DOE Comments to ExxonMobil's Responses (and the Attached Proposed Work Plan) to NRC's Request for Additional Information Regarding the *Highland Uranium Mine and Millsite Request for Amendment to Radioactive Materials License SUA-1139; Application to Amend Existing Alternate Concentration Limits* (and Proposal to Include the "Pit Lake" within the Long-Term Surveillance Boundary)

The purpose of this review, performed at the request of the U.S. Nuclear Regulatory Commission (NRC), is to provide comments on ExxonMobil Environmental Services Company's (EMES) responses to NRC's Request for Additional Information (RAIs), dated April 12, 2013, and its attached proposed work plan titled *Highland Mine & Millsite Supplemental Hydrologic and Geochemical Characterization Work Plan* (work plan). NRC developed the RAIs in response to EMES's May 2011 license amendment request titled *Highland Uranium Mine and Millsite Request for Amendment to Radioactive Materials License SUA-1139; Application to Amend Existing Alternate Concentration Limits* (ACL application), which includes a proposal to include the incompletely reclaimed and contaminated former open pit uranium mine, referred to as the "pit lake", within a revised long-term surveillance boundary (LTSB).

The U.S. Department of Energy (DOE) will ultimately become the long-term custodian of the Highland uranium mill tailings disposal site upon final regulatory closure, and will be responsible for long-term custody and care (i.e., site surveillance and maintenance). DOE therefore provides these requested comments for NRC's consideration in their sole determination as to an appropriate reply to EMES's responses to the RAI and the associated proposed work plan. For many of the RAIs, DOE notes that the response defers to the work plan. In many of these responses, DOE suggests that an adequate answer to the RAI is not provided in the proposed work plan. For these RAIs DOE offers a justification/reason as to the importance of responding fully to the specific RAI and reflecting that position in the proposed work plan.

DOE's comments are divided into three groups; comments to specific RAIs, comments to the proposed work plan, and general comments.

Comments to Specific RAIs

RAI-Geo1 – The NRC asks EMES to provide additional information regarding the location of all "known" boreholes (wells and investigation boreholes) on the site and neighboring mine sites used to develop the geological framework model discussed in the ACL application (Section 1.2.2.6) … EMES must also state whether the 3-dimensional geologic framework model provides any additional or new insights or a change in the understanding of the lateral extent and continuity of the strata in the LTSB… Additionally, the impact of these insights on the hydrological and geochemical processes should be discussed.

If the model was based on existing data and did not change any previously assumed hydrostratigraphic conceptualizations, and was not used to develop additional or new insights in the lateral extent and continuity of the geologic strata at the site, DOE would propose that the licensee better characterize the flow and extent of any tailing-related contamination (i.e., hazardous constituents) in the Tailings Dam Sandstone (TDSS) and backfill material between the tailings impoundment and the pit lake (a proposed POE) in order to support the revised conceptual model that claims the TDSS (and any hazardous constituents from the tailings cell within it) have indeed migrated to the pit lake rather than having attenuated as presented in the earlier conceptual model.

RAI-H1 – The NRC asks EMES to "Clarify the current conceptualization of the regional groundwater flow." In the response EMES discusses "additional data collection on water level and water quality within the primary Ore Body Sandstone (OBSS) units in the vicinity of the existing pit lake and tailings impoundment from newly-installed monitoring wells." There is no mention about collecting additional data from the TDSS unit, and Section 2.1 (Monitoring Well Installation) of the proposed work plan clearly states that all new wells will be installed in the OBSS units. As stated above, the TDSS is the formation that EMES suggests is the conduit for contamination from the tailings impoundment to the pit lake. Without additional TDSS data an important aspect of an updated conceptual model is missing. DOE questions why none of the new wells are targeted to provide additional information about the TDSS formation, particularly between the tailings impoundment and the pit lake. There also is concern that there are not proposed data collection points (i.e., borings and wells) in the vicinity of POC well 0175, particularly between the tailings impoundment and the pit lake. The locations of the seven nested wells proposed to be installed are shown on Figure 1 of the work plan; all are in the OBSS, none are in the TDSS. DOE suggests that this is an important oversight of the proposed work plan.

RAI-H2 – The NRC inquires about "excluding explicit effects of in situ recovery (ISR) or oil and gas exploration operations in the flow analyses." EMES in the response agrees to review publicly available data and documents related to these external activities. ISR activities typically occur over an extended period of time such that these operations could have an effect on the integrity of the site. Any activity which has the potential to change the flow for any length of time should be included for calibrating a model which is going to be used for predictive purposes. If it is determined that ISR effects provide important changes to the conceptual site model, DOE would suggest it should also trigger the need for an updated groundwater flow model.

RAI-H4 – Two comments to consider are discussed. (1) The pre-mining historical water elevation in the area of the pit lake is estimated to be approximately 5110 to 5120 feet above mean sea level (amsl). The groundwater model presented in the ACL application (Tetra Tech 2007)[1] predicted the pit lake would reach a long-term elevation of 5060 feet amsl. This is a difference of 50 to 60 feet between historical and predicted long-term water levels. This is a significant difference considering that water levels eventually naturally return to pre-existing conditions in the absence of outside influences (extraction/injection). Net evaporation, precipitation minus evaporation, is approximately 34 in/yr. If the pit lake eventually returns to a historical water level of ~5110 amsl minus the net evaporation (~3 ft/yr), the water level would be well above the model predicted value and a flow through condition could occur. DOE notes that previous modeling, Exxon Production Research Company (1983)[2], Shepherd Miller

[1] Tetra Tech, 2007 "*Long Term Geochemical Evolution of the Highland Pit Lake*". Highland Uranium Project Request for Amendment To Radioactive Materials License SUA-1139.
[2] Exxon Production Research Company (1983*) Surface Mine Reclamation Lake Study for Highland Uranium Operations (Updated)*. April.

Incorporated (1998)[3], and Carovillano (1998)[4], predicted the long-term steady-state pit lake elevation would be at least 5117 feet amsl which is close to the historical pre-mining water level elevation. (2) Figure 1-8 of the ACL application show four cross-section locations across the site. Figure 1-12, which is cross-section D-D' indicates the potential for water to discharge toward North Fork Box Creek through the Pit 1/Pit 2 backfill areas and then the OBSS-50 and OBSS-40 formations, well below the model predicted long-term water elevation of 5060 feet amsl. DOE suggests these discrepancies should be rectified.

RAI-H5 – The NRC asks EMES to "Clarify the technical basis for the boundary conditions used in groundwater modeling." Water level data can be used to estimate the 'head' parameter required for using General-Head Boundary (GHB) as a boundary condition. DOE suggests that the use of GHBs is somewhat subjective at the discretion of the modeler and is typically based on experience. Layer 2 in the ACL application's groundwater flow model (Tetra Tech 2007) represents the TDSS formation. DOE also suggests that without additional data from the TDSS formation realistic values for 'head' becomes problematic.

RAI-H6 – In RAI-H6 the NRC asks EMES to "Clarify the influence of drainages east of the tailings basin on groundwater flow paths, geochemistry, and radionuclide transport". In the response, EMES states it will "include a detailed review of potential influence of regional OBSS outcrops on groundwater flow and long-term behavior of the Highland Pit Lake in the updated site conceptual model." DOE notes that there is no mention in the responses about geochemistry or radionuclide transport, and therefore thinks that the RAI is not fully answered. In the response to H15 there is mention of "The conceptual geochemical site model . . ." and "the geochemical conceptual model." DOE again notes that there is no mention in the responses to the H-series RAIs about updating or potentially updating the ACL application's geochemical model titled "Long Term Geochemical Evolution of Highland Pit Lake" (Tetra Tech 2007).

RAI-H9 – The NRC ask EMES to "Provide a technical basis for considering the Pit Lake hydrology and Southeast Drainage independently." DOE suggest that these are not independent hydrologic systems. Although there is a subsurface groundwater flow divide, mounding caused by tailing impoundment seepage and the subsequent dissipation of that mound will likely change the location of that divide over time. DOE submits that data collected from the proposed work plan characterization should be reassessed and revised to include the development of a single conceptual site model that includes flow in all directions from the tailings impoundment. DOE points out that a similar situation existed at the Panna Maria site (an UMTRCA Title II site in Texas) where three disconnected models were used to model the tailings impoundment. DOE highlighted numerous problems with these models and communicated them to the licensee and the Texas Commission on Environmental Quality. Eventually a model that encompassed the entire site and an extensive area beyond that included in the separate models was developed. The results of the single model were significantly different that those of the separate models.

