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NUCLEAR REGULATORY COMMISSION
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January 5, 2017

MEMORANDUM TO: Gregory Suber, Chief
Low Level Waste Branch
Division of Decommissioning,
Uranium Recovery, and Waste Programs

THRU: Christopher McKenney, Chief /RA/
Performance Assessment Branch
Division of Decommissioning,
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FROM: Karen Pinkston, Systems Performance Analyst /RA/
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SUBJECT: TECHNICAL REVIEW: IODINE SORPTION COEFFICIENTS FOR USE
IN PERFORMANCE ASSESSMENTS FOR THE SALTSTONE
DISPOSAL FACILITY (DOCKET NO. PROJ0734)

The U.S. Nuclear Regulatory Commission (NRC) staff has performed a technical review of the sorption of iodine-129 (I-129) to support the NRC's review of performance assessments (PAs) and Special Analyses (SAs) for the Saltstone Disposal Facility (SDF) at the Savannah River Site (SRS). As part of this review, the NRC staff reviewed new information on the leaching of iodine from saltstone and the sorption of iodine in the subsurface. This technical review report supports Monitoring Factors (MFs) 5.04, "Certain Risk-Significant Kd Values for Saltstone", 6.01, "Certain Risk-Significant Kd Values in Disposal Structure Concrete", 7.01, "Certain Risk-Significant Kd Values in Site Sand and Clay", 10.04, "Kd Values for Saltstone", 10.06, "Kd Values for Disposal Structure Concrete", and 10.09, "Kd Values for SRS Soil", as detailed in the NRC staff's plan for monitoring the SDF (Agencywide Documents Access and Management System [ADAMS] Accession No. ML13100A076).

The NRC staff concluded that the preliminary leaching data from cores from saltstone in Saltstone Disposal Structure (SDS) 2A indicate that the iodine is not retained significantly by field-emplaced saltstone and the Kd values assumed for iodine for saltstone in the FY2014 SA are higher than are justified. Because of the risk-significance of the assumed saltstone Kd values for iodine, the NRC is expanding MF 5.04, "Certain Risk-Significant Kd Values for Saltstone", to include the sorption of iodine on saltstone. The NRC is also expanding MF 6.01, "Certain Risk-Significant Kd Values in Disposal Structure Concrete", to include sorption of iodine

on disposal structure concrete because the Kd values assumed by DOE appear to be slightly higher than is justified and the low sorption of iodine in saltstone may cause the chemical performance of the disposal structure concrete to control the release of iodine from the engineered system.

The NRC staff concluded that the use of the new proposed subsurface Kd values for iodine in the Saltstone PAs and SAs is not adequately supported. The NRC staff also concluded that the basis for the assumed leachate impact factors and resulting leachate impacted subsurface Kd values is unclear. The NRC staff is therefore expanding MF 7.01 to include the subsurface Kd values for iodine and the leachate impact factors and the leachate impacted subsurface Kd values for iodine.

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Enclosure:

Technical Review of the Sorption of Iodine at the Saltstone Disposal Facility at the Savannah River Site

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Technical Review of the Sorption of Iodine at the Saltstone Disposal Facility at the Savannah River Site

Date: January 5, 2017

Reviewers

Karen Pinkston, Systems Performance Analyst, U.S. Nuclear Regulatory Commission

Primary Documents

SRNL-STI-2016-00106, Revision 0, "Results and Analysis of Saltstone Cores Taken from Saltstone Disposal Unit Cell 2A", March 2016. ML16173A174.

SRR-CWDA-2016-00053, Revision 1, "Research Results/Status-Savannah River Site Salt Waste Disposal NRC Onsite Observation Visit", April 2016. ML16134A186

SRNL-STI-2012-00518, Revision 0, "Radioiodine Geochemistry in the SRS Subsurface Environment", May 2013. ML14196A201.

