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# INVESTIGATING URANIUM CONCENTRATIONS, ISOTOPIC RATIOS, AND ACTIVITY RATIOS IN GROUNDWATER IN THE STATE OF IDAHO

by

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## **Table of Contents**

| List of Figures |  | vii  |
|-----------------|--|------|
| List of Tables  |  | vii  |
| Abstract        |  | viii |
| Chapter 1.      | Introduction                                 | 1    |
| 1.1.            | Overview                                     | 1    |
| 1.2.            | Hypotheses                                   | 3    |
| Chapter 2.      | Background and Theory                        | 5    |
| 2.1.            | Motivation                                   | 5    |
| 2.2.            | Regulations and Uranium Measurement Methods  | 7    |
|                 | 2.2.1. EPA Methods 1620 and 900              | 8    |
|                 | 2.2.2. EPA Method 908.0                      | 9    |
|                 | 2.2.3. EPA Method 908.1                      | 10   |
|                 | 2.2.4. EPA Method 200.8                      | 11   |
| 2.3.            | Uranium Activity Ratios                      | 12   |
| 2.4.            | Previous Work                                |      |
| Chapter 3.      | Materials and Methods                        | 18   |
| 3.1.            | Introduction                                 | 18   |
| 3.2.            | Sample Preparation                           | 21   |
| 3.3.            | Kinetic Phosphorescence Analysis             | 22   |
| 3.4.            | Inductively Coupled Plasma Mass Spectrometry |      |
| 3.5.            | Quality Control                              | 28   |
|                 | 3.5.1. Split Samples                         | 28   |

|                                      | 3.5.2. Spiked Samples                                      | 30 |
|--------------------------------------|--|----|
| 3.6.                                 | Detection Limit  | 32 |
| 3.7.                                 | Statistical Tests and Error Propagation                    | 33 |
|                                      | 3.7.1. Dixon's Q-test                                      | 33 |
|                                      | 3.7.2. Error Propagation                                   | 34 |
| Chapter 4.                           | Results and Discussion                                     | 35 |
| 4.1.                                 | Introduction   |    |
| 4.2. Uranium Concentrations in Idaho |  | 35 |
|                                      | 4.2.1. Total Uranium in Idaho                              | 35 |
|                                      | 4.2.2. Isotopic Uranium in Idaho                           | 38 |
| 4.3.                                 | Enriched and Depleted Uranium                              | 42 |
| 4.4.                                 | Uranium Activity Ratios                                    |    |
| 4.5.                                 | 4.5. Lower Limits of Detection                             |    |
|                                      | 4.5.1. Determining the LLDs for KPA and ICP MS             | 45 |
|                                      | 4.5.2. LLDs for KPA and ICP MS vs. Radiochemical Technique | 47 |
| 4.6.                                 | Quality Control  | 49 |
| Chapter 5.                           | Conclusions and Future Work                                | 50 |
| 5.1.                                 | Conclusions  | 50 |
| 5.2.                                 | Future Work  | 51 |
| References                           |  | 52 |
| Appendix A                           |  | 55 |
| Appendix B                           |  | 72 |
| ppendix C                            |  | 80 |

## List of Figures

| Figure 2.1. | Recoil into Water   | 12 |
|-------------|---|----|
| Figure 2.2. | Etching of Tracks   | 13 |
| Figure 3.1. | Detailed Steps of Investigating Uranium in Idaho Using ICP MS     | 19 |
| Figure 3.2. | Detailed Steps of Investigating Uranium in Idaho Using KPA        | 20 |
| Figure 3.3. | Fluorescence  | 22 |
| Figure 3.4. | Phosphorescence   | 22 |
| Figure 3.5. | Three-Point Calibration for Low and High Ranges on KPA            | 24 |
| Figure 3.6. | Outline of ICP MS   | 26 |
| Figure 3.7. | Thermo Xseries 2 Calibration Curve for U-238                      | 27 |
| Figure 4.1. | Total Uranium Concentrations in Idaho Measured by KPA             | 37 |
| Figure 4.2. | <sup>238</sup> Uranium Concentrations in Idaho Measured by ICP MS | 39 |
| Figure 4.3. | UARs in Groundwater in Idaho                                      | 44 |

## List of Tables

| Table 2.1. | City of Pocatello Water Quality Table for Total Uranium                | 6  |
|------------|--|----|
| Table 3.1. | Three-Point Calibration for Low and High Ranges on KPA                 | 23 |
| Table 4.1. | Elevated Uranium Concentrations in Idaho                               | 41 |
| Table 4.2. | Samples with high UARs   | 45 |
| Table 4.3. | Lower Levels of Detection for KPA and ICP MS                           | 46 |
| Table 4.4. | <sup>234</sup> U concentrations as measured by Radiochemical Technique | 47 |
| Table 4.5. | <sup>235</sup> U concentrations as measured by Radiochemical Technique | 48 |
| Table 4.6. | <sup>238</sup> U concentrations as measured by Radiochemical Technique | 48 |

# INVESTIGATING URANIUM CONCENTRATIONS, ISOTOPIC RATIOS, AND ACTIVITY RATIOS IN GROUNDWATER IN THE STATE OF IDAHO

Dissertation Abstract – Idaho State University (2011)

Analyzing uranium isotope concentrations can give valuable information for hydrologic and environmental studies such as insights to weathering processes, estimating water mixing ratios, and identifying water sources. Besides naturally occurring uranium, depleted or enriched uranium can also be present in environmental samples if human activities have taken place in the vicinity. We employed an inductively coupled plasma mass spectrometer (ICP-MS) and a kinetic phosphorescence analyzer (KPA) to perform environmental level uranium concentration measurements on three hundred and eighty groundwater samples from various locations within the State of Idaho. These techniques can be used for detection of ground and surface water contaminations with high sensitivity and great precision. The lower limits of detection (LLD) were determined to be 0.532µg/L and 0.099µg/L for ICP-MS and KPA respectively. As a reference, the Environmental Protection Agency (EPA) drinking water limit for uranium is 30µg/L. Also, for the first time uranium concentrations were mapped on the Idaho grid. Some parts of the state exhibit elevated uranium concentrations (over  $30\mu g/L$  in some cases). The uranium activity ratios (UAR) for these samples range between 0.91 and 6.21, which suggests that the parent <sup>238</sup>U is not in equilibrium with its daughter product <sup>234</sup>U. Therefore, uranium is mobile in the groundwaters in Idaho. Determining the UARs for the samples that represent the whole state was accomplished for the first time as well in the frame of this project. In addition, a novel approach for determining the total uranium concentration was proposed. EPA uses a few different methodologies to determine the uranium concentrations in aqueous samples. But all these methodologies assume the presence of only natural uranium in the medium. Hence, in the presence of depleted/enriched uranium in the sample, all the EPA methods overestimate/underestimate the true uranium concentration. We propose a new method which determines the true uranium concentration even in the presence of depleted/enriched uranium. Finally, no enriched or depleted uranium was detected outside the Idaho National Laboratory (INL) as determined within the framework of this project.

## **CHAPTER 1**

## Introduction

#### 1.1. Overview

Uranium is a terrestrial naturally occurring radioactive material. The average concentration of uranium in the earth's crust corresponds to  $2\mu$ g uranium per gram of soil and it is mainly concentrated in the acidic magmatic rocks. Soluble uranium carbonate complexes produced in soil are commonly transferred in natural waters (El Himri et al, 2000). Natural uranium consists of 99.2742% of <sup>238</sup>U, 0.7204%, of <sup>235</sup>U, and 0.0054% of <sup>234</sup>U (Environmental Protection Agency). Despite its very small fraction in natural uranium, measuring <sup>234</sup>U concentrations can be a very useful tool because of the high specific activity of this isotope (1.8x10<sup>4</sup> higher than that of <sup>238</sup>U).

The determination of uranium concentrations in natural water samples is of a great interest due to the well documented human health effects and environmental consequences of this radionuclide. Uranium concentrations as low as  $500\mu g/L$  can affect the reproductive capability of aquatic organisms (El Himri et al, 2000). EPA established the drinking water limit to be  $30\mu g/L$ .

The slight fractionation of <sup>234</sup>U over <sup>238</sup>U that takes place during weathering is the main source of information for weathering processes, estimating water mixing ratios, and identifying water sources (Kraemer et al, 2002). Physical and chemical changes in host material due to the alpha decay of <sup>238</sup>U make the daughter product <sup>234</sup>U more susceptible to leaching and removal than the parent <sup>238</sup>U. Consequently, the <sup>234</sup>U/<sup>238</sup>U activity ratio (UAR) in natural waters

commonly exceeds 1, the equilibrium value found in closed systems. As a result, waters contained in diverse rock environment acquire distinct UARs, which can subsequently be used for their identification as they emerge and move as surface water or groundwater. These UARs then can be used as an additional tool to be applied to environmental problems such as identifying source areas of water containing contaminant materials or assisting to predict future transport of a contaminant by establishing the present flow path (Kraemer et al, 2002).

Also, mapping uranium concentrations can be beneficial to identify areas of elevated contaminant concentrations. Both methodologies used in this study are suitable for mapping purposes. Besides mapping, determining the lower limits of detection (LLD) for these two methods and comparing them to the other methodologies can assist in establishing a cost effective and local uranium measurement program at Idaho State University.

Creating a database with the uranium ratios is a prerequisite to identifying activity alternation in the uranium concentrations in the groundwaters of the state of Idaho.

Finally, developing a new methodology for measuring the total uranium concentration in the presence of depleted or enriched uranium is of a great interest. All the accepted EPA methodologies assume no presence of depleted or enriched uranium in the medium and therefore overestimate or underestimate the true total uranium concentration in the presence of depleted or enriched uranium.

2

#### 1.2. Hypothesis

Besides mapping the uranium concentrations in the State of Idaho and developing a new method for estimating the total uranium concentrations, four sets of hypothesis were tested in this project. All three hundred and eighty samples were analyzed for depleted and enriched uranium. The hypothesis tested for these samples were:

- **H10:** Uranium depleted or enriched in <sup>235</sup>U is not found in the 380 samples from Idaho wells outside the INL reservation.
- **H1A:** Uranium depleted or enriched in <sup>235</sup>U is found in the 380 samples from Idaho wells outside the INL reservation.

For the first set of Hypotheses the  ${}^{235}\text{U}/{}^{238}\text{U}$  concentration ratio was determined for each sample and the resulted value was compared to the ratio of natural abundances of these isotopes. Since the  ${}^{235}\text{U}/{}^{238}\text{U}$  concentration ratio for natural uranium is expected to be between 0.006 and 0.008, the samples exhibiting values lower than 0.006 were accepted as the samples depleted in  ${}^{235}\text{U}$  and the samples exhibiting values higher than 0.008 were accepted as the samples enriched in  ${}^{235}\text{U}$ .

Analyzing isotopic uranium in groundwater, gives information about the uranium equilibrium state. Therefore, the second set of the hypothesis tested were:

- **H20:** <sup>238</sup>U is in equilibrium with its daughter product <sup>234</sup>U in the groundwaters in the State of Idaho.
- H2A: <sup>238</sup>U is not in equilibrium with its daughter product <sup>234</sup>U in the groundwaters in the State of Idaho.

For the second set of Hypotheses <sup>234</sup>U/<sup>238</sup>U activity ratio (UAR) was determined for each sample and the resulted value was compared to the unity. The sample was accepted as "in equilibrium" if the sample UAR was within two standard deviations of the unity. Similarly, the sample was accepted as "out of equilibrium" if the sample UAR was outside two standard deviations of the unity. The two standard deviation rule corresponds to the 95% confidence level.

Furthermore, by comparing the KPA and ICP MS methods to the radiochemical method of measuring uranium concentrations the following hypothesis have been tested:

- **H30:** The lower limit of detection of the KPA method for determining natural uranium concentrations is comparable to or worse than that obtained with the radiochemical method.
- **H3A:** The lower limit of detection of the KPA method for determining natural uranium concentrations is better than that obtained with the radiochemical method.
- **H40:** The lower limit of detection of the ICP MS method for determining uranium isotope concentrations is comparable to or worse than that obtained with the radiochemical method.
- **H4A:** The lower limit of detection of the ICP MS method for determining uranium isotope concentrations is better than that obtained with the radiochemical method.

For the third and fourth sets of Hypotheses the lower limits of detection (LLD) were determined for both instruments and these values were compared to the published minimum detectable concentrations (MDC) for the radiochemical analysis methods.

4

## **CHAPTER 2**

## **Background and Theory**

#### 2.1. Motivation

Natural uranium is one of the major radionuclides monitored in drinking water. Because exposure to natural uranium in drinking water can cause toxic effects to the kidney, the Environmental Protection Agency in 1991 proposed a Maximum Concentration Level (MCL) of 20µg/L. Later, in 2000, because the additional cost to reduce uranium concentration in drinking water to 20µg/L could not be technically justified, the MCL was changed to 30µg/L. EPA determined that the new value was the appropriate MCL, because it maximizes the net benefit, while providing an adequate margin of safety for protecting against kidney toxicity and carcinogenicity (Environmental Protection Agency).

Water monitoring programs are usually conducted by local water treatment authorities, and then results are reported to the governing institutions. Each year city water departments are required to present the city water quality report to the Mayor, City Council, and citizen stakeholders.

Uranium is one of the radionuclides monitored by the City of Pocatello in the drinking water. The following table summarizes the analysis results of monitoring natural uranium in Pocatello drinking water in the years between 2006 and 2010.

| Year | Month/Year Sampled | Minimum conc. (ppb) | Maximum conc. (ppb) |
|------|--------------------|---------------------|---------------------|
| 2006 | March 2005         | 1.6                 | 3.0                 |
| 2007 | June 2005          | 1.72                | 3.86                |
| 2008 | June 2005          | 1.72                | 3.86                |
| 2009 | June 2005          | 1.72                | 3.86                |
| 2010 | June 2005          | 1.72                | 3.86                |
| 2011 | 2010               | 1.82                | 1.82                |
|      |                    |                     |                     |

 Table 2.1. City of Pocatello Water Quality Table for Total Uranium

(Pocatello Water Reports, 2010)

It can be noted that there has been no analysis conducted between 2005 and 2010 and the same results were reported for those five years. The regulations do not require the drinking water to be reanalyzed every year, but it is possible that the uranium concentrations in the drinking water are altered due to human activities and/or natural phenomena. Hence developing a cost effective and local expertise for measuring uranium concentrations in water could greatly assist monitoring uranium in drinking water.

Also, determining the concentrations of different uranium isotopes can give information about the presence of any depleted or enriched uranium in the water, or changes in the groundwater system.

#### 2.2. Regulations and Uranium Measurement Methods

All the waters in the United States are regulated for contaminants. The Environmental Protection Agency (EPA) is the governing body that sets the limits for different contaminants and monitors the water quality. The Clean Water and Safe Drinking Water Acts are the EPA regulations for navigable and drinking waters.

The Clean Water Act (WCA) is the standard regulating discharges of pollutants into the waters of the United States. Within the framework of the CWA, EPA implements pollution control programs including setting wastewater standards for industry. According to the CWA, it is illegal to discharge any pollutant from a point source into navigable waters without a permit. The program that controls these discharges is EPA's National Pollutant Discharge Elimination System (NPDES) (Environmental Protection Agency).

The Safe Drinking Water Act (SDWA) is the main federal law that ensures the quality of drinking water in the United States. Within the framework of the SDWA, EPA sets standards for drinking water quality and oversees the states, localities and water suppliers who implement those standards (Environmental Protection Agency). The SDWA also establishes concentration limits for different radionuclides in drinking water. Uranium is one of the radionuclides that EPA uses in its monitoring programs. The MCL for natural uranium is  $30\mu g/L$  (Environmental Protection Agency).

EPA uses a number of different methodologies to analyze uranium in aquatic samples. Although some of the methodologies are capable of distinguishing between the different uranium isotopes, EPA assumes all uranium in water to be natural and bases all the concentration calculations on the natural ratios of the uranium isotopes. Therefore, in case of the presence of depleted or enriched uranium, none of the EPA methods give accurate estimates of the uranium concentrations. The following is a summary of the EPA methodologies for measuring uranium in water.

#### 2.2.1. EPA Methods 1620 and 900

EPA Methods 1620 – 'Inductively Coupled Plasma Atomic Emission Spectroscopy' and Method 900 – 'Gross Alpha and Gross Beta Radioactivity in Drinking Water' are general methods for determination of trace elements in solutions. A number of elements can be detected using these two methods. Uranium is one of the elements to which these methods may be applied.

Method 1620 is based on the measurement of atomic emission using optical spectroscopic techniques. Analyses using this technique employ the following general sequence:

- The samples are nebulized and the resulted aerosols are transported to the plasma torch where excitation occurs
- A radio-frequency inductively coupled plasma is then used to produce characteristic atomic-line emission spectra
- Further dispersion of the spectra by a grating spectrometer makes it possible to monitor the intensities of the lines using photomultiplier tubes (PMTs)
- The Processed photocurrents from the PMTs are then correlated to the concentrations of the contaminants in the sample (Methods and Guidance for the Analysis of Water, 1999).

The Minimum Detectable Concentration for uranium measured by this method is 1mg/L.

Method 900 is another screening technique for monitoring drinking water supplies for alpha and beta particle activities. Analyses with EPA Method 900 employ the following general sequence:

- An aliquot of a preserved drinking water sample is evaporated and transferred to a 2-inch stainless steel counting planchet
- The sample residue is dried to constant weight, reweighed to determine dry residue weight, and counted for alpha and/or beta radioactivity using gas flow proportional counters (Methods and Guidance for the Analysis of Water, 1999).

#### 2.2.2. EPA Method 908.0

EPA Method 908.0 is the 'Radiochemical Method for Uranium in Drinking Water'. This method is specifically developed to measure natural uranium in drinking water. Analyses with EPA Method 908.0 employ the following general sequence:

- The aqueous sample is acidified by adding HCl and the resulted acid is boiled to eliminate carbonate and bicarbonate ions
- Uranium is then separated from the sample by coprecipitation with ferric hydroxide
- Further separation of the uranium from other radionuclides which were precipitated with the ferric hydroxide is achieved by dissolving the hydroxide precipitate in 8N HCl, putting the solution through an anion exchange column, washing the column with 8N HCl, and finally eluting the uranium with 0.1N HCl

The resulting elute is then evaporated and the uranium chemical form is converted to nitrate. The residue is transferred to a stainless steel planchet, dried, flamed, and counted for alpha particle activity

Spiking blank samples with known amounts of uranium and taking them through the procedure as regular samples gives information about uranium recovery.

Counting efficiencies are established by transferring measured aliquots of a uranium standard to a planchet, diluting with 6 to 8 ml of a 1 mg/ml HIO<sub>3</sub> solution in 4N HNO<sub>3</sub>, evaporating to dryness, flaming the planchet, and counting in an alpha counter (Methods and Guidance for the Analysis of Water, 1999).

#### 2.2.3. EPA Method 908.1

EPA Method 908.1 is the 'Fluorometric Method for Uranium in Drinking Water'. This method like Method 908.0 is developed specifically for measuring uranium in drinking water, but does not give any information about isotopic uranium. The MDC for this method is  $0.1\mu g/L$  and there is no upper limit. If the uranium concentration exceeds the upper detection limit of the method, the sample needs only be diluted. Method 908.1 is applicable for the monitoring of water discharges from industries related to the uranium fuel cycle. Analyses with EPA Method 908.1 employ the following general sequence:

- Uranium is concentrated by coprecipitation with aluminum phosphate.
- The aluminum phosphate is dissolved in dilute nitric acid containing magnesium nitrate as a salting agent and the coprecipitated uranium is extracted into ethyl acetate
- > The ethyl acetate is removed by evaporation

- The extracted residue is dissolved in nitric acid and diluted to volume in a small volumetric flask
- Aliquots are transferred to each of two fusion dishes and dried
- $\blacktriangleright$  Known mass of uranium, usually 0.1µg/L, is added to one of the dishes and dried
- Solution containing sodium fluoride is added to both dishes, fused at a prescribed temperature, cooled and analyzed using a fluorometer
- Interferences that may coextract with uranium are corrected by the use of the standard addition technique

(Methods and Guidance for the Analysis of Water, 1999)

#### 2.2.4. EPA Method 200.8

EPA Method 200.8 is 'Determination of Trace Elements in Waters and Wastes by Inductively Coupled Plasma – Mass Spectrometry'. It provides procedures for determination of dissolved elements in ground waters, surface waters and drinking water. Uranium is one of the elements that can be tested using this method. Although ICP MS can provide information on each isotope, EPA assumes that all the uranium in the samples is natural and it only determines the concentration of <sup>238</sup>U. <sup>234</sup>U and <sup>235</sup>U concentrations can be calculated with the assumption of natural abundances of uranium isotopes. The MDC for this method is 0.1µg/L (Methods and Guidance for the Analysis of Water, 1999).

#### **2.3. Uranium Activity Ratios**

Isotopic ratios have great power to detect environmental processes and to reveal alternations caused by human activities. The <sup>234</sup>U/<sup>238</sup>U activity ratio is a much-studied term when investigating uranium in different media (Fleischer 2008). <sup>234</sup>U is a product of <sup>238</sup>U by an alpha decay followed by two rather prompt beta decays. Water flowing through porosity in rock or soil will preferentially pick up <sup>234</sup>U and hence be increased in the <sup>234</sup>U/<sup>238</sup>U ratio. Subsequently, the rock or soil the water passed through is decreased in the <sup>234</sup>U/<sup>238</sup>U ratio. If the enriched water later evaporates in a different geological environment, it will cause the enrichment of that geological material. Depending on its detailed history, a material may have a <sup>234</sup>U/<sup>238</sup>U activity ratio which is greater than or less than unity; the value found in the closed geological systems (Fleischer 2008).

There are two main physical mechanisms that can account for the  $^{234}U/^{238}U$  disequilibrium. Figure 2.1. shows the first of the two.



Adopted from Fleischer 2008

Figure 2.1. Recoil into Water

In this mechanism, the <sup>234</sup>Th recoil from alpha decay of <sup>238</sup>U is stopped in the water provided the decay is close to the surface of the solid particle (Fleischer 2008). Decay of <sup>234</sup>Th into <sup>234</sup>U enriches the water in the <sup>234</sup>U/<sup>238</sup>U ratio.

Gas α particle <sup>238</sup>U Decay <sup>234</sup>Th Recoil

Adopted from Fleischer et al 2008

#### Figure 2.2. Etching of Tracks

The second mechanism is shown on Figure 2.2.

There is no intervening fluid between the grains in this second mechanism. As a result, the recoil nucleus crosses the space and causes radiation damage in the neighboring grain. Later when pore fluid enters the system, it leaches the <sup>234</sup>Th recoil or its decay product <sup>234</sup>U. Both mechanisms, depending on circumstances, result in the enrichment of water in the <sup>234</sup>U/<sup>238</sup>U ratio.

The common methods for detecting uranium isotopes include highly precise thermal ionization mass spectrometry (TIMS) and alpha particle spectrometry (APS). Another method that is widely used is inductively coupled plasma mass spectrometry. The advantages of the latter

methods compare to TIMS and APS is lower cost and high sample through-put (Jarvis et al, 1992).

The common approach to estimating the  ${}^{234}U/{}^{238}U$  activity ratio includes determining  ${}^{234}U^+$  and  ${}^{235}U^+$  ion intensities instead of measuring the extreme differences in  ${}^{234}U^+$  and  ${}^{238}U^+$  ion intensities that occur in natural uranium samples (Kraemer et al, 2002). Taking into account that the  ${}^{235}U/{}^{238}U$  ratio is essentially invariant in natural uranium at a value of  $7.253 \times 10^{-3}$ , the  ${}^{234}U/{}^{238}U$  ratio can be obtained as follows:

$$UAR_{samp} = \frac{\left[\frac{2^{34}U}{2^{35}U}\right]_{samp} \left[\frac{2^{35}U}{2^{38}U}\right]_{N}}{\left[\frac{2^{34}U}{2^{38}U}\right]_{N}}$$
(2.1)

Where  $[^{234}U/^{235}U]_{samp}$  is the mass ration of  $^{234}U$  to  $^{235}U$  measured for the sample,  $[^{235}U/^{238}U]_N$  is the mass ration of  $^{235}U$  to  $^{238}U$  (7.253x10<sup>-3</sup>) for natural uranium, and  $[^{234}U/^{238}U]_N$  is the mass ration of  $^{234}U$  to  $^{238}U$  (5.591x10<sup>-5</sup>) for natural uranium in secular equilibrium. The values presented for the natural ratios are based on published data (Kraemer et al, 2002).

By stating that  $[^{235}U/^{238}U]_N$  in Equation 2.1 is constant, it is assumed that the samples contain only natural uranium. This method should not be used if samples are likely to contain depleted or enriched uranium (Kraemer et al, 2002).

Because of high sensitivity, good accuracy of isotope measurements and relatively simple sample preparation procedures, ICP-MS is becoming one of the main tools for routine long-lived radionuclide isotopic ratio determinations in environmental samples (Izmer et al, 2003).

