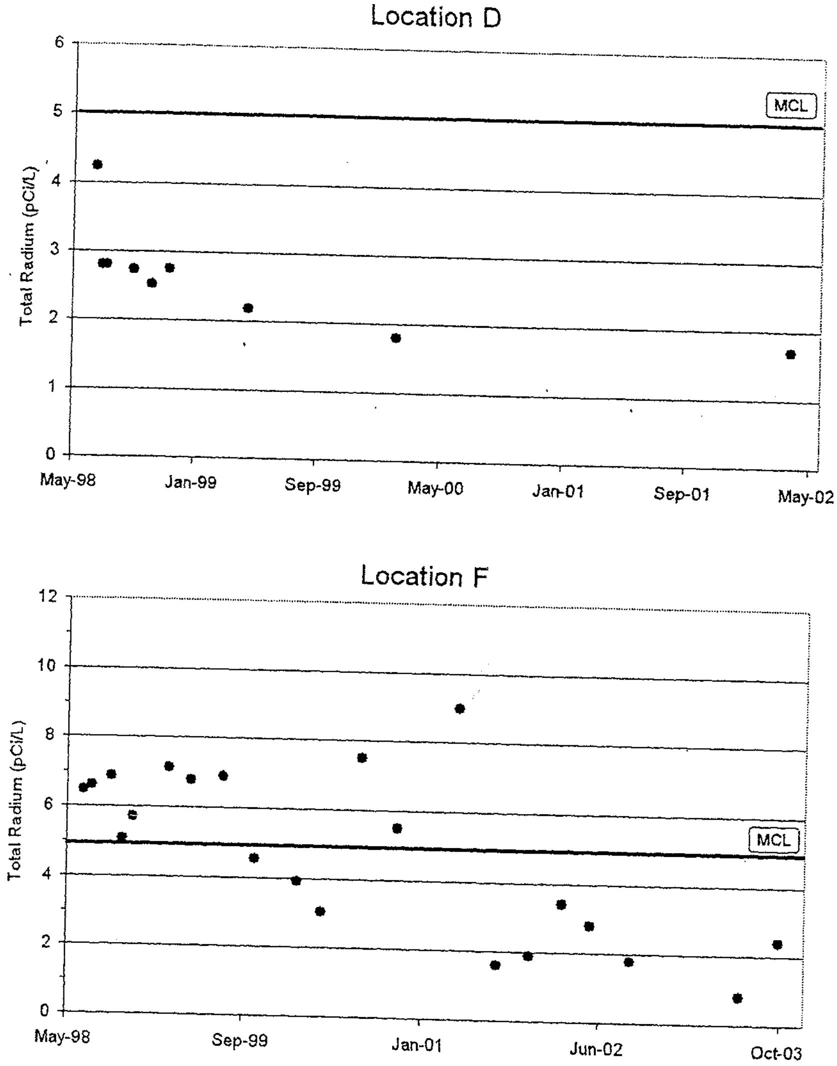


Figure 2: Total radium time series examples.

Uncertainty in Radium Measurements

The laboratory reports an analytical error for each radium measurement. This error quantifies the confidence in the reported result and is based on an estimate of the random error in counting radioactive disintegrations with the calibrated analytical equipment (U.S. EPA, 1980). The total analytical uncertainty in the reported analytical results can also be estimated by examining the observed variability in the results of duplicate (split and co-located samples) and replicate (re-analysis of the same sample) analyses. The observed variability is generally used to check the precision of a set of measurements against quality control limits. For this study we use observed variability for duplicates and replicates to characterize the precision of the data by comparing it to various criteria as used by the EPA and national laboratories (Brookhaven and Lawrence Livermore National Laboratories).

Observed variability can also be compared to the reported uncertainty to see whether the reported uncertainty explains the variability seen in the data. In this paper we use the laboratory analytical uncertainty as the error bar on individual values for the trend analyses.

