



UNITED STATES ENVIRONMENTAL PROTECTION
AGENCY

Washington, DC 20460

July 17, 2020



OFFICE OF
AIR AND
RADIATION

Mr. Mike Brown
Carlsbad Field Office
U.S. Department of Energy
P.O. Box 3090
Carlsbad, New Mexico 88221-3090

Dear Mr. Brown:

The U.S. Environmental Protection Agency is working on its completeness review of the U.S. Department of Energy's (DOE) 2019 Compliance Recertification Application (CRA-2019) for the Waste Isolation Pilot Plant (WIPP). This letter is accompanying the fourth set of Agency completeness comments and questions (see attached). We request that DOE respond in a timely manner. If you have any questions concerning this request, please contact Ingrid Rosencrantz at (202) 343-9286 or rosencrantz.ingrid@epa.gov.

Sincerely,

Tom Peake
Director
Center for Waste Management and Regulations

Enclosure

cc: Electronic Distribution
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FOURTH SET OF EPA COMPLETENESS COMMENTS AND QUESTIONS FOR CRA 2019

OVERVIEW

Completeness questions are organized by theme or CRA section. The naming convention is as follows:

- 1) Round of questions from EPA (e.g. CC3 = Completeness Comment 3rd Round)
- 2) Section/appendix/theme (33 = Section 33)
- 3) Comment/question number and subcomment/question (1, 2, 3a, 3b, etc.).
- 4) Brief title or topic

The comment or question is listed below the heading. Further descriptions and explanations may follow and are indicated by the italicized text.

GENERAL COMMENTS AND QUESTIONS

QUESTIONS AND COMMENTS BY CRA SECTION

QUESTIONS AND COMMENTS BY APPENDICES

CC4-GEOCHEM-22 Appendix GEOCHEM Colloid Parameter Table

Please note that some of the colloid parameters in Table GEOCHEM-61 were not used in the CRA-2014 PA and CRA-2019 DPA as stated in the title. The Pu(III) intrinsic colloid, U(IV) intrinsic colloid, and Pu(III) microbial colloid proportionality constant parameter values in this table are not the CRA-2019 DPA values presented in Kim and Feng (2019). The Am(III) microbial colloid proportionality constant and the U(IV) intrinsic colloid parameter values in this table are not the CRA-2014 PA values presented in Kicker and Herrick (2013). Such corrections should be documented in an errata sheet or similar.

There are inconsistencies between two parameters listed in Appendix GEOCHEM Table GEOCHEM-61 for the CRA-2014 PA and the parameter values used in the CRA-2014 PA (Kicker and Herrick 2013). There are additional inconsistencies between three parameters listed in Appendix GEOCHEM Table GEOCHEM-61 for the CRA-2019 DPA and the parameter values used in the CRA-2019 DPA (Kim and Feng 2019). These inconsistencies occur because of the differences between the oxidation-state specific colloid parameters listed in Appendix GEOCHEM Table GEOCHEM-61 and the element-specific microbial and intrinsic colloid parameters used in the CRA-2014 PA and the CRA-2019 DPA.

Differences Between Parameters in Appendix GEOCHEM Table GEOCHEM-61 and Parameters Used in the CRA-2014 PA (Kicker and Herrick 2013)

| <i>Appendix GEOCHEM Table GEOCHEM-61</i> | | <i>Kicker and Herrick (2013) Table 4 and 27</i> | |
|--|--|---|--|
| <i>Am(III) PROPMIC</i> | <i>0.32</i> | <i>AM:PROPMIC</i> | <i>1.76</i> |
| <i>U(IV) CONCINT</i> | <i>$2 \times 10^{-8} M$</i> | <i>U:CONCINT</i> | <i>$3 \times 10^{-8} M$</i> |

Differences Between Parameters in Appendix GEOCHEM Table GEOCHEM-61 and Parameters Used in the CRA-2019 DPA (Kim and Feng 2019)

| <i>Appendix GEOCHEM Table GEOCHEM-61</i> | | <i>Kim and Feng (2019) Table 27</i> | |
|--|------------------------|-------------------------------------|------------------------|
| <i>Pu(III) CONCINT</i> | $9.5 \times 10^{-9} M$ | <i>PU:CONCINT</i> | $4.3 \times 10^{-8} M$ |
| <i>U(IV) CONCINT</i> | $4.3 \times 10^{-8} M$ | <i>U:CONCINT</i> | $1.4 \times 10^{-6} M$ |
| <i>Pu(III) PROPMIC</i> | 0.03 | <i>PU:PROPMIC</i> | 0.21 |

Kicker, D.C., and C.G. Herrick. 2013. *Parameter Summary Report for the Compliance Recertification Application, Rev. 0*. Sandia National Laboratories, Carlsbad, NM. ERMS 560298.

Kim, S. and L. Feng. 2019. *Input Parameter Report for the 2019 Compliance Recertification Application Performance Assessment (CRA-2019 PA), Revision 1*. Sandia National Laboratories, Carlsbad, NM. ERMS 571660.

CC4-MON-2 Updated map of potash mining

Please provide a more recent, updated map of potash operations with potash areas outlined but not obscuring the mine labels and include the boundary of the Delaware Basin where it crosses the map area.

The Delaware Basin Monitoring Annual Report (DBMAR) includes diagrams and maps of various extractive industries and infrastructure, including drilling for oil and gas, and potash mining. A map of current potash mining has been provided in Figure 7 for the last several years of the DBMAR. This figure is useful; however, it has not been updated since 2007 (according to the legend) and does not include all of the features that are referred to in the text. For example, the orange layer of potash mining completely obscures the names and locations of mine facilities referenced in the text.

CC4-MON-3 Status of proposed new potash mine east of WIPP

Please provide the status, lease locations, and location data of any new or proposed mines not described in CRA-2019. This information may be included with the updated potash mining map requested by the previous comment.