#### 2.4. Previous Work

Several research groups have employed and developed different methodologies for measuring uranium concentrations.

Kraemer et al. described a method that allows precise determination of <sup>234</sup>U/<sup>238</sup>U activity ratios (UAR) in natural water samples by ICP-MS. The instrument used in this study was a Perkin-Elmer ELAN 6000 ICP-MS with CETAC U-6000 AT+ ultrasonic nebulizer and CETAC membrane desolvator (model MSX-200).

Kraemer et al. used NIST 4321B (Natural Uranium Standard) to determine optimal operation parameters for the instrument. This standard has an adequately known UAR of  $0.963 \pm 0.0003$ . The main goal of the optimization process was to maximize the <sup>234</sup>U<sup>+</sup> ion count rate, while minimizing the sample consumption. Two different sets of operational parameters were determined for the <sup>234</sup>U/<sup>235</sup>U isotopic determination mode and for the dissolved uranium concentration analysis mode. The latter step is performed as soon as the samples reach the laboratory and is mainly used to estimate the amount of sample needed to produce an acceptable <sup>234</sup>U<sup>+</sup> ion intensity during the isotopic ratio phase of analysis. Therefore, any other technique for determination of total uranium concentration can be used.

Kraemer et al. found that the precision achieved by the ICP-MS technique is  $\pm$  0.5% RSD, 1 sigma, which lies in between TIMS ( $\pm$  0.25% RSD, 1 sigma) and APS ( $\pm$  0.5% RSD, 1 sigma). The group also determined the transport path of the surface and ground waters on the sample locations, based on the measured values of <sup>234</sup>U/<sup>235</sup>U ratios.

El Himri et al. developed a simple strategy for determination of uranium in tap and natural waters using ICP-MS. The measurements were performed at the mass number 238 and Rh was used as an internal standard. The instrument used in the study was Perkin-Elmer Sciex ELAN<sup>TM</sup>

5000 ICP-MS spectrometer equipped with a Perkin-Elmer Ryton Scott-type spray chamber and cross-flow nebulizer. The ICP-MS data was validated by alpha spectrometric measurements of the same samples using a Tennelec instrument.

El Himri et al. observed a linear relationship between U/Rh relative signals versus uranium concentration when using external solutions of uranium in the presence of 100ng/mL Rh. Seventeen standard solutions with uranium concentrations ranging from  $0.05\mu$ g/L to  $125\mu$ g/L were analyzed in the quantitative mode. It was concluded that analytical sensitivity of uranium using ICP-MS provided a limit of detection in the order of 3ng/L uranium with a confidence level of 99.6%.

A number of studies have been conducted to determine  ${}^{234}U/{}^{238}U$  uranium activity ratios in different media as well. Thurber (1963) reported UARs up to 2.1 for tufas and 2.3 for oolites. UAR values of 1.4 in sandstones were found by Rosholt et al. (1963) and later Rosholt found UARs ranging between 0.51 and 1.65 in tuffs (1980). Szabo (1969) reported UARs ranging between 0.5 and 3.3 and later studies by Szabo (1982) revealed UAR values to be 3.4 in clays and 14 in sands. The values up to 12 were found in water by Osmond and Cowart (1976). Kronfeld (1974) recorded the  ${}^{234}U/{}^{238}U$  alpha-activity disequilibrium as great as 1100% within the Cretaceous Trinity aquifer of central Texas (Kronfeld 1974).

Roback et al. investigated the uranium isotopic evidence for groundwater chemical evolution and flow patterns in the eastern Snake River Plain aquifer in Idaho. The research group used isotope-dilution thermal ionization mass spectrometry to determine uranium concentrations and isotopic composition. The study was mainly conducted in and around the Idaho National Laboratory (INL), which has a long history of storing and disposing of radioactive waste, some of which has entered the aquifer (Roback et al, 2001). Roback et al. reported uranium concentrations in the INL groundwater to range between 0.3µg/L and 3.6µg/L, and the <sup>234</sup>U/<sup>238</sup>U uranium activity ratios to range between 1.5 and 3.1. No depleted or enriched uranium was found in this study and <sup>236</sup>U was not detected either. Hence it was concluded that the <sup>234</sup>U/<sup>238</sup>U UARs reflect natural variations in the aquifer. The findings were interpreted to identify chemically distinct water masses, to assess mixing and water-rock interaction, and to delineate detailed groundwater flow pathways in and around INL (Roback et al, 2001).

## **CHAPTER 3**

## **Materials and Methods**

#### **3.1. Introduction**

The purpose of the study was to investigate uranium activities and their ratios in groundwaters in the State of Idaho. Three hundred and eighty aqueous samples were obtained for this reason from all around the state from the Idaho Department of Water Resources and analyzed by two different methods. The methodologies used were Kinetic Phosphorescence Analysis (KPA) and Inductively Coupled Plasma Mass Spectrometry (ICP MS). The work was performed in the Environmental Monitoring Laboratory (EML) and in the Center of Archaeology, Materials and Applied Spectroscopy (CAMAS) at Idaho State University. Thirty one samples were split and a number of spikes were prepared for quality assurance and quality control. The splits and spikes were analyzed as independent samples. Triplicate measurements were performed for each sample on each machine. Then the average concentration was calculated and the uncertainty was propagated for each data point. Dixon's Q test was used to reject the outliers from the data.

The sensitivities of the both techniques employed were compared to each other, and also to a radiochemical analysis technique. A number of samples previously analyzed by the radiochemical analysis technique at a vendor laboratory were reanalyzed using KPA and ICP MS. The results acquired from the KPA and ICP MS analysis were then compared to those reported by the radiochemical measurements.

The detailed steps of the project are shown on the following flow charts:



Figure 3.1. Detailed Steps of Investigating Uranium in Idaho Using ICP MS



Figure 3.2. Detailed Steps of Investigating Uranium in Idaho Using KPA

#### **3.2. Sample Preparation**

Each sample was acidified with a concentrated nitric acid to obtain 2% acid solutions. The heavy metals, including uranium, tend to get attached to the walls of sample containers. Acidification of the samples is necessary to remove uranium from the container walls and make sure all the uranium in the sample is accounted for when analyzing the sample matrix. Each acidified sample was left in its container for 24 hours before the analysis. After this period of time the samples were filtered using 0.45-µm pore size filters. The filtration step is necessary to remove any residue from the sample that could result in quenching when performing the kinetic phosphorescence analysis. Also for quality control and to evaluate reproducibility, two samples in every twenty five samples were split and treated as separate samples when performing analysis.

To analyze the samples on both KPA and ICP MS a number of natural uranium standards were prepared. Two different original standards used were: 1) 735µg/L natural uranium standard by Environmental Resource Associates (ERA), and 2) 0.01998g/g natural uranium standard by National Institute of Standards and Technology (NIST). The standard concentrations prepared for KPA were  $1 \pm 0.1\mu g/L$ ,  $5 \pm 0.1\mu g/L$ ,  $10 \pm 0.1\mu g/L$ ,  $50 \pm 0.1\mu g/L$ ,  $100 \pm 0.1\mu g/L$  all from the ERA standard. The standard concentrations for the ICP MS analysis were  $0.1 \pm 0.01\mu g/L$ ,  $1 \pm 0.01\mu g/L$ ,  $10 \pm 0.01\mu g/L$ , and  $100 \pm 0.01\mu g/L$  prepared from both ERA and NIST standards.

#### **3.3. Kinetic Phosphorescence Analysis**

The KPA analysis method uses a pulsed-laser phosphorimeter to determine total uranium in water. When a photon strikes a molecule, a possible scenario involves the molecule absorbing the photon and becoming exited. The return of the molecule to the ground state is accompanied by emission of a photon, which has energy equal to the difference between the corresponding energies of the excited and the ground state. The process is called fluorescence and the relaxation lifetime is on the order of nanoseconds (Figure 3.3).



#### **Figure 3.3. Fluorescence**

Phosphorescence is a similar process, but in this case, the molecule's return to the ground state by the emission of a photon is forbidden. As a result, the molecule has to get to a different state before returning to ground state, so the relaxation lifetimes range from microseconds to hours (Figure 3.4).



**Figure 3.4.** Phosphorescence

A pulsed laser is used as the excitation source for uranium atoms in the kinetic phosphorescence analysis technique. The amount of light emitted by the atoms when returning to the ground state is collected using photomultiplier tubes and then correlated to the uranium concentration in the sample. Since the laser photons interact with the electron shell of an atom, this technique does not distinguish between different isotopes of uranium.

The KPA analysis was performed with a Kinetic Phosphorescence Analyzer model 11 (KPA-11) by Chemchek Instruments\*. The instrument was calibrated in two ranges. A three-point calibration was performed for both low and high ranges (Table 3.1):

| Low Range (µg/L) | High Range (µg/L) |
|------------------|-------------------|
| 1                | 10                |
| 5                | 50                |
| 10               | 100               |

 Table 3.1. Three-Point Calibration for Low and High Ranges on KPA

The resulting calibration is shown on the following Figure:

\*Chemchek Instruments, Inc. 1845 Terminal Dr. Suite #101, Richland, WA 99354



Figure 3.5. Three-Point Calibration for Low and High Ranges on KPA\*

Each sample was measured three times using the KPA-11 and the average concentrations were calculated alongside with the propagated uncertainties. A number of calibration check standards and reagent blanks were analyzed alongside the samples to guarantee the quality control and reproducibility of the measurements.

\*KPAWin Software by Chemchek Instruments, Inc. 1845 Terminal Dr. Suite #101, Richland, WA 99354

#### 3.4. Inductively Coupled Plasma Mass Spectrometry

The second type of analysis employed in this study was Inductively Coupled Plasma Mass Spectrometry. Unlike the KPA method, the ICP MS technique can differentiate between the different isotopes of chemical elements. A sample entering the mass spectrometer is decomposed into ions in high temperature argon plasma. The ions of specific mass-to-charge ratios are then transmitted by the quadrupole mass filter to the detection system, which detects the number of ions transmitted. The detection system comprises a channel electron multiplier with proper amplification facilities. The pulses from the detection system are transmitted to a multi-channel scaler (MSC) data acquisition unit. The instrument is controlled by a PC running special interface Software. The data from the MSC is also transferred to the PC for storage and further analysis (PQ 2 User's Manual).

Hence the four main processes in any Mass Spectrometry Systems are:

- Sample introduction and aerosol generation
- Ionization by an argon plasma source
- Mass discrimination
- Detection of the ions of specific mass-to-charge ratios

The following figure represents this sequence of processes:



Figure 3.6. Outline of ICP MS

(Adopted from Plasma Quad 2 User's Manual)

The apparatus used in this study was a Thermo Xseries 2 ICP MS with integrated autosampler located in the CAMAS laboratory of Idaho State University. The Xseries 2 ICP-MS is a productive quadrupole ICP MS available for routine and high performance analytical work (Thermoscientific). The ease of use and practical design of the instrument guarantees that any analytical objective is achieved faster, with greater confidence and less 'hands-on' time from the operator. The instrument is an ergonomically designed quadrupole ICP MS with a low background specification (Thermoscientific). The low background is achieved by the Protective Ion Extraction and Infinity II ion optics, based upon a hexapole design with an ion deflector. The high performance quadrupole analyzer is pumped by a split flow turbo pump backed by a single

rotary. The simultaneous analog/PC detector with real time multi-channel analyzer electronics provides more than eight orders of dynamic range ensuring the spectrometer is suitable for both steady state and transient signal analysis. The instrument and accessories are entirely PC controlled by PlasmaLab\* software (Thermoscientific).

The instrument was calibrated using two sets of standards. A four-point calibration was performed for each set. The concentrations prepared were  $0.1 \pm 0.01 \mu g/L$ ,  $1 \pm 0.01 \mu g/L$ ,  $10 \pm 0.01 \mu g/L$ , and  $100 \pm 0.01 \mu g/L$  of natural uranium. The calibration curve for U-238 is shown on Figure 3.7.



Figure 3.7. Thermo Xseries 2 Calibration Curve for U-238\*

\*PlasmaLab Software by Thermoscientific. 81 Wyman Street, Waltham, MA 02454
Similarly, to the KPA measurements, triplet analysis of all the samples alongside with the splits and spikes was carried out on the ICP MS. The average concentration of each isotope for each sample was calculated and the uncertainty was propagated. Dixon's Q test was used to reject outliers.

# 3.5. Quality Control

Quality control is used in every kind of analysis to produce credible results. A good quality control program has to consist of the following elements (Standard Methods, 1989):

- Analysis of externally supplied standards
- Analysis of reagent blanks
- Calibration with standards
- Analysis of duplicates
- Recovery of known additions

The externally supplied standards and calibration with standards have already been discussed in this chapter. The following section focuses on the last two elements in the quality control program list.

#### **3.5.1. Split Samples**

To achieve high quality 10% of all the samples were split into A and B aliquants and analyzed as separate samples. The split samples were used to check the overall reproducibility of the analyses. The two tests for reproducibility for the individual pairs of samples are:

- Agreement to within a set percentage
- Agreement to within a set number of standard deviations

Both tests are applied to each pair of the split samples and the pair shall pass at least one of these tests to be judged "in agreement". The advantage of this approach is that even samples with low concentrations can be tested for statistical agreement. The set percentage for acceptability in this study was arbitrarily chosen to be 20% and the set number of standard deviations was arbitrarily chosen to be three (Standard Methods, 1989). The percentage test is considered met only when the absolute difference between the two values of the pair of split samples does not differ by more than 20% from the average of the two values:

$$\frac{\left|C_{1}-C_{2}\right|}{\frac{C_{1}+C_{2}}{2}} \le 0.2 \tag{3.1}$$

Where

 $C_1$  = Uranium concentration measured for A split

 $C_2$  = Uranium concentration measured for B split

The standard deviation test is considered met only when the absolute difference between the two values of the pair of split samples is less than or equal to three times the root mean square of standard deviations of the results:

$$|AD| \le 3 \times \sqrt{\sigma_A^2 + \sigma_B^2} \tag{3.2}$$

Where

AD = Acceptable Difference  $\sigma_A$  = Standard deviation of the A split measurement  $\sigma_B$  = Standard deviation of the B split measurement

#### **3.5.2. Spiked Samples**

Spiked samples are used to check the overall accuracy of analytical methods internally. Spiked aqueous samples of known concentration are prepared from either a NIST or a commercial standard. The spiked samples are then analyzed as unknowns. The two tests for reproducibility for the individual spike samples are:

- Agreement with the known concentration to within a set percentage
- > Agreement with the known concentration to within a set number of standard deviations

Both tests are applied to each spike sample and the sample shall pass at least one of these tests to be judged "in agreement". The advantage of this approach is that even samples with low concentrations can be tested for statistical agreement. In this study, the set percentage was arbitrarily chosen to be 20% and the set number of standard deviations was arbitrarily chosen to be three (Standard Methods, 1989). The criterion used for the spike samples for percentage test is considered met only when the absolute difference between the measured and known values of the spiked sample does not differ by more than 20% from the known value:

$$\frac{\left|C_{Measured} - C_{STD}\right|}{C_{STD}} \le 0.2 \tag{3.3}$$

Where

 $C_{Measured}$  = Measured concentration of the spiked sample

 $C_{STD}$  = Known concentration of the spiked sample

The criterion used for the spike samples for three standard deviations test is identical to that used for the split samples. The standard deviation test is considered met only when the absolute difference between the measured and known values is less than or equal to three times the root mean square of standard deviations of the values:

$$|AD| \le 3 \times \sqrt{\sigma_{Measured}^2 + \sigma_{STD}^2}$$
(3.4)

Where

*AD* = Acceptable Difference

 $\sigma_{Measured}$  = Standard deviation of the measurement value

 $\sigma_{STD}$  = Standard deviation of the known value

(Standard Methods, 1989).

# **3.6. Detection Limit**

Analytical instruments produce a signal even when analyzing a blank sample or no sample at all. This is also called noise. Since any quality control program requires analysis of blanks, the mean and standard deviation of this kind of analyses become well known. Hence the Gaussian curve of the blank distribution becomes well known and ideally narrow.

The lower detection limit (LLD) is defined as the amount of constituent that produces a signal sufficiently large that some pre-established fraction (i.e. 95% or 99%) of the analyses with that test amount will lead to a detectable signal. The LLD is usually determined using following steps (Standard Methods, 1989):

- > Multiple injections of a standard at near zero concentration
- > Determining the standard deviation of the resulted distribution
- Multiplying the determined standard deviation by 1.645 from a cumulative normal probability table to reduce Type I – false positive error to 5%.
- Similarly, to reduce the probability of Type II false negative errors to 5% doubling this number to 3.290

The LLDs were calculated for both instruments. Solutions in concentrations near zero for each instrument were prepared and analyzed at least ten times. The concentrations analyzed on KPA and ICP MS were  $0.15\mu$ g/L and  $1\mu$ g/L respectively. The standard deviation for each set was calculated and the LLD for each instrument was determined by multiplying the respective standard deviation by 3.290.

#### **3.7. Statistical Tests and Error Propagation**

#### 3.7.1. Dixon's Q-test

The Dixon's Q-test was employed to reject outliers for both KPA and ICP MS measurements. This test can be used to examine if one of the measurements from a small set of replicate measurements can be rejected or not. Typically, it is employed for the number of observations between 3 and 10. In case of our triplicate measurements on both KPA and ICP MS the test was appropriate.

The Dixon's Q-test is based on the statistical distribution of subrange ratios of ordered data samples, drawn from the same normal population. Therefore, a normal or Gaussian distribution is presumed when this test is employed.

The steps to apply the test are the following (Ellison et al, 2009):

> The experimentally observed values are arranged in ascending order:

$$x_1 < x_2 < \dots < x_n$$

> The  $Q_{Exp}$  parameter is calculated by dividing the difference of the suspect value from its nearest one by the range of the values:

$$Q_{Exp} = \frac{x_n - x_{n-1}}{x_n - x_1} \tag{3.5}$$

- > The resulted  $Q_{Exp}$  value is compared to a critical  $Q_{Crit}$  value from tables. The critical value corresponds to the confidence level of CL = 95%.
- > In case of  $Q_{Exp} > Q_{Crit}$  the suspect value is identified as an outlier and subsequently rejected.

The  $Q_{Crit}$  in case of n = 3 is 0.97. Every data point in both KPA and ICP MS measurements was tested by the Dixon's Q-test and all the outliers were rejected.

# **3.7.2. Error Propagation**

The standard error propagation technique was used to calculate the uncertainty of the average values for the triplicate measurements. The technique states that in case of

$$y = a(x_1 + x_2 + ... + x_n)$$
 (3.6)

the propagated error  $\sigma_y$  is calculated as follows:

$$\sigma_{y} = a_{\sqrt{\sigma_{x_{1}} + \sigma_{x_{2}} + \dots + \sigma_{x_{n}}}}$$
(3.7)

(Bevington et al, 2003)

In case of triplicate measurements, a = 1/3 and n = 3.

Both KPA and ICP MS report the statistical uncertainty of each separate measurement. The total uncertainty was propagated for each data point for both instrument measurements from the reported uncertainty values.

# **CHAPTER 4**

# **Results and Discussion**

#### 4.1. Introduction

Three hundred and eighty samples from various locations within the State of Idaho were analyzed using KPA and ICP MS. The uranium concentrations were mapped for both cases and the LLDs were compared. The <sup>234</sup>U/<sup>238</sup>U activity ratio was determined for each location and it was shown that these ratios differ from unity, which means that uranium is mobile in the groundwaters in Idaho. Further analysis of the samples previously analyzed by the radiochemical method made it possible to compare LLDs of KPA and ICP MS to the MDC of the radiochemical method of measuring uranium. All the results and discussions are presented in this chapter.

#### 4.2. Uranium Concentrations in Idaho

#### 4.2.1. Total Uranium in Idaho

The total uranium measurements were conducted using KPA in the EML of ISU. All three hundred and eighty samples, thirty one split samples and twenty spiked samples were analyzed for total uranium concentrations. A triplicate analysis was performed for each sample and an average concentration was calculated in each case. The Dixon's Q Test was applied to the data to reject outliers. A total of nine data points was rejected at 95% confidence level using this criterion. The uncertainty for each calculated average concentration was propagated from the uncertainties of each individual measurement reported by KPA. Equation 3.7 was used for these propagations.

The measured concentrations ranged from below the lower level of detection (LLD) to up and over the EPA Safe Drinking Water limit in some cases. Each data point was assigned to one of the three arbitrary ranges. The ranges chosen were:

 $\geq$  Low – 0 - 10µg/L

- ➢ High 10 30µg/L
- $\blacktriangleright$  Over the EPA Limit over  $30\mu g/L$

The reasoning behind choosing  $10\mu g/L$  as the border between the Low and High ranges lies in the operational specifications of KPA. The instrument operates in two ranges and the border point between the ranges is  $10\mu g/L$ . Since uranium concentrations over  $10\mu g/L$  were arbitrarily defined as high concentrations,  $10\mu g/L$  was accepted as the action level.

The resulted concentrations were mapped on the Idaho grid. Different symbols were employed for the different ranges.

Most of the samples analyzed had uranium concentrations below the action level. But twenty of the measured samples exhibited the concentrations over  $10\mu g/L$ . Nineteen out of these twenty samples were collected in the western part of the state and the last one was from northern Idaho.

Figure 4.1 represent the uranium concentrations in Idaho as measured by KPA:

36



Figure 4.1. Total Uranium Concentrations in Idaho Measured by KPA

## 4.2.2. Isotopic Uranium in Idaho

Isotopic uranium measurements were conducted using ICP MS at the CAMAS facility at ISU. Similarly, to the KPA measurements, triplicate measurements were performed in this case for all the original samples, split samples and spiked samples for each of the three uranium isotopes. The Dixon's Q Test was applied to the data to reject outliers. None of the measured ICP MS values were rejected. The instrument estimated and reported a standard deviation ( $\sigma$ ) and percent relative standard deviation (%RSD) for each triplicate measurement.

Analyzing the uranium isotopic ratios showed that all the uranium in the samples was natural (section 4.2). Since <sup>238</sup>U accounts for 99.2742% of the natural uranium (Environmental Protection Agency), a decision was made to map only <sup>238</sup>U for ICP MS measurements.

To be consistent with the methodology used in the KPA analysis, the data from the ICP MS analysis was fractionated into the same three ranges:

- $\blacktriangleright$  Low 0 10µg/L
- > High 10 30µg/L
- $\blacktriangleright$  Over the EPA Limit over  $30\mu g/L$

Concentration of <sup>238</sup>U equal to10µg/L was accepted as the action level for the ICP MS analysis.

Similarly, to the total uranium mapping the resulted concentrations of <sup>238</sup>U were mapped on the Idaho grid. The same symbols used in case of the total uranium mapping were employed for the <sup>238</sup>U mapping.

Figure 4.2 represent the <sup>238</sup>U concentrations in Idaho as measured by ICP MS:



Figure 4.2. <sup>238</sup>U Concentrations in Idaho Measured by ICP MS

It can be noted that the measured uranium concentrations for the two instruments fall in different ranges for some data points. The measurements were in good agreement for the low range data. The results diverted more from each other for the high range measurements. ICP MS measured five data points over the EPA limit, while KPA measured only two data points above that limit. To ensure that the measurements from different instruments did not differ statistically, the set percentage statistical test was applied to the data. Similarly, to the split and spike samples, the set percentage was chosen to be 20% (Standard Methods, 1989).

