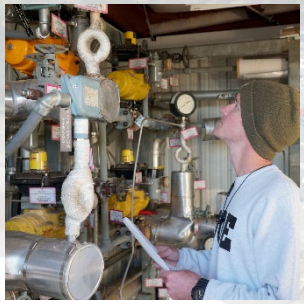
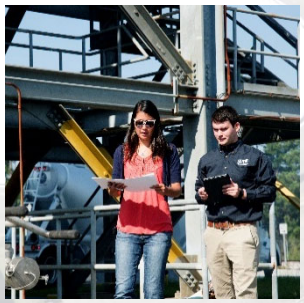
Savannah River Remediation
AMENTUM | BECHTEL | JACOBS | BWXT

December 09, 2020

SRR-CWDA-2020-00088





LONG TERM RADIOLOGICAL LYSIMETER PROGRAM

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Safety SRR Integrity SRR Ownership SRR Teamwork SRR Continuous Improvement

RadFLEx Background

- **What:**
  - A multi-year study is being performed at the Radionuclide Field Lysimeter Experiment Facility (RadFLEx) to evaluate radionuclide mobility from sources emplaced in lysimeters exposed to the outside environment.

Figure 1-1: SRNL-STI-2013-00446

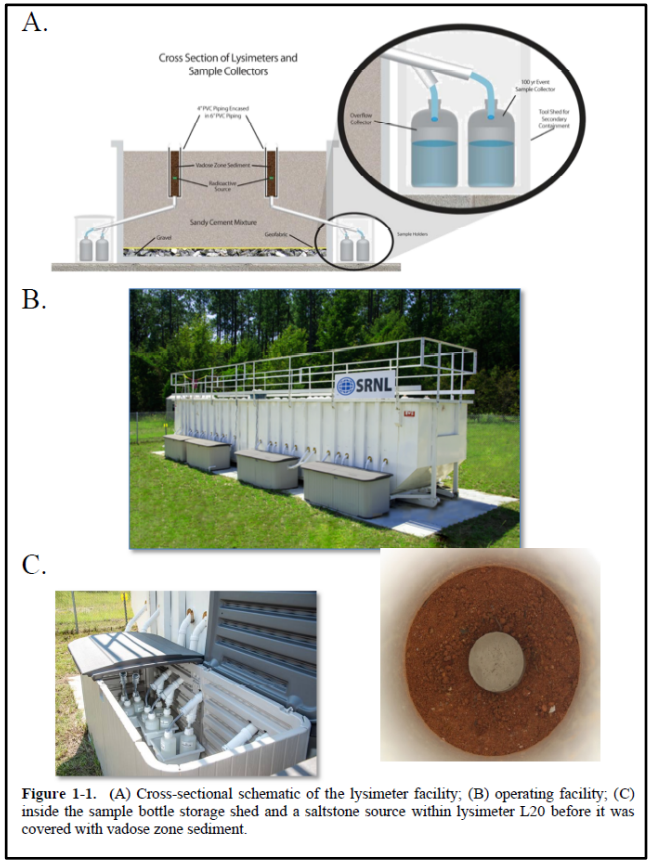


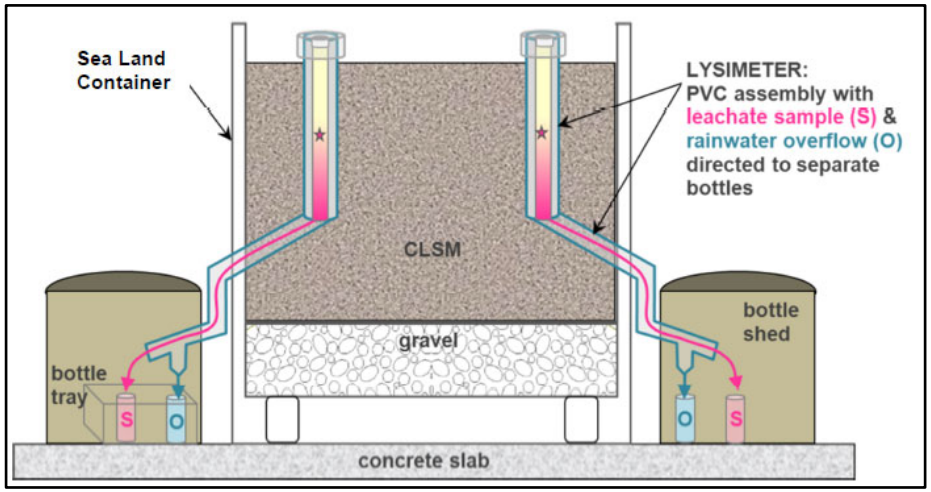
Figure 1-1. (A) Cross-sectional schematic of the lysimeter facility; (B) operating facility; (C) inside the sample bottle storage shed and a saltstone source within lysimeter L20 before it was covered with vadose zone sediment.

