TECHNICAL COMMENT

Pinyon Plain Mine Aquifer Protection Permit No. P-100333
Proposed Minor Amendment: Revision of AQL for Arsenic at MW-02 (POC #2)
Submitted to: Arizona Department of Environmental Quality (ADEQ)
Commenter: Dr. Bradley K. Esser

Introduction and Key Points

I am submitting this technical comment to identify serious deficiencies in the investigation and technical demonstration submitted by Energy Fuels Resources (USA) Inc. (EFRI) in support of a proposed minor amendment to Aquifer Protection Permit No. P-100333 for the Pinyon Plain Mine, Coconino County, Arizona. EFRI is requesting that the Aquifer Quality Limit (AQL) for dissolved arsenic at monitoring well MW-02 (Point of Compliance #2, Perched Coconino Sandstone Aquifer) be raised from 50 µg/L to 55 µg/L. This comment concerns the adequacy of the investigation and the technical merit of EFRI's "natural phenomenon" justification for the proposed AQL revision. The mine site is approximately 9 miles south of the South Rim of the Grand Canyon, and the springs and water sources of the Grand Canyon, including sources of drinking water for the Havasupai Tribe, depend on the quality of the perched and regional aquifer system that this mine penetrates.

Qualifications of the Commenter

I am a retired scientist, group leader and project leader from Lawrence Livermore National Laboratory (LLNL), where I worked for more than 28 years. I hold a Ph.D. in Geology and Geophysics from Yale University (1991) and dual Bachelor of Science degrees from the University of Arizona in Geosciences and in Ecology and Evolutionary Biology (1980, 1983). My complete curriculum vitae is appended to this comment.

My research career has been devoted primarily to environmental isotope geochemistry and groundwater science, with particular emphasis on:

- **Groundwater age-dating** using environmental tracers, including tritium/helium-3 ($^3H/^3He$), radiogenic ^4He, cosmogenic ^{35}S, chlorofluorocarbons (CFCs), sulfur hexafluoride (SF_6), radiocarbon (^{14}C), and dissolved atmospheric volatile organic compounds (VOCs). I developed and led the application of LLNL's groundwater age dating capability across California and the western United States.
- **Isotope geochemistry** applied to contaminant source attribution and fate-and-transport in groundwater, including uranium, nitrate, arsenic, chromium, mercury, and radionuclides. I have published on the use of isotopic tools to distinguish natural from anthropogenic sources of groundwater contamination.

- **The California Groundwater Ambient Monitoring and Assessment (GAMA) Program:** I was Technical Lead for the GAMA Special Studies Program from 2007 to 2018, delivering more than 40 technical reports on groundwater age, quality, and contamination across California. The GAMA program applies a suite of passive environmental tracer methods (VOCs, CFCs, SF_6, tritium, noble gases, stable isotopes) that I recommend in this comment for characterization of MW-02. I was also Technical Lead for age-dating public drinking supply wells in California in collaboration with the USGS under the main GAMA program.
- **Uranium contamination on tribal lands**: I led LLNL's technical collaboration with the Navajo Nation to characterize and remediate uranium contamination from legacy uranium mining and milling. I also led a small public interest project characterizing deep drinking water supply wells, shallow livestock wells and springs in the Ute Mountain Ute White Mesa community. These waters sources are directly downgradient from the .EFRI White Mesa Mill. We were able to show that the deep municipal wells produced very old water and were safe and that the shallow wells and springs produced young water and were potentially vulnerable.
- **Oil and gas groundwater monitoring**: I chaired the multi-institution State of California Water Board committee that developed the model criteria for groundwater sampling, testing, and monitoring of oil and gas well stimulation operations (SB4, 2015), leading a team of national laboratory scientists and university researchers.

I have authored or co-authored 112 peer-reviewed publications (h-index 34, Google Scholar; 3,486 citations) and more than 50 technical reports. Selected publications directly relevant to the subject matter of this comment include work on groundwater age distributions and contamination history, multi-tracer groundwater age methods, VOC tracers of modern recharge, and contaminant attribution using isotope methods. I received the Hitchon Award from the International Association of Geochemistry and Cosmochemistry (IAGC) in 2007.

I have no financial interest in the outcome of this permit proceeding. I am submitting this comment in my capacity as a private citizen and scientist with expertise directly relevant to the technical questions at issue.

Key Points

My principal concerns regarding EFRI's investigation and the proposed AQL revision are as follows:

- The arsenic concentrations recently observed in MW-02 (30 to 43 µg/L, persistently since mid-2024) are 20 to 30 times the regional natural background median (~1.5 µg/L; Tillman et al., 2023) and 3 to 4 times the upper statistical fence of the regional background distribution. They are not consistent with natural background in the perched Coconino Sandstone aquifer.
- EFRI's proposed explanation that the cone of depression caused by mine shaft seepage is drawing naturally elevated-arsenic off-site groundwater toward MW-02 is an internally

consistent hypothesis but remains entirely untested. The single off-site USGS well cited by EFRI shows arsenic of only 0.51–0.84 µg/L, which directly contradicts rather than supports the hypothesis.

- The co-trending of eight constituents at MW-02 (arsenic, antimony, barium, fluoride, gross alpha, uranium, nickel, thallium) is geochemically inconsistent with import of distal regional groundwater. This element suite is the signature of breccia pipe ore mineralization; it is not the signature of the regional perched Coconino aquifer.
- Redox indicator data (dissolved oxygen, oxidation-reduction potential) are already being collected under the permit compliance program and are present in the ADEQ QWQD database. They show oxidizing conditions in MW-02 (DO ~5.5 mg/L, ORP ~+175 mV) that are mechanistically consistent with sulfide mineral oxidation, which is the exact reaction pathway demonstrated experimentally by Tillman et al. (2023) to produce arsenic concentrations of the magnitude observed at MW-02. EFRI's investigation does not present or interpret these data.
- The ancient radiocarbon age of the perched Coconino groundwater (~10,000 years) does not rule out fast-flow pathway connectivity. In carbonate-dominated aquifer systems, radiocarbon ages are systematically inflated by exchange with "dead" geogenic carbon. More importantly, in dual-porosity or conduit-flow systems, even a small fast-flow component can transport contaminants at disproportionately high concentrations relative to the young-water volume fraction. The question of fast-flow connectivity has not been tested with appropriate passive environmental tracers.
- The January 2025 arsenic measurement of 206 µg/L at MW-02 was dismissed as a statistical outlier. This value falls within the lower range of measured sump arsenic concentrations and could represent a transient pulse from a fast-flow pathway. Dismissal without investigation of that possibility is not scientifically or regulatorily adequate.
- No water isotope data (δ^2H, δ^{18}O) have been collected at MW-02 during the period of rising arsenic concentrations, despite these being inexpensive and informative tracers for source attribution. There is no scientific justification for omitting them.
- The proposed AQL revision would set a regulatory precedent in which a facility can revise upward the compliance threshold based on a statistical representation of its own rising contamination data, effectively allowing the threshold to chase concentrations. The correct regulatory response to a documented increasing trend is intensified investigation, not threshold normalization.
- A USGS investigation formally identifying the fast-flow pathway question as unresolved is currently underway (USGS, 2025), with results expected in 2026–2027. ADEQ should not approve a permanent AQL revision premised on an inadequately characterized hydrogeological conceptual model that the USGS has formally identified as requiring investigation.

Regulatory Background and Triggering Events

The APP permit requires investigation when monitoring results exceed the Alert Level (AL). The sequence at MW-02 is as follows:

- January 22, 2025: arsenic at MW-02 measured at 0.206 mg/L (206 µg/L), exceeding both the AL (0.04 mg/L) and the AQL (0.05 mg/L). EFRI rejected this result using the Dixon extreme outlier criterion (INTERA, 2025).
- February 7, 2025 (verification sample): arsenic confirmed at 0.0434 mg/L (43.4 µg/L), exceeding the AL. Five-day notification submitted to ADEQ February 24, 2025.
- Accelerated monthly monitoring commenced March 2025. Continued AL exceedances in May, July, and September 2025 triggered the second-investigation requirement under Section 2.6.2.3.2(7).
- EFRI submitted a combined second investigation report and minor amendment request (November 19, 2025; Revision 1 January 12, 2026), proposing to raise the AQL from 50 to 55 µg/L based on a statistical analysis by INTERA (2025) and a “natural phenomenon” technical demonstration.

Section 2.6.2.3.2(3) of the permit allows an operator to demonstrate, in lieu of implementing the contingency plan, that an exceedance is due to a “natural phenomenon” unrelated to facility discharge. That demonstration must receive written ADEQ approval. EFRI has elected this pathway. As detailed below, the demonstration does not meet the required evidentiary standard.

EFRI's Cone-of-Depression Hypothesis

EFRI attributes the MW-02 arsenic exceedance to the following mechanism (EFRI/HGC, 2025; HGC, 2024):

- Continuous seepage of perched Coconino groundwater into the mine shaft since late 2016 has caused the shaft to act as an over-pumped well, generating an expanding cone of depression in the C-aquifer.
- The cone of depression has grown to encompass all three on-site Coconino monitoring wells (MW-01, MW-02, MW-03), so that all perched groundwater in the monitoring network now flows toward the shaft.
- MW-02 is therefore no longer sampling near-mine water but is progressively capturing distal off-site C-aquifer water with naturally elevated arsenic, importing regional background chemistry from greater distances.
- The concurrent significant trends in eight co-monitored constituents (antimony, arsenic, barium, fluoride, gross alpha, uranium, nickel, and thallium) are attributed to the same non-steady-state hydrogeologic effect (HGC, 2024).

