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An evaluation of health risk to the public as a consequence of *in situ* uranium mining in Wyoming, USA

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Highlights

- Most groundwater constituents do not change as a result of alkali ISR mining
- After restoration, ^{226}Ra decreases by about one half, ^{nat}U increases by factor of about five
- Radiological risk decreases, nephrotoxic biomarkers increase, clinical significance unclear

Abstract

In the United States there is considerable public concern regarding the health effects of in situ recovery uranium mining. These concerns focus principally on exposure to contaminants mobilized in groundwater by the mining process. However, the risk arising as a result of mining must be viewed in light of the presence of naturally occurring uranium ore and other constituents which comprise a latent hazard. The United States Environmental Protection agency recently proposed new guidelines for successful restoration of an in situ uranium mine by limiting concentrations of thirteen groundwater constituents: arsenic, barium, cadmium, chromium, lead, mercury, selenium, silver, nitrate (as nitrogen), molybdenum, radium, total uranium, and gross α activity. We investigated the changes occurring to these constituents at an ISR uranium mine in Wyoming, USA by comparing groundwater quality at baseline measurement to that at stability (post-restoration) testing. Of the groundwater constituents considered, only uranium and radium-226 showed significant ($p<0.05$) deviation from site-wide baseline conditions in matched-wells. Uranium concentrations increased by a factor of 5.6 (95% CI 3.6-8.9 times greater) while radium-226 decreased by a factor of about one half (95% CI 0.42-0.75 times less). Change in risk was calculated using the RESRAD (onsite) code for an individual exposed as a resident-farmer; total radiation dose to a resident farmer decreased from pre- to post-mining by about 5.2 mSv y^{-1}. Higher concentrations of uranium correspond to increased biomarkers of nephrotoxicity, however the clinical significance of this increase is unclear.

1. Introduction

Approximately one eighth of the world's electricity (Nuclear Energy Institute 2014) is supplied by 435 nuclear reactors (Nuclear Energy Agency and International Atomic Energy Agency 2014). Over 58,000 tonnes of uranium ore were mined in 2014 to supply fuel for these reactors (Nuclear Energy Institute 2014). In 2013, 47% of world-wide uranium production was the result of *in situ* recovery (ISR) mining. In the United States, the fraction of uranium production attributable to ISR is much higher (World Nuclear Association 2015a). Domestically, seven operational ISR mines produce 2300 tonnes of uranium per year, approximately 11% of domestic uranium consumption (United States Energy Information Administration 2014), which powdered 100 reactors generating one fifth of the nation's electricity (Nuclear Energy Institute 2014). ISR is the most economically efficient method of uranium extraction in the United States and an important generator of economic activity in rural parts of the country (e.g., Wyoming and South Dakota). However, there are risks associated with ISR uranium mining, most notably the contamination of a drinking-water aquifer with uranium or other heavy metals (United States Environmental Protection Agency 2008).

The ISR process utilizes a series of injector and recovery wells to access, without excavation, below-ground uranium orebodies. The chemistry of the ISR process varies with both geological and regulatory conditions; in the United States groundwater is pumped from recovery wells to the surface, where it is fortified with dissolved oxygen (or, less commonly hydrogen peroxide), carbon dioxide, and/or sodium bicarbonate, and then re-injected. Following each subsurface pass, the groundwater, now laden with uranium, is sent through ion-exchange resins for uranium recovery and re-fortified/rejuvenated. The circulation is a closed loop except for a small "bleed" (typically 0.5 to 1% of the total flow) maintained to prevent mine water from leaving the mining zone. When the ion exchange bed is filled to capacity with uranium, it is taken off-line and eluted; the resulting eluent is chemically treated to produce uranyl peroxide. A more detailed description of the ISR process can be found in Davis and Curtis (2007).

The ISR mining process may be conducted with either an acid or alkali agent. Currently, only the alkali process is used in the United States, although other countries such as Australia and Khazakstan employ acid leach processes. Here, we only consider the alkali processes as these are the techniques used at our field site.

The goal of this study was to quantify the risk resulting from changes in groundwater, induced by ISR at a uranium mine in Wyoming, USA. Our hypothesis was that post-restoration (stability) conditions on-site represent either (1) a significant increase in groundwater constituents beyond the range of conditions found naturally on-site prior to mining, and/or (2) a significant increase in risk to a resident farmer.

2. **Material and Methods**

2.1. Description of Site

The Smith Ranch-Highlands ISR uranium mine is located in Converse County, Wyoming, USA (Figure 1). The site is at 1500 m elevation and experiences a semi-arid climate, with an average annual temperature of 7°C and average annual precipitation of 319 mm (National Oceanic and Atmospheric Administration 2002). Wyoming is sparsely populated, with 584,000 inhabitants occupying 250,000 square kilometers, an area roughly the size of the United Kingdom. The dominant ground cover is mixed grass prairie (State of Wyoming 2010). Pronghorn antelope (*Antilocapra americana),* mule deer (*Odocoileus hemionus*), white tailed deer (*Odocoileus virginianus*), and game birds such as wild turkey (*Meleagris gallopavo)*, ducks, and Canadian geese (*Branta canadensis*), which are consumed by some local residents*,* are frequently found on-site.

Borch et al (2012) provide a detailed description of subsurface conditions at Smith Ranch-Highlands. Generally, uranium ore has been deposited in a sandy layer approximately 150-200 m below ground. The ore bearing sands are bounded above and below by shale deposits. Groundwater flow is estimated to be approximately 2-3 m per year. Uranium concentrations in ore body solids vary from a few hundredths of a percent to about one percent, with the bulk average typically being approximately 0.1%. Uranium ore deposits develop in permeable formations that are generally sandy. Borch et al (2012) describe the hydrogeological processes that deposit mineable uranium: within groundwater, soluble and mobile ^{nat}U(VI) moves along a hydrologic gradient. If it encounters reducing conditions, ^{nat}U(VI) precipitates to eventually form minerals such as uraninite, coffinite, and uranophane. The zone of solid uranium within the aquifer is known as a roll front. Figure 2, adapted from Davis and Curtis (2007), illustrates the formation of a uranium roll front.

Four well fields were considered for this study, each consisting of multiple wells in a set pattern. The well fields were brought online asynchronously, and so are different stages of their lifecycle. Because of the asynchronicity of the wells' life stages, and the fact that mining is still ongoing at Smith Ranch Highlands, not all wells have been in each phase of operation.

2.2. Source of Data

Baseline groundwater quality data was provided by Cameco Resources for well fields 1, 3, 4, and 4A. Stability (a regulatory phase wherein a post-production well field is monitored for geochemical stability prior to being released) testing has only been conducted for a subset

of wells in well field 1; this data was also provided by Cameco Resources. The baseline and stability samples were collected as part of site characterization and routine monitoring to satisfy regulatory requirements. Samples were collected and analyzed over an approximately eighteen-year period; the majority of these analyses were performed by Energy Labs Inc., in Casper, Wyoming. More recently, Intermountain Laboratories in Sheridan, Wyoming has been used for analysis.

We performed statistical analysis on thirteen groundwater constituents considered to be of interest to the United States Environmental Protection Agency (USEPA), as well as a handful of other indicators of groundwater quality (total dissolved solids, alkalinity, pH) to ascertain the scope of the change induced to groundwater by the ISR process.

2.3. Statistical Analysis

Figure 3 shows our process for determining whether a statistically robust change occurred in concentrations of a groundwater constituent between baseline and stability operational phases. Detailed analysis has been conducted on ground water, as described in section 2.2. Many of the constituents of interest to the USEPA are below minimum detectable concentrations (MDCs) in the groundwater at the Smith Ranch-Highlands site. Others were not tested for in the data provided to us by Cameco Resources. Those species with concentrations below the experimental MDC, in addition to those species for which analysis was not performed, were eliminated from consideration; table 1 details which constituents were eliminated at this stage. It should be noted that all MDCs were below US Safe Water Drinking Act (SWDA) concentrations except for Pb; SWDA concentrations are reported in table 2. Because the Pb MDC was above the SWDA regulation, it is possible that concentrations of Pb at Smith Ranch Highlands exceed regulatory limits for potable water. However, we lacked data to make this determination and opted to exclude Pb from our analysis.

A series of Welch's t-tests were run to examine whether stability (post-restoration) conditions in well field 1 (the only well field for which stability testing has been conducted) differed significantly from (1) baseline conditions in the same well field and (2) baseline conditions throughout the site. The results of these tests indicate whether or not the concentrations found in stability differ significantly from those at baseline, or from naturally

occurring variability throughout the Smith Ranch-Highlands site. In order to obtain a normal distribution suitable for a parametric test, concentrations of ^{226}Ra and ^{nat}U were log transformed prior to t-testing.

Finally, we retested those constituents that had a significant change from baseline to stability using the subset of wells for which both baseline and stability concentration data were available. We will refer to this subset of wells as matched wells.

R[1] version 3.1.2 was used for all statistical computations.

2.4. Biosphere Transport Model

Simply measuring the scope of groundwater change as a result of ISR mining is insufficient for estimating the change in risk to humans. Food chain transfer as well as natural environmental processes influence the quantity of a contaminant to which humans are exposed.

A resident farmer scenario was modelled at Smith Ranch Highlands using the US Department of Energy's RESRAD (onsite) code[2]. A resident farmer is an individual who moves onto the former mine site at some unspecified time in the future and engages in subsistence agricultural practice using a groundwater well. This scenario is not implausible; there are a handful of abandoned homes located on the Smith Ranch-Highland site used by homesteaders during the 20th century. The former occupants of these homes were likely engaged in activities similar to those selected for our model: ranching of livestock such as cattle and gardening to produce vegetables for human consumption. We modeled the source well as located within the uranium ore body in order to maximize the exposed resident farmer's exposure to groundwater contaminants[3].

The resident farmer was modeled as exposed through consumption of food, water and soil, with food separately including dairy, meat, and plants. No aquatic foods were considered due to the lack of natural surface water on or near Smith-Ranch Highland. All produce

[1] http://www.r-project.org/
[2] https://web.evs.anl.gov/resrad/
[3] Input parameters for our model are available online as electronic supplement 1.

consumed was assumed to have come from a garden within the mined area. Ingestion and inhalation rates were set to the values recommended by the USEPA's exposure factor handbook (United States Environmental Protection Agency 2011), with the exception of meat consumption, which was increased to compensate for the lack of aquatic foods. Climate data for Converse County, Wyoming was taken from the reports of the United States National Oceanic and Atmospheric Administration (National Oceanic and Atmospheric Administration 2002).

