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1.0 Introduction

This document will describe the types of methods approved to analyze uranium in groundwater and the effects of converting between the units of picocuries per liter and micrograms per liter for the various methods. To this end, a water sample was analyzed by three different methods at the Wisconsin State Laboratory of Hygiene. The results were reported in both picocuries per liter and micrograms per liter for each method. A discussion of the results and recommendations for public water suppliers is included.

There are three naturally occurring isotopes of uranium in ground water: uranium-238, uranium-235, and uranium-234. The atom percent natural abundances for these isotopes are 99.27, 0.72, and 0.0055, respectively. However, due to chemical and radiochemical processes that occur in the aquifer, the natural distribution of these isotopes is not maintained and the condition is referred to as a state of disequilibrium.

Although secular equilibrium exists in the U-238 and Th-232 decay series, the amount of a particular radionuclide found in water will depend to a large extent on some of the physical and chemical properties of the radionuclides. For example, when a radionuclide undergoes alpha decay, it emits an alpha particle with several million electron-volts of energy. To conserve momentum, the nucleus that emits the alpha particle will recoil in the direction opposite to the direction in which the alpha particle was emitted. If the parent nuclide started in the solid phase, the progeny nucleus may have recoiled far enough for it to have reached the aqueous phase. For example, suppose that in an aquifer U-238 is present in an ore, which may be a small grain of material. When the U-238 decays, the recoiling Th-234 nucleus may reach the aqueous phase and precipitate from solution. The Th-234 atom then undergoes two successive beta decays forming a U-234 atom, which readily goes into the aqueous phase. Without the recoil step, this atom of U-234 would have remained in the parent ore. This process tends to enrich the aqueous phase in U-234 relative to U-238.1

A more detailed explanation of the mechanism responsible for disequilibrium is beyond the scope of this document; however, a list of references on this topic is provided in the references^{2,3}. Figure 1 shows the decay series for uranium-238 and its progeny.

When a water sample contains uranium there are different analytical approaches that can be taken in order to determine the level of uranium. Since uranium is radioactive, analysts can use either techniques that measure radioactivity or techniques that measure the concentration. There are three types of methods for measuring the level of uranium in groundwater: (1) total activity methods, (2) total concentration methods, and (3) isotopic uranium methods.

The total activity method (1) is a technique whereby the uranium in the sample is chemically separated from all other chemical substances in the water sample. Once the uranium is isolated, it is analyzed on an instrument designed specifically to measure the alpha radiation that is emitted from the decay of any of the three uranium isotopes. The instrument, called an alphabeta proportional counter, can not distinguish one isotope from another. The data output from the instrument is the gross number of alpha particles emitted from the sample over a given period of time, or the count rate. Since the sample was purified before analysis it is assumed that all of the alpha particles measured are from uranium. The count rate is used to calculate the radioactivity of the sample and reported in the units of picocuries per liter (pCi/L). The calculated activity is the total activity of all three uranium isotopes. If the activity is converted into a mass concentration, the assumption is made that the uranium isotopes are present in natural abundance. As will be discussed in later sections, this is a poor assumption and generally the mass concentration is biased high.

Total concentration methods (2) measure the mass of uranium in the sample on a per liter basis. The number of preparatory steps required will depend on which instrument is used to measure the mass. All of the total concentration methods measure only the uranium 238 in the sample. The results are reported in micrograms per liter (μ g/L) of total uranium (all three isotopes) using the assumption that the isotopes are present in natural abundance. This assumption is acceptable because even under conditions of extreme disequilibrium, uranium-238 accounts for more than 99% of the mass. To convert the concentration results into activity, it is assumed once again that the isotopes are present in natural abundance. This, however, is a poor assumption because a tiny increase above the expected mass of uranium-234 leads to a large increase in the total activity. Since the mass concentration methods can not measure these small changes, the conversion of the results to activity is biased low.