This hypothesis deserves fair consideration. A cone of depression driven by shaft seepage is physically plausible and consistent with observed water-level drawdown. However, as a “natural phenomenon” justification, it must be supported by evidence demonstrating that (a) off-site C-aquifer water contains arsenic at concentrations consistent with MW-02, and (b) alternative explanations, particularly in-situ mobilization driven by mine operations, are excluded. Neither condition is met.

MW-02 Arsenic Concentrations Are Inconsistent with Regional Natural Background

Regional background: Tillman et al. (2023)

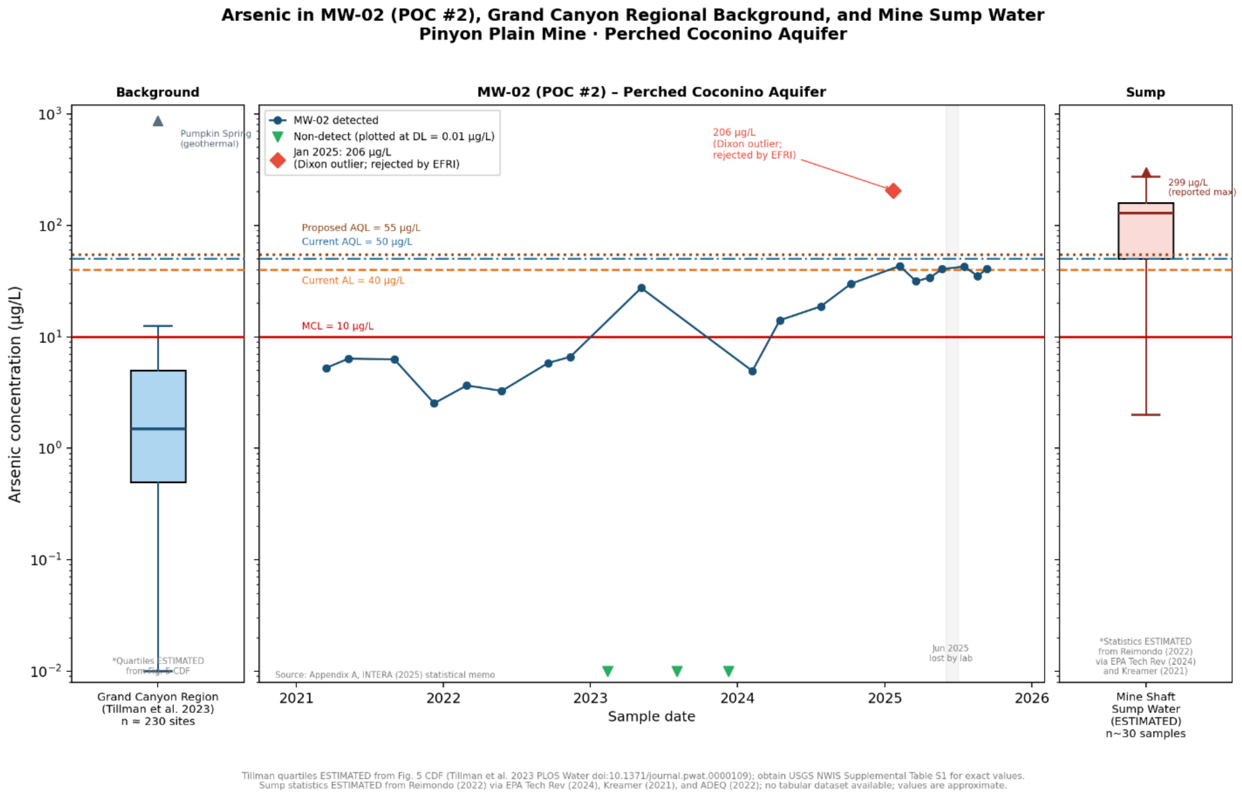
Tillman et al. (2023) is a comprehensive regional characterization. The study compiled 652 arsenic measurements from 230 sites (springs and wells) across the Grand Canyon region, collected between 1977 and 2022. Key statistics:

- Regional median arsenic: approximately 1.5 µg/L (estimated from Fig. 5 cumulative frequency distribution; exact values in USGS NWIS Supplemental Table S1).
- Upper fence of the regional distribution (Q75 + 1.5×IQR): approximately 12.5 µg/L.
- Fraction of sites below the MCL (10 µg/L): 88% (202 of 230 sites).
- Only a handful of sites exceed 40 µg/L; virtually all are geothermally influenced (Pumpkin Spring: 875 µg/L) or associated with travertine spring systems with deep mantle-fluid input. No ordinary perched Coconino Sandstone groundwater site approaches 40 µg/L.

MW-02 time series: 2021–2025

- 2021–2022: arsenic ranged from 2.54 to 6.63 µg/L, which is below the MCL and within the upper regional background range.
- Early 2023: two consecutive non-detects (below MDL 4.4 µg/L) followed by an anomalous detection of 27.5 µg/L in May 2023.
- 2024: concentrations rose steadily from 4.95 µg/L (February) to 30.0 µg/L (October), crossing the MCL by approximately April 2024.
- 2025 (accelerated monthly monitoring): arsenic persistently 31–43 µg/L, representing 20–30× the regional median and 3–4× the upper statistical fence of the regional background distribution.
- January 2025 primary sample: 206 µg/L, which was subsequently rejected by EFRI as a Dixon extreme outlier; this is addressed separately below.

Figures 1 and 2 show the MW-02 arsenic time series against regional background statistics and against regulatory thresholds.


Arsenic in MW-02 (POC #2), Grand Canyon Regional Background, and Mine Sump Water
Pinyon Plain Mine · Perched Coconino Aquifer

Background

MW-02 (POC #2) – Perched Coconino Aquifer

Sump

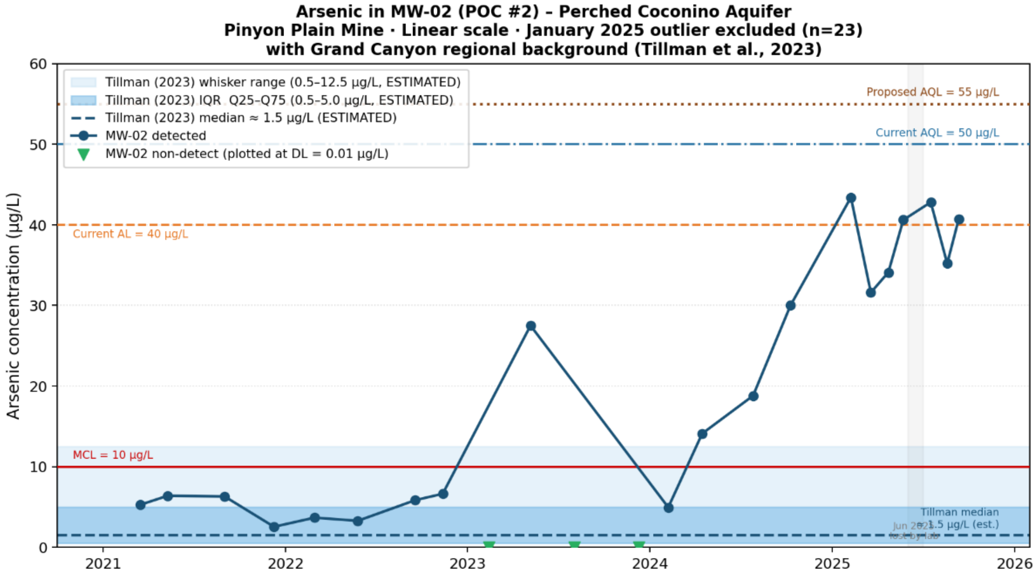
Source: Appendix A, INTERA (2025) statistical memo

*Quartiles ESTIMATED from Fig. 5 CDF

*Statistics ESTIMATED from Reimondo (2022) via EPA Tech Rev (2024) and Kreamer (2021)

Tillman quartiles ESTIMATED from Fig. 5 CDF (Tillman et al. 2023 PLOS Water doi:10.1371/journal.pwat.0000109); obtain USGS NWIS Supplemental Table S1 for exact values.
Sump statistics ESTIMATED from Reimondo (2022) via EPA Tech Rev (2024), Kreamer (2021), and ADEQ (2022); no tabular dataset available; values are approximate.

Figure 1. Arsenic time series at MW-02 (POC #2), Center: Arsenic time series on log scale from 01/2021 to 09/205 in MW-02, a monitor well in the perched Coconino Sandstone Aquifer, Current AL and AQL and proposed AQ shown. Non-detects plotted at MDL. The 2025/01 value was flagged as an extreme outlier and is shown for reference only. Left Regional background distribution (Tillman et al.,2023) shown as a box plot. Right: Sump water concentrations shown as box plot.