Background

Iodine-129 (I-129) was identified by the Department of Energy (DOE) as being a risk-significant radionuclide for the disposal of salt waste at the Saltstone Disposal Facility (SDF) at the Savannah River Site (SRS) in the 2009 Performance Assessment (PA) for the Saltstone Disposal Facility (SRR-CWDA-2009-00017). In the NRC 2012 Technical Evaluation Report (TER) (Agencywide Documents Access and Management System [ADAMS] Accession No. ML121020140) documenting the Nuclear Regulatory Commission (NRC) review of the 2009 Saltstone PA, the NRC staff concluded that "the Kd values assumed for iodine for cementitious materials seem to be slightly higher than measured values". The NRC staff also concluded that the Kd value of 0 mL/g assumed for vadose zone/sandy soil in the 2009 PA Evaluation Case was appropriate and the Kd value of 0.6 mL/g assumed for the backfill clayey soil in the Evaluation Case was reasonably based on site-specific data. The NRC staff further concluded that site specific observations supported the slightly higher Kd values (i.e., a Kd value of 0.3 mL/g for sandy soil and 0.9 mL/g for clayey soil) assumed for the subsurface in Case K in DOE's response to NRC's request for additional information (RAI) (SRR-CWDA-2011-00044).

In Appendix A of the 2012 TER and in Revision 1 to the NRC Plan for Monitoring Disposal Actions taken by DOE at the SRS SDF (ML13100A113) the NRC staff developed Monitoring Factors (MFs) related to concerns identified in the 2012 TER. MFs related to sorption coefficients include: MFs 5.04, "Certain Risk-Significant Kd Values for Saltstone", 6.01, "Certain Risk-Significant Kd Values in Disposal Structure Concrete", 7.01, "Certain Risk-Significant Kd Values in Site Sand and Clay", 10.04, "Kd Values for Saltstone", 10.06, "Kd Values for Disposal

Structure Concrete”, and 10.09, “Kd Values for SRS Soil”. The most risk-significant Kd values were monitored under MFs 5.05, 6.01, and 7.01, while the less risk-significant Kd values were monitored under MFs 10.04 and 10.06. In the monitoring plan, MFs 5.04 and 6.01 were limited to the saltstone and disposal structure concrete Kd values for radium and selenium and MF 7.01 was limited to the subsurface soil Kd values for selenium. In the monitoring plan, the NRC staff notes that these monitoring factors may be expanded to include additional radionuclides if their Kd values prove to be risk-significant and are not adequately supported.

DOE assumed the same Kd values for saltstone and disposal structure concrete in the 2009 PA Evaluation Case and Case K, the 2013 Special Analysis (SA) for the SDF (SRR-CWDA-2013-00062, Rev. 2), and the 2014 SA for the SDF (SRR-CWDA-2014-00006, Rev 2) (Table 1). In the RAI response for the 2014 SA (SRR-CWDA-2016-00004), DOE also performed sensitivity analyses with Kd values of 0 mL/g and 1 mL/g for saltstone and disposal structure concrete.

Table 1 Kd Values Assumed for Iodine in Cementitious Materials (mL/g)

	Reduced Region II	Oxidized Region II	Oxidized Region III
2009 PA, 2009 Case K, 2013 SA, 2014 SA	9	15	4

In the 2009 PA, DOE assumed that iodine did not sorb onto sandy soil and that it had a Kd value of 0.6 mL/g for clayey soil (Table 2). The clayey soil Kd value was based on site-specific data reported in WSRC-TR-2006-00004. Revised Kd values were assumed in the Case K evaluation based on research in SRNL-STI-2009-00473. In the 2013 SA, DOE added leachate impacted Kd values to their analysis to account for changes in sorption in areas with high concentrations of leachate from the cementitious materials that is highly basic and has a high ionic strength. The leachate impacted Kd values were based on leachate impact factors from SRNL-STI-2009-00473 and were used for any unsaturated soil. The iodine Kd values for subsurface soils were updated again in the 2014 SA based on research in SRNL-STI-2012-00518.