Table 4.1. represents all the locations in Idaho with the uranium activities either in High or Over the EPA Limit range as measured by at least one of the instruments. The list of all the samples with concentrations above LLD and their measured uranium concentrations can be found in Appendix A.

| Sample ID | County  | Lo   | cation | TU (µg/L) | Error (µg/L) | <sup>238</sup> U (µg/L) | Error (µg/L) |
|-----------|---------|------|--------|-----------|--------------|-------------------------|--------------|
| CG181     | Canyon  | 03 N | 04 W   | 18.315    | 0.382        | 26.870                  | 0.480        |
| CG183     | Canyon  | 03 N | 03 W   | 11.094    | 0.242        | 15.940                  | 0.386        |
| CG186     | Canyon  | 03 N | 02 W   | 33.367    | 0.632        | 43.840                  | 1.281        |
| CG187     | Ada     | 03 N | 01 E   | 25.599    | 0.496        | 33.530                  | 1.372        |
| CG191     | Ada     | 03 N | 01 W   | 11.933    | 0.241        | 17.600                  | 0.484        |
| CG192     | Ada     | 03 N | 02 E   | 14.880    | 0.284        | 21.130                  | 0.660        |
| CG193     | Canyon  | 03 N | 03 W   | 9.086     | 0.720        | 10.170                  | 0.317        |
| CG194     | Canyon  | 03 N | 02 W   | 15.828    | 0.308        | 23.910                  | 0.641        |
| CG196     | Ada     | 03 N | 01 E   | 9.098     | 0.174        | 13.250                  | 0.279        |
| CG200     | Ada     | 04 N | 01 E   | 18.867    | 0.358        | 28.140                  | 0.758        |
| CG203     | Ada     | 04 N | 01 E   | 10.680    | 0.208        | 15.910                  | 0.564        |
| CG220     | Ada     | 04 N | 01 E   | 9.548     | 0.181        | 13.850                  | 0.369        |
| CG221     | Ada     | 04 N | 01 W   | 29.312    | 1.065        | 35.980                  | 0.849        |
| CG222     | Canyon  | 04 N | 05 W   | 17.004    | 1.163        | 20.200                  | 0.547        |
| CG232     | Ada     | 05 N | 01 W   | 11.053    | 0.211        | 14.890                  | 0.219        |
| CG236     | Canyon  | 05 N | 03 W   | 27.342    | 0.700        | 36.610                  | 0.652        |
| CG239     | Canyon  | 05 N | 04 W   | 13.143    | 0.337        | 18.490                  | 0.422        |
| CG260     | Payette | 06 N | 04 W   | 42.494    | 0.780        | 43.880                  | 0.702        |
| CG288     | Payette | 09 N | 05 W   | 10.681    | 0.209        | 11.550                  | 0.242        |
| CG379     | Boundry | 61 N | 01 E   | 11.641    | 0.220        | 13.200                  | 0.403        |

Table 4.1. Elevated Uranium Concentrations in Idaho

## 4.3. Enriched and Depleted Uranium

All three hundred and eighty samples from the Idaho Department of Water Resources were analyzed for isotopic uranium using ICP MS. ICP MS is capable of calculating isotopic ratios between different isotopes. The <sup>234</sup>U/<sup>235</sup>U, <sup>234</sup>U/<sup>238</sup>U, and <sup>235</sup>U/<sup>238</sup>U ratios were calculated for each sample. The <sup>235</sup>U/<sup>238</sup>U ratio is the best source of information for uranium enriched or depleted in <sup>235</sup>U. The <sup>235</sup>U/<sup>238</sup>U ratio for natural uranium is 7.253x10<sup>-3</sup>. Hence uranium depleted in <sup>235</sup>U will exhibit a lower value for this ratio. Similarly, uranium enriched in <sup>235</sup>U will exhibit a higher value for this ratio. As it was discussed in the Section 2.2., every EPA methodology for measuring uranium in water is based on the assumption that all uranium in water is natural. According to these methods <sup>238</sup>U concentration is determined in the medium and the other uranium isotope concentrations are calculated based on the percent isotopic composition of natural uranium. Hence in the presence of depleted or enriched uranium, the EPA methods do not give the true value for the isotopic and total uranium concentrations. It can be shown that an EPA estimate for total uranium concentration can differ from the true value by about 4.5% in the case of 5% enrichment. Up to 5% enrichment is typical for reactor grade uranium fuel. The difference becomes even more extreme for weapons grade uranium which can be enriched to as high as 90%.

According to the ICP MS analysis results, all the samples with uranium concentrations above the LLD exhibit the  ${}^{235}\text{U}/{}^{238}\text{U}$  ratio values between 0.006 and 0.008. Therefore, it was concluded that all the uranium in the analyzed samples is natural and the null hypothesis **H10** was accepted for the first set of the hypothesis. The detailed table of the isotopic ratios for each sample is presented in the Appendix B.

# 4.4. Uranium Activity Ratios

The <sup>234</sup>U/<sup>238</sup>U uranium activity ratios were calculated for each of the three hundred and eighty samples. The values ranged between 0.91 and 6.21, with most of the UARs differing from the unity. Each data point was assigned to one of the three arbitrary ranges. The ranges chosen were: Low (UAR < 2), Medium (2 < UAR < 4), and High (UAR > 4). The resulted values were mapped on the county map of Idaho. Different symbols were employed for the different ranges, where circles correspond to the low-level values, diamonds correspond to the medium level values, and squares correspond to the high level values.

Majority of the samples analyzed had uranium concentrations within the low range. But seventy of the measured samples exhibited the UARs between 2 and 4. Only six samples were found to have UARs higher than 4.

Figure 4.3 shows the analyses results mapped on the Idaho grid and Table 4.2 lists the six samples in the high range:



Figure 4.3. UARs in Groundwater in Idaho

| Sample ID | County   | Location | Ì    | <sup>235</sup> U/ <sup>238</sup> U | %RSD | UAR  | %RSD  |
|-----------|----------|----------|------|------------------------------------|------|------|-------|
| CG007     | Franklin | 15 S     | 40 E | 0.006                              | 0.99 | 4.54 | 0.22  |
| CG011A    | Oneida   | 15 S     | 35 E | 0.006                              | 0.76 | 5.45 | 6.42  |
| CG011B    | Oneida   | 15 S     | 35 E | 0.006                              | 0.96 | 5.58 | 1.43  |
| CG056     | Bannock  | 09 S     | 38 E | 0.006                              | 1.30 | 6.23 | 13.48 |
| CG138     | Camas    | 01 S     | 15 E | 0.006                              | 3.05 | 4.28 | 3.04  |
| CG142     | Owyhee   | 01 S     | 03 W | 0.006                              | 2.44 | 4.28 | 6.31  |
| CG345     | Benewah  | 44 N     | 03 W | 0.007                              | 0.28 | 6.21 | 3.86  |

Table 4.2. Samples with high UARs

Hence it was concluded that <sup>238</sup>U is not in equilibrium with its daughter product <sup>234</sup>U in the groundwaters in the State of Idaho and the alternative hypothesis **H2A** was accepted for the second set of hypothesis. The detailed table of the UARs for all three hundred and eighty samples is presented in Appendix B.

## **4.5.** Lower Limits of Detection

#### 4.5.1. Determining the LLDs for KPA and ICP MS

The lower limits of detection were determined for KPA and ICP MS. Near zero uranium concentrations were analyzed on each instrument a number of times and the LLDs were determined as described in Section 3.6.

By analyzing multiple low concentration standard solutions on KPA it was determined that the amount of uranium concentration producing a signal that could be distinguished from the noise was around  $0.15\mu$ g/L. Similarly, analyzing near zero uranium concentrations for ICP MS proved 1µg /L to be the concentration producing a signal just above the noise. In the case of KPA, the standard solution was analyzed ten times and LLD was determined (KPA User's Manual 2006). The standard solution for ICP MS was analyzed thirty five times for sufficient statistics and the LLD was determined in the similar manner. The following table represents the analyses results for both instruments and estimated LLDs:

|         | KPA        |       |            |       |            |       |            |       |
|---------|------------|-------|------------|-------|------------|-------|------------|-------|
|         | # of trial | μg/L  |
|         | 1          | 0.196 | 1          | 1.063 | 13         | 0.900 | 25         | 0.933 |
|         | 2          | 0.164 | 2          | 1.175 | 14         | 0.834 | 26         | 0.894 |
|         | 3          | 0.111 | 3          | 1.108 | 15         | 0.971 | 27         | 0.992 |
|         | 4          | 0.157 | 4          | 1.114 | 16         | 0.867 | 28         | 0.965 |
|         | 5          | 0.192 | 5          | 1.123 | 17         | 0.845 | 29         | 0.957 |
|         | 6          | 0.154 | 6          | 1.337 | 18         | 0.994 | 30         | 1.026 |
|         | 7          | 0.187 | 7          | 1.303 | 19         | 0.905 | 31         | 0.997 |
|         | 8          | 0.112 | 8          | 1.091 | 20         | 0.880 | 32         | 0.924 |
|         | 9          | 0.148 | 9          | 1.388 | 21         | 0.980 | 33         | 1.012 |
|         | 10         | 0.111 | 10         | 1.405 | 22         | 0.929 | 34         | 0.972 |
|         |            |       | 11         | 1.409 | 23         | 0.863 | 35         | 0.967 |
|         |            |       | 12         | 0.989 | 24         | 1.013 |            |       |
| Average |            | 0.153 |            |       |            | 1.032 |            |       |
| σ       |            | 0.033 |            |       |            | 0.162 |            |       |
| LLD     |            | 0.099 |            |       |            | 0.532 |            |       |

Table 4.3. Lower Levels of Detection for KPA and ICP MS

#### 4.5.2. LLDs for KPA and ICP MS vs. Radiochemical Analysis Technique

The calculated LLDs for KPA and ICP MS were compared to that achieved by the. radiochemical method. The Environmental Monitoring Laboratory of ISU does not perform this type of radiochemical analysis. The samples that require this kind of analysis are shipped to a vendor laboratory\*. After analyzing the samples using the radiochemical Method (RCM), the vendor laboratory reports the analyses results and minimum detectable concentration for each sample to EML of ISU.

A total of five samples previously analyzed for isotopic uranium concentrations at the vendor laboratory were reanalyzed at EML and CAMAS using KPA and ICP MS respectively. The radiochemical analysis results were reported to EML in the units of in pCi/L. Taking into account that the specific activities for <sup>234</sup>U, <sup>235</sup>U, and <sup>238</sup>U are 2.313x10<sup>8</sup>Bq/g, 8.001x10<sup>4</sup>Bq/g 1.2445x10<sup>4</sup>Bq/g respectively, the radiochemical analysis results were converted into µg/L. The original and converted results are presented in the following tables (EML Data and Quality Assurance Report, 2007):

| <sup>234</sup> U (pCi/L) | σ (pCi/L)  | MDC (pCi/L)  | <sup>234</sup> U (µg/L)  | σ (µg/L)   | MDC (µg/L)  |
|--------------------------|--|--|--|--|---|
| 1.970                    | 0.215  | 0.160  | 3.15x10 <sup>-4</sup>  | 3.44x10 <sup>-5</sup>  | 2.56x10 <sup>-5</sup>   |
| 2.360                    | 0.235  | 0.070  | 3.78x10 <sup>-4</sup>  | 3.76x10 <sup>-5</sup>  | 1.12x10 <sup>-5</sup>   |
| 1.770                    | 0.190  | 0.070  | 2.83x10 <sup>-4</sup>  | 3.04x10 <sup>-5</sup>  | 1.12x10 <sup>-5</sup>   |
| 0.660                    | 0.095  | 0.100  | 1.06x10 <sup>-4</sup>  | 1.52x10 <sup>-5</sup>  | $1.60 \times 10^{-5}$   |
| 1.550                    | 0.175  | 0.110  | 2.48x10 <sup>-4</sup>  | 2.80x10 <sup>-5</sup>  | 1.76x10 <sup>-5</sup>   |
|                          | <ul> <li><sup>234</sup>U (pCi/L)</li> <li>1.970</li> <li>2.360</li> <li>1.770</li> <li>0.660</li> <li>1.550</li> </ul> | 234U (pCi/L)         σ (pCi/L)           1.970         0.215           2.360         0.235           1.770         0.190           0.660         0.095           1.550         0.175 | 234U (pCi/L)         σ (pCi/L)         MDC (pCi/L)           1.970         0.215         0.160           2.360         0.235         0.070           1.770         0.190         0.070           0.660         0.095         0.100           1.550         0.175         0.110 | $^{234}$ U (pCi/L) $\sigma$ (pCi/L)MDC (pCi/L) $^{234}$ U (µg/L)1.9700.2150.160 $3.15x10^{-4}$ 2.3600.2350.070 $3.78x10^{-4}$ 1.7700.1900.070 $2.83x10^{-4}$ 0.6600.0950.100 $1.06x10^{-4}$ 1.5500.1750.110 $2.48x10^{-4}$ | $^{234}$ U (pCi/L) $\sigma$ (pCi/L)MDC (pCi/L) $^{234}$ U (µg/L) $\sigma$ (µg/L)1.9700.2150.160 $^{3.15x10^{-4}}$ $^{3.44x10^{-5}}$ 2.3600.2350.070 $^{3.78x10^{-4}}$ $^{3.76x10^{-5}}$ 1.7700.1900.070 $^{2.83x10^{-4}}$ $^{3.04x10^{-5}}$ 0.6600.0950.100 $^{1.06x10^{-4}}$ $^{1.52x10^{-5}}$ 1.5500.1750.110 $^{2.48x10^{-4}}$ $^{2.80x10^{-5}}$ |

Table 4.4. <sup>234</sup>U concentrations as measured by Radiochemical Analysis Technique

\*Paragon Analytics, 225 Commerce Drive, Fort Collins, CO 80524

| ISU ID  | <sup>235</sup> U (pCi/L) | σ (pCi/L) | MDC (pCi/L) | <sup>235</sup> U (µg/L) | σ (µg/L) | MDC (µg/L) |
|---------|--------------------------|-----------|-------------|-------------------------|----------|------------|
| RS33917 | 0.071                    | 0.036     | 0.109       | 0.032                   | 0.016    | 0.05       |
| RS33918 | 0.042                    | 0.021     | 0.028       | 0.019                   | 0.01     | 0.013      |
| RS33919 | 0.027                    | 0.022     | 0.066       | 0.012                   | 0.01     | 0.03       |
| RS34060 | 0.039                    | 0.024     | 0.067       | 0.018                   | 0.011    | 0.031      |
| RS34061 | 0.026                    | 0.021     | 0.064       | 0.012                   | 0.009    | 0.029      |

Table 4.5. <sup>235</sup>U concentrations as measured by Radiochemical Analysis Technique

Table 4.6. <sup>238</sup>U concentrations as measured by Radiochemical Analysis Technique

| ISU ID  | <sup>238</sup> U (pCi/L) | σ (pCi/L) | MDC (pCi/L) | <sup>238</sup> U (µg/L) | σ (μg/L) | MDC (µg/L) |
|---------|--------------------------|-----------|-------------|-------------------------|----------|------------|
| RS33917 | 1.400                    | 0.165     | 0.110       | 4.162                   | 0.491    | 0.327      |
| RS33918 | 0.970                    | 0.120     | 0.070       | 2.884                   | 0.357    | 0.208      |
| RS33919 | 0.690                    | 0.100     | 0.030       | 2.051                   | 0.297    | 0.089      |
| RS34060 | 0.380                    | 0.070     | 0.060       | 1.130                   | 0.208    | 0.178      |
| RS34061 | 0.800                    | 0.105     | 0.050       | 2.378                   | 0.312    | 0.149      |
|         |                          |           |             |                         |          |            |

It can be noted that the radiochemical analysis technique is capable of measuring much lower concentrations than KPA and ICP MS. Hence, the null hypothesis **H30** and the null hypothesis **H40** were accepted for the third and the fourth sets of the hypothesis.

Although it was established that radiochemical analysis technique has a lower MDC when compared to KPA and ICP MS, the latter two methods have their advantages over the

radiochemical method. Both these instruments have a large sample throughput and take one to two minutes per sample analysis, while radiochemical method involves a chemical treatment of the samples which could take up to a few days. Also, higher LLDs for KPA and ICP MS can be justified by the lower cost of the sample analyses by these methods.

# 4.6. Quality Control

Thirty one samples were split and a number of spiked samples were prepared for quality control purposes. The criteria applied to the split and spiked samples were three standard deviation and 20% set percentage difference as explained in Sections 3.5.1. and 3.5.2. All the split samples except of one that had uranium concentrations above the LLD were "in agreement" when analyzed using KPA. The failed split sample was reanalyzed and it resulted in "in agreement". All the samples that had uranium concentrations above the LLD passed the split test in the case of ICP MS.

Every spiked sample was "in agreement" for both KPA and ICP MS measurements.

The detailed tables of all the split and spiked samples are given in Appendix C.

# **CHAPTER 5**

# **Conclusions and Future Work**

#### **5.1.** Conclusions

A complete investigation of uranium concentration distributions was conducted for the groundwater in the State of Idaho. It was shown that most of the state has total uranium concentrations well below the EPA Safe Drinking Water Limit. But some parts of the state (western and northern) exhibit elevated concentrations in uranium.

No enriched or depleted uranium was found during the study. Even the locations with elevated uranium concentrations have the  ${}^{235}\text{U}/{}^{238}\text{U}$  ratios that are in the natural uranium range.

Uranium activity ratios were calculated for the  ${}^{234}U/{}^{238}U$  ratio and it was concluded that the parent  ${}^{238}U$  is not in equilibrium with its daughter product  ${}^{234}U$ . Therefore, uranium is mobile in the groundwaters in Idaho.

Comparison of the methods used in this study (KPA and ICP MS) to the radiochemical analysis technique showed that the latter is capable of detecting lower concentrations of uranium. But KPA and ICP MS are much faster and more cost effective methodologies.

Isotopic analysis of uranium using ICP MS showed that this technique is capable of detecting the  $^{238}$ U isotope down to about 0.5µg/L in environmental samples.

Also, a novel methodology for determining the isotopic and total uranium concentrations by directly measuring <sup>235</sup>U concentration was proposed. This method could account for the presence of depleted or enriched uranium, unlike the EPA methods that assume natural uranium in the medium. Furthermore, the study results suggest that although KPA is not one of the EPA standard methods for measuring uranium concentrations, it can readily be employed to detect total uranium concentrations as low as  $0.1\mu g/L$  in environmental samples. Hence building an environmental assessment program for total uranium concentrations can be successfully achieved by this method.

# 5.2. Future Work

Some of the future work of this project may involve:

- Building a comprehensive, cost effective and readily available environmental assessment program for uranium using KPA and ICP MS
- Modeling uranium migration in the groundwater in Idaho based on the <sup>234</sup>U/<sup>238</sup>U activity ratios
- > Investigating uranium concentrations and activity ratios in surface water and soil
- Investigating uranium concentrations and activity ratios inside Idaho National Laboratory
- Investigating other long term contaminants (i.e. <sup>127</sup>I, etc.) using the ICP MS technique

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# **APPENDIX** A

| Sample ID | County     | Loc  | ation | Total U(µg/L) | σ (μg/L) |
|-----------|------------|------|-------|---------------|----------|
| CG001     | Bear Lake  | 16 S | 43 E  | 1.186         | 0.013    |
| CG002     | Oneida     | 16 S | 32 E  | 0.250         | 0.005    |
| CG003     | Oneida     | 16 S | 36 E  | 1.992         | 0.039    |
| CG005     | Franklin   | 16 S | 39 E  | 0.878         | 0.034    |
| CG006     | Bear Lake  | 15 S | 44 E  | 3.165         | 0.043    |
| CG007     | Franklin   | 15 S | 40 E  | 1.602         | 0.019    |
| CG008     | Oneida     | 15 S | 36 E  | 1.767         | 0.020    |
| CG009     | Franklin   | 15 S | 40 E  | 1.740         | 0.019    |
| CG010     | Oneida     | 15 S | 36 E  | 0.569         | 0.009    |
| CG011A    | Oneida     | 15 S | 35 E  | 0.480         | 0.014    |
| CG011B    | Oneida     | 15 S | 35 E  | 0.499         | 0.012    |
| CG012A    | Franklin   | 14 S | 38 E  | 1.179         | 0.012    |
| CG012B    | Franklin   | 14 S | 38 E  | 1.005         | 0.017    |
| CG013     | Cassia     | 14 S | 25 E  | 3.308         | 0.031    |
| CG014     | Oneida     | 14 S | 36 E  | 1.164         | 0.012    |
| CG015     | Cassia     | 14 S | 27 E  | 0.843         | 0.017    |
| CG016     | Bear Lake  | 14 S | 43 E  | 0.868         | 0.008    |
| CG017     | Bear Lake  | 14 S | 44 E  | 1.054         | 0.018    |
| CG018     | Oneida     | 14 S | 35 E  | 0.992         | 0.021    |
| CG019     | Cassia     | 13 S | 25 E  | 2.297         | 0.051    |
| CG020     | Oneida     | 13 S | 36 E  | 0.996         | 0.016    |
| CG021     | Bear Lake  | 13 S | 43 E  | 0.390         | 0.004    |
| CG023     | Cassia     | 12 S | 28 E  | 1.592         | 0.028    |
| CG024     | Bear Lake  | 12 S | 43 E  | 4.209         | 0.051    |
| CG025     | Twin Falls | 12 S | 18 E  | 2.975         | 0.026    |
| CG026A    | Franklin   | 12 S | 40 E  | 0.981         | 0.009    |
| CG026B    | Franklin   | 12 S | 40 E  | 0.844         | 0.008    |
| CG027A    | Bannock    | 12 S | 36 E  | 0.507         | 0.008    |
| CG027B    | Bannock    | 12 S | 36 E  | 0.545         | 0.009    |
| CG028     | Twin Falls | 11 S | 16 E  | 2.938         | 0.082    |
| CG029     | Bannock    | 12 S | 36 E  | 0.704         | 0.011    |
| CG030     | Cassia     | 11 S | 22 E  | 2.069         | 0.063    |
| CG032     | Cassia     | 11 S | 23 E  | 1.721         | 0.027    |
| CG033     | Cassia     | 10 S | 26 E  | 1.120         | 0.021    |
| CG034     | Twin Falls | 11 S | 18 E  | 4.154         | 0.076    |
| CG035     | Twin Falls | 10 S | 16 E  | 2.410         | 0.128    |
| CG036     | Cassia     | 10 S | 22 E  | 3.601         | 0.095    |

Total Uranium Concentrations as Measured by KPA

| Sample ID | County     | Loc  | ation | Total U(µg/L) | σ (µg/L) |
|-----------|------------|------|-------|---------------|----------|
| CG037     | Twin Falls | 10 S | 17 E  | 4.348         | 0.112    |
| CG038     | Twin Falls | 10 S | 13 E  | 1.400         | 0.036    |
| CG039     | Cassia     | 10 S | 22 E  | 1.469         | 0.049    |
| CG040     | Twin Falls | 10 S | 16 E  | 0.836         | 0.033    |
| CG041     | Twin Falls | 10 S | 13 E  | 0.101         | 0.002    |
| CG042     | Cassia     | 10 S | 24 E  | 1.773         | 0.020    |
| CG043     | Twin Falls | 10 S | 16 E  | 3.546         | 0.083    |
| CG044     | Minidoka   | 10 S | 23 E  | 3.452         | 0.078    |
| CG045     | Minidoka   | 10 S | 24 E  | 1.910         | 0.034    |
| CG046     | Jerome     | 10 S | 21 E  | 3.258         | 0.102    |
| CG047     | Cassia     | 10 S | 25 E  | 0.591         | 0.016    |
| CG048     | Minidoka   | 10 S | 23 E  | 0.553         | 0.004    |
| CG049     | Caribou    | 10 S | 41 E  | 0.635         | 0.011    |
| CG050     | Cassia     | 10 S | 27 E  | 0.364         | 0.012    |
| CG051A    | Twin Falls | 09 S | 19 E  | 2.777         | 0.075    |
| CG051B    | Twin Falls | 09 S | 19 E  | 3.155         | 0.075    |
| CG052A    | Twin Falls | 09 S | 17 E  | 6.979         | 0.337    |
| CG052B    | Twin Falls | 09 S | 17 E  | 6.268         | 0.322    |
| CG054     | Caribou    | 09 S | 42 E  | 1.482         | 0.038    |
| CG055     | Minidoka   | 09 S | 22 E  | 2.393         | 0.067    |
| CG056     | Bannock    | 09 S | 38 E  | 1.079         | 0.013    |
| CG057     | Twin Falls | 09 S | 16 E  | 4.897         | 0.169    |
| CG058     | Cassia     | 09 S | 26 E  | 1.773         | 0.050    |
| CG059     | Minidoka   | 09 S | 23 E  | 0.579         | 0.011    |
| CG060     | Caribou    | 09 S | 40 E  | 0.959         | 0.033    |
| CG061     | Caribou    | 09 S | 41 E  | 0.531         | 0.008    |
| CG062     | Jerome     | 09 S | 20 E  | 1.193         | 0.046    |
| CG063     | Power      | 09 S | 34 E  | 1.258         | 0.035    |
| CG065     | Minidoka   | 09 S | 23 E  | 1.236         | 0.024    |
| CG066     | Twin Falls | 09 S | 14 E  | 5.809         | 0.299    |
| CG067     | Blaine     | 09 S | 27 E  | 1.494         | 0.028    |
| CG068     | Minidoka   | 08 S | 24 E  | 2.119         | 0.048    |
| CG069     | Twin Falls | 08 S | 12 E  | 2.402         | 0.030    |
| CG070     | Jerome     | 08 S | 17 E  | 0.820         | 0.019    |
| CG071     | Minidoka   | 08 S | 24 E  | 1.119         | 0.044    |
| CG072     | Jerome     | 08 S | 18 E  | 1.387         | 0.033    |
| CG074     | Power      | 08 S | 30 E  | 2.251         | 0.051    |
| CG075     | Minidoka   | 08 S | 25 E  | 1.914         | 0.030    |
| CG076     | Blaine     | 08 S | 26 E  | 2.487         | 0.212    |
| CG077     | Caribou    | 08 S | 39 E  | 0.252         | 0.018    |
| CG078     | Caribou    | 08 S | 39 E  | 0.701         | 0.009    |
| CG079     | Gooding    | 08 S | 14 E  | 1.624         | 0.017    |

