

Use of Probability Distributions to Define Baseline Ground Water Quality at a Uranium Mill Site

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ABSTRACT

Background ground water quality is often used to drive remediation programs at sites with waters affected by mining activities. In the past these levels were sometimes chosen without rigorous statistical and geochemical analysis. A probability distribution approach was used at the Sweetwater Uranium Project to separate a large set of geochemical data into populations that represent contributions from different sources. Natural background populations were identified for major ions, metals, and radionuclides, and were submitted to the Nuclear Regulatory Commission to modify the extent of a Corrective Action Program and to establish control limits for future monitoring.

INTRODUCTION

Overview

The Sweetwater Uranium Project (Site) is in a remote area within the Red Desert portion of the Wyoming Great Divide Basin. It is located approximately 40 air miles northwest of Rawlins. The nearest permanent residence is approximately 17 air miles east of the Site. The general region in which the project area lies is used primarily for livestock grazing, dispersed recreation, wildlife range, oil and gas productions and mineral exploration. The Site uranium mill operated from 1981 through 1983 and is currently in standby status pending resumed operations.

The Great Divide basin is a closed basin. The Site is located within the Battle Spring Draw watershed, which empties into Battle Spring Flat, a playa located approximately six miles southwest of the Site. Regional wells are completed in either the Battle Spring or the Wasatch Formations, which are the two most important aquifers in the Great Divide Basin. The Battle Spring Formation, which underlies the Site and interfingers with the Wasatch Formation to the southwest of the Site, is comprised of thick Cenozoic sediments. The Site is primarily underlain by sandstones, with numerous thin, discontinuous claystone lenses. Uses of the aquifers include potable water supplies for industry, stock/game watering, dewatering, domestic, and miscellaneous (e.g. stock watering pipelines or monitoring). There are no domestic or potable water supplies not owned by the project within 10 miles of the Site. Furthermore, because the hydrologic low point of the basin, Battle Spring Flat, is within this 10-mile radius, there are no domestic or potable water supplies downgradient of the Site.

In 1989 a Corrective Action Program (CAP) was instituted at the site. The Site license with the Nuclear Regulatory Commission (NRC) currently specifies ground water protection standards under the CAP based on a determination of background from data that are limited in both time and areal extent. These protection standards were determined based on a limited number of samples taken from a cross-gradient well. As a consequence, ground water protection standards for certain radionuclides were set at a level lower than natural background, causing these standards to be exceeded by concentrations of unaffected waters in the area. Specifying protection standards from background concentrations that are

based on such limited data is inconsistent with currently accepted baseline (or background) determination approaches.

Planned resumption of operations at the Site will entail the construction of new tailings impoundments adjacent to the existing tailings impoundment. Prior to resumed operations, the Site license will be amended to include detection standards to allow identification of contaminated ground water in the unlikely case of a leak in the new impoundments.

Therefore, a more comprehensive determination of background ground water quality for the Site was conducted for two purposes: (1) to establish reasonable ground water protection standards and (2) to establish detection standards for resumed operations.

Data summary

The locations of wells used to determine background concentrations at the Site range from several on-site tailings monitoring wells to livestock watering wells located approximately 12 miles from the Site. Wells were divided into three categories based on their proximity to the Site: (1) on-site wells within 1.5 miles of the mill (termed site wells), (2) slightly remote wells within 5 miles of the mill (local wells), and (3) remote wells within 12 miles of the mill (regional wells). When weighing the significance of ground water quality data as background, the proximity of wells to the Site was taken into consideration for several constituents. Figure 1 is a regional map which shows the locations of the wells used in this study.

Data were analyzed from 65 wells (15 site wells, 44 local wells, and 6 regional wells) over a period of up to 20 years, for 33 constituents (major ions, trace metals, and radionuclides), for a total of 1082 records. A total of approximately 19,000 data values exists in the database. The database evaluated for background determination was selected to ensure that both spatial and temporal variability in natural ground water quality could be assessed.

The period of record for site wells was primarily 1979 to the present (but including data from as early as 1976). The local wells had a period of record for 1980 and 1981 only. Regional wells included samples from the time period 1981 to 1990.