Trend Analysis

To test for the presence of trends in data time series at individual locations we used the Mann-Kendall test. This test is commonly relied upon in the interpretation of environmental data (Maidment, 1993; Helsel & Hirsch, 2002). For example, the test is often applied on data sets from monitoring wells to assess the stability of a groundwater contaminant plume. The Mann-Kendall test is an important tool that is relied upon in the decision making process in matters that relate to water quality (Lee et al., 2003; Aziz et al., 2003). The Mann-Kendall test is non-parametric and therefore does not assume an underlying distribution in the data set. It can address missing data values and be modified to account for seasonality or predictable fluctuations. In the application of the test, the data is first ranked according to an independent variable (i.e. date for time series data), and a statistical parameter (S) is calculated by comparing each data point to the data points that occur after it in the ranked series (Gilbert, 1987). The S parameter in the Mann-Kendall test is calculated as:

$$S = \sum_{k=1}^{n-1} \sum_{j=k+1}^{n} \operatorname{sgn}(x_j - x_k) \qquad \text{Equation 1}$$

where,
n is the number of data points,
x is a value in the ranked series, and

$$\operatorname{sgn}(x_j - x_k) = \begin{cases} 1 & \text{If } x_j - x_k > 0 \\ 0 & \text{If } x_j - x_k = 0 \\ -1 & \text{If } x_j - x_k < 0 \end{cases}$$

S and n are then used to read a p-value from a statistics table (e.g. Table A18 in Gilbert, 1987). The sign of S indicates the direction of the trend (i.e. positive S indicates an upward trend) and the p-value is a measure of the significance of the trend. The p-value represents the probability that the data set could have been arranged randomly rather than as a statistical trend. The p-values that are usually tabulated for the Mann-Kendall test are for a one-tailed test, in which the null hypothesis is no trend and the alternative hypothesis must be specified as either an upward trend or a downward trend. A two-tailed test is used to test for both upward and downward trends in a single alternative hypothesis. The one-tailed p-values must be doubled for a two-tailed test (Gilbert, 1987; Gibbons and Coleman, 2001).

For large data sets ($n > 10$) S can be modified to be approximated by a normal distribution. To do this a test statistic Z_S is calculated (Equation 2) and a p-value is generated from a table of the normal distribution (Helsel and Hirsch, 2002). A correction for tied values is included in the modification of S. Tied values are data points with the same value for the independent or dependent variable, that therefore contribute 0 to S.

$$Z_S = \begin{cases} \dfrac{S-1}{\sigma_S} & \text{If } S>0 \\ 0 & \text{If } S=0 \\ \dfrac{S+1}{\sigma_S} & \text{If } S<0 \end{cases} \qquad \text{Equation 2}$$

where,

$$\sigma_S = \sqrt{\frac{\left[n(n-1)(2n+5) - \sum_{i=1}^{n} t_i(i)(i-1)(2i+5)\right]}{18}},$$

t is the number of tied values, and
i is the "extent" of the tie, or the number of data points with the same value.

For radium data the Mann-Kendall test is used to assess the presence of a statistical trend on the reported activity values of time series at individual locations without considering the error bar on the values (analytical uncertainty). To test the significance of the analytical uncertainty on the statistical trends obtained applying the Mann-Kendall test, we are proposing a secondary test that is applied as an option to the Mann-Kendall test. This proposed secondary test takes into consideration the analytical error bars on the radium values to evaluate trends. The proposed test is referred to as the "Alternating Min/Max Test" in this paper for convenience. The approach is further described below.

Applied Methodology

For the uncertainty analysis, all samples with multiple measurements of either Ra-226 or Ra-228 are considered. Ra-226 and Ra-228 are examined independently, as they are analyzed by different analytical methods resulting in different analytical uncertainty. The trend analysis is performed on time series at individual locations for total radium values created by adding Ra-226 and Ra-228 activities. This procedure takes into account the fact that the number of radium measurements varies from location to location.

