The Relationship Between Deuterium Excess and Uranium Groundwater Concentrations in Whitehall, Montana

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Abstract

Uranium can become elevated in the environment and pose a human health risk to water resources. Therefore, it is important to understand the origin, transport and concentration of uranium in the environment. With this exploratory study we propose that deuterium (δ^2H) excess may be a viable way to study uranium groundwater concentrations that originate from the surface. The Boulder Batholith of southwestern Montana has been associated with varying levels of uranium. The Whitehall, MT area includes the Boulder Batholith as a geologic unit. We collected 23 water samples and compiled data from a study done by the United States Geological Survey (USGS) within the area of Whitehall, MT. With a linear regression we show that there is a significant negative relationship between deuterium excess and uranium water concentrations. As expected we showed that there is a significant positive, linear relationship between uranium water concentrations and nitrate water concentrations. As expected we also showed that there is a significant positive, linear relationship between calcium carbonate and uranium groundwater concentrations for the compiled USGS data. The results of a Wilcoxon rank sum test showed that median uranium concentration for wells close to ponds is 14.5 $\mu g/L$ higher for our data added to the USGS data. These results may be consistent with a model that involves insoluble uranium being transported in suspension and settling out in ponds where an influx of nitrates and/or carbonates react to produce higher concentrations of water-soluble forms of uranium. We suggest that the relationship found between uranium concentration and deuterium excess in water samples may be related to the higher rates of evaporation in ponds. Thus, in this specific hydrologic environment of valley ponds close to the Boulder Batholith source, deuterium excess may serve to help predict levels of uranium concentrations.

Key Words: groundwater, uranium concentrations, water isotopes, carbonates, nitrates, Jefferson County

Introduction

Uranium (U) exists in varying levels in the soil, air and water of the natural environment. When uranium levels become elevated in water it can pose certain environmental problems. When uranium is dissolved in drinking water, at and above $30\mu g/L$, it poses a significant health risk to humans. These health risks include a slight increased risk of cancer, and kidney complications for drinking water at and above $30\mu g/L$ (CDC 2016). This is important for area populations that rely on groundwater as a main drinking water source. The Whitehall, MT area of Jefferson County mainly relies on groundwater for the population's residential and municipal drinking water needs. A past water quality study done by the United States Geological Survey (USGS) showed that Jefferson County, as well as surrounding counties in Southwestern MT, have groundwater extents with dissolved uranium present (Caldwell R. et al. 2013).

The potential for uranium to enter and become elevated in the groundwater can depend on many different factors. Granitic batholiths can often, but not always, contain detectable levels of uranium. The Boulder Batholith in Southwestern Montana is one geologic unit known to contain varying levels of uranium (Roberts, et al. 1951). The USGS conducted a three-year-long study to assess the uranium (and other radionuclides) groundwater concentrations of wells within Jefferson County, MT and surrounding areas that contain the Boulder Batholith as a geologic unit (Caldwell, et al. 2013). The Elkhorn volcanic range is related to the Boulder Batholith and may be a source of uranium for this area (Caldwell R. et al. 2013, Roberts et al. 1951, Smedes 1966). The alluvium and Boulder Batholith below the surface were also identified as potential sources of uranium for this area.

In this exploratory study, we attempt to use deuterium excess to study uranium concentrations in the Whitehall area. This exploratory study is based on environmental data and therefore not definitive research. There is also no specific known past research into the use of deuterium excess to study groundwater concentrations of uranium. However, the use of stable water isotopes to study chemical groundwater concentrations has been researched in the past (Sidle 1998). This is because the heavier water isotopes become enriched or depleted in natural waters relative to the lighter water isotopes. This process is referred to as isotope fractionation. One way that the heavier water isotopes become enriched in natural waters is through the preferential evaporation of the lighter water isotopes. Ponds for example have a higher ratio of stable water isotopes in comparison to rainwater and streams due to the preferential evaporation of the heavier water isotopes. Because, deuterium excess is calculated from both the stable water isotopes of oxygen (^{18}O) and deuterium (^{2}H or δD) from isotopic data it may be a more sensitive measure of evaporation (Frohlich et al. 2002, Keesari et al. 2017). Past studies have attempted to understand uranium groundwater concentrations using stable water isotopes to produce mixing models based on the relative age of water sources as well as an attempt to produce a direct relationship to ^{18}O (Helling 2000, Lonschinski et al. 2010, Kumar et al. 2014, Pant et al. 2017). In this exploratory study deuterium excess is used to attempt to understand uranium groundwater concentrations based on evaporation.