DBMAR 2011 through 2015 mentioned new potash leases acquired by Intercontinental Potash Corp east of WIPP and an Environmental Impact Statement (EIS) that was approved by the BLM in 2016 for a new underground mine. This proposed new mine is not mentioned in subsequent reports or any other CRA documentation that EPA has reviewed.

CC4-SCR-6 N FEPS updates

Please update the natural FEPS with new information and confirm that screening decisions remain the same.

Since the CCA was submitted in 1996 and first recertified in 2004, only a handful (7) of Natural (N) FEPS appear have received any updates, and most of these updates were to eliminate individual FEPS by combining them with others or to fix errors. The only FEPS to have received regular updates are N2 Brine Reservoirs, updated in 2014 and 2019 with new GLOBAL:PBRINE parameters, and N12 Seismicity, which is periodically updated with data from the Delaware Basin Monitoring Program.

It appears that there is no new information relating to the natural FEPS list that might be used to change, update, or even potentially strengthen screening arguments. In addition, post-CCA, DOE-funded studies directly relating to several natural FEPs are neither mentioned nor referenced in Appendix SCR. Substantial new information on the geology of the Delaware Basin has emerged from increased scientific study over the past decade in parallel with the boom of oil and gas production in the region. This information also has relevance for many Human-induced (H) events and processes. Although no changes to screening decisions are immediately apparent, EPA expects DOE to revisit the N FEPs list regularly, including for the 2024 CRA.

One comprehensive source of updated or new information on the geology and resources of the Delaware Basin and surrounding region is the recently published two-volume compilation by the Texas Bureau of Economic Geology on the Permian Basin which is co-published as an AAPG Memoir:

Ruppel, S. C. 2019. *Anatomy of a Paleozoic Basin: The Permian Basin, USA (Vol. 1)*. The University of Texas at Austin, Bureau of Economic Geology Report of Investigations 285, AAPG Memoir 118, 399 p. doi: doi.org/10.23867/RI0285-1.

Ruppel, S. C., 2020. *Anatomy of a Paleozoic Basin: The Permian Basin, USA (Vol. 2)*. The University of Texas at Austin, Bureau of Economic Geology Report of Investigations 285, AAPG Memoir 118, 538 p. doi:doi.org/10.23867/RI0285-2.

The following are some examples:

SCR-6a: Tectonics, faulting FEPs

FEPs documentation is missing significant, relevant updates from the published literature for SCR-4.1.2 Tectonics: N3 Changes in Regional Stress, N4 Regional Tectonics, N5 Regional Uplift and Subsidence.

These sections rely on decades-old references and do not mention newer published studies. Even if the screening argument does not change, this suggests that DOE has neglected to stay up on relevant literature that has come out since the original certification. For example, “there are no reported stress measurements from the Delaware Basin” (pp. SCR-28) isn’t completely true—there have been a number of studies reporting stress orientation measurements within and surrounding the Delaware Basin over the past decade. Several references in the current FEPs list are made to work by Zoback and colleagues in the 1980s to early 1990s, but a large body of later, more sophisticated, and widely disseminated work on regional stress states in the US, including the Worldwide Stress Map (WSM), and specific work on the Permian Basin region (e.g. Lund-Snee and Zoback, 2016, 2018) have not been incorporated into this FEP. These kinds of information are also essential for assessing seismic hazard due to induced earthquakes, and also can influence oil & gas activities.

One particular finding from the Delaware Basin is that maximum horizontal stress orientations vary from north to south. Around WIPP, maximum horizontal stress is roughly NE-SW, and the overall stress (faulting) regime is normal with a minor strike-slip component. In the middle of the Delaware Basin, it is roughly E-W, and in the south, it is roughly NW-SE. Interpreting the orientation of the NE-SW striking, ~30 Ma igneous dike identified in the early WIPP site characterization work as a paleostress indicator suggests that the overall stress field in the WIPP region has not significantly changed since that time, strengthening the claim of long term tectonic

stability (tens of millions of years) and therefore the safety case for WIPP. This may also have impacts on FEPS N10, N11 (faulting).

Lund Snee, J.-E. and M. D. Zoback. 2018. State of stress in the Permian Basin, Texas and New Mexico: Implications for induced seismicity. *The Leading Edge*: 810-819.

Lund Snee, J.-E. and M. D. Zoback. 2016. "State of stress in Texas: Implications for induced seismicity." *Geophysical Research Letters*, 43(19): 10,208-210, 214.

Forand, D., et al. (2017). Constraints on Natural Fracture and In-situ Stress Trends of Unconventional Reservoirs in the Permian Basin, USA. *Proceedings of the 5th Unconventional Resources Technology Conference*. URTEC 2669208

Heidbach, O., Rajabi, M., Reiter, K., Ziegler, M., and WSM Team. 2016. *World Stress Map Database Release 2016. V. 1.1*. GFZ Data Services. <http://doi.org/10.5880/WSM.2016.001>

The reference discussing Quaternary Faulting (Muehlberger et al., 1978) that has been relied on since the early site characterization studies of WIPP is very dated. More recent published literature, including the USGS Quaternary Faulting Database, has identified normal faults immediately south of the southern tip of the Delaware Mountains and just outside the Delaware Basin, just over 110 km WSW from the WIPP boundary. This information may have relevance to the arguments of FEPS N10 and N11.

U.S. Geological Survey. 20188. *Quaternary fault and fold database for the United States*. <https://www.usgs.gov/natural-hazards/earthquake-hazards/faults>.

SCR-6b. Please update discussions of karst-related FEPs (N18, 19, 21) and manmade dissolution features (i.e. sinkholes) (H34, H59) with relevant, recent information, including an EPA-mandated, DOE-funded, SNL study.