Table 2 Kd Values Assumed for Iodine in Subsurface Soils (mL/g)

	Clayey Soil		Sandy Soil	
	Without Leachate	Leachate Impacted	Without Leachate	Leachate Impacted
2009 PA	0.6		0	
2009 Case K	0.9		0.3	
2013 SA	0.9	0.1	0.3	0
2014 SA	3	0.3	1	0.1

Recent DOE Research

Saltstone Core Analyses

DOE recently analyzed six core samples of field-emplaced saltstone from Saltstone Disposal Structure (SDS) 2A (SRNL-STI-2016-00106). One of the objectives in this research was to validate property values assumed for the SDF PA modeling. As part of these analyses, desorption leaching experiments for I-129, Tc-99, and Sr-90 were performed under both anoxic and oxic conditions. The total activity of these radionuclides in the core samples was also measured. The total activity measured for I-129 was 4.13 pCi/g, which corresponds to 87% of the predicted activity in the core based on the measured Tank 50 salt solution activity. A summary of these analyses was presented by DOE during the April 2016 NRC onsite observation (SRR-CWDA-2016-00053). Additional results from the core sample analyses were subsequently provided to the NRC (R-16-0003). However, this paper did not include any new information on I-129 leaching and the paper stated that analysis of I-129 released from the SDS samples in the experiments described in R-16-0003 is pending.

In the leaching experiments described in SRNL-STI-2016-00106, ground core sample was mixed with leachate and the samples were equilibrated for 7 days. Two groups of samples were considered, one that was leached in a nitrogen atmosphere (anoxic conditions) and one that was leached under oxic conditions. Reverse (desorption) distribution ratio¹ (Rd) values were calculated from the leaching data. In the PA and SA transport models, DOE assumes that the I-129 leaching is controlled by adsorption, rather than solubility, in both oxic and anoxic environments. The researchers therefore expressed the leachate data results for iodine in terms of sorption rather than solubility.

The calculated Rd values for I-129 were between -5 to 4 mL/g. The average measured Rd values were 0.3 mL/g for anoxic environments and -2 mL/g for oxic environments. The lowest possible value for an Rd or Kd is 0, and a negative value is not a physically possible result. The researchers attributed the measured negative values to the uncertainty in the radiochemical analyses for I-129 at the low concentrations observed in these experiments. Based on these

¹ The distribution ratio (Rd), as used in SRNL-STI-2016-00106, is equal to the activity in the solid divided by the activity in the liquid (C_s/C_w). DOE uses the notation Rd, or distribution ratio, to present the desorption data in SRNL-STI-2016-00106 rather than reporting it as a Kd value because Kd values represent the amount of sorption at equilibrium and there was no verification that equilibrium was reached in these experiments. This Rd is not the same as the "retardation factor", which is commonly abbreviated as R.

experiments, DOE concluded that the I-129 Kd for saltstone is <1 mL/g and is potentially 0 mL/g (SRR-CWDA-2016-0005). These values are lower than what was assumed in the 2014 SA.

Subsurface Iodine Kd values

A literature review of subsurface iodine Kd values was conducted in SRNL-STI-2012-00518. The research reviewed in SRNL-STI-2012-00518 included studies of iodine speciation and transformation in SRS groundwater near the F-Area Seepage Basins, the speciation transformation of iodine in contact with SRS subsurface sediments, adsorption and desorption Kd values for iodine, and iodine Kd values as a function of geochemical conditions (i.e., pH, oxidizing or reducing conditions) and the form of iodine added to the system.

The observations from the F-Area Seepage Basins show that iodine exists in the subsurface in that area of the site as iodate and organo-iodine, in addition to iodide, even though the iodine appeared to have been introduced to the basins primarily as iodide. The pH appeared to affect the speciation of the iodine, with a greater iodate fraction existing in elevated pH (pH>6) systems.

Adsorption and desorption Kd values were measured using an organic-rich and organic-poor sediment using iodine that was either initially in the form of iodide or iodate. When iodide was added to the SRS subsurface sediment, negligible amounts of iodate were observed in the aqueous phase. When iodate was added to the sediment, about 20% of the iodine in the aqueous phase was iodide. Due to the very low sediment organic matter concentrations in the sediment, little organo-iodine was observed in the aqueous phase. In all cases, the measured desorption Kd values were higher than the measured adsorption Kd values.