STUDY METHODOLOGY

When estimating natural background concentrations in ground water, it is important to utilize methods which consider temporal, lateral and vertical variability in natural ground water quality. The use of a limited number of samples from a single well as a measure of natural background cannot yield valid background parameters because samples from one well cannot be representative of local ground water over a broad area or a large vertical extent. Mineralized areas such as the Great Divide Basin are variable geologically, structurally, hydrologically and geochemically, and selection of a single specific well for characterizing natural background neglects that inherent variability. It is not uncommon to identify multiple natural

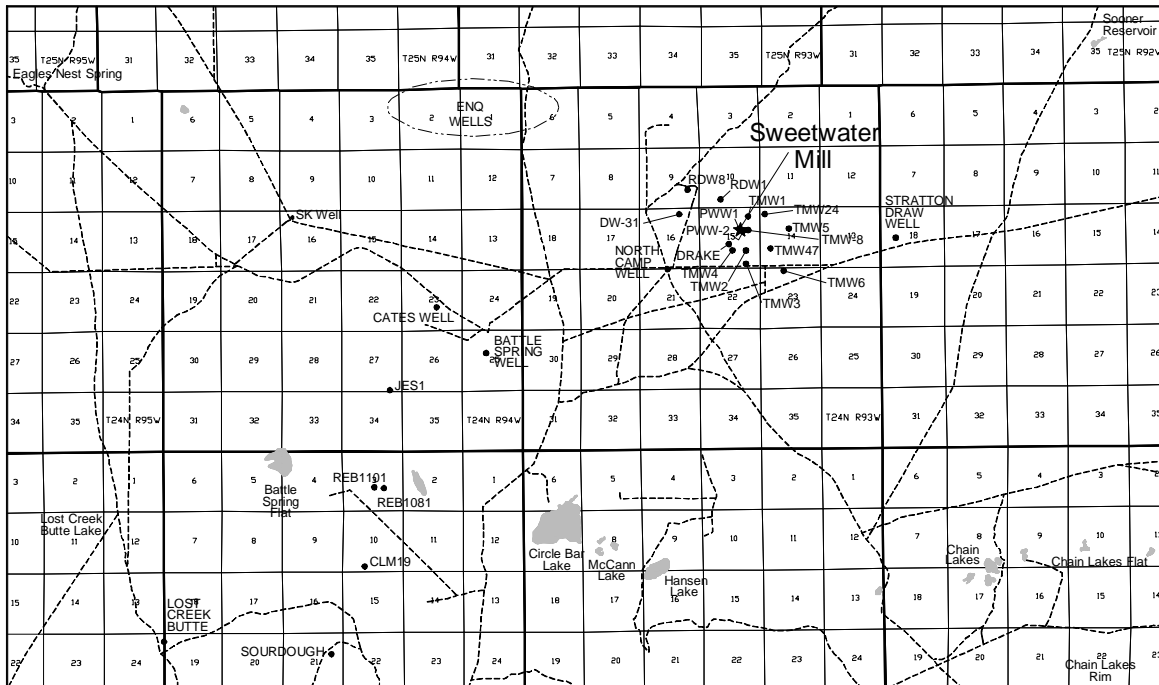


Figure 1. Project Area Well Map

background populations in a mineralized region, and therefore it is essential to consider a broad range of ground water data to allow assessment of variability over time and location and to use a proven and accepted technique which can identify different ground water quality populations.

This paper therefore presents an evaluation of background ground water quality for the Site by evaluating data which (1) include the entire period of record available for the data set, and (2) include all regional wells with meaningful data.

Probability distribution approach

Probability distribution diagrams, which have many applications in engineering and geology, are a time-tested method for summarizing large amounts of data in a comprehensive and understandable manner (Runnells and others, 1997; Sinclair, 1976; Saager and Sinclair, 1974). The greatest value of the approach for the application presented here is that it allows complex mixtures of geochemical data to be separated into categories, or families, that represent contributions from different sources.

The authors have used the probability distribution technique for identifying, separating and displaying chemical families of ground waters at mine and mill sites. The populations that result from the probability graphs are interpretable in terms of geographic locations, relative ages, aquifers, sampling depths, and contaminant source terms. Using this technique to display and separate different families of ground water in the vicinity of the Site may distinguish waters containing only natural background concentrations from waters showing impact from anthropogenic activities, such as leaks from tailings impoundments, effects from flooding of open pits, or effects

from pit dewatering operations. However, the technique allows all data to remain in the database; no groups of data are eliminated solely because of their deviation from the mean.