Appendix SCR fails to discuss or even mention multiple recent studies on karst in the WIPP region, including what may be considered as the most comprehensive, authoritative, DOE-funded, SNL study, which was mandated and reviewed by EPA. With the exception of H59, it appears that no changes have been made to these FEPs in any way since at least CRA-2004. EPA expects reevaluation of the FEPs for each recertification cycle to determine their continued relevance, including these and related FEPs.

Lorenz, J.C. 2006a. *Assessment of the Potential for Karst in the Rustler Formation at the WIPP Site*. Albuquerque, New Mexico: Sandia National Laboratories. SAND2005-7303.

Lorenz, J.C. 2006b. Assessment of the Geological Evidence for Karst in the Rustler Formation at the WIPP Site. *Caves and Karst of Southeastern New Mexico. L. Land, V.W.* pp. 243–52.

The only specific mention and discussion of these karst studies in CRA-2019 is in Appendix PA under Groundwater Flow in the Culebra (Section PA-2.1.2.6), as it was for CRA-2014, and Section 23 Computer Codes in CRA-2014. A reason for its omission in Appendix SCR is not clear.

Additionally, EPA notes that evaporite karst in and around the Delaware Basin is still an active area of research in the scientific community. EPA regulations require that DOE track and incorporate relevant findings as appropriate, whether or not they strengthen or change the screening argument. A few examples:

Land, L. 2013. Evaporite karst in the Permian Basin region of west Texas and southeastern New Mexico: The human impact. In: *Proceedings of the 13th Sinkhole Conference, Carlsbad, New Mexico, 2013, Volume Sinkholes and the engineering and environmental impacts of karst*. National Cave and Karst Research Institute.

Stafford, K. W., Rosales-Lagarde, L., and P.J. Boston. 2008. Castile evaporite karst potential map of the Gypsum Plain, Eddy County, New Mexico and Culberson County, Texas: A GIS methodological comparison. *Journal of Cave and Karst Studies*, v. 70, no. 1, p. 35-46.

Stafford, K. W., Nance, R., Rosales-Lagarde, L., and P.J. Boston. 2008. Epigene and Hypogene Gypsum Karst Manifestations of the Castile Formation: Eddy County, New Mexico and Culberson County, Texas. *International Journal of Speleology*, v. 37, no. 2, p. 83-98.

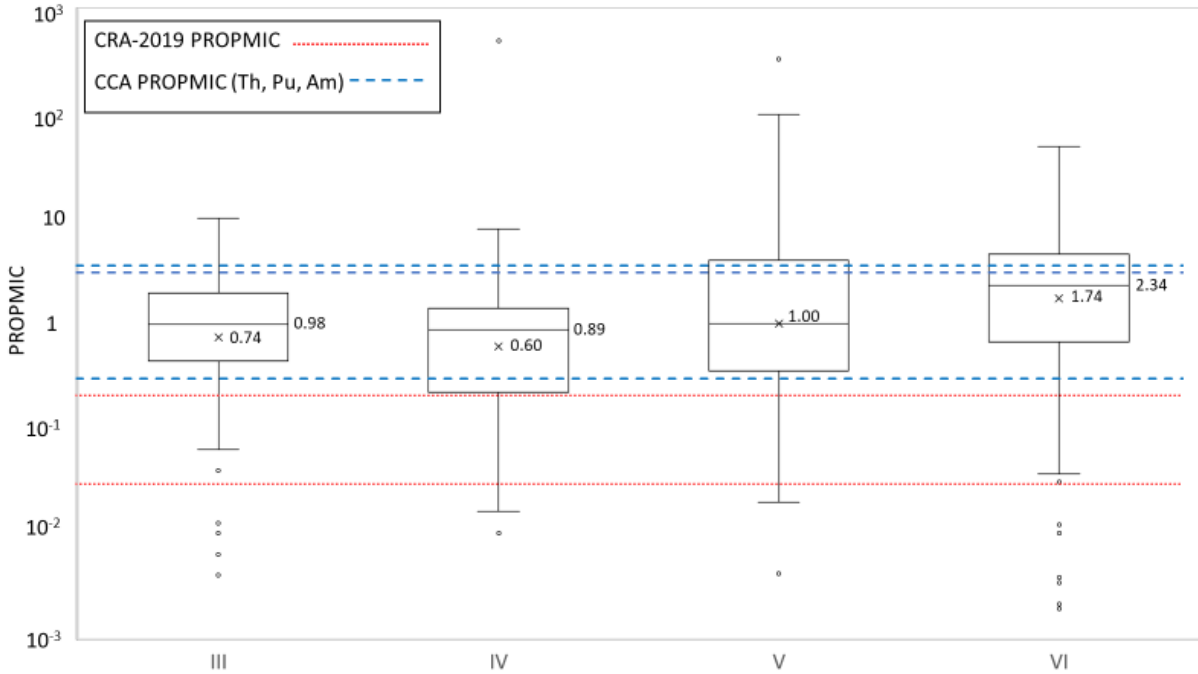
Stafford, K., Ehrhart, J., Majzoub, A., Shields, J., and W. Brown. 2018. Unconfined hypogene evaporite karst: West Texas and southeastern New Mexico. *USA: International Journal of Speleology*, v. 47, no. 3, p. 293-305.