| Sample ID | County   | Loc  | ation | Total U(µg/L) | σ (μg/L) |
|-----------|----------|------|-------|---------------|----------|
| CG080     | Minidoka | 08 S | 24 E  | 1.288         | 0.046    |
| CG081     | Lincoln  | 07 S | 21 E  | 3.649         | 0.410    |
| CG082A    | Power    | 07 S | 31 E  | 6.777         | 0.520    |
| CG082B    | Power    | 07 S | 31 E  | 6.489         | 0.475    |
| CG083     | Power    | 07 S | 30 E  | 3.669         | 0.292    |
| CG084     | Caribou  | 07 S | 41 E  | 0.287         | 0.039    |
| CG085     | Power    | 07 S | 29 E  | 1.200         | 0.042    |
| CG086     | Caribou  | 07 S | 38 E  | 1.967         | 0.069    |
| CG087     | Bannock  | 07 S | 36 E  | 0.608         | 0.013    |
| CG088     | Bannock  | 07 S | 35 E  | 2.389         | 0.300    |
| CG090     | Owyhee   | 07 S | 06 E  | 0.859         | 0.008    |
| CG091     | Lincoln  | 07 S | 23 E  | 1.194         | 0.010    |
| CG092     | Jerome   | 07 S | 17 E  | 1.373         | 0.010    |
| CG093     | Gooding  | 07 S | 13 E  | 2.434         | 0.020    |
| CG094     | Caribou  | 07 S | 41 E  | 1.647         | 0.020    |
| CG095     | Minidoka | 06 S | 24 E  | 1.129         | 0.013    |
| CG096     | Lincoln  | 06 S | 20 E  | 1.277         | 0.016    |
| CG097     | Power    | 06 S | 29 E  | 2.426         | 0.217    |
| CG098     | Power    | 06 S | 32 E  | 2.688         | 0.387    |
| CG099     | Owyhee   | 06 S | 05 E  | 9.327         | 0.460    |
| CG101     | Power    | 06 S | 33 E  | 1.523         | 0.061    |
| CG102     | Power    | 06 S | 32 E  | 1.358         | 0.022    |
| CG103     | Bannock  | 06 S | 34 E  | 3.014         | 0.159    |
| CG104     | Power    | 06 S | 34 E  | 2.363         | 0.301    |
| CG105     | Gooding  | 05 S | 15 E  | 5.588         | 0.231    |
| CG108     | Elmore   | 05 S | 10 E  | 2.079         | 0.086    |
| CG109     | Owyhee   | 05 S | 03 E  | 8.742         | 0.361    |
| CG110     | Lincoln  | 05 S | 17 E  | 4.705         | 0.030    |
| CG112     | Lincoln  | 04 S | 16 E  | 2.442         | 0.019    |
| CG113     | Lincoln  | 04 S | 19 E  | 5.105         | 0.041    |
| CG115     | Lincoln  | 04 S | 20 E  | 2.328         | 0.025    |
| CG116A    | Elmore   | 04 S | 07 E  | 2.204         | 0.255    |
| CG116B    | Elmore   | 04 S | 07 E  | 2.050         | 0.184    |
| CG117A    | Elmore   | 04 S | 07 E  | 3.070         | 0.306    |
| CG117B    | Elmore   | 04 S | 07 E  | 3.518         | 0.417    |
| CG118     | Elmore   | 04 S | 07 E  | 2.326         | 0.149    |
| CG119     | Bingham  | 03 S | 34 E  | 4.458         | 0.124    |
| CG120     | Elmore   | 03 S | 06 E  | 0.680         | 0.107    |
| CG121     | Bingham  | 02 S | 34 E  | 3.973         | 0.627    |
| CG122     | Bingham  | 02 S | 33 E  | 5.619         | 1.044    |
| CG123     | Bingham  | 02 S | 36 E  | 3.199         | 0.385    |
| CG124     | Bingham  | 02 S | 35 E  | 1.433         | 0.169    |

| Sample ID | County     | Loc  | ation | Total U(µg/L) | σ (μg/L) |
|-----------|------------|------|-------|---------------|----------|
| CG125     | Bingham    | 02 S | 35 E  | 1.598         | 0.188    |
| CG126A    | Bingham    | 02 S | 34 E  | 3.203         | 0.038    |
| CG126B    | Bingham    | 02 S | 34 E  | 2.905         | 0.034    |
| CG127A    | Blaine     | 02 S | 29 E  | 3.152         | 0.028    |
| CG127B    | Blaine     | 02 S | 29 E  | 3.327         | 0.029    |
| CG128     | Bingham    | 02 S | 33 E  | 2.971         | 0.048    |
| CG129     | Blaine     | 02 S | 21 E  | 4.361         | 0.034    |
| CG130     | Elmore     | 02 S | 05 E  | 6.438         | 0.057    |
| CG131     | Blaine     | 01 S | 21 E  | 3.476         | 0.026    |
| CG132     | Owyhee     | 01 S | 02 W  | 2.128         | 0.017    |
| CG133     | Bingham    | 01 S | 37 E  | 3.048         | 0.027    |
| CG134     | Elmore     | 01 S | 05 E  | 2.365         | 0.018    |
| CG135     | Bingham    | 01 S | 34 E  | 2.869         | 0.034    |
| CG136     | Bingham    | 01 S | 37 E  | 4.718         | 0.039    |
| CG137     | Camas      | 01 S | 15 E  | 2.156         | 0.018    |
| CG138     | Camas      | 01 S | 15 E  | 2.211         | 0.018    |
| CG139     | Blaine     | 01 S | 20 E  | 4.820         | 0.040    |
| CG140     | Bingham    | 01 S | 37 E  | 3.396         | 0.044    |
| CG141     | Canyon     | 01 S | 02 W  | 7.681         | 0.083    |
| CG142     | Owyhee     | 01 S | 03 W  | 2.934         | 0.024    |
| CG143     | Blaine     | 01 S | 19 E  | 3.547         | 0.028    |
| CG144     | Camas      | 01 N | 13 E  | 2.187         | 0.017    |
| CG145     | Canyon     | 01 N | 02 W  | 3.166         | 0.030    |
| CG146     | Elmore     | 01 N | 04 E  | 3.046         | 0.027    |
| CG147     | Blaine     | 01 N | 18 E  | 4.177         | 0.032    |
| CG149     | Bingham    | 01 N | 35 E  | 3.039         | 0.029    |
| CG150     | Bonneville | 01 N | 44 E  | 2.425         | 0.019    |
| CG151A    | Camas      | 01 N | 14 E  | 0.730         | 0.006    |
| CG151B    | Camas      | 01 N | 14 E  | 0.717         | 0.005    |
| CG152A    | Bonneville | 01 N | 37 E  | 1.390         | 0.012    |
| CG152B    | Bonneville | 01 N | 37 E  | 1.476         | 0.019    |
| CG153     | Bonneville | 01 N | 43 E  | 0.638         | 0.010    |
| CG154     | Bonneville | 01 N | 38 E  | 1.794         | 0.026    |
| CG155     | Ada        | 01 N | 01 E  | 3.319         | 0.204    |
| CG156     | Blaine     | 01 N | 19 E  | 1.426         | 0.018    |
| CG157     | Canyon     | 02 N | 02 W  | 1.366         | 0.012    |
| CG158     | Ada        | 02 N | 01 W  | 9.039         | 0.066    |
| CG159     | Canyon     | 04 N | 04 W  | 5.689         | 0.040    |
| CG160     | Ada        | 02 N | 03 E  | 2.045         | 0.014    |
| CG161     | Ada        | 02 N | 01 E  | 4.698         | 0.033    |
| CG162     | Ada        | 02 N | 01 E  | 5.170         | 0.070    |
| CG163     | Canyon     | 02 N | 02 W  | 8.157         | 0.058    |

| Sample ID | County     | Loc  | ation | Total U(µg/L) | σ (μg/L) |
|-----------|------------|------|-------|---------------|----------|
| CG164     | Canyon     | 02 N | 02 W  | 4.597         | 0.033    |
| CG165     | Bonneville | 02 N | 38 E  | 1.060         | 0.036    |
| CG166     | Bonneville | 02 N | 39 E  | 3.311         | 0.611    |
| CG167     | Owyhee     | 02 N | 05 W  | 7.354         | 0.075    |
| CG168     | Owyhee     | 02 N | 04 W  | 10.661        | 0.759    |
| CG169     | Canyon     | 02 N | 03 W  | 5.426         | 0.151    |
| CG170     | Ada        | 02 N | 01 E  | 0.503         | 0.008    |
| CG172     | Canyon     | 02 N | 03 W  | 11.087        | 0.209    |
| CG174     | Ada        | 02 N | 01 W  | 9.565         | 0.187    |
| CG175     | Ada        | 02 N | 03 E  | 1.268         | 0.014    |
| CG176A    | Bonneville | 03 N | 38 E  | 0.995         | 0.015    |
| CG176B    | Bonneville | 03 N | 38 E  | 1.087         | 0.026    |
| CG178     | Ada        | 03 N | 03 E  | 5.910         | 0.041    |
| CG179     | Canyon     | 03 N | 02 W  | 9.262         | 0.181    |
| CG180     | Ada        | 03 N | 02 E  | 4.256         | 0.030    |
| CG181     | Canyon     | 03 N | 04 W  | 18.315        | 0.382    |
| CG182     | Canyon     | 03 N | 03 W  | 5.288         | 0.048    |
| CG183     | Canyon     | 03 N | 03 W  | 11.094        | 0.242    |
| CG184     | Boise      | 03 N | 04 E  | 0.755         | 0.038    |
| CG185     | Canyon     | 03 N | 01 W  | 8.194         | 0.102    |
| CG186     | Canyon     | 03 N | 02 W  | 33.367        | 0.632    |
| CG187     | Ada        | 03 N | 01 E  | 25.599        | 0.496    |
| CG188     | Ada        | 03 N | 01 E  | 6.720         | 0.057    |
| CG189     | Canyon     | 03 N | 03 W  | 4.655         | 0.033    |
| CG191     | Ada        | 03 N | 01 W  | 11.933        | 0.241    |
| CG192     | Ada        | 03 N | 02 E  | 14.880        | 0.284    |
| CG193     | Canyon     | 03 N | 03 W  | 9.086         | 0.720    |
| CG194     | Canyon     | 03 N | 02 W  | 15.828        | 0.308    |
| CG195     | Ada        | 03 N | 02 E  | 4.978         | 0.037    |
| CG196     | Ada        | 03 N | 01 E  | 9.098         | 0.174    |
| CG197     | Jefferson  | 03 N | 40 E  | 1.060         | 0.149    |
| CG198     | Canyon     | 03 N | 02 W  | 0.654         | 0.037    |
| CG199     | Jefferson  | 04 N | 37 E  | 1.101         | 0.227    |
| CG200     | Ada        | 04 N | 01 E  | 18.867        | 0.358    |
| CG201A    | Jefferson  | 04 N | 39 E  | 0.876         | 0.190    |
| CG201B    | Jefferson  | 04 N | 39 E  | 0.754         | 0.061    |
| CG203     | Ada        | 04 N | 01 E  | 10.680        | 0.208    |
| CG204     | Blaine     | 04 N | 18 E  | 0.249         | 0.060    |
| CG205     | Canyon     | 04 N | 03 W  | 1.171         | 0.015    |
| CG206     | Jefferson  | 04 N | 37 E  | 1.177         | 0.189    |
| CG207     | Boise      | 04 N | 03 E  | 0.898         | 0.010    |
| CG208     | Canyon     | 04 N | 02 W  | 3.804         | 0.452    |

| Sample ID | County    | Loc  | ation | Total U(µg/L) | σ (μg/L) |
|-----------|-----------|------|-------|---------------|----------|
| CG209     | Canyon    | 04 N | 02 W  | 0.567         | 0.086    |
| CG210     | Blaine    | 04 N | 26 E  | 2.218         | 0.018    |
| CG211     | Canyon    | 04 N | 03 W  | 2.751         | 0.019    |
| CG213     | Canyon    | 04 N | 05 W  | 3.054         | 0.021    |
| CG214     | Ada       | 04 N | 01 W  | 1.821         | 0.101    |
| CG215     | Teton     | 04 N | 44 E  | 0.873         | 0.086    |
| CG216     | Ada       | 04 N | 01 E  | 1.031         | 0.020    |
| CG217     | Blaine    | 04 N | 17 E  | 1.928         | 0.020    |
| CG218     | Canyon    | 04 N | 02 W  | 0.396         | 0.047    |
| CG219     | Canyon    | 04 N | 03 W  | 0.300         | 0.054    |
| CG220     | Ada       | 04 N | 01 E  | 9.548         | 0.181    |
| CG221     | Ada       | 04 N | 01 W  | 29.312        | 1.065    |
| CG222     | Canyon    | 04 N | 05 W  | 17.004        | 1.163    |
| CG223     | Madison   | 05 N | 40 E  | 0.825         | 0.008    |
| CG224     | Jefferson | 05 N | 38 E  | 0.776         | 0.049    |
| CG225     | Canyon    | 05 N | 02 W  | 0.737         | 0.006    |
| CG226A    | Canyon    | 05 N | 05 W  | 4.428         | 0.032    |
| CG226B    | Canyon    | 05 N | 05 W  | 4.860         | 0.054    |
| CG227A    | Canyon    | 04 N | 02 W  | 4.160         | 0.033    |
| CG227B    | Canyon    | 04 N | 02 W  | 4.134         | 0.030    |
| CG228     | Ada       | 05 N | 01 E  | 3.374         | 0.174    |
| CG230     | Ada       | 05 N | 02 E  | 1.390         | 0.011    |
| CG231     | Ada       | 05 N | 01 E  | 3.106         | 0.022    |
| CG232     | Ada       | 05 N | 01 W  | 11.053        | 0.211    |
| CG233     | Butte     | 05 N | 26 E  | 1.587         | 0.011    |
| CG236     | Canyon    | 05 N | 03 W  | 27.342        | 0.700    |
| CG237     | Teton     | 05 N | 46 E  | 1.093         | 0.008    |
| CG238     | Madison   | 05 N | 39 E  | 1.077         | 0.068    |
| CG239     | Canyon    | 05 N | 04 W  | 13.143        | 0.337    |
| CG240     | Ada       | 05 N | 01 W  | 2.990         | 0.025    |
| CG241     | Canyon    | 05 N | 04 W  | 4.793         | 0.039    |
| CG242     | Madison   | 05 N | 41 E  | 3.894         | 0.574    |
| CG243     | Teton     | 05 N | 45 E  | 2.740         | 0.019    |
| CG244     | Butte     | 05 N | 26 E  | 1.784         | 0.013    |
| CG245     | Canyon    | 05 N | 03 W  | 6.217         | 0.526    |
| CG246     | Butte     | 05 N | 30 E  | 1.611         | 0.124    |
| CG247     | Jefferson | 05 N | 35 E  | 2.546         | 0.429    |
| CG248     | Madison   | 05 N | 39 E  | 1.134         | 0.269    |
| CG249     | Jefferson | 06 N | 38 E  | 1.014         | 0.182    |
| CG251A    | Canyon    | 06 N | 05 W  | 8.610         | 0.155    |
| CG251B    | Canyon    | 06 N | 05 W  | 8.394         | 0.128    |
| CG252A    | Madison   | 06 N | 39 E  | 1.009         | 0.007    |

| Sample ID | County     | Location |      | Total U(µg/L) | σ (μg/L) |
|-----------|------------|----------|------|---------------|----------|
| CG252B    | Madison    | 06 N     | 39 E | 0.999         | 0.007    |
| CG253     | Payette    | 06 N     | 05 W | 3.440         | 0.031    |
| CG254     | Teton      | 06 N     | 44 E | 0.614         | 0.005    |
| CG256     | Jefferson  | 06 N     | 35 E | 3.442         | 0.022    |
| CG258     | Madison    | 06 N     | 40 E | 1.458         | 0.009    |
| CG259     | Madison    | 06 N     | 41 E | 2.697         | 0.017    |
| CG260     | Payette    | 06 N     | 04 W | 42.494        | 0.780    |
| CG261     | Madison    | 06 N     | 39 E | 0.427         | 0.003    |
| CG262     | Jefferson  | 06 N     | 35 E | 0.955         | 0.006    |
| CG263     | Gem        | 06 N     | 03 W | 0.152         | 0.001    |
| CG264     | Gem        | 06 N     | 01 W | 0.587         | 0.005    |
| CG265     | Madison    | 07 N     | 39 E | 1.370         | 0.009    |
| CG266     | Jefferson  | 06 N     | 35 E | 2.880         | 0.018    |
| CG269     | Custer     | 07 N     | 24 E | 3.463         | 0.022    |
| CG270     | Gem        | 07 N     | 03 W | 0.434         | 0.003    |
| CG271     | Fremont    | 07 N     | 40 E | 0.603         | 0.007    |
| CG272     | Jefferson  | 07 N     | 35 E | 0.883         | 0.006    |
| CG273     | Jefferson  | 07 N     | 36 E | 1.220         | 0.008    |
| CG274     | Custer     | 07 N     | 24 E | 1.238         | 0.008    |
| CG275     | Butte      | 07 N     | 27 E | 0.771         | 0.005    |
| CG277A    | Jefferson  | 08 N     | 33 E | 4.127         | 0.406    |
| CG277B    | Jefferson  | 08 N     | 33 E | 4.647         | 0.583    |
| CG278     | Fremont    | 08 N     | 44 E | 2.166         | 0.014    |
| CG281     | Payette    | 08 N     | 05 W | 6.122         | 0.039    |
| CG282     | Custer     | 08 N     | 22 E | 0.985         | 0.006    |
| CG283     | Custer     | 09 N     | 14 E | 6.913         | 0.044    |
| CG284     | Payette    | 08 N     | 05 W | 2.130         | 0.014    |
| CG285     | Fremont    | 09 N     | 44 E | 0.501         | 0.008    |
| CG286     | Fremont    | 09 N     | 40 E | 0.511         | 0.004    |
| CG288     | Payette    | 09 N     | 05 W | 10.681        | 0.209    |
| CG289     | Fremont    | 09 N     | 44 E | 0.421         | 0.057    |
| CG291     | Fremont    | 09 N     | 42 E | 1.361         | 0.021    |
| CG294     | Custer     | 10 N     | 18 E | 2.175         | 0.014    |
| CG295     | Clark      | 10 N     | 37 E | 0.775         | 0.017    |
| CG298     | Custer     | 11 N     | 17 E | 0.481         | 0.003    |
| CG300     | Clark      | 12 N     | 36 E | 0.764         | 0.071    |
| CG302A    | Custer     | 13 N     | 23 E | 1.648         | 0.012    |
| CG302B    | Custer     | 13 N     | 23 E | 1.362         | 0.009    |
| CG303A    | Washington | 14 N     | 03 W | 0.543         | 0.004    |
| CG303B    | Washington | 14 N     | 03 W | 0.612         | 0.005    |
| CG304     | Lemhi      | 15 N     | 22 E | 0.180         | 0.002    |
| CG305     | Lemhi      | 15 N     | 43 E | 1.465         | 0.010    |

| Sample ID | County     | Location |      | Total U(µg/L) | σ (µg/L) |
|-----------|------------|----------|------|---------------|----------|
| CG307     | Valley     | 16 N     | 03 E | 0.146         | 0.001    |
| CG308     | Lemhi      | 16 N     | 25 E | 1.637         | 0.020    |
| CG309     | Valley     | 17 N     | 03 E | 0.181         | 0.001    |
| CG310     | Lemhi      | 19 N     | 24 E | 1.946         | 0.017    |
| CG311     | Lemhi      | 22 N     | 22 E | 1.514         | 0.067    |
| CG312     | Lemhi      | 23 N     | 22 E | 2.367         | 0.017    |
| CG313     | Lemhi      | 24 N     | 21 E | 5.919         | 0.038    |
| CG314     | Idaho      | 28 N     | 01 E | 0.246         | 0.002    |
| CG315     | Idaho      | 29 N     | 02 E | 0.271         | 0.002    |
| CG316     | Idaho      | 30 N     | 03 E | 0.262         | 0.002    |
| CG317     | Idaho      | 30 N     | 03 E | 0.159         | 0.001    |
| CG320     | Idaho      | 31 N     | 01 E | 0.281         | 0.002    |
| CG321     | Idaho      | 32 N     | 01 E | 1.860         | 0.013    |
| CG322     | Lewis      | 32 N     | 03 W | 0.343         | 0.002    |
| CG323     | Lewis      | 33 N     | 01 W | 1.109         | 0.008    |
| CG324     | Lewis      | 33 N     | 02 W | 8.103         | 0.068    |
| CG325     | Lewis      | 34 N     | 02 E | 0.367         | 0.003    |
| CG326A    | Idaho      | 34 N     | 03 E | 0.612         | 0.010    |
| CG326B    | Idaho      | 34 N     | 03 E | 0.680         | 0.011    |
| CG327A    | Lewis      | 35 N     | 02 E | 0.820         | 0.008    |
| CG327B    | Lewis      | 35 N     | 02 E | 0.940         | 0.009    |
| CG328     | Nez Perce  | 35 N     | 06 W | 1.632         | 0.033    |
| CG329     | Nez Perce  | 35 N     | 05 W | 0.424         | 0.004    |
| CG330     | Nez Perce  | 35 N     | 04 W | 0.892         | 0.009    |
| CG331     | Nez Perce  | 37 N     | 02 W | 0.385         | 0.003    |
| CG332     | Clearwater | 37 N     | 02 E | 0.474         | 0.003    |
| CG333     | Nez Perce  | 37 N     | 05 W | 1.012         | 0.011    |
| CG334     | Nez Perce  | 37 N     | 03 W | 1.380         | 0.015    |
| CG335     | Latah      | 38 N     | 05 W | 0.382         | 0.003    |
| CG336     | Latah      | 38 N     | 02 W | 0.830         | 0.010    |
| CG337     | Latah      | 39 N     | 03 W | 2.741         | 0.044    |
| CG338     | Latah      | 39 N     | 04 W | 1.478         | 0.011    |
| CG339     | Latah      | 40 N     | 03 W | 0.711         | 0.007    |
| CG340     | Latah      | 40 N     | 02 W | 0.345         | 0.002    |
| CG341     | Latah      | 40 N     | 01 W | 0.437         | 0.003    |
| CG342     | Latah      | 41 N     | 05 W | 0.390         | 0.003    |
| CG343     | Latah      | 41 N     | 05 W | 0.618         | 0.005    |
| CG344     | Latah      | 41 N     | 04 W | 0.325         | 0.002    |
| CG345     | Latah      | 44 N     | 03 W | 2.205         | 0.015    |
| CG346     | Benewah    | 44 N     | 05 W | 0.263         | 0.002    |
| CG347     | Benewah    | 45 N     | 01 W | 0.181         | 0.001    |
| CG348     | Benewah    | 45 N     | 04 W | 0.306         | 0.002    |