Ground water quality analyses were entered into the program PROBLOT (Stanley, 1987) for analysis to identify distinct families of waters. PROBLOT produces frequency distribution diagrams and probability graphs for either arithmetic or logarithmic-transformed data. Distinct populations on a probability graph can be identified by an inflection in the distribution of data. Although PROBLOT includes algorithms for automatic selection of inflections between identified populations, in this study the selection of inflection points was performed by visual assessment, which allows better differentiation between populations. Interpreting inflections in a probability diagram allows the separation of multiple families of ground water and derivation of a best-fit to the distribution of data points, so that PROBLOT can determine summary statistics (i.e., mean and standard deviation) for each family.

Screening the data

The data set provided by the Site licensee was screened using an approach which combines the method described in Guideline 4 from the Wyoming Department of Environmental Quality with a point-by-point analysis of identified outliers. Guideline 4 provides a statistical method for analyzing water quality databases. It is intended to be sensitive to significant departures from a normal distribution and/or temporal variability. This is accomplished through the identification of outlier values which exceed statistically calculated upper control limits.

Parameters were analyzed for outlier values using a 95% confidence level. For several of the screened constituents, a group of outlying values was identified which represented a separate population. However, in the majority of instances, these outlying values were retained in the database because they represented a distinct population and not errors in quality of analysis or transcription. Isolated outliers were compared to original laboratory data to determine if an error in transcription had occurred. If a transcription error was detected, the database was corrected. Where isolated values were identified which obviously represented errors in transcription or poor quality of analysis, but the nature of the error could not be determined, these values were eliminated. Approximately 10 values were eliminated from the database as obvious errors.

Most of the metals and radionuclides were comprised of a significant proportion of below detection limit values. These constituents were not screened using the method outlined in Guideline 4, but rather were screened visually for obvious errors.

Below detection limit values

As with other statistical methods, probability distribution analysis fails to produce an accurate estimate of summary statistics where a significant (greater than about 15%) of the values are reported as “non-detect,” or below detection limit values. In using PROBPLOT to provide mean estimates for a population comprised of a significant proportion of below detection limit values, the resulting mean value will usually be biased on the high side; this occurs because PROBPLOT ignores below detection limit values when defining a population.

A common approach for including below detection limit values into summary statistic calculations is the substitution of given values (such as one-half of the detection limit) for each below detection limit value, followed by calculation of summary statistics for this modified data set. The substitution approach was not considered for this analysis because it lacks a sound theoretical basis.

The problem of below detection limit values is further compounded when multiple detection limits are part of a database, as was the case for the project database. As described by Helsel (1990), populations of data comprised of significant proportions of multiple below detection limit values can be analyzed by synthetically generating below detection limit values based on a rank determined from the probability of exceeding each detection limit (see Helsel, 1988 for an example). This method utilizes all data for all detection limits. A computer program, MDL (Multiple Detection Limits), uses this method of “filling in” the below detection limit values before calculating arithmetic summary statistics for normal or log-normal distributions. The program MDL was applied only to the lowest population identified for each parameter.

Figure 2 is a histogram of natural uranium concentrations and exhibits the three distinct detection limits that have existed for this constituent over its period of record. Thirteen percent of the natural uranium data values collected were less than the detection limit applicable at the time of sampling. MDL was

used in this case to generate synthetic data below the multiple detection limits assuming a log-normal distribution.

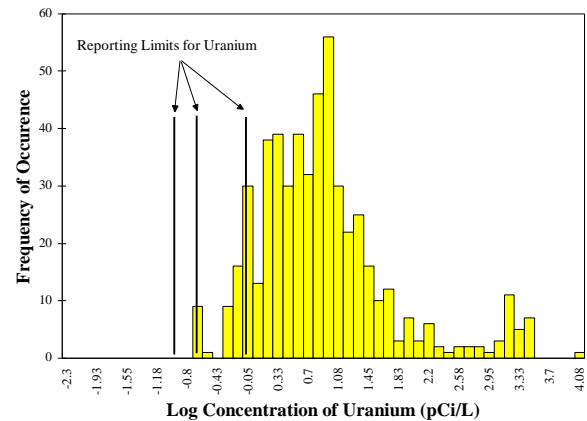


Figure 2. Natural Uranium Histogram

Geometric and arithmetic averages

For all components except pH, the data were approximated by log-normal distributions because natural geochemical data tend to be distributed log-normally. PROBPLOT provides geometric summary statistics for log-normally distributed data. In order to compare statistics from logarithmic probability distributions to regulatory values (such as compliance limits), it is necessary to convert geometric statistics to arithmetic statistics.