Our RESRAD (onsite) model took advantage of a source override procedure that allows a radionuclide source to be placed directly in groundwater. The model then ignores any below-ground geochemistry, holding the concentration of a contaminant in groundwater constant over time by adjusting the distribution coefficient (K_D) to an appropriate value. This is reasonable for our exposure scenario: before site release can occur, the operator must establish that the belowground environment has reached chemical equilibrium, and that concentrations of groundwater contaminants are constant. Horizontal and vertical transport through the belowground environment were neglected, as these processes are unimportant for our exposure scenario and, due to site hydrogeology, insignificantly small over a human's lifespan. Direct external exposure and inhalation radiation dose were included in the model, but these contributed negligibly to overall radiation dose.

3. *Results*

3.1. Effect of ISR Mining on Groundwater Quality

3.1.1. Constituents Above the Experimental MDC

Of the thirteen species considered to be of interest to USEPA, only As, Se, total N (NO_3+NO_2), ^{226}Ra (as total Ra), and ^{nat}U were detectable above analytic limits. We chose to include TDS, alkalinity, and pH in this analysis as general indicators of change in groundwater quality. Per the process depicted in Figure 3, all these constituents were passed on to the next level of analysis.

3.1.2. Same Well Field Comparison, Baseline to Stability

Stability concentrations of As, Se, total N (NO_3+NO_2), ^{226}Ra, and ^{nat}U, TDS, alkalinity, and pH were compared to baseline concentrations in well field 1, to determine if groundwater conditions after mining were within the baseline natural variation of well field 1. Stability concentrations were significantly different ($p<0.05$) from baseline

variability within well field 1, and all these constituents were passed on to the next level of analysis. The results of statistical testing are presented in table 3.

3.1.3. Well Field to Site Wide Comparison, Baseline to Stability

Stability concentrations of As, Se, total N (NO_3+NO_2), ^{226}Ra, and ^{nat}U, TDS, alkalinity, and pH were compared to baseline concentrations across the Smith Ranch-Highlands site, to determine if changes in groundwater quality due to mining were within site-wide natural variation at baseline. The statistical tests demonstrated that only alkalinity, As, ^{226}Ra, TDS, and ^{nat}U in the stability well field differed significantly from the naturally occurring variation across the Smith Ranch-Highlands site at baseline. The results of the tests are presented in table 3. Boxplots of ^{nat}U and ^{226}Ra concentrations, in baseline and stability, are given in figure 4.

3.1.4. Matched Well Comparison, Baseline to Stability

Stability concentrations of As, Se, total N (NO_3+NO_2), ^{226}Ra, and ^{nat}U, TDS, alkalinity, and pH were compared to baseline concentrations across the Smith Ranch-Highlands site, including only matched wells, that subset of wells for which both baseline and stability data was available.

Figure 5 depicts concentrations for matched wells at baseline and at stability. The t-test results were identical to those comparing stability to site-wide baseline: alkalinity, As, ^{226}Ra, TDS, and ^{nat}U showed significant variation. It should be noted that alkalinity is not regulated by the USEPA and median concentrations of As were well below the USEPA's regulatory limits for potable water. Only TDS, ^{nat}U, and ^{226}Ra exceeded USEPA's regulatory limits for potable water and showed variation between site-wide baseline and well field stability testing concentrations. As TDS is not one of the thirteen constituents of interest to USEPA, only ^{nat}U and ^{226}R were included in the risk assessment.

Table 4 presents the change in groundwater concentrations of ^{nat}U and ^{226}Ra from baseline to stability, as well as the results of the statistical test. As expected, the mining process oxidized ^{nat}U(IV) to ^{nat}U(VI), mobilizing the uranium in groundwater. More surprising was a dramatic reduction in ^{226}Ra in groundwater. We speculate that this may have occured because radium is mobile in groundwater prior to the onset of mining, and

subsequently removed from groundwater by the ion exchange process. This is unlike uranium, which is actively mobilized from ore by the mining process, thereby introducing additional uranium into groundwater. However, it is difficult to propose a mechanism of change that accounts for the totality of changes in the subsurface environment as a result of mining, as changes in groundwater concentrations were not consistent. At Smith Ranch Highlands, we found that after mining, As increased relative to pre-mining (levels remained below regulatory limits for potable water, both before and after mining), while TDS and alkalinity decreased. Median concentrations of Se stayed approximately the same, but the distribution of concentrations spread significantly[4]. Subsurface geochemistry is enormously complex, and influenced by both physical and biological conditions (Campbell et al 2012), further obscuring the exact mechanism by which geochemical changes occur. Such uncertainties underscore the need for the detailed measurement and modelling of the ISR process on a site-specific basis.

3.2. **RESRAD (onsite) Results**

Model results demonstrated that ^{226}Ra contributes far more to radiation dose than ^{238}U for a resident farmer at the Smith Ranch-Highlands site. Given that uranium is understood to be more chemically than radiologically toxic, this is expected. A detailed description of total dose from each radionuclide, based on concentrations in matched-wells at baseline and stability, is presented as table 4. The model unsurprisingly identified water as the primary exposure pathway; a detailed description of total dose from each pathway is presented as table 5.

3.3. Risk Analysis

Radium and uranium represent different types of risk to exposed populations. While exposure to ^{226}Ra results in a radiological risk, uranium's toxic mechanism in the body is principally chemical, not radiological (Wrenn et al 1985, Leggett 1989, Taylor and Taylor 1997, Guseva Canu et al 2011, Kurttio et al 2002). Biological endpoints for radiation exposure are either stochastic (increased risk of cancer) at low doses or deterministic (direct damage) at high doses. This is in contrast to uranium, which like many heavy metals, causes kidney damage in exposed humans (Zamora et al 1998, Mao et al 1995, Thun et al 1985,

[4] Detailed boxplots of groundwater concentrations are available as electronic supplement 2.

Russell et al 1996, Kurttio et al 2002). This damage occurs at uranium exposures far below those necessary to induce stochastic or deterministic radiation effects due to radioactive decay (Guseva Canu et al 2011).

Because of the chemotoxic mechanism of uranium, it is difficult to harmonize the risk arising from increased exposure to uranium with the risk from decreased exposure to radium: exposure to radium at the concentrations found at Smith Ranch Highlands confers a risk of cancer years in the future, while exposure to uranium confers a risk of near-term kidney damage. Thus, we have elected to consider each component of risk separately, commenting on both the magnitude of the change in risk as well as expected public health outcomes.

3.3.1. Cancer risk following exposure to ^{nat}U and ^{226}Ra

Table 6 summarizes radiation dose rate results, which were calculated with RESRAD (onsite) using concentrations of ^{226}Ra and ^{nat}U from groundwater in matched wells, at baseline and in stability. Uranium concentrations increased from baseline to stability, resulting in a 0.36 mSv y^{-1} increase in radiation dose to the resident farmer. ^{226}Ra concentrations decreased from baseline to stability, resulting in a 5.6 mSv y^{-1} decrease in radiation dose rate. Overall, mining resulted in a 5.2 mSv y^{-1} decrease in radiation dose to the resident farmer. This corresponds to a reduction of 260 mSv over a 50 year adult lifetime. The BEIR VII report (2006) concludes that every additional 100 mSv of radiation dose received corresponds to a 1% increase in cancer (solid cancer or leukemia) incidence (Biological Effects of Ionizing Radiation VII 2006). Thus the reduction in dose rate as a result of mining corresponds to a 2.6% decrease in cancer incidence compared to a resident farmer consuming water from the pre-mining aquifer.

3.3.2. Nephrotoxicity of ^{nat}U

Guseva Canu et al (2011) undertook a review of available studies on uranium toxicity as a result of consuming naturally contaminated water. In total they identified 27 "peer-reviewed published reports of original epidemiological studies, including studies of uranium, radium, and radon" from the period of 1970-2011 (Guseva Canu et al, 2011). Of these studies, seven assessed the incidence of renal damage as a function of exposure to naturally occurring uranium (Mao et al 1995, Zamora et al 1998, Kurttio et al 2002, Kurttio et al 2005, Kurttio et al 2006, Seldén et al 2009,

306 Zamora et al 2009) with approximately 1500 subjects across all seven studies. The
307 average concentration of uranium consumed in water in these studies varied between
308 25 μgU L^{-1} and 180 μgU L^{-1}; with this level of uranium exposure statistically correlated
309 with various biomarkers of renal damage. The values reported are comparable to our
310 study site's baseline median of 73 μgU L^{-1} and stability median of 411 μgU L^{-1} in
311 matched wells. In short, while an exact threshold for uranium nephrotoxicity has not
312 been established, it likely occurs somewhere around the baseline median value at
313 Smith Ranch-Highlands. Kurttio et al (2006) report a modest (on the order of 10%)
314 increase in biomarkers of kidney dysfunction as concentrations of uranium in water
315 increase from tens of μgU L^{-1} to hundreds or thousands of μgU L^{-1}, but also note that:

316 > The clinical significance of [our] results is not easily established… Tubular
317 > dysfunction manifested within the normal physiological range, but occurred
318 > without an apparent threshold. Excretion of calcium, phosphate, and glucose
319 > remained within normal range in most subjects, even for persons with very
320 > high and long-lasting exposure to uranium. These findings are consistent with
321 > studies of occupational exposure to uranium failing to demonstrate overt
322 > kidney disease among workers exposed to uranium. (Kurttio et al, 2006)

323 Additionally, the authors note that renal failure is associated with only very high (10-
324 25 mgU kg^{-1} body weight) acute exposures to uranium (Kurttio et al 2006). For an
325 adult human of 60 kg, this corresponds to a 0.6-1.5gU ingestion – the equivalent of
326 drinking, at a minimum, 1500 L of stability mine water in a single, acute exposure.