| Sample ID | County   | Location |      | Total U(µg/L) | σ (μg/L) |
|-----------|----------|----------|------|---------------|----------|
| CG349     | Shoshone | 45 N     | 03 E | 0.768         | 0.006    |
| CG350     | Shoshone | 45 N     | 03 E | 0.341         | 0.002    |
| CG351A    | Shoshone | 45 N     | 02 E | 0.373         | 0.003    |
| CG351B    | Shoshone | 45 N     | 02 E | 0.380         | 0.003    |
| CG352A    | Benewah  | 46 N     | 02 W | 0.893         | 0.007    |
| CG352B    | Benewah  | 46 N     | 02 W | 0.852         | 0.007    |
| CG353     | Benewah  | 46 N     | 04 W | 0.353         | 0.003    |
| CG354     | Kootenai | 47 N     | 02 W | 0.248         | 0.002    |
| CG355     | Kootenai | 47 N     | 04 W | 1.013         | 0.008    |
| CG356     | Shoshone | 48 N     | 06 E | 0.269         | 0.002    |
| CG357     | Kootenai | 48 N     | 04 W | 1.665         | 0.014    |
| CG358     | Shoshone | 48 N     | 03 E | 0.340         | 0.003    |
| CG359     | Kootenai | 49 N     | 03 W | 0.593         | 0.005    |
| CG360     | Shoshone | 50 N     | 04 E | 0.304         | 0.002    |
| CG361     | Kootenai | 50 N     | 04 W | 0.821         | 0.007    |
| CG362     | Kootenai | 50 N     | 05 W | 3.004         | 0.036    |
| CG363     | Kootenai | 51 N     | 05 W | 0.968         | 0.008    |
| CG364     | Kootenai | 53 N     | 03 W | 2.577         | 0.018    |
| CG365     | Kootenai | 53 N     | 02 W | 7.451         | 0.047    |
| CG366     | Bonner   | 55 N     | 03 E | 0.737         | 0.006    |
| CG367     | Bonner   | 55 N     | 02 W | 0.308         | 0.002    |
| CG368     | Bonner   | 55 N     | 02 E | 0.290         | 0.002    |
| CG369     | Bonner   | 56 N     | 05 W | 0.370         | 0.003    |
| CG370     | Bonner   | 56 N     | 05 W | 4.716         | 0.032    |
| CG371     | Bonner   | 56 N     | 01 W | 0.151         | 0.001    |
| CG372     | Bonner   | 56 N     | 02 E | 0.257         | 0.002    |
| CG373     | Bonner   | 56 N     | 04 W | 2.077         | 0.015    |
| CG374     | Bonner   | 57 N     | 05 W | 0.832         | 0.008    |
| CG375     | Bonner   | 58 N     | 01 W | 0.428         | 0.004    |
| CG376     | Bonner   | 59 N     | 01 W | 0.341         | 0.003    |
| CG377     | Bonner   | 59 N     | 01 W | 0.663         | 0.005    |
| CG378     | Bonner   | 61 N     | 04 W | 0.548         | 0.004    |
| CG379A    | Boundry  | 61 N     | 01 E | 11.596        | 0.218    |
| CG379B    | Boundry  | 61 N     | 01 E | 11.641        | 0.220    |
| CG380     | Boundry  | 62 N     | 02 E | 0.985         | 0.008    |
| CG381     | Boundry  | 62 N     | 02 E | 4.885         | 0.031    |
| CG382     | Boundry  | 63 N     | 01 W | 0.132         | 0.001    |
| Sample ID | County     | Loc  | ation | $^{238}U (\mu g/L)$ | σ (μg/L) | %RSD  |
|-----------|------------|------|-------|---------------------|----------|-------|
| CG001     | Bear Lake  | 16 S | 43 E  | 0.986               | 0.001    | 0.080 |
| CG002     | Oneida     | 16 S | 32 E  | 3.518               | 0.128    | 3.649 |
| CG003     | Oneida     | 16 S | 36 E  | 2.427               | 0.021    | 0.846 |
| CG004     | Franklin   | 16 S | 38 E  | 0.813               | 0.005    | 0.636 |
| CG005     | Franklin   | 16 S | 39 E  | 2.012               | 0.006    | 0.288 |
| CG006     | Bear Lake  | 15 S | 44 E  | 3.537               | 0.071    | 2.015 |
| CG007     | Franklin   | 15 S | 40 E  | 1.414               | 0.010    | 0.715 |
| CG008     | Oneida     | 15 S | 36 E  | 1.299               | 0.004    | 0.336 |
| CG009     | Franklin   | 15 S | 40 E  | 1.418               | 0.009    | 0.602 |
| CG010     | Oneida     | 15 S | 36 E  | 1.106               | 0.005    | 0.410 |
| CG011A    | Oneida     | 15 S | 35 E  | 1.017               | 0.002    | 0.235 |
| CG011B    | Oneida     | 15 S | 35 E  | 1.016               | 0.005    | 0.525 |
| CG012A    | Franklin   | 14 S | 38 E  | 1.076               | 0.032    | 2.978 |
| CG012B    | Franklin   | 14 S | 38 E  | 1.187               | 0.007    | 0.552 |
| CG013     | Cassia     | 14 S | 25 E  | 1.348               | 0.002    | 0.175 |
| CG014     | Oneida     | 14 S | 36 E  | 0.808               | 0.005    | 0.601 |
| CG015     | Cassia     | 14 S | 27 E  | 3.829               | 0.038    | 0.982 |
| CG016     | Bear Lake  | 14 S | 43 E  | 0.683               | 0.003    | 0.476 |
| CG017     | Bear Lake  | 14 S | 44 E  | 1.356               | 0.003    | 0.218 |
| CG018     | Oneida     | 14 S | 35 E  | 1.540               | 0.010    | 0.647 |
| CG019     | Cassia     | 13 S | 25 E  | 3.130               | 0.014    | 0.434 |
| CG020     | Oneida     | 13 S | 36 E  | 1.304               | 0.014    | 1.086 |
| CG022     | Cassia     | 13 S | 26 E  | 9.041               | 0.101    | 1.122 |
| CG023     | Cassia     | 12 S | 28 E  | 1.896               | 0.005    | 0.256 |
| CG024     | Bear Lake  | 12 S | 43 E  | 2.995               | 0.019    | 0.632 |
| CG025     | Twin Falls | 12 S | 18 E  | 2.281               | 0.007    | 0.309 |
| CG028     | Twin Falls | 11 S | 16 E  | 4.126               | 0.073    | 1.758 |
| CG029     | Bannock    | 12 S | 36 E  | 0.721               | 0.001    | 0.191 |
| CG030     | Cassia     | 11 S | 22 E  | 3.113               | 0.025    | 0.816 |
| CG031     | Bannock    | 11 S | 36 E  | 4.018               | 0.016    | 0.390 |
| CG032     | Cassia     | 11 S | 23 E  | 1.919               | 0.022    | 1.154 |
| CG033     | Cassia     | 10 S | 26 E  | 3.830               | 0.032    | 0.845 |
| CG034     | Twin Falls | 11 S | 18 E  | 4.122               | 0.028    | 0.679 |
| CG035     | Twin Falls | 10 S | 16 E  | 9.964               | 0.042    | 0.422 |
| CG036     | Cassia     | 10 S | 22 E  | 4.427               | 0.036    | 0.805 |
| CG037     | Twin Falls | 10 S | 17 E  | 5.259               | 0.054    | 1.018 |
| CG038     | Twin Falls | 10 S | 13 E  | 1.862               | 0.013    | 0.687 |
| CG039     | Cassia     | 10 S | 22 E  | 3.024               | 0.024    | 0.790 |
| CG040     | Twin Falls | 10 S | 16 E  | 1.819               | 0.008    | 0.459 |
| CG042     | Cassia     | 10 S | 24 E  | 1.865               | 0.025    | 1.317 |

<sup>238</sup>U Concentrations as measured by ICP-MS

| Sample ID | County     | Loc  | ation | $^{238}$ U (µg/L) | σ (μg/L) | %RSD  |
|-----------|------------|------|-------|-------------------|----------|-------|
| CG043     | Twin Falls | 10 S | 16 E  | 3.985             | 0.032    | 0.810 |
| CG044     | Minidoka   | 10 S | 23 E  | 4.008             | 0.020    | 0.509 |
| CG045     | Minidoka   | 10 S | 24 E  | 1.688             | 0.011    | 0.644 |
| CG046     | Jerome     | 10 S | 21 E  | 5.001             | 0.057    | 1.135 |
| CG047     | Cassia     | 10 S | 25 E  | 4.475             | 0.027    | 0.596 |
| CG049     | Caribou    | 10 S | 41 E  | 0.675             | 0.002    | 0.333 |
| CG050     | Cassia     | 10 S | 27 E  | 1.388             | 0.008    | 0.586 |
| CG051A    | Twin Falls | 09 S | 19 E  | 3.054             | 0.195    | 6.393 |
| CG051B    | Twin Falls | 09 S | 19 E  | 2.367             | 0.164    | 6.937 |
| CG052A    | Twin Falls | 09 S | 17 E  | 6.434             | 0.388    | 6.035 |
| CG052B    | Twin Falls | 09 S | 17 E  | 5.368             | 0.242    | 4.503 |
| CG054     | Caribou    | 09 S | 42 E  | 0.971             | 0.018    | 1.829 |
| CG055     | Minidoka   | 09 S | 22 E  | 1.846             | 0.044    | 2.371 |
| CG056     | Bannock    | 09 S | 38 E  | 0.705             | 0.007    | 1.024 |
| CG057     | Twin Falls | 09 S | 16 E  | 3.412             | 0.099    | 2.905 |
| CG058     | Cassia     | 09 S | 26 E  | 1.520             | 0.024    | 1.566 |
| CG059     | Minidoka   | 09 S | 23 E  | 0.728             | 0.007    | 0.907 |
| CG060     | Caribou    | 09 S | 40 E  | 1.270             | 0.015    | 1.144 |
| CG061     | Caribou    | 09 S | 41 E  | 0.696             | 0.006    | 0.819 |
| CG062     | Jerome     | 09 S | 20 E  | 1.476             | 0.024    | 1.655 |
| CG063     | Power      | 09 S | 34 E  | 1.258             | 0.014    | 1.094 |
| CG064     | Caribou    | 09 S | 42 E  | 1.330             | 0.021    | 1.588 |
| CG065     | Minidoka   | 09 S | 23 E  | 1.071             | 0.017    | 1.563 |
| CG066     | Twin Falls | 09 S | 14 E  | 7.605             | 0.128    | 1.683 |
| CG067     | Blaine     | 09 S | 27 E  | 1.233             | 0.024    | 1.906 |
| CG068     | Minidoka   | 08 S | 24 E  | 1.629             | 0.044    | 2.682 |
| CG069     | Twin Falls | 08 S | 12 E  | 1.437             | 0.020    | 1.380 |
| CG070     | Jerome     | 08 S | 17 E  | 1.658             | 0.033    | 2.011 |
| CG071     | Minidoka   | 08 S | 24 E  | 1.729             | 0.037    | 2.169 |
| CG072     | Jerome     | 08 S | 18 E  | 1.448             | 0.037    | 2.526 |
| CG074     | Power      | 08 S | 30 E  | 2.218             | 0.074    | 3.357 |
| CG075     | Minidoka   | 08 S | 25 E  | 1.377             | 0.036    | 2.647 |
| CG076     | Blaine     | 08 S | 26 E  | 1.534             | 0.031    | 2.030 |
| CG077     | Caribou    | 08 S | 39 E  | 1.622             | 0.015    | 0.932 |
| CG078     | Caribou    | 08 S | 39 E  | 1.126             | 0.009    | 0.798 |
| CG079     | Gooding    | 08 S | 14 E  | 1.673             | 0.055    | 3.310 |
| CG080     | Minidoka   | 08 S | 24 E  | 1.942             | 0.064    | 3.293 |
| CG081     | Lincoln    | 07 S | 21 E  | 1.888             | 0.051    | 2.691 |
| CG082A    | Power      | 07 S | 31 E  | 3.483             | 0.102    | 2.932 |
| CG082B    | Power      | 07 S | 31 E  | 3.471             | 0.106    | 3.042 |
| CG083     | Power      | 07 S | 30 E  | 2.065             | 0.044    | 2.122 |
| CG084     | Caribou    | 07 S | 41 E  | 0.780             | 0.003    | 0.429 |

| Sample ID | County   | Loc  | ation | <sup>238</sup> U (µg/L) | σ (μg/L) | %RSD  |
|-----------|----------|------|-------|-------------------------|----------|-------|
| CG085     | Power    | 07 S | 29 E  | 1.852                   | 0.048    | 2.577 |
| CG086     | Caribou  | 07 S | 38 E  | 1.845                   | 0.034    | 1.824 |
| CG087     | Bannock  | 07 S | 36 E  | 1.291                   | 0.018    | 1.390 |
| CG088     | Bannock  | 07 S | 35 E  | 1.456                   | 0.026    | 1.756 |
| CG090     | Owyhee   | 07 S | 06 E  | 1.350                   | 0.018    | 1.343 |
| CG091     | Lincoln  | 07 S | 23 E  | 1.639                   | 0.040    | 2.459 |
| CG092     | Jerome   | 07 S | 17 E  | 1.677                   | 0.036    | 2.123 |
| CG093     | Gooding  | 07 S | 13 E  | 2.492                   | 0.057    | 2.301 |
| CG094     | Caribou  | 07 S | 41 E  | 1.860                   | 0.028    | 1.530 |
| CG095     | Minidoka | 06 S | 24 E  | 1.580                   | 0.039    | 2.472 |
| CG096     | Lincoln  | 06 S | 20 E  | 1.659                   | 0.036    | 2.167 |
| CG097     | Power    | 06 S | 29 E  | 1.732                   | 0.035    | 2.039 |
| CG098     | Power    | 06 S | 32 E  | 1.729                   | 0.042    | 2.453 |
| CG099     | Owyhee   | 06 S | 05 E  | 9.412                   | 0.189    | 2.005 |
| CG100     | Bannock  | 06 S | 35 E  | 4.960                   | 0.138    | 2.785 |
| CG101     | Power    | 06 S | 33 E  | 1.768                   | 0.030    | 1.688 |
| CG102     | Power    | 06 S | 32 E  | 1.860                   | 0.033    | 1.798 |
| CG103     | Bannock  | 06 S | 34 E  | 2.546                   | 0.057    | 2.246 |
| CG104     | Power    | 06 S | 34 E  | 1.898                   | 0.032    | 1.668 |
| CG105     | Gooding  | 05 S | 15 E  | 4.507                   | 0.119    | 2.637 |
| CG106     | Owyhee   | 05 S | 03 E  | 0.648                   | 0.000    | 0.053 |
| CG107     | Elmore   | 05 S | 08 E  | 0.756                   | 0.001    | 0.146 |
| CG108     | Elmore   | 05 S | 10 E  | 3.787                   | 0.095    | 2.511 |
| CG109     | Owyhee   | 05 S | 03 E  | 6.499                   | 0.161    | 2.475 |
| CG110     | Lincoln  | 05 S | 17 E  | 4.781                   | 0.111    | 2.326 |
| CG111     | Elmore   | 04 S | 04 E  | 0.986                   | 0.006    | 0.615 |
| CG112     | Lincoln  | 04 S | 16 E  | 2.845                   | 0.068    | 2.405 |
| CG113     | Lincoln  | 04 S | 19 E  | 4.758                   | 0.127    | 2.673 |
| CG114     | Elmore   | 04 S | 05 E  | 0.891                   | 0.005    | 0.546 |
| CG115     | Lincoln  | 04 S | 20 E  | 2.741                   | 0.059    | 2.144 |
| CG116A    | Elmore   | 04 S | 07 E  | 1.819                   | 0.031    | 1.686 |
| CG116B    | Elmore   | 04 S | 07 E  | 1.797                   | 0.029    | 1.626 |
| CG117A    | Elmore   | 04 S | 07 E  | 2.177                   | 0.042    | 1.939 |
| CG117B    | Elmore   | 04 S | 07 E  | 2.165                   | 0.045    | 2.073 |
| CG118     | Elmore   | 04 S | 07 E  | 1.990                   | 0.032    | 1.615 |
| CG119     | Bingham  | 03 S | 34 E  | 3.660                   | 0.096    | 2.619 |
| CG120     | Elmore   | 03 S | 06 E  | 1.095                   | 0.007    | 0.617 |
| CG121     | Bingham  | 02 S | 34 E  | 2.089                   | 0.037    | 1.751 |
| CG122     | Bingham  | 02 S | 33 E  | 2.704                   | 0.053    | 1.945 |
| CG123     | Bingham  | 02 S | 36 E  | 2.055                   | 0.034    | 1.639 |
| CG124     | Bingham  | 02 S | 35 E  | 1.394                   | 0.015    | 1.109 |
| CG125     | Bingham  | 02 S | 35 E  | 1.421                   | 0.018    | 1.246 |

| Sample ID | County     | Loc  | ation | $^{238}$ U (µg/L) | σ (μg/L) | %RSD  |
|-----------|------------|------|-------|-------------------|----------|-------|
| CG126A    | Bingham    | 02 S | 34 E  | 2.004             | 0.036    | 1.773 |
| CG126B    | Bingham    | 02 S | 34 E  | 1.985             | 0.030    | 1.532 |
| CG127A    | Blaine     | 02 S | 29 E  | 2.051             | 0.034    | 1.681 |
| CG127B    | Blaine     | 02 S | 29 E  | 2.018             | 0.035    | 1.720 |
| CG128     | Bingham    | 02 S | 33 E  | 2.985             | 0.062    | 2.085 |
| CG129     | Blaine     | 02 S | 21 E  | 2.839             | 0.063    | 2.214 |
| CG130     | Elmore     | 02 S | 05 E  | 2.826             | 0.037    | 1.317 |
| CG131     | Blaine     | 01 S | 21 E  | 2.031             | 0.034    | 1.650 |
| CG132     | Owyhee     | 01 S | 02 W  | 0.754             | 0.000    | 0.027 |
| CG133     | Bingham    | 01 S | 37 E  | 1.579             | 0.014    | 0.909 |
| CG134     | Elmore     | 01 S | 05 E  | 0.856             | 0.001    | 0.124 |
| CG135     | Bingham    | 01 S | 34 E  | 1.881             | 0.030    | 1.589 |
| CG136     | Bingham    | 01 S | 37 E  | 1.217             | 0.008    | 0.694 |
| CG137     | Camas      | 01 S | 15 E  | 0.846             | 0.001    | 0.144 |
| CG138     | Camas      | 01 S | 15 E  | 0.766             | 0.000    | 0.027 |
| CG139     | Blaine     | 01 S | 20 E  | 3.455             | 0.088    | 2.558 |
| CG140     | Bingham    | 01 S | 37 E  | 2.605             | 0.052    | 1.999 |
| CG141     | Canyon     | 01 S | 02 W  | 3.709             | 0.061    | 1.643 |
| CG142     | Owyhee     | 01 S | 03 W  | 0.768             | 0.001    | 0.096 |
| CG143     | Blaine     | 01 S | 19 E  | 2.227             | 0.027    | 1.196 |
| CG144     | Camas      | 01 N | 13 E  | 0.965             | 0.004    | 0.377 |
| CG145     | Canyon     | 01 N | 02 W  | 1.683             | 0.022    | 1.313 |
| CG146     | Elmore     | 01 N | 04 E  | 1.325             | 0.011    | 0.839 |
| CG147     | Blaine     | 01 N | 18 E  | 2.724             | 0.067    | 2.470 |
| CG148     | Bingham    | 01 N | 36 E  | 1.964             | 0.030    | 1.542 |
| CG149     | Bingham    | 01 N | 35 E  | 1.823             | 0.025    | 1.349 |
| CG150     | Bonneville | 01 N | 44 E  | 1.170             | 0.009    | 0.741 |
| CG151A    | Camas      | 01 N | 14 E  | 1.502             | 0.013    | 0.895 |
| CG151B    | Camas      | 01 N | 14 E  | 1.491             | 0.015    | 1.031 |
| CG152A    | Bonneville | 01 N | 37 E  | 2.355             | 0.046    | 1.965 |
| CG152B    | Bonneville | 01 N | 37 E  | 2.313             | 0.037    | 1.612 |
| CG153     | Bonneville | 01 N | 43 E  | 1.401             | 0.012    | 0.827 |
| CG154     | Bonneville | 01 N | 38 E  | 2.511             | 0.056    | 2.249 |
| CG155     | Ada        | 01 N | 01 E  | 3.257             | 0.062    | 1.919 |
| CG156     | Blaine     | 01 N | 19 E  | 2.370             | 0.040    | 1.695 |
| CG157     | Canyon     | 02 N | 02 W  | 2.170             | 0.035    | 1.598 |
| CG158     | Ada        | 02 N | 01 W  | 11.400            | 0.305    | 2.676 |
| CG159     | Canyon     | 04 N | 04 W  | 6.790             | 0.148    | 2.181 |
| CG160     | Ada        | 02 N | 03 E  | 2.840             | 0.049    | 1.715 |
| CG161     | Ada        | 02 N | 01 E  | 5.786             | 0.126    | 2.182 |
| CG162     | Ada        | 02 N | 01 E  | 5.803             | 0.167    | 2.871 |
| CG163     | Canyon     | 02 N | 02 W  | 9.771             | 0.210    | 2.149 |

| Sample ID | County     | Loc  | ation | <sup>238</sup> U (µg/L) | σ (μg/L) | %RSD  |
|-----------|------------|------|-------|-------------------------|----------|-------|
| CG164     | Canyon     | 02 N | 02 W  | 5.681                   | 0.166    | 2.921 |
| CG165     | Bonneville | 02 N | 38 E  | 1.693                   | 0.022    | 1.287 |
| CG166     | Bonneville | 02 N | 39 E  | 2.676                   | 0.059    | 2.216 |
| CG167     | Owyhee     | 02 N | 05 W  | 8.617                   | 0.182    | 2.111 |
| CG168     | Owyhee     | 02 N | 04 W  | 8.410                   | 0.279    | 3.316 |
| CG169     | Canyon     | 02 N | 03 W  | 5.519                   | 0.140    | 2.531 |
| CG170     | Ada        | 02 N | 01 E  | 1.295                   | 0.012    | 0.890 |
| CG171     | Canyon     | 02 N | 03 W  | 0.827                   | 0.000    | 0.037 |
| CG172     | Canyon     | 02 N | 03 W  | 13.310                  | 0.366    | 2.747 |
| CG173     | Owyhee     | 02 N | 04 W  | 0.828                   | 0.001    | 0.172 |
| CG174     | Ada        | 02 N | 01 W  | 11.510                  | 0.325    | 2.826 |
| CG175     | Ada        | 02 N | 03 E  | 2.112                   | 0.027    | 1.266 |
| CG176A    | Bonneville | 03 N | 38 E  | 0.950                   | 0.025    | 2.667 |
| CG176B    | Bonneville | 03 N | 38 E  | 0.938                   | 0.023    | 2.480 |
| CG178     | Ada        | 03 N | 03 E  | 7.837                   | 0.292    | 3.729 |
| CG179     | Canyon     | 03 N | 02 W  | 12.760                  | 0.248    | 1.940 |
| CG180     | Ada        | 03 N | 02 E  | 5.490                   | 0.110    | 1.996 |
| CG181     | Canyon     | 03 N | 04 W  | 26.870                  | 0.480    | 1.786 |
| CG182     | Canyon     | 03 N | 03 W  | 6.863                   | 0.180    | 2.630 |
| CG183     | Canyon     | 03 N | 03 W  | 15.940                  | 0.386    | 2.421 |
| CG184     | Boise      | 03 N | 04 E  | 0.692                   | 0.016    | 2.260 |
| CG185     | Canyon     | 03 N | 01 W  | 11.840                  | 0.353    | 2.977 |
| CG186     | Canyon     | 03 N | 02 W  | 43.840                  | 1.281    | 2.921 |
| CG187     | Ada        | 03 N | 01 E  | 33.530                  | 1.372    | 4.092 |
| CG188     | Ada        | 03 N | 01 E  | 9.470                   | 0.224    | 2.363 |
| CG189     | Canyon     | 03 N | 03 W  | 6.124                   | 0.234    | 3.816 |
| CG191     | Ada        | 03 N | 01 W  | 17.600                  | 0.484    | 2.752 |
| CG192     | Ada        | 03 N | 02 E  | 21.130                  | 0.660    | 3.125 |
| CG193     | Canyon     | 03 N | 03 W  | 10.170                  | 0.317    | 3.112 |
| CG194     | Canyon     | 03 N | 02 W  | 23.910                  | 0.641    | 2.682 |
| CG195     | Ada        | 03 N | 02 E  | 6.511                   | 0.210    | 3.221 |
| CG196     | Ada        | 03 N | 01 E  | 13.250                  | 0.279    | 2.106 |
| CG197     | Jefferson  | 03 N | 40 E  | 0.737                   | 0.012    | 1.668 |
| CG198     | Canyon     | 03 N | 02 W  | 0.679                   | 0.011    | 1.627 |
| CG199     | Jefferson  | 04 N | 37 E  | 0.838                   | 0.022    | 2.577 |
| CG200     | Ada        | 04 N | 01 E  | 28.140                  | 0.758    | 2.694 |
| CG201A    | Jefferson  | 04 N | 39 E  | 0.711                   | 0.016    | 2.266 |
| CG201B    | Jefferson  | 04 N | 39 E  | 0.708                   | 0.014    | 2.031 |
| CG202A    | Owyhee     | 04 N | 05 W  | 0.819                   | 0.015    | 1.772 |
| CG202B    | Owyhee     | 04 N | 05 W  | 0.584                   | 0.006    | 0.946 |
| CG203     | Ada        | 04 N | 01 E  | 15.910                  | 0.564    | 3.544 |
| CG205     | Canyon     | 04 N | 03 W  | 1.310                   | 0.021    | 1.591 |