A straightforward approach was applied for converting geometric means to arithmetic means. After the appropriate inflections were manually applied to the logarithmic probability graphs in PROBPLOT, the same inflections were then used to differentiate the arithmetic distributions of the data. This method identifies multiple populations for log-transformed data then calculates the corresponding arithmetic statistics for these populations. In using this approach, PROBPLOT automatically calculated arithmetic summary statistics for the identified logarithmic populations in a simple, defensible approach. In cases where the proportion of below detection limit values was significant, MDL was used to calculate summary statistics.

RESULTS

Thirty-three constituents, consisting of 14 ions and other non-metals, 13 trace metals, and 6 radionuclides, were evaluated for background ground water quality. Table 1 lists these constituents and summarizes the background ground water quality estimates for each, listing arithmetic mean values as well as the mean plus two standard deviations. Values for background are reported in Table 1 to two significant figures. In several instances for trace metals, use of PROBPLOT or MDL could not yield an arithmetic mean for the Sweetwater data because so few above detection limit values were present. For these parameters the natural background mean

concentration is given as less than the most recent, or sensitive, detection limit.

Example constituents

Discussion is provided below of the determination of background for two example constituents. Magnesium is included because interpretation of the populations of this ion is instructive of the use of the technique to identify multiple populations. Natural uranium is included because it provides a

Table 1. Natural Background Ground Water Concentrations at the Site

Constituent	Units	N	ND (%)	Arithmetic Mean	Arithmetic Std. Dev.	Mean plus 2 Std. Dev.
<i>Non-metals</i>						
HCO3	mg/l	621	0	110	33	170
Ca	mg/l	517	0	19	14	47
Cl	mg/l	748	1	7.4	7.9	23
F	mg/l	728	0	0.28	1.1	2.5
Mg	mg/l	246	0	1.4	0.71	2.8
NO3-N	mg/l	482	25	0.99	3.1	7.2
K	mg/l	693	1	2.1	1.3	4.7
Na	mg/l	728	0	42	12	66
SO4	mg/l	574	1	45	16	77
pH	s.u.	553	0	7.71	0.52	6.67
TDS	mg/l	577	0	170	30	230
Cond	mho/cm	511	0	250	49	350
Alk	mg/l	600	0	99	26	150
CN	mg/l	124	99	<0.005	na	na
<i>Metals</i>						
As	mg/l	628	93	<0.001	na	na
Ba	mg/l	379	81	0.019	0.016	0.05
Be	mg/l	129	98	<0.01	na	na
Cd	mg/l	527	97	<0.001	na	na
Cr	mg/l	522	98	<0.01	na	na
Fe	mg/l	575	52	0.13	0.25	0.63
Hg	mg/l	473	89	0.0003	0.0017	0.004
Mo	mg/l	523	92	<0.01	na	na
Ni	mg/l	531	92	<0.01	na	na
Pb	mg/l	534	96	<0.01	na	na
Se	mg/l	620	87	0.001	0.004	0.01
Tl	mg/l	153	96	<0.015	na	na
Zn	mg/l	509	40	0.023	0.032	0.09
<i>Radiometrics</i>						
U,nat	pCi/l	573	14	8.0	14	36
Ra226	pCi/l	662	5	2.0	2.1	6.2
Pb210	pCi/l	586	17	2.5	3.2	8.9
Th230	pCi/l	632	20	1.6	2.7	7.0
Alpha	pCi/l	154	40	1.8	1.3	4.4
Ra226/228	pCi/l	167	16	2.0	1.9	5.8
na	not applicable					
N	number of values representing background population					
ND	percentage of background values which are below detection limits					
upper limit	based on arithmetic mean and two times the standard deviation					

good example of the use of MDL to determine representative statistics considering below detection limit data where multiple detection limits exist, and because it is an important constituent for establishment of reasonable ground water protection standards for the Site.