327

328 4. ***Discussion***

329 **4.1. Implications of Risk Analysis**

330 The results of this study indicate that it is possible that radiation dose (from radium) is
331 actually slightly decreased as a result of ISR uranium mining. The increased concentration
332 of uranium in groundwater due to ISR uranium mining would point to an increased risk of
333 nephrotoxicity, but not cancer, with no impact on mortality (Agency for Toxic Substances
334 and Disease Registry 2013). Since radium and uranium each have an independent biological
335 endpoint for risk, it difficult to harmonize the two changes in risk. We did not find that other
336 groundwater constituent concentrations changed significantly as a result of mining,

although they may have been present in sufficient quantities – before and/or after mining – to impact human health.

It should be emphasized that this model was a "worst case" analysis of wellfield 1, based on the most contaminated groundwater, which is found directly within the ore body. Due to high site variability, drilling a well just tens of meters away from the ore body may dramatically lower an individual's risk.

4.2. Limitations and Uncertainties

Limitations to this analysis can be loosely grouped into three categories: dose/effects relationships, characterization uncertainties, and uncertainties associated with variability/heterogeneity.

Dose/effects uncertainties are those uncertainties that are present in epidemiologic or toxicologic studies on the effects of a stressor on humans. For example, the BEIR VII (2006) report dataset has some important confounders: acute exposures to radiation may be more harmful than a continuous low dose exposure, which is the exposure scenario more applicable to our resident farmer scenario. We have also not considered the increased sensitivity associated with exposure to radiation or other stressors during sensitive life stages, such as exposure to children or to the fetus *in utero*, principally because there is limited data concerning uranium toxicity variability between life stages. Finally, the interactions between multiple stressors may be important for understanding the net effect of the consumption of uranium mine water on an individual. Multi-stressor theory holds that the net effect of multiple, simultaneous exposures may be greater than the sum of exposures given individually. Given the number of constituents in mine water, such analysis is relevant to conducting risk analysis at an ISR mine.

Characterization uncertainty arises as a result of limitations in the sampling scheme or analytic techniques used. In this study, well water samples were not continuously collected and analyzed for their chemical content. Rather, well water was collected and analyzed at discrete times, sometimes years apart; how groundwater quality varied between time points is not known. Stability testing is meant occur over several years, in part to address this concern by establishing that groundwater concentrations remain nearly-constant after

restoration is complete. Additionally, samples were not analyzed for the chemical speciation of uranium; some uranium complexes are not cytotoxic, e.g, calcium-uranyl-carbonato complexes (Prat et al, 2009). Depending upon the speciation of uranium in groundwater at Smith Ranch-Highlands, the risk of groundwater consumption may be lower than estimated here.

Site wide variability should also be considered when interpreting the results of this study. Our results were for a single well field (the only well field for which stability testing has been conducted). Given the variability of groundwater quality across the Smith Ranch Highlands site (for e.g., uranium, as shown in Figure 4), it is likely that our result, or any single result, is not generalizable between well fields or mine sites. To understand the changes to groundwater resulting from ISR uranium mining requires a complete understanding of site-conditions.

5. Conclusions

Our statistical analysis and risk-based impact assessment at an ISR mine in Wyoming, USA had some unexpected results: chiefly, that ^{226}Ra concentrations in groundwater decreased dramatically as a result of the mining process. This type of analysis may prove useful to regulators and operators alike, providing a paradigm to arrive at site closure criteria that is more flexible than the current approach, which requires adherence to rigid groundwater standards and does not account for natural background or variability. Geoscience Australia (2010) recommends that "For lease relinquishment, regulators should be confident that the rehabilitated site does not present any significant radiation exposure risks, impacts on groundwater quality are as limited as is practicable, and the site will be fit for agreed future land uses." Risk analysis, such as that undertaken here, would ideally be based on data coupled with geo- and biosphere transport codes, as appropriate and should be used to assist in setting site closure criteria. A data-driven approach is necessary at ISR uranium mines, where predictive modelling of changes to subsurface geochemistry resulting from mining may not be reliable, and intra- and inter- site variability is extremely high, making adherence to a generalized standard at sites with diverse geochemical conditions impractical.

Finally**,** the USEPA notes that there is "only very limited data in the open literature" concerning the stability of restored ISR well fields in the United States (USEPA 2014). This lack of data makes

it difficult to establish regulatory endpoints and is damaging both to both regulators and mining operations. While it may be tempting to require mining companies to bear the financial burden for generating this data, such work is better funded and conducted by impartial institutions, e.g., federal or state governments, universities, or other independent agencies. Such a separation would ensure the integrity and pedigree of reported data by preventing conflicts of interest.

6. Future Work

The results of this study were unexpected and have underscored how poorly studied ISR uranium operations are in the United States. Further work investigating the magnitude of impacts that ISR uranium mining has on the geosphere and biosphere – both in the short and the long term – is critical to the establishment of a reasonable regulatory structure and the protection of the public.

Within the next decade, additional data will become available as more ISR mine units near the end of their life cycle: such data should be the subject of additional analysis, similar to the one conducted as a part of this study, so that the impacts of ISR uranium mining on groundwater quality may be better understood.

7. Acknowledgements

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8. Conflict of Interest

This study was financed by the State of Wyoming. Cameco Resources provided no financial assistance. The authors maintain technical and personal relationships with employees of Cameco Resources but have never been employed by nor accepted any remuneration from that organization.

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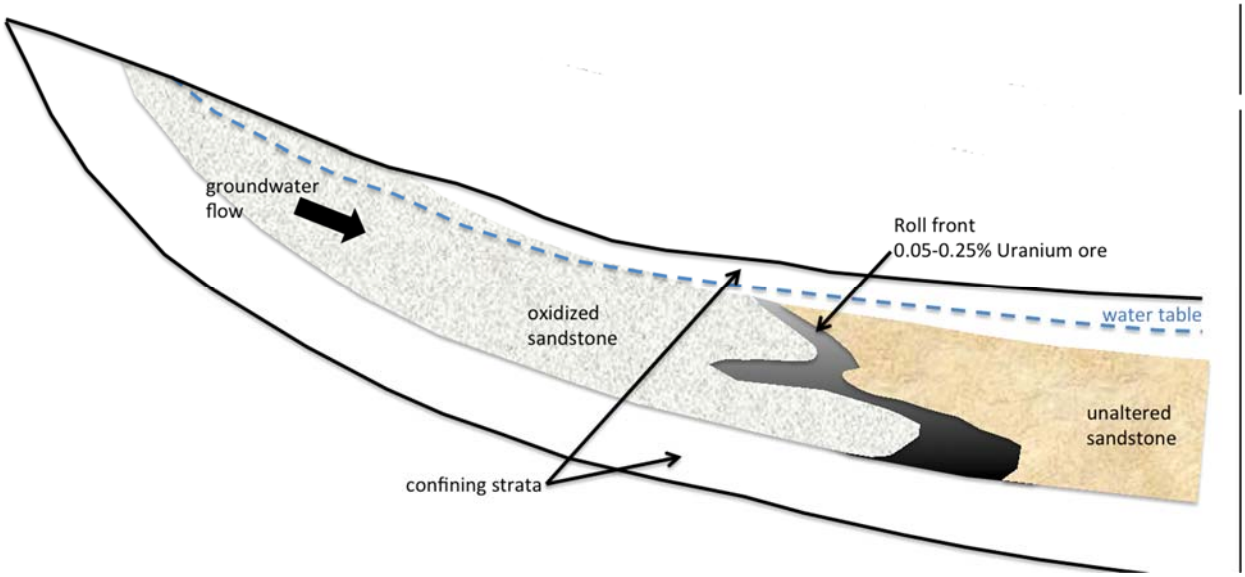
Figure 1: Map showing location of the Smith Ranch-Highlands site and photograph depicting site landscape and a typical well-field.

Figure 2: The formation of a uranium roll front. Adapted from Davis and Curtis (2007).



546 **Figure 3** Flowchart describing how contaminants were eliminated from risk analysis.

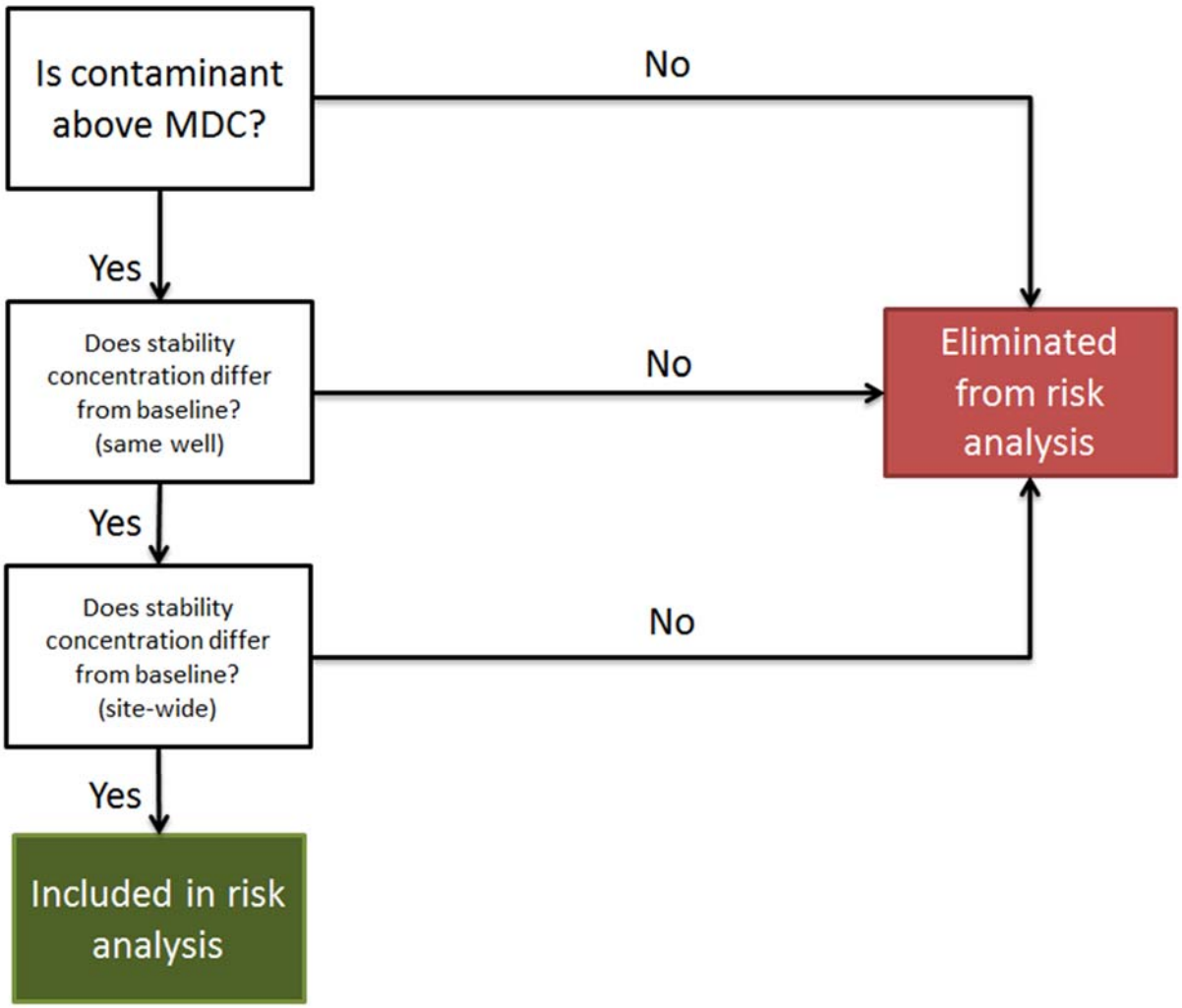


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Figure 4 Stability concentrations of ^{226}Ra and ^{nat}U in well field 1 compared to baseline conditions site-wide. USEPA's limit appears as a pink horizontal line.