Assessing nitrate (NO_3^-) and nitrite (NO_2^-) concentrations for this study is important as they have the potential to increase the solubility of uranium minerals through oxidative dissolution (Gronowski, A. 2013, Nolan et al. 2015). Carbonates may also play a role in reacting with insoluble uranium minerals and producing more water-soluble complexes (Goodwin 1981, Abdelouas et al. 1998, Chau et al. 2011, Gronowski, A. 2013).

The purpose of this study was to study the spatial distribution of uranium groundwater concentrations in the Whitehall area with stable water isotopes as well as nitrate and carbonate concentrations. The study was completed as an undergraduate research project and funded by the University of Montana Western Student Senate through a learning grant. The future application of this study is a basis for a better understanding of the variables that affect uranium groundwater concentrations. This study is primarily an exploratory investigation into the uranium concentrations of wells in a specific area of the Boulder Batholith of Whitehall, Montana.

Study Area

This study was done in the area of Whitehall, MT. The approximately 61km^2 study area included 18 groundwater samples, 2 snow samples, and 3 surface water samples. These samples come from private and municipal wells as well as public access sites. The elevation within the study area ranges from 1200m to 1400m with most of the sample points located in the valley of Whitehall, MT area. Whitehall, MT and the surrounding area is classified

as having a semi-arid-climate. The average annual precipitation in Whitehall, MT is 24.13cm. The common native plants in the study area includes Big Sagebrush (*Artemisia tridentata*), Blue-bunch Wheatgrass (*Pseudoroegneria spicata*), Broom Snakeweed (*Gutierrezia sarothrae*) and Fringed Sagewort (*Artemisia frigida*). The sampling points are shown below over a geologic map of the area in Figure 1.

Geologic Setting

The main geologic feature in the area that is pertinent to this study is the Elkhorn Mountain range, which is mostly composed of Tertiary through Cretaceous aged extrusive igneous rocks. These extrusive and intrusive igneous rocks of the Elkhorn Range are closely related to the Boulder Batholith (Roberts et al. 1951, Smedes 1966). The generation of magma of the Boulder Batholith was caused by the subduction of the Farlon oceanic plate under the North American plate during the Cretaceous. The extrusive igneous rocks of this area resulted from a conduit of the mainly quartz monzonite Boulder Batholith. These volcanic igneous rocks of the Elkhorn Range intruded through older Mesoproterozoic to Mesozoic sedimentary and metamorphic rocks of the range (Roberts et al. 1951, Smedes 1966). The Boulder Batholith has been known to contain elevated levels of radionuclides (Caldwell R. et al. 2013). The valley of Whitehall area is mainly composed of undifferentiated gravel or alluvium of the Quaternary. This Quaternary as well as Tertiary sediment is the source of many of the wells in the valley of Whitehall area. There are also many wells in this area that draw from fractures in igneous or metamorphic source rock. The depth of the wells in this study area ranged from 8m to 107m.
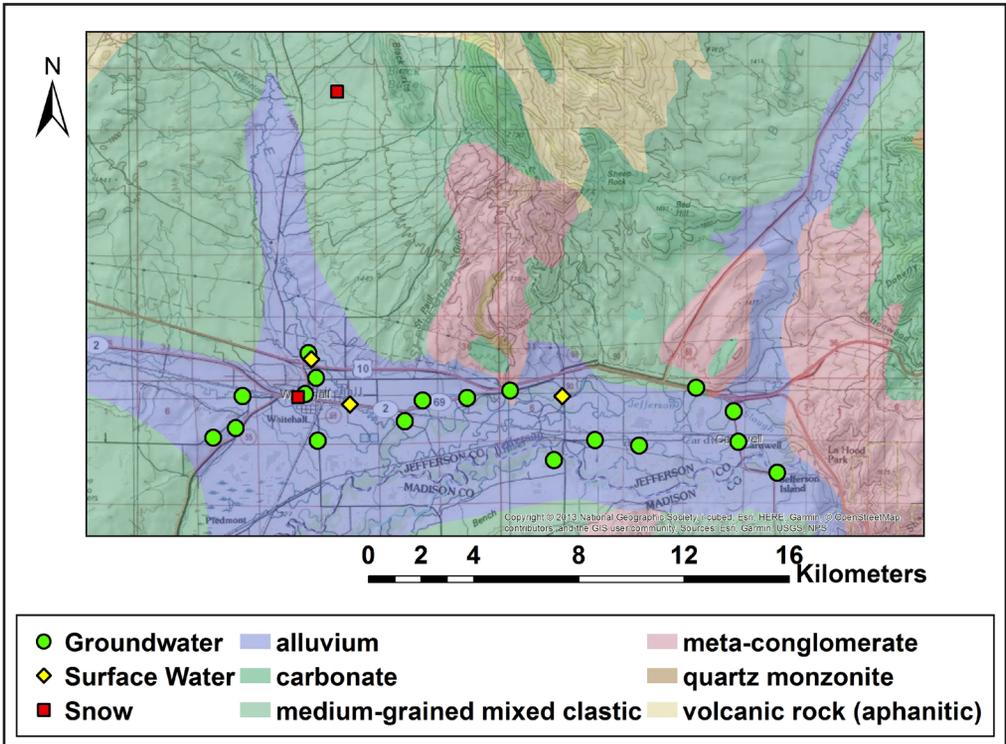
Figure 1. Geologic map of the Whitehall, MT area with the general sampling locations for the field data we collected. These points are generalized and do not reflect definitive sampling locations for this study (ESRI ArcMap 10.6, 2017).