Lysimeter	Rad	Source	Description	Uncapped - start of infiltration
1	EMPTY	N/A	EMPTY	N/A
2	EMPTY	N/A	EMPTY	N/A
3	Control	Cement	Cement Control, 10 yr	7/5/2012
4	Gamma	Cement	Cement Gamma suite, 2 yr	7/5/2012
5	Gamma	Cement	Cement Gamma suite, 4 yr	7/5/2012
6	Gamma	Cement	Cement Gamma suite, 10 yr	7/5/2012
7	Tc&I	Cement	Cement Tc&I, 4 yr	7/5/2012
8	Np	Soil	Np(IV)O_2, 2 yr	10/24/2019
9	Ra	Soil	Ra-226/Sand	10/24/2019
10	Pu	Soil	Pu(IV)$(C_2O_4)_2$, Grass, 4 yr	7/5/2012
11	Pu	Soil	Pu(IV)$(C_2O_4)_2$, Grass, 10 yr	7/5/2012
12	Control	Soil	Grass Control	7/5/2012
13	Pu	Soil	Pu(IV)$(C_2O_4)_2$, Grass, 2 yr	7/5/2012
14	Pu	Soil	Pu(V)NH4 (CO3)/OM, 2 yr	10/24/2019
15	Control	N/A	Saltstone Control, 10 yr	7/5/2012
16	Gamma	Saltstone	Saltstone Gamma suite, 2 yr	7/5/2012
17	Gamma	Saltstone	Saltstone Gamma suite, 4 yr	7/5/2012
18	Gamma	Saltstone	Saltstone Gamma suite, 10 yr	7/5/2012
19	Tc&I	Saltstone	Saltstone Tc&I, 4 yr	7/5/2012
20	Pu	Soil	Pu(V)NH4 (CO3)/OM, 4 yr	10/24/2019
21	Pu	Soil	Pu(V)NH4 (CO3)/OM, 10 yr	10/24/2019
22	Pu	Soil	Pu(V)$NH_4(CO_3)$/OM, 4 yr	7/5/2012
23	Pu	Soil	Pu(V)$NH_4(CO_3)$/OM, 10 yr	7/5/2012
24	Control	Soil	Instrumented Control	7/5/2012
25	Control	Soil	Sediment Control	7/5/2012
26	Ra	Soil	Ra-226	7/8/2020
27	Gamma	Soil	Gamma suite, 4 yr	7/5/2012
28	Gamma	Soil	Gamma suite, 10 yr	7/5/2012
29	Np	Soil	Np(IV)O_2, 6 yr	10/24/2019
30	Np	Soil	Np(V)O_2NO_3, 4 yr	7/5/2012
31	Np	Soil	Np(V)-nitrate, 2 yr	10/24/2019
32	Np	Soil	Np(IV)O_2, 10 yr	7/5/2012
33	Np	Soil	Np(V)-nitrate, 6 yr	10/24/2019
34	Pu	Soil	Pu(III)$_2(C_2O_4)_3$, 4 yr	7/5/2012
35	Pu	Soil	Pu(III)$_2(C_2O_4)_3$, 10 yr	7/5/2012
36	Ra	Soil	Ra-226	7/8/2020
37	Control	Soil	Instrumented Control	7/5/2012
38	Pu	Soil	Pu(V)NH4 (CO3)/Sand, 2 yr	10/24/2019
39	Pu	Soil	Pu(IV)$(C_2O_4)_2$, 4 yr	7/5/2012
40	Pu	Soil	Pu(IV)$(C_2O_4)_2$, 10 yr	7/5/2012
41	Pu	Soil	Pu(V)NH4 (CO3)/Sand, 4 yr	10/24/2019
42	Pu	Soil	Pu(V)$NH_4(CO_3)$, 4 yr	7/5/2012
43	Pu	Soil	Pu(V)$NH_4(CO_3)$, 10 yr	7/5/2012
44	Pu	Soil	Pu(V)NH4 (CO3)/Sand, 10 yr	10/24/2019
45	Pu	Soil	Pu(IV)O_2 colloids, 4 yr	7/5/2012
46	Pu	Soil	Pu(IV)O_2 colloids, 10 yr	7/5/2012
47	I-129	Cement	Cement I-129	8/25/2020
48	I-129	Cement	Cement I-129	8/25/2020

Lysimeter Effluent Analysis

- Lysimeter effluents sampled and analyzed routinely
- Can help in ascertaining mechanism(s) controlling radionuclide mobility in the vadose zone (e.g., sorption, solubility, colloid-facilitated transport)
  - Pu concentrations of 10^{-15} – 10^{-13} mol/L measured in lysimeter effluent.
    - Values are close to the solubility limits for Pu(IV) hydroxide phases.
  - Np breakthrough for Np(IV) and Np(V) sources
    - [Np(V)] ≈ 100 – 10,000 [Np(IV)]
    - Data suggests:
      - Np(V) sorption controlled
      - Np(IV) solubility controlled