The isotopic method (3) for the analysis of uranium is similar to the total activity method, in that the sample is taken through a number of purification steps prior to being analyzed to isolate the uranium. The instrument used for the analysis, an alpha spectrometer, is designed to detect the alpha particles coming from the sample. In addition to measuring the alpha count rate of the sample, the alpha spectrometer is also able to distinguish one uranium isotope from another because the alpha particles from each isotope are emitted with different energies. These energies are given in Table 2. Each time an alpha particle of a given energy is detected by the spectrometer, the count will be displayed in a specific region of the spectrum. As more and more counts are detected a peak forms in the spectrum that is proportional to the amount of radioactivity from that isotope. The count rate from this sample is used to calculate the radioactivity of each uranium isotope in the sample and reported in the units of picocuries per liter (pCi/L). The individual activities can be summed to determine the total uranium activity in pCi/L. Since the activity of each isotope has been determined it is possible to directly calculate the mass of each isotope. The individual masses can be summed to determine the total mass of uranium in $(\mu g/L)$. The equations needed to perform this calculation can be found in Attachment 2. The spectrum from the sample analyzed for this document is shown in Figure 2.

The isotopic method is the only method capable of accurately determining uranium mass concentration and activity. This is because no assumptions need to be made to determine either concentration or activity.

The Safe Drinking Water Act (SDWA) regulation sets a limit of 30 micrograms per liter ($\mu g/L$) of water for total uranium. This standard is a total concentration standard, whereas all other standards for radionuclides are based on activity. Additionally, the SDWA allows the measured gross alpha activity to be adjusted by subtracting the activity of total uranium to determine compliance with the gross alpha standard of 15 pCi/L. So, a water utility may need the uranium level in pCi/L, µg/L, or both depending on its situation. There is at least one approved method for each of the three types of methods described above. However, the regulation does not specify which type of method to use in given situations. The regulation does allow for the conversion between pCi/L and μ g/L using the factor of 0.67 pCi/ μ g⁴ using any of the approved methods. A derivation of how this factor is determined is given in Attachment 1. This is a valid conversion factor only when the three isotopes are dissolved in the water in natural abundance ratios. As stated above due to secular disequilibrium between uranium-238 and uranium-234 in groundwater this is a poor assumption. To demonstrate how using the conversion factor can affect the uranium level reported, a sample was analyzed by one of each of the three types of methods and shown in Table 1. Only one of the three analytically methods, isotopic uranium, can accurately produce both pCi/L and µg/L results. This is because the conversion to micrograms is not based on any assumptions.



Figure 1 U-238 Decay Series



Figure 2 Alpha Spectrum of a Groundwater Sample

2.0 Methods

2.1 Total Activity (1): EPA Method 908.0 Total Uranium

2.1.1 Summary

In this method the uranium in an acidified water sample is scavenged by iron hydroxide precipitation. The precipitate is dissolved in a strong acid solution and then passed through an anion exchange resin. Interfering ions are eluted from the resin and then the uranium is eluted and collected. The isolated uranium is transferred to a planchet and analyzed on a low background alpha beta proportional counter in the alpha only mode. The results are reported in pCi/L as total uranium. This is a labor intensive technique and the fiscal year 2005 price at the Wisconsin State Laboratory of Hygiene is \$153.95.

2.1.2 Limitations

Due to the fact that there are no stable isotopes of uranium, a gravimetric yield can not be used to measure recovery for each sample. Nor can an isotopic tracer be used for recovery determination, since a proportional counter can not distinguish one isotope from another. Therefore, an average recovery factor, determined yearly by experiment, is used in the calculation of each sample and this can lead to error in the result.