The measured Kd values for iodate were much higher than for iodide. The sorption was also higher for organic rich sediments than organic poor sediments. The sorption was sensitive to pH values and decreased with increasing pH values. The Kd values were similar under reducing and oxidizing conditions for clayey and sandy sediments and were lower under reducing conditions for wetland sediments. The measured desorption Kd values were 4 to 5 times greater than the adsorption Kd values.

In the 2014 SA, the subsurface iodine Kd values for areas not impacted by leachate were revised based on the literature review in SRNL-STI-2012-00518. The new best-estimate values were used for the 2014 SA and were higher than the Kd values previously assumed in saltstone performance assessments (Table 2). The bases provided for increasing the subsurface iodine Kd values included: 1) the more strongly sorbing iodate species exists in SRS groundwater in addition to iodide in some locations (average total iodine in three F area background wells = 15% iodide, 42% iodate, and 43% organoiodine), 2) when iodine was added as iodate, the

measured Kd values were 2 to 6 times greater than when the iodine was added as iodide, and 3) higher desorption (10 to 20 mL/g) than adsorption Kd values were measured.

The subsurface Kd values in the 2014 SA were multiplied by cement leachate impact factors from SRNL-STI-2009-00473 to determine the Kd values for leachate impacted areas. These leachate factors were based on values reported for the Hanford site in PNNL-16663.

NRC Staff Evaluation

Iodine Kd values in Saltstone and Disposal Structure Concrete

The NRC staff agrees that it is acceptable to use the measured Rd values from the saltstone core samples to draw conclusions regarding the Kd values selected for saltstone in the model because the method for determining the Rd and Kd values is essentially the same and both are calculated by dividing the concentration in the solid by the concentration in the water. The difference between the Rd and Kd values is that the Kd values represent a system that is at equilibrium, while Rd values may or may not represent a system at equilibrium. In the iodine leaching experiments performed on the saltstone core samples, nearly all of the iodine had leached from the solid, which indicates that in the experiments there was sufficient time for the iodine to leach and equilibrium was likely reached. The NRC staff also agrees with DOE's conclusion that, based on the core leaching data, the saltstone Kd for iodine seems to be less than 1 mL/g and may be 0 mL/g.

In the TER for the 2009 PA (ML121020140), the NRC staff concluded that the Kd values assumed for iodine for cementitious materials seem to be slightly higher than measured values. There have been no new experimental results for the sorption of iodine on disposal structure concrete under conditions expected in saltstone disposal structures since the NRC made this conclusion. Therefore, the NRC staff continues to conclude that the Kd values assumed for disposal structure concrete in the evaluation case may be higher than is justified.

The NRC is expanding MF 5.04, "Certain Risk-Significant Kd Values for Saltstone" to include the sorption of iodine on saltstone based on the Rd values measured in the saltstone core samples that were lower than values DOE assumed for the Kd in the 2009 PA and FY2014 Special Analysis. The lower Kd values correspond to a more rapid leaching of the iodine from saltstone and, therefore, a larger potential dose. Also, the low sorption of iodine in saltstone may cause the chemical performance of the disposal structure concrete to become more risk significant. If the iodine is not being retained by the saltstone, then the sorption of iodine on the disposal structure concrete likely would control the transport of iodine from the engineered system into the environment, making it more risk-significant. Therefore, the NRC is also expanding MF 6.01, "Certain Risk-Significant Kd Values in Disposal Structure Concrete", to include sorption of iodine on disposal structure concrete. As the sorption of iodine on disposal

structure concrete and saltstone is now included in MFs 5.04 and 6.01, the NRC staff is no longer monitoring these items under MFs 10.04 and 10.06.

Subsurface Iodine Kd values

As part of its review of the "Tank 16 Special Analysis for the Performance Assessment for the H-Tank Farm at the Savannah River Site", the NRC previously reviewed SRNL-STI-2012-00518 and the new proposed subsurface Kd values for iodine presented in SRNL-STI-2012-00518. Although the NRC staff agreed that the potential exists for I-129 released from the tanks to be converted from iodide to iodate and organo-iodide, the NRC staff concluded that the new proposed subsurface Kd values for iodine assumed in the Tank 16 SA are not adequately supported (ML15301A710). This conclusion was based on concerns related to: (i) the potential for iodate to be unstable, (ii) the use of a composite Kd value to represent multiple iodine species, (iii) an apparent disconnect between pH, speciation, and sorption, (iv) the spatial and temporal variability of chemical conditions, and therefore the speciation and sorption of iodine, under field conditions, (v) representativeness of F-Area Seepage Basin aquifer sediments to the SRS Tank Farm far-field environments, and (vi) chemical form of iodine expected to be released from the tanks.

