Rocky Flats Environmental Technology Sites: Actinide Migration Evaluation

Meetings January 8-9, 2001

Advisory Group

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Summary and recommendations for path forward

The Pathway Report is making progress (overview, air, chemistry/-geochemistry, biological, groundwater) An important component will be the strong technical basis with clear separation of basic information from model description and applications to specific RFETS areas and problems The document is building upon the solid basis of the Conceptual Model Report, with careful definition of the technical aspects of actinide chemistry, treatment of misguided simplification approaches to some of the problems (e g ^{KD} usage for Plutonium$^{(Pu)}$ and Americium$^{(Am)}$ groundwater migration evaluation) and collection of documentation of RFETS monitoring It is critical that this document be the technical basis for Sampling and Analysis Plans for RFETS Industrial Area, Protected Area, Process Waste Line, and buffer zone – integrated characterization, D&D remedial actions, and long-term stewardship

Stewardship needs to be more carefully promulgated through the chain of work at RFETS (ER, D&D, and communications)

Uranium$^{(U)}$ sources and old borings data found by Laurie Gregory-Frost and Bob Smith is great We really appreciate the diligence of the RFETS personnel that work on AME projects, especially such efforts to find and integrate historical data to provide a fully textured view for AME

Progress and integration

Land configuration is an area that we are pleased to see being considered in the context of the Pathway Analysis and in relationship to the erosion and water balance modeling efforts It is good to see a contract in place for integrated emphasis at the Sites Participation by the RFETS contractors in this AME meeting was an important initial step in making sure that actinide migration evaluation issues are fully coupled We are very interested in hearing about their plans, procedures and progress during future AME advisory meetings

We feel that Sites Management is positive and proactive in its approach to stewardship and stakeholder participation The AME advisors view this as a critical component of successful closure of the RFETS Sites

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Results and Discussions

Updates on the Water Balance Modeling – Dayton

As part of developing a detailed design basis for closure activities RFETS is conducting a Sites-wide Water Balance (SWWB) Project The SWWB will develop a management tool in the form of a physically-based, integrated model This model may be applied to various RFETS Closure decisions

Following development of the conceptual hydrologic model for RFETS, a draft, comparative study was completed to select an appropriate model code The MIKE SHE software was selected as the best system to meet the project objectives

Most of the Sites environmental data collected during the calibration period has been compiled Data analysis at this stage of model calibration consists of preparation of data inputs (i e model drivers) and calibration targets Listed below are the status of the wind, surface water and groundwater data inputs

- Wind data from the perimeter wind and air quality monitoring stations maintained by CDPHE were analyzed
- All surface water data have been compiled, and responses for the entire year at all gauging stations have been plotted against event duration and magnitude
- All quarterly groundwater level data up to and including 10/00 have been collected, and Hermit continuous water level data have been retrieved, but not been checked for accuracy and completeness

Update on the Erosion and Sediment Transport Modeling –Wetherbee

There was a discussion of representation of rip/rap drop structures in the sediment transport model, HEC-6T Previous modeling efforts used two models and averaged the results Subsequent review comments suggested this was arbitrary The most recent modeling represented the rip/rap structures as a serrated configuration with and without channel erosion Computed sediment yields are now between those of the original drop structure modeling results and those from modeling the channels without drop structures The flow velocities now look reasonable, and they are examining the maximum velocities for the 100 yr event, Some of the velocities are on the order of some 20 ft/sec

The relative proportions of channel and hillslope sediment yields are now being studied Channel erosion is now integrated into all sediment yield models Some interesting features have been found that were not included in previous models (e g stock pond in Noname gulch)

Efforts are underway to evaluate taking out some ponds at closure Changes in simulated sediment yield with and without Pond C-1 are being investigated Simulated sediment yields increased and additional analyses are needed for actinide transport evaluation Removing all but terminal ponds in Walnut Creek resulted in a 30% increase in sediment yield Removing all but

Pond B-5 resulted in a 50% to 90% increase in sediment yield With no detention ponds, sediment yields were estimated to increase by about 60-94%

Wright Water Engineers personnel summarized future efforts as follows Greg Weatherbee will program some future scenarios and continue modeling channel processes, Ian Paton will model actinide transport, Chris Hawley will conduct additional erosion simulation, and Margaret Herzog will be writing programs to automate linking the erosion, sediment transport, and actinide transport calculations

Leonard Lane suggested that they develop data management routines to track changes in erosion modeling through the sediment transport calculations and then through the actinide transport calculations Greg Wetherbee responded that they are working to accomplish this automated linking but have not completed it and referred to the work by Margaret Herzog Greg Wetherbee also stated that they are working to link QA/QC to the data management programs and will get together with personnel conducting the land use/configuration work next week