Magnesium (Mg): A total of 630 magnesium values were utilized for statistical analysis. Inflection points along the best fit line depicted in Figure 3 were identified at 25%, 65%, and

98.5% of the sample population that separate the sample into four distinct populations (see Figure 3). The four populations (616 above detection limit values) have arithmetic means of 0.17, 1.4, 7.7 and 50 mg/l (Tables 1 and 2). Lines that describe the statistics for the four populations of magnesium concentrations are also included in Figure 3. Because there were 14 below detection limit values observed, MDL was used to calculate summary statistics for the lowest population.

Table 2. Statistical Summary for Magnesium

Constituent	units	No. of Values	Non-Detects (%)	Populations (% of observed values)	Arithmetic Mean	Arithmetic Standard Deviation	Background Mean plus 2 Std. Dev. (see text)
Mg	mg/l	630	14(2)	25	0.17	0.10	-
				40	1.4	0.71	2.8
				33.5	7.7	4.5	-
				1.5	50	11	-

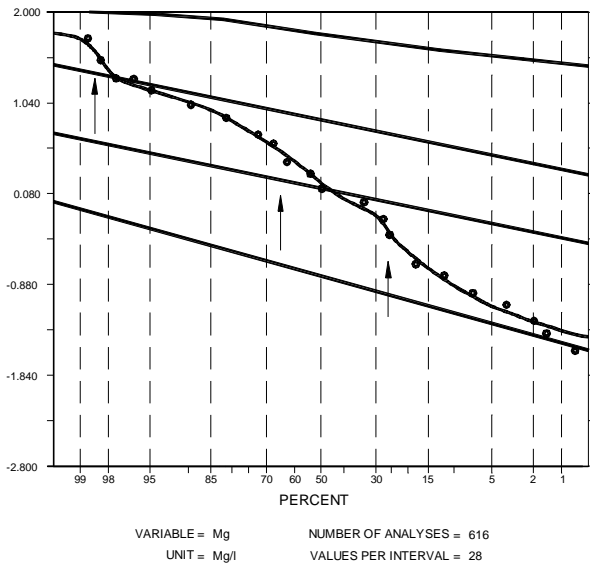


Figure 3. Probability Distribution for Magnesium

The upper population of magnesium values corresponds to regional wells (located near Battle Spring Flat approximately 11 miles southwest of the Site), while the next highest population corresponds to the earliest years for several site monitoring wells and some regional wells. Because the earliest dates of sampling for site monitoring wells may represent contamination related to well construction, the second highest population of magnesium (with a mean of 7.7 mg/l) is not considered to be background. The lowest magnesium population (mean value of 0.17 mg/l) corresponds specifically to local wells approximately 5 miles northeast of the Site and represents natural concentrations for that specific area. The second lowest population corresponds to stable concentrations in site monitoring wells during the most recent years of sampling. Therefore, the second population, with an arithmetic mean estimate of 1.4 mg/l, represents natural background for magnesium in the vicinity of the Site.

Natural uranium (Unat): A total of 610 uranium values were utilized for statistical analysis. Two populations were identified for uranium (Figure 4); the upper population arithmetic mean is 1500 pCi/l, and the lower population arithmetic mean is 8.0 pCi/l, as listed in Tables 1 and 3. Because there were 79 below detection limit values observed, MDL was used to calculate summary statistics for the lower population.

The upper population of natural uranium values is found primarily in one site well. Therefore, the upper population may not represent a site-wide natural background. However, the

mean value is still less than the Wyoming domestic standard. The lower population (93% of above detection limit values) is found in site monitoring wells, local wells, and regional wells. Because the values which comprise the lower population are found randomly in several wells for the entire time period of sampling, they are considered to be representative of natural background. MDL was used to combine the 494 above detection limit values with 79 below detection limit values for calculating an arithmetic mean of 8.0 pCi/l; this represents natural background for natural uranium in the vicinity of the Site