Uranium Groundwater Concentration Factors

The main variables that affect uranium groundwater concentrations from the surface include origin, liberation, transport, and accumulation. The process that liberates uranium from the origin is often the same process that allows it to become mobile (Gronowski, A. 2013). The accumulation phase simply identifies the point at which uranium enters the groundwater aquifer and begins to accumulate over time. The element uranium exists in the natural environment as an isotopic mix of ^{234}U, ^{235}U and ^{238}U. The uranium element has six valence electrons and is a highly reactive metal in the natural environment. The two most common oxidative states of uranium that are found in the environment are the U(VI) and U(IV) valence forms (Gronowski, A 2013). These two valence forms that uranium exists in the natural environment serve as the base for numerous uranium minerals, and complexes. The uranium minerals associated with the Boulder Batholith in this area have been studied in the past and include pitchblende, torbernite, rutherfordine and others (Roberts and Gude, 1953). These uranium complexes and minerals have various solubility characteristics (Závodská et al. 2008). It should be noted that many other uranium complexes can form in the natural environment (Erikson et al. 1990, Závodská et al. 2008, Chau et al. 2011, Nielsen, E. et al. 2013,). The uranium complexes that form through reactions with carbonates and nitrates have been identified as important on how uranium becomes water soluble (Gronowski A. 2013). It should be noted that any one of these uranium minerals can become transported in suspension (Gronowski, A 2013).

METHODS AND MATERIALS

The study area was first established as an area within the valley from Whitehall to Cardwell. The sample points for this study consist of 18 groundwater, two snow, and three stream samples. One snow sample was new snow that precipitated the day of collection and the second was not. The materials used in this study included general field equipment such as a Garmin handheld GPS unit, data sheets, a pH and TDS meter, a YSI meter, and 65 sterile Nalgene bottles of 40ml, 300ml, and 500ml.

The 23 sample sites included the collection of a GPS point, water sample pH, temperature, total dissolved solids (TDS), and a general site description. The water sampling procedures of this study followed the standards outlined by the USGS (U.S. Geological Survey 2017). The 23 water samples were stored in a refrigerator for no longer than ten days before being sent to the lab in Butte for analysis. The 23 water samples were sent to the Montana Bureau of Mines and Geology lab in Butte, Montana for the analysis of water isotope ratios, total nitrate and nitrite concentrations in milligrams per liter, and uranium in micrograms per liter. The isotopic data is represented in standard percent per million as and denoted as δ. An example of the equations used to calculate deuterium (D) excess is shown below.

$$(\delta \text{ or } \permil \text{ of deuterium}) = \left(\frac{\frac{^2H}{H}(Sample)}{\frac{^2H}{H}(VSMOW\ Std.)} - 1 \right) \cdot 1000, (D\ excess = \delta D - 8\ \delta^{18}O)$$

(Kendell et al. 2001, Lee et al. 2003, Shama 2018)

Statistical Analysis

The data used for this study was analyzed statistically using Microsoft Excel 2016 and MATLAB 2018. The descriptive statistics were calculated in Excel and the statistical tests were conducted in MATLAB. Uranium groundwater concentrations and δD excess for this study were run through a regression. The data for the uranium water concentrations and the nitrate/nitrite ground and surface-water concentrations were also run through a regression with one outlier. The 8 nitrate/nitrite samples below the detection limit were run as 0 for both nitrate/nitrite and uranium. The data from the USGS study for uranium groundwater concentrations and hardness as calcium carbonate was also run through a regression. The uranium water concentration data was categorized by pond distance based on the mean distance (>168m or <168m) and run through a Wilcoxon Rank-Sum (WRS) test as the data was not from a normal distribution. Ponds were identified as approximately larger than a 100-meter perimeter using Google Earth version 9.2.93.1. The data for all tests used a 0.05 level of significance ($\propto$ =0.05).