RAI-H10 through H14 – The NRC ask EMES to "clarify the technical basis", "provide technical basis", and "justify" concerns related to the ACL application's groundwater flow

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[3] Shepherd Miller, Inc. (1998) *Hydrologic and Chemical Evolution of Highland Pit Lake, Converse County, Wyoming*. November 12.
[4] Carovillano, R. (1998) *Highland Tailings Basin Groundwater Study, Final Report*. Prepared for Exxon Production Research Company and Exxon Coal and Minerals Company. August.

model (Tetra Tech 2007). EMES's response is that "additional water level and water quality data" and "publically- available regional data" "will be reviewed and used to update the site conceptual model" and "will form the basis for updated assessments and potentially an updated groundwater flow model . . .". DOE suggests there is considerable uncertainty in this response which is at the discretion of EMES. If EMES decides that the data does not justify updating the conceptual site model and an updated groundwater flow model is not developed, then DOE suggests that these RAIs are not fully answered. DOE recommends the collection of site-specific water level and water quality data be collected (per a revised work plan) and used in an updated comprehensive site-wide groundwater flow model.

RAI-H15 – NRC asks EMES to provide technical justification for not evaluating a potential flow pathway north of the tailings dam.

EMES indicates that contaminants are not likely to be transported in the TDSS to the north because of reducing conditions that exist (see Figure H15-2 of the RAI responses). It is noted that contaminants in the TDSS would be expected to rapidly attenuate in this flow direction. It is unclear why the same would not be expected to have occurred for fluids moving to the west toward the pit lake, consistent with the early conceptual model for the site (i.e., that attenuation occurred within a relatively short distance from the tailings impoundment and that the elevated concentrations found within the pit lake were a result of oxidation of the exposed ore body). If elevated iron is indicative of reducing conditions, it would appear that these same conditions occur to the west based on iron concentrations in wells 114 and 175 (see Figure H15-3 of the RAI responses). It is not clear how the TDSS could have served as the conduit for significant contaminant movement to the pit lake based on these limited analytical data. Consistently low uranium concentrations observed in wells adjacent to the tailings impoundment, particularly when compared to concentrations observed in the pit lake suggest the same rapid attenuation proposed in the northerly flow direction. DOE suggests that additional geochemical characterization of the TDSS to the north and to the west of the impoundment would also be appropriate.

RAI-H17 – The NRC ask EMES to "Clarify the calculations and conclusions of the verification analysis of the estimated groundwater flux from the GHB in Tetra Tech (2007)." DOE suggests that the response by EMES does not directly answer this RAI. DOE notes that it defers to the proposed work plan, which also does not answer the RAI. See comments above for H5 and H10 through H14.

RAI – GC-1 – The NRC ask EMES to provide data and assumptions associated with modeling of surface reactions involving the sorption and desorption of uranium, selenium, nickel, and chromium. The data should include; (1) the reactions postulated; (2) the equilibrium constants assumed and the basis for those constants; and (3) the MINTEQ database used in the calculations.

Regarding EMES's response to this NRC comment, the final sentence of the response reads: "The assumed HFO surface area and surface site densities are the best estimates for these parameters developed through a comprehensive literature review and evaluation (Dzombak and

Morel, 1990)." However, Mahoney et al., (2009)[5] provides a reevaluation of the Dzombak and Morel database which corrects the problems associated with the linear free energy relationship method of calculating the reaction constants used to calculate the reaction constants for their model (D-M model). The more recent work of Mahoney et al., (2009) used experimental data and an inverse fitting routine which calls the USGS code PHREEQC, and optimizes the log K values of the surface complexation reactions for uranium adsorption on HFO. Comparison of the D-M model to the experimental data shows that D-M over predicts uranium adsorption at pH values less than 7 and under predicts at pH greater than 7.

Furthermore, Mahoney et al., (2009) expanded the D-M model to include uranyl-carbonate surface complexes. Contrary to the statement on page 2-12 of the application amendment which states: "The presence of uranium-carbonate complexes limits the extent of additional uranium adsorption and therefore contributes to uranium mobility (Langmuir, 1997)." Figure 4 in Mahoney et al., (2009) demonstrated that uranium-carbonate surface complexation with HFO are important across a broad range of pH, and that these complexes cannot be neglected in carbonate bearing systems.

While the Dzombak and Morel model is pedigreed and has a long legacy in the modeling of surface complexation reactions more recent and improved data exist which build upon and expand the D-M model. The example provided, Mahoney et al., (2009) provides an improved and expanded data set which better fits experimental data and should be used in future modeling efforts.

DOE suggests additional issues with this model include:

- The PHREEQC model. Why was mixing of the contaminated tailings solution with the water of the TDSS neglected? The water native to the TDSS would not be 100% displaced from the TDSS with the influx of contaminated water, rather there would be a long term mixing that would continue until some pseudo steady state mixing of the two fractions was realized. Mixing ratios of 25%, 50%, 75% and 100% contaminated water could have been tested to better understand the role of this process in the attenuation of the contaminants of interest.
- It is stated on page 2-11: "the partial pressure of CO2(g) was adjusted to obtain an equilibrium pH of 7.0 commonly observed in the impacted groundwater." First of all there is no analysis provided of the "impacted" groundwater for comparison purposes, and secondly a high pCO2 = 10-1.9 is required to attain a pH of 7, this may not be a realistic value of pCO2, which begs the question what is the calculated pCO2 of the "impacted" groundwater based on its measured analysis?
- In the SCM, ferrihydrite, molecular weight 107 g/mol, was used to represent HFO. The D-M model is built around Goethite as HFO, molecular weight 89 g/mol. Thus, the precipitated mass of HFO would be greater with Goethite, and the SCM could have been coupled to Goethite formation to render a more coupled model. Also, pre-equilibrating the adsorbing phase with the natural background groundwater makes little sense since the HFO presumably precipitates from the contaminated tailings solution.

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[5] Mahoney, J.J., S.A. Cadle, and R.T. Jakubowski. 2009. Uranyl Adsorption onto Hydrous Ferric OxidesA Re-Evaluation for the Diffuse Layer Model Database. Environ. Sci. Technol. 43, 9260–9266.

- The PHREEQC input file included in Appendix D does not contain the key words "Exchange_Master_Species" and "Exchange_Species", and neither does the Minteq.v4.dat database. Without these key words in the input file or database the code will fail to execute the run.
- The phases, Al(OH)3(a) and Cr(OH)3(a), included under the key word “Equilibrium_Phases”, as written in the provided input file do not exist in Minteq.v4.dat. For both of these phases the “(a)” is replaced with “(am)” in Minteq.v4.dat, the provided spelling will also cause the run to fail.

DOE also suggests that the above comments and the use of incomplete and obsolete surface complexation reaction constant data, sheds significant doubt on the results of the PHREEQC transport model as presented in permit amendment. DOE thinks that resolution of these issues would likely change the results of the model to lessen the impact of the tailings solution downgradient of the tailings impoundment.

RAI-HAZ3 – Clarify whether the bases for the proposed ACL for well 175 included considerations of ecological hazards, and if so provide the information or references that support the no environmental hazard determination for the proposed well 175 ACL.

As mentioned previously, DOE notes that the proposed work plan does not include any new wells constructed in the TDSS between POC well 175 and the pit lake. DOE would propose that more than one TDSS wells be constructed down gradient of well 175, between it and the pit lake (the proposed POE), such that the revised conceptual model can be validated (i.e., that the contamination in the pit lake is tailings-related rather than ore-body-related as was thought in the original conceptual model). DOE offers that the migration of hazardous constituents from the tailings liquids could be better monitored and environmental risk at the POE (pit lake) could be better understood. In conjunction with installing these wells, DOE suggests that EMES should collect data which would demonstrate that tailing-related contamination within this formation would not attenuate before reaching the pit lake. DOE also thinks that EMES should also be expected to demonstrate that the TDSS is hydraulically connected to the pit lake and that tailings fluids would preferentially flow from the tailings impoundment to the pit lake. DOE recommends that calculations which include documented historical flows, groundwater mixing, and actions taken to prevent tailings fluid migration should be incorporated to more accurately determine the volume and concentration of the contaminated groundwater, which EMES indicates in their revised conceptual model has reached the pit lake.