The first four concerns are all directly applicable to the iodine Kd value used in the saltstone analysis. Additionally, the representativeness of the F-Area Seepage Basin aquifer sediments to the SDF far-field environments is also a concern for the performance assessments for the SDF. Variability in the clay and organic matter content of the soils can lead to different amounts of sorption of iodine. Finally, similar to the tank farms, it is not clear what form of iodine is expected to be released from saltstone and it is not clear what effect the chemical form of iodine released from the engineered system has on the speciation of iodine in the natural system. For example, it is indicated in SRNL-STI-2012-00518 that once iodide was added to the organic-poor "North Borrow" subsurface sediment, negligible amounts of iodate were formed. Additionally, due to the very low sediment organic matter concentrations, little organo-iodine was observed in the aqueous phase. These results show that the chemical form in the leachate may be an important factor with respect to the speciation of iodine in the natural system once released from the engineered system. That is, the natural system may not form significant iodate or organo-iodine in all locations on the site so the speciation in the natural system may be more likely to reflect the speciation upon release from the engineered system.

The NRC staff is also concerned that the measured higher amounts of adsorption than desorption may be attributable to an experimental artifact. In these experiments, iodine, in the form of iodide or iodate, was initially adsorbed onto two different sediments, an organic-rich sediment and an organic-poor sediment. The NRC staff is concerned that the species of I-129 adsorbed in the adsorption phase of the experiment would have disproportionately been the species that tend to sorb more (e.g., iodate or organo-iodide). After the adsorption portion of

the experiment was completed, the sediment with the sorbed iodine was placed in contact with a desorbing solution with 0.1 N KCl² to measure desorption K_d values. If the species that sorb more strongly had disproportionately sorbed during the adsorption phase, the sorbed iodine could primarily consist of the more strongly sorbing species and might not be representative of the species present in the aqueous phase in the subsurface. The desorption K_d values measured in these experiments might therefore represent the desorption of the more strongly sorbing species and might not be representative of the overall behavior of all of the iodine species expected to exist in the subsurface. Although very little iodate or organo-iodide was measured in the aqueous phase in the experiments in which the iodine was added as iodide, it is possible that a small amount of the iodide was converted to a more strongly sorbing species. The higher desorption K_d values could therefore be just due to differences in the speciation of I-129 in the original solution versus the speciation sorbed on the sediment.

Also, the K_d value is an equilibrium constant that represents the amount of the species expected to be sorbed under equilibrium conditions and is a function of both the rates of adsorption and desorption. In SRNL-STI-2012-00518, the researchers concluded that the 4 day equilibration time used in the experiments was sufficient for the organic-poor sediment to reach steady state but it was probably insufficient for the organic-rich sediment to reach steady state. This conclusion was based on observations in a different experiment that steady state was reached within four days for the sorption onto sandy soils. The soil samples that were used in this experiment appear to be different samples than were used in the adsorption and desorption experiments, and it is not clear if the results are applicable. Additionally, the fact that the batch studies were not measuring the same value for adsorption and desorption indicates that the measured K_d value may not have been a true equilibrium constant. For these reasons, the applicability of the sorption observed in the adsorption/desorption batch studies to the sorption during transport in the subsurface is not clear. Column experiments to evaluate the effect of the sorption of iodine onto sediment on the transport of iodine might provide more useful information on the expected transport of iodine in the subsurface at SRS.