In a follow up, Dave Clark suggested they carefully examine reversals in sediment yields with changing assumptions to see if they are reasonable and consistent across all watersheds Leonard Lane suggested comparing their reservoir sediment routing results with some standard reservoir trap efficiencies as a generalized test of the sediment transport modeling

Wright Water Engineers personnel stated that they hope to have the sediment transport modeling and revisions in place within the next 4-6 weeks and then move forward to the actinide evaluations

Overview of the Pathway Analysis Report – Paton

A general overview was presented on the general outline for the pathway report The general approach to the pathway report is to follow the general outline of the conceptual model document that has been used effectively as a tool for public communication and involvement The amount of material is so large, that it was wisely decided to break the report down into two documents The primary document will be a summary report outlining the general philosophy, conceptual model, and scientific understanding of actinide chemistry, geochemistry, and transport that affect actinide migration at RFETS The secondary document will be a sizeable technical appendix that covers the following 1) background, 2) actinide geochemistry, 3) RFETS actinide data, 4) pathway analysis based on measured data, 5) pathway analysis based on modeled data, 6) a comparison of measured/modeled data, 7) links to comprehensive risk assessment, and 8) overall summary and comparison of major pathways

We strongly agree with this two-report philosophy We also suggest that the primary document should remain relatively short (50-100 pages) and should be produced with the assistance of a professional editor, and make good use of color graphics This should be viewed as a high-profile glossy marketing

document for use in marketing the underlying scientific understanding of actinide migration on which future decisions will be based An example of what we have in mind can be found in the recent volume of *Los Alamos Science*, *26*, **2000** "Challenges in Plutonium Science" We realize that this represents a sizeable effort and cost, but also suggest that such a document will be more readable and useable by the general public, stakeholders, regulators, and political representatives

The technical appendix can and should contain the bulk of the technical information, and is expected to get quite large We also suggest that appendix section 3, "Measured RFETS Actinide Data" be moved forward to follow the introduction and background (section 1) In this way, the major discussion of actinide geochemistry can follow the discussion of measured data, and our understanding of actinide geochemistry can refer to the measured data

Actinide Pathway Sources and Surface Water Pathway (status and path forward) – Paton & WWE staff

A major focus of discussion by Ian Paton in his presentation was the need for a more quantitative aspect in the Pathway Report, especially in the surface water transport analysis This had been the major concern of Tom Hakonson in his comments (10/17/99) The original intent was to develop a qualitative analysis as a guide to further AME activities R&D However, appropriate quantitative data with references are needed to provide a convincing basis for choice of major versus minor pathways and the relative importance of the subpathways within these two classifications (major versus minor) Such a quantitative analysis would be important in choice of topics and the relative emphasis on these in the final report However, to develop a useful quantitative analysis requires more data on actinide behavior in neutral and basic solutions in different environmental media This includes good knowledge of the nature of the actinide species present and the thermodynamic and kinetic parameters of the sorption to particulates and surfaces, colloidal formation, migration (dissolved and colloidal), etc

It seems that more emphasis should be devoted to defining the different behavior patterns of U, Pu, and Am U is likely to be in the VI oxidation state which would be associated with a significant solubility as simple, molecular sized species (e g , $UO_2(OH)^+$, $UO_2(CO_3)_3^{4-}$), etc Americium would be present in the trivalent state with a lower molecular species solubility than U as hydrolysis is the major speciation pathway unless fluoride concentrations are unusually high Plutonium would be, predominately, in the IV oxidation state This results in a very strong hydrolysis reaction to form $Pu(OH)_4$ which ages to the even more insoluble PuO_2 in H_2O There is a small solubility of the PuO_2^+ species but its concentration is controlled by the insolubility of $Pu(OH)_4$ and is limited to ca 10^{-8}M Consequently, the majority of Pu transported in surface water is present as intrinsic colloids of PuO_2 in H_2O or as pseudo colloids in which PuO_2 is sorbed to other mineral (e g Fe_2O_3) and organic (e g humic) colloids

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This difference is very important to recognize if quantitative analysis (i e , modeling) is to be applied to the Pathway Report U data can be used with models for soluble species (e g RESRAD) but such models are not applicable to analyses of Am or Pu behavior in surface water pathways Use of such soluble species models for Am(III) and Pu(IV) could be successfully challenges legally as they are scientifically incorrect

Pathway Analysis Report, Section 2 Geochemistry

The Pathway Report is well done It discusses most issues well and gives pertinent references The discussion on the merits and limitations of K_D values usage should be useful to other Sitess in their reports as K_D's are valuable when used within their limitations The Resume on pages 2 2-2 7 is valuable and should be well cited in the future