| Sample ID | County    | Loc  | ation | $^{238}$ U (µg/L) | σ (μg/L) | %RSD  |
|-----------|-----------|------|-------|-------------------|----------|-------|
| CG206     | Jefferson | 04 N | 37 E  | 0.815             | 0.018    | 2.219 |
| CG207     | Boise     | 04 N | 03 E  | 1.015             | 0.023    | 2.237 |
| CG208     | Canyon    | 04 N | 02 W  | 3.169             | 0.072    | 2.274 |
| CG210     | Blaine    | 04 N | 26 E  | 2.642             | 0.055    | 2.088 |
| CG211     | Canyon    | 04 N | 03 W  | 3.327             | 0.043    | 1.300 |
| CG213     | Canyon    | 04 N | 05 W  | 3.901             | 0.101    | 2.576 |
| CG214     | Ada       | 04 N | 01 W  | 1.688             | 0.039    | 2.303 |
| CG215     | Teton     | 04 N | 44 E  | 0.738             | 0.003    | 0.387 |
| CG216     | Ada       | 04 N | 01 E  | 1.125             | 0.013    | 1.124 |
| CG217     | Blaine    | 04 N | 17 E  | 1.429             | 0.035    | 2.417 |
| CG220     | Ada       | 04 N | 01 E  | 13.850            | 0.369    | 2.663 |
| CG221     | Ada       | 04 N | 01 W  | 35.980            | 0.849    | 2.359 |
| CG222     | Canyon    | 04 N | 05 W  | 20.200            | 0.547    | 2.709 |
| CG223     | Madison   | 05 N | 40 E  | 0.852             | 0.011    | 1.275 |
| CG224     | Jefferson | 05 N | 38 E  | 0.710             | 0.014    | 2.042 |
| CG225     | Canyon    | 05 N | 02 W  | 0.806             | 0.009    | 1.163 |
| CG226A    | Canyon    | 05 N | 05 W  | 6.028             | 0.114    | 1.883 |
| CG226B    | Canyon    | 05 N | 05 W  | 5.990             | 0.118    | 1.968 |
| CG227A    | Canyon    | 04 N | 02 W  | 5.595             | 0.127    | 2.261 |
| CG227B    | Canyon    | 04 N | 02 W  | 5.540             | 0.103    | 1.865 |
| CG228     | Ada       | 05 N | 01 E  | 3.512             | 0.085    | 2.413 |
| CG230     | Ada       | 05 N | 02 E  | 1.551             | 0.030    | 1.937 |
| CG231     | Ada       | 05 N | 01 E  | 3.523             | 0.073    | 2.061 |
| CG232     | Ada       | 05 N | 01 W  | 14.890            | 0.219    | 1.473 |
| CG233     | Butte     | 05 N | 26 E  | 1.821             | 0.027    | 1.496 |
| CG236     | Canyon    | 05 N | 03 W  | 36.610            | 0.652    | 1.782 |
| CG237     | Teton     | 05 N | 46 E  | 1.296             | 0.002    | 0.192 |
| CG238     | Madison   | 05 N | 39 E  | 0.833             | 0.008    | 1.019 |
| CG239     | Canyon    | 05 N | 04 W  | 18.490            | 0.422    | 2.282 |
| CG240     | Ada       | 05 N | 01 W  | 3.504             | 0.058    | 1.661 |
| CG241     | Canyon    | 05 N | 04 W  | 5.769             | 0.149    | 2.589 |
| CG242     | Madison   | 05 N | 41 E  | 2.631             | 0.047    | 1.771 |
| CG243     | Teton     | 05 N | 45 E  | 3.416             | 0.062    | 1.826 |
| CG244     | Butte     | 05 N | 26 E  | 2.136             | 0.023    | 1.091 |
| CG245     | Canyon    | 05 N | 03 W  | 4.494             | 0.088    | 1.959 |
| CG246     | Butte     | 05 N | 30 E  | 1.230             | 0.016    | 1.289 |
| CG247     | Jefferson | 05 N | 35 E  | 1.609             | 0.019    | 1.195 |
| CG248     | Madison   | 05 N | 39 E  | 0.727             | 0.011    | 1.473 |
| CG249     | Jefferson | 06 N | 38 E  | 0.700             | 0.010    | 1.393 |
| CG251A    | Canyon    | 06 N | 05 W  | 8.130             | 0.166    | 2.043 |
| CG251B    | Canyon    | 06 N | 05 W  | 8.117             | 0.146    | 1.803 |
| CG252A    | Madison   | 06 N | 39 E  | 1.233             | 0.018    | 1.436 |

| Sample ID | County    | Loc  | ation | <sup>238</sup> U (µg/L) | σ (μg/L) | %RSD  |
|-----------|-----------|------|-------|-------------------------|----------|-------|
| CG252B    | Madison   | 06 N | 39 E  | 1.244                   | 0.017    | 1.391 |
| CG253     | Payette   | 06 N | 05 W  | 4.163                   | 0.092    | 2.200 |
| CG254     | Teton     | 06 N | 44 E  | 0.741                   | 0.010    | 1.349 |
| CG256     | Jefferson | 06 N | 35 E  | 3.580                   | 0.063    | 1.773 |
| CG258     | Madison   | 06 N | 40 E  | 1.502                   | 0.035    | 2.324 |
| CG259     | Madison   | 06 N | 41 E  | 2.689                   | 0.029    | 1.080 |
| CG260     | Payette   | 06 N | 04 W  | 43.880                  | 0.702    | 1.600 |
| CG262     | Jefferson | 06 N | 35 E  | 1.046                   | 0.019    | 1.810 |
| CG264     | Gem       | 06 N | 01 W  | 0.624                   | 0.013    | 2.125 |
| CG265     | Madison   | 07 N | 39 E  | 1.415                   | 0.029    | 2.033 |
| CG266     | Jefferson | 06 N | 35 E  | 3.029                   | 0.070    | 2.306 |
| CG269     | Custer    | 07 N | 24 E  | 3.840                   | 0.095    | 2.473 |
| CG270     | Gem       | 07 N | 03 W  | 0.613                   | 0.006    | 1.000 |
| CG271     | Fremont   | 07 N | 40 E  | 0.738                   | 0.015    | 2.029 |
| CG272     | Jefferson | 07 N | 35 E  | 1.034                   | 0.017    | 1.689 |
| CG273     | Jefferson | 07 N | 36 E  | 1.393                   | 0.026    | 1.855 |
| CG274     | Custer    | 07 N | 24 E  | 1.433                   | 0.037    | 2.557 |
| CG275     | Butte     | 07 N | 27 E  | 0.991                   | 0.017    | 1.749 |
| CG277A    | Jefferson | 08 N | 33 E  | 3.130                   | 0.082    | 2.615 |
| CG277B    | Jefferson | 08 N | 33 E  | 3.074                   | 0.077    | 2.490 |
| CG278     | Fremont   | 08 N | 44 E  | 2.392                   | 0.043    | 1.814 |
| CG281     | Payette   | 08 N | 05 W  | 5.656                   | 0.104    | 1.842 |
| CG282     | Custer    | 08 N | 22 E  | 1.301                   | 0.022    | 1.657 |
| CG283     | Custer    | 09 N | 14 E  | 8.049                   | 0.168    | 2.083 |
| CG284     | Payette   | 08 N | 05 W  | 2.363                   | 0.055    | 2.324 |
| CG285     | Fremont   | 09 N | 44 E  | 0.646                   | 0.013    | 2.055 |
| CG286     | Fremont   | 09 N | 40 E  | 0.597                   | 0.012    | 1.988 |
| CG288     | Payette   | 09 N | 05 W  | 11.550                  | 0.242    | 2.094 |
| CG291     | Fremont   | 09 N | 42 E  | 1.482                   | 0.038    | 2.579 |
| CG294     | Custer    | 10 N | 18 E  | 2.250                   | 0.059    | 2.633 |
| CG295     | Clark     | 10 N | 37 E  | 0.797                   | 0.015    | 1.924 |
| CG298     | Custer    | 11 N | 17 E  | 0.583                   | 0.016    | 2.804 |
| CG300     | Clark     | 12 N | 36 E  | 0.544                   | 0.011    | 1.966 |
| CG302A    | Custer    | 13 N | 23 E  | 1.633                   | 0.027    | 1.668 |
| CG302B    | Custer    | 13 N | 23 E  | 1.607                   | 0.035    | 2.169 |
| CG305     | Lemhi     | 15 N | 43 E  | 1.382                   | 0.032    | 2.302 |
| CG308     | Lemhi     | 16 N | 25 E  | 1.776                   | 0.043    | 2.445 |
| CG310     | Lemhi     | 19 N | 24 E  | 1.772                   | 0.036    | 2.020 |
| CG311     | Lemhi     | 22 N | 22 E  | 3.490                   | 0.066    | 1.891 |
| CG312     | Lemhi     | 23 N | 22 E  | 2.165                   | 0.040    | 1.851 |
| CG313     | Lemhi     | 24 N | 21 E  | 5.790                   | 0.160    | 2.766 |
| CG321     | Idaho     | 32 N | 01 E  | 1.537                   | 0.038    | 2.487 |

| Sample ID | County    | Loc  | ation | <sup>238</sup> U (µg/L) | σ (μg/L) | %RSD  |
|-----------|-----------|------|-------|-------------------------|----------|-------|
| CG323     | Lewis     | 33 N | 01 W  | 0.845                   | 0.014    | 1.652 |
| CG324     | Lewis     | 33 N | 02 W  | 9.431                   | 0.264    | 2.799 |
| CG327A    | Lewis     | 35 N | 02 E  | 0.748                   | 0.016    | 2.165 |
| CG327B    | Lewis     | 35 N | 02 E  | 0.754                   | 0.012    | 1.553 |
| CG328     | Nez Perce | 35 N | 06 W  | 2.229                   | 0.040    | 1.812 |
| CG330     | Nez Perce | 35 N | 04 W  | 0.701                   | 0.013    | 1.837 |
| CG333     | Nez Perce | 37 N | 05 W  | 0.915                   | 0.013    | 1.388 |
| CG334     | Nez Perce | 37 N | 03 W  | 1.412                   | 0.029    | 2.078 |
| CG336     | Latah     | 38 N | 02 W  | 0.849                   | 0.022    | 2.572 |
| CG337     | Latah     | 39 N | 03 W  | 3.567                   | 0.062    | 1.730 |
| CG338     | Latah     | 39 N | 04 W  | 1.337                   | 0.022    | 1.616 |
| CG345     | Latah     | 44 N | 03 W  | 1.956                   | 0.041    | 2.089 |
| CG355     | Kootenai  | 47 N | 04 W  | 0.869                   | 0.006    | 0.681 |
| CG357     | Kootenai  | 48 N | 04 W  | 1.547                   | 0.047    | 3.038 |
| CG362     | Kootenai  | 50 N | 05 W  | 3.508                   | 0.078    | 2.227 |
| CG363     | Kootenai  | 51 N | 05 W  | 0.677                   | 0.009    | 1.386 |
| CG364     | Kootenai  | 53 N | 03 W  | 2.565                   | 0.054    | 2.093 |
| CG365     | Kootenai  | 53 N | 02 W  | 8.555                   | 0.190    | 2.217 |
| CG370     | Bonner    | 56 N | 05 W  | 5.091                   | 0.110    | 2.157 |
| CG371     | Bonner    | 56 N | 01 W  | 4.504                   | 0.170    | 3.766 |
| CG373     | Bonner    | 56 N | 04 W  | 2.063                   | 0.040    | 1.925 |
| CG374     | Bonner    | 57 N | 05 W  | 0.655                   | 0.007    | 1.011 |
| CG379A    | Boundry   | 61 N | 01 E  | 13.200                  | 0.403    | 3.053 |
| CG379B    | Boundry   | 61 N | 01 E  | 12.800                  | 0.380    | 3.022 |
| CG380     | Boundry   | 62 N | 02 E  | 0.952                   | 0.011    | 1.140 |
| CG381     | Boundry   | 62 N | 02 E  | 5.261                   | 0.111    | 2.114 |

## **APPENDIX B**

| Sample ID | <sup>234</sup> U/ <sup>235</sup> U | σ        | %RSD   | <sup>235</sup> U/ <sup>238</sup> U | σ        | %RSD  | UAR  | σ    |
|-----------|------------------------------------|----------|--------|------------------------------------|----------|-------|------|------|
| CG001     | 0.015                              | 1.76E-03 | 11.739 | 0.006                              | 7.34E-05 | 1.223 | 1.95 | 0.23 |
| CG002     | 0.029                              | 1.08E-03 | 3.733  | 0.006                              | 4.94E-05 | 0.823 | 3.76 | 0.14 |
| CG003     | 0.015                              | 1.57E-03 | 10.490 | 0.006                              | 5.04E-05 | 0.840 | 1.95 | 0.20 |
| CG004     | 0.015                              | 8.65E-04 | 5.769  | 0.006                              | 7.58E-05 | 1.263 | 1.95 | 0.11 |
| CG005     | 0.012                              | 9.77E-04 | 8.138  | 0.006                              | 5.77E-05 | 0.962 | 1.56 | 0.13 |
| CG006     | 0.024                              | 4.59E-04 | 1.914  | 0.006                              | 1.63E-05 | 0.272 | 3.11 | 0.06 |
| CG007     | 0.035                              | 1.05E-04 | 0.300  | 0.006                              | 5.96E-05 | 0.993 | 4.54 | 0.01 |
| CG008     | 0.015                              | 6.87E-04 | 4.579  | 0.006                              | 3.52E-05 | 0.587 | 1.95 | 0.09 |
| CG009     | 0.019                              | 1.18E-03 | 6.196  | 0.006                              | 1.51E-05 | 0.252 | 2.46 | 0.15 |
| CG010     | 0.018                              | 5.35E-04 | 2.971  | 0.006                              | 1.37E-05 | 0.228 | 2.34 | 0.07 |
| CG011A    | 0.042                              | 2.68E-03 | 6.374  | 0.006                              | 4.57E-05 | 0.762 | 5.45 | 0.35 |
| CG011B    | 0.043                              | 5.84E-04 | 1.358  | 0.006                              | 5.77E-05 | 0.961 | 5.58 | 0.08 |
| CG012A    | 0.015                              | 1.47E-03 | 9.775  | 0.006                              | 6.47E-05 | 1.079 | 1.95 | 0.19 |
| CG012B    | 0.015                              | 7.39E-04 | 4.927  | 0.006                              | 4.25E-05 | 0.709 | 1.95 | 0.10 |
| CG013     | 0.030                              | 5.70E-04 | 1.899  | 0.006                              | 2.41E-05 | 0.402 | 3.89 | 0.07 |
| CG014     | 0.026                              | 1.84E-03 | 7.087  | 0.006                              | 8.61E-05 | 1.435 | 3.37 | 0.24 |
| CG015     | 0.015                              | 5.49E-04 | 3.658  | 0.007                              | 5.88E-05 | 0.840 | 1.95 | 0.07 |
| CG016     | 0.023                              | 1.71E-03 | 7.415  | 0.006                              | 5.98E-05 | 0.997 | 2.98 | 0.22 |
| CG017     | 0.015                              | 1.09E-03 | 7.246  | 0.006                              | 2.89E-05 | 0.482 | 1.95 | 0.14 |
| CG018     | 0.022                              | 8.98E-04 | 4.081  | 0.006                              | 8.55E-05 | 1.425 | 2.85 | 0.12 |
| CG019     | 0.019                              | 4.22E-04 | 2.219  | 0.007                              | 1.69E-05 | 0.241 | 2.46 | 0.05 |
| CG020     | 0.018                              | 1.67E-03 | 9.298  | 0.006                              | 4.19E-05 | 0.698 | 2.34 | 0.22 |
| CG022     | 0.018                              | 1.95E-04 | 1.081  | 0.007                              | 1.77E-05 | 0.253 | 2.34 | 0.03 |
| CG023     | 0.024                              | 8.83E-04 | 3.679  | 0.006                              | 5.16E-05 | 0.860 | 3.11 | 0.11 |
| CG024     | 0.012                              | 1.12E-03 | 9.318  | 0.007                              | 4.52E-05 | 0.645 | 1.56 | 0.15 |
| CG025     | 0.019                              | 6.53E-04 | 3.439  | 0.006                              | 4.09E-05 | 0.681 | 2.46 | 0.08 |
| CG028     | 0.012                              | 5.01E-04 | 4.172  | 0.007                              | 4.20E-06 | 0.060 | 1.56 | 0.06 |
| CG029     | 0.017                              | 3.97E-04 | 2.335  | 0.006                              | 5.11E-05 | 0.852 | 2.21 | 0.05 |
| CG030     | 0.012                              | 3.24E-04 | 2.696  | 0.007                              | 5.38E-05 | 0.768 | 1.56 | 0.04 |
| CG031     | 0.025                              | 1.22E-03 | 4.862  | 0.007                              | 4.37E-05 | 0.624 | 3.24 | 0.16 |
| CG032     | 0.011                              | 4.40E-04 | 3.996  | 0.007                              | 2.52E-05 | 0.360 | 1.43 | 0.06 |
| CG033     | 0.015                              | 8.65E-04 | 5.769  | 0.007                              | 2.81E-05 | 0.402 | 1.95 | 0.11 |
| CG034     | 0.012                              | 2.50E-04 | 2.081  | 0.007                              | 3.96E-05 | 0.565 | 1.56 | 0.03 |
| CG035     | 0.012                              | 2.80E-04 | 2.330  | 0.007                              | 3.91E-05 | 0.559 | 1.56 | 0.04 |
| CG036     | 0.012                              | 2.85E-04 | 2.373  | 0.007                              | 2.62E-05 | 0.374 | 1.56 | 0.04 |
| CG037     | 0.011                              | 3.23E-04 | 2.937  | 0.007                              | 4.49E-05 | 0.641 | 1.43 | 0.04 |
| CG038     | 0.012                              | 1.13E-03 | 9.441  | 0.007                              | 5.22E-05 | 0.746 | 1.56 | 0.15 |

Uranium Isotopic ratios and activity ratios in the State of Idaho

| Sample ID | <sup>234</sup> U/ <sup>235</sup> U | σ        | %RSD   | <sup>235</sup> U/ <sup>238</sup> U | σ        | %RSD  | UAR  | σ    |
|-----------|------------------------------------|----------|--------|------------------------------------|----------|-------|------|------|
| CG039     | 0.016                              | 1.20E-03 | 7.488  | 0.007                              | 2.05E-05 | 0.293 | 2.08 | 0.16 |
| CG040     | 0.012                              | 1.25E-03 | 10.399 | 0.007                              | 7.94E-05 | 1.134 | 1.56 | 0.16 |
| CG042     | 0.012                              | 7.94E-04 | 6.616  | 0.007                              | 7.29E-05 | 1.041 | 1.56 | 0.10 |
| CG043     | 0.012                              | 3.22E-04 | 2.683  | 0.007                              | 7.91E-06 | 0.113 | 1.56 | 0.04 |
| CG044     | 0.011                              | 3.50E-04 | 3.182  | 0.007                              | 5.16E-05 | 0.737 | 1.43 | 0.05 |
| CG045     | 0.012                              | 1.24E-03 | 10.367 | 0.007                              | 2.24E-05 | 0.320 | 1.56 | 0.16 |
| CG046     | 0.016                              | 2.67E-04 | 1.666  | 0.007                              | 3.00E-05 | 0.429 | 2.08 | 0.03 |
| CG047     | 0.011                              | 4.59E-04 | 4.177  | 0.007                              | 5.38E-05 | 0.769 | 1.43 | 0.06 |
| CG049     | 0.016                              | 3.03E-03 | 18.943 | 0.007                              | 9.46E-05 | 1.351 | 2.08 | 0.39 |
| CG050     | 0.015                              | 7.51E-04 | 5.009  | 0.007                              | 6.00E-05 | 0.857 | 1.95 | 0.10 |
| CG051A    | 0.015                              | 5.88E-04 | 3.919  | 0.006                              | 8.36E-05 | 1.394 | 1.95 | 0.08 |
| CG051B    | 0.015                              | 5.95E-04 | 3.968  | 0.006                              | 1.34E-04 | 2.230 | 1.95 | 0.08 |
| CG052A    | 0.014                              | 4.37E-04 | 3.120  | 0.007                              | 3.44E-05 | 0.491 | 1.82 | 0.06 |
| CG052B    | 0.014                              | 3.50E-04 | 2.497  | 0.007                              | 3.79E-05 | 0.541 | 1.82 | 0.05 |
| CG054     | 0.011                              | 1.11E-03 | 10.105 | 0.006                              | 1.50E-04 | 2.505 | 1.43 | 0.14 |
| CG055     | 0.013                              | 1.02E-03 | 7.872  | 0.006                              | 7.79E-05 | 1.299 | 1.69 | 0.13 |
| CG056     | 0.048                              | 6.44E-03 | 13.413 | 0.006                              | 7.81E-05 | 1.302 | 6.23 | 0.84 |
| CG057     | 0.012                              | 7.06E-04 | 5.880  | 0.006                              | 6.17E-05 | 1.028 | 1.56 | 0.09 |
| CG058     | 0.013                              | 8.05E-04 | 6.195  | 0.006                              | 6.78E-05 | 1.130 | 1.69 | 0.10 |
| CG059     | 0.013                              | 9.54E-04 | 7.337  | 0.006                              | 2.07E-04 | 3.452 | 1.69 | 0.12 |
| CG060     | 0.011                              | 1.55E-03 | 14.133 | 0.006                              | 3.77E-05 | 0.629 | 1.43 | 0.20 |
| CG061     | 0.019                              | 2.26E-03 | 11.879 | 0.006                              | 2.13E-04 | 3.549 | 2.46 | 0.29 |
| CG062     | 0.014                              | 7.40E-04 | 5.289  | 0.006                              | 9.80E-05 | 1.633 | 1.82 | 0.10 |
| CG063     | 0.019                              | 1.91E-03 | 10.057 | 0.006                              | 1.27E-05 | 0.211 | 2.46 | 0.25 |
| CG064     | 0.018                              | 5.99E-04 | 3.330  | 0.006                              | 1.03E-04 | 1.716 | 2.34 | 0.08 |
| CG065     | 0.015                              | 8.85E-04 | 5.899  | 0.006                              | 8.59E-05 | 1.432 | 1.95 | 0.11 |
| CG066     | 0.013                              | 4.70E-04 | 3.612  | 0.007                              | 1.39E-05 | 0.198 | 1.69 | 0.06 |
| CG067     | 0.012                              | 7.92E-04 | 6.603  | 0.006                              | 6.05E-05 | 1.009 | 1.56 | 0.10 |
| CG068     | 0.012                              | 1.03E-03 | 8.559  | 0.006                              | 1.11E-04 | 1.847 | 1.56 | 0.13 |
| CG069     | 0.015                              | 1.41E-03 | 9.382  | 0.006                              | 6.79E-05 | 1.132 | 1.95 | 0.18 |
| CG070     | 0.014                              | 2.18E-04 | 1.558  | 0.006                              | 5.91E-05 | 0.985 | 1.82 | 0.03 |
| CG071     | 0.013                              | 6.65E-04 | 5.116  | 0.006                              | 5.26E-05 | 0.876 | 1.69 | 0.09 |
| CG072     | 0.016                              | 4.36E-04 | 2.728  | 0.006                              | 4.12E-05 | 0.686 | 2.08 | 0.06 |
| CG074     | 0.018                              | 9.86E-04 | 5.478  | 0.006                              | 8.66E-05 | 1.444 | 2.34 | 0.13 |
| CG075     | 0.015                              | 2.01E-03 | 13.409 | 0.006                              | 8.12E-05 | 1.354 | 1.95 | 0.26 |
| CG076     | 0.015                              | 8.33E-04 | 5.556  | 0.006                              | 6.07E-05 | 1.011 | 1.95 | 0.11 |
| CG077     | 0.016                              | 2.50E-04 | 1.565  | 0.006                              | 2.47E-05 | 0.412 | 2.08 | 0.03 |
| CG078     | 0.021                              | 1.24E-03 | 5.894  | 0.006                              | 5.88E-05 | 0.980 | 2.72 | 0.16 |
| CG079     | 0.016                              | 5.92E-04 | 3.703  | 0.006                              | 1.15E-04 | 1.921 | 2.08 | 0.08 |
| CG080     | 0.013                              | 5.89E-04 | 4.533  | 0.006                              | 1.11E-04 | 1.858 | 1.69 | 0.08 |
| CG081     | 0.014                              | 2.76E-04 | 1.968  | 0.006                              | 1.20E-04 | 2.005 | 1.82 | 0.04 |
| CG082A    | 0.024                              | 6.82E-04 | 2.841  | 0.006                              | 4.88E-05 | 0.813 | 3.11 | 0.09 |