CC4-SOTERM-6 Comparison of PROPMIC Values to Literature Values

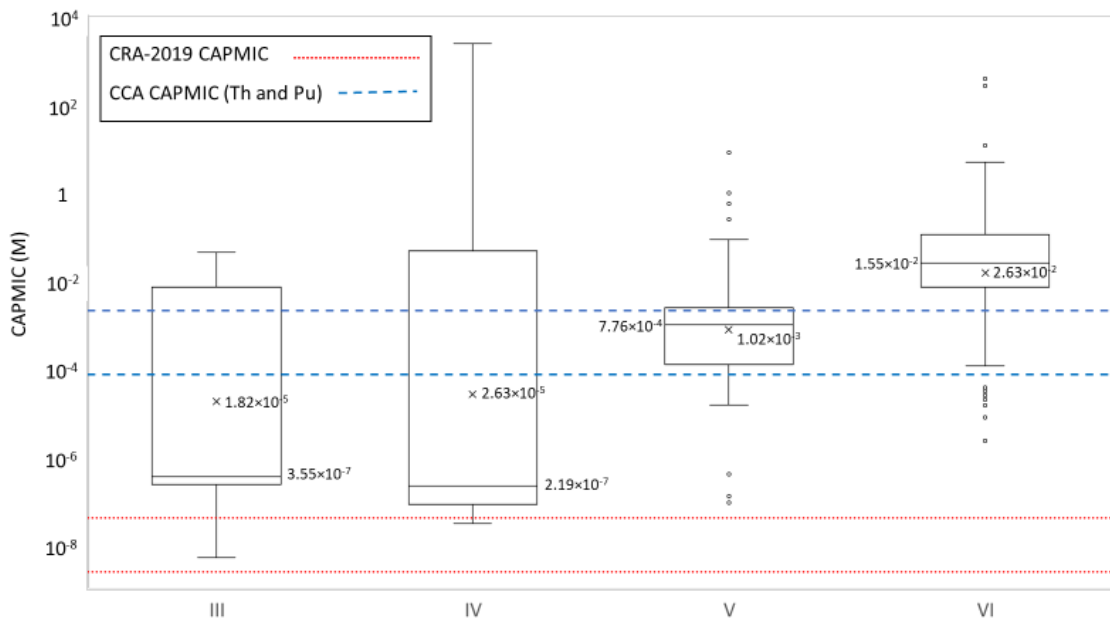
Please use the biomass-based approach to compare the differences in mobilized actinides using the proposed updated CRA-2019 CAPMIC parameters and a recalculated CCA CAPMIC parameter as specifically identified below. EPA has conducted its own independent investigation of the microbial colloids parameters and has found that the values DOE used in the 2019 DPA do not appropriately bound the variability presented in the literature (see writeup below).

CC4-SOTERM-6a. Please recalculate the CCA CAPMIC parameter values using the 10^9 cells/ml proposed biomass-based approach that was used to calculate the CRA-2019 CAPMIC and PROPMIC values.

CC4-SOTERM-6b. Using the CCA PROPMIC values as well as the recalculated CCA CAPMIC, please compare the effects on repository releases to that of the CRA-2019 microbial colloid enhancement parameters.



EPA-calculated PROP MIC values derived from a review of laboratory and field studies in the literature.



EPA-calculated CAP MIC values derived from a review of laboratory and field studies in the literature.

Reed et al. (2019) notes “EPA comments on these [microbial colloid] changes were received as part of their CRA-2014 review [EPA 2017] but the updated parameters submitted were accepted.” The Agency has not accepted the 2014 parameter updates and continues to assert that

until the updates are defensibly implemented, EPA will expect the use of the original CCA data and approach (EPA 2017, page 148; Peake 2018).

The study used to derive the 2019-CRA DPA proposed microbial colloid enhancement parameters are based on Nd bioassociation experiments on the WIPP-specific isolate Chromohalobacter (Swanson 2018). It is likely that the WIPP environment will contain a diversity of microorganisms beyond a single isolate, which means that the extent of bioassociation will also likely fall within a range of values due to the variation of microbe-actinide bioassociation. The experiments used to determine the microbial colloid enhancement parameters in the CCA were based on both the pure WIPP-1A culture as well as the mixed BAB culture, which appears more representative of the variety of organisms expected at the WIPP.

Given the paucity of WIPP microbe-actinide interaction data, EPA conducted an independent review of microbial colloids, using expanded criteria that include conditions beyond that of the WIPP. From this search, EPA reviewed 39 studies in both laboratory and field settings that examined biosorption, bioaccumulation, and biomineralization. PROPMIC and CAPMIC values were calculated from these studies and then further screened to include only experiments that quantified interactions at pH values of 6.0 or higher, in defined media, and greater than 2-hour timescales. This resulted in 20 studies in solutions ranging from dilute solutions to 4 M ionic strength, and in experimental timescales from 2 hours to 150 days. Of these studies, 5 experiments represented +III Am and Eu, 7 experiments represented +IV Pu and Th, 7 experiments represented +V Pu and Np, and 14 experiments represented +VI Pu and U. Organisms represented include bacteria, archaea, eukarya (algae, fungi, and yeast), and mixed cultures.

Results of the study are shown in the figures above. The data reveal a wide variability in PROPMIC values for all oxidation states. Both median (lines inside the boxes) and mean (x symbols inside the boxes) PROPMIC values are between 0.5 to 1.5 orders of magnitude greater than the values selected for CRA-2019 DPA (red dashed lines) but are bounded by the Th and Pu CCA values (blue dashed lines).

CAPMIC values, calculated using 10^9 cells/ml, are also highly variable, especially for the +III and +IV actinides. The mean CAPMIC concentrations for An(III) and An(IV) (x symbols) vary by three to four orders of magnitude from the CRA-2019 DPA values, while median concentrations (lines inside boxes) vary by one to two orders of magnitude. Both the An(III) and An(IV) CAPMIC median and mean values are bounded by the Th and Pu CAPMIC concentrations used in the CCA.

Some caveats to the data: many of these studies fall below the expected pH range and ionic strengths of WIPP brine and also often ignore important issues like filtration. Many studies, such as those with Pu, may also involve mixed oxidation states. Nevertheless, these studies provide much needed context into the possible extent of actinide/microbe interactions and provide a range of values that can be used to select adequately bounding PROPMIC values that incorporate the variability in microorganisms in nature as well as variability resulting from the range of geochemical conditions expected at the WIPP. Because the CCA CAPMIC and PROPMIC parameters are based on experiments with more diverse organisms than the CRA-2019 DPA parameters and because these parameters provide an upper bound for the available literature values (see figures above), the CCA PROPMIC and recalculated CAPMIC parameters should be used to calculate mobilization of actinides by microbial colloids.

