# EPA-Nuclear Metals: Campbell comments August 2020

I have reviewed Appendix E of the Remedial Design Work Plan, AECOM's comments, and the responses to those comments. Please find comments outlined below. The proposed experiments are well-designed, detailed, and a scientifically compelling set of tests that will answer the key questions of which amendments and what dose should be considered for the three *in situ* treatment zones (holding basin, overburden, bedrock). I also think that these tests would be very interesting to the greater scientific community (e.g., presented in a journal article form). There may be a few additional or specialized analyses needed, but the work as described has potential to be written up as a paper. I'm happy to discuss further if this is of interest to the team.

1. **AECOM comments**
   a. AECOM comment 2: AECOM recommended a possible alternative amendment (Calcium chloride, trisodium citrate, dibasic sodium phosphate amendment) for this system. Although this approach in many treatment environments has merit, it may not be ideal for NMI. As described in the responses to comments, calcium citrate is mainly used to amend the groundwater aquifer with calcium, so that the phosphate addition creates an apatite (or other Ca-phosphate mineral) barrier that reacts with uranium. Given the concentrations of Ca at the site, additional amendment of Ca is not likely to be needed. In addition, citrate forms complexes with metals, including iron and uranium, which may substantially alter the solubility of uranium, also mentioned in the responses. The complexation of uranium with citrate may potentially decrease the effectivity of the phosphate treatments. Francis et al. (1991) found that U-citrate complexes are resistant to microbial degradation, while Huang et al. (1998) found that microbial degradation of uranium-citrate complexes is pH dependent; however, both studies suggest that microbial degradation is likely to be limited at the pH of the aquifer. The literature on this subject is slightly outdated, given relatively new information about U-carbonato ternary complexes (ca., 2008-9), which are undoubtably important in this context. Some geochemical aqueous speciation modeling could be relatively easily used to determine the expected speciation of uranium in the presence of a citrate amendment under groundwater conditions. Generally, although an interesting approach to creating U-reactive Ca-phosphate precipitates in situ, it is not likely to be the best option for the NMI site.

      RESPONSE: We agree that the in-situ approach proposed by AECOM is not the best option for the NMI site.

   b. AECOM comment 4: Gamma spectrometry is an acceptable method for screening U concentrations. An alternate approach (mainly for future reference and which may have already been considered) is handheld XRF. Although it needs to be calibrated specifically for the matrix to be analyzed, it has the advantage of screening multiple elements simultaneously on a semi-quantitative to quantitative basis.

RESPONSE: Thank you for the recommendation.

c. AECOM comment 5: I would like to second this comment – vacuum sealing in the field is easy, inexpensive, and reliable when a double bag method is used. It is worth considering. Refrigeration of core is also highly recommended.

   RESPONSE: Preservation of the in-situ redox conditions of soil samples is not a priority for the soil sampling protocol since the soils will be exposed to oxygenated solutions during the batch and column tests. Based on this consideration, we believe that double bagging the soil in zip top bags and expelling ambient air within the bags will be sufficient to preserve the geochemistry of the samples. Additionally, soil samples will be packed on ice during transport to the lab to limit geochemical alteration.

d. AECOM comment 6: I agree that a consistent panel of metal(loids) should be analyzed throughout. Al, Ca, Mn, Fe, As, U – it looks like this was corrected in the edits. Consider also any other potential metals of concern (e.g., Sb, Ba, Be, Cd, Cr, Co, Cu, Pb, Mo, Ni, Se, Ag, Tl, Th, V, Zn – measured in GW) at this stage.

   RESPONSE: Concerning the other potential metals of concern: Sb, Be, Cd, Pb, Ni, Se, Ag, Tl, V, and Zn are not on the EPA Groundwater Cleanup List for the site. Ba, Cr, Co, Cu, have not been detected at levels of concern. Th has been detected in overburden groundwater above the cleanup level, but at locations far away from the uranium plume. Mo has been detected in groundwater at elevated levels in the upgradient portion of the uranium plume in overburden and has been added to the analyte list. Similar to arsenic, molybdenum will be measured in select batch reactor and column effluent samples at a frequency less than uranium.

e. AECOM comment 10: An important point is to have background phosphate and nitrate data in the baseline data.