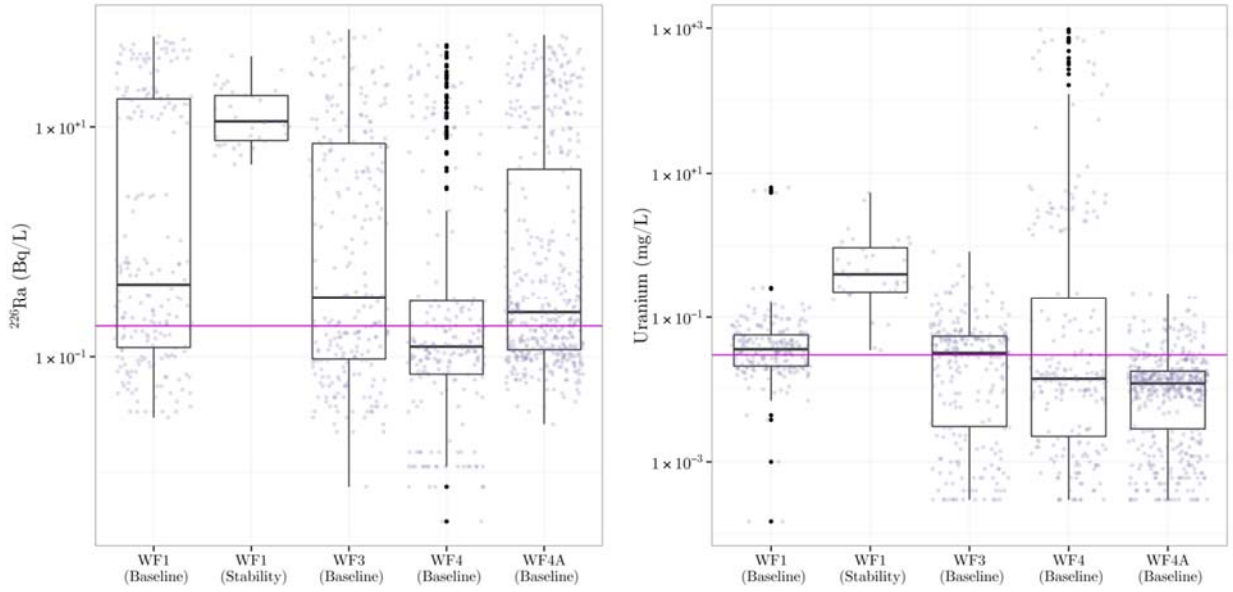

Figure 5 Stability concentrations of ^{226}Ra and ^{nat}U in well field 1 compared to baseline concentrations in well field 1. Only matched wells - tested both at baseline and in stability - are displayed. USEPA's limit appears as a pink horizontal line.

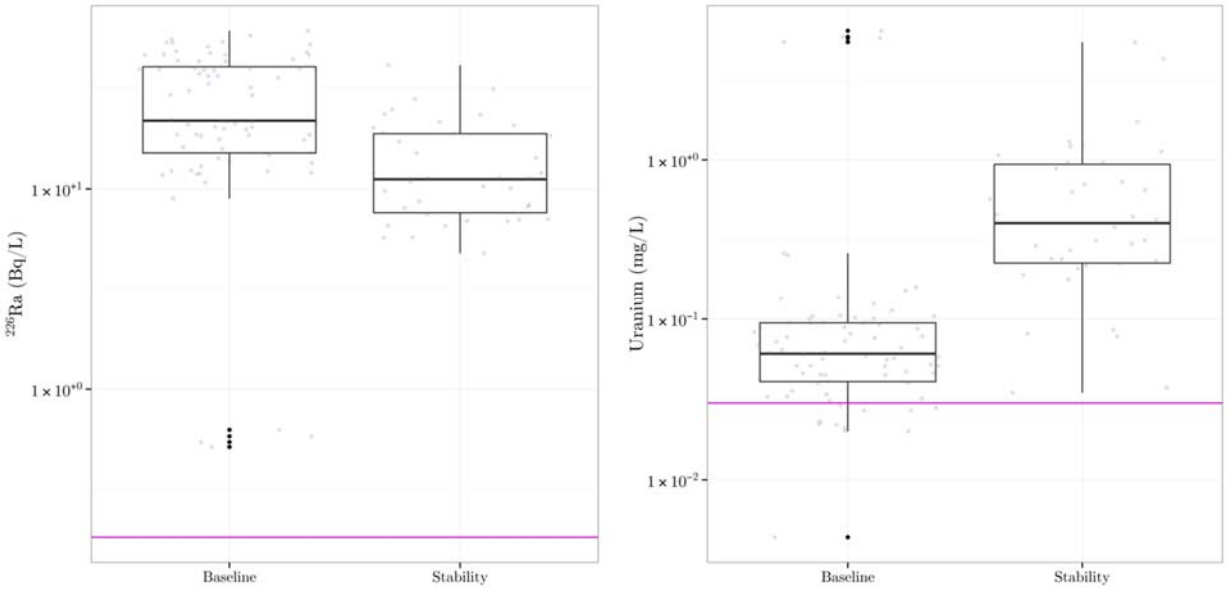

Table 1: Abundance of groundwater constituents of interest to USEPA, and others selected for study by the authors. Underlined text indicates a constituent was present in multiple wells above the minimum detectable concentration (MDC) in groundwater from Smith Ranch-Highlands. We have also included data about the total number of wells sampled, and the total number of samples acquired. Note that these data describe the results of the first round of stability (post-mining) testing, completed in September 2014.

Constituent	**Regulated by USEPA**	**Well Field 1 (stability)**	**Well Field 1 (baseline)**	**Well Field 3 (baseline)**	**Well Field 4 (baseline)**	**Well (ba**
		n_{wells}=20 $n_{measurement}$=40	n_{wells}=66 $n_{measurement}$=275	n_{wells}=69 $n_{measurement}$=248	n_{wells}=72 $n_{measurement}$=220	n_{v} n_{meas}
As	yes	> MDC	> MDC	> MDC	> MDC	>
Ba	yes	< MDC	not analyzed	not analyzed	< MDC	not
Cd	yes	< MDC	< MDC	< MDC	< MDC	<
Cr	yes	< MDC	< MDC	< MDC	< MDC	<
Pb	yes	< MDC	not analyzed	not analyzed	< MDC	not
Hg	yes	not analyzed	not analyzed	not analyzed	> MDC	not
Se	yes	> MDC	> MDC	> MDC	> MDC	>
Ag	yes	not analyzed	not analyzed	not analyzed	not analyzed	not
NO_3+NO_2	yes	> MDC	> MDC	> MDC	> MDC	>
Mo	yes	< MDC	< MDC	< MDC	< MDC	<
^{226}Ra	yes (as total Ra)	> MDC	> MDC	> MDC	> MDC	>
U	yes	> MDC	> MDC	> MDC	> MDC	>
gross alpha	yes	not analyzed	not analyzed	not analyzed	not analyzed	not
TDS	no	> MDC	> MDC	> MDC	> MDC	>
Alkalinity	no	> MDC	> MDC	> MDC	> MDC	>
pH	no	> MDC	> MDC	> MDC	> MDC	>

566 **Table 2**: US Safe Water Drinking Act Standards for 13 groundwater constituents proposed by the
567 USEPA to be of interest at ISR mine facilities.

Constituent	**SWDA Limit**
Arsenic	0.010 mg L^{-1}
Barium	2 mg L^{-1}
Cadmium	0.005 mg L^{-1}
Chromium	0.1 mg L^{-1}
Lead	0.015 mg L^{-1}
Mercury	0.002 mg L^{-1}
Selenium	0.05 mg L^{-1}
Silver	0.1 mg L^{-1}
Nitrate (as nitrogen)	10 mg L^{-1}
Molybdenum	Unregulated contaminant
Radium (226+228)	0.185 Bq L^{-1}
Uranium	0.030 mg L^{-1}
Gross-□	0.555 Bq L^{-1}

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571 **Table 3**: Comparison of stability (post-restoration) conditions in well field 1 with baseline conditions
572 in well field 1 and site-wide baseline conditions.

Groundwater constituent	Stability conditions differ from MU1 baseline?	Stability conditions differ from site-wide baseline?
Alkalinity	Yes p<0.001	Yes p<0.001
As	Yes p<0.001	Yes p<0.001
NO_3+NO_2	Yes p=0.02	No
pH	Yes p<0.001	No
^{226}Ra	Yes p<0.001	Yes p<0.001
Se	Yes p=0.02	No
TDS	Yes p<0.001	Yes p<0.001
^{nat}U	Yes p<0.001	Yes p<0.001

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575 **Table 4:** Magnitude and significance of change from baseline to stability in groundwater
576 concentrations of ^{226}Ra and ^{nat}U for matched well measurements. Twenty matched wells were
577 measured. In total, 40 measurements were made at baseline (December 1996 – January 1997), and
578 40 have been made so far in stability (January 2014 – September 2014).