- Ams, D. A., Swanson, J. S., Szymanowski, J. E., Fein, J. B., Richmann, M., and D.T. Reed. 2013. The effect of high ionic strength on neptunium (V) adsorption to a halophilic bacterium. *Geochimica et Cosmochimica Acta*, 110, 45-57.
- Bader, M., Müller, K., Foerstendorf, H., Drobot, B., Schmidt, M., Musat, N., Swanson, J. S., Reed, D. T., Stumpf, T., and A. Cherkouk. 2017. Multistage bioassociation of uranium onto an extremely halophilic archaeon revealed by a unique combination of spectroscopic and microscopic techniques. *Journal of hazardous materials*, 327, 225–232.
<https://doi.org/10.1016/j.jhazmat.2016.12.053>
- Bader, M., Müller, K., Foerstendorf, H., Schmidt, M., Simmons, K., Swanson, J. S., Reed, D. T., Stumpf, T., and A. Cherkouk. 2018. Comparative analysis of uranium bioassociation with halophilic bacteria and archaea. *PloS one*, 13(1), e0190953.
<https://doi.org/10.1371/journal.pone.0190953>
- Bader, M., Rossberg, A., Steudtner, R., Drobot, B., Großmann, K., Schmidt, M., Musat, N., Stumpf, T., Ikeda-Ohno, A., and A. Cherkouk. 2018. Impact of Haloarchaea on Speciation of Uranium-A Multispectroscopic Approach. *Environmental science & technology*, 52(21), 12895–12904. <https://doi.org/10.1021/acs.est.8b02667>
- Boggs, M. A., Jiao, Y., Dai, Z., Zavarin, M., and A.B. Kersting. 2016. Interactions of plutonium with *Pseudomonas* sp. strain EPS-1W and its extracellular polymeric substances. *Applied and Environmental Microbiology*, 82(24), 7093-7101.
<https://aem.asm.org/content/aem/82/24/7093.full.pdf>
- Ding, C., Feng, S., Cheng, W., Zhang, J., Li, X., Liao, J., Yuanyou, Y., Zhu, A., Shunzhong, L., Yang, Jijun, Tang, J., and N. Liu. 2014. Biosorption behavior and mechanism of thorium on *Streptomyces sporoverrucosus* dwc-3. *Journal of Radioanalytical Nuclear Chemistry*, 301(1), 237-245.
- EPA (United States Environmental Protection Agency). 2017. *Technical Support Document For Section 194.24, Evaluation Of The Compliance Recertification Actinide Source Term, Gas Generation, Backfill Efficacy, Water Balance And Culebra Dolomite Distribution Coefficient Values*. Office of Radiation and Indoor Air, Washington, D.C., EPA-HQ-OAR-2014-0609, June 2017.
- Erkaya, I. A., Arica, M. Y., Akbulut, A., and G. Bayramoglu. 2014. Biosorption of uranium (VI) by free and entrapped *Chlamydomonas reinhardtii*: kinetic, equilibrium and thermodynamic studies. *Journal of Radioanalytical Nuclear Chemistry*, 299(3), 1993-2003.
- Dodge, C. J., Dunn, M., Francis, A. J., Gillow, J. B., Mantione, K., Pansoy-Hjelvik, M. E., and B.A. Strietelmeier. 1998. *Role of Microbes as Biocolloids in the Transport of Actinides from a Deep Underground Radioactive Waste Repository* (No. SAND98-0370J). Sandia National Laboratories, Albuquerque, NM, and Livermore, CA.
- Gorman-Lewis, D., Elias, P. E., and J.B. Fein. 2005. Adsorption of aqueous uranyl complexes onto *Bacillus subtilis* cells. *Environmental science & technology*, 39(13), 4906-4912.
- Gorman-Lewis, D., Fein, J. B., Soderholm, L., Jensen, M. P., and M.H. Chiang. 2005. Experimental study of neptunyl adsorption onto *Bacillus subtilis*. *Geochimica et cosmochimica acta*, 69(20), 4837-4844.