The SDWA allows for the conversion of the results from this method to μ g/L using a conversion factor of 0.67 pCi/ μ g. As long as the isotopes of uranium are present in natural abundance, this conversion is accurate. As stated above, disequilibrium of the uranium-238 and uranium-234 often occurs in groundwater making this conversion unreliable. When disequilibrium of the isotopes exists, using the conversion factor of 0.67 pCi/ μ g usually has the effect of biasing the mass concentration high. This is due to the fact that although uranium-234 accounts for only a tiny fraction of the total uranium mass, its activity is often more than double the activity of uranium-238 which accounts for more than 99% of the mass.

2.2 Total Concentration (2): ICP-MS EPA 200.8

2.2.1 Summary

This method prepares liquid samples for total metals analysis by ICP-MS. Digestion can reduce interferences from organic matter and convert metals to a form that can be determined by instrumentation.

The sample is cautiously evaporated to a low volume and refluxed with nitric acid until the digestate is light or stable in color. The digestate is then refluxed with hydrochloric acid, allowed to cool and brought up to volume. The results are reported as $\mu g/L$.

The method itself is not very labor intensive and the cost per analysis is \$29.80 for fiscal 2005 at the Wisconsin State Laboratory of Hygiene. However, the instrument is often off-line for repairs and requires highly trained analysts for proper operation.

2.2.2 Limitations

The SDWA allows for the conversion of the results from this method to pCi/L using a conversion factor of 0.67 pCi/ μ g. As long as the isotopes of uranium are present in natural abundance, this conversion is accurate. As stated above, disequilibrium of the uranium-238 and uranium-234 often occurs in groundwater making this conversion unreliable. When disequilibrium of the isotopes exists, using the conversion factor of 0.67 pCi/ μ g generally has the effect of biasing the radioactivity concentration low. This is due to the fact that the ICP method only measures the uranium-238 isotope which accounts for more than 99% of the mass of the three uranium isotopes.

2.3 Isotopic (3) : Eichrom Method ACW01 Revision 1.5

2.3.1 Summary

In order to concentrate the various actinides (thorium, uranium, neptunium, plutonium, and americium), the volume of a water sample is reduced to less than one liter by evaporation. Further, a sample with a low sulfate ion concentration is evaporated to dryness, the actinides being concentrated in the residue. When a sample has a high sulfate ion concentration, calcium sulfate would normally precipitate well before the sample had been evaporated to dryness. The calcium sulfate, which cannot be re-dissolved at any pH, will co-precipitate with the various actinides. Since the actinides must be re-dissolved for the chromatography steps, formation of a calcium sulfate precipitate is unacceptable. In this case phosphate anion is added to the high-sulfate sample, and the solution pH is adjusted to about 9 bringing about the precipitation of a calcium phosphate precipitate which contains the actinides.

Any calcium sulfate present in the sample is converted to calcium phosphate, and the sulfate ion is discarded with the supernatant. Next, the residue from the low-sulfate sample or the calcium phosphate from the high-sulfate sample is prepared for the chromatography steps by redissolution in a strong nitric acid solution. When this solution is poured through the UTEVA column, thorium, plutonium, and neptunium are eluted while uranium and americium remain on the column. The uranium is eluted at a pH of about 2, leaving the americium on the UTEVA column. Finally, the uranium is co-precipitated with cerium fluoride, the precipitate is collected on a filter, and the filter is mounted on a disc, ready for analysis by α -spectroscopy.

The results are reported in pCi/L. However, since the radioactivity of each isotope is determined, it is possible to directly convert the results from pCi/L to μ g/L. The individual values can then be summed for the total pCi/L or total μ g/L of uranium. A sample calculation can be found in attachment 1.

The fiscal year 2005 price at the Wisconsin State Laboratory of Hygiene is \$210.00.