   RESPONSE: We agree that background phosphate concentrations are important for evaluating the results of the study; phosphate is measured during background characterization of all soil and groundwater. Nitrate has been measured throughout the site in groundwater, and we have added nitrate to the baseline groundwater characterization and select batch reactor and column effluent samples in each treatability study.

f. AECOM comment 11: I definitely agree that Fe(II) data will be important to these studies. Phenanthroline or FerroZine are both relatively easy colorimetric methods for this determination.

   RESPONSE: We agree on the importance of measuring Fe(II) during the column study. We will consult with the laboratory on the use of colorimetric methods to directly measure Fe(II).

g. AECOM comment 18: Regarding solubility of apatite II, hydroxyapatite: Some experiments in my lab (unpublished, as of yet) found a poorly crystalline Ca-P-U precipitate that had a higher solubility than both apatite II and hydroxyapatite, and which slowly recrystallized into U-substituted hydroxyapatite over time. It may or may not be directly applicable to this situation, but this kind of phase may precipitate in the phosphate amended experiments. I am happy to discuss further if it would be helpful, or if you see evidence of it in the experiments.

   RESPONSE: Thank you for this insight. We will consider the possibility of this phase during evaluation of the treatability study results.

**2. General comments**

a. Analytical comments
   i. Consistency: The document still has a few incontinences with analytes. It might be good to double check the analytical parameters. I tried to point out many of them in the specific comments below.
   ii. Mineralogy
      1. Pre-reaction sediments: consider including particle size (PSA), XRD, sequential extractions to understand the baseline sediments.

         RESPONSE: During the Remedial Investigation (RI), soil cores were collected from the uranium plume in overburden. Analyses of these cores included particle size determinations using a Beckman-Coulter laser coulter counter (LS-230), XRD for major constituent minerals, and sequential extractions to qualitatively determine iron mineralogy and concentrations of uranium associated with different mineral fractions. Results can be found in Appendix G of the RI Report (de maximis, Geosyntec, 2011[1]). This document has been included with the response to comments transmittal.

         In general, the study found that the relative importance of system variables on uranium transport included: 1) sediment specific surface area; 2) solution alkalinity; 3) solution pH; and 4) calcium concentration.

      2. Homogeneity: It would be good to clarify how the homogeneity of the sediments used in the experiments will be ensured.

         RESPONSE: Soil samples will be homogenized by the laboratory prior to baseline characterization and column packing. Additional details regarding the soil homogenization procedure have been added throughout the text:

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[1] *de maximis*, Geosyntec, 2011, Remedial Investigation Report (Appendix G: Dartmouth College Uranium Adsorption Study - Nielsen, L.C.., and Bostick, B.C., *Integrating Variable Aquifer Geochemistry and Sediment Properties into Models of Uranium Retention at the Nuclear Meals Incorporated Superfund Site.*

Loose or sandy soil will be blended via cloth blending, use of a V-blender, or cone-and-quartering techniques. Following homogenization, representative sub-samples will be collected for use in the batch and column tests based on visual inspection.

3. Bicarbonate extraction: Kohler et al. (Environ. Sci. Technol. 2004, 38, 240-247) developed an extraction with bicarbonate at pH ~9 that estimates desorbable U. It is no harder to do than the modified Tessier extractions. Consider incorporating this method.