- Haas, J. R., Dichristina, T. J., and R. Wade Jr. 2001. Thermodynamics of U (VI) sorption onto *Shewanella putrefaciens*. *Chemical Geology*, 180(1-4), 33-54.
- Luk'yanova, E., Zakharova, E., Konstantinova, L., and T. Nazina. 2008. Sorption of radionuclides by microorganisms from a deep repository of liquid low-level waste. *Radiochemistry*, 50(1), 85-90.
- Markai, S., Andres, Y., Montavon, G., and B. Grambow. 2003. Study of the interaction between europium (III) and *Bacillus subtilis*: fixation sites, biosorption modeling and reversibility. *Journal of Colloid Interface Science*, 262(2), 351-361.
- Nazina, T. N., Luk'yanova, E. A., Zakharova, E. V., Konstantinova, L. I., Kalmykov, S. N., Poltarau, A. B., and A.A. Zubkov. 2010. Microorganisms in a disposal site for liquid radioactive wastes and their influence on radionuclides. *Geomicrobiology Journal*, 27(5), 473-486.
- Panak, P., and H. Nitsche. 2001. Interaction of aerobic soil bacteria with plutonium (VI). *Radiochimica Acta*, 89(8), 499-504.
- Pang, C., Liu, Y.H., Cao, X.H., Li, M., Huang, G.L., Hua, R., Wang, C.X., Liu, Y.T. and X.F. An. 2011. Biosorption of uranium (VI) from aqueous solution by dead fungal biomass of *Penicillium citrinum*. *Chemical Engineering Journal*, 170(1), pp.1-6.
- Peake, T. 2018. Re: Status of PA issues 2 and 3, actinide uncertainty distribution and microbial colloids. *US Environmental Protection Agency Letter to M. Brown, US Department of Energy*, November 20, 2018.
- Reed, D.T., J.S. Swanson, J.-F. Lucchini, and M.K. Richmann. 2013. *Intrinsic, Mineral, and Microbial Colloid Enhancement Parameters for the WIPP Actinide Source Term*. Los Alamos National Laboratory; Carlsbad, NM. LCO-ACP-18, LA-UR-13-20858
- Reed, D. T., J. Swanson, and F. Stanley. 2019. *LANL/ACRSP Parameter Recommendations for the CRA-2019 Deferred Performance Assessment*. Los Alamos National Laboratory, Los Alamos, NM. LANL-CO ACRSP/CP-ACP-24. LA-UR-19-22787
- Sar, P., and S.F. D'Souza, S. F. 2001. Biosorptive uranium uptake by a *Pseudomonas* strain: characterization and equilibrium studies. *Journal of Chemical Technology Biotechnology*, 76(12), 1286-1294.
- Sar, P., Kazy, S. K., & S.F. D'Souza. 2004. Radionuclide remediation using a bacterial biosorbent. *International Biodeterioration Biodegradation*, 54(2-3), 193-202.
- Sasaki, T., Kubota, T., Mito, S., Kauri, T., and A. Kudo. 2002. Radionuclide sorption to a mixture of anaerobic bacteria in the repository environment. *Journal of Nuclear Science Technology*, 39(3), 954-957.
- Swanson, J., C. Hazelton, F. Stanley, E. Yalcintas, and D. T. Reed. 2019. *Review of the Microbial Colloid Contribution to the WIPP Actinide Source Term*. Los Alamos National Laboratory, NM, USA. Los Alamos Report LA-UR-19-22310.
- Takenaka, Y., Saito, T., Nagasaki, S., Tanaka, S., Kozai, N., and T. Ohnuki. 2007. Metal sorption to *Pseudomonas fluorescens*: influence of pH, ionic strength and metal concentrations. *Geomicrobiology Journal*, 24(3-4), 205-210.

Zheng, X., Shen, Y., Wang, X., and T. Wang. 2018. Effect of pH on uranium (VI) biosorption and biomineralization by *Saccharomyces cerevisiae*. *Chemosphere*, 203, 109-116.

CC4-SOTERM-7 Missing Reference

Please confirm that Nishikaya et al. (2018), cited in SOTERM-5.4.2, is the following reference:

Nishikawa, S., T. Kobayashi and T. Sasaki. 2018. Solubilities and solubility products of thorium hydroxide under moderate temperature conditions. *Radiochimica Acta*, 106:2017-2917.

Nishikaya et al. (2018) is referenced on page SOTERM-57 (Section 5.4.2) but does not appear in the reference list and there is an apparent typographical error in the first author's name.

CC4-SOTERM-8 Pu(III) Intrinsic Colloid Concentrations

Please address the following discrepancies identified in Pu(III) intrinsic colloid concentrations from the tables presented in Reed et al. (2019) as well as their effects on the PA :

SOTERM-8a. Please resolve the discrepancy between intrinsic colloid concentrations cited on page 23 and in Table A-1 of Reed et al. (2019). The Pu(III) intrinsic colloid concentration (1.5×10^{-9} M) cited in the discussion on page 23 differs from the corresponding concentration in Table A-1 (1.49×10^{-8} M). The higher concentration in Table A-1 appears to be correct, based on the data presented in Figure 4-4 of Reed et al. (2013).

SOTERM-8b. Please assess the effects on repository releases due to a higher AM:CONCINT parameter as a result of the changes in the previous question. If the Pu(III) intrinsic colloid concentration of 1.49×10^{-8} M identified in Table A-1 (Reed et al. 2019) is correct, the maximum observed Pu(III) intrinsic colloid concentration will increase from 9.5×10^{-9} M to 1.49×10^{-8} M. Using the oxidation state analogy, the AM:CONCINT parameter for the concentration of Am(III) intrinsic colloids will increase to 1.49×10^{-8} M.

SOTERM-8c. Please explain the differences in some of the Pu(III) data presented by Reed et al. (2019), and data previously included in Reed et al. (2013) (see table below). Please explain whether the 2019 data are a correction of the 2013 data or if the 2013 data are from different samples that were excluded from the 2019 data. Also, please explain the reasons for the data correction or exclusion.

Pu(III) Intrinsic Colloid and Dissolved Concentration Data

| pH | Pu(III) intrinsic colloid (M) | Pu(III) dissolved (M) | Brine | Source |
|-----|-------------------------------|-----------------------|--------|------------------------------|
| 7 | 9.53E-09 | 4.08E-10 | GWB | Reed et al. (2019) Table A-1 |
| 7.7 | 1.00E-08 | 1.10E-09 | GWB | Reed et al. (2013) Table 4-4 |
| 8.3 | 1.49E-08 | 1.14E-09 | ERDA-6 | Reed et al. (2019) Table A-1 |
| 8.4 | 1.20E-08 | 4.10E-10 | ERDA-6 | Reed et al. (2013) Table 4-4 |

Reed, D.T., J.S. Swanson, J.F. Lucchini and M.K. Richmann. 2013. *Intrinsic, Mineral and Microbial Colloid Enhancement Parameters for the WIPP Actinide Source Term. LCO-ACP-18, Revision 0*. Los Alamos National Laboratory, Carlsbad, New Mexico.

Reed, D., J. Swanson, and F. Stanley. 2019. *LANL/ACRSP Parameter Recommendations for the CRA-2019 Deferred Performance Assessment LCO-ACP-18, Revision 0*. Los Alamos National Laboratory, Carlsbad, New Mexico.