RESULTS

The uranium water concentrations for all samples had a maximum of 44.4$\mu g/L$ and a minimum of <0.2$\mu g/L$. The total nitrite and nitrate for all samples had a maximum of 10.3 (mg/L) and a minimum of <0.2(mg/L). The complete list of parameters analyzed including pH, total dissolved solids (TDS), temp (Celsius), δO^{18}, δD (or ‰), uranium ($\mu g/L$) and total nitrite and nitrate (mg/L), can be found in Appendix A.

The isotopic data for this study is a plot of the $\delta^{18}O$, and δ^2H (δD) for the groundwater, surface water, and snow samples(Fig. 2).

Scatter plots for the uranium water levels vs the δ^2H (δD) excess levels, and the uranium water concentrations and total nitrate/nitrite water levels for the Whitehall are depected in Figure 3 and Figure 4.

There is a significant negative linear relationship between uranium water concentrations and δD excess levels (t_{18} = - 3.18, $p = 0.0058$, $R^2 = 0.38$). The linear regression line for this data is $y = 11.65 - 3.540x$. This data includes the 18 groundwater samples from the Whitehall area.

There is a significant positive relationship between uranium water concentrations and total nitrate nitrite concentrations ($t_{20} = 3.663$, $p = 0.00178$, $R^2 = 0.42$). An outlier was removed for this regression. The outlier was discounted due to a potential influx of nitrates at the site where it was collected. The linear regression line for this uranium and nitrate scatter plot is $y = 5.49 + 9.24x$ (Fig. 5).

The results for the linear regression of the USGS groundwater data showed that there is a significant positive relationship between uranium water concentrations and hardness as calcium carbonate concentrations ($t_{66} = 6.295$, $p = 2.846e^{-8}$, $R^2 = 0.38$). The linear regression line for this USGS data is $y = -6.03 + 0.107x$. This regression consists of data compiled from the USGS 2007-2010 study as calcium carbonate was not measured in this study.

Table 1. The descriptive statistics for the uranium and total nitrate and nitrite concentrations of the surface water and groundwater samples for the Whitehall, MT area (2019).

Sample Source (U)	Mean ($\mu g/L$)	SD	Sample Size
Groundwater Samples	14.9	12.6	18
Surface Water Samples	14.6	1.76	3