Constituent	Change in median concentration, baseline to stability	95% Confidence Interval	p-value
^{226}Ra	0.56 times less	0.42-0.75 times less	<0.001
^{nat}U	5.61 times greater	3.6-8.9 times greater	<0.001

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581 **Table 5**: Sources of total radiation dose, as a fraction of total dose and by pathway, calculated by
582 RESRAD. Note that this is per nuclide, as nuclide concentrations (and therefore contribution to total
583 dose) change between baseline and stability.

Species	Ground Shine	Inhalation	Water	Plants	Meat	Milk
^{238}U	0	0	0.60	0.35	0.01	0.04
^{226}Ra	0	0	0.57	0.34	0.03	0.06

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586 **Table 6**: RESRAD calculated radiation dose rates to resident farmer based on concentrations of
587 radionuclides in groundwater at baseline and stability, for matched wells.

Species	**Baseline median concentration (Bq L^{-1})**	**Baseline median dose rate (mSv y^{-1})**	**Stability median concentration (Bq L^{-1})**	**Stability median dose rate (mSv y^{-1})**	**Δdose rate (mSv y^{-1})**
^{238}U	0.9	0.08	5.1	0.44	0.36
^{226}Ra	21.4	12.6	11.9	7.0	-5.6

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Electronic supplement 1: RESRAD parameters for risk model.

All parameters not detailed here were left as RESRAD defaults.

Contaminated Zone
Not considered in model. Our model scenario considered groundwater contamination only, with no contaminated zone near the surface.

Cover / Hydrology		
Evapotranspiration coefficient	0.2	National Oceanic and Atmospheric Administration Technical report NWS-33. http://www.wrds.uwyo.edu/sco/climateatlas/evaporation.html
Wind speed	4.5 m/s	https://weatherspark.com/averages/30046/Douglas-Wyoming-United-States
Precipitation	0.012 m/yr	National Oceanic and Atmospheric Administration Climatography of the United States No. 81 http://cdo.ncdc.noaa.gov/climatenormals/clim81/WYnorm.pdf
Irrigation	0.65 m/yr	Enough to grow corn, minus natural precipitation http://www.extension.org/pages/14080/corn-water-requirements#.VCSCho_F-5I
Runoff coefficient	0.2	RESRAD user manual table E.1, for "rolling land"

Saturated Zone
Groundwater transport not considered; only pumping from groundwater to the biosphere. These parameters were left as default and do not affect the outcome of the model.

Unsaturated Zone
Left as default

Occupancy		
Inhalation rate	2591 m^3/yr	USEPA Exposure Factors Handbook http://www.epa.gov/ncea/efh/pdfs/efh-complete.pdf
Indoor dust filtration	0.4	Alzona et al. 1979 : Alzona, J., et al., 1979, "Indoor-Outdoor Relationships for Airborne Particulate Matter of Outdoor Origin," Atmospheric Environment 13:55–60.
External gamma shielding	0.68	Dickson, Elijah. Experimental shielding evaluation of the radiation protection provided by residential structures. Dissertation, Oregon State University 2013. https://ir.library.oregonstate.edu/xmlui/handle/1957/38431
Indoor time fraction	0.5	Consistent with farmer
Outdoor time fraction	0.5	Consistent with farmer

Ingestion - Dietary

Fruit, veg, grain	330 kg/yr	USEPA Exposure Factors Handbook http://www.epa.gov/ncea/efh/pdfs/efh-complete.pdf ; 70kg adult
Leafy veg	50 kg/yr	USEPA Exposure Factors Handbook http://www.epa.gov/ncea/efh/pdfs/efh-complete.pdf ; 70kg adult
Milk	250 L/yr	USEPA Exposure Factors Handbook http://www.epa.gov/ncea/efh/pdfs/efh-complete.pdf ; 70kg adult
Meat and poultry	150 kg/yr	USEPA Exposure Factors Handbook http://www.epa.gov/ncea/efh/pdfs/efh-complete.pdf ; 70kg adult
Fish	0	No surface water - no source of fish, take assumed poundage instead to be beef
Other seafood	0	No surface water - no source of fish, take assumed poundage instead to be beef
Soil	7.3 g/yr	USEPA Exposure Factors Handbook http://www.epa.gov/ncea/efh/pdfs/efh-complete.pdf ; 70kg adult
Drinking water	1095 L/yr	USEPA Exposure Factors Handbook http://www.epa.gov/ncea/efh/pdfs/efh-complete.pdf ; 70kg adult
Contaminated fraction	1	No other source for food, all food comes from areas/foods irrigated with mine water

Storage Times		
Fruit, veg, grain	14d	RESRAD Default
Leafy veg	1d	RESRAD Default
Milk	1d	RESRAD Default
Meat and poultry	20d	RESRAD Default
fish	n/a	No seafood consumption
Crustacean and mollusks	n/a	No seafood consumption
Well water	1d	RESRAD Default
Surface water	1d	RESRAD Default
Livestock fodder	140d	Roughly half the time (winter), livestock are on fodder that has been stored from the summer harvest.

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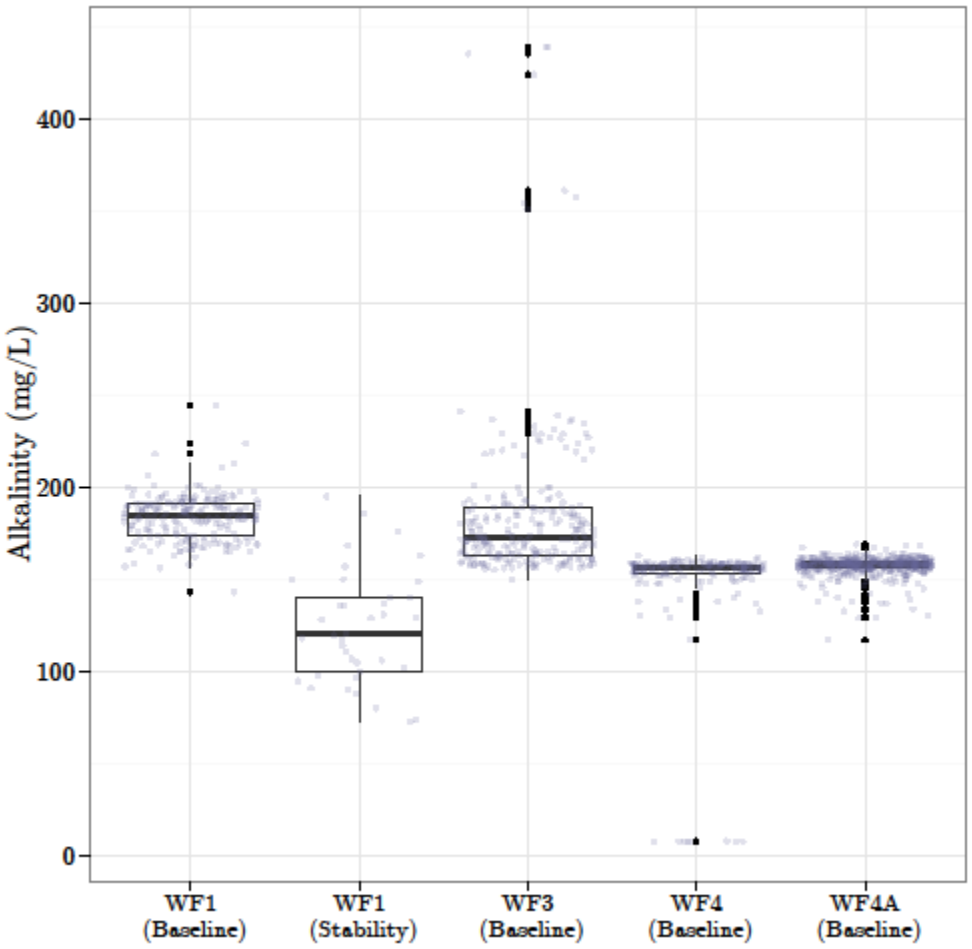
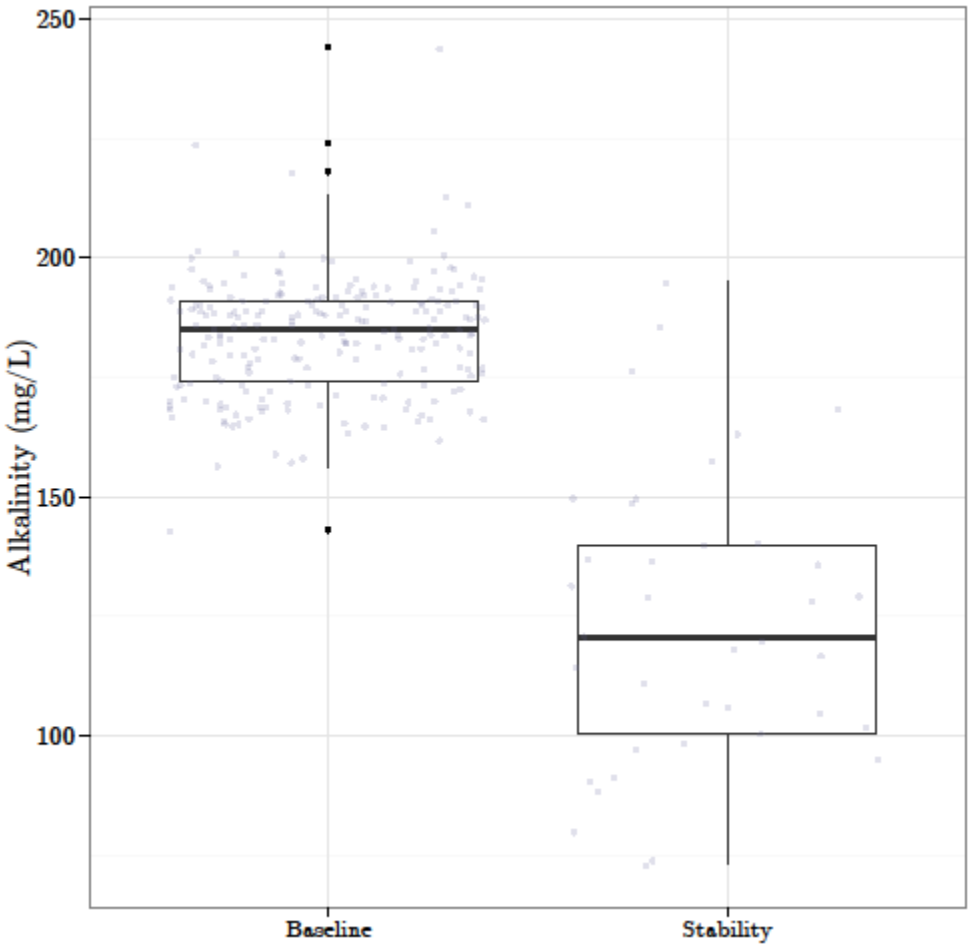
Electronic supplement 2: All boxplot data

The first boxplot displayed for each constituent includes all data points, the second boxplot is matched wells – those wells that have entered the stability phase and been characterized. The pink line represents the US EPA's potable water limit – where it is not displayed, no regulatory limit exists.