   RESPONSE: We appreciate this suggestion. The Kohler et al. work employs a 1,500 mg/L bicarbonate/carbonate extraction fluid at pH 9.45 to quantitatively desorb uranium from sediment surfaces by strong aqueous complexation of U(VI) in order to develop a distribution coefficient for uranium partitioning between sediment and aqueous phases. We propose two extraction steps that target adsorbed uranium (weakly sorbed and strongly sorbed), which we believe are sufficient to quantify the fraction of uranium sorbed to soils. Additionally, we propose a bicarbonate extraction as part of the evaluation of the leachability of immobilized uranium after the column tests using a 70 mg/L bicarbonate solution adjusted to pH 7 to represent site groundwater conditions. We believe this to be more relevant to the site than the higher concentrations of bicarbonate used by Kohler et al.

iii. Alkalinity. This measurement is generally absent from the groundwater sampling, but this is a key measurement. Groundwater in ambient atmosphere will likely degas $CO_2$, changing the pH and potentially precipitating calcite. Also, microbial activity will generate $CO_2$, so monitoring alkalinity is part of understanding the redox question (see below) as well as affecting uranium speciation. In addition, phosphate amendments may change the alkalinity as well. Good alkalinity and pH measurements will be a key part of having enough geochemistry to interpret changes between amendment and control batch reactors and columns.

RESPONSE: We agree alkalinity is an important parameter to assess and monitor; we propose in the workplan to use inorganic carbon measurements (by coulometry) for baseline groundwater characterization and during batch and column tests to determine inorganic carbon concentration, which is the primary component of alkalinity that affects uranium mobility and is equivalent to alkalinity measurements. Phosphorus measurements during the column study will allow us to evaluate the effect of phosphate-based treatments on alkalinity. Directly measuring inorganic carbon and phosphorus provides more information and is more practical than alkalinity titrations.

iv. Consider following nitrate, Fe(II), and sulfide in addition to total Al, Ca, Mn, Fe, As, and U

RESPONSE: We will measure nitrate (colorimetry), Fe(II) (colorimetry, or indirectly by dissolved iron), and sulfide (by zinc acetate/turbidimetric method) in select samples as redox indicators. These analytes have also been added to the baseline characterization.

b. Redox: As we are all well-aware, reduction can be beneficial for U in short term (insoluble U(IV) precipitates) but the extent of reduction can also be hard to control. If U reduction is a mode of removal, the extent of U reduction and co-precipitation of "redox buffers" (e.g., FeS) need to be considered and matched as closely as possible to what is achievable *in situ*, especially during reduction/reoxidation tests for the holding basin.

i. How reducing should each batch/column test go (how is the system poised in Eh space)? How will it be monitored? With different treatments will come different redox conditions (especially since Apatite II has organic carbon residual in it). Do the redox conditions in the experiments mimic what is expected in the aquifer during treatment?

RESPONSE: The only test in which we are manipulating the redox conditions will be for the holding basin (HB) soil in TS ISS-1; we expect that field conditions in the HB will be anaerobic after capping and vertical barrier wall installation. We will simulate these conditions in the lab by amending the column influent with glucose and ammonium chloride to stimulate consumption of dissolved oxygen and nitrate and to achieve iron reducing/fermentative conditions. We will monitor the change in redox conditions via measurement of DO, ORP, nitrate, Fe(II) (directly, or as dissolved iron), and sulfide in select column effluent samples. The goal will then be to reintroduce oxidizing conditions to simulate a breach in the HB containment, to evaluate uranium dissolution and to determine which amendment works best to prevent resolubilizaiton of the uranium. It is possible that the reducing conditions in the TS ISS-1 tests will lead to uranium reduction and formation of uraninite. We may add QEMSCAN analysis of selected samples to determine the mineralogical distribution of solid-associated uranium.

We will not seek to lower the ORP in the overburden groundwater study outside the Holding Basin (ISS-2) since the groundwater system is generally oxidizing, however, changes in redox conditions may occur due to the amendment. We have revised the work plan to monitor redox indicator species (nitrate, iron, sulfide) concentrations throughout the batch and column tests to evaluate if we see development of reducing conditions.

ii. Glucose: this will drive fermentative microbial pathways. I would think you would be more interested in dissimilatory metal reduction, so lactate or acetate may be an alternative electron donor.