| Sample ID | <sup>234</sup> U/ <sup>235</sup> U | σ        | %RSD   | <sup>235</sup> U/ <sup>238</sup> U | σ        | %RSD  | UAR  | σ    |
|-----------|------------------------------------|----------|--------|------------------------------------|----------|-------|------|------|
| CG082B    | 0.023                              | 9.29E-04 | 4.041  | 0.006                              | 1.45E-05 | 0.241 | 2.98 | 0.12 |
| CG083     | 0.019                              | 5.74E-04 | 3.022  | 0.006                              | 8.00E-05 | 1.334 | 2.46 | 0.07 |
| CG084     | 0.011                              | 1.91E-03 | 17.328 | 0.006                              | 9.57E-05 | 1.595 | 1.43 | 0.25 |
| CG085     | 0.015                              | 1.29E-03 | 8.587  | 0.006                              | 1.01E-04 | 1.677 | 1.95 | 0.17 |
| CG086     | 0.019                              | 3.31E-04 | 1.742  | 0.006                              | 8.74E-05 | 1.457 | 2.46 | 0.04 |
| CG087     | 0.013                              | 1.57E-03 | 12.105 | 0.006                              | 2.32E-05 | 0.387 | 1.69 | 0.20 |
| CG088     | 0.029                              | 1.30E-03 | 4.469  | 0.006                              | 1.19E-04 | 1.990 | 3.76 | 0.17 |
| CG090     | 0.012                              | 8.47E-04 | 7.059  | 0.006                              | 8.29E-05 | 1.381 | 1.56 | 0.11 |
| CG091     | 0.015                              | 5.28E-04 | 3.519  | 0.006                              | 7.80E-05 | 1.300 | 1.95 | 0.07 |
| CG092     | 0.015                              | 2.93E-04 | 1.952  | 0.006                              | 6.45E-05 | 1.075 | 1.95 | 0.04 |
| CG093     | 0.014                              | 1.07E-03 | 7.631  | 0.006                              | 7.30E-05 | 1.216 | 1.82 | 0.14 |
| CG094     | 0.016                              | 8.39E-04 | 5.243  | 0.006                              | 4.46E-05 | 0.743 | 2.08 | 0.11 |
| CG095     | 0.015                              | 1.69E-03 | 11.236 | 0.006                              | 9.07E-05 | 1.512 | 1.95 | 0.22 |
| CG096     | 0.016                              | 8.91E-04 | 5.566  | 0.006                              | 8.12E-05 | 1.353 | 2.08 | 0.12 |
| CG097     | 0.015                              | 1.82E-04 | 1.215  | 0.006                              | 3.30E-05 | 0.550 | 1.95 | 0.02 |
| CG098     | 0.018                              | 7.69E-04 | 4.274  | 0.006                              | 1.11E-04 | 1.843 | 2.34 | 0.10 |
| CG099     | 0.011                              | 9.16E-05 | 0.833  | 0.007                              | 1.73E-05 | 0.247 | 1.43 | 0.01 |
| CG100     | 0.016                              | 3.50E-04 | 2.190  | 0.006                              | 2.30E-05 | 0.383 | 2.08 | 0.05 |
| CG101     | 0.015                              | 5.17E-04 | 3.449  | 0.006                              | 7.09E-05 | 1.182 | 1.95 | 0.07 |
| CG102     | 0.015                              | 5.71E-04 | 3.805  | 0.006                              | 7.32E-06 | 0.122 | 1.95 | 0.07 |
| CG103     | 0.014                              | 5.37E-04 | 3.834  | 0.006                              | 8.70E-06 | 0.145 | 1.82 | 0.07 |
| CG104     | 0.016                              | 1.46E-03 | 9.114  | 0.006                              | 1.57E-05 | 0.261 | 2.08 | 0.19 |
| CG105     | 0.012                              | 9.85E-04 | 8.206  | 0.006                              | 6.84E-06 | 0.114 | 1.56 | 0.13 |
| CG106     | 0.014                              | 8.67E-03 | 61.924 | 0.006                              | 9.64E-05 | 1.606 | 1.82 | 1.12 |
| CG107     | 0.014                              | 4.67E-03 | 33.382 | 0.006                              | 9.52E-05 | 1.587 | 1.82 | 0.61 |
| CG108     | 0.010                              | 6.06E-05 | 0.606  | 0.006                              | 2.66E-05 | 0.443 | 1.30 | 0.01 |
| CG109     | 0.011                              | 4.77E-04 | 4.335  | 0.006                              | 3.73E-05 | 0.622 | 1.43 | 0.06 |
| CG110     | 0.012                              | 4.12E-04 | 3.436  | 0.006                              | 2.78E-05 | 0.464 | 1.56 | 0.05 |
| CG111     | 0.014                              | 2.65E-03 | 18.894 | 0.006                              | 5.35E-05 | 0.892 | 1.82 | 0.34 |
| CG112     | 0.010                              | 5.36E-05 | 0.536  | 0.006                              | 6.35E-05 | 1.059 | 1.30 | 0.01 |
| CG113     | 0.011                              | 4.42E-04 | 4.020  | 0.006                              | 3.26E-05 | 0.544 | 1.43 | 0.06 |
| CG114     | 0.017                              | 1.29E-03 | 7.599  | 0.006                              | 2.23E-04 | 3.721 | 2.21 | 0.17 |
| CG115     | 0.014                              | 9.50E-04 | 6.784  | 0.006                              | 4.63E-05 | 0.772 | 1.82 | 0.12 |
| CG116A    | 0.019                              | 1.02E-03 | 5.394  | 0.006                              | 4.83E-05 | 0.805 | 2.46 | 0.13 |
| CG116B    | 0.019                              | 1.02E-03 | 5.389  | 0.006                              | 7.78E-05 | 1.297 | 2.46 | 0.13 |
| CG117A    | 0.014                              | 7.15E-04 | 5.107  | 0.006                              | 2.28E-05 | 0.380 | 1.82 | 0.09 |
| CG117B    | 0.014                              | 7.37E-04 | 5.263  | 0.006                              | 1.86E-05 | 0.310 | 1.82 | 0.10 |
| CG118     | 0.014                              | 7.28E-04 | 5.200  | 0.006                              | 6.78E-05 | 1.130 | 1.82 | 0.09 |
| CG119     | 0.011                              | 9.32E-04 | 8.476  | 0.006                              | 1.06E-05 | 0.176 | 1.43 | 0.12 |
| CG120     | 0.014                              | 1.76E-03 | 12.593 | 0.006                              | 1.03E-05 | 0.171 | 1.82 | 0.23 |
| CG121     | 0.012                              | 4.07E-04 | 3.388  | 0.006                              | 2.32E-05 | 0.386 | 1.56 | 0.05 |
| CG122     | 0.012                              | 3.01E-04 | 2.508  | 0.006                              | 4.68E-06 | 0.078 | 1.56 | 0.04 |

| Sample ID | <sup>234</sup> U/ <sup>235</sup> U | σ        | %RSD   | <sup>235</sup> U/ <sup>238</sup> U | σ        | %RSD  | UAR  | σ    |
|-----------|------------------------------------|----------|--------|------------------------------------|----------|-------|------|------|
| CG123     | 0.018                              | 1.75E-03 | 9.730  | 0.006                              | 6.08E-05 | 1.014 | 2.34 | 0.23 |
| CG124     | 0.011                              | 5.15E-04 | 4.686  | 0.006                              | 8.77E-05 | 1.462 | 1.43 | 0.07 |
| CG125     | 0.014                              | 1.02E-03 | 7.265  | 0.006                              | 1.54E-04 | 2.561 | 1.82 | 0.13 |
| CG126A    | 0.012                              | 1.22E-03 | 10.195 | 0.006                              | 2.06E-05 | 0.344 | 1.56 | 0.16 |
| CG126B    | 0.012                              | 4.02E-04 | 3.351  | 0.006                              | 8.86E-05 | 1.476 | 1.56 | 0.05 |
| CG127A    | 0.019                              | 7.90E-04 | 4.156  | 0.006                              | 2.98E-05 | 0.496 | 2.46 | 0.10 |
| CG127B    | 0.019                              | 4.11E-04 | 2.161  | 0.006                              | 3.67E-05 | 0.611 | 2.46 | 0.05 |
| CG128     | 0.013                              | 6.12E-04 | 4.710  | 0.006                              | 7.07E-05 | 1.178 | 1.69 | 0.08 |
| CG129     | 0.015                              | 8.70E-04 | 5.802  | 0.006                              | 4.88E-05 | 0.814 | 1.95 | 0.11 |
| CG130     | 0.013                              | 4.04E-04 | 3.109  | 0.006                              | 2.56E-05 | 0.426 | 1.69 | 0.05 |
| CG131     | 0.015                              | 5.26E-04 | 3.508  | 0.006                              | 7.85E-05 | 1.308 | 1.95 | 0.07 |
| CG132     | 0.014                              | 5.49E-03 | 39.231 | 0.006                              | 1.03E-04 | 1.724 | 1.82 | 0.71 |
| CG133     | 0.010                              | 4.84E-04 | 4.838  | 0.006                              | 6.74E-05 | 1.124 | 1.30 | 0.06 |
| CG134     | 0.012                              | 5.27E-03 | 43.899 | 0.006                              | 1.54E-04 | 2.571 | 1.56 | 0.68 |
| CG135     | 0.012                              | 1.31E-03 | 10.924 | 0.006                              | 6.86E-05 | 1.144 | 1.56 | 0.17 |
| CG136     | 0.010                              | 6.93E-04 | 6.928  | 0.006                              | 2.46E-04 | 4.102 | 1.30 | 0.09 |
| CG137     | 0.012                              | 9.51E-03 | 79.286 | 0.006                              | 1.69E-04 | 2.818 | 1.56 | 1.23 |
| CG138     | 0.033                              | 9.97E-04 | 3.021  | 0.006                              | 1.83E-04 | 3.048 | 4.28 | 0.13 |
| CG139     | 0.016                              | 5.34E-04 | 3.339  | 0.006                              | 4.69E-05 | 0.781 | 2.08 | 0.07 |
| CG140     | 0.013                              | 6.38E-04 | 4.906  | 0.006                              | 8.06E-05 | 1.344 | 1.69 | 0.08 |
| CG141     | 0.014                              | 2.47E-04 | 1.764  | 0.006                              | 3.83E-05 | 0.639 | 1.82 | 0.03 |
| CG142     | 0.033                              | 2.06E-03 | 6.236  | 0.006                              | 1.46E-04 | 2.437 | 4.28 | 0.27 |
| CG143     | 0.015                              | 1.01E-03 | 6.736  | 0.006                              | 5.92E-05 | 0.987 | 1.95 | 0.13 |
| CG144     | 0.014                              | 4.48E-03 | 32.011 | 0.006                              | 2.09E-04 | 3.478 | 1.82 | 0.58 |
| CG145     | 0.012                              | 6.34E-04 | 5.287  | 0.006                              | 4.85E-05 | 0.808 | 1.56 | 0.08 |
| CG146     | 0.008                              | 7.62E-04 | 9.531  | 0.006                              | 1.46E-04 | 2.440 | 1.04 | 0.10 |
| CG147     | 0.012                              | 1.09E-03 | 9.055  | 0.006                              | 4.30E-05 | 0.716 | 1.56 | 0.14 |
| CG148     | 0.013                              | 6.07E-04 | 4.671  | 0.006                              | 3.33E-05 | 0.555 | 1.69 | 0.08 |
| CG149     | 0.012                              | 2.67E-04 | 2.229  | 0.006                              | 6.66E-05 | 1.110 | 1.56 | 0.03 |
| CG150     | 0.009                              | 1.06E-03 | 11.758 | 0.006                              | 1.25E-04 | 2.091 | 1.17 | 0.14 |
| CG151A    | 0.027                              | 1.25E-03 | 4.643  | 0.006                              | 1.18E-04 | 1.959 | 3.50 | 0.16 |
| CG151B    | 0.028                              | 8.83E-04 | 3.153  | 0.006                              | 1.08E-04 | 1.807 | 3.63 | 0.11 |
| CG152A    | 0.013                              | 3.73E-04 | 2.866  | 0.006                              | 8.05E-05 | 1.341 | 1.69 | 0.05 |
| CG152B    | 0.013                              | 5.34E-04 | 4.104  | 0.006                              | 7.70E-05 | 1.283 | 1.69 | 0.07 |
| CG153     | 0.011                              | 1.92E-03 | 17.491 | 0.006                              | 3.20E-05 | 0.533 | 1.43 | 0.25 |
| CG154     | 0.015                              | 3.24E-04 | 2.163  | 0.006                              | 6.95E-05 | 1.158 | 1.95 | 0.04 |
| CG155     | 0.021                              | 3.34E-04 | 1.592  | 0.006                              | 6.32E-05 | 1.053 | 2.72 | 0.04 |
| CG156     | 0.015                              | 5.85E-04 | 3.897  | 0.006                              | 4.84E-05 | 0.806 | 1.95 | 0.08 |
| CG157     | 0.014                              | 8.65E-04 | 6.180  | 0.006                              | 6.40E-05 | 1.066 | 1.82 | 0.11 |
| CG158     | 0.013                              | 6.81E-05 | 0.524  | 0.007                              | 3.23E-05 | 0.462 | 1.69 | 0.01 |
| CG159     | 0.010                              | 1.40E-04 | 1.402  | 0.006                              | 2.08E-05 | 0.346 | 1.30 | 0.02 |
| CG160     | 0.011                              | 3.80E-04 | 3.451  | 0.006                              | 2.78E-05 | 0.464 | 1.43 | 0.05 |

| Sample ID | <sup>234</sup> U/ <sup>235</sup> U | σ        | %RSD   | <sup>235</sup> U/ <sup>238</sup> U | σ        | %RSD  | UAR  | σ    |
|-----------|------------------------------------|----------|--------|------------------------------------|----------|-------|------|------|
| CG161     | 0.021                              | 4.52E-04 | 2.150  | 0.006                              | 3.31E-05 | 0.551 | 2.72 | 0.06 |
| CG162     | 0.010                              | 1.21E-04 | 1.207  | 0.006                              | 7.92E-06 | 0.132 | 1.30 | 0.02 |
| CG163     | 0.009                              | 8.31E-05 | 0.923  | 0.007                              | 2.17E-05 | 0.310 | 1.17 | 0.01 |
| CG164     | 0.012                              | 3.52E-04 | 2.935  | 0.006                              | 1.46E-05 | 0.243 | 1.56 | 0.05 |
| CG165     | 0.012                              | 6.20E-04 | 5.169  | 0.006                              | 7.09E-05 | 1.182 | 1.56 | 0.08 |
| CG166     | 0.016                              | 1.04E-03 | 6.504  | 0.006                              | 2.18E-05 | 0.363 | 2.08 | 0.13 |
| CG167     | 0.010                              | 4.71E-04 | 4.711  | 0.006                              | 4.12E-05 | 0.686 | 1.30 | 0.06 |
| CG168     | 0.017                              | 4.44E-04 | 2.614  | 0.006                              | 1.13E-05 | 0.189 | 2.21 | 0.06 |
| CG169     | 0.020                              | 3.89E-04 | 1.944  | 0.006                              | 2.51E-05 | 0.419 | 2.59 | 0.05 |
| CG170     | 0.008                              | 8.95E-04 | 11.190 | 0.006                              | 5.38E-05 | 0.896 | 1.04 | 0.12 |
| CG171     | 0.019                              | 3.26E-03 | 17.156 | 0.006                              | 1.86E-04 | 3.095 | 2.46 | 0.42 |
| CG172     | 0.010                              | 3.56E-04 | 3.557  | 0.007                              | 5.04E-06 | 0.072 | 1.30 | 0.05 |
| CG173     | 0.025                              | 3.10E-03 | 12.381 | 0.006                              | 2.17E-04 | 3.615 | 3.24 | 0.40 |
| CG174     | 0.013                              | 3.53E-04 | 2.717  | 0.007                              | 2.67E-05 | 0.381 | 1.69 | 0.05 |
| CG175     | 0.010                              | 5.76E-04 | 5.760  | 0.006                              | 2.16E-05 | 0.360 | 1.30 | 0.07 |
| CG176A    | 0.014                              | 9.43E-04 | 6.542  | 0.008                              | 8.83E-05 | 1.104 | 1.87 | 0.12 |
| CG176B    | 0.015                              | 6.73E-04 | 4.580  | 0.008                              | 1.23E-04 | 1.543 | 1.90 | 0.09 |
| CG178     | 0.009                              | 2.41E-04 | 2.782  | 0.007                              | 2.99E-05 | 0.411 | 1.13 | 0.03 |
| CG179     | 0.014                              | 1.28E-05 | 0.093  | 0.007                              | 1.09E-05 | 0.152 | 1.78 | 0.00 |
| CG180     | 0.012                              | 3.33E-04 | 2.779  | 0.007                              | 3.42E-05 | 0.468 | 1.55 | 0.04 |
| CG181     | 0.012                              | 1.31E-04 | 1.125  | 0.007                              | 3.56E-05 | 0.514 | 1.51 | 0.02 |
| CG182     | 0.016                              | 5.45E-04 | 3.462  | 0.007                              | 4.40E-05 | 0.602 | 2.04 | 0.07 |
| CG183     | 0.018                              | 4.01E-04 | 2.256  | 0.007                              | 4.09E-05 | 0.576 | 2.31 | 0.05 |
| CG184     | 0.010                              | 1.27E-03 | 12.596 | 0.008                              | 9.15E-05 | 1.144 | 1.30 | 0.16 |
| CG185     | 0.014                              | 1.63E-04 | 1.127  | 0.007                              | 3.43E-05 | 0.481 | 1.88 | 0.02 |
| CG186     | 0.012                              | 9.63E-05 | 0.800  | 0.008                              | 6.09E-05 | 0.791 | 1.56 | 0.01 |
| CG187     | 0.016                              | 1.90E-04 | 1.201  | 0.008                              | 1.31E-05 | 0.172 | 2.05 | 0.02 |
| CG188     | 0.013                              | 3.03E-04 | 2.304  | 0.007                              | 1.06E-05 | 0.147 | 1.71 | 0.04 |
| CG189     | 0.021                              | 4.02E-04 | 1.959  | 0.007                              | 1.03E-04 | 1.406 | 2.66 | 0.05 |
| CG191     | 0.013                              | 4.04E-05 | 0.312  | 0.007                              | 3.97E-05 | 0.561 | 1.68 | 0.01 |
| CG192     | 0.010                              | 1.97E-04 | 1.944  | 0.007                              | 3.60E-05 | 0.514 | 1.31 | 0.03 |
| CG193     | 0.017                              | 1.19E-04 | 0.692  | 0.007                              | 3.95E-05 | 0.551 | 2.24 | 0.02 |
| CG194     | 0.012                              | 5.52E-04 | 4.695  | 0.007                              | 2.60E-05 | 0.372 | 1.53 | 0.07 |
| CG195     | 0.010                              | 1.94E-04 | 1.930  | 0.007                              | 4.85E-06 | 0.067 | 1.30 | 0.03 |
| CG196     | 0.011                              | 3.15E-04 | 2.865  | 0.007                              | 1.46E-05 | 0.204 | 1.43 | 0.04 |
| CG197     | 0.013                              | 1.95E-03 | 14.933 | 0.008                              | 4.07E-05 | 0.539 | 1.69 | 0.25 |
| CG198     | 0.013                              | 6.35E-04 | 4.841  | 0.008                              | 5.21E-05 | 0.687 | 1.70 | 0.08 |
| CG199     | 0.014                              | 1.56E-03 | 11.106 | 0.007                              | 8.03E-05 | 1.072 | 1.82 | 0.20 |
| CG200     | 0.010                              | 1.09E-04 | 1.111  | 0.007                              | 2.07E-06 | 0.030 | 1.27 | 0.01 |
| CG201A    | 0.012                              | 1.15E-03 | 9.495  | 0.007                              | 5.85E-05 | 0.787 | 1.57 | 0.15 |
| CG201B    | 0.012                              | 4.28E-04 | 3.608  | 0.007                              | 1.67E-04 | 2.278 | 1.54 | 0.06 |
| CG202A    | 0.012                              | 7.28E-04 | 6.024  | 0.007                              | 3.77E-05 | 0.513 | 1.57 | 0.09 |

| Sample ID | <sup>234</sup> U/ <sup>235</sup> U | σ        | %RSD   | <sup>235</sup> U/ <sup>238</sup> U | σ        | %RSD  | UAR  | σ    |
|-----------|------------------------------------|----------|--------|------------------------------------|----------|-------|------|------|
| CG202B    | 0.013                              | 8.76E-04 | 6.875  | 0.008                              | 2.84E-05 | 0.355 | 1.65 | 0.11 |
| CG203     | 0.010                              | 2.83E-04 | 2.824  | 0.007                              | 4.05E-05 | 0.572 | 1.30 | 0.04 |
| CG205     | 0.013                              | 1.19E-03 | 8.960  | 0.008                              | 4.88E-05 | 0.620 | 1.73 | 0.15 |
| CG206     | 0.014                              | 1.30E-03 | 9.287  | 0.008                              | 9.79E-05 | 1.224 | 1.81 | 0.17 |
| CG207     | 0.011                              | 4.98E-04 | 4.574  | 0.008                              | 4.50E-05 | 0.567 | 1.41 | 0.06 |
| CG208     | 0.014                              | 5.91E-04 | 4.239  | 0.007                              | 2.40E-05 | 0.323 | 1.81 | 0.08 |
| CG210     | 0.019                              | 1.94E-04 | 1.022  | 0.007                              | 2.80E-05 | 0.374 | 2.46 | 0.03 |
| CG211     | 0.015                              | 4.47E-04 | 3.059  | 0.007                              | 5.33E-05 | 0.723 | 1.90 | 0.06 |
| CG213     | 0.017                              | 9.45E-04 | 5.707  | 0.007                              | 2.82E-05 | 0.381 | 2.15 | 0.12 |
| CG214     | 0.010                              | 5.57E-04 | 5.401  | 0.008                              | 2.82E-05 | 0.372 | 1.34 | 0.07 |
| CG215     | 0.018                              | 1.74E-03 | 9.759  | 0.008                              | 4.66E-05 | 0.588 | 2.31 | 0.23 |
| CG216     | 0.011                              | 3.72E-04 | 3.360  | 0.008                              | 1.54E-05 | 0.200 | 1.44 | 0.05 |
| CG217     | 0.015                              | 2.99E-04 | 1.964  | 0.008                              | 4.98E-05 | 0.657 | 1.98 | 0.04 |
| CG220     | 0.012                              | 2.11E-04 | 1.733  | 0.007                              | 1.14E-05 | 0.161 | 1.58 | 0.03 |
| CG221     | 0.012                              | 1.12E-04 | 0.949  | 0.008                              | 6.03E-07 | 0.008 | 1.54 | 0.01 |
| CG222     | 0.012                              | 2.32E-04 | 1.880  | 0.007                              | 2.64E-05 | 0.379 | 1.60 | 0.03 |
| CG223     | 0.012                              | 1.09E-03 | 9.205  | 0.008                              | 5.25E-05 | 0.691 | 1.53 | 0.14 |
| CG224     | 0.013                              | 4.84E-04 | 3.828  | 0.008                              | 1.04E-05 | 0.137 | 1.64 | 0.06 |
| CG225     | 0.015                              | 9.14E-04 | 6.221  | 0.007                              | 8.12E-05 | 1.083 | 1.91 | 0.12 |
| CG226A    | 0.014                              | 3.87E-04 | 2.749  | 0.007                              | 5.36E-05 | 0.746 | 1.83 | 0.05 |
| CG226B    | 0.015                              | 2.10E-04 | 1.434  | 0.007                              | 2.10E-06 | 0.029 | 1.90 | 0.03 |
| CG227A    | 0.015                              | 3.82E-04 | 2.537  | 0.007                              | 3.20E-05 | 0.443 | 1.95 | 0.05 |
| CG227B    | 0.015                              | 5.66E-04 | 3.697  | 0.007                              | 3.13E-05 | 0.435 | 1.99 | 0.07 |
| CG228     | 0.010                              | 8.68E-04 | 8.622  | 0.007                              | 5.60E-05 | 0.772 | 1.31 | 0.11 |
| CG230     | 0.007                              | 3.25E-04 | 4.611  | 0.008                              | 3.68E-06 | 0.048 | 0.91 | 0.04 |
| CG231     | 0.012                              | 4.63E-04 | 3.933  | 0.007                              | 2.46E-05 | 0.332 | 1.53 | 0.06 |
| CG232     | 0.010                              | 4.36E-05 | 0.444  | 0.007                              | 8.47E-06 | 0.120 | 1.27 | 0.01 |
| CG233     | 0.016                              | 7.14E-04 | 4.420  | 0.008                              | 8.39E-05 | 1.107 | 2.09 | 0.09 |
| CG236     | 0.011                              | 2.32E-04 | 2.041  | 0.008                              | 9.99E-06 | 0.133 | 1.48 | 0.03 |
| CG237     | 0.010                              | 1.19E-03 | 11.708 | 0.008                              | 9.81E-05 | 1.275 | 1.32 | 0.15 |
| CG238     | 0.012                              | 4.92E-04 | 4.213  | 0.008                              | 4.18E-05 | 0.528 | 1.51 | 0.06 |
| CG239     | 0.012                              | 2.67E-04 | 2.215  | 0.007                              | 4.47E-06 | 0.064 | 1.56 | 0.03 |
| CG240     | 0.011                              | 4.76E-04 | 4.178  | 0.007                              | 3.06E-05 | 0.417 | 1.48 | 0.06 |
| CG241     | 0.012                              | 1.57E-04 | 1.340  | 0.007                              | 2.84E-05 | 0.393 | 1.52 | 0.02 |
| CG242     | 0.016                              | 6.16E-04 | 3.856  | 0.007                              | 4.75E-05 | 0.646 | 2.07 | 0.08 |
| CG243     | 0.012                              | 3.30E-04 | 2.762  | 0.007                              | 6.35E-05 | 0.864 | 1.55 | 0.04 |
| CG244     | 0.017                              | 3.68E-04 | 2.119  | 0.007                              | 3.80E-05 | 0.508 | 2.25 | 0.05 |
| CG245     | 0.014                              | 1.09E-03 | 7.680  | 0.007                              | 6.22E-05 | 0.856 | 1.83 | 0.14 |
| CG246     | 0.022                              | 3.52E-04 | 1.569  | 0.008                              | 1.97E-05 | 0.262 | 2.91 | 0.05 |
| CG247     | 0.016                              | 6.95E-04 | 4.403  | 0.007                              | 2.53E-05 | 0.340 | 2.05 | 0.09 |
| CG248     | 0.015                              | 5.45E-04 | 3.585  | 0.008                              | 5.23E-05 | 0.685 | 1.97 | 0.07 |
| CG249     | 0.012                              | 1.34E-03 | 11.013 | 0.008                              | 8.58E-05 | 1.122 | 1.58 | 0.17 |