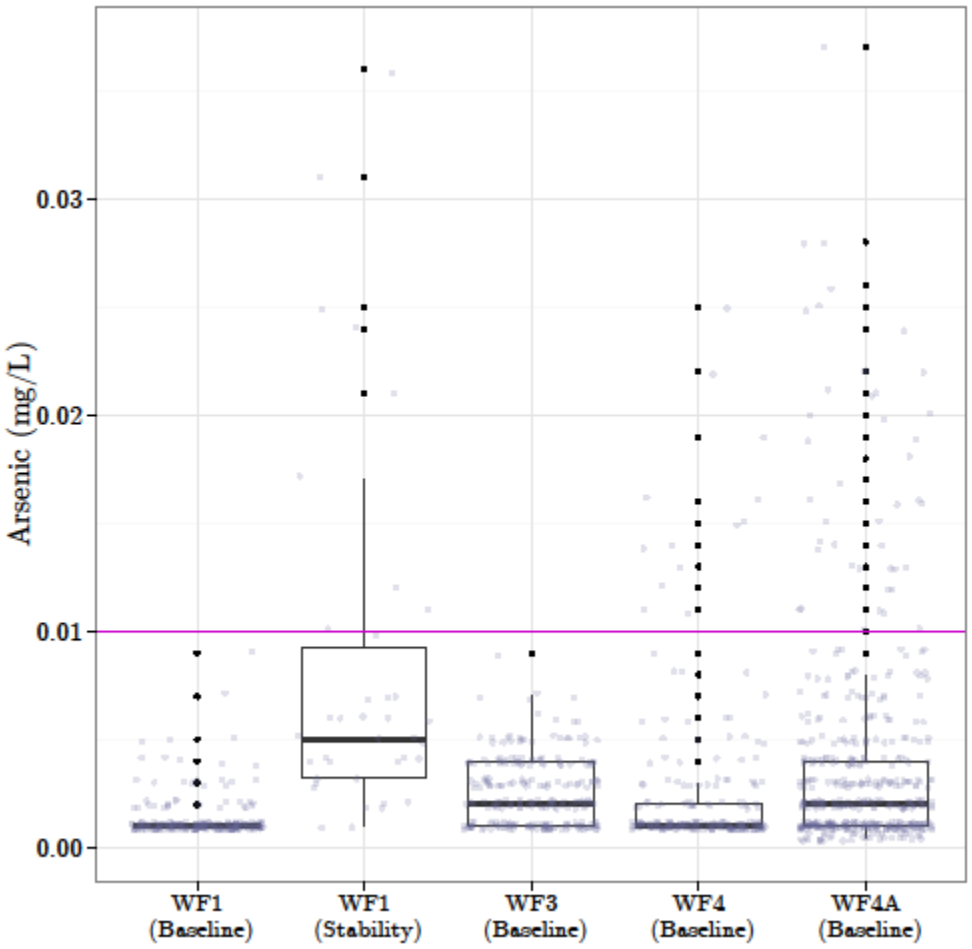
Alkalinity – all wells



Alkalinity – matched wells



606 **Arsenic – all wells**



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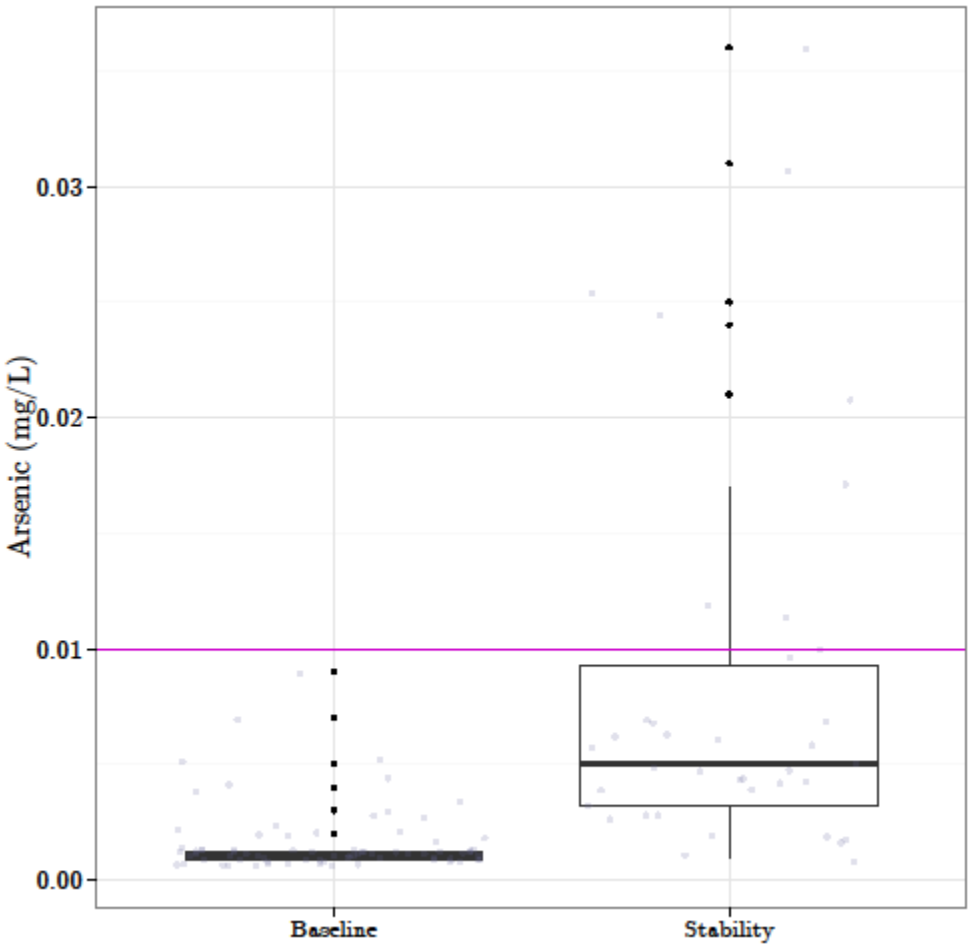
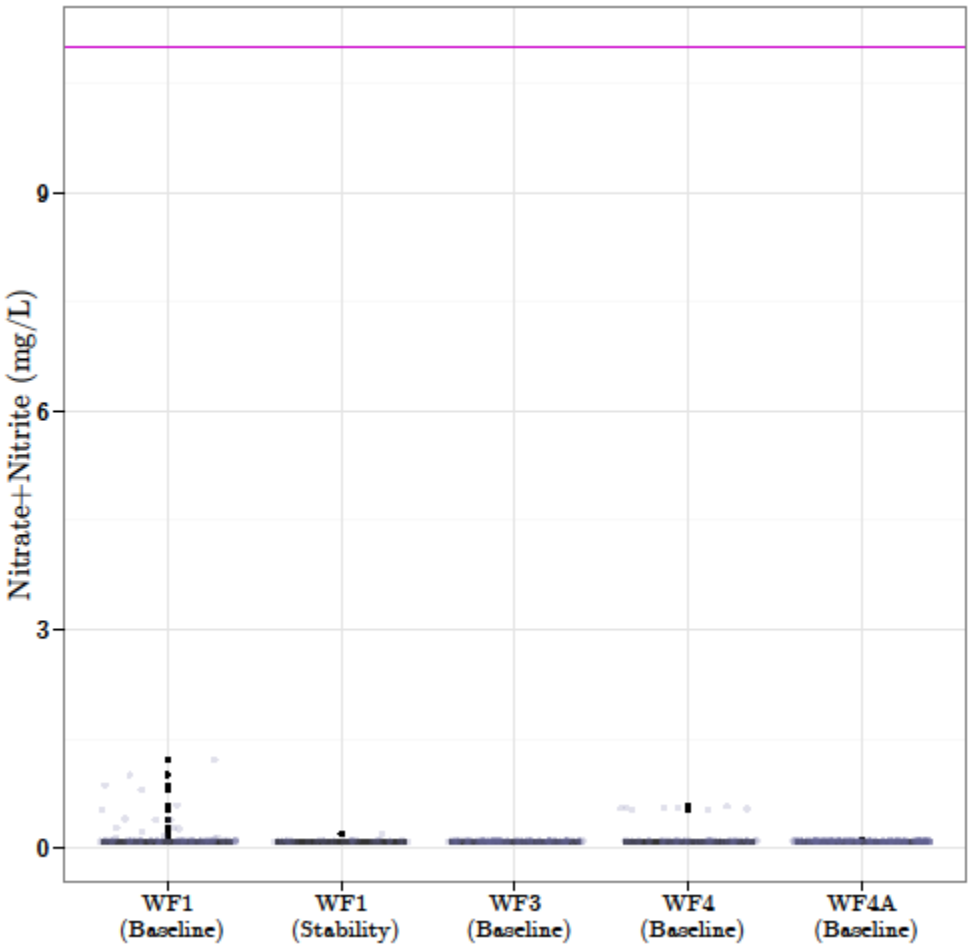
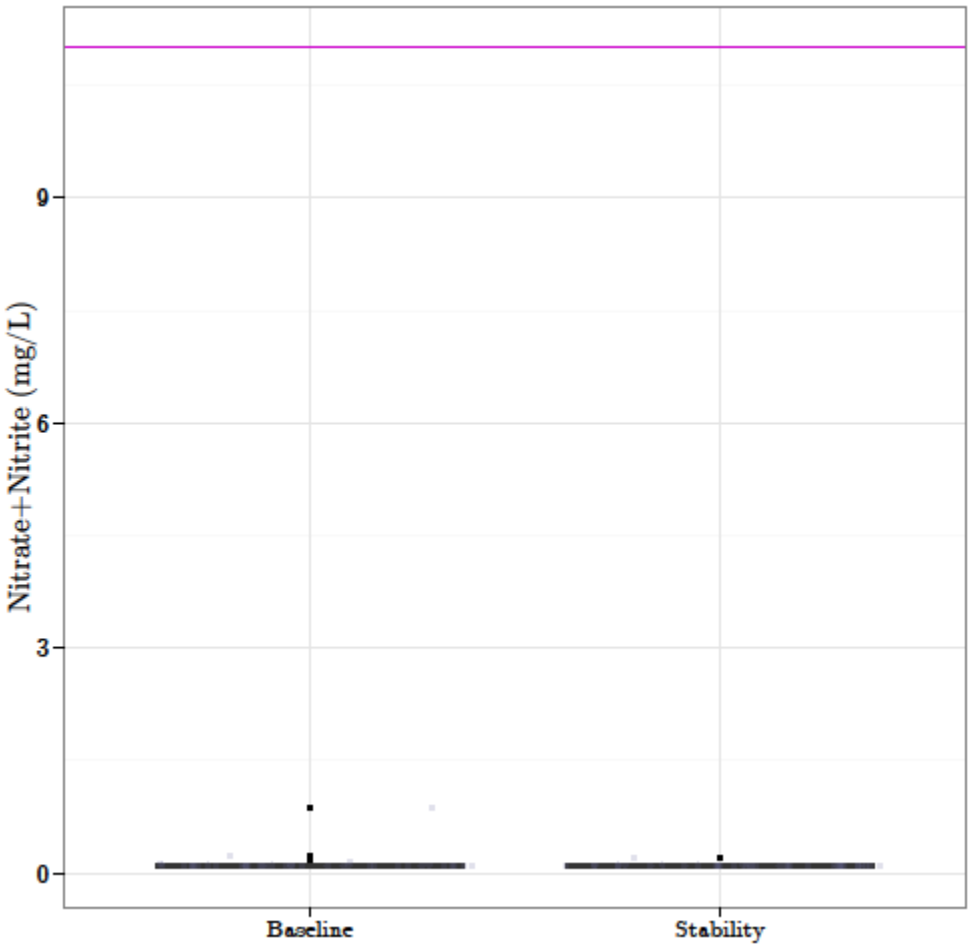
Arsenic – matched wells