RESPONSE: Glucose fermentation will produce lactate and acetate and associated iron reduction, so we believe this will stimulate a range of microbial activity relevant in the HB. We hope to avoid promoting dissimilatory uranium reduction with the redox amendment in the Treatability Study. Dissimilatory uranium reduction is not a primary sequestration mechanism of any of our proposed in situ amendments, and reduced uranium is susceptible to reoxidation and remobilization even when reducing conditions are maintained.[2]

iii. Question of long-term stability: One of the big questions is if aquifer returns to oxic conditions and the reduced U reoxidizes, is the Ca-phosphate sufficient to sequester U? If the experiments are too reducing and the reoxidation test indicates no pulse of U because of the "redox buffering", there is the possibility that the phosphate question will not quite be answered. Just something to think about – this does not change the experimental design fundamentally. Also, maybe this is made clear in the larger document (my apologies if I've missed it), but what is the decision tree for whether possible follow-up injection may be needed if oxidation of reduced U becomes an issue in the aquifer?

RESPONSE: We agree that a return to oxic conditions within the columns during TS ISS-1 will be necessary to evaluate whether the amendments prevent remobilization of oxidized U(IV) minerals. Section 3.2.4 states that "if by week 4 the effluent is not indicative of a return to oxidizing conditions within the column (return to baseline ORP and dissolved iron concentrations), this phase of the column test will continue."

Injection remedies rely on contact with impacted groundwater and are a function of distribution, seepage rates, and hydraulic gradients. Whatever amendment and dose are selected for the final remedy in overburden outside the Holding Basin, groundwater in and downgradient of the current plume area will be monitored closely. If uranium concentrations do not decline significantly or decline and rebound, then follow-up injections will be implemented.

c. Column experiments
  i. Tracers: Consider monitoring/adding a tracer. You will need it to determine porosity of the column (so results can be translated to pore volumes). Also, a tracer is needed for any geochemical modeling efforts. E.g., Br addition to influent. This would also add an additional analyte.

RESPONSE: Thank you for this suggestion; we agree that addition of a conservative tracer to the column tests will provide valuable information. We have revised the text to state that bromide (or a suitable tracer based on laboratory analytical

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[2] Wan et al., 2005. *Reoxidation of bioreduced uranium under reducing conditions.* Environmental Science and Technology.

capabilities) will be added to the column influent solution and measured in the column effluent.

d. Arsenic. It is clear this team is well-aware of likelihood of arsenic release from sediments, especially with high levels of phosphate competitively desorbs arsenic from the sediments, along with the arsenic introduced with amendments. It is not clear to me exactly what the plan is to deal with arsenic – it sounds like there may be enough Fe-oxide content downstream of treatment to re-adsorb the arsenic, but it would be worth adding modeling or other planning tools for understanding arsenic mobility during and after treatment. The comments about redox state also applies to arsenic, as strongly sulfidic conditions in a column may sequester As in sulfide precipitates, but is the aquifer expected to go that reducing during treatment conditions?

   RESPONSE: We do not expect sulfidic conditions to develop within the aquifer during treatment, especially if apatite or soluble phosphate is used, however, we will monitor redox conditions during the treatability tests to evaluate this. We expect there will be enough iron oxide mineral content in the aquifer downstream of the injections to sequester any arsenic that might be mobilized through phosphate emplacement. We will evaluate the iron content of the soil and the iron mineral phases through selective extractions as part of the TS. This information can be used to assess the sorptive capacity of the aquifer for arsenic.

## 3. Specific comments

[p2] Amendment selection Matrix:
#1: additional disadvantage: contains organic matter – reduction potential likely.
#4: additional advantage: As may be retained on ZVI; ZVI not likely to introduce As

RESPONSE: The amendment selection matrix has been updated to include these additional advantages and disadvantages.

[p3]. Other possibilities include U-substituted apatite, and possible poorly crystalline Ca-U-P precursor phases. There is uncertainty in the actual mineralogical products.

RESPONSE: The text has been updated to include these additional uranium sequestration products.

[p7 #8] I would think that mineralogy, particle size would be as important to this experiment as U concentration. How homogeneous are these cores expected to be?