Sample Source (N)	Mean (mg/L)	SD	Sample Size
Groundwater Samples	2.18	3.008	18
Surface Water Samples	.48	0.0316	3
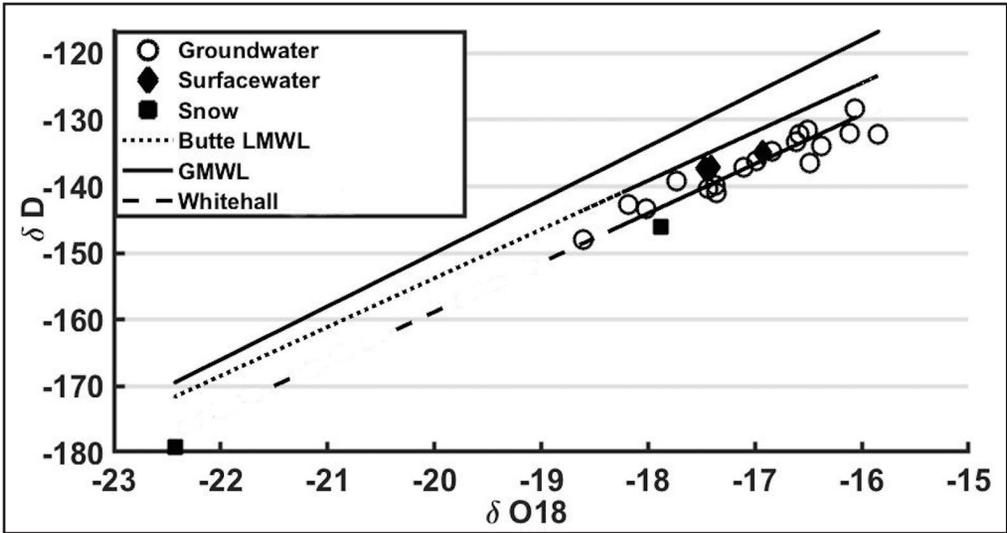
Figure 2. Isotopic data collected in our study which is displayed with the LMWL for the Butte, MT area based on past research (Gammons et al. 2006) and the GMWL (Craig H. 1961), (2019). The dotted line depicts the LMWL for the Butte area for comparison ($\delta^2 H = 7.32\ \delta O^{18} -7.32$). The GMWL is also depicted in this figure as the solid line for comparison ($\delta^2 H = 8\ \delta O^{18} + 10$).
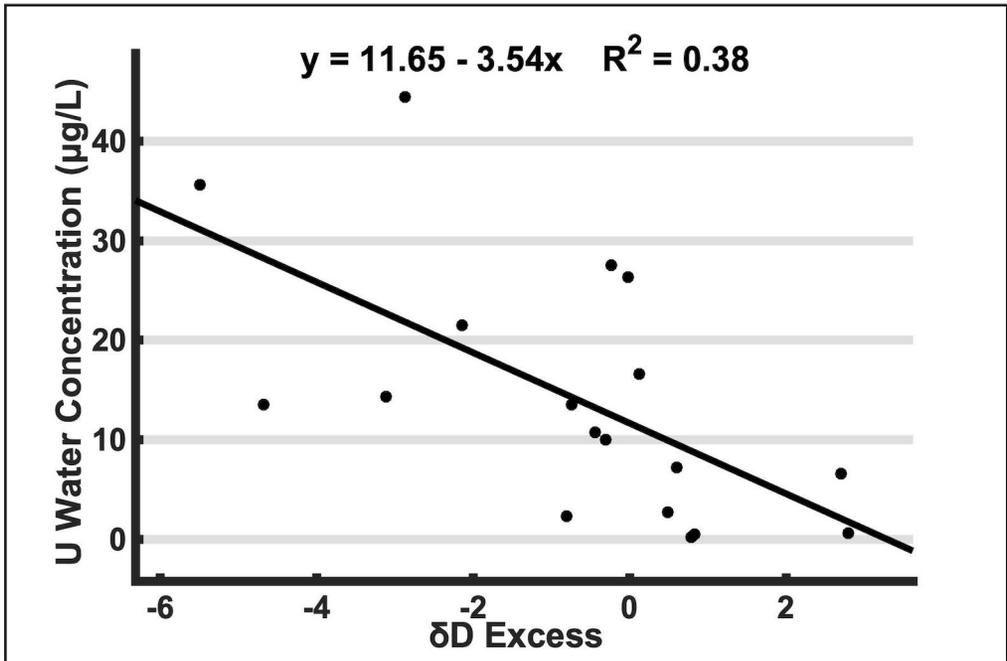
Figure 3. Scatter plot for the uranium water concentrations and deuterium excess of the 18 groundwater samples collected in the Whitehall, MT area (2019).

The figure referenced in this section is a boxplot that depicts the median uranium water concentrations for wells close to ponds (≤168m, 8 samples) and the median uranium water concentrations for wells far from ponds (>168m, 12 samples).(Fig. 6)