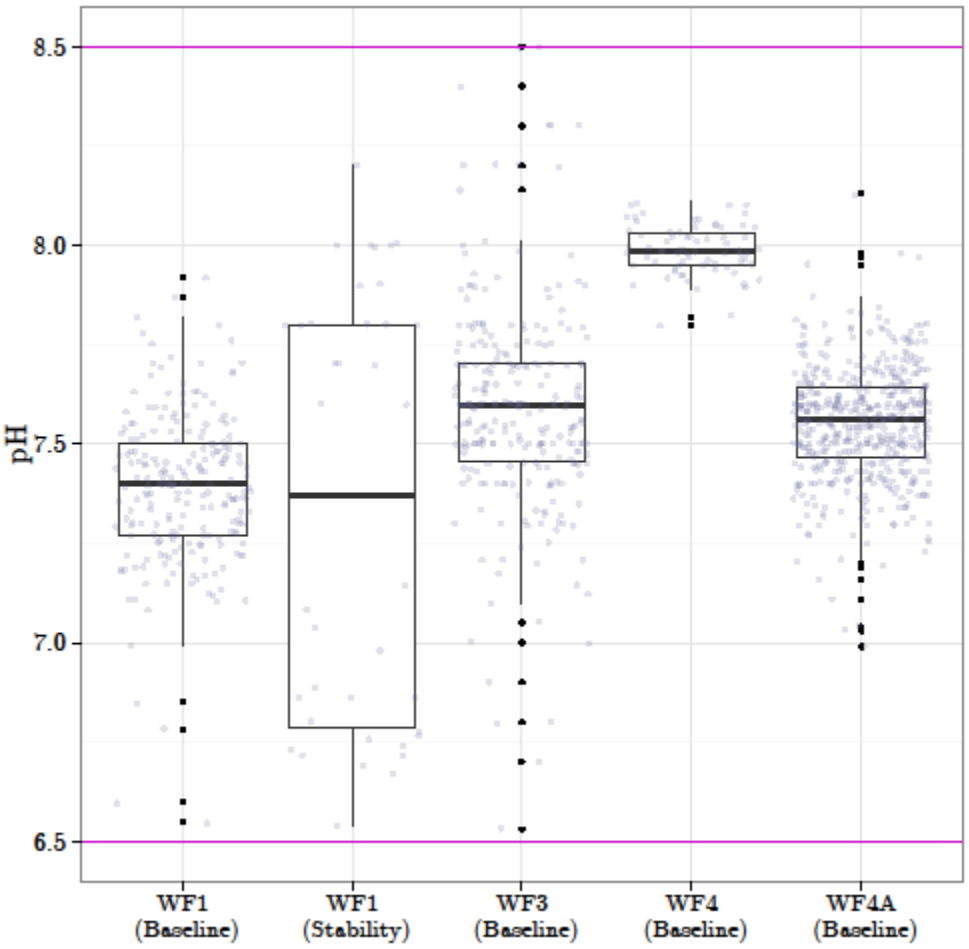
Total N – all wells



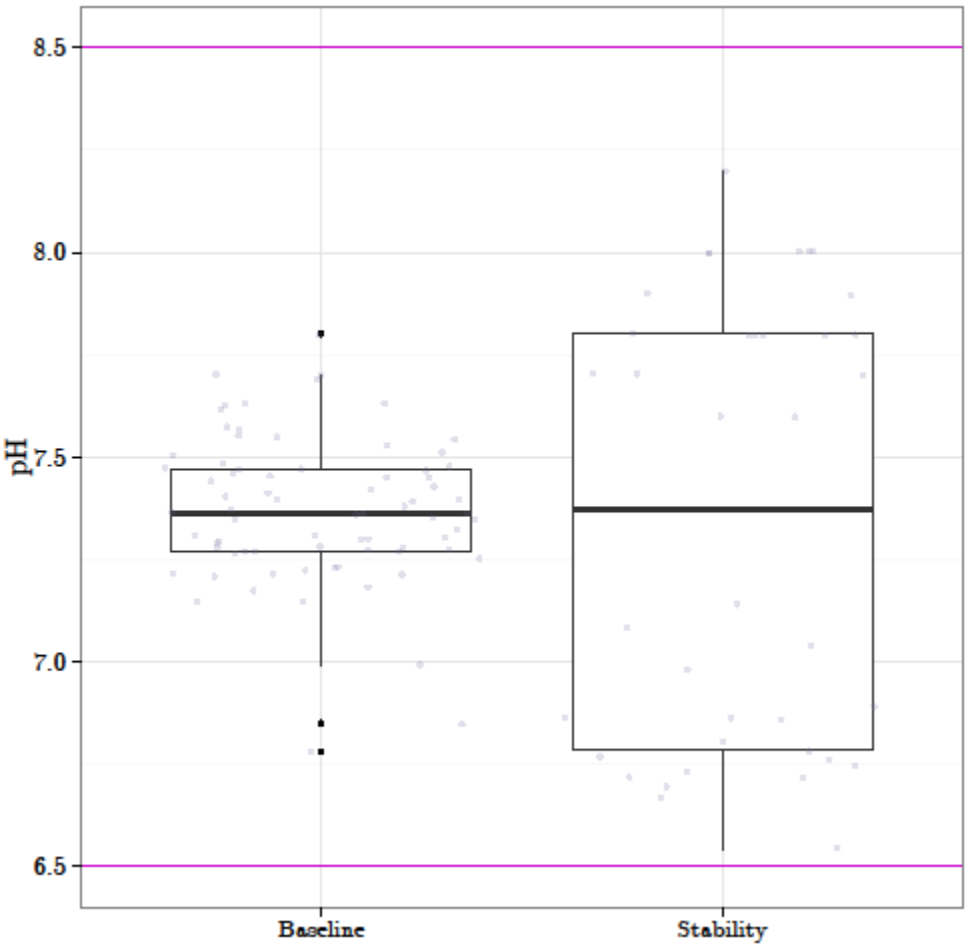
Total N – matched wells



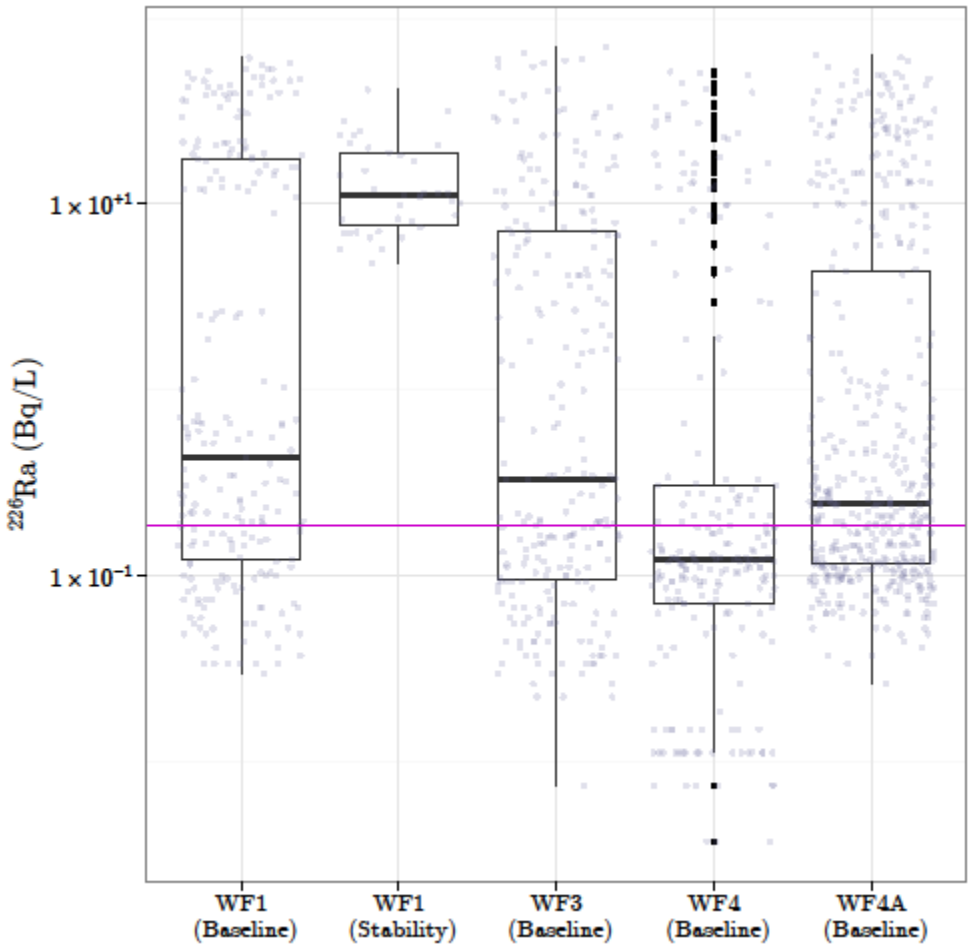
pH – all wells


8.5
8.0
7.5
7.0
6.5
pH
WF1 (Baseline)
WF1 (Stability)
WF3 (Baseline)
WF4 (Baseline)
WF4A (Baseline)