CC4-SOTERM-9 Th(IV) Intrinsic Colloid Concentrations Error

Please evaluate the effects on predicted repository releases as a result of increasing the TH:CONCINT, NP:CONCINT, PU:CONCINT, and U:CONCINT parameters to 3.08×10^{-6} M.

In a previous evaluation of colloid concentrations obtained from the Th(IV) filtration data (USEPA 2017), the Agency accepted DOE's explanation that two samples from the Reed et al. (2013) experiment E10C3-1 (ERDA-6 brine from oversaturation) were likely too far from equilibrium solubility to reliably determine Th(IV) intrinsic colloid concentrations (see table below). However, the Agency found that dissolved Th(IV) concentrations in samples from oversaturation in GWB brine (experiment G8C2-1) were very close to equilibrium based on EQ3/6 modeled concentrations (see table below), so these samples should be included in the calculation of Th(IV) intrinsic colloid concentrations. The Agency also noted that the intrinsic colloid parameters should reflect upper bounding values (USEPA 2017).

The 10 nm-filtered Th(IV) concentration data in Table A-1 of Reed et al. (2019) are incorrect and are actually the 5 nm-filtered data from Borkowski (2012, page 19). Using the correct Borkowski (2012) data for samples from the experiments close to equilibrium solubility in GWB (two samples each from UG8C0-2, G8C2-1, and UG9C2-2, see table below), the upper bounding value for Th(IV) intrinsic increases to 3.08×10^{-6} M (highlighted in yellow). This value is significantly higher than the TH:CONCINT value of 4.3×10^{-8} M used for the CRA-2019 DPA. The Th(IV) intrinsic colloid concentration of 3.08×10^{-6} M is consistent with the upper bound observed for Th(IV) intrinsic colloids in NaCl and MgCl₂ brine experiments reported by Altmaier et al. (2004) and consequently is a reasonable upper bound for TH:CONCINT. Based on the assumption that actinides in the same oxidation state will have similar chemical behavior that was used by Reed et al. (2019), the values of the intrinsic colloid parameters for Pu(IV), Np(IV), and U(IV) will also increase to 3.08×10^{-6} M.

Th(IV) Colloidal and Dissolved Concentrations (Borkowski 2012) and Modeled Dissolved Th(IV) Concentrations (CRA-2019 Appendix SOTERM, Table SOTERM-30)

| Brine | Experiment | pH | Th(IV) Intrinsic Colloids (M) | Dissolved Th(IV) (M) |
|------------------------------------|-------------------|-------------|-------------------------------|----------------------|
| GWB | UG8CO-2 (1) | 8.13 | 3.58E-08 | 1.41E-08 |
| GWB | UG8CO-2 (2) | 8.13 | 3.97E-08 | 1.27E-08 |
| GWB | G8C2-1 (1) | 8.54 | 3.00E-06 | 6.17E-08 |
| GWB | G8C2-1 (2) | 8.54 | 3.08E-06 | 5.43E-08 |
| GWB | UG9C2-2 (1) | 9.11 | 9.40E-09 | 3.63E-08 |
| GWB | UG9C2-2 (2) | 9.11 | 2.24E-08 | 2.68E-08 |
| GWB modeled baseline solubility | | | | 5.45E-08 |
| ERDA-6 | E10C3-1 (1) | 9.18 | 1.08E-06 | 5.02E-07 |
| ERDA-6 | E10C3-1 (2) | 9.18 | 7.56E-07 | 8.14E-07 |
| ERDA-6 modeled baseline solubility | | | | 5.44E-08 |

Altmaier, M., V. Neck, and T. Fanghänel. 2004. Solubility and colloid formation of Th(IV) in concentrated NaCl and MgCl₂ solution. *Radiochimica Acta* 92:537-543.

Borkowski, M. 2012. *Numerical Values for Graphs Presented in Report LCO-ACP-08, Rev. 0, Entitled Solubility of An(IV) in WIPP Brine: Thorium Analog Studies in WIPP Simulated Brine, and for Graphs. Published in: Borkowski, M., et al. Radiochimica Acta 98 (9-11), 577-582, (2010). LA-UR- 12-26640; LANL-CO ACRSP ACP-01/2012, Rev. 0. Los Alamos National Laboratory, Carlsbad, New Mexico.*

Reed, D., J. Swanson, and F. Stanley. 2019. *LANL/ACRSP Parameter Recommendations for the CRA-2019 Deferred Performance Assessment, LCO-ACP-18, Revision 0.* Los Alamos National Laboratory, Carlsbad, New Mexico.

U.S.E.P.A. (United States Environmental Protection Agency). 2017. *Technical Support Document For Section 194.24, Evaluation Of The Compliance Recertification Actinide Source Term, Gas Generation, Backfill Efficacy, Water Balance And Culebra Dolomite Distribution Coefficient Values.* Office of Radiation and Indoor Air, Washington, D.C., EPA-HQ-OAR-2014-0609, June 2017.

CC4-SOTERM-10 Th(IV) Concentration Data Revisions

Please explain the differences between Th(IV) data presented for two samples by Reed et al. (2013 and 2019), and data presented by Borkowski (2012) (see table below). Please explain whether the Reed et al. (2013 and 2019) data are a correction of the Borkowski (2012) data or if the Reed et al. (2013 and 2019) data represent different samples than the Borkowski (2012) samples.