There are 80 non-detect radium values in the data set. These are attributed an arbitrary activity value that corresponds to half the detection limit. The mean reported detection limit is applied to non-detects without available detection limit information (9 analyses). It is recognized that other methods of dealing with censored data are available (Helsel, 2004), and this issue is not addressed in this paper.

Reported error was recorded in the data set for 36 pairs of Ra-226 and Ra-228 analyses. The uncertainty for total radium is calculated as the sum of the uncertainties for Ra-226 and Ra-228(Taylor, 1982). The propagated uncertainties show a correlation with total radium concentration as illustrated in Figure 3. Typically for radium data, a 2-sigma error is reported by the laboratory (i.e. reported value +/- 2-sigma). Care must be taken to ensure consistency in the level of error when comparing data from multiple laboratories, as a 1-

sigma error is sometimes reported. As illustrated in Figure 3, the error bar data as a function of total radium activity values fits a logarithmic function:

Relative Error = -0.1920 ln(Total Radium) + 0.5822 Equation 3

This relationship was used to calculate a total radium error for measurements where no reported error was recorded.

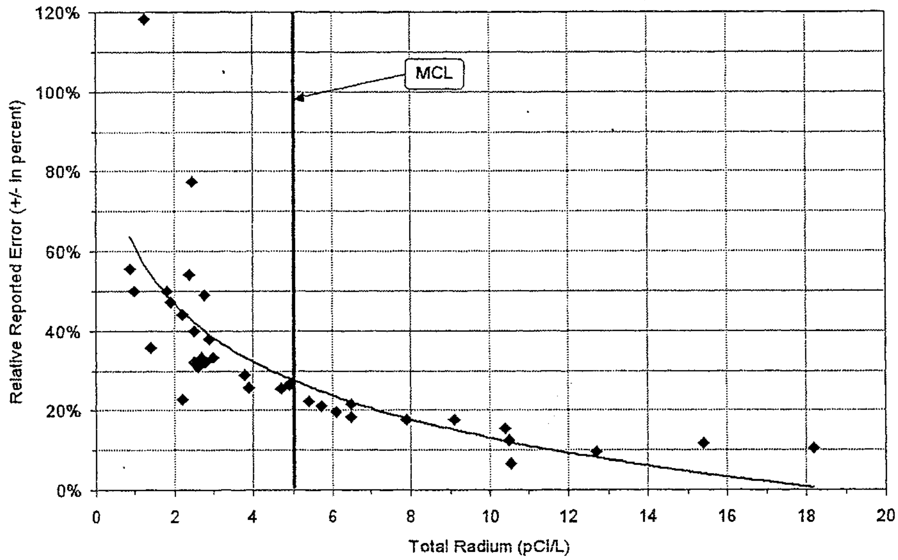


Figure 3: Relative reported error (error divided by reported value) vs. Total Radium

Assessment of Uncertainty in the Analytical Data

To compare the observed variability among the samples, we used two approaches. First, the Relative Percent Difference (RPD) is calculated for each sample by dividing the range in results by the mean result for that sample. This variability is expressed as a percentage (U.S. EPA, 1994). For example, in terms of radium activity, an RPD of 100 percent on the MCL activity value of 5 pCi/L corresponds to a variability of +/- 2.5 pCi/L. Second, the relative standard deviation (RSD) is calculated (Garcia and MacQueen, 1999) to allow comparison with the range of variability for parameters other than radium in the data set (Equation 4).

$$RSD = \frac{200}{\sqrt{2}} \times \frac{|x_1 - x_2|}{x_1 + x_2}$$ Equation 4

For samples with multiple measurements (duplicates, triplicates, replicates, etc.), x_1 and x_2 are equated to the minimum and maximum values for a given location at a given date. This is done to avoid including a measurement in the analysis more than once, as well as to avoid giving a heavier weighting to samples with more than two measurements. For Example, a sample with triplicate measurements could have three pairs of measurements for RSD calculation, as opposed to 1 pair for a sample with duplicate measurements. By applying this methodology, there is only one RPD and one RSD for each sample, even if a sample was measured multiple times.