RAI-CA1 – NRC asks EMES to provide additional information or bases for not evaluating corrective action for the uranium exceedance at well 175.

EMES in their response indicates that groundwater flow in the TDSS in the vicinity of POC well 175 “reports” to the backfill and eventually to the pit lake. DOE offers that data to substantiate this premise is not present in the available documentation to date, nor is it addressed in any of the RAI responses or through additional data collection included in the proposed work plan. DOE points out that for almost thirty years EMES refuted this scenario (i.e., the early conceptual model indicated that the tailing fluids would attenuate well before reaching the pit lake and that the elevated concentration observed in the pit lake were the result of oxidation of the exposed ore body). DOE suggests that EMES should provide new data, not just revised models, that clearly and decisively demonstrate how the previous modeling was in error and that hazardous constituents (tailing fluid) from the tailings impoundment have now reached the pit lake rather than attenuate as they indicate is occurring north of the impoundment.

RAI-RA1 – The NRC asks EMES the impact of the pit lake on migratory birds.

The 2007 Ecological Risk Assessment (Arcadis 2007)[6] provides evidence that reclamation of the Highland disposal site is incomplete. Ecological exposures were discounted based on the lack of habitat, although considerable uncertainty exists as to whether the habitat is lacking because of the lack of a primary food source and the immaturity of the ecosystem or because of the contaminants currently present in the water and sediments. DOE thinks that an ecosystem will establish at the pit lake over time, and other flooded open pit mines may provide valid analogs to the Highland site for predicting the characteristics of an established ecosystem. DOE suggests the ecological risk assessment for the Highland site should be based on these analog sites.

DOE also suggests that the EMES has not adequately proven that the ore-derived contaminants of potential ecological concern in the pit lake will not pose a substantial future risk to human health or the environment. The U.S. Fish and Wildlife Service in a letter to NRC dated October 28, 2011 expressed concern with the concentrations of selenium in the pit lake with regard to posing an ecological risk, indicating that concentrations in invertebrates ranged from 6 to 287 ug/g and that dietary selenium concentrations greater than 3 ug/g are known to cause adverse effects (DOE notes that the Colorado aquatic acceptable level for selenium from an eco-risk standpoint is 0.005 mg/L).

Additionally, EMES has not provided an adequate technical basis for asserting that the newly proposed uranium ACL or the current selenium ground water protection standard will not continue to be exceeded in the pit lake (a proposed POE) under long-term management. Both of these constituents currently exceed their respective groundwater protection standards in the pit lake (see trend graphs of data through 2010 in Attachment 1), and long-term concentrations of uranium are predicted to go even higher. According to EMES's 2006 presentation regarding the Highland site, EMES predicted long-term uranium concentrations in the pit lake would reach 4.5 mg/L and indicated that measured concentrations were at 3.12 mg/L, both of which are greater than the newly proposed uranium ACL of 3.0 mg/L for POC well 175.

Based on the above comments, DOE recommends that NRC consider rejecting the proposal to include the incompletely reclaimed and contaminated pit lake within the LTSB (as presented in EMES's ACL application).

Comments to the Proposed Work Plan

General

The proposed work plan does address collecting any additional data to substantiate EMES's claim that significant concentrations of tailings fluids have migrated to the pit lake through the TDSS as suggested in their revised conceptual model (i.e., the work plan does not propose any

[6] Arcadis U.S. Incorporated, 2007. Final Highland Pit Lake Ecological Risk Assessment, April.

additional data collection points between the tailings impoundment and the pit lake which would validate the claim that significant concentrations of tailings fluids occur in the TDSS and have migrated to the pit lake and not attenuated; particularly in light of the concentrations observed in POC well 175 as compared to concentrations observed in the pit lake, see Attachment 1). DOE suggests that the work plan be revised to collect additional data which would validate the claim that significant concentrations of tailings fluids occur in the TDSS and have migrated to the pit lake, and that the elevated concentrations observed in the pit lake are not the result of oxidation caused by the exposed ore body.

Updated Groundwater Flow and Geochemical Models Comment

It is not clear to DOE if an updated numerical groundwater flow model is going to be developed. In Section 2.0 (Supplemental Hydrological Characterization) of the proposed work plan it states that "ExxonMobil has agreed to update the conceptual site model and numerical groundwater flow model for the site." However, throughout the responses to the H-series RAIs EMES uses statements such as "ExxonMobil has agreed to reassess the conceptual and numerical groundwater flow model for the site" and "newly collected data and the updated site conceptual model will form the basis for updated assessments and potentially an updated groundwater flow model that will supersede results and assumptions made during development of the Tetra Tech (2007) model". Based on additional comments regarding the modeling provided, DOE recommends that an updated comprehensive site-wide numerical groundwater flow model be developed that would support the revised conceptual model.

Geochemistry Comment

Section 3.0 (Supplemental Geochemical Characterization) of the proposed work plan contains subsections on the Eastern Drainage (Section 3.1), the Southeast Drainage (Section 3.2), and the North Area (Section 3.3). DOE notes that there is nothing related to the geochemistry between the tailing impoundment and the pit lake or between the OBSS units and the pit lake. Based on the EMES argument that a significant portion of the contamination in the pit lake originated from the tailings impoundment via the TDSS formation, DOE offers that this is a major oversight in the work plan with regard to the additional data collection that is being proposed.

General Comments

ExxonMobil's ACL Amendment application proposes to more than double the area contained within the long-term surveillance boundary (LTSB) for transfer to DOE. As it is DOE's policy to maintain as small a footprint as necessary, DOE has concerns with this proposal. Particularly troubling to DOE is the licensee's proposal to extend the site boundary westward to include an incompletely reclaimed and contaminated pit lake within the LTSB. The basis for this proposal involves a complete revision of the conceptual site model for the site that has been in place for more than two decades. This model revision does not appear to be based on new data or information that was obtained to refute the original model, but rather is proposed by the licensee as an alternative to backfilling the pit lake now that the water quality is found to be unacceptable for the originally intended uses (i.e., initially for recreation, then for livestock watering).

The original conceptual model indicated that contaminant movement from tailings seepage would be strongly attenuated by subsurface formations and be very limited in extent. It was not anticipated that contamination would enter the pit lake. When high concentrations of certain constituents, particularly uranium, were observed in pit lake samples, this was ascribed to oxidation and leaching of natural ore-bearing formations exposed on the sides of the excavated pit. Monitoring data collected during and after groundwater corrective action was consistent with the original conceptual model. There is no monitoring data to suggest that the contaminant seepage has migrated beyond the originally proposed transfer boundary in the direction of the pit lake. POC wells at this boundary have remained below groundwater protection standards, with the exception of a single minor excursion.

DOE offers the following information that supports this original conceptual model:

- Only a small portion of the TDSS was actually in direct contact with the tailings.
- No concentrations of uranium in groundwater have been observed that approach that of the pit lake (around 3 mg/L) let alone that observed in tailings fluids (up to 13 mg/L). POC well 175 uranium concentrations have remained less than the 0.03 mg/L standard for over 20 years and have only recently had one exception—the 0.0381 mg/L result in August 2010 (see Attachment 1 trend graphs).
- Pumping mill tailings to the tailings basin ceased in 1984. Seepage from the tailings impoundment was controlled from early on in the history of the site by a groundwater corrective action that was operating from November 1989 and continued through May 1999 (according to RAI-CA1).
- Seepage from the tailings impoundment continued to decline over time as the clay fraction of the tailings and the precipitation of gypsum decreased permeability of the TDSS.
- The potentially hazardous constituents from the tailings were said to be confined to an area close to the tailings basin in an area between the tailings basin and the mine backfill. This was corroborated by monitoring data (1998 ACL application).
- $^{234}U/^{238}U$ ratios observed in the pit lake are used as evidence that the uranium is most likely milling-related, with ratios close to 1.0. However, it has been demonstrated that leaching of uranium ore bodies under oxidizing conditions also produces high concentrations of uranium with $^{234}U/^{238}U$ ratios close to unity (Caine et al. 2011)[7]. This interpretation is consistent with the originally proposed model for pit lake water quality and appears to be validated by over 20 years of monitoring data (see Attachment 1 trend graphs).
- The lack of other processing-related constituents in the pit lake concentrations compared to background levels concentrations observed in POC well 175.