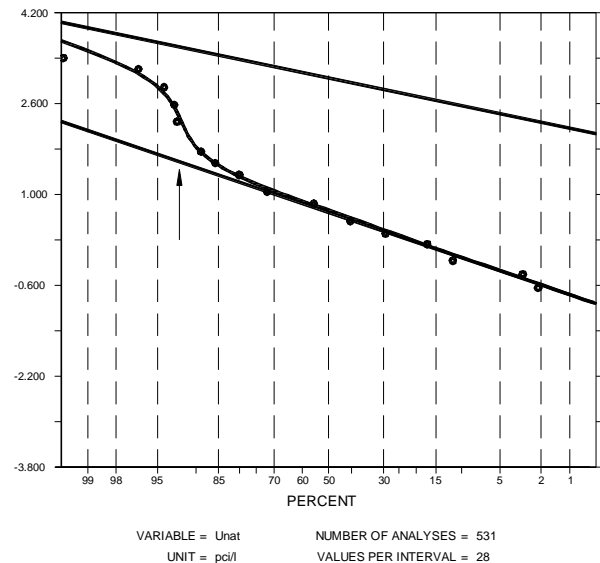


Figure 4. Probability Distribution for Natural Uranium

Ground water protection standards

The Sweetwater Uranium Facility has an ongoing ground water Corrective Action Program administered under the Site license. Table 4 summarizes the constituents and ground water protection standards specified in the license. Table 4 also presents for each license constituent the mean background value plus two standard deviations determined as presented above and current drinking water standards for each constituent where applicable.

As shown in Table 4, natural background values of several constituents, as defined by the mean plus two standard deviations, exceed the ground water protection standards. These constituents are lead-210, mercury, combined radium-226/228, and uranium. Therefore, with ground water protection standards that are more stringent than background,

remediation of the aquifer to current ground water protection

standards is impossible for these constituents.

Table 3. Statistical Summary for Uranium

Constituent	units	No. of Values	Non-Detects (%)	Populations (% of observed values)	Arithmetic Mean	Arithmetic Standard Deviation	Background Mean plus 2 Std. Dev. (see text)
Unat	pCi/l	610	79(13)	93	8.0	14	36
				7	1500	1500	-

The Site licensee has proposed to increase the ground water protection standards in the license for lead-210, combined radium-226/228, and uranium to the mean background concentration of these parameters plus two standard deviations. This would eliminate the current situation in which the license specifies ground water protection standards that are more stringent than natural background. Table 5 presents the proposed ground water protection standards list. Table 5 also lists applicable drinking water standards.

Currently the area of ground water defined by the 1.7 pCi/l natural uranium ground water protection standard includes a

significant portion of ground water that is within normal background variability. An area of ground water defined, as proposed, by a mean plus two standard deviations standard would more accurately reflect the actual affected area. It would eliminate areas with unaffected uranium concentrations that vary normally within the range of natural background between 1.7 pCi/l and the mean plus two standard deviations and the licensee would be allowed to focus efforts on the actual affected area.

Table 4. Summary of Existing Protection Standards for License Constituents

Constituent	Current NRC Ground Water Protection Standard	Drinking Water Standard	Background, Mean + 2 Std. Dev.
Arsenic	0.05 mg/l	0.05 mg/l (1)	na
Barium	1.0 mg/l	1.0 mg/l (1)	0.05 mg/l
Beryllium	0.01 mg/l	na	na
Cadmium	0.01 mg/l	0.01 mg/l (1)	na
Chromium	0.05 mg/l	0.05 mg/l (1)	na
Cyanide	0.005 mg/l	0.2 mg/l (2)	na
Lead	0.05 mg/l	0.05 mg/l (1)	na
Lead ²¹⁰	1.4 pCi/l	na	8.9 pCi/l
Mercury	0.002 mg/l	0.002 mg/l (1)	0.004 mg/l
Molybdenum	0.04 mg/l	na	na
Nickel	0.01 mg/l	na	na
Ra ²²⁶ /Ra ²²⁸	2.8 pCi/l	5 pCi/l (1)	5.8 pCi/l
Selenium	0.01 mg/l	0.01 mg/l (1)	0.01 mg/l
Silver	0.05 mg/l	0.05 mg/l (2)	na
Thallium	0.01 mg/l	na	na
Thorium ²³⁰	10.0 pCi/l	na	7.0 pCi/l
Natural Uranium	1.7 pCi/l	na	36 pCi/l
Gross Alpha	6.6 pCi/l	15 pCi/l (1)	4.4 pCi/l