Trend Analysis

Trend analysis is performed for all locations with at least 4 data points. Prior to the analysis the median is taken where multiple values exist for a single day. The Mann-Kendall test is applied to the reported values for each location. Then a secondary test is applied, which assesses the robustness of the Mann-Kendall result by including the analytical error bars. This test involves creating two new time series for each location by alternately adding or subtracting the error bar from the reported values, creating a maximum or minimum value – one of the two new data sets starts with the minimum value and the other starts with the maximum value at the earliest data point in the time series. We will refer to this test as the "Alternating Min/Max Test", for convenience. The time series of reported values will be called the "Reported Value Scenario." The calculated new data sets will be termed "Alternating Scenario 1" (starting with the minimum value) and "Alternating Scenario 2" (starting with the maximum value). The Mann-Kendall test is applied to the alternating scenarios generating a total of three p-values for each location. The maximum p-value is taken to represent the "worst-case scenario" for trend evaluation at a given location. We use Equation 1 to generated S parameters, and Equation 2 to generate the two-tailed p-values for each scenario. For locations with 10 or fewer data points the calculated p-value is checked against the tabulated p-values for the corresponding S and n.

Results and Discussion

Observed variability

Of the 409 samples collected for radium analysis in the Escambia County data set, 130 had multiple analyses for Ra-226, and 129 had multiple analyses for Ra-228. The median RPD is 37 percent for Ra-226 and 42 percent for Ra-228 (Table 1a). There are 9 samples that were analyzed multiple times by a single laboratory for Ra-226 and 7 for Ra-228. The median RPD is 36 percent for Ra-226 and 50 percent for Ra-228 in the intra-laboratory samples (Table 1b).

Table 1a: Observed variability for all samples with multiple analyses

	Ra-226	Ra-228
Number of Samples	130	129
Number of Analyses	316	315
Minimum RPD	0%	0%
Maximum RPD	385%	183%
Median RPD	37%	42%
90th Percentile RSD	76%	80%

Table 1b: Observed variability for samples with multiple analyses by the same lab

	Ra-226	Ra-228
Number of Samples	9	7
Number of Analyses	18	15
Minimum RPD	14%	15%
Maximum RPD	162%	118%
Median RPD	36%	50%

The observed variability in the radium data here is large relative to other parameters or constituents that can be measured with more accuracy and precision. For comparison, in the data set the median observed variability for chloride measurements taken over the same time period (1992-2003) is 2 percent. This is more than an order of magnitude lower than that seen for radium measurements. This highlights the difficulty of measuring low radium activities and the importance of accounting for uncertainty in interpreting radium data.

For comparison with other published data we considered two studies. With the first study, we considered the EPA criteria for evaluating duplicate analyses using the RPD approach (U.S. EPA, 1994, as discussed above). This approach results in 28 percent of the Ra-226 duplicate/split data pairs to fail for the Escambia County data set. This high rate of failure compares to a 3 percent failure rate for a radionuclide data set consisting of 288 duplicate pairs of analyses of groundwater samples collected from the Brookhaven National Laboratory site in 2001 (Brookhaven National Laboratory, 2002). Second, we considered the data set evaluated by Lawrence Livermore National Laboratories, which comprises data collected in 1999 from the "Livermore site" and "Site 300" (Garcia and MacQueen, 1999). That study used a different validation procedure and characterized a set of duplicates measured for various analytes, including selected radionuclides, as having "generally good agreement" since 90% of duplicate sets had a better than 27 percent RSD. The corresponding value for the Escambia County data set is 76 percent RSD for Ra-226 and 80 percent RSD for Ra-228 (Table 1a). The comparisons indicate the Escambia County data set has a high degree of variability in samples that were analyzed more than once. This high variability is partly explained by the analytical uncertainty of the data set and can be taken into account with the proposed Alternating Min/Max Test.