Despite the above information and in the absence of new data, EMES recently proposed an alternative conceptual model in which substantial amounts of highly concentrated tailings fluids migrated from the tailings pile through the TDSS unit and discharged into the pit lake. Geochemical modeling was presented in the ACL application (Tetra Tech 2007) in an attempt to demonstrate that an influx of tailings fluids to the pit lake was required to produce the observed

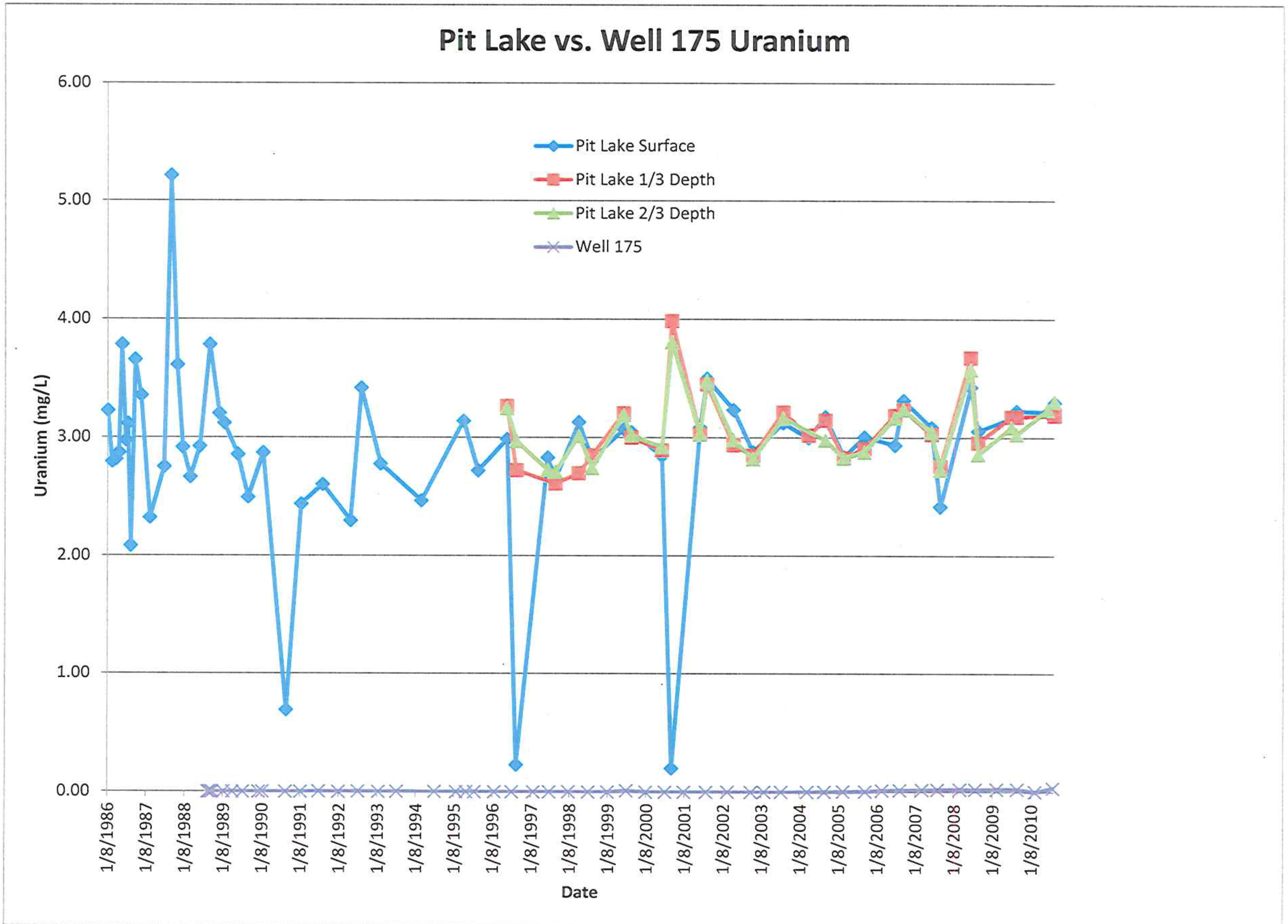
[7] Caine, J.S., R.H. Johnson, E.C. Wild, 2011. Review and Interpretation of Previous Work and New Data on the Hydrogeology of the Schwartzwalder Uranium Mine and Vicinity, Jefferson County, Colorado; United States Geological Survey Open File Report 2011-1092.

pit lake water chemistry. DOE reviewed this model (see Attachment 2) and found a number of deficiencies, including the following:

- Influx of highly concentrated tailings fluids to the pit lake is purely speculative and not based on any empirical monitoring data.
- If the TDSS served as a conduit for the transport of highly concentrated tailings fluids, it would be expected that some remnant contamination would be present in this formation. This is based on DOE’s experience at other sites with uranium contamination in groundwater. No such remnant contamination has been observed.
- The most recent geochemical conceptual model of the pit lake may be too over simplified to adequately represent actual conditions in the lake.
- The flux values from the various sources into the lake are highly uncertain, the perched aquifer contribution for example is based on a personal communication with no documentation.
- Verification/validation of the hydrologic budget is plagued with problems. The equation (3-3) used for mass balance of chloride is incorrect. The composition of the tailings influent used in the model was not realistic, and concentration values were changed to fit the measured lake concentration data.
- Water level elevation data for the pit lake have been collected since 1986, and this data could have been used in combination with the chloride data to validate the hydrologic model, however, it was not.
- To match the observed pit lake uranium, selenium, and sulfate data, many liberties were taken with the model input concentrations to force the model into a match the measurements.
- The modeling report points to gypsum precipitation as an important chemical process to remove sulfate from the pit lake. However, PHREEQC model calculations show that calcite precipitation suppresses gypsum saturation and subsequent precipitation.

Based on DOE’s technical review of the modeling, it was concluded that by manipulating model input parameters to unrealistic values, it was possible to fit the observed chemical trends in the pit lake. However, DOE offers that other explanations for the pit lake chemistry are just as likely, including the original site conceptual model. DOE does not believe that the rationale for extending the current site boundary to include the pit lake is adequate to justify such a large change in the remedy.