Notes: (1) EPA Primary Drinking Water Standard
(2) Wyoming DEQ Domestic Standard

Table 5. Proposed Ground Water Protection Standards

Constituent (1)	Proposed Standard	Source of Standard (2)	Drinking Water Standard
Arsenic	0.05 mg/l	Table 5C (3)	0.05 mg/l
Beryllium	0.01 mg/l	Current GPS	na
Cadmium	0.01 mg/l	Table 5C	0.01 mg/l
Chromium	0.05 mg/l	Table 5C	0.05 mg/l
Lead	0.05 mg/l	Table 5C	0.05 mg/l
Lead ²¹⁰	8.9 pCi/l	Background (4)	na
Nickel	0.01 mg/l	Current GPS	na
Ra ²²⁶ /Ra ²²⁸	5.8 pCi/l	Background	5 pCi/l
Selenium	0.01 mg/l	Table 5C	0.01 mg/l
Thorium ²³⁰	7.0 pCi/l	Background	na
Natural Uranium	36 pCi/l	Background	na
Gross Alpha	15 pCi/l	Table 5C	15 pCi/l

Notes (1) The following constituents are proposed to be removed from the list of ground water protection standards because they are not found in appreciable quantities in the tailings fluid: barium, cyanide, mercury, molybdenum, silver and thallium

(2) EPA Primary Drinking Water Standard

(3) Table 5C from 10 CFR 40 Appendix A.

(4) Background mean plus 2 standard deviations.

Detection Standards

The Site licensee also proposed detection standards for resumed Site operations for three constituents that would best indicate that an excursion from the tailings impoundment has occurred: pH, conductivity and chloride. The standards would allow detection monitoring within the aquifer immediately downstream of the tailings impoundment. The parameter pH is included because the tailings are acidic due to the acid leach milling process and the fact that the natural background is approximately neutral (pH = 7.7). Conductivity is included because it is proportional to the total amount of dissolved ions in solution and thus would be indicative of the contrast between ion concentrations in tailings fluid and those in natural ground water. Chloride is also chosen because it is conservative and therefore would be likely to be detected at the compliance well should a tailings impoundment leak occur. Other constituents are likely to be present in lower concentrations or would be more easily affected by oxidation reduction or adsorption and would therefore be less likely to be detected.

The detection standards were proposed to be established at the mean background value plus two standard deviations. Should a standard be exceeded for two of the three detection parameters in one sampling episode, monthly monitoring would commence and the licensee would notify the NRC in writing. Because the mean plus/minus two standard deviations represents approximately 95% of normally distributed data, is probable that a small amount of data under natural conditions would exceed the mean plus two standard deviations level. Because natural variability would result in some concentrations greater than a mean plus two standard deviation detection standard, a single measurement higher than the standard would not necessarily indicate a leak. Therefore, an excursion would be considered to have occurred when three consecutive months

of monitoring show higher than standard concentrations for any two of the three parameters. It was estimated that the probability, under normal data variability, of a false positive (i.e., an identified leak where none has occurred) is on the order of 1 in 300 million.

With the current state of the art of double-lined impoundments leaks are highly unlikely, but if an excursion is determined, a corrective action program would be established. Monitoring frequency would return to quarterly if three consecutive months of measurements show the standards to have not been exceeded. Table 6 summarizes the proposed detection standards, at the background mean plus two standard deviations (minus two standard deviations in the case of pH), for the proposed Sweetwater Uranium Project tailings impoundments.

Table 6. Proposed Detection Standards for Resumed Operation

Constituent	Detection Standard
pH	6.67 *
Conductivity	350 mho/cm
Chloride	23 mg/l

* The standard for pH is a minimum; a detection notice would exist if the pH falls below 6.67 standard pH units.

Note: Exceedence of the standard for two of the three detection parameters would trigger monthly monitoring. Exceedence of the standards for two of the three parameters for three consecutive months would require notification of the NRC.

CONCLUSIONS

A rigorous methodology was employed to determine background ground water quality statistics for the Sweetwater Uranium Project. The methodology is valuable for its ability to utilize all the available data for a designated area and to separate the data for a constituent into multiple populations that represent contributions from different sources. The methodology also uses a technique to calculate meaningful statistics that include synthetically generated data below multiple detection limits. Alternative ground water protection standards were proposed based on the background determination methodology presented that are more theoretically sound than those based on a limited number of samples from a single cross-gradient well. Approval of the proposed protection standards would allow the licensee to focus remediation efforts on the actual affected area rather than on an area with concentrations that vary within the normal range of natural background. Additionally, reasonable standards are proposed to establish control limits for future monitoring.

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