2.3.2 Limitations

The method is labor intensive but produces results that can be reported as pCi/L or as μ g/L. Additionally, an isotopic tracer is used in the method to determine the recovery, making this method more reliable.

| Method No | Parameter | Results (pCi/L) Directly from method | Results (µg/L) Directly from method | convert results using 0.67 pCi/µg |
|--------------------------------|----------------|--|---|---|
| EPA 908.0 Total Uranium | Uranium, total | 32.8 ± 1.1 | NA | 48.9 μg/L |
| Eichrom ACW01 Alpha Spec | Uranium-238 | 9.7 ± 0.34 | 28.9 | |
| Éichrom ACW01 Alpha Spec | Uranium-235 | 2.1 ± 0.13 | 0.97 | |
| Eichrom ACW01 Alpha Spec | Uranium-234 | 24.3 ± 0.7 | 0.004 | |
| Eichrom ACW01 Alpha Spec | Uranium Total | 36.1 ± 0.8 | 29.9 | |
| EPA 200.8 ICP MS | Uranium-238 | NA | 25.7 ± 0.381 | 17.2 pCi/L |

Table 1 Results of a Sample Analyzed by Three Methods

3.0 Results

The data in Table 1 is from an actual sample taken from a water utility in Wisconsin. The sample was analyzed at the Wisconsin State Laboratory of Hygiene using three different methods. Using EPA method 908.0 the total uranium activity was determined to be 32.8 pCi/L. This activity was converted to µg/L using the conversion factor of 0.67 pCi/µg and was calculated to be 48.9 µg/L. The sample was then analyzed using the Eichrom ACW01 method and an activity was determined for each of the three isotopes. The activities of the three isotopes were summed for a total uranium activity of 36.1 pCi/L. Since the activity of each isotope was known it was possible to directly calculate the mass from the activity using the values given in Table 2 for specific activity. The individual results in μ g/L were then summed for a total uranium mass concentration of 29.9 μ g/L. Note that the mass concentration from the method 908.0 does not agree well with the mass concentration from the alpha spectroscopy method. Lastly, the sample was analyzed by EPA 200.8 ICP-MS and the mass concentration was determined to be 25.7 µg/L. This value compares well with the mass concentration determined from the alpha spectroscopy method of 29.9 µg/L. The mass concentration from the ICP-MS method was converted to activity using the conversion factor of 0.67 pCi/µg and was calculated to be 17.2 pCi/L. Note that this result does not compare well to the total activity determined by method 908.0 nor the alpha spectroscopy method.

4.0 Discussion

The data from Table 1 shows that each method adequately measures the amount of uranium, provided the results are reported in the units that the method was designed for. Biased data results from the conversion of one unit into another. The cause of the bias is the enrichment of uranium-234. In this sample, the activity of the uranium-234 is more than double the activity of uranium-238. Under conditions of natural abundance the uranium-238 and uranium-234 activities would be approximately equal. Referring to table 2, it can be seen that a tiny enrichment in the mass of uranium-234 will greatly increase the radioactive concentration of this isotope.

In the case of the analysis by ICP-MS, only the uranium-238 mass is detected. The tiny amount of uranium-234 present is not detected by the ICP-MS. The conversion of the mass concentration into activity was 17.2 pCi/L of total uranium. Under conditions of natural abundance the activities of uranium-238 and uranium-234 are about the same. Therefore, in this case one would predict 8.6 pCi/L of uranium-238 and 8.6 pCi/L of uranium-234 given a total activity of 17.2 pCi/L (17.2/2 = 8.6). Since this method can not measure the mass of uranium-234 the conversion of the results from mass to activity are biased low. The method does adequately measure the mass concentration because even when there is enrichment in uranium-234, more than 99% of the mass is still from the uranium-238.

In the case of the radiochemical method, EPA 908.0, the activity from all uranium isotopes is detected and reported as total uranium. The conversion of the activity concentration into mass was 48.69 μ g/L of total uranium. Under conditions of natural abundance the ratios of the mass of uranium-238 and uranium-234 are 99.27% and 0.0055% respectively. Therefore, in this case one would predict 48.3 μ g/L of uranium-238 and 0.0027 μ g/L of uranium-234 given a total mass of 48.69 μ g /L (48.69 X 0.9927= 48.3 and 48.6 X 0.000055=0.0027). Since this method can not measure the activity of the individual isotopes the conversion of the results from activity to mass are biased high. The method does adequately measure the total activity because it is not necessary to distinguish between the isotopes to get the total activity.