Attachment 1
Trend Graphs


Pit Lake vs. Well 175 Uranium
Pit Lake Surface
Pit Lake 1/3 Depth
Pit Lake 2/3 Depth
Well 175
Uranium (mg/L)
6.00
5.00
4.00
3.00
2.00
1.00
0.00
1/8/1986
1/8/1987
1/8/1988
1/8/1989
1/8/1990
1/8/1991
1/8/1992
1/8/1993
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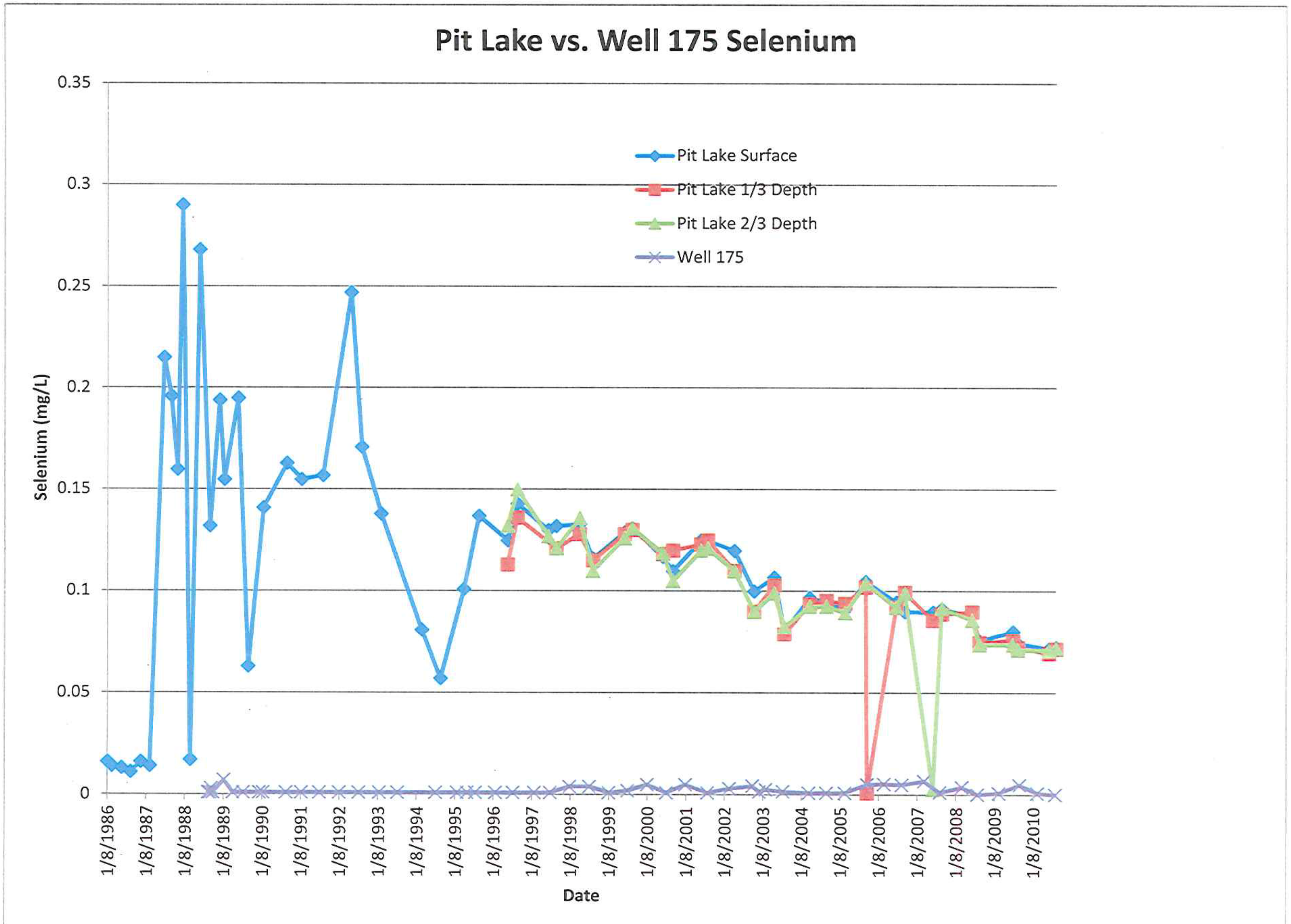
1/8/2009
1/8/2010
Date


Pit Lake vs. Well 175 Selenium
Selenium (mg/L)
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Pit Lake Surface
Pit Lake 1/3 Depth
Pit Lake 2/3 Depth
Well 175
1/8/1986
1/8/1987
1/8/1988
1/8/1989
1/8/1990
1/8/1991
1/8/1992
1/8/1993
1/8/1994
1/8/1995
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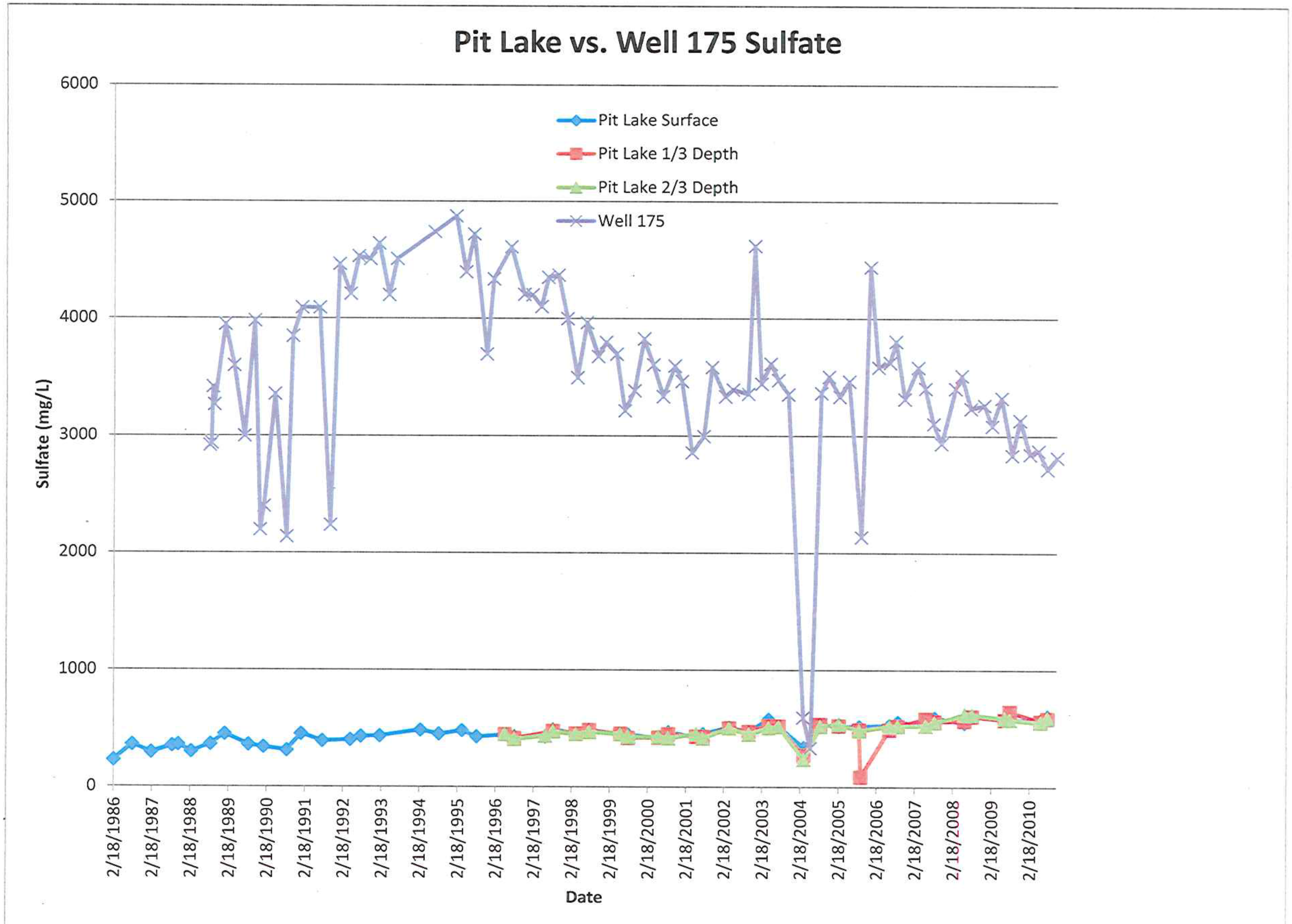
1/8/2007
1/8/2008
1/8/2009
1/8/2010
Date