MW-02 source: Appendix A, INTERA (2025) statistical memo. Non-detects plotted at reported value (0.01 µg/L = DL). Tillman bands: Q25/median/Q75/whiskers ESTIMATED from Fig. 5 CDF in Tillman et al. (2023) PLOS Water doi:10.1371/journal.pwat.0000109; obtain USGS NWIS Supplemental Table S1 for exact quartiles.

Figure 2. Arsenic time series at MW-02, linear scale, with current AQL (50 µg/L, blue dashed), proposed revised AQL (55 µg/L, orange dotted), Alert Level (40 µg/L), and MCL (10 µg/L) shown. The proposed 10% AQL increase provides negligible regulatory buffer against continuing arsenic increases.

Mine shaft sump water as geochemical comparator

Sump water (dewatered perched Coconino groundwater that has been in direct contact with the mineralized breccia pipe) provides a direct geochemical comparator. ADEQ QWQD permit compliance data show:

Period	Sump As (µg/L)	Context
Q1 2024	446	~15× current MW-02 max
Q2 2024	14,400	Ore contact event
Q3 2024	8,490	
Q4 2024	4,130	
Q1 2025	1,290	
Q2 2025	6,890	
Q4 2025	214,000	Extraordinary escalation; consistent with major ore-body breakthrough

Table 1. Sump arsenic concentrations, Pinyon Plain Mine, Q1 2024–Q4 2025. Source: ADEQ QWQD permit compliance database.

These values demonstrate that the perched Coconino water in direct contact with breccia pipe mineralization is capable of producing arsenic concentrations five orders of magnitude above the regional background median. Dilution of even a small volume of this mine-proximal water into the surrounding aquifer would be sufficient to generate the concentrations observed at MW-02. EFRI's investigation does not quantitatively evaluate this mixing scenario.

Multi-Element Co-Trending Points to a Mineralized Source, Not Distal Background Import

The co-trending element suite is geochemically diagnostic

HGC (2024) identified statistically significant trends in eight constituents at the on-site Coconino wells: antimony, arsenic, barium, fluoride, gross alpha, uranium, nickel, and thallium. EFRI attributes all eight to the cone-of-depression mechanism, arguing that the expanding capture zone is importing off-site C-aquifer water enriched in all of these elements simultaneously. This explanation is geochemically implausible for the following reasons:

- Breccia pipe uranium deposits in the Grand Canyon region are specifically characterized by enrichment in uranium, arsenic, antimony, barium, nickel, and thallium — exactly the suite showing significant trends (Tillman et al., 2023; Wenrich, 1985). This elemental combination is the fingerprint of breccia pipe ore mineralogy, not of background Coconino Sandstone groundwater.
- Thallium is the most diagnostic indicator. It is detected above reporting limits in all three on-site Coconino monitoring wells (MW-01: 0.000681–0.001030 mg/L; MW-02: 0.000532–0.000662 mg/L; MW-03: 0.000216–0.000661 mg/L) but is consistently below reporting limits in the off-site R-aquifer well (RW-01: < 0.000130 mg/L). In low-temperature aquifer systems, thallium in the concentration range observed here is virtually exclusively derived from sulfide mineral weathering. Its spatial association with the mine-proximate Coconino wells and its absence from the regional R-aquifer is inconsistent with regional background import.
- Antimony shows a similar pattern: 0.00221–0.00304 mg/L at MW-01 but below reporting limits at MW-02, MW-03, and RW-01 in most periods. This is a pattern that is consistent with spatially heterogeneous near-mine mineralization influence rather than a homogeneous off-site background source.
- Sulfate at MW-03 (776 mg/L) is dramatically higher than at MW-02 (125 mg/L) or MW-01 (226 mg/L). The R-aquifer well (RW-01) has sulfate of only 19.3 mg/L. This large within-site variation in sulfate, co-occurring with high nickel at MW-03 (0.199–0.265 mg/L), is consistent with variable degrees of sulfide mineral oxidation at different well locations (a near-mine, in-situ process) and not with the import of a uniform off-site background signature.

Redox data are in the compliance record but absent from EFRI's analysis

Field measurements of dissolved oxygen (DO) and oxidation-reduction potential (ORP) are included in the permit compliance monitoring program and are present in the ADEQ QWQD database and the 2024 Annual Report. These data directly address the operative arsenic mobilization mechanism. EFRI's investigation does not present or interpret them. Key observations:

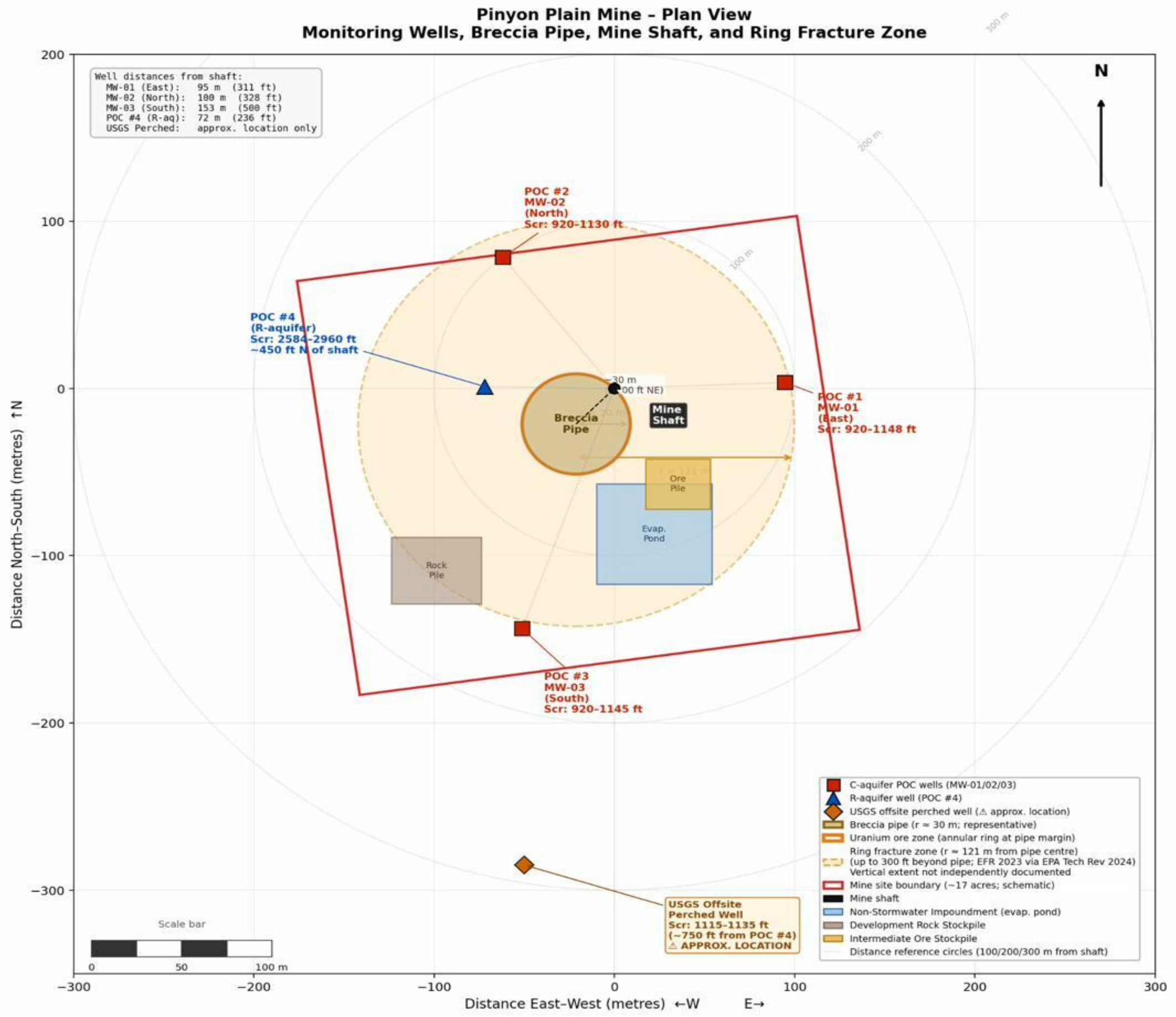
- MW-02 (Q4 2024): DO = 5.53 mg/L; ORP = +175 mV. These values indicate moderately oxidizing conditions, conditions under which sulfide mineral oxidation (arsenopyrite, gersdorffite) is the thermodynamically favored arsenic mobilization mechanism. They are definitively inconsistent with reductive dissolution of iron oxyhydroxides, which requires strongly anoxic conditions (DO < 0.5 mg/L, ORP < 0 mV typically).
- MW-03 (Q4 2024): DO = 0.16 mg/L; ORP = +30 mV. Near-anoxic, with very high sulfate (776 mg/L) and the highest nickel (0.230 mg/L) of any well. This well is geochemically distinct from MW-02 and is consistent with a flow path experiencing more extensive historical sulfide oxidation with consequent oxygen depletion.
- MW-01 (Q4 2024): DO = 2.51 mg/L; ORP = +165 mV. Suboxic-to-oxic; intermediate sulfate (226 mg/L).
- RW-01 R-aquifer (Q4 2024): DO = 4.05 mg/L; ORP = +193 mV. Clearly oxic, but sulfate of only 19.3 mg/L, which is less than 3% of the sulfate at MW-03 and less than 16% at MW-02. This is the most direct chemical evidence that the R-aquifer is not in geochemical communication with a sulfide-oxidizing environment, whereas all three Coconino wells are.
- Q2 2025 data quality anomaly: MW-01 reported DO = 36.1 mg/L and MW-02 reported DO = 41.6 mg/L. Both values are physically impossible for groundwater (atmospheric saturation at this elevation and temperature is approximately 8–9 mg/L). These values indicate instrument malfunction or calibration failure and represent a data quality issue requiring investigation and correction.