5.0 Conclusion

From a regulatory standpoint both the ICP-MS method and methods based on alpha spectroscopy will produce adequate results for mass concentration. However, for water utilities with gross alpha violations, the ICP-MS method may not help bring the utility into compliance when adjusting the gross alpha value because the activity will likely be biased low. From a public health standpoint, if the ICP-MS method is used, it will be easy to loose sight of the actual radioactivity of the sample because of the low bias. In an earlier study, the Wisconsin State Laboratory of Hygiene analyzed 70 groundwater samples for uranium-238 and uranium-234 by alpha spectroscopy (method 3). The ratio of uranium-234 to uranium-238 was computed and plotted in the histogram shown in Figure 3. Recall that under conditions of natural abundance the activity of uranium-238 and uranium-234 are equal and therefore, the ratio of uranium-234 to uranium-238 would be one. Of the 70 water samples analyzed, none of the ratios were equal to one. The largest ratio was 44.5 and the average was 13.5. In the case of the municipal water sample in this study, the activity of the uranium-234 is more than double that predicted by natural abundance.

Generally, the chemical toxicity of uranium is considered to be more of a concern than the radiotoxicity. However, there may be extreme cases where the level of radioactivity may warrant a concern but would be missed by the ICP-MS method.

The radiochemical method, EPA 908.0 will produce adequate results for radioactive concentrations. This activity can be used in the adjustment of the gross alpha value and may bring the water utility into compliance. However, the conversion of activity into mass will likely be biased high and may cause a violation of the uranium standard. The activity would have to be greater than 20 pCi/L for this to occur.

Finally, methods that use alpha spectroscopy produce the most accurate results for both activity concentrations and mass concentration. It is unfortunately, an expensive technique.



Figure 3 Histogram of U-234/U-238 Ratios

6.0 **Recommendations**

For groundwater samples requiring the results to be reported in both concentration (μ g/L) and activity (pCi/L), a conservative approach would be to have the sample analyzed by both a total concentration method and by a total activity method. Using the conversion of 0.67 pCi/ μ g often leads to inaccurate results. The approximate cost of a total concentration method is \$30 per sample and the approximate cost of a total activity method is \$150 per sample for a total of about \$180 per sample. There is probably a slight cost advantage for the previous test panel over the alpha spectroscopy method, which is \$210. The alpha spectroscopy method is the most sensitive and accurate of all of the techniques.

7.0 References

¹ Arndt, M. F., and West, L., A Study of the Factors Affecting the Gross Alpha Measurement, and a Radiochemical Analysis of some Groundwater Samples from the State of Wisconsin Exhibiting an Elevated Gross Alpha Activity, Final Report for Wisconsin Department of Natural Resources Groundwater Management Practice Monitoring Program, Project #176, Wisconsin Department of Natural Resources, 101 South Webster St, Madison WI 53703 (2004)

²Kigoshi, K., *Science* **173**, 47-48 (1971).

³Gilkeson, R. H., and E. C., Cowart, *Radium, Radon, and Uranium isotopes in ground water from Cambrian-Ordovician Sandstone Aquifers in Illinios* in *Radon in Ground Water-Hydrogeologic Impact and Indoor Air Contamination*, p. 402, Graves, Barbara, ed., Chelsea Mich., Lewis Publishers Inc. (1987).

⁴Federal Register / Vol. 65, No. 236 / Thursday, December 7, 2000 / Rules and Regulations 40 CFR Parts 9, 141, and 142 National Primary Drinking Water Regulations; Radionuclides; Final Rule

⁵Methods for the Determination of Metals in Environmental Samples. USEPA, 200.7, 1994

⁶ Krieger and E. L. Whittaker, *Prescribed Procedures for Measurement of Radioactivity in Drinking Water*, United States Environmental Protection Agency, Environmental Monitoring and Support Laboratory, EPA-6000/4-80-032, August 1980, p. 103-109.