For these reasons, the NRC staff concludes that the use of the new proposed subsurface K_d values for iodine in the Saltstone PA and Special Analyses is not adequately supported. Because I-129 is a risk-significant radionuclide in the PA and Special Analyses, the NRC staff is expanding MF 7.01 to include the subsurface K_d value for iodine. Because the subsurface K_d value for iodine is now being monitored under MF 7.01, this K_d value will no longer be monitored under 10.09, "K_d Values for SRS Soil".

² The use of a 0.1 N KCl solution is not representative of field conditions, but it is expected that the use of this solution would increase the amount of desorption.

The NRC staff requested additional information regarding the origin of the leachate impacted Kd values in the RAI for the 2013 SA and the 2014 SA (ML14148A153 and ML15161A541). DOE's response to both RAIs was minimal and did not provide a traceable description of the basis for the values selected. The NRC staff additionally asked for the basis for the cement leachate factors from SRNL-STI-2009-00473 during the April 2016 OOV (ML16147A197). In its response to this action item (SRR-CWDA-2016-00060), DOE stated that

*“an update to *Geochemical Data Package for Performance Assessment Calculations Related to the Savannah River Site*, SRNL-STI-2009-00473, is in progress and will be provided to the NRC when available. The document is being revised to incorporate recent documented geochemical results including radionuclide Kd values, solubility values, and cementitious impact factors, and includes a critical evaluation of these values with respect to existing values to assess potential impacts.”*

DOE has not provided any additional basis for the leachate impact factors at this time. Although the assumed value for the leachate impact factor for iodine results in a lower amount of modeled sorption, it is possible that the true amount of sorption on leachate impacted soil could be lower than the assumed amount. The staff cannot assess the acceptability of the leachate impact factor without a traceable description of the basis for the value selected. For this reason, the NRC staff is also expanding MF 7.01 to include the leachate impact factor for iodine and the iodine leachate impacted Kd values.

Follow-up Actions

There are no Follow-up Actions

Open Issues

There are no Open Issues.

Conclusions

The NRC staff concludes that the saltstone core leaching data indicate that the iodine may not be retained significantly by field-emplaced saltstone and the Kd values assumed for iodine for saltstone in the 2014 SA are higher than are justified. Because of the increased risk-significance of the iodine Kd value for saltstone, the NRC is expanding MF 5.04, “Certain Risk-Significant Kd Values for Saltstone”, to include the sorption of iodine on saltstone. The NRC is also expanding MF 6.01, “Certain Risk-Significant Kd Values in Disposal Structure Concrete”, to include sorption of iodine on disposal structure concrete because the Kd values assumed by DOE may be non-conservative and the low sorption of iodine in saltstone may cause the chemical performance of the disposal structure concrete to become more risk significant

The NRC staff concluded that the use of the new proposed subsurface Kd values for iodine in the Saltstone PAs is not adequately supported. The NRC staff also concluded that the basis for the assumed leachate impact factors and resulting leachate impacted subsurface Kd values is unclear. The NRC staff is therefore expanding MF 7.01 to include the subsurface Kd values for iodine and the leachate impact factors and the leachate impacted subsurface Kd values for iodine.

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_____ SRR-CWDA-2016-0004, Rev. 1, "Comment Response Matrix for NRC Request for Additional Information on the Fiscal Year 2014 Special Analysis for the Saltstone Disposal Facility at the Savannah River Site," March 2016. ML16105A033.

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_____ SRR-CWDA-2016-00060, Revision 0, "Action Item Follow-up in Support of U.S. Nuclear Regulatory Commission Onsite Observation Visit April 19 - 21, 2016", June 2016. ML16180A311.

U.S. Nuclear Regulatory Commission (NRC). "Technical Evaluation Report for the Revised Performance Assessment for the Saltstone Disposal Facility at the Savannah River Site, South Carolina," April 2012. ML121020140.

_____ "NRC Plan for Monitoring Disposal Actions Taken by DOE at the Savannah River Site Saltstone Disposal Facility in Accordance with the National Defense Authorization Act for Fiscal Year 2005," Rev. 1, September 2013. ML13100A113.

_____ "NRC Staff Comments and Requests for Additional Information on the Fiscal Year 2013 Special Analysis for the Saltstone Disposal Facility at the Savannah River Site", June 2014. ML14148A153.

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