| Sample ID | <sup>234</sup> U/ <sup>235</sup> U | σ        | %RSD   | <sup>235</sup> U/ <sup>238</sup> U | σ        | %RSD  | UAR  | σ    |
|-----------|------------------------------------|----------|--------|------------------------------------|----------|-------|------|------|
| CG251A    | 0.013                              | 1.15E-04 | 0.875  | 0.007                              | 4.19E-05 | 0.585 | 1.70 | 0.01 |
| CG251B    | 0.013                              | 5.73E-04 | 4.270  | 0.007                              | 3.03E-05 | 0.422 | 1.74 | 0.07 |
| CG252A    | 0.014                              | 1.51E-03 | 10.977 | 0.007                              | 6.98E-05 | 0.945 | 1.78 | 0.20 |
| CG252B    | 0.015                              | 1.26E-03 | 8.409  | 0.007                              | 7.84E-05 | 1.059 | 1.94 | 0.16 |
| CG253     | 0.019                              | 3.68E-04 | 1.970  | 0.007                              | 3.63E-05 | 0.503 | 2.42 | 0.05 |
| CG254     | 0.014                              | 4.50E-04 | 3.172  | 0.007                              | 1.36E-04 | 1.852 | 1.84 | 0.06 |
| CG256     | 0.012                              | 2.98E-04 | 2.461  | 0.007                              | 6.86E-05 | 0.955 | 1.57 | 0.04 |
| CG258     | 0.011                              | 8.65E-04 | 7.526  | 0.008                              | 4.54E-05 | 0.599 | 1.49 | 0.11 |
| CG259     | 0.021                              | 6.67E-04 | 3.206  | 0.007                              | 4.85E-05 | 0.654 | 2.70 | 0.09 |
| CG260     | 0.010                              | 2.10E-04 | 2.068  | 0.007                              | 6.59E-06 | 0.089 | 1.32 | 0.03 |
| CG262     | 0.017                              | 7.80E-04 | 4.527  | 0.008                              | 8.97E-05 | 1.183 | 2.24 | 0.10 |
| CG264     | 0.009                              | 5.89E-04 | 6.339  | 0.008                              | 7.37E-05 | 0.931 | 1.21 | 0.08 |
| CG265     | 0.021                              | 9.43E-04 | 4.521  | 0.008                              | 7.79E-05 | 1.038 | 2.71 | 0.12 |
| CG266     | 0.014                              | 2.90E-04 | 2.055  | 0.007                              | 3.71E-05 | 0.508 | 1.83 | 0.04 |
| CG269     | 0.017                              | 3.62E-04 | 2.156  | 0.007                              | 3.20E-05 | 0.439 | 2.18 | 0.05 |
| CG270     | 0.010                              | 1.12E-03 | 11.678 | 0.008                              | 3.63E-05 | 0.467 | 1.24 | 0.15 |
| CG271     | 0.014                              | 1.85E-03 | 13.127 | 0.008                              | 4.75E-05 | 0.624 | 1.83 | 0.24 |
| CG272     | 0.015                              | 8.86E-04 | 5.793  | 0.008                              | 3.84E-05 | 0.510 | 1.98 | 0.11 |
| CG273     | 0.017                              | 2.60E-04 | 1.490  | 0.007                              | 6.06E-05 | 0.817 | 2.27 | 0.03 |
| CG274     | 0.018                              | 5.60E-04 | 3.174  | 0.007                              | 3.93E-05 | 0.528 | 2.29 | 0.07 |
| CG275     | 0.021                              | 1.16E-03 | 5.617  | 0.007                              | 8.12E-05 | 1.090 | 2.68 | 0.15 |
| CG277A    | 0.017                              | 4.35E-04 | 2.518  | 0.007                              | 4.40E-05 | 0.607 | 2.24 | 0.06 |
| CG277B    | 0.018                              | 2.85E-04 | 1.589  | 0.007                              | 1.73E-05 | 0.238 | 2.33 | 0.04 |
| CG278     | 0.023                              | 8.93E-04 | 3.884  | 0.007                              | 2.46E-05 | 0.339 | 2.98 | 0.12 |
| CG281     | 0.009                              | 2.23E-04 | 2.520  | 0.007                              | 5.99E-05 | 0.829 | 1.15 | 0.03 |
| CG282     | 0.015                              | 5.61E-04 | 3.868  | 0.007                              | 1.63E-05 | 0.224 | 1.88 | 0.07 |
| CG283     | 0.019                              | 5.82E-04 | 3.112  | 0.007                              | 2.71E-05 | 0.379 | 2.43 | 0.08 |
| CG284     | 0.010                              | 5.27E-04 | 5.492  | 0.007                              | 2.57E-05 | 0.353 | 1.24 | 0.07 |
| CG285     | 0.011                              | 7.86E-04 | 7.349  | 0.007                              | 7.24E-05 | 0.996 | 1.39 | 0.10 |
| CG286     | 0.015                              | 5.09E-04 | 3.321  | 0.008                              | 8.32E-05 | 1.053 | 1.99 | 0.07 |
| CG288     | 0.010                              | 1.69E-04 | 1.752  | 0.007                              | 1.43E-05 | 0.201 | 1.25 | 0.02 |
| CG291     | 0.019                              | 1.30E-03 | 6.776  | 0.007                              | 1.37E-05 | 0.184 | 2.49 | 0.17 |
| CG294     | 0.016                              | 8.66E-04 | 5.356  | 0.007                              | 1.89E-05 | 0.257 | 2.10 | 0.11 |
| CG295     | 0.018                              | 1.21E-03 | 6.848  | 0.008                              | 1.26E-05 | 0.167 | 2.29 | 0.16 |
| CG298     | 0.030                              | 6.16E-04 | 2.035  | 0.008                              | 7.90E-05 | 1.041 | 3.93 | 0.08 |
| CG300     | 0.018                              | 2.26E-03 | 12.896 | 0.008                              | 4.54E-05 | 0.596 | 2.27 | 0.29 |
| CG302A    | 0.016                              | 8.03E-04 | 5.010  | 0.007                              | 5.25E-05 | 0.711 | 2.08 | 0.10 |
| CG302B    | 0.016                              | 3.65E-04 | 2.334  | 0.007                              | 4.12E-05 | 0.558 | 2.03 | 0.05 |
| CG305     | 0.016                              | 1.01E-03 | 6.247  | 0.007                              | 7.03E-05 | 0.957 | 2.10 | 0.13 |
| CG308     | 0.016                              | 1.69E-03 | 10.341 | 0.007                              | 7.28E-06 | 0.099 | 2.11 | 0.22 |
| CG310     | 0.018                              | 1.48E-04 | 0.827  | 0.007                              | 2.81E-05 | 0.386 | 2.32 | 0.02 |
| CG311     | 0.013                              | 6.08E-04 | 4.783  | 0.007                              | 3.86E-05 | 0.536 | 1.65 | 0.08 |

| Sample ID | <sup>234</sup> U/ <sup>235</sup> U | σ        | %RSD   | <sup>235</sup> U/ <sup>238</sup> U | σ        | %RSD  | UAR  | σ    |
|-----------|------------------------------------|----------|--------|------------------------------------|----------|-------|------|------|
| CG312     | 0.008                              | 1.38E-04 | 1.681  | 0.007                              | 1.73E-05 | 0.239 | 1.07 | 0.02 |
| CG313     | 0.010                              | 5.65E-04 | 5.814  | 0.007                              | 5.36E-05 | 0.746 | 1.26 | 0.07 |
| CG321     | 0.008                              | 2.29E-04 | 3.016  | 0.007                              | 7.45E-05 | 1.015 | 0.99 | 0.03 |
| CG323     | 0.014                              | 1.59E-03 | 11.285 | 0.007                              | 1.73E-05 | 0.237 | 1.83 | 0.21 |
| CG324     | 0.019                              | 6.44E-04 | 3.422  | 0.007                              | 4.66E-05 | 0.651 | 2.44 | 0.08 |
| CG327A    | 0.017                              | 4.11E-04 | 2.458  | 0.007                              | 1.04E-04 | 1.430 | 2.17 | 0.05 |
| CG327B    | 0.018                              | 9.63E-04 | 5.461  | 0.007                              | 1.06E-04 | 1.453 | 2.29 | 0.12 |
| CG328     | 0.012                              | 1.35E-03 | 11.452 | 0.007                              | 3.66E-05 | 0.501 | 1.53 | 0.18 |
| CG330     | 0.018                              | 1.36E-03 | 7.626  | 0.007                              | 1.09E-04 | 1.490 | 2.31 | 0.18 |
| CG333     | 0.012                              | 8.75E-04 | 7.467  | 0.007                              | 5.60E-05 | 0.760 | 1.52 | 0.11 |
| CG334     | 0.012                              | 9.87E-04 | 8.525  | 0.007                              | 2.92E-05 | 0.400 | 1.50 | 0.13 |
| CG336     | 0.011                              | 7.50E-04 | 6.936  | 0.007                              | 7.12E-05 | 0.971 | 1.40 | 0.10 |
| CG337     | 0.010                              | 3.70E-04 | 3.564  | 0.007                              | 2.09E-05 | 0.288 | 1.35 | 0.05 |
| CG338     | 0.018                              | 2.19E-04 | 1.237  | 0.007                              | 1.26E-04 | 1.733 | 2.30 | 0.03 |
| CG345     | 0.048                              | 1.88E-03 | 3.924  | 0.007                              | 2.02E-05 | 0.279 | 6.21 | 0.24 |
| CG355     | 0.016                              | 1.27E-03 | 7.922  | 0.007                              | 6.57E-05 | 0.905 | 2.07 | 0.16 |
| CG357     | 0.010                              | 7.23E-04 | 7.599  | 0.007                              | 4.53E-05 | 0.625 | 1.24 | 0.09 |
| CG362     | 0.009                              | 8.83E-04 | 10.276 | 0.007                              | 1.05E-04 | 1.457 | 1.11 | 0.11 |
| CG363     | 0.011                              | 8.51E-04 | 7.581  | 0.007                              | 9.24E-05 | 1.263 | 1.46 | 0.11 |
| CG364     | 0.008                              | 9.64E-04 | 11.555 | 0.007                              | 3.90E-05 | 0.540 | 1.08 | 0.13 |
| CG365     | 0.008                              | 2.88E-04 | 3.441  | 0.007                              | 7.10E-06 | 0.099 | 1.09 | 0.04 |
| CG370     | 0.008                              | 6.34E-04 | 8.170  | 0.007                              | 1.56E-05 | 0.216 | 1.01 | 0.08 |
| CG371     | 0.012                              | 5.25E-05 | 0.444  | 0.007                              | 5.10E-05 | 0.705 | 1.53 | 0.01 |
| CG373     | 0.008                              | 3.81E-04 | 4.775  | 0.007                              | 7.39E-05 | 1.017 | 1.04 | 0.05 |
| CG374     | 0.008                              | 7.51E-04 | 9.679  | 0.007                              | 2.42E-05 | 0.328 | 1.01 | 0.10 |
| CG379     | 0.008                              | 5.87E-05 | 0.781  | 0.007                              | 2.53E-05 | 0.356 | 0.97 | 0.01 |
| CG380     | 0.012                              | 1.29E-03 | 10.859 | 0.007                              | 7.34E-05 | 1.017 | 1.54 | 0.17 |
| CG381     | 0.013                              | 1.84E-04 | 1.379  | 0.007                              | 5.76E-05 | 0.796 | 1.73 | 0.02 |

## **APPENDIX C**

Split Samples on KPA

| Sample ID | TU (µg/L) | 3σ (μg/L) | Sample ID | TU (µg/L) | 3σ (μg/L) | % diff | pass? |
|-----------|-----------|-----------|-----------|-----------|-----------|--------|-------|
| CG011A    | 0.480     | 0.042     | CG011B    | 0.499     | 0.036     | 3.9    | Yes   |
| CG012A    | 1.179     | 0.036     | CG012B    | 1.005     | 0.051     | 15.9   | Yes   |
| CG026A    | 0.981     | 0.026     | CG026B    | 0.844     | 0.023     | 15.0   | Yes   |
| CG027A    | 0.507     | 0.024     | CG027B    | 0.545     | 0.027     | 7.2    | Yes   |
| CG051A    | 2.777     | 0.225     | CG051B    | 3.155     | 0.226     | 12.7   | Yes   |
| CG052A    | 6.979     | 1.012     | CG052B    | 6.268     | 0.966     | 10.7   | Yes   |
| CG082A    | 6.777     | 1.561     | CG082B    | 6.489     | 1.425     | 4.3    | Yes   |
| CG116A    | 2.204     | 0.764     | CG116B    | 2.050     | 0.553     | 7.2    | Yes   |
| CG117A    | 3.070     | 0.917     | CG117B    | 3.518     | 1.252     | 13.6   | Yes   |
| CG126A    | 3.203     | 0.113     | CG126B    | 2.905     | 0.103     | 9.8    | Yes   |
| CG127A    | 3.152     | 0.085     | CG127B    | 3.327     | 0.088     | 5.4    | Yes   |
| CG151A    | 0.730     | 0.017     | CG151B    | 0.717     | 0.016     | 1.8    | Yes   |
| CG152A    | 1.390     | 0.037     | CG152B    | 1.476     | 0.057     | 6.0    | Yes   |
| CG176A    | 0.995     | 0.045     | CG176B    | 1.087     | 0.078     | 8.8    | Yes   |
| CG201A    | 0.876     | 0.570     | CG201B    | 0.754     | 0.183     | 15.0   | Yes   |
| CG226A    | 4.428     | 0.095     | CG226B    | 4.860     | 0.162     | 9.3    | Yes   |
| CG227A    | 4.160     | 0.099     | CG227B    | 4.134     | 0.091     | 0.6    | Yes   |
| CG251A    | 8.610     | 0.465     | CG251B    | 8.394     | 0.385     | 2.5    | Yes   |
| CG252A    | 1.009     | 0.021     | CG252B    | 0.999     | 0.021     | 1.1    | Yes   |
| CG277A    | 4.127     | 1.218     | CG277B    | 4.647     | 1.749     | 11.9   | Yes   |
| CG302A    | 1.648     | 0.035     | CG302B    | 1.362     | 0.026     | 19.0   | Yes   |
| CG303A    | 0.543     | 0.013     | CG303B    | 0.612     | 0.016     | 11.9   | Yes   |
| CG326A    | 0.612     | 0.030     | CG326B    | 0.680     | 0.033     | 10.5   | Yes   |
| CG327A    | 0.820     | 0.024     | CG327B    | 0.940     | 0.026     | 13.6   | Yes   |
| CG351A    | 0.373     | 0.009     | CG351B    | 0.380     | 0.009     | 1.8    | Yes   |
| CG352A    | 0.893     | 0.021     | CG352B    | 0.852     | 0.020     | 4.7    | Yes   |
| CG379A    | 11.596    | 0.655     | CG379B    | 11.641    | 0.659     | 0.4    | Yes   |

Split Samples on ICP-MS

| Sample ID | $^{238}U (\mu g/L)$ | 3σ (μg/L) | Sample ID | $^{238}$ U (µg/L) | 3σ (μg/L) | % diff | pass? |
|-----------|---------------------|-----------|-----------|-------------------|-----------|--------|-------|
| CG011A    | 1.017               | 0.006     | CG011B    | 1.016             | 0.015     | 0.1    | Yes   |
| CG012A    | 1.076               | 0.096     | CG012B    | 1.187             | 0.021     | 9.8    | Yes   |
| CG026A    | 0.444               | 0.009     | CG026B    | 0.437             | 0.006     | 1.6    | Yes   |
| CG051A    | 3.054               | 0.585     | CG051B    | 2.367             | 0.492     | 25.3   | Yes   |
| CG052A    | 6.434               | 1.164     | CG052B    | 5.368             | 0.726     | 18.1   | Yes   |
| CG082A    | 3.483               | 0.306     | CG082B    | 3.471             | 0.318     | 0.3    | Yes   |
| CG089A    | 0.589               | 0.000     | CG089B    | 0.593             | 0.000     | 0.0    | Yes   |
| CG116A    | 1.819               | 0.093     | CG116B    | 1.797             | 0.087     | 1.2    | Yes   |
| CG117A    | 2.177               | 0.126     | CG117B    | 2.165             | 0.135     | 0.6    | Yes   |
| CG126A    | 2.004               | 0.108     | CG126B    | 1.985             | 0.090     | 0.9    | Yes   |
| CG127A    | 2.051               | 0.102     | CG127B    | 2.018             | 0.105     | 1.6    | Yes   |
| CG151A    | 1.502               | 0.039     | CG151B    | 1.491             | 0.045     | 0.7    | Yes   |
| CG152A    | 2.355               | 0.138     | CG152B    | 2.313             | 0.111     | 1.8    | Yes   |
| CG176A    | 0.950               | 0.075     | CG176B    | 0.938             | 0.069     | 1.3    | Yes   |
| CG201A    | 0.711               | 0.048     | CG201B    | 0.708             | 0.042     | 0.4    | Yes   |
| CG226A    | 6.028               | 0.342     | CG226B    | 5.990             | 0.354     | 0.6    | Yes   |
| CG227A    | 5.595               | 0.381     | CG227B    | 5.540             | 0.309     | 1.0    | Yes   |
| CG251A    | 8.130               | 0.498     | CG251B    | 8.117             | 0.438     | 0.2    | Yes   |
| CG252A    | 1.233               | 0.054     | CG252B    | 1.244             | 0.051     | 0.9    | Yes   |
| CG277A    | 3.130               | 0.246     | CG277B    | 3.074             | 0.231     | 1.8    | Yes   |
| CG302A    | 1.633               | 0.081     | CG302B    | 1.607             | 0.105     | 1.6    | Yes   |
| CG303A    | 0.501               | 0.018     | CG303B    | 0.512             | 0.024     | 2.2    | Yes   |
| CG326A    | 0.170               | 0.009     | CG326B    | 0.175             | 0.015     | 2.9    | Yes   |
| CG327A    | 0.748               | 0.048     | CG327B    | 0.754             | 0.036     | 0.8    | Yes   |
| CG352A    | 0.503               | 0.027     | CG352B    | 0.511             | 0.045     | 1.6    | Yes   |

Spiked Samples on KPA

| Sample ID | Analysis | Conc. (µg/L) | 3σ (μg/L) | Measured (µg/L) | 3σ (μg/L) | % diff | pass? |
|-----------|----------|--------------|-----------|-----------------|-----------|--------|-------|
| Blank1    | Total U  | 0.000        | 0.000     | 0.000           | 0.000     | 0.0    | Yes   |
| 5ppb1     | Total U  | 5.000        | 0.300     | 5.198           | 0.168     | 4.0    | Yes   |
| 20ppb1    | Total U  | 20.000       | 0.300     | 20.652          | 1.953     | 3.3    | Yes   |
| 50ppb1    | Total U  | 50.000       | 0.300     | 49.862          | 4.728     | 0.3    | Yes   |
| Blank2    | Total U  | 0.000        | 0.000     | 0.000           | 0.000     | 0.0    | Yes   |
| 5ppb2     | Total U  | 5.000        | 0.300     | 4.939           | 0.189     | 1.2    | Yes   |
| 20ppb2    | Total U  | 20.000       | 0.300     | 18.164          | 1.782     | 9.2    | Yes   |
| 50ppb2    | Total U  | 50.000       | 0.300     | 51.262          | 4.941     | 2.5    | Yes   |
| Blank3    | Total U  | 0.000        | 0.000     | 0.000           | 0.000     | 0.0    | Yes   |
| 5ppb3     | Total U  | 5.000        | 0.300     | 5.300           | 0.207     | 6.0    | Yes   |
| 20ppb3    | Total U  | 20.000       | 0.300     | 20.859          | 2.013     | 4.3    | Yes   |
| 50ppb3    | Total U  | 50.000       | 0.300     | 53.755          | 5.190     | 7.5    | Yes   |
| Blank4    | Total U  | 0.000        | 0.000     | 0.000           | 0.000     | 0.0    | Yes   |
| 5ppb4     | Total U  | 5.000        | 0.300     | 5.300           | 0.171     | 6.0    | Yes   |
| 20ppb4    | Total U  | 20.000       | 0.300     | 18.597          | 1.794     | 7.0    | Yes   |
| 50ppb4    | Total U  | 50.000       | 0.300     | 48.440          | 4.590     | 3.1    | Yes   |
| Blank5    | Total U  | 0.000        | 0.000     | 0.000           | 0.000     | 0.0    | Yes   |
| 5ppb5     | Total U  | 5.000        | 0.300     | 5.064           | 0.165     | 1.3    | Yes   |
| 20ppb5    | Total U  | 20.000       | 0.300     | 18.972          | 1.803     | 5.1    | Yes   |
| 50ppb5    | Total U  | 50.000       | 0.300     | 48.117          | 4.578     | 3.8    | Yes   |

Spiked Samples on ICP MS

| Sample ID    | Analysis | Conc. (µg/L) | <b>3σ (μg/L)</b> | Measured (µg/L) | <b>3σ (μg/L)</b> | % diff | pass? |
|--------------|----------|--------------|------------------|-----------------|------------------|--------|-------|
| Blank_1      | U-238    | 0.000        | 0.000            | 0.043           | 0.115            | 0.0    | Yes   |
| 100ppt 71A_1 | U-238    | 0.100        | 0.030            | 0.194           | 0.042            | 94.0   | No    |
| 1ppb71A_1    | U-238    | 1.000        | 0.030            | 1.063           | 0.030            | 6.3    | Yes   |
| 10ppb71A_1   | U-238    | 10.000       | 0.030            | 8.855           | 0.372            | 11.5   | Yes   |
| 100ppb71A_1  | U-238    | 100.000      | 0.030            | 95.930          | 3.243            | 4.1    | Yes   |
| Blank_1      | U-238    | 0.000        | 0.000            | 0.034           | 0.102            | 0.0    | Yes   |
| 100pptERA_1  | U-238    | 0.100        | 0.030            | 0.553           | 0.006            | 453.0  | No    |
| 1ppbERA_1    | U-238    | 1.000        | 0.030            | 1.175           | 0.006            | 12.5   | Yes   |
| 10ppbERA_1   | U-238    | 10.000       | 0.030            | 9.061           | 0.204            | 9.4    | Yes   |
| 100ppbERA_1  | U-238    | 100.000      | 0.030            | 99.380          | 1.347            | 0.6    | Yes   |
| Blank_1      | U-238    | 0.000        | 0.000            | 0.027           | 0.055            | 0.0    | Yes   |
| 100pptNIST_1 | U-238    | 0.100        | 0.030            | 0.511           | 0.000            | 411.0  | No    |
| 1ppbNIST_1   | U-238    | 1.000        | 0.030            | 1.108           | 0.003            | 10.6   | Yes   |
| 10ppbNIST_1  | U-238    | 10.000       | 0.030            | 9.186           | 0.411            | 8.1    | Yes   |
| 100ppbNIST_1 | U-238    | 100.000      | 0.030            | 100.000         | 0.588            | 0.0    | Yes   |
| Blank_2      | U-238    | 0.000        | 0.000            | 0.032           | 0.650            | 0.0    | Yes   |
| 100ppt71A_2  | U-238    | 0.100        | 0.030            | 0.197           | 0.030            | 97.0   | No    |
| 1ppb 71A_2   | U-238    | 1.000        | 0.030            | 1.114           | 0.036            | 9.1    | Yes   |
| 10ppb 71A_2  | U-238    | 10.000       | 0.030            | 8.903           | 0.990            | 11.0   | Yes   |
| 100ppb 71A_2 | U-238    | 100.000      | 0.030            | 99.220          | 11.043           | 0.8    | Yes   |
| Blank_2      | U-238    | 0.000        | 0.000            | 0.041           | 0.06             | 0.0    | Yes   |
| 100pptERA _2 | U-238    | 0.100        | 0.030            | 0.549           | 0.006            | 449.0  | No    |
| 1ppbERA_2    | U-238    | 1.000        | 0.030            | 1.123           | 0.069            | 14.6   | Yes   |
| 10ppbERA_2   | U-238    | 10.000       | 0.030            | 9.159           | 0.828            | 8.4    | Yes   |
| 100ppb ERA_2 | U-238    | 100.000      | 0.030            | 100.900         | 9.897            | 0.9    | Yes   |
| Blank_2      | U-238    | 0.000        | 0.000            | 0.046           | 0.21             | 0.0    | Yes   |
| 100pptNIST_2 | U-238    | 0.100        | 0.030            | 0.532           | 0.006            | 432.0  | No    |
| 1ppbNIST_2   | U-238    | 1.000        | 0.030            | 1.091           | 0.087            | 14.0   | Yes   |
| 10ppbNIST_2  | U-238    | 10.000       | 0.030            | 9.143           | 0.939            | 8.6    | Yes   |
| 100ppbNIST_2 | U-238    | 100.000      | 0.030            | 99.900          | 10.119           | 0.1    | Yes   |

Note: The concentration of the spiked samples that did not pass is  $0.1\mu$ g/L, which is below the LLD for ICP MS.