The oxidizing conditions in MW-02 are exactly the geochemical environment in which Tillman et al. (2023) laboratory leaching experiments produced arsenic concentrations up to 224 µg/L from Coconino Sandstone in contact with breccia pipe ore material. EFRI's failure to present and interpret the available redox data in the technical demonstration is a significant evidentiary deficiency.

Alternative Mechanisms Not Evaluated by EFRI

In-situ mobilization driven by cone-of-depression-induced flow path changes

The breccia pipe at Pinyon Plain Mine is surrounded by a ring fracture zone extending up to approximately 300 feet (91 m) beyond the pipe boundary (EFR, 2023; as cited in EPA Tech Review, 2024, p. 11). The ore mineralization column spans from approximately 650 to more than 2,100 feet depth, a vertical extent that encompasses the perched Coconino Sandstone aquifer at which MW-02 is screened. Figure 3 shows the plan view of the mine site with the ring fracture zone extent.



Coordinates from ADEQ Permit P-100333 (2022, 2024). Pipe centre inferred from shaft offset (~100 ft NE of pipe; permit text).
Breccia pipe diameter representative (50-75 m typical; exact Pinyon Plain diameter not in available records). Mine site boundary schematic (17 acres; no polygon in record).
Ring fracture zone lateral extent (up to 300 ft / ~91 m beyond pipe): operator-reported per EFR (2023), cited in EPA Tech Rev (2024, p.11); stated as upper bound only.
Ring fracture zone vertical extent: NOT independently documented in available records; co-extensive with pipe column inferred from geology but not confirmed.
USGS offsite perched well location approximate (no precise coordinates in available records). All schematic features labelled accordingly.

Figure 3. Plan view of Pinyon Plain Mine showing monitoring well locations, breccia pipe boundary, uranium ore zone (annular ring at pipe margin), and ring fracture zone extent (dashed tan circle, up to 300 ft / 91 m beyond pipe center; operator-reported, stated as upper bound). All three Coconino monitoring wells (MW-01, MW-02, MW-03) plot within or near the ring fracture zone. RW-01 (R-aquifer) is shown for reference. Source: Constructed from ADEQ Permit P-100333 (2022, 2024) and EPA Tech Review (2024).

As the cone of depression expands, driven by the very operational effect that EFRI invokes, hydraulic gradients shift and previously quiescent portions of the ring fracture zone and surrounding mineralized rock are brought into the active flow field. Water that was not previously flowing through mineralized fractures is now directed through them by the mine-induced hydraulic sink. This constitutes an in-situ mechanism for arsenic and trace element mobilization into the monitoring wells, a mechanism that is operationally induced even if the arsenic source is geological. Critically, all three Coconino monitoring wells are positioned within the approximate outer extent of the ring fracture zone, as shown in Figure 3. EFRI's investigation does not evaluate or exclude this mechanism.

Fast-flow pathways in a karst and fractured-rock setting

EFRI's conceptual model treats the perched C-aquifer as a porous-medium system in which flow directions can be reliably inferred from water-level gradients among three closely spaced wells. This is an inappropriate assumption in a geological setting with documented fracture and karst conduit flow, unless explicitly validated. Multiple lines of evidence indicate this validation has not been performed:

- The EPA Technical Review (2024) specifically notes that the conceptual flow model for the Grand Canyon South Rim involves a multi-permeability system with fast-traveled groundwater along fault and karst conduit pathways.
- Fluorescent dye tracer studies on the Kaibab Plateau (Jones et al., 2017; Beisner et al., 2023b) have demonstrated groundwater transport of thousands of feet vertically and tens of miles horizontally within weeks to months via structurally controlled conduit pathways at velocities orders of magnitude faster than porous-medium Darcy flow would predict.
- Beisner et al. (2023b) demonstrated that Grand Canyon Village wastewater tracers appeared at Monument Spring via a preferential pathway that violates the surface topographic watershed divide and constitues direct evidence that hydraulic connectivity in this system does not follow surface drainage patterns or simple hydraulic gradient directions.
- Kreamer (2021) specifically warned in public comment that standard porous-medium monitoring well placement is inadequate in karst settings, where conduit flow can bypass the capture zone of a monitoring well entirely.
- The USGS proposed investigation (USGS, 2025) lists as its primary Task 1 objective the characterization of fast-flow pathways in the vicinity of the Pinyon Plain Mine using CSAMT geophysical surveys. The formal identification of this as an unresolved scientific question by an independent federal agency constitutes acknowledgment that the hydrogeological framework assumed by EFRI is not established.

Operationally induced mobilization of naturally occurring constituents

EFRI argues that because all mine water is managed in a lined impoundment, there is no "discharge" that could have caused the MW-02 exceedance. This framing is too narrow. The mine shaft penetrates the perched Coconino aquifer and the surrounding mineralized geological column, fundamentally altering the hydraulic and geochemical environment in ways that mobilize naturally occurring arsenic from the ring fracture zone and adjacent mineralized rock into the active flow field. This is not a discharge from a surface facility. But it is an operationally induced impact on groundwater quality that falls squarely within the protective intent of the Aquifer Protection Permit program. The regulatory framework should not be interpreted to exempt the most geochemically consequential operational impacts simply because they are mediated through subsurface hydrogeological changes rather than through a direct surface-to-groundwater pathway.

Critical Evidentiary Gaps in EFRI's Technical Demonstration

The following evidentiary deficiencies, individually and collectively, prevent EFRI's technical demonstration from satisfying the standard required by APP Section 2.6.2.3.2(3):

Redox data are available but uninterpreted

Field DO, ORP, pH, and sulfate data are present in the ADEQ QWQD database and 2024 Annual Report but are not presented or discussed in EFRI's technical demonstration. These data distinguish between competing arsenic mobilization mechanisms. Additionally, arsenic speciation (As(III)/As(V)) has not been analyzed — a low-cost, high-diagnostic-value measurement that would confirm whether oxidative or reductive mobilization is dominant.

Arsenic geochemistry is not considered

A number of mechanisms potentially provide pathways for in-situ mobilization of arsenic, including

- **Oxidative sulfide dissolution:** Arsenic in breccia pipe deposits is hosted primarily in sulfide minerals (arsenopyrite, As-rich pyrite, orpiment, realgar). As Eh rises with oxygenation of the shaft, these minerals dissolve, releasing As into the aqueous phase as arsenate ($HAsO_4^{2-}$ or $H_2AsO_4^-$). The Pinyon Plain deposit has an arsenic concentration of 105,000 mg/kg (orders of magnitude above background) providing enormous source-term potential.
- **Desorption from oxyhydroxides**: ADEQ has argued that the "*abundance of iron oxide rich sediments throughout the stratigraphic column*" will attenuate dissolved metals by sorption. Sorption, however, is a conditional and reversible process (Kreamer, 2021; Tillman et al. 2023; and EPA Tech Review 2024) If pH rises above ~8.5 (common in carbonate-buffered Grand Canyon groundwaters) or if reducing conditions develop in stagnant zones, arsenate desorbs from ferrihydrite/goethite surfaces. The EPA Tech Review specifically recommends PHREEQC modeling to evaluate iron mobilization during leaching as a function of time, and notes this has not been done.,
- **pH-driven speciation shifts**, Carbonate dissolution in the Kaibab/Toroweap/Redwall system can buffer pH upward. At pH > 8, arsenate speciation shifts toward the more mobile divalent $HAsO_4^{2-}$, and sorption to iron oxides decreases. Mixing of oxidized

shallow water with deeper, potentially more reducing R-M aquifer water could create geochemical conditions where neither end-member behavior is predictive.

- **Hydraulic mobilization**: The expanding cone of depression (>82 million gallons pumped) induces convergent flow from all directions toward the shaft. As water table gradients steepen and previously static pore waters are mobilized, geochemical disequilibrium occurs, i.e. pore waters with different redox histories mix, and previously sorbed contaminants can be kinetically released. This is the mechanism EFRI implicitly acknowledges in their cone-of-depression argument, but they frame it as dilution rather than mobilization.

These scenarios are not rigorously considered. Redox and pH data are available to enable geochemical modeling of arsenic mineral and iron oxide stability and sorption/desorption of arsenic from iron oxide surfaces.

No spatial characterization of upgradient water quality at the scale of the cone of depression

EFRI's hypothesis predicts that off-site C-aquifer water at the expanding cone-of-depression margin contains arsenic at 30–43 µg/L. The single off-site USGS observation well (arsenic 0.51–0.84 µg/L) contradicts this prediction. No data are presented from multiple azimuths around the mine at distances commensurate with the claimed cone-of-depression radius.