There are several minor and one major concern First the minor ones On page 2 10, at the bottom, in the discussion of aqueous complexes, hydrolytic species should - *must* -be included Presumably, they were omitted because of the assumption that they are insoluble, however, in contact with insoluble $Pu(OH)_4$ are the soluble $Pu(OH)n^{(4-n)+}$ species where n=1-3 Also, the most soluble species in neutral/basic systems are the PuO_2^+ species such as $PuO_2Cl_2^-$ Am^{3+} has much more $Am(OH)^{(3-n)+}$ present and these species usually are quite significant in the net solubility It is stated that complexation increases the solubility - true but oxidation of Pu from IV to V is much more important than complexation for increasing Pu concentration in neutral/basic waters On page 2 19, Fig 2 4-1, why are all the species Pu^{4+}(except a little Pu^{3+}at low pH)? It is very well established that in natural and sea surface waters, $Pu(V)O^+$ is present at many orders of magnitude higher concentrations than any Pu(IV) species Also Pu(III) is present only in very acidic or highly reducing solutions I suggest getting a more recent diagram than one from 1985

A discussion of K_D values emphasized the importance of recognizing the correct models to use for actinide species K_D values may represent thermodynamic (reversible) binding of ions to anionic Sitess on solids or extractant ligands soluble in organic phase However, also, they may reflect sorption of insoluble species to solid surfaces, colloidal sorption to surfaces, precipitation of insoluble species, etc , all of which may be irreversible reactions Rarely are measurements done by methods which define the species involved or which measure the kinetics of sorption/desorption involved in the K_D values Without such knowledge, K_D values cannot be used in models which require species reversibility For example, without understanding the kinetics related to the K_D measurements, models using multiplate sorption/desorption for migration are scientifically invalid Consequently, based on experimental data correlated with speciation calculations using stability constant, solubility products, the redox speciation calculations both for RF natural waters as well as other Sitess (Hanford, INEEL, sea water, fresh water lakes, etc), U in near surface oxic waters is present at $U(VI)O_2^{2+}$ species In this oxidation state, U can be present

in concentrations of 10^{-6}-10^{-8}M as carbonato and mixed hydroxy/carbonato species In anoxic waters, it would be present as U(IV) and this state is highly insoluble (# 10^{-12}M) Consequently, in oxic waters, U behavior can be modeled as soluble species and K_D values can be used

By contrast, Am exists as Am(III) in both oxic and anoxic waters The hydrolysis of Am(III) is very strong so the soluble Am(III) is very low relative to colloidal species K_D values, relevant to behavior of soluble monocationic Am(III) species are irrelevant for modeling colloidal Am(III) migration

Plutonium favors Pu(IV) in oxic, natural waters Like Am(III), the hydrolyzed $Pu(OH)_4$ is extremely insoluble but sorbs to colloidal material Pu(IV) does have a redox equilibrium with more soluble $Pu(V)O_2^+$ but the latter, in the presence of $Pu(OH)_4$, the concentration of Pu(V) is limited to #10^{-8}M The concentration of insoluble Pu(IV) species sorbed on and transported by colloids can be #10^{-6} M Consequently, modeling of Pu in natural waters must focus on transport of colloidal Pu(IV) and use of K_D values for soluble Pu equilibria behavior is not acceptable scientifically

Pathway Analysis Report, Biological Pathway

Biological pathway report dealing with the uptake by plants and animals needs to be revised and presented in a concise and coherent manner with supporting documentation showing the soil concentration and bioavailability and the role of microorganism in regulating the bioavailability to higher plants For example, the report should include the concentrations of Pu, Am, and U in the soil, in plants, and animals This section should high light the mechanisms of uptake of the actinides from RFETS soils by plants based on the studies conducted at the Sites

The new section on TA - 1 6 3 Microbiology should address the presence, abundance and distribution of microorganisms at RFETS and its potential role in the mobilization or stabilization of U, Pu, and Am Briefly discuss the role of soil and rhizosphere microorganisms in regulating the bioavailabilty of actinides to higher plants

The section TA - 2 6 Microbiological Transformations of the report should address the key microbial processes which affect the mobility and stability of actinides as well as the long-term-management and stewardship of the Sites It should discuss the microbially mediated redox reactions and how it affects the dissolution and precipitation of Pu, and U under wet and dry cycles, bioaccumulation and biosorption of actinides resulting in the immobilization or mobilization as biocolloids in porus media, biotransformation of actinide-organic and inorganic complexes and the fate of released actinide

This section of the report should be integrated with the TA-2 Pu, Am, and U Geochemical, Transport Processes section and should take advantage of the background information discussed in the report which are pertinent to microbial transformations of Pu, Am, and U

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Industrial Area Sampling and Analysis Plan status and schedule – Serreze