The results of the Wilcoxon Rank-Sum test showed that the median uranium concentration for wells close to ponds was significantly higher than the median uranium
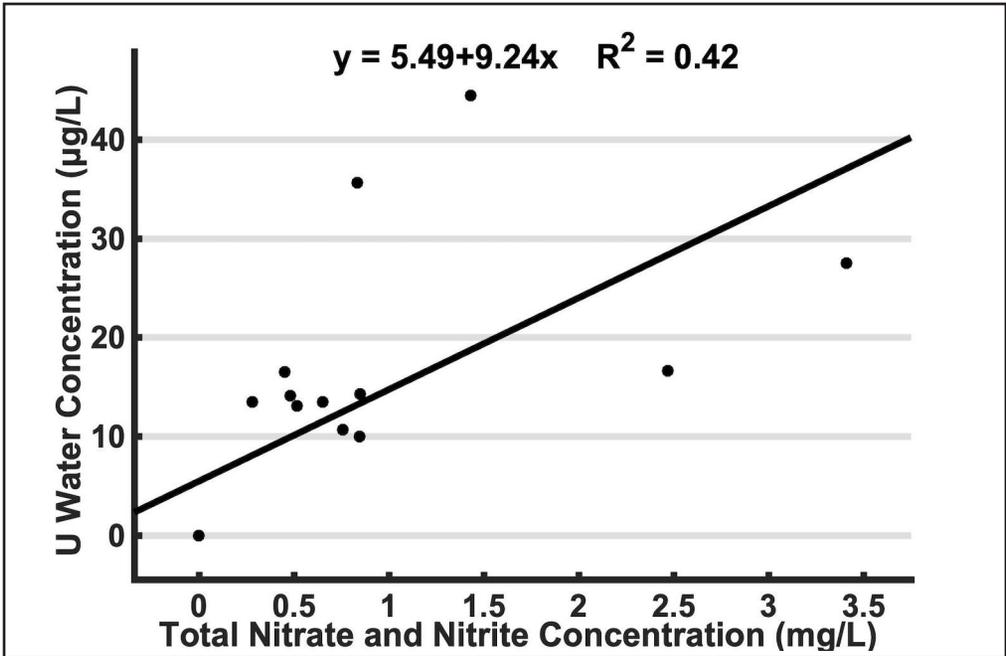
Figure 4. Scatter plot for the uranium water concentrations and total nitrate and nitrite concentrations of 20 surface water and groundwater samples collected in the Whitehall, MT area (2019). One groundwater sample had an abnormally high nitrate concentration (10.3mg/L compared a mean of 2.18mg/L for all other samples) and was not included in this linear regression and scatter plot. The outlier was discounted due to a potential influx of nitrates at the site where it was collected. The 8 samples below the detection limit of 0.02 were run as 0 for both uranium water concentrations and nitrate/nitrite concentrations for this test.
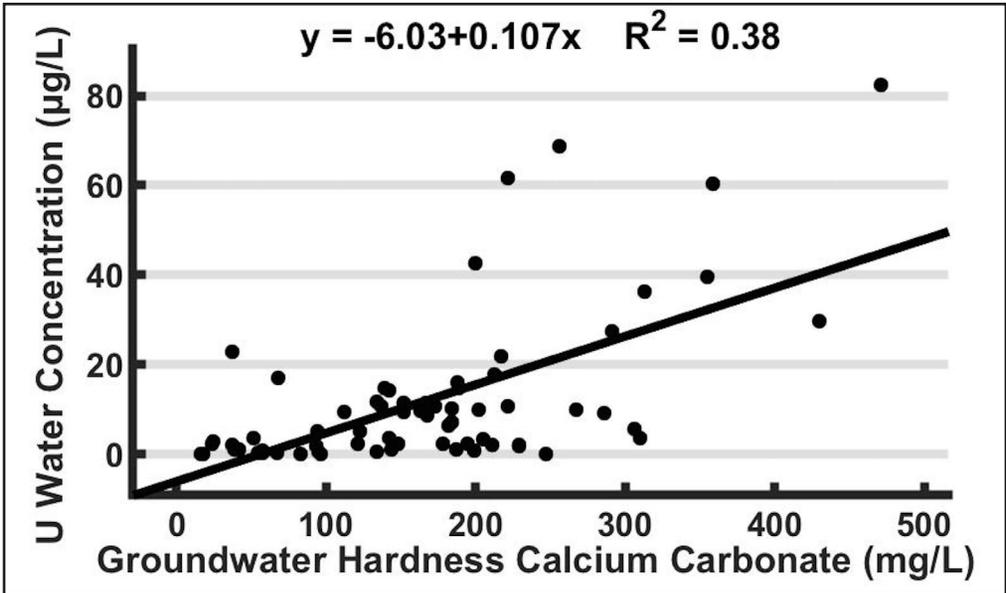
Figure 5. Scatter plot for the uranium water concentrations and calcium carbonate hardness of 66 of the groundwater samples collected in Jefferson County, MT by the United States Geological Survey from 2007 to 2010.
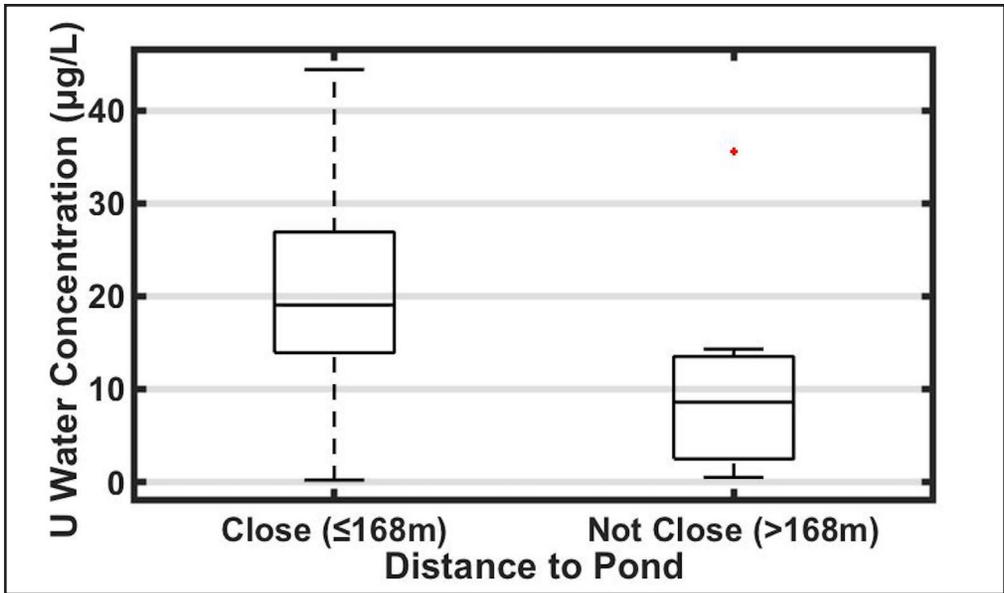
Figure 6. Box plot of median uranium groundwater concentrations for wells close to (≤168m) and not close to ponds (>168m). This distance was based on the average distance from the ponds to the wells for this data. This figure displays data collected for this study and data collected by the United States Geological Survey from 2007 to 2010. The data was collected from the Whitehall, MT area (2019).