No stable isotope data for water source attribution

No stable isotope data (δ^2H, δ¸O) have been collected at MW-02 or other on-site wells during the period of rising arsenic concentrations. The isotopic composition of water extremely inexpensive and very informative tracers for water source attribution in the Grand Canyon system, where the isotopic compositions of perched C-aquifer and deeper R-aquifer waters have been characterized (Solder and Beisner, 2020). If the cone of depression is progressively importing isotopically distinct distal C-aquifer water, a systematic isotopic shift should be detectable. The absence of these data from EFRI's investigation is scientifically indefensible given their cost and availability.

No evaluation of fast-flow connectivity using appropriate tracers

EFRI relies on the radiocarbon age of the perched C-aquifer to argue against fast-flow connectivity. This argument is flawed. Radiocarbon ages in carbonate-hosted aquifers are systematically inflated by isotopic exchange with geogenic "dead" carbon from the host rock (Szabo et al., 1986). And significantly in dual-porosity systems, a minor fast-flow component is not detectable in bulk radiocarbon measurements. Appropriate tracers for detecting modern atmospheric communication (CFCs, SF_6, tritium, dissolved industrial VOCs) have not been applied at MW-02.

The January 2025 outlier (206 µg/L) was not adequately investigated

The January 2025 arsenic measurement of 206 µg/L was rejected on the basis of disagreement with a verification sample collected 16 days later (43.4 µg/L). In a system with documented high temporal variability and evidence of episodic processes in the sump (Q4 2025 sump As = 214 mg/L; Q2 2024 sump As = 14.4 mg/L), a 16-day interval is not trivially representative. The value 206 µg/L lies within the lower end of the measured sump arsenic distribution and is consistent with a transient pulse from a fast-flow preferential pathway that was subsequently diluted. Rejection without investigation of this possibility is not adequate.

Uranium activity ratios are available but not reported for monitoring wells

The activity ratio of ^{234}U to ^{238}U ("AR") in groundwater is a well-established tracer of uranium source and water-rock interaction history (which are directly relevant to identifying the mechanism for arsenic enrichment in MW-02). The AR tracer has been applied in exactly this geological setting by the USGS. Beisner et al. (2023a) explicitly used ^{234}U/^{238}U ARs to discriminate ore-proximal from natural background groundwater at the Orphan Mine (a geologically analogous breccia pipe on the South Rim of the Grand Canyon). Liebe (2003) similarly used the AR to characterize springs and surface waters across Grand Canyon National Park, and the USGS investigation proposal for Pinyon Plain (USGS, 2025) explicitly identifies isotopic characterization, including uranium isotope ratios, as a priority task. The evolution of uranium activity radios in Pinyon Plain Mine sump water demonstrates the utility of the measurement. I have attached Appendix A: Uranium Activity Ratios with further discussion of the utility of the measurement and its potential application to Pinyon Plain Mine monitoring.

Uranium isotope activities are determined individually by alpha spectrometry and are reported individually in the sump compliance annual reports. For the monitoring wells, however, only the aggregated sum "Uranium Isotopes (pCi/L)" is reported in the QWQD permit compliance database. Individual isotope activities for MW-01, MW-02, MW-03, and RW-01 are not accessible in any publicly reported compliance record. These individual isotope activities however should be available in the analytical reports.

Recommendations

ADEQ should decline to approve the proposed AQL revision until an expanded investigation that affirmatively demonstrates that the arsenic exceedance in MW-02 is attributable to natural phenomenon rather than to operationally induced mobilization. This will require a more comprehensive investigation of existing data and additional analyses, characterization and modeling specific recommended requirements are listed below. These are ordered roughly by cost and immediacy; items 1–9 can be initiated within the current accelerated monitoring schedule with minimal additional cost.

A. Interpret existing data (no additional field work required)

1. **Present and interpret all existing redox indicator data** from the ADEQ permit compliance monitoring record, including dissolved oxygen, ORP, pH, and sulfate for MW-01, MW-02, MW-03, and RW-01. These data are already collected and present in the QWQD database; they have simply not been analyzed in the context of the investigation. The investigation report must address what these data imply about the operative arsenic mobilization mechanism.
2. **Investigate and resolve the physically impossible dissolved oxygen values** reported for MW-01 (36.1 mg/L) and MW-02 (41.6 mg/L) in Q2 2025 (atmospheric saturation at this elevation is ~8–9 mg/L). These values indicate an instrument calibration or protocol failure and constitute a data quality issue requiring formal documentation and correction.
3. **Present a quantitative comparison of the major ion chemistry and trace element signatures** of MW-02 with sump water and with the off-site USGS C-aquifer well, using Piper diagram analysis. This comparison is a basic geochemical discriminant that EFRI's investigation omits.
4. **Present and interpret ^{234}U and ^{238}U activities in monitoring well sample** as required by the permit. These data are available in the analytical data reports and should be compiled into compliance data tables. Time-series ^{234}U/^{238}U activity ratios should be compared to literature data for Grand Canyon water (both impacted and not impacted by breccia pipe mineralization) and to sump water activity ratios
5. **Geochemically model arsenic and uranium behavior** under current and projected subsurface conditions**.** EPA's 2024 Technical Review explicitly recommended this, and it was not required in the original permit. The cone of depression and oxygenation from dewatering represent ongoing perturbations to the geochemical system. Iron oxyhydroxide sorption capacity (ADEQ's stated attenuation mechanism) is demonstrably pH- and Eh-dependent. Without speciation modeling, ADEQ cannot evaluate whether the "attenuation" buffer remains effective under current conditions and cannot evaluate the role of sorption in elevated arsenic concentrations in MW-02. The model (PRHEEQC or equivalent) should explicitly evaluate: (a) fate of arsenic as a function of measured Eh/pH ranges in the system; (b) saturation indices for arsenate-bearing phases; (c) sensitivity of iron oxyhydroxide sorption capacity to pH perturbations in the 7.5–9.0 range; (d) projected arsenic concentrations under continued dewatering vs. post-closure scenarios.

B. Expand geochemical characterization at each accelerated monitoring event

6. **Collect and analyze stable isotope samples** (δ^{2}H, δ¸O) at every monitoring event at MW-01, MW-02, MW-03, the off-site USGS perched well, and the mine sump. This means monthly collection during the current accelerated monitoring period, reverting to quarterly only when ADEQ approves reduced monitoring frequency. Collection is easy, analysis is inexpensive, and interpretation is robust. Such data could directly test whether MW-02 is evolving toward a distinct isotopic signature consistent with the cone-of-depression import hypothesis (Solder and Beisner, 2020). There is no scientific or logistical justification for not collecting these data at every event.
7. **Conduct arsenic speciation analysis** (As(III)/As(V)) at MW-02 at a minimum of two monthly accelerated monitoring events. Under the oxidizing conditions documented in MW-02 (DO ~5.5 mg/L), arsenic from sulfide oxidation should be predominantly As(V). A

significant As(III) fraction would indicate different chemistry. This analysis directly tests the operative mobilization mechanism.

8. **Analyze dissolved iron speciation** (Fe(II)/Fe(III)) at MW-02 at a minimum of two events to further constrain redox conditions in the aquifer as sampled.
9. **Continue to analyze uranium isotopic composition** ($^{234}U/^{238}U$ activity ratios) at MW-01, MW-02, MW-03, the off-site USGS perched well, and the sump at a minimum in every sampling event These ratios are sensitive to water-rock interaction history and are useful for distinguishing water sources in uranium-bearing aquifer systems.

C. Conduct an age tracer study at MW-02 to test for fast-flow connectivity

The two studies below address the same fundamental question: whether any component of modern recharge reaches MW-02 via fast-flow pathways or whether the waters are isolated from direct communication with the surface. Together they constitute the minimum scientifically adequate response to EFRI's argument that the ancient radiocarbon age of the perched C-aquifer water precludes fast-flow connectivity. Both are standard, non-invasive methods that require only water sample collection; no artificial tracers are introduced. The recommendation is for a single analysis at each well. The rationale and methods are discussed in an attached Appendix B: Age Tracers.

10. **Collect and analyze groundwater samples for dissolve tracers of the modern atmosphere** to detect fast-flow pathway connectivity. Collect water samples at MW-01, MW-02, MW-03, RW-01, and the off-site USGS perched well for analysis of dissolved industrial volatile organic compounds (VOCs; EPA Method 524 or equivalent), chlorofluorocarbons (CFC-11, CFC-12, CFC-113), and sulfur hexafluoride (SF_6). These compounds were absent from the atmosphere prior to approximately 1940 (VOCs and CFCs) or 1960 (SF_6). Detection of any of these compounds above reporting limits in any monitoring well would constitute definitive evidence of fast-flow connectivity to the modern land surface, independent of bulk groundwater age. This study addresses a fundamentally different question from the tritium/noble gas study below: not whether the water was recharged during the Last Glacial Maximum or during the modern era, but whether the aquifer at the well screen has any hydraulic communication, however minor in volumetric terms, with a pathway that has transported modern atmospheric gases.
11. ***Collect and analyze groundwater samples for tritium and noble gases to assess groundwater age and recharge temperature.*** Collect water samples at MW-01, MW-02, MW-03, RW-01, and the off-site USGS perched well for analysis of the full dissolved noble gas suite (*^3He, ^4He, Ne, Ar, Kr, Xe*) and tritium (*^3H*). Analysis should be performed at a laboratory with demonstrated capability for low-level tritium by the ^3He ingrowth method (electrolytic enrichment plus mass spectrometry), with a detection limit of ≤0.3 TU. This study serves three analytically independent purposes: tritium as a proxy for post-1963 recharge, noble gas recharge temperature to assess recharge during the Last Glacial Maximum, and terrigenic helium as an independent indicator of old water..