Trend Analysis

For the trend analysis, the Mann-Kendall's is applied to the total radium data at 35 locations that have at least four measurements (249 samples in total). At the 90% confidence level, there are 2 locations with positive trends and 8 with negative trends for the Reported Value Scenario (Table 2). There are 3 significant trends for Alternating Scenario 1 and 4 significant trends for Alternating Scenario 2. Considering the largest p-value of the three statistical data sets as the worst-case scenario, 8 of the 10 trends from the Mann-Kendall test are no longer significant at the 90% confidence level when the Aternating Min/Max Test is applied (there is also one time series, Location C, which exhibits a significant trend in one of the two alternating scenarios while no trend exists for the reported values under the Mann-Kendall test).

The p-values in Table 2 were calculated from Equation 2. For the 7 locations with 10 or fewer data points, the calculated p-values were checked against the tabulated values (Helsel and Hirsch, 2002). The two sets of p-values show a very good agreement, with differences seen only in the third decimal place for 6 of the 7 locations, and the second decimal place in one location. The differences do not affect any determinations of significance of the trend.

Table 2: Results of the Mann-Kendall tests on the Reported Value and Alternating Scenarios.

Location	n	Reported Value Scenario									Alternating 1		Alternating 2	
		Min	Max	Mean	Median	SD	CV	E	S	p	S	p	S	p
A	5	10.4	18.2	13.5	12.7	3.3	0.2	1.6	8	**0.086**	6	0.221	6	0.221
B	5	1.5	10.5	7.1	7.9	3.4	0.5	1.4	8	**0.086**	4	0.462	8	**0.086**
C	10	0.9	4.1	2.3	2.2	1.0	0.4	0.9	-10	0.419	-3	0.858	-23	**0.049**
D	9	1.7	4.2	2.6	2.7	0.7	0.3	0.9	-31	**0.002**	-14	0.175	-10	0.348
E	4	1.1	1.9	1.6	1.7	0.4	0.2	0.9	-6	**0.089**	0	1.000	-4	0.308
F	21	0.8	9.0	4.8	5.1	2.3	0.5	1.2	-94	**0.005**	-92	**0.006**	-94	**0.005**
G	6	1.5	4.5	3.0	2.8	1.0	0.4	1.0	-11	**0.060**	-5	0.452	-7	0.260
H	8	1.6	5.1	3.3	3.3	1.1	0.3	1.1	-16	**0.063**	-14	0.108	-14	0.108
I	13	1.1	6.1	2.9	2.6	1.5	0.5	1.0	-29	**0.087**	-28	**0.100**	-24	0.161
J	11	0.7	4.9	1.8	1.4	1.2	0.7	0.7	-23	**0.083**	-15	0.276	-17	0.213
K	13	1.0	7.5	3.1	2.1	2.0	0.6	0.9	-29	**0.087**	-41	**0.014**	-29	**0.087**

n – Number of data points
SD - Standard deviation
CV - Coefficient of variation (standard deviation divided by the mean)
E - Median error (+/-) used for the time series
S - Kendall's S statistic (sign indicates the direction of the trend)
p - Two-tailed p-value. Bold values are significant at the 90% confidence level
Alternating 1 and 2 - Scenarios created as described in the text starting with the minimum (1) or maximum (2) value of the first data point in the series