pH – matched wells



624 **^{226}Ra - all wells**



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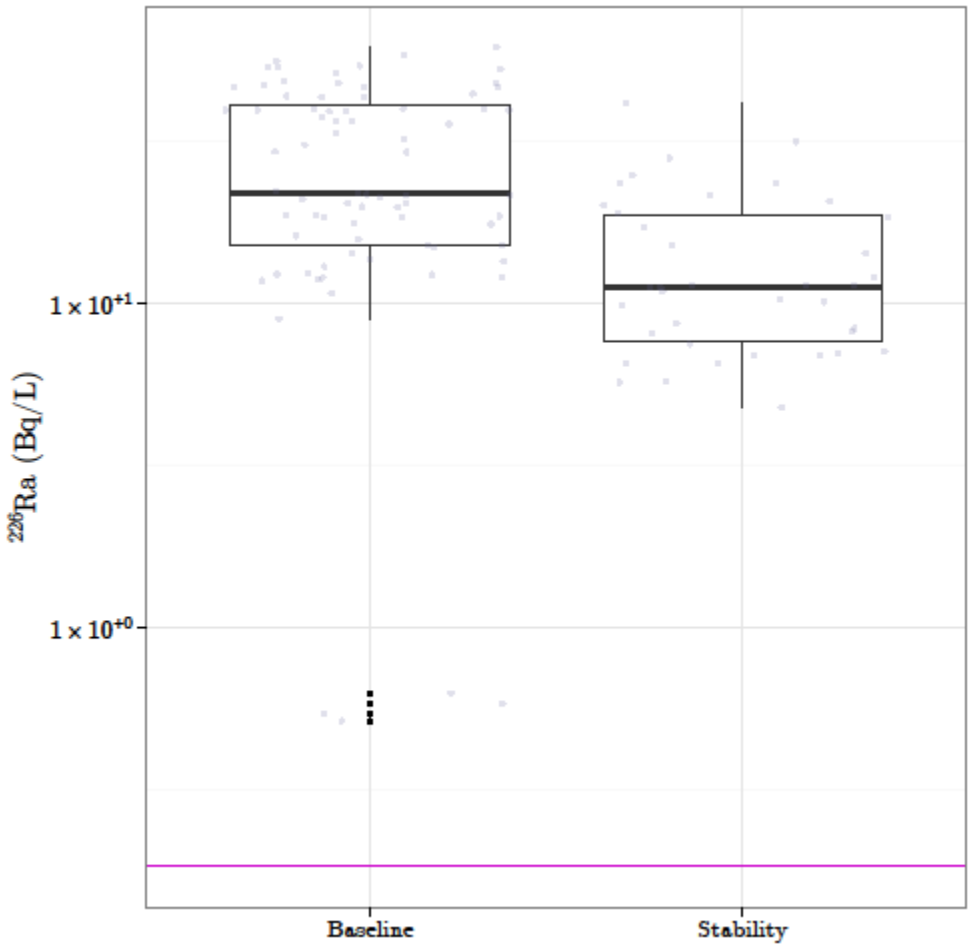
627 **^{226}Ra - matched wells**



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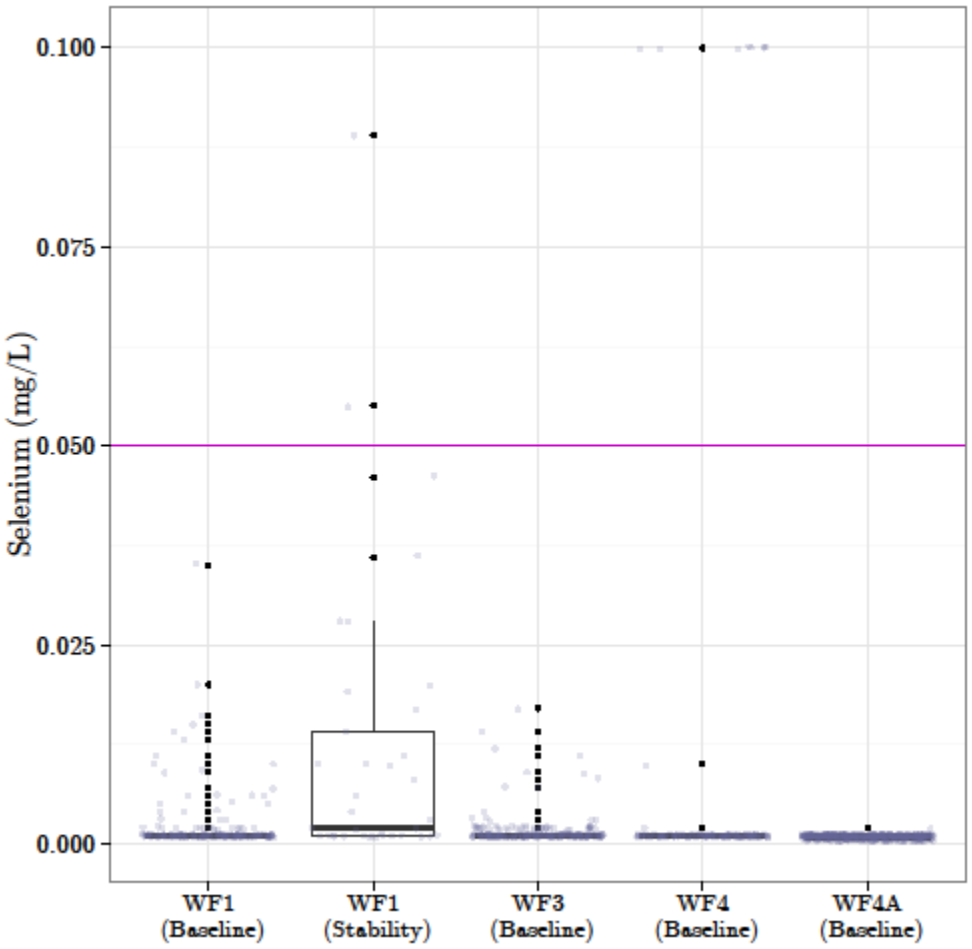
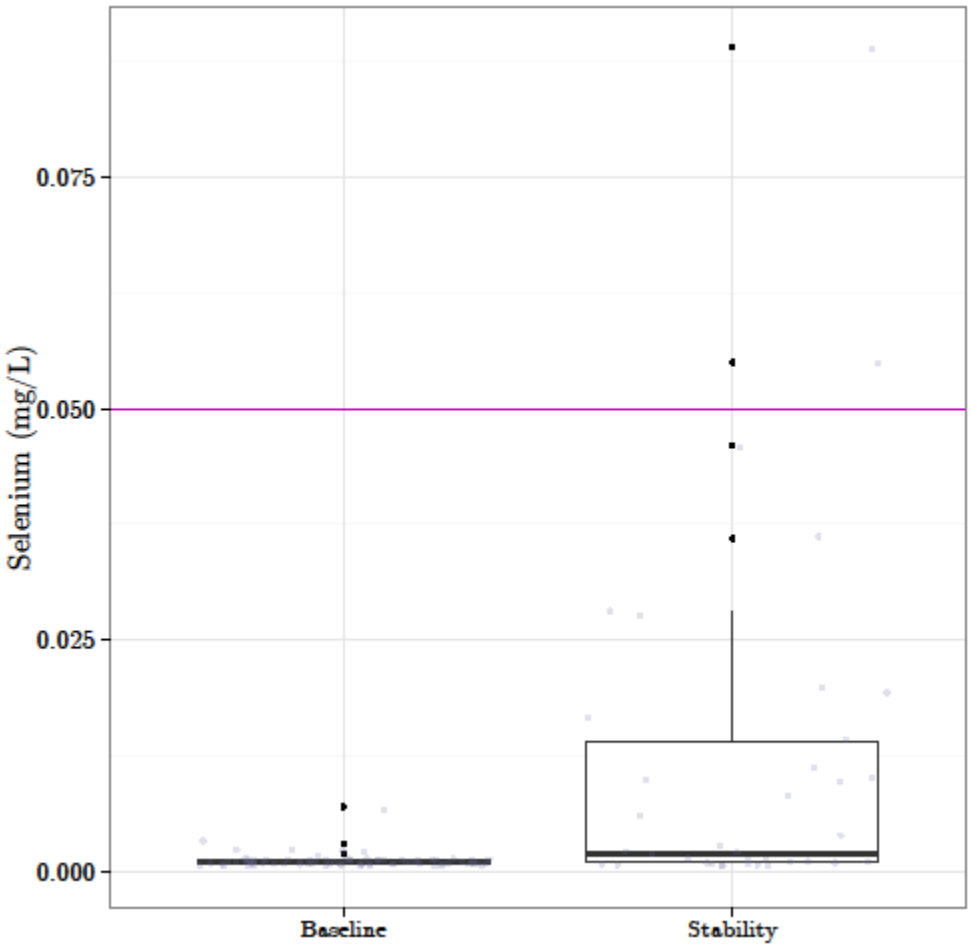
Se – all wells


0.100
0.075
0.050
0.025
0.000
Selenium (mg/L)
WF1 (Baseline)
WF1 (Stability)
WF3 (Baseline)
WF4 (Baseline)
WF4A (Baseline)

Se – matched wells



TDS – all wells