The sampling plan describes approaches to determination of the contamination of the Industrial Area D&D aspects looks like a good plan, but ER needs to be more strongly integrated In the ER plan, in particular, background and differentiation of background from contamination is used extensively, however how background will be determined is not defined For example, it is well documented that natural U is present across the Sites and variable in concentration, so a background cannot be generally defined The AME Advisory Group needs to continue to be updated on current status, data, interpretations and plans for underbuilding contamination and old process waste lines

Uranium sources, transport and disposal – Gregory-Frost

The project to examine historical data for insight into the potential U source term associated with old and new Sites Ponds has progressed rapidly This is largely due to the hard work and dedication of Laurie Gregory-Frost Historical U analyses were presented for soil and pore-water samples analyzed in 1993-1994 These analyses were performed following a remedial action at the Sites Ponds to remove the sludge (pondcrete) and liners Some samples were taken from directly under the ponds, while others were from the adjacent hillside In general the U concentrations found in and around the Sites ponds were very low, and in the pCi/g (soil) or pCi/L (water) range None of the soil samples exceeded the Tier I action level, and pore-water samples only exceeded action levels at three sample locations All three of these were located within the Sites pond boundaries Most important is the fact that the soil cores were sampled all the way down to the bedrock layer, and in no case was a large deposit of U observed Soil samples were all within the 1-60 pCi/g range, while pore-water samples were all within the 1-3,700 pCi/L range Recall that the natural background U in Rock Creek is on the order of 1,200 pCi/L This is a very small amount of U

These historical data go a long way toward establishing the magnitude of the U source term under the Sites pond as a result of past Sites activities It appears that there is in fact, only a small quantity of U present This is consistent with the geochemical modeling results of Ball (2000) that suggested that groundwater samples near the Sites ponds were all under-saturated with respect to common U solids Therefore, the observed retardation of U relative to nitrate is more consistent with sorption/desorption processes This is also consistent with our expectations for U geochemical behavior, namely that it will be relatively soluble and mobile under the soil and groundwater conditions at RFETS The fact that there is only a small amount of U present beneath the Sites ponds suggests that the reactive barrier presently installed downslope of the Sites ponds should continue to capture and remediate U as an ancillary role to the treatment of nitrate

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Stewardship – Shelton

We had a very insightful and illuminating discussion with Kaiser-Hill senior management on long-term stewardship It was useful to understand that management acknowledges that some actinides will remain on the Sites, and that the South Interceptor Ditch and ponds will likely remain at Sites closure We like this view from the Senior Management of Kaiser-Hill, but are concerned about how to convey this stewardship concept and approach down to workers For example, how do we keep balance in areas like decisions on Old Process Waste Lines and under building contamination The Advisory Group supports use of the long-term Vulnerability Assessment as an integration process This will enhance perception and accomplishment of early emphasis on stewardship, and alignment with the stakeholders The Sites believes it is built into documents, but maybe not as explicitly as required and fully integrated to end-state

The AME Advisory Group will continue to incorporate this perspective into our activities and evaluations to make sure we are also communicating effectively

Documents provided to advisory group

Meijer, A (1990) Yucca Mountain Project Far-Field Sorption Studies and Data Needs
Ian Paton – Pathway report activity viewgraphs
Martha Hyder – air transport pathway viewgraphs
Pathway report section 3 viewgraphs and maps
Comments on section 2 of technical appendix
Comments on Hersman's text by AJ Francis
R Smith viewgraphs on groundwater pathway progress
Draft Industrial Area Characterization Schedule
Laurie Gregory-Frost -- Sites evaporation ponds soil and water sampling viewgraphs
Ableson stewardship viewgraphs
Summary of NRC August 2000 report on stewardship

Documents and information requested for advisory group

Meyers/McKenna presentation and analysis papers on smart sampling, IA white space

Requests for Future Presentations and Information

Land Configuration team – need early plan presentation and updates on progress, constraints and concerns often after that
SAP continuation with team, not separate representative each time

Participants in AMS technical meetings

Name	Organization
Greg Choppin	FSU
David Clark	LANL
David Janecky	LANL
Lane Leonard	Tuscon
A J Francis	BNL
Anne Kersting	LLNL
Chris Dayton	K-H
Greg Wetherbee	WWE
Ian Paton	WWE
Mike Peters	RMC Consultants
Larry Hersman	LANL
Russell McCallister	DOE/RFFO
Lynn Kidman	IT-Los Alamos
Chris Hawley	IE
John Anthony	Parsons
Bruce Curtis	Parsons
Martha Hyder	Radian
Susan Serreze	Acradia
Laurie Gregory-Frost	E2
Bob Nininger	K-H

Future Meetings

March 27 -- advisors conference call
April 30-May2 – third quarter Sites meeting
July 23-25 – fourth quarter Sites meeting

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