D. Characterize the special heterogeneity of upgradient C-aquifer water quality

12. **Install or sample groundwater monitoring wells** at multiple azimuths from the mine at distances consistent with the postulated cone-of-depression capture zone (minimum 500

m, 1,000 m, and 2,000 m in at least three azimuthal directions). EFRI's hypothesis makes a specific, testable prediction: that off-site C-aquifer water at these distances contains arsenic at 30–43 µg/L. This prediction must be tested before the hypothesis can be considered demonstrated.

13. **Provide arsenic data** from at least three additional off-site C-aquifer wells or springs with flow paths documented to be directed toward the mine, to replace the single off-site data point currently cited.

E. Coordinate with the USGS to characterize subsurface features

14. **Coordinate with the pending USGS CSAMT geophysical survey** (USGS, 2025, Task 1) to characterize subsurface structural features (faults, fractures, and potential karst conduits) in the vicinity of the mine shaft and MW-02. The ring fracture zone and existing exploration borehole network should be explicitly evaluated as potential fast-flow pathways.

F. Regulatory recommendations

15. **Do not approve revising the arsenic AQL until the expanded investigation described above is complete** and the results, taken together, affirmatively demonstrate that the exceedance is attributable to natural phenomenon rather than to operationally induced mobilization. The current investigation does not meet this standard.
16. **Continue accelerated monthly monitoring at the current AQL (50 µg/L) until the USGS investigation results (expected 2026–2027) can be incorporated into the record** before any permanent AQL change is approved.
17. **Formally evaluate the APP mechanism for revising an AQL.** The current mechanism allows a facility to propose raising the compliance threshold based on a statistical representation of its own monitoring data while those data are trending upward. This risks sequential normalization of contaminant concentrations and loss of the ability to detect contamination from facility operators. The appropriate regulatory response to a documented increasing trend in a potential contaminant is intensified investigation, not threshold revision.
18. **ADEQ should clarify, through written guidance, that the definition of "discharge" encompasses mobilization of naturally occurring constituents** driven by operationally induced alteration of subsurface conditions (not only direct surface-to-groundwater releases from lined impoundments or other engineered structures). The penetration of the mine shaft through the perched Coconino Sandstone aquifer and the surrounding ring fracture zone, and the consequent hydrologic reorganization described in this comment (including cone-of-depression expansion, rerouting of groundwater through mineralized fractures and the alteration halo, and oxidative exposure of reduced sulfide mineral phases) constitutes a facility-induced alteration of groundwater quality that is causally attributable to mine operations regardless of whether any water from a surface impoundment has entered the subsurface. The current regulatory framing, in which EFRI can argue that the absence of a leaking surface impoundment is equivalent to the absence of a discharge, creates a regulatory exemption for the most geochemically consequential pathway by which breccia pipe mining affects groundwater quality. ADEQ should confirm, on the record, whether operationally induced subsurface

mobilization of naturally occurring constituents falls within the scope of the permit. If current regulatory language does not reach this mechanism, ADEQ should identify what regulatory authority exists to address it.

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TECHNICAL COMMENT APPENDIX A: URANIUM ACTIVITY RATIOS

Pinyon Plain Mine Aquifer Protection Permit No. P-100333
Proposed Minor Amendment: Revision of AQL for Arsenic at MW-02 (POC #2)
Submitted to: Arizona Department of Environmental Quality (ADEQ)
Commenter: Dr. Bradley K. Esser

Background: The Geochemistry and Utility of Uranium Activity Ratios

The activity ratio of ^{234}U to ^{238}U (“AR”) in groundwater is a well-established tracer of uranium source and water-rock interaction history. Its utility in this context rests on two well-understood physical principles.

- Secular equilibrium: In a uranium mineral that has been a closed system for several half-lives of ^{234}U (~245,000 years), the activities of parent (^{238}U) and granddaughter (^{234}U) are equal by definition: AR = 1.0. The Grand Canyon breccia pipe ore bodies, which formed in Proterozoic time, are in secular equilibrium. Direct stoichiometric dissolution of fresh uranium ore mineral (uraninite, coffinite) therefore produces AR ~1.0 in solution.
- ^{234}U enrichment in groundwater: Alpha-recoil fractionation enriches ^{234}U in natural groundwater. ^{234}U is produced by alpha decay of ^{238}U. The daughter nucleus receives a recoil of ~70 keV and is displaced ~30–40 nm through the crystal lattice, placing it in a structurally damaged, weakly bonded lattice site near grain surfaces. Contact with groundwater preferentially leaches this recoil-damaged ^{234}U relative to lattice-bound ^{238}U. In natural, unperturbed aquifer systems — including sandstone aquifers analogous to the perched Coconino — sustained alpha-recoil accumulation produces AR consistently above 1.0, typically in the range 1.5–3.0, depending on grain size, flow rate, and residence time. A key corollary: if a water sample has AR significantly below the expected natural background value, it indicates interaction with a uranium mineral whose surface-proximal ^{234}U inventory has been depleted by prior leaching, or congruent dissolution of a uranium mineral whose ^{234}U inventory has already been stripped.

The AR tracer has been applied in exactly this geological setting by the USGS. Beisner et al. (2023a) explicitly used ^{234}U/^{238}U ARs to discriminate ore-proximal from natural background groundwater at the Orphan Mine, a geologically analogous breccia pipe on the South Rim of the Grand Canyon. They found ARs of approximately 0.85–0.95 at springs emerging directly from or near the mineralized zone, compared with higher values at springs on natural background flow paths. Liebe (2003) similarly used the AR to characterize springs and surface waters across Grand Canyon National Park, and the USGS investigation proposal for Pinyon Plain (USGS, 2025) explicitly identifies isotopic characterization, including uranium isotope ratios, as a priority task.

The sump data demonstrate the discriminating power of the AR

The mine shaft sump provides a directly observable, time-resolved record of AR evolution as the shaft has progressively deepened toward and into the ore body. Individual uranium isotope activities (^{234}U, ^{235}U, ^{238}U, in pCi/L by alpha spectrometry) are reported for sump samples in the permit compliance monitoring record. The data are summarized in Table A-1.

Period	U (mg/L)	^{234}U (pCi/L)	^{238}U (pCi/L)	AR (^{234}U/^{238}U)	Phase / Interpretation
Q1 2023	0.0841	70.3	25.5	2.76	Pre-contact baseline: normal alpha-recoil enrichment
Q2 2023	0.163	122	59.2	2.06	Pre-contact baseline
Q3 2023	0.0574	40.7	19.7	2.07	Pre-contact baseline
Q4 2023	0.169	106	56.2	1.89	Pre-contact baseline
Q1 2024	0.130	95.3	49.0	1.94	Pre-contact baseline (mean AR ~2.1)
Q2 2024	54.5	13,000	1,100	11.8	First ore contact: extreme ^{234}U pulse — recoil zone stripped
Q3 2024	55.3	20,800	17,200	1.21	Sustained ore dissolution: AR converges toward secular eq.
Q4 2024	14.4	1,470	1,740	0.84	Post-contact: AR sub-unity — ^{234}U-depleted mineral dissolving

Table A-1. ^{234}U/^{238}U activity ratios in Pinyon Plain Mine sump water, Q1 2023–Q4 2024, calculated from individual uranium isotope activities reported in permit compliance monitoring. Source: EFRI Annual Reports 2023–2024; ADEQ QWQD.

This five-quarter sequence is a textbook illustration of the AR tracer's discriminating power. Three geochemically distinct phases are identifiable:

- Pre-ore-contact baseline (Q1 2023 – Q1 2024): AR = 1.89–2.76, mean ~2.1. This represents ordinary alpha-recoil fractionation in Coconino Sandstone perched groundwater interacting with non-ore breccia pipe wall rock. This is the expected signature of natural perched C-aquifer water at this site — the geochemical analog of what the monitoring wells should be sampling if they reflect undisturbed background Coconino water.
- First ore-body contact (Q2 2024): U increases from 0.13 mg/L to 54.5 mg/L (420-fold) and AR spikes to 11.8. The extraordinarily high AR reflects a transient flush of the accumulated recoil-damaged ^{234}U inventory from freshly exposed ore grain surfaces — a surface reservoir that had been building since the last episode of active dissolution. This AR value is diagnostic of first contact with a previously unweathered uranium mineral surface.

- Sustained congruent dissolution (Q3 2024 – Q4 2024): As ore-mineral dissolution becomes dominant, the AR collapses. Q3 2024 AR = 1.21 (near secular equilibrium, consistent with stoichiometric dissolution of bulk mineral). Q4 2024 AR = 0.84 (below secular equilibrium), indicating that dissolution has progressed into the mineral interior, now depleted in ^{234}U at grain surfaces from the prior leaching episode. This depleted AR is a fingerprint of advanced ore-body dissolution.

The AR data in the sump therefore provide a time-stamped record of the mine's progressive penetration of the breccia pipe ore body, independently of the uranium mass concentration data. They would provide the same discriminating power in monitoring well waters if they were reported.