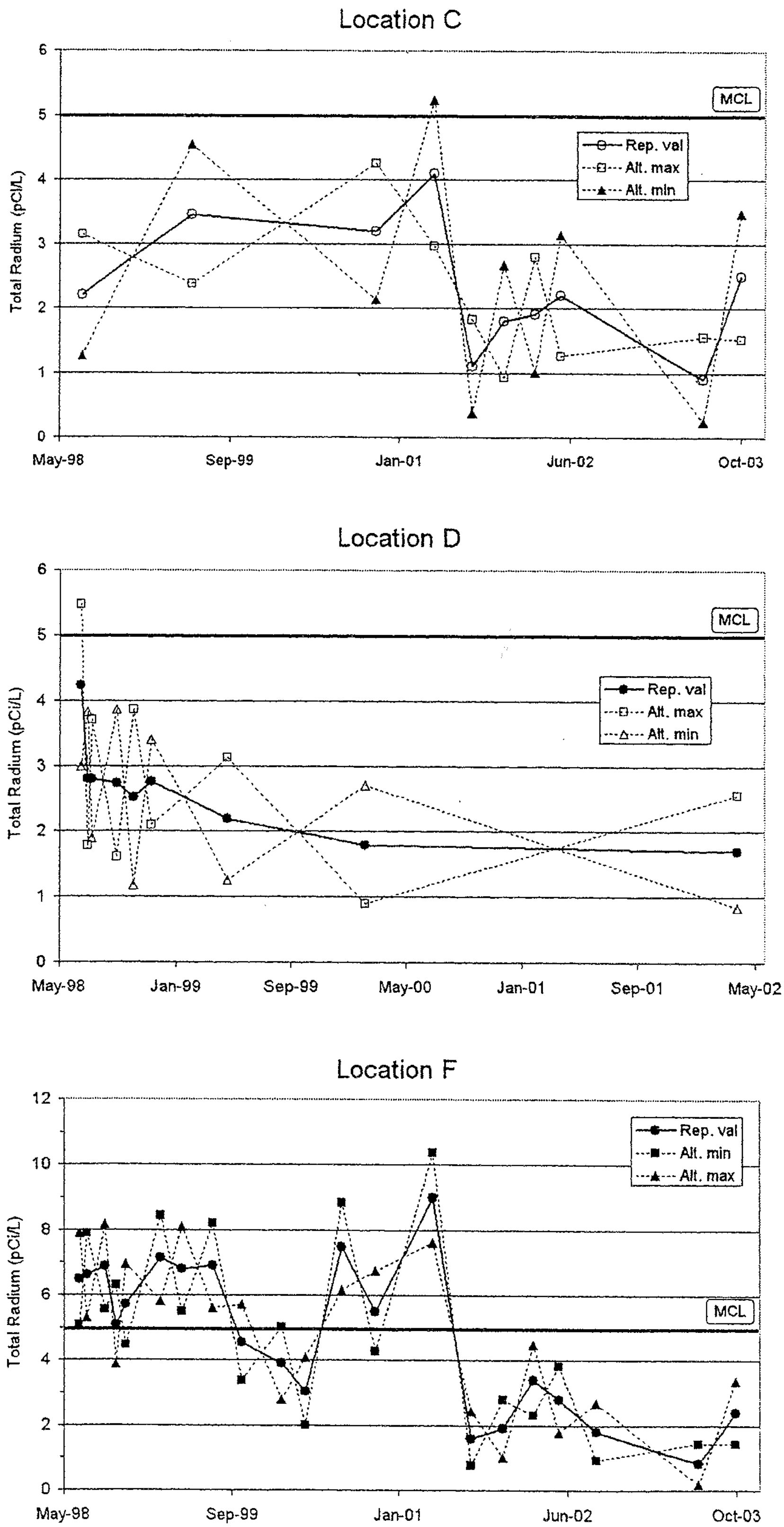


Figure 4: Examples of total radium time series. Solid lines are reported values and dashed

lines are alternating scenarios. Scenarios with significant trends at the 90% confidence level have filled in markers.

The comparison of the Mann-Kendall test results for the Reported Value Scenario and the Alternating Scenarios indicates that when the reported error in a time series is relatively large, Mann-Kendall results for the evaluation of trends can be unreliable. That is, looking only at the Reported Value Scenario can lead to the determination that statistically significant trends are present; whereas the trends are not significant in the Alternating Scenarios, where the analytical error bars are considered. This indicates a lack of robustness for the trends revealed by the original Mann-Kendall test. For most locations, the three scenarios show a large range in S and p-values. For example, Location E shows p-values ranging from 0.089 for the Reported Value Scenario to 1.000 for Alternating Scenario 1. This means that the Reported Value Scenario has less than a 10 percent probability of being random; while Alternating Scenario 1 indicates that when the analytical error bars are taken into consideration the probability of being random is 100 percent. Location K, which shows a significant trend under all the three scenarios, and all three S values (ranging from –29 to –41) support a robust downward trend at this particular location.