⁷ *Ibid.* p. 96-102.

⁸Uranium in Water, Method ACW02, Rev. 1.3, Eichrom Technologies, Inc., Darien IL (2001).

8.0 Glossary

Activity - The rate of disintegration (transformation) or decay of radioactive material. The units of activity are the curie (Ci) and the becquerel (Bq).

Source: http://www.nrc.gov/reading-rm/doc-collections/cfr/part020/part020-1003.html

Alpha Particle - One of the particles emitted in radioactive decay. It is identical with the nucleus of the helium atom and consists, therefore, of two protons plus two neutrons bound together. A moving alpha particle is strongly ionizing and so loses energy rapidly in traversing through matter. Natural alpha particles will traverse only a few centimeters of air before coming to rest.

Source: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 57th Edition, CRC Press, Boca Raton, Florida (1976).

Anion - A negatively charged ion.

Source: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 57th Edition, CRC Press, Boca Raton, Florida (1976).

Anion exchange resin - An insoluble organic polymer containing cation groups that attract and hold anions present in a surrounding solution in exchange for anions previously held.

Source: http://www.answers.com/anion%20exchange%20resin

Beta Particle - One of the particles which can be emitted by a radioactive atomic nucleus. It has a mass about 1/1837 that of a proton. The negatively charged beta particle is identical with the ordinary electron, while the positively charged type (positron) differs from the electron om having equal but opposite electrical properties. The emission of an electron entails the change of a neutron into a proton inside the nucleus. The emission of a positron is similarly associated with the change of a proton into a neutron. Beta particles have no independent existence inside the nucleus, but are created at the instant of emission.

Source: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 70th Edition, CRC Press, Boca Raton, Florida (1990).

Concentration - The amount of a substance in weight, moles, or equivalents contained in unit volume.

Source: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 57th Edition, CRC Press, Boca Raton, Florida (1976).

Curie - A unit of radioactivity, equal to the amount of a radioactive isotope that decays at the rate of 3.7×10^{10} disintegrations per second.

Source: http://www.answers.com/curie

Decay - The spontaneous transformation of one radioactive nuclide into a daughter nuclide, which may be radioactive or may not, with the emission of one or more particles or photons. The decay of N_0 nuclides to give N nuclides after time t is given by $N = N_0 \exp(-\lambda t)$, where λ is called the decay constant or the disintegration constant. The reciprocal of the decay constant is the mean life. The time required for half of the original nuclides to decay (i.e., $N=\frac{1}{2}N_0$) is called the half-life of the nuclide. The same terms are applied to elemental particles that spontaneously transform into other particles. For example, a free neutron decays into a proton and an electron.

Source: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 70th Edition, CRC Press, Boca Raton, Florida (1990).

Electron-volt (symbol eV) – A unit of energy equal to the work done on an electron in moving it through a potential difference of one volt. It is used as a measure of particle energies although it is not an SI unit. 1 eV = 1.602×10^{-19} joule

Source: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 70th Edition, CRC Press, Boca Raton, Florida (1990).

Gram - One thousandth of a kilogram. The gram is the fundamental unit of mass in c.g.s units and was formerly used in such units as the gram-atom, gram-molecule, and gram-equivalent weight, which have now been replaced by the mole.

Source: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 70th Edition, CRC Press, Boca Raton, Florida (1990).

Gravimetric - Of or relating to measurement by weight.

Source: http://www.answers.com/gravimetric

Half-life - The time required for half of the nuclei in a sample of a specific isotopic species to undergo radioactive decay.