Pit Lake vs. Well 175 Sulfate
Pit Lake Surface
Pit Lake 1/3 Depth
Pit Lake 2/3 Depth
Well 175
Sulfate (mg/L)
6000
5000
4000
3000
2000
1000
0
2/18/1986
2/18/1987
2/18/1988
2/18/1989
2/18/1990
2/18/1991
2/18/1992
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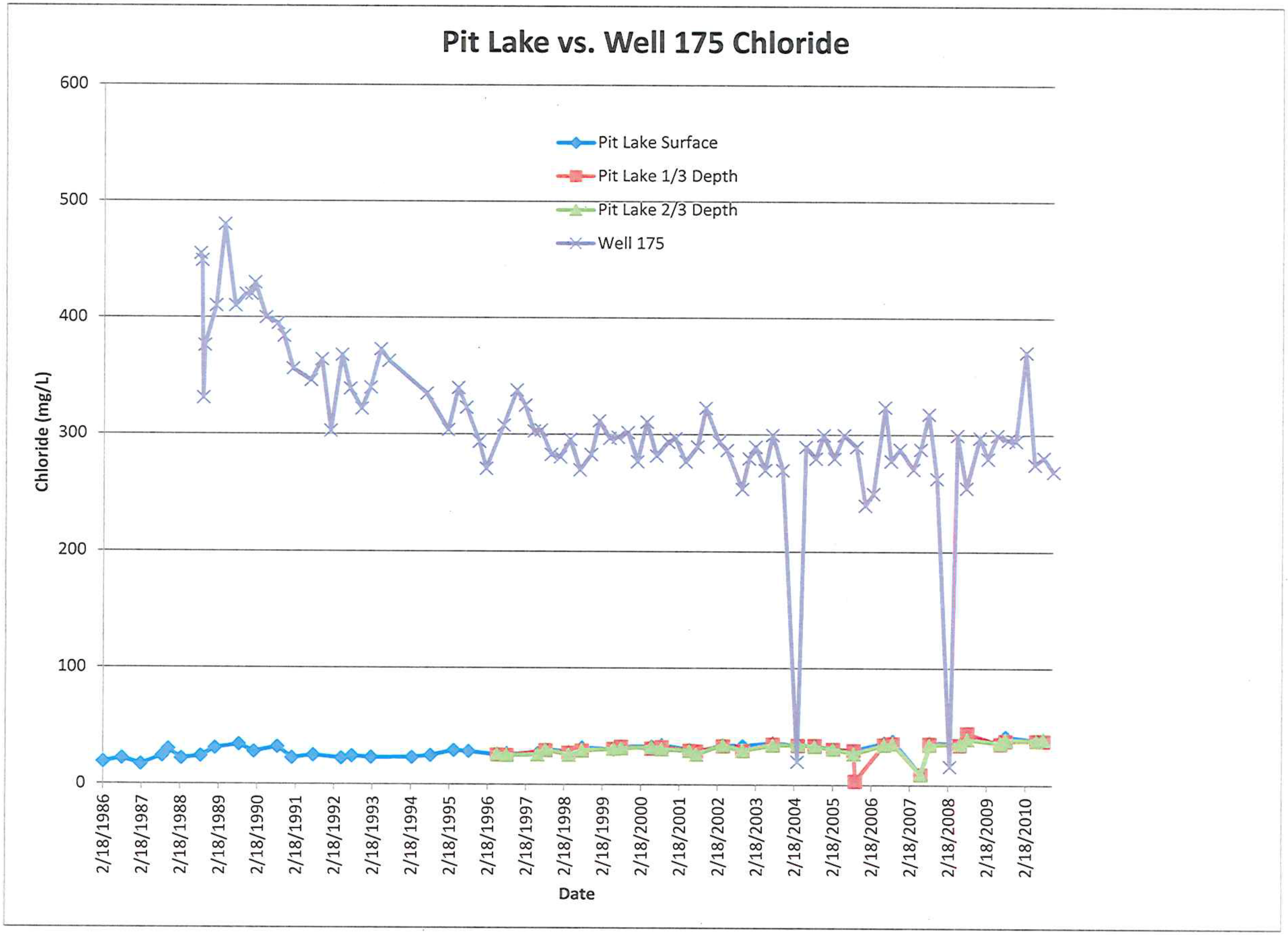
Date


Pit Lake vs. Well 175 Chloride
Chloride (mg/L)
600
500
400
300
200
100
0
Pit Lake Surface
Pit Lake 1/3 Depth
Pit Lake 2/3 Depth
Well 175
2/18/1986
2/18/1987
2/18/1988
2/18/1989
2/18/1990
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Attachment 2
Critical Review of "Long-Term Geochemical Evolution of the Highland Pit Lake", Tetra Tech (2007)

Critical Review of "Long Term Geochemical Evolution of the Highland Pit Lake", Tetra Tech (2007)

Introduction

The Tetra Tech (2007a) document "Long Term Geochemical Evolution of the Highland Pit Lake" is used as an argument by ExxonMobil to seek alternate concentration limits, and amend the regulatory permit for the Highland Mine site. The model used by Tetra Tech examines the mass loading of the Highland Pit Lake by the various sources of inflow and outflow to the lake. Model calibration was achieved by manipulating the model input parameters to match the first 20 years of the measured chloride data of the Pit Lake. The model was then used to extrapolate these chemical trends 1000 years into the future. In a final step Tetra Tech uses the USGS aqueous geochemical code, PHREEQC, to set simple mineral equilibrium constraints on the calculated time-series compositions output by their model. The following review will examine the model inputs, their implementation, and the model output. Note that for this review no model runs of Tetra Tech's dynamic site model (DSM) were performed, neither with the Tetra Tech's input parameters, nor with any updated input parameters, only a few simple PHREEQC calculations were performed using the output of the DSM provided in Attachment 2 of Tetra Tech (2007a).

Code

Tetra Tech used the code Stella©, available from ISEE Systems (http://www.iseesystems.com/) to create a DSM of the evolution of the Pit Lake chemistry. Software such as Stella is useful for simulating complex systems with many input variables, variables that may carry large uncertainties, because of the ease in which parameters may be varied to best fit the measured data. A dynamic system model was created to simulate the inflows, outflows and track the chemistry of the Pit Lake as it developed over the 1000 year model period. The DSM does not perform any chemical manipulations with the water composition, it simply maintains totals of all of the chemical constituents that have entered or exited the lake.

The geochemical manipulations of the water compositions that were output by the DSM were performed using the well known USGS code PHREEQC. PHREEQC was used to perform simple mineral equilibrium calculations on the water chemistry to precipitate calcite and gypsum while maintaining a constant partial pressure of carbon dioxide. There was no mention of which thermodynamic database was used to perform these calculations, which makes it difficult to exactly reproduce the results obtained by Tetra Tech.

Conceptual Model

The hydrologic conceptual model of the Pit Lake is that of a closed basin lake, where it receives water and chemical inputs from groundwater inflow, surface runoff, and direct precipitation, and the only outflow is from evaporation, there are no outflows in the form of either surface or groundwater. The report was reviewed assuming this conceptual model is correct.

Geochemically, the Pit Lake is treated like a well mixed reservoir, i.e., homogenous chemical conditions exist throughout, of constant temperature where the only chemical process effecting the concentration of constituents is mineral precipitation. The viability of this conceptual model is supported by the relatively constant chemistry with depth, however the changing temperature and lack of carbonate data with depth, as well as the changing dissolved oxygen content with depth could influence the local chemistry by creating microenvironments, and contribute to uncertainty in the conceptual model.

Water Balance

The water balance for the Pit Lake is based on the estimates of flow from the geologic units, surface run off, and direct precipitation. The hydrologic study of MFG (2003) provided the groundwater flux values for the ore-body sand stone (OBSS), which comprise ~30% of the total flow, and the Tailings-dam sand stone (TDSS) at 10% of the total. The perched aquifer contributes 23% of the flow, a value that is documented in a personal communication by Range (1998) with very little rationale provided in support of this flow value. Values for tailings basin seepage vary considerably between the four studies cited in the report (Table 3.4), the value used in the DSM was set to 30 gpm for the first 20 years (1984 – 2004) of the modeled period, after which it was set to zero. Regarding tailings seepage, Tetra Tech 2007b states:

> *"Early estimates of this groundwater mound suggested that the draindown from the tailings reached a maximum of approximately 180 gpm (34,500 cubic feet per day [ft3/day]) in 1984 as the Pit Lake initially began to fill, decreasing to 3.5 gpm (670 ft3/day) by 1992 (WWL, 1984). Recent estimates suggest that the seepage mound under the tailings will naturally disappear in 20 to 60 years, and that a portion of the seepage mound water will flow through the TDSS and OBSS aquifers to backfilled areas of the Pit Lake, eventually reaching the Pit Lake itself (WWL, 1989)."* Page 19

Based on this statement it is unclear how the flux value of 30 gpm for 20 years was selected. The quoted maximum, 180 gpm, is a total flux from the tailings, not necessarily that which was believed to have entered the pit lake, and by 1992, eight years later, this flux purportedly had decreased to 3.5 gpm. The value of 30 gpm for 20 years does not appear to be a conservative estimate of flux entering the pit lake, but rather a maximum flux, see Figure 1.