^{234}U/^{238}U activity ratios are available for monitoring wells but are not reported

The APP permit (Tables 7 and 8, both the 2022 and 2024 versions) requires the following radionuclide measurements for the monitoring wells and the sump discharge:

- Adjusted Gross Alpha Activity (pCi/L)
- Radium-226 (pCi/L)
- Radium-228 (pCi/L)
- Uranium Isotopes (234, 235, 238) (pCi/L)

The critical point regarding the last entry is that uranium isotope activities are measured as individual isotopes, not as a pre-summed total. Standard methods for uranium isotope analysis in water (EPA Method 908.0 or equivalent; ASTM D3972; USGS methods) use alpha spectrometry following radiochemical separation. The alpha spectrum resolves ^{234}U (~4.77 MeV), ^{235}U (~4.40 MeV), and ^{238}U (~4.20 MeV) as distinct peaks. The laboratory therefore measures and records individual isotope activities (^{234}U, ^{235}U, ^{238}U in pCi/L) for every sample analyzed.

This is confirmed by the sump compliance record, in which individual isotope activities are explicitly tabulated in the Annual Reports (^{234}U, ^{235}U, and ^{238}U separately in pCi/L; Table A-1 above). For the monitoring wells, however, only the aggregated sum "Uranium Isotopes (pCi/L)" is reported in the QWQD permit compliance database. Individual isotope activities for MW-01, MW-02, MW-03, and RW-01 are not accessible in any publicly reported compliance record.

This is not an analytical limitation. The individual isotope activities were measured and are in the laboratory's possession. They are simply not being reported as separate fields for the monitoring wells. There is no additional laboratory cost to requesting disaggregated reporting.

The utility of uranium activity ratios at Pinyon Plain Mine

Below are recommendations for the incorporation of uranium activity ratios into monitoring and compliance at Pinyon Plain Mine. Since they are not directly relevant to the current amendment request to revise the AQL for arsenic and to change sampling intervals for monttoring wells, I have not included them in the main text of the the Technical Comment. However I feel strongly that uranium activity ratios should have a role in monitoring and compliance at the mine site, especially considering that they are informative and the measurements are already being made

1. Require EFRI to report ^{234}U, ^{235}U, and ^{238}U as separate values (pCi/L) in the QWQD compliance database for all monitoring wells (MW-01, MW-02, MW-03, and RW-01) for all future sampling events, retroactively for all existing samples if laboratory records are available, and at the accelerated monthly frequency currently in effect for the Coconino monitoring wells. This is not an additional analytical requirement: alpha spectrometry already resolves the individual isotopes, and the laboratory records these values as a matter of routine analytical practice. The requirement is for disaggregated reporting of data that the laboratory already possesses.

2. Once individual isotope activities are reported, require EFRI to calculate and report the ^{234}U/^{238}U activity ratio for each monitoring well at each sampling event and to include time-series plots of the AR alongside the dissolved uranium concentration in each quarterly and annual report. Require that these plots span the full monitoring record, beginning with the ambient groundwater monitoring period.

3. Require EFRI to evaluate the temporal co-trends among the ^{234}U/^{238}U AR and the following co-monitored constituents that have been identified as showing statistically significant trends at one or more Coconino monitoring wells: arsenic, uranium mass concentration, thallium, nickel, antimony, barium, fluoride, and adjusted gross alpha activity. The AR provides geochemical context for interpreting whether co-trending in this element suite reflects (a) import of a natural off-site water source with a distinct AR signature, (b) in-situ mobilization driven by changing redox conditions at ore-proximal sites, or (c) dilute mixing with ore-contact water from the shaft. These three scenarios have different and distinguishable AR signatures:

    - *Natural off-site Coconino C-aquifer import:* AR expected ~1.5–3.0, comparable to or above the pre-contact sump baseline of ~2.1. If the cone-of-depression hypothesis is correct and MW-02 is increasingly sampling distal background Coconino water, a shift toward or above the pre-contact sump AR should be detectable.
    - *In-situ desorption or secondary mineral dissolution from ore-proximal aquifer materials*: AR expected near secular equilibrium (~1.0) or in the natural alpha-recoil range (~1.5–2.0), depending on the degree of prior leaching of the local aquifer matrix. This would produce a different AR trajectory than ore-body dissolution.
    - *Mixing with ore-contact shaft water:* AR expected to evolve in parallel with the sump AR time series (Q4 2024 sump AR = 0.84; Q4 2025 sump chemistry indicates continuing ore-body contact). If MW-02 receives a fast-flow contribution of sump-

proximal water, a shift toward sub-unity AR would be expected and would be diagnostic.

4. Require that the AR evaluation extend to the R-aquifer well (RW-01) to test whether uranium in the deeper aquifer shows any evolution in isotopic signature over the monitoring record. The pre-contact sump AR (~2.1) and the expected natural R-aquifer AR provide reference end-members for any mixing calculation.

5. Require comparison of the MW-02 AR time series with the AR data from the USGS regional spring and well network (Liebe, 2003; Beisner et al., 2023a; Tillman et al., 2021), which provide the regional natural background AR distribution for springs and groundwaters in the Grand Canyon region. This comparison will establish whether MW-02 falls within the normal range for natural Coconino C-aquifer groundwater, or whether it plots as an outlier consistent with ore-contact influence. The USGS investigation currently underway (USGS, 2025) is expected to generate additional AR data for regional sites that should also be incorporated.

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TECHNICAL COMMENT APPENDIX B: AGE TRACERS

Pinyon Plain Mine Aquifer Protection Permit No. P-100333
Proposed Minor Amendment: Revision of AQL for Arsenic at MW-02 (POC #2)
Submitted to: Arizona Department of Environmental Quality (ADEQ)
Commenter: Dr. Bradley K. Esser

Age Tracers to Assess Fash Path Contamination and Groundwater age

Recommendation C recommends collecting monitoring well samples for analysis of industrial VOCs and for analysis of tritium, noble gas, helium isotope and industrial VOC analyses. This Appendix expands on the methodology and use of these tracers.

Tracers of Modern Atmosphere

Industrial VOCs": A broad-spectrum dissolved VOC scan targets compounds including chlorinated solvents (trichloroethylene, tetrachloroethylene, 1,1,1-trichloroethane), halogenated compounds (chloroform, carbon tetrachloride), fuel hydrocarbons (BTEX), and refrigerants (freons). These compounds have been present in the global troposphere since the mid-20th century and are now detectable at sub-ppb concentrations in precipitation, soil gas, and shallow groundwater worldwide. Their presence in deep confined groundwater is physically impossible unless a connected pathway for modern atmospheric gas exchange exists. The State of California Water Board GAMA Program, which I led as Technical Lead for Groundwater Age Determination and Special Studies for eleven years, routinely applies this suite as a first-pass screen for fast-flow connectivity in confined aquifer systems (Fram et al., 2012; Moran et al., 2015; Singleton et al., 2012). No artificial tracer introduction is required. The analytical cost is low.

Chlorofluorocarbons (CFC-11, CFC-12, CFC-113) and sulfur hexafluoride (SF_6). CFCs and SF_6 are quantitative atmospheric age tracers (Plummer and Busenberg, 2000). Their dissolved concentrations in groundwater at the time of recharge are controlled by solubility equilibrium with the atmosphere, and the atmospheric concentrations of these gases are precisely known as a function of time from continuous records beginning in 1940 (CFCs) and 1953 (SF_6). In undisturbed ancient groundwater recharged before 1940, CFC concentrations should be at or below the detection limit. Detection above the detection limit would indicate a component of post-1940 recharge. Even trace amounts at parts-per-quadrillion concentrations are interpretable against the atmospheric input function. In combination with the tritium data from C1, a positive CFC or SF_6 detection in a tritium-free sample would suggest water containing a small fraction of modern recharge from a period before weapons-test tritium was added to the hydrological cycle (pre-1952), which is still informative about conduit connectivity on decadal timescales.

Together, the VOC scan and the CFC/SF_6 measurements cover different sensitivity ranges and different time windows for fast-flow connectivity. Their strength in this regulatory context is their binary character: detection is affirmative evidence of modern atmospheric communication via a preferential pathway, regardless of what fraction of the bulk flow that pathway represents. Even if 99% of the water at MW-02 is ancient matrix flow, a 1% component of modern recharge delivered via a conduit connecting the shaft to the land surface is hydrologically significant for contaminant transport.

Important caveat: This tracer suite tests for atmospheric communication, not for mine-derived contamination per se. A non-detect result at all monitoring wells would constitute the strongest available evidence that the aquifer system at these locations is not in fast-flow communication with the modern atmosphere, supporting EFRI's ancient-water argument. A non-detect result does not, however, rule out contaminant transport via processes that do not require atmospheric connectivity (e.g., advective transport of dissolved metals through fracture networks from ore-contact water, which involves no atmospheric gas exchange). The tracer results must therefore be interpreted in conjunction with the multi-element geochemical data, redox parameters, and stable water isotope data from the other recommended measurements.