RESPONSE: Soil samples will be collected over a 20-foot interval, which will likely include the range of grain sizes known to be present in the holding basin. Homogeneity in the lab will be achieved by mixing the soils in the laboratory prior to constructing the columns. Assessments of mineralogy and particle size within the depleted uranium plume have been performed previously.[3]

[p7 #10]
Particle size? XRD? Please see general comment about Mineralogy above. Sometimes the controlling phases are minor in content but key to overall geochemistry. E.g., Fe oxides.
Are there any other elements of concern that should be measured at this time? Co was mentioned in the call. How about Al, Sb, Ba, Be, Cd, Cr, Co, Cu, Pb, Mn, Mo, Ni, Se, Ag, Tl, Th, V, Zn, as for groundwater? Consider measuring in starting material.

RESPONSE: Mineralogy and particle size have been previously measured,[4] and results indicated that particle size and Fe oxides have strong controls on the magnitude of uranium sorption at the site. The target soil collection areas for the Holding Basin should have very representative particle size and Fe oxide concentrations, and therefore, particle size determination on the homogenized soils will not be critical for the study. We have added molybdenum to the list of metals for baseline characterization and select batch and column effluent samples; the remaining metals are not regulated at the site or have not been detected at levels of concern.

[p8 3.2.2] Raw and a 0.45um filtered sample at time of sampling would be useful. Although O2 will be minimized, as will CO2 degassing, you may want to know if calcite precipitated before starting your experiments. Calcite precipitation may also affect [U]. By having an initial acidified sample for analysis, you know how much your water changed. Also, measure alkalinity. Having a handle on the initial water conditions, including pCO2, calcite, and effect on U speciation and conc seems important. Add Mn to analyte list. Consider turbidity for consistency. Sample preservation protocols (e.g., acidification)?

RESPONSE: We added additional characterization of the groundwater at the time of sampling (unfiltered and 0.45 µm filtered) to identify potential changes in groundwater composition between sampling and initiation of the column and batch studies (baseline characterization). In addition, we added Mn to the analyte list and turbidity to the field measurements.

[p9 analytes] Sorry for nit-picking here, but As is not a cation. Also, Fe is not always reliable by ICP-MS. Many of these elements are better by ICP-OES, which is included in the rest of the document, so perhaps this is just a typo.

RESPONSE: Thank you for pointing out these typos. We have revised the text to add arsenic to the list of metals and to add ICP-OES as a potential analytical technique for cation analysis.

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[3] *de maximis*, Geosyntec, 2011, Remedial Investigation Report (Appendix G: Dartmouth College Uranium Adsorption Study - Nielsen, L.C.., and Bostick, B.C., *Integrating Variable Aquifer Geochemistry and Sediment Properties into Models of Uranium Retention at the Nuclear Meals Incorporated Superfund Site.*

[4] *de maximis*, Geosyntec, 2011, Remedial Investigation Report (Appendix G: Dartmouth College Uranium Adsorption Study - Nielsen, L.C.., and Bostick, B.C., *Integrating Variable Aquifer Geochemistry and Sediment Properties into Models of Uranium Retention at the Nuclear Meals Incorporated Superfund Site.*

[p10 3.2.4 column test] Consider monitoring/adding a tracer. You will need it to determine porosity of the column (so results can be translated to pore volumes). Also needed for any geochemical modeling efforts.

RESPONSE: We have revised the text to include amendment of bromide (or a similar conservative tracer) to the column influent solution and measurement of bromide in the column effluent.

[p10 column test, weeks 2-3] Consider redox carefully. Glucose will drive fermenters. So, it will go anaerobic, but perhaps by a slightly different mechanism than intended (e.g., dissimilatory Fe reduction)? Also, it is hard to control/stop at Fe reduction and the system may go more reducing, especially with apatite II amendment. Vitamins are sometimes added in nutrient amendments, but it may or may not be necessary. It sounds like the groundwater will be sterilized in addition to filtration – if so, how will the GW be sterilized? 0.22um filtration usually does the trick. Note that sterilization can introduce O2 and degas CO2. That's expected and it's better to sterilize but knowing how the processing of the water affects the geochemistry is necessary. Consider measuring Fe(II), sulfide. May need closer monitoring and redox decision-making determination in this step (see redox comments above).