groundwater concentration for wells not close to ponds ($WRS_{8,12}$, $W = 111.5$, $p = 0.0338$). The wells close to ponds were 14.5 $\mu g/L$ higher for uranium than wells far from ponds.

Discussion

The understanding of the transport, source, and concentration of uranium in the environment is important for populations that rely on groundwater as a main source of drinking water. Our exploratory study might show that there is some influx of uranium into the groundwater aquifer from the surface. We were able to show this based on the significant value from comparing the deuterium excess and the uranium water concentrations through a linear regression. This relationship can be related to the differing isotopic ratios of water influx from the surface into the groundwater aquifer. The study done by researchers in Punjab Pakistan was not able to find a relationship between ^{18}O and uranium groundwater concentrations (Pant et al. 2017). It may be that deuterium excess

might serve as a more suitable comparison to understanding uranium water concentrations for this area because of its close relationship with evaporation (Frohlich et al. 2002, Lee et al. 2003, Keesari et al. 2017). Or there is less connectivity from the surface to the groundwater in their study area. A relationship between uranium in groundwater and an enrichment in the heavier water isotopes recharging the groundwater may be possible if we can relate it to evaporation. However, it should be noted that the isotopic data suggests that recharge to the aquifer during the short sampling period of this study was affected less by evaporation than the average LMWL which may be why our data falls below it (Gammons et al. 2006). It's also likely that deuterium excess may not be a good fit because it is typically calculated from meteoric water instead of groundwater and there is little past research to support a relationship (Lee et al. 2003, Penga et al. 2012, Pant et al. 2017). It should also be noted that water isotope data is typically collected over a longer period than the time period of this study.

The positive relationship between uranium water concentration and total nitrate (NO_3^-) and nitrite (NO_2^-) concentration has been found by researchers at the University of Nebraska (Nolan et al. 2015) and has also been linked to nitrate dependent microbes that may oxidize uranium into a more water soluble form U(VI) (Senko et al. 2005). These authors speculate that insoluble or less soluble uranium minerals are being oxidized while the nitrates and nitrites are being reduced. The positive relationship between uranium concentrations and groundwater hardness as calcium carbonate ($CaCO_3$) has also been studied as a potential mode for uranium to become more water soluble (Goodwin 1981, Erikson et al. 1990, Gronowski, A. 2013).

The significantly higher median for uranium groundwater concentration of wells close to (≤168m) ponds compared to wells not close to ponds (>168m), may indicate how uranium is being transported on the surface for the Whitehall area. This might suggest that there are uranium minerals being transported in suspension from the source on the surface. The insoluble uranium compounds then settle out in the valley ponds. The Piedmont Pond in the Whitehall area may fit this model due to its relatively elevated uranium concentration of 23.1$\mu g/L$, as well as being relatively enriched in the stable water isotopes and the presence of calcium carbonate (182.7mg/L). The Piedmont Pond does not however appear to be affected by an influx of nitrates or nitrites (<0.02mg/L) (MBMG et al. 2012).