Tracers of Groundwater Age and Recharge Temperature

Tritium as a test for post-1952 recharge: Tritium (half-life 12.32 years) was elevated in global precipitation by atmospheric nuclear weapons testing beginning in 1952, reaching peak values of 1,000–6,000 TU in the northern hemisphere in 1963–1964. Tritium in modern precipitation in northern Arizona is currently ~2–5 TU (Harms et al., 2016). Any detection of dissolved tritium above the analytical detection limit (~0.3 TU by ^{3}He ingrowth) in MW-02 would provide unambiguous evidence that a component of the water entered the aquifer after 1952, i.e. that MW-02 is in hydraulic communication with a fast-flow pathway connected to the modern land surface. This conclusion is independent of, and complementary to, the radiocarbon age argument: even if the bulk radiocarbon age is ~10,000 years, a 1% admixture of modern tritiated water would be detectable at approximately 0.03–0.05 TU with enrichment methods and would have significant implications for contaminant transport velocities. The expected result for truly isolated ancient perched Coconino groundwater is non-detect at all wells. Any deviation from non-detect at any Coconino well requires mechanistic explanation.

Noble gas recharge temperature (NGT) as evidence for Pleistocene recharge: The concentrations of dissolved Ne, Ar, Kr, and Xe in groundwater are controlled at the time of recharge by temperature-dependent gas solubility and the amount of excess air trapped during infiltration (Aeschbach-Hertig et al., 2000). These concentrations are preserved unchanged in confined groundwater and can be inverted to recover the soil temperature at the time of recharge. If the C-aquifer water was recharged during the late Pleistocene, as the radiocarbon ages (~10,000–16,000 yr) suggest, the NGT should be detectably cooler than modern mean annual soil temperature at the Coconino Plateau. The most directly analogous published study is that of Zhu and Kipfer (2010), who measured NGTs 5–6°C below modern in the confined Navajo (N) Aquifer of the Black Mesa basin, ~100 km northeast of Pinyon Plain, in a comparable confined aeolian sandstone aquifer with similar radiocarbon ages. The current mean annual soil temperature at Pinyon Plain (~2,100 m elevation) is approximately 6–8°C; Pleistocene recharge would be expected to yield NGTs in the range of roughly 0–3°C. A measured NGT in that range would independently corroborate the radiocarbon age interpretation and confirm that the bulk water is not recently recharged. The NGT measurement provides information no other tracer in this suite provides: direct evidence of the paleotemperature at the time of recharge.

Terrigenic helium ($^4He_{terr}$) as an independent indicator of old water and a test for anomalous helium input: In groundwater older than a few thousand years, radiogenic ^4He accumulates from in-situ alpha decay of uranium and thorium in the aquifer matrix, producing dissolved ^4He concentrations measurably above the atmospheric equilibrium value ($\sim 4.0\times10^{-8}$ ccSTP/g). For groundwater of ~10,000–16,000-year age, terrigenic ^4He excess above atmospheric equilibrium is expected to be detectable, depending on the U/Th content of the aquifer matrix (Andrews, 1985; Castro et al., 2000). Detection of significant $^4He_{terr}$ at MW-02 or the R-aquifer well (RW-01) would provide a third independent indicator of old water, internally consistent with the radiocarbon ages.

Crossey et al. (2009) have successfully measured helium isotopic composition in Grand Canyon spring waters. The R-M aquifer well (RW-01) is particularly favorable for this measurement because it is artesian and deeply confined (screened at 788–902 m depth), providing a high-quality dissolved gas sample environment free of atmospheric contamination and degassing.

Sample collection for dissolved noble gas analysis to determine both noble gas recharge temperature and terrigenic helium concentration requires specialized copper tube or steel cylinder containers that preserve dissolved gas concentrations without atmospheric exchange; these are standard equipment at isotope hydrology laboratories and add only modest incremental cost to a sampling event. The USGS Reston Groundwater Dating Laboratory, Lawrence Livermore National Laboratory Noble Gas Mass Spectrometry Group, and several universities maintain this capability.

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Dr. Bradley K. Esser, PhD

Positions (Lawrence Livermore National Laboratory):

Retired (2021-present)
Visiting Scientist – LLNL Environmental Isotope Systems, Retiree (2018-2021)
Lead – LLNL Environmental Radiochemistry Group Leader, Retired (2001-2018)
Lead – LLNL Environmental Monitoring Radioanalytical Laboratory (EMRL), Retired (2002-2018)
Lead – State of California Groundwater Ambient Monitoring and Assessment (GAMA) Special Studies Program, Retired (2007-2018)

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EDUCATION:

1991, Yale University, Ph. D. (Geology and Geophysics; advisor, K. K. Turekian)
Dissertation: *"Osmium isotope geochemistry of terrigenous and marine sediments"*
1983, U. of Arizona, B. S. with High Distinction (Geosciences).
1980, U. of Arizona, B. S. with Highest Distinction (Ecology and Evolutionary Biology)

RESEARCH EXPERIENCE:

Lawrence Livermore National Laboratory, Chemical Biology & Nuclear Science Division

Visiting & Contract Scientist, Environmental Isotope Systems (2018-2021)
Group Lead, Environmental Radiochemistry (2001-2018)
Lead –Environmental Monitoring Radioanalytical Laboratory (2002-2018)
Technical Lead – California GAMA Special Studies program (2007-2018)
Staff Scientist (1993-2001)
Postdoctoral Research Assistant (1990–1993); Sid Niemeyer, advisor.

Yale University, Department of Geology and Geophysics

Doctoral Research Assistant (1985-1990), K. K. Turekian, advisor

HONORS

Hitchon Award, IAGC (2007)
Dissertations Symposium on Chemical Oceanography (1991)
Philip M. Orville Prize, Yale University (1991)
Bateman Prize, Yale University (1983)

PROFESSIONAL MEMBERSHIPS

American Geophysical Union (active)
Geochemical Society (active)
Geological Society of America
American Chemical Society
National Ground Water Association
Groundwater Resources Association of California (active)

EXPERTISE AND GENERAL RESEARCH INTERESTS:

- **Groundwater age-dating**, isotope biogeochemistry, and reactive transport modeling in support of water resource management, especially with regards to groundwater nitrate contamination.
- **Isotope geochemistry** to constrain the fate and transport of natural and anthropogenic elements, radionuclides and nutrients in natural waters and sediments.
- **Inorganic mass spectrometry and radiation counting** applied to environmental science and water resource management.

RESEARCH HIGHLIGHTS:

- **The California drought and groundwater:** Through five years of drought, California relied more heavily on groundwater. LLNL used decadal-scale groundwater age tracers and noble gas tracers to understand the delayed effects of the drought on high-elevation streamflow and high-volume spring discharge. LLNL also developed new short timescale groundwater age tracers to support the use of recycled wastewater for groundwater recharge.
- **Nitrate contamination of California groundwater:** Nitrate is the most prevalent contaminant of groundwater in California, the nation and the world. I led an LLNL strategic initiative to develop better methods for characterizing the source, transport and fate of nitrate in groundwater; gave invited talks and chaired symposia on groundwater nitrate; and worked with the agricultural community on implementing new monitoring requirements. Until recently, the focus of the GAMA Special Studies project, for which I was technical lead, was nitrate in groundwater. LLNL, under my leadership, used groundwater age to determine the history, nitrate isotopic composition to determine the source, and excess nitrogen to assess denitrification and determine the fate of nitrate in groundwater.
- **Uranium contamination on tribal lands:** Uranium contamination of soil and water on Navajo tribal lands is the legacy of uranium mining and milling decades ago. LLNL worked with the Navajor Nation to better characterize and remediate uranium contamination. The effort included validation of landfill performance and development of portable radiation monitoring.

MANAGEMENT HIGHLIGHTS

- **State of California Water Board Committee Lead:** California recently passed legislation (SB4) to regulate well stimulation. I chaired a multi-institution (national labs and universities) committee that made recommendations to the State on groundwater monitoring of oil & gas operations. In a very short time (9 months), we worked with environmental groups and with industry to craft recommendations for regulation that were largely adopted by the State.
- **LLNL/State of California Program Lead:** As program lead for the California Water Board GAMA Special Studies program for the past 12 years, I managed a budget from $500K to $2M (for a total of over $13M), several employees, students, and UC/CSU collaborators and delivered dozens of reports.
- **LLNL Analytical Capability Lead:** As lead for the Environmental Monitoring Radioanalytical Laboratory from 2002 to 2018, I managed a $1M (unburdened) budget, 4 laboratories and 5 employees, and provided radioanalytical data for radioactivity on the two LLNL sites. Over the those 16 years, sample load increased by 20%, turn-around times decreased and cost to the customer remained constant.
- **LLNL Group Leader:** Lead for Environmental Radiochemistry for 16 years, with line management responsibilities for 25 employees and students.

PUBLICATION METRICS (AS OF 5/15/2025)

- Publications: 112 (ResearchGate)
- Reads: 14,438 (ResearchGate)
- Citations: 2645 (ResearchGate)
- h-index: 34 (Google Scholar)
- i10-index: 61 (Google Scholar)
- Citations: 3486 (Google Scholar)

SELECT AND RECENT JOURNAL ARTICLES

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Esser B. K, Volpe A, Kenneally J. M. and Smith D. K. (1994) Preconcentration and Purification of Rare-Earth Elements in Natural-Waters Using Silica-Immobilized 8-Hydroxyquinoline and a Supported Organophosphorus Extractant. *Analytical Chemistry* **66**, 1736–1742.

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