Surface runoff was set to 10% of the annual total inflow, and precipitation 27% of the total. The only outflow from the Pit Lake is through direct evaporation, and a value of 45 inches/year was used in the DSM, and which when combined with the surface area provides a flux.

A cursory examination of the Tetra Tech (2007b) report, *Long Term Pit Lake and Groundwater Hydrology at the Highland Mine Site*, seems to show inconsistencies in the flux values used in the DSM compared with those in the hydrology report. A detailed critical review of the hydrology and DSM should be carried out to determine if the steady-state values used in the DSM are consistent with the transient flux values of the hydrologic model.

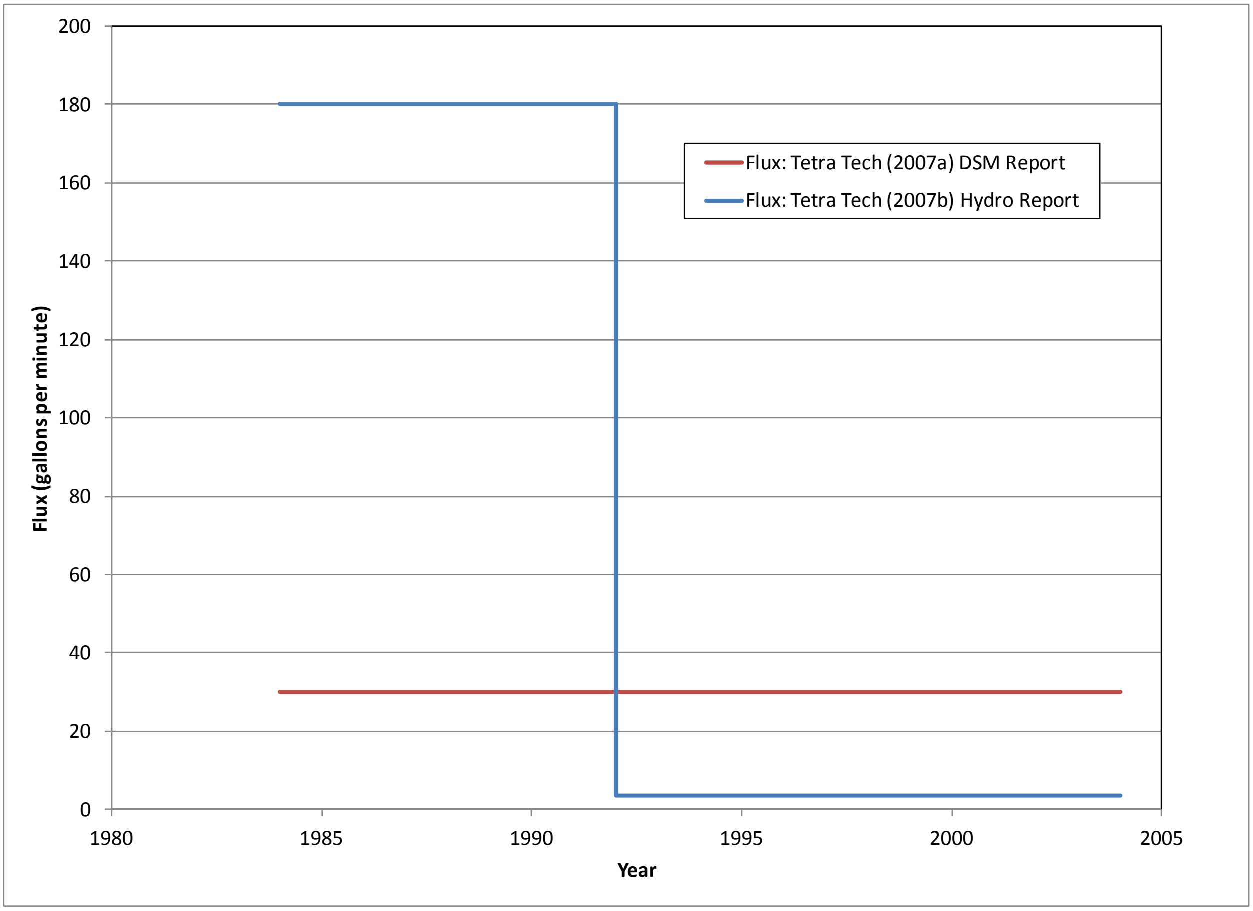

Figure 1. Tailings flux values from Tetra Tech (2007a) and Tetra Tech (2007b).

Verification of the Hydrologic Budget

In addition to assigning the flux values coming into the Pit Lake from the various sources it was also necessary to set the composition of each of the contributing sources to the lake. Based on its conservative geochemical behavior chloride was used to verify the hydrologic budget by matching the output from the DSM to the first 20 years of chloride data measured in the Pit Lake.

Equation 3-3 (page 22) is the mass balance equation for chloride in the Pit Lake. The equation states that the mass of chloride in the Pit Lake is:

$$\text{Mass } Cl_{PL} = C_{PL}\, V_{PL} = \sum Q_X\, C_X \qquad \text{[equation 3-3]}$$

Where: C_{PL} = chloride concentration in the Pit Lake; V_{PL} = volume of the Pit Lake
And Q_X = Inflow from the various sources; C_X = the Cl concentration form the various sources.

There is a problem with the dimensionality of this equation, the left side is (M/V) X (V) = M Where, M= mass, and V = volume. The right side of the equation is (V/T) X (M/V) = M/T, where V = volume, T = time, and M = mass.

While the Stella code probably works in the correct units, the fact that this equation is not correct decreases confidence in the report. In light of this, examination of figure 3-8 shows the measured concentrations of chloride (mg/L) versus the sampling date, however, equation 3-3 is not in terms of concentration, and more importantly the Pit Lake chloride concentration is effected by evaporation from the lake. There is no term to account for evaporative concentration in equation 3-3. Equation 3-3 is not correct for this process, rather equation 3-2 is the correct expression describing the relationship between concentration, lake volume, and time (see below).

$$\frac{d(C_K V)}{dt} = \sum_{i=1}^{n} C_{k,i} Q_i^{inflow} - \sum_{j=1}^{n} C_{k,j} Q_j^{outflow} \quad \text{[equation 3-2]}$$

Where $C_k V$ is the mass of constituent k dissolved in the Pit Lake water, and $C_{k,i(j)}$ are the concentrations of constituent k in the source of inflow i and source of outflow j (in units of mass per unit volume).

The chloride concentration value used in the DSM is that for the pure tailings solution, table 3.6 in the report, which was "from a sample collected at the mouth of the discharge tube leading to the Tailings Basin (EPRC 1982).", and was not a measured groundwater chloride concentration value. Tetra Tech used the chloride value of 217.5 mg/L to represent the tailings basin seepage, assuming that this was the chloride concentration of the groundwater entering the Pit Lake. The use of this composition is curious given that monitoring well 175, which is close to the Pit Lake and next to the tailings impoundment, and has been monitored since 1988, has recorded Cl values ranging from ~250 mg/L to ~450 mg/L. It appears the reason for using the 217.5 value was not because it represents water that Tetra Tech believed entered the Pit Lake, but rather it was a value that worked in the DSM to fit the measured Pit Lake chloride data. The source of the elevated chloride in well 175 is not discussed in Tetra Tech (2007a), and whether it was a byproduct of the mining/refining processes, such as barium chloride which have been used to precipitate radium.

Regardless of their reasons for using this composition, Tetra Tech was able to fit the DSM to the measured Pit Lake chloride data using a flux of 26 gpm and Cl = 217.5 mg/L. Based on the DSM to match the measured Pit Lake chloride data the hydrologic model was considered validated. If the flows from the various inputs were indeed accurate why then wasn't the DSM output compared to the measured water level of the Pit Lake? Had Tetra Tech matched both the chloride and Pit Lake's water levels, then confidence in the hydrologic model would be much greater than simply matching the lake's chloride values.