The higher median for uranium groundwater concentration of wells close to (≤168m) ponds compared to wells not close to ponds (>168m), suggests that insoluble uranium minerals are being transported to the valley ponds. The Piedmont Pond in the Whitehall area has been measured by the Montana Bureau of Mines and Geology (MBMG) and they found a relatively elevated uranium concentration of 23.1$\mu g/L$, relatively enriched stable water isotopes (δD: -104.0, $\delta^{18}O$: -10.5) and the presence of calcium carbonate (182.7 mg/L). The Piedmont Pond does not however appear to

be affected by an influx of nitrates or nitrites (<0.02mg/L), (MBMG et al. 2012).

We then may relate the significant result of pond distance to wells back to our first three significant test results displayed. The ponds might have a higher ratio of the stable water isotopes compared to their mainly snow melt source, and other surface water bodies due to evaporation (Penga et al. 2012). We see this for the Piedmont pond in the Whitehall area which is relatively enriched in the stable water isotopes (MBMG et al. 2012). This might relate to decreasing deuterium excess (which is calculated from the isotopic data) being related to increasing uranium concentrations. If we assume that some amount of insoluble uranium is settling out in the ponds, we might also assume that it becomes soluble at some point to enter the groundwater aquifer. An influx of nitrates, nitrites and calcium carbonate into the ponds may react with the insoluble uranium compounds and allow them to become water soluble. It's also likely that the relationship between uranium and ponds has nothing to do with insoluble uranium settling out in the ponds as they often have little to no surface flow influx. It may be more related to the ponds being a sink for nitrate/nitrites, and carbonates which might increase the solubility of uranium.

We can now propose a potential model for an influx of uranium from the surface into groundwater of the Whitehall area.

1. The uranium minerals may be liberated from the Elkhorn volcanic rock of the Boulder Batholith on the surface.
2. The uranium minerals are transported from the source to the valley
3. An influx of nitrates, nitrites and carbonates react with the insoluble uranium compounds and form water soluble uranium compounds.
4. The water-soluble uranium compounds then enter the groundwater aquifer from the valley ponds which can be directly related **to** deuterium excess.

This model is based on an exploratory study based on environmental data and therefore not definitive research. We suggest that future research related to this exploratory study include samples from the ponds in the Whitehall, MT area. The analysis of calcium carbonate, nitrate, and uranium concentration as well as water isotope ratios for future samples to test our model presented in this study. Future research related to this study should also include water isotope data collected over a longer period, and groundwater samples to compare to the pond data in the Whitehall, MT area. The model presented in this exploratory study may also apply to further research in other study areas where uranium is present in the groundwater. The model presented might only account for one part of how uranium enters the groundwater in this area. Other sources of uranium discussed most likely play a part in how uranium enters the groundwater and this study should be viewed solely as an exploratory analysis.

ACKNOWLEDGEMENTS

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Appendix of Results A.

Sample #	Temp C°	pH	TDS (ppm)	Uranium μg/L	NO3/NO2⁻ N mg/L	$\delta^{18}O$ (‰)	$\delta^{2}H$ (‰)
1	10	7.17	164	2.67	< 0.2	-16.5	-131.5
2	11	7.1	165	2.29	< 0.2	-17.4	-139.8
3	11	7.15	166	16.6	2.47	-16.1	-128.4
4	9	7.25	169	27.5	3.41	-17.0	-136.2
5	11	7.31	370	13.5	0.281	-16.5	-136.5
6	13	7.56	202	< 0.2	< 0.2	-18.0	-143.4
7	12	7.41	186	26.3	< 0.2	-16.8	-134.7
8	12	7.2	175	44.4	1.43	-16.4	-133.9
9	11	7.23	170	0.503	< 0.2	-18.6	-148.0
10	11	7.18	150	35.6	0.834	-15.8	-132.2
11	11	7.21	14	NA	NA	-17.9	-146.1
12	11	7.22	131	7.18	< 0.2	-16.6	-132.1
13	11	7.21	143	21.5	10.3	-17.4	-141.0
14	16	7.56	301	0.565	< 0.2	-18.2	-142.7
15	17	7.16	206	10	0.847	-17.1	-137.1
16	17	7.11	189	6.54	< 0.2	-17.7	-139.2
17	17	7.12	153	10.7	0.758	-16.6	-133.4
18	17	7.18	123	13.5	0.652	-17.4	-140.3
19	17	7.14	205	14.3	0.848	-16.1	-132.0
20	17	7.18	127	13.08	0.516	-17.5	-137.3
21	17	7.16	141	14.1	0.481	-17.4	-137.2
22	17	7.26	186	16.5	0.453	-16.9	-134.8
23	17	7.35	10	NA	NA	-22.4	-179.1