RESPONSE: We agree that these are important considerations during the anaerobic phase of the column test. Glucose will stimulate a range of microbial activity including iron reduction and we believe this is appropriate to simulate anaerobic conditions in the HB since we want to avoid promoting dissimilatory uranium reduction. As noted above, we have added measurement of redox indicators (nitrate, dissolved iron, sulfide) to select samples. Turbidity has been added to the field parameters measured during sample collection. The text has been revised to clarify that groundwater will be filter sterilized.

[week 4] This will depend greatly on how reducing the column went in the previous step. If a lot of reduced Fe, U, S, biomass is present, then there could be a large redox buffer in the column.

RESPONSE: We agree. As noted above, the column test will continue until oxidizing conditions are observed in the effluent.

[p11] Analyte list: Mn, alkalinity. Microbial activity will produce inorganic C, and P amendment can affect alkalinity. I would be good to have enough geochemistry to interpret changes between amended and control columns, especially changes in U concentrations. Another thought: consider raw/filtered analysis for Fe as well as U, as colloidal Fe is a possibility.

RESPONSE: We have added Mn to the analyte list and will be measuring inorganic carbon and phosphorus during the test to assess alkalinity. If effluent iron concentrations are elevated, we will consider analysis of filtered and unfiltered column effluent for iron to assess the possibility of colloidal iron in the column effluent.

[p13 4.2.1] sediment analyses: Mn, As – other elements? XRD? PSA?

RESPONSE: We have revised the text to include manganese and arsenic in the baseline soil characterization. We have historical XRD data on the soils;[5] particle size determination on the homogenized soils will not be critical data for the study.

[p14 4.2.2] ditto water analysis comments above.

RESPONSE: We added additional characterization of the groundwater at the time of sampling (unfiltered and 0.45 µm filtered) to identify potential changes in groundwater composition between sampling and initiation of the column and batch studies (baseline characterization). In addition, we added Mn, nitrate, and sulfide to the analyte list and turbidity to the field measurements.

[p14 4.2.3] Glass is not typically used for metal work as it may have some sorption capacity. Acid-washed plastic is standard, but given the size of the batch experiments, it is likely that a glass bottle effect is small.

RESPONSE: We will discuss with the laboratory the decision to switch to plastic bottles for this work.

[p15 4.2.3] These experiments could have wide divergence in redox conditions in the treatments. Is it valuable to control redox? Would that mimic the site conditions best? This comment relates to the general redox comment above. Fe(II), sulfide could be useful measurements to pinpoint process (not necessarily just relying on ORP) even if redox state is allowed to float depending on amendment. I assume the rubber stoppers are fixed to the bottles (e.g., serum bottles with sealed stoppers) - if you get any microbial activity, you may see flying rubber stoppers!

RESPONSE: The redox state of the batch and column experiments will depend on the treatment amendment. Since the redox conditions in the field will not be controlled, the redox state of the batch and column tests will also not be poised. We agree that analyzing nitrate, Fe(II) (directly or indirectly as dissolved Fe), and sulfide in the batch reactor and column effluent samples will provide valuable information regarding the geochemical processes controlling the redox conditions within the reactors/columns. We have added these analyses to the text and the sample matrix.

[p16 4.2.4] ditto tracer comment

RESPONSE: We have revised the text to state that bromide (or a similar conservative tracer) will be added to the column influent solution and measured in the column effluent.

[p16] Ditto comments about O2/CO2 exchange, alkalinity, U, calcite, etc.

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[5] *de maximis*, Geosyntec, 2011, Remedial Investigation Report (Appendix G: Dartmouth College Uranium Adsorption Study - Nielsen, L.C.., and Bostick, B.C., *Integrating Variable Aquifer Geochemistry and Sediment Properties into Models of Uranium Retention at the Nuclear Meals Incorporated Superfund Site.*

RESPONSE: As noted above, we have included additional baseline characterization of groundwater at the time of sampling to evaluate changes in composition (e.g., precipitation, degassing of CO2) during transport and storage.