Chemical Balance

In a similar fashion used to verify the hydrologic budget, Tetra Tech compared output from the DSM to the measured values of the various constituents present in the Pit Lake. The first step in this process is assigning compositions to all of the inflows. In the case of the tailings seepage (see section 3.3.4) Tetra Tech chose well TDM 37 (Table 3.9) as the representative composition. However, rather than use the measured TDM 37 chloride value of 23.16 mg/L they used the pure tailings solution value of 217.5 mg/L. Likewise, instead of the measured TDM 37 sodium value of 92.24 mg/L, a value of 50% of the measured value was used instead. It is stated in the report that the 50% sodium value was used "in order to better reproduce the measured concentrations."

The measured TDM 37 composition (Table 3.9) has a charge-balance error (CBE) of 0.03%, and the altered composition used by Tetra Tech has a CBE equal to -34.5% due to the excess chloride and reduced sodium values. By changing the composition of the TDM 37 Tetra Tech created a fictional solution that does not meet the requirement of electroneutrality, the composition they used could never exist in the natural world. The reason they changed the composition was simply for the convenience of fitting the model to the measured data.

In the case of uranium Tetra Tech was able to match the measured Pit Lake uranium values by using two different combinations of uranium concentration in the tailing solution and OBSS, and two different volumes, i.e., flux values, of solution from the tailings solution and OBSS. This demonstrates the non-uniqueness of this problem, in other words there are an infinite number of combinations of flux values and uranium concentration that will fit the observed Pit Lake uranium concentration. In the baseline DSM Tetra Tech used the 9.1 mg/L at 26 gpm for 20 years for the tailings solution uranium input, and a value of 2.6 mg/L uranium from the OBSS – a higher value than any observed for the OBSS.

Likewise, with selenium, Tetra Tech took liberties with the concentrations in order to match the measured data. They used the measured value of the pure tailing solution, 0.126 mg/L to represent the tailings water, and used an initial pulse of 0.45 – 0.50 mg/L from the OBSS to achieve the concentrations in the Pit Lake. If there was no data to back up the use of this "pulse" of selenium from the OBSS, then how is this legitimate?

With regard to sulfate, which is a major contributor to TDS, Tetra Tech used the pure tailing solution value of 7580 mg/L (Table 3.6) in the DSM which overestimated that observed in the Pit Lake. They cite gypsum precipitation as the process that removes sulfate from solution to account for the discrepancy. However, their PHREEQC calculations show that gypsum doesn't become saturated until the year 2550.

Predicted Long-term Water Quality

This section summarizes the results of running the DSM out to 1000 years, and provides an explanation (Appendix A) of how the geochemical code, PHREEQC, was used to equilibrate the DSM output compositions with calcite, CO_2(g), and gypsum. From the information provided in Appendix A it was possible to scan the DSM output compositions into an Excel spreadsheet and run PHREEQC simulations for comparison with the results provided in Tetra Tech (2007a). Unfortunately, the thermodynamic database used by Tetra Tech was not documented, so it was not possible to exactly reproduce their results. There were 40 water compositions provided in Appendix A, and each composition was equilibrated with calcite at a saturation of 0.05, gypsum saturation at 0.0, and carbon dioxide partial pressure of $10^{-2.82}$ atm, note that the PHREEQC model was set up such that neither calcite nor gypsum were present in the system prior to the equilibration. The results showed, prior to calcite – carbon dioxide – gypsum equilibrium, that all 40 of the compositions were over saturated with respect to calcite, and that 12 (year 2400 to year 2984) were over saturated with respect to gypsum. Upon equilibration with calcite, CO_2(g) and gypsum only calcite was observed to precipitate, no gypsum precipitated from solution. The reason for this is the common ion effect, where calcite and gypsum share calcium in common, and as the calcium concentration decreased as calcite precipitated the saturation of gypsum was decreased below saturated concentrations. These results contradict those provided in the Tetra Tech (2007a) report where they observed gypsum precipitation.

While the geochemical model runs do not influence the constituents of concern, the fact that different results were obtained using the same inputs casts doubt on the modeling efforts presented in Tetra Tech (2007a).

Summary

A dynamic system model was created by Tetra Tech to simulate the long-term geochemical evolution of the Pit Lake at the Highland, WY uranium mine. This model is comprised of numerous inputs, both physical parameters (flux values and surfaces area) and chemical properties (multi component water compositions) all of the inputs carry a relatively high level of uncertainty. The model was calibrated to 20 years of measured chemistry of the Pit Lake, and then the calibrated model was used to simulate the Pit lake chemistry 1000 years into the future. The following are potential sources of uncertainty in the model:

- The geochemical conceptual model of the Pit Lake may be too over simplified to adequately represent actual conditions in the lake.
- The flux values from the various sources into the lake are highly uncertain, the perched aquifer contribution for example is based on a personal communication with no documentation.
- Verification/validation of the hydrologic budget is plagued with problems. The equation (3-3) used for mass balance of chloride is incorrect. The composition of the tailings influent used in the model was not realistic, and concentration values were changed to fit the measured lake concentration data. Water level elevation data for the Pit Lake have been collected since 1986, and this data could have been used in combination with the chloride data to validate the hydrologic model, however, it was not.
- To match the observed Pit Lake uranium, selenium, and sulfate data many liberties were taken with the model input concentrations to force the model into a match the measurements.
- The report points to gypsum precipitation as an important chemical process to remove sulfate from the Pit Lake, however, PHREEQC calculations show that calcite precipitation suppresses gypsum saturation and subsequent precipitation.

The DSM has many input parameters each with a high level uncertainty, and Tetra Tech adjusted the input parameters to match the measured 20 years of Pit Lake chloride and other chemical constituent data, for example using an unrealistic tailings solution composition. Having calibrated the model to the first twenty years of pit lake chloride data Tetra Tech then executed the model out 1000 years into the future as a predictive tool. However, the solution to this problem is non-unique, there are an infinite number of input parameter combinations that would result in matching the 20 years of measured data, none of them more or less correct than the next set of parameters. Model uncertainty could have been reduced, and greater confidence built into the model had Tetra Tech simultaneously matched the pit lake chloride data and the measured lake water level, however, this additional step was not performed and the model remains but one of an infinite number of possible solutions. Therefore, predicting long-term trends with this model will produce results which are essentially meaningless. Additionally, the report lacks transparency, it is often unclear what manipulations were performed, and why they were performed to the input parameters, which is bad protocol in a modeling study.

While this review does not speculate on the observed chemistry of the Pit Lake, the authors of the Tetra Tech (2007a) report do present a plausible explanation:

"One geochemically plausible explanation for the discrepancy between observed and modeled results is that groundwater entering the Pit Lake from the OBSS contacts oxidized portions of the roll-front deposits adjacent to the pit walls, enriching the influent groundwater with radionuclides and selenium. While historical well data from the OBSS in the vicinity of the Site (EPRC, 1983) suggest that the activities of uranium and radium were quite high in the excavated area, none of the current monitoring wells penetrate the un-mined ore body that occurs in close proximity to the Pit Lake to provide an estimate of the potential enrichment." From Section 3.3.6.

As the above quote from Tetra Tech (2007a) clearly states, water entering the Pit Lake could have reacted with the "oxidized portions of the roll-front deposits adjacent to the pit walls, enriching the influent water with radionuclides and selenium." Tetra Tech, in their own words, posed a perfectly legitimate alternative for the source of the Pit Lake contamination other than the tailings solution. Indeed, the DSM was a simple exercise in curve fitting a model by manipulating the key input parameters to unrealistic values until the model matched the measured data, as such does not prove that the source of the contamination observed in the Highland Pit Lake originated from the mine tailings. The only conclusion that can be drawn from the DSM is that it is possible, by manipulating the model input parameters to unrealistic values, to fit the observed chemical trends in the Pit Lake.

References

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Tetra Tech, 2007b. "*Long Term Pit Lake and Groundwater Hydrology at the Highland Mine Site*". Prepared by Tetra Tech Fort Collins, CO 80525. Project number 180549.