Data from Location D best illustrates the effect of the analytical error bars on the trend analysis. This location time series has 10 data points, and visually the time series appears to show a downward trend (Figure 2). The Mann-Kendall test on the Reported Value Scenario indicates that there is a significant downward trend with a p-value of 0.002. However, when the analytical error bars are accounted for, the Alternating Min/Max Test indicates that the trend is no longer significant, with a maximum p-value of 0.348 (Figure 4).

Location F shows the smallest variability in S and p-values and all statistical tests agree on a significant downward trend for these data. This particular time series has 21 data points, which is significantly more than the other examples discussed above. Results for F show that for time series with numerous data points (i.e. more than 10 data points) there is a large decrease in uncertainty for trend analysis and a better agreement between the Reported Value Scenario and the Alternating Scenarios.

Generally the two Alternating Scenarios tend to increase the p-value, showing the trend to be less significant than when considering the Reported Value Scenario only. However, the two Alternating Scenarios do not affect the p-values by the same magnitude (this occurs only 1 time for the 11 time series presented in Table 2). Interestingly, Alternating Scenario 2 reveals a trend at Location C that is not significant in the Reported Value Scenario. We do not consider this result to indicate that there is a significant trend, as it is not seen in either of the other scenarios (see Figure 4).

Based on the results of this study, it is apparent that the level of robustness of a trend, as tested by the Mann-Kendall approach, depends greatly on the analytical error bar. Therefore, the confidence level used by the laboratory in determining and reporting the analytical error (e.g. 1-sigma vs. 2-sigma) should be noted in the data. In the case of radium, the analytical error bar is assumed to represent a normal distribution with the reported value as the mean. The Alternating Min/Max test becomes more important if the error bar is considered to depict a uniform distribution, meaning that the reported value is a random value within the range of uncertainty. In this case the reported value has the same probability of being the true value as any other number that falls within the range of the analytical error bar.

Another check that has been applied to the Mann-Kendall test is the Coefficient of Variation (standard deviation divided by the mean; CV). The CV should be less than 1 for a "no upward or downward trend" result of a Man-Kendall test to be considered stable (Robb and Moyer, 2001; Aziz et al., 2003). This check includes some of the uncertainty in the data indirectly in that the analytical uncertainty can lead to more noise in the reported values, but stops short of directly assessing the impact of the uncertainty around each reported value.

Conclusion

The Mann-Kendall test for the presence of a trend can provide misleading results when applied only to the reported values for data with large uncertainty and a small number of data points in a time series. This is demonstrated in the radium data set for the Sand-and-Gravel aquifer in Escambia County, Florida. For this data set, the median reported error of 1.2 pCi/L for a median total radium activity of 3.0 pCi/L. Of the 35 radium time-series analyzed, 10 have trends at the 90 percent confidence level when examined with the Mann-Kendall test on reported activities. However, after applying a secondary test incorporating uncertainty around the reported values, 8 of the 10 trends are no longer significant. This rate of failure is likely to decrease with larger data sets. With more data at each location, more analytical variability will be represented in the reported values, as is apparent for the time series data for Location F. However, regulatory or managing decisions are commonly required upon review of time series with that include only a small number of sampling events. For radium data sets with small numbers of data the chance of a misleading Mann-Kendall result for trends can be important. The radium data also exhibit uncertainty in replicate analyses, with a median relative percent difference of 37 percent and 42 percent for Ra-226 and Ra-228, respectively. Replicate analyses can be a useful check indicating potentially unreliable Mann-Kendall results for trends; this is especially important if no reported errors are available for a data set.

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