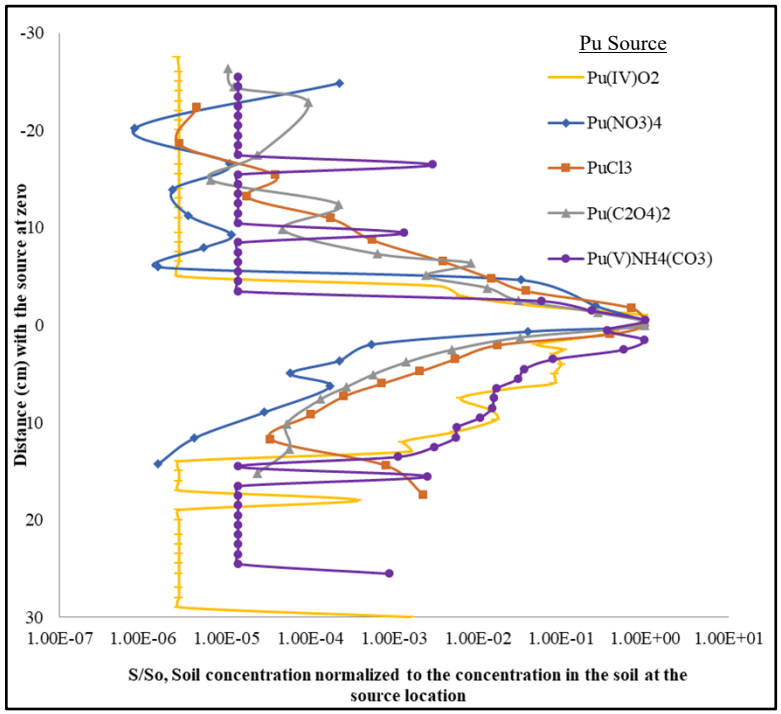
Figure 2-1(A): SRNL-STI-2013-00446



Lysimeter Solid Phase Analysis

- Used to determine radionuclide migration from the emplaced source
- Performed after several years of lysimeter exposure to the outside environment
- Results can help in determining mechanism(s) responsible for contaminant transport
  - Varied Pu transport for $Pu(IV)O_2(s)$ and $NH_4Pu(V)O_2CO_3(s)$ sources
    - Hypothesis #1: Transport of Pu as $PuO_2(s)$ colloids
    - Hypothesis #2: Differing solubilities of source material

Figure 6: SRRA021685-000010



Desorption Studies

- **Use contaminated sediment from dissected lysimeters to perform batch desorption experiments**
  - Provides site-specific K_d values
  - Cs and Eu have demonstrated the potential for aging-effects (i.e., increased affinity over time) with SRS sediment

Element	K_d (mL/g)	
	Measured	Modeled [c]
Pu	25,000 [a]	650
Pu	1,600 [b]	650
Co	29	40
Ba	29	20
Cs	2,200	10
Eu	4,300	1000

a) $Pu(IV)O_2$ lysimeter

b) $Pu(V)NH_4(CO_3)$ lysimeter

c) Table 4.3-4 SRR-CWDA-2019-00001, Rev. 0

Figure 1: SRRA021685-000009

References

- *SRNL Radionuclide Field Lysimeter Experiment: Baseline Construction and Implementation* (SRNL-STI-2012-00603)
- *Status of SRNL Radiological Field Lysimeter Experiment – Year 1* (SRNL-STI-2013-00446)
- *Radionuclide Field Lysimeter Experiment (RadFLEx): Geochemical and Hydrological Data for SRS Performance Assessments* (SRNL-STI-2017-00677)
- *Status of SRNL Radiological Field Lysimeter Experiment – Year 7* (SRNL-L3230-2019-00005)
- *Determination of constituent concentrations in field lysimeter effluents – FY13 Final Report* (SRR-CWDA-2013-00121)
- *Determination of constituent concentrations in field lysimeter effluents – FY14 Final Report* (SRRA021685SR)
- *Determination of constituent concentrations in field lysimeter effluents – FY15 Final Report* (SRRA021685-000007)
- *Determination of constituent concentrations in field lysimeter effluents – FY15 Final Report* (SRRA021685SR-2016)
- *Determination of constituent concentrations in field lysimeter effluents – FY17 Final Report* (SRRA021685-000008)
- *Determination of constituent concentrations in field lysimeter effluents – FY18 Final Report* (SRRA021685-000011)
- *Determination of constituent concentrations in field lysimeter effluents – FY19 Final Report* (SRRA021685-000013)
- *Analysis of plutonium soil concentrations in field lysimeter experiments* (SRRA021685-000009)
- *Analysis of plutonium soil concentrations in field lysimeter experiments: Soil Pu concentration profile from a $NH_4Pu(V)O_2CO_3(s)$ source* (SRRA021685-000010)
- *Partitioning of Cesium-137 and Other Gamma-Emitting Radionuclides to SRS Sediments Recovered from Field Lysimeter Experiments at the Savannah River Site* (SRRA021685-000012)