Source: http://www.answers.com/half-life



Isotope - (1) One of several nuclides having the same number of protons in their nuclei, and hence belonging to the same element, but differing in the number of neutrons and therefore in mass number A, or in energy content (isomers). For example ${}^{12}_{6}$ C, ${}^{13}_{6}$ C, and ${}^{14}_{6}$ C are carbon isotopes. Small quantitative differences in chemical properties exist between isotopes or orientation.

Source: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 70th Edition, CRC Press, Boca Raton, Florida (1990).

Isotopic tracer - An isotopic tracer, (also "isotopic marker" or "isotopic label"), is used in chemistry and biochemistry to help understand chemical reactions and interactions. In this technique, one or more of the atoms of the molecule of interest is substituted for an atom of the same element, but of a different (often radioactive) isotope. Because the atom has the same number of protons, it will behave in almost exactly the same way chemically as other atoms in the compound, and with few exceptions will not interfere with the reaction under investigation. The difference in the number of neutrons, however, means that it can be detected separately from the other atoms of the same element.

Mass spectrometry can also be used with this technique, since mass spectra recorded with sufficiently high resolution can distinguish among isotopes based on the different masses resulting from the different number of neutrons.

Source: http://www.answers.com/isotopic%20tracer

Natural abundance - The relative abundance of an isotope in nature compared to other isotopes of the same element is relatively constant. The Chart of the Nuclides presents the relative abundance of the naturally occurring isotopes of an element in units of atom percent. Atom percent is the percentage of the atoms of an element that are of a particular isotope. Atom percent is abbreviated as a/o.

Source: http://www.tpub.com/content/doe/h1019v1/css/h1019v1_39.htm

Nuclide - A species of atom distinguished by the constitution of its nucleus. The nuclear constitution is specified by the number of protons, Z; number of neutrons, N; and energy content. (Or, by the atomic number, Z; mass number A(= N + Z) and atomic mass).

Source: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 70th Edition, CRC Press, Boca Raton, Florida (1990).

Picocurie - One trillionth (10^{-12}) of a curie.

Planchet - A small shallow metal container in which a radioactive substance is deposited for measurement of its activity Source: <u>http://www.answers.com/planchet</u>

Precipitation - The process of separating a substance from a solution as a solid. Source: <u>http://www.answers.com/precipitation</u>

Radioactivty - The spontaneous disintegration of certain atomic nuclei accompanied by the emission of alpha-particles (helium nuclei), beta-particles (electrons), or gamma-radiation (short-wave electromagnetic waves).

Natural radioactivity is the result of the spontaneous disintegration of naturally occurring radioisotopes: most radioisotopes can be arranged in three radioactive series. The rate of disintegration is uninfluenced by chemical changes or any normal changes in their environment. However, radioactivity can be induced in many nuclides by bombarding then with neutrons or other particles.

Adapted From: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 70th Edition, CRC Press, Boca Raton, Florida (1990).

Spectrometer - An instrument which separates radiation into energy bands (or, in a mass spectrometer, particles into mass groups) and indicates the relative intensities in each band or group. See, for example, Figure 2.

Source: http://history.nasa.gov/EP-95/glossary.htm

Attachment 1

| Isotope | Half-life in years ($t_{1/2}$) | Atom % natural | Specific Activity | Alpha particle energies in MeV | Branching Ratios |
|---------|----------------------------------|-------------------|----------------------|-----------------------------------|---------------------|
| U 238 | 4.47 billion | 99.27 | 0.336 | 4.039 | 0.0023 |
| | | | | 4 147 | 0.22 |
| | | | | 4.14/ | 0.25 |
| | | | | 4.196 | 0.77 |
| U 235 | 700 million | 0.72 | 2.17 | 4.1525 | 0.009 |
| | | | | 4.2157 | 0.057 |
| | | | | 4.3237 | 0.046 |
| | | | | 4.3641 | 0.11 |
| | | | | 4.370 | 0.06 |
| | | | | 4.3952 | 0.55 |
| | | | | 4.4144 | 0.021 |
| | | | | 4.5025 | 0.017 |
| | | | | 4.5558 | 0.042 |
| | | | | 4.5970 | 0.050 |
| U 234 | 246,000 | 0.0055 | 6208.2 | 4.604 | 0.0024 |
| | | | | 4.7231 | 0.275 |
| | | | | 4.776 | 0.725 |

Table 2Uranium Isotopes

Half-life data taken from http://www.epa.gov/radiation/radionuclides/uranium.htm.