[p17] Consider at least one filtered/unfiltered comparison, and adding Alkalinity, Mn

RESPONSE: We added manganese to the analyte list for the column effluent. As noted above, inorganic carbon and phosphorus are already being analyzed to monitoring changes in alkalinity. If effluent iron concentrations are elevated, we will consider measuring iron and filtered effluent samples to assess the possibility of colloidal iron in column effluent.

[p18] May be hard to measure to amount of uranium sequestered by measuring changes in solid phase U concentration. Consider calculating this value by difference if you know influent/effluent concentration and pore volumes put through the column during the 6 weeks.

RESPONSE: Thank you for this recommendation. The text has been revised to include this calculation.

[p18 Bicarb leach] How will this be done? My apologies if I'm not understanding this procedure, but if you add bicarb and pH increases, then you decrease pH with acid, you will just degas CO2. Consider sparging with CO2 to hit alkalinity and pH targets. Or, use a published carb/bicarb leach method (e.g., Kohler et al 2004), which is higher pH, but is a well characterized method for quantifying for labile U.

RESPONSE: A bicarbonate solution will be mixed with soil and the pH will be adjusted to 7.0. Small or no headspace in the container will minimize the amount of degassing from the bicarbonate solution following pH adjustment, allowing the bicarbonate concentration and pH to be relatively constant during the extraction. The objective of this leaching test is to mimic potential changes in groundwater composition in the overburden, therefore, we believe the proposed bicarbonate leach test is more relevant than the Kohler et al. extraction.

[p19] A Kohler bicarb leach (above) would complement this sequential extraction suite. Also, Step 3 is commonly done with Hydroxylamine + HCl to target Fe, Mn oxides.

RESPONSE: Thank you for this recommendation. We believe the proposed bicarbonate leach test is more suitable for replicating site conditions and will provide more relevant information on the potential for uranium remobilization than the Kohler et al. extraction.

Step 3 follows the standard Tessier extraction procedure for iron and manganese oxides,[6] which has a similar extraction efficiency as hydroxylamine in HCl.[7]

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[6] Tessier et al., 1979. Sequential extraction procedure for the speciation of particulate trace metals. Analytical Chemistry.

[7] La Force, M.J. and S. Fendorf, 2000. Solid-phase iron characterization during common selective extractions. Soil Science Society of America Journal.

[p20] As you are aware, U phase identification may hard by XRD, even at these concentrations. Also, be on the lookout for amorphous/poorly crystalline U-containing phases that can be challenging (but not impossible!) by XRD. SEM, although expensive, could help understand where U is found in solids.

RESPONSE: Thank you for these considerations – we agree that analytical techniques in addition to XRD will likely be necessary to identify the precipitated uranium phase. We have proposed QEMSCAN analysis, which is based on Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM-EDS), for samples that do not have a sufficient uranium concentration to resolve the identity of the solid phase by XRD.

[Section 5: Bedrock experiments] Many of the comments above about groundwater sampling, bedrock homogenized chip mineralogical analysis, batch reactors, extractions, tracers, etc., apply to this section as well.

RESPONSE: We have revised the text and sampling matrix to be consistent with changes made to the other treatability studies (e.g., additional groundwater characterization at time of sampling, analysis of manganese and redox indicators [nitrate, iron, sulfide] during baseline characterization and during the batch tests). Tracers will not be necessary for TS-3 since only batch tests will be performed.

[p22 5.2.1] To what size will the bedrock chips be crushed? Also, additional analytes may be needed here for consistency. As, Mn are missing in solids characterization, as is mineralogy (XRD). Alkalinity and possibly turbidity should be added during groundwater collection. Mn is missing from the batch analyte list.

RESPONSE: The crushed bedrock will be sieved in the laboratory to target the sand fraction. We have updated the baseline characterization to be consistent with the other treatability studies.