Th(IV) Intrinsic Colloid and Dissolved Concentration data

| pCH | Th(IV) intrinsic colloid (M) | Th(IV) dissolved (M) | Brine | Source |
|------|------------------------------|----------------------|-------|---|
| 9.1 | 3.60E-08 | 1.00E-08 | GWB | Reed et al. (2013) Table A2-1, Reed et al. (2019) Table A-2 |
| 9.1 | 2.70E-08 | 2.20E-08 | GWB | Reed et al. (2013) Table A2-1, Reed et al. (2019) Table A-2 |
| 9.11 | 9.40E-09 | 3.63E-08 | GWB | Borkowski (2012) |
| 9.11 | 2.24E-08 | 2.68E-08 | GWB | Borkowski (2012) |

Borkowski, M. 2012. *Numerical Values for Graphs Presented in Report LCO-ACP-08, Rev. 0, Entitled Solubility of An(IV) in WIPP Brine: Thorium Analog Studies in WIPP Simulated Brine, and for Graphs. Published in: Borkowski, M., et al. Radiochimica Acta 98 (9-11), 577-582, (2010). LA-UR- 12-26640; LANL-CO ACRSP ACP-01/2012, Rev. 0. Los Alamos National Laboratory, Carlsbad, New Mexico.*

Reed, D.T., J.S. Swanson, J.F. Lucchini and M.K. Richmann. 2013. *Intrinsic, Mineral and Microbial Colloid Enhancement Parameters for the WIPP Actinide Source Term, LCO-ACP-18, Revision 0.* Los Alamos National Laboratory, Carlsbad, New Mexico.

Reed, D., J. Swanson, and F. Stanley. 2019. *LANL/ACRSP Parameter Recommendations for the CRA-2019 Deferred Performance Assessment, LCO-ACP-18, Revision 0*. Los Alamos National Laboratory, Carlsbad, New Mexico.

CC4-SOTERM-11 Th(IV) Mineral Fragment Colloid Parameter

Please assess whether the mineral fragment colloid parameter for Th(IV) (TH:CONCMIN) should be increased from 2.6×10^{-8} M to an upper bounding value of 6.2×10^{-7} M.

Two samples measuring Th(IV) solubility in oversaturated GWB (Borkowski 2012, experiment G8C2-1) had Th(IV) concentrations of 3.3×10^{-7} M and 6.2×10^{-7} M in the size range consistent with the mineral fragment colloid size range (10 nm to 450 nm, see table below, samples are highlighted in yellow).

Th(IV) Mineral Fragment Colloid and Dissolved Concentration Data

| Brine | Experiment | pH | Th(IV) 450 nm filtration (M) | Th(IV) 10 nm filtration (M) | Th(IV) mineral fragment colloids (M) | Th(IV) dissolved (M) |
|------------|------------------|-------------|---|---|--|---|
| GWB | UG8C0-2(1) | 8.13 | 5.47×10^{-8} | 4.99×10^{-8} | 4.8×10^{-9} | 1.41×10^{-8} |
| GWB | UG8C0-2(2) | 8.13 | 5.07×10^{-8} | 5.24×10^{-8} | 0 | 1.27×10^{-8} |
| GWB | G8C2-1(1) | 8.54 | 3.68×10^{-6} | 3.06×10^{-6} | 6.2×10^{-7} | 6.17×10^{-8} |
| GWB | G8C2-1(2) | 8.54 | 3.46×10^{-6} | 3.13×10^{-6} | 3.3×10^{-7} | 5.43×10^{-8} |
| GWB | UG9C2-2(1) | 9.11 | 4.95×10^{-8} | 4.57×10^{-8} | 3.8×10^{-9} | 3.63×10^{-8} |
| GWB | UG9C2-2(2) | 9.11 | 4.60×10^{-8} | 4.92×10^{-8} | 0 | 2.68×10^{-8} |
| ERDA-6 | E10C3-1(1) | 9.18 | 1.58×10^{-6} | 1.58×10^{-6} | 0 | 5.02×10^{-7} |
| ERDA-6 | E10C3-1(2) | 9.18 | 1.58×10^{-6} | 1.57×10^{-6} | 1×10^{-8} | 8.14×10^{-7} |

Borkowski, M. 2012. *Numerical Values for Graphs Presented in Report LCO-ACP-08, Rev. 0, Entitled Solubility of An(IV) in WIPP Brine: Thorium Analog Studies in WIPP Simulated Brine, and for Graphs Published in Borkowski, M., et al. Radiochimica Acta 98 (9-11), 577-582 (2010)*. LA-UR- 12-26640; LANL-CO ACRSP ACP-01/2012, Rev. 0. Los Alamos National Laboratory, Carlsbad, New Mexico.

CC4-SOTERM-12 Actinide(IV)-Silica Colloids

Please evaluate and summarize in a writeup the possible importance of An(IV)-silicate colloids to mobilization of Th(IV), U(IV), and Pu(IV) in WIPP brines. Please include any relevant tables or figures in this writeup as needed.

Recent publications, for example, Zänker et al. (2016), Neil et al. (2018), and Neil et al. (2019), have described the potential mobilization of +IV actinides by colloids that include structural An(IV) and silica. Possible formation of these colloids under WIPP repository conditions was not addressed in the CRA-2019.

Neill, T.M., K. Morris, C.I. Pearce, N.K. Sherriff, M.G. Burke, P.A. Chater, A. Janssen, L. Natrajan, and S. Shaw. 2018. Stability, composition, and core-shell particle structure of uranium(IV)-silicate colloids. *Environmental Science and Technology* 52:9118-9127.

7/17/2020

Neill, T.M., K. Morris, C.I. Pearce, L. Abrahamsen-Mills, L. Kovarik, S. Kellet, B. Rigby, T. Vitova, B. Schacherl, and S. Shaw. 2019. Silicate stabilization of colloidal UO_2 produced by uranium metal corrosion. *Journal of Nuclear Materials* 526.

Zänker, H. S. Weiss, C. Henning, V. Brendler, and A. Ikeda-Ohno. 2019. Oxyhydroxy silicate colloids: a new type of waterborne actinide(IV) colloids. *ChemistryOpen* 5:174-182.