Alpha Particle Energies taken from: Weast, R. C., *CRC Handbook of Chemistry and Physics*, 70th Edition, CRC Press, Boca Raton, Florida (1990).

Derivation of factor used to convert between pCi and µg assuming natural abundance.

U-238 0.9927 X 0.336 pCi/µg = 0.333 pCi/µg

U-234 0.000055 X 6208.2 pCi/µg = 0.341 pCi/µg

U-235 0.0072 X 2.17 = 0.016 pCi/µg

If the isotopes are present in natural abundance then the ratio of pCi/µg is; 0.333+0.341+0.016=0.69~pCi/µg

Attachment 2

Derivation of specific activity

 $t_{\frac{1}{2}}$ = half-life of the isotope given in units of seconds

N = number of nuclides in units of nuclides per gram

 $\lambda = \frac{\ln(2)}{t_{\frac{1}{2}}} = \text{decay constant in units of s}^{-1}$

 $A = \lambda \times N$ = specific activity in units of pCi/ µg

U-238

$$t_{\frac{1}{2}} = 1.41 \times 10^{17}$$
 seconds (s)

$$\lambda = \frac{\ln(2)}{1.41 \times 10^{17}} = 4.91 \times 10^{-18} \text{ s}^{-1}$$

$$N = \frac{6.02 \times 10^{23} nuclides / mole}{238 grams / mole} = 2.53 \times 10^{21} \text{ nuclides/gram}$$

$$A = (4.91 \times 10^{-18} \, \text{s}^{-1}) \times (2.53 \times 10^{21} \, \text{nuclides} \, / \, \text{g}) \times \left(\frac{1 \, pCi}{3.7 \times 10^{-2} \, \text{decays} \, / \, \text{s}}\right) = 336,000 \, pCi \, / \, \text{g}$$

or 0.336 pCi/µg for U-238

U-235

$$t_{\frac{1}{2}} = 2.21 \times 10^{16} \text{ seconds (s)}$$

$$\lambda = \frac{\ln(2)}{2.21 \times 10^{16}} = 3.14 \times 10^{-17} \text{ s}^{-1}$$

$$N = \frac{6.02 \times 10^{23} \text{ nuclides / mole}}{235 \text{ grams / mole}} = 2.56 \times 10^{21} \text{ nuclides/gram}$$

$$A = (3.14 \times 10^{-17} \text{ s}^{-1}) \times (2.56 \times 10^{21} \text{ nuclides / g}) \times \left(\frac{1\text{ pCi}}{3.7 \times 10^{-2} \text{ decays / s}}\right) = 2,170,000 \text{ pCi / g}$$

or 2.17 pCi/µg for U-235

U-234

 $t_{\frac{1}{2}} = 7.76 \times 10^{12}$ seconds (s)

$$\lambda = \frac{\ln(2)}{7.76 \times 10^{12}} = 8.93 \times 10^{-14} \text{ s}^{-1}$$

 $N = \frac{6.02 \times 10^{23} \text{ nuclides / mole}}{234 \text{ grams / mole}} = 2.57 \times 10^{21} \text{ nuclides/gram}$

$$A = (8.93 \times 10^{-14}) \times (2.573 \times 10^{21} \, nuclides \, / \, g) \times \left(\frac{1 \, pCi}{3.7 \times 10^{-2} \, decays \, / \, s}\right) = 6208200000 \, pCi \, / \, g$$

or 6208.2 pCi/µg for U-234