

Evaluation of EBS Processes:  
Near field geochemical environment,  
Waste form degradation/mobilization,  
EBS transport

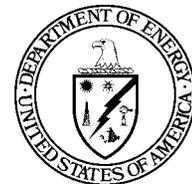
Presented to:

Nuclear Waste Technical Review Board  
Performance Assessment Panel  
Albuquerque, New Mexico

Presented by:

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Duke Engineering and Services  
Las Vegas, Nevada

April 23-24, 1998



U.S. Department of Energy  
Office of Civilian Radioactive  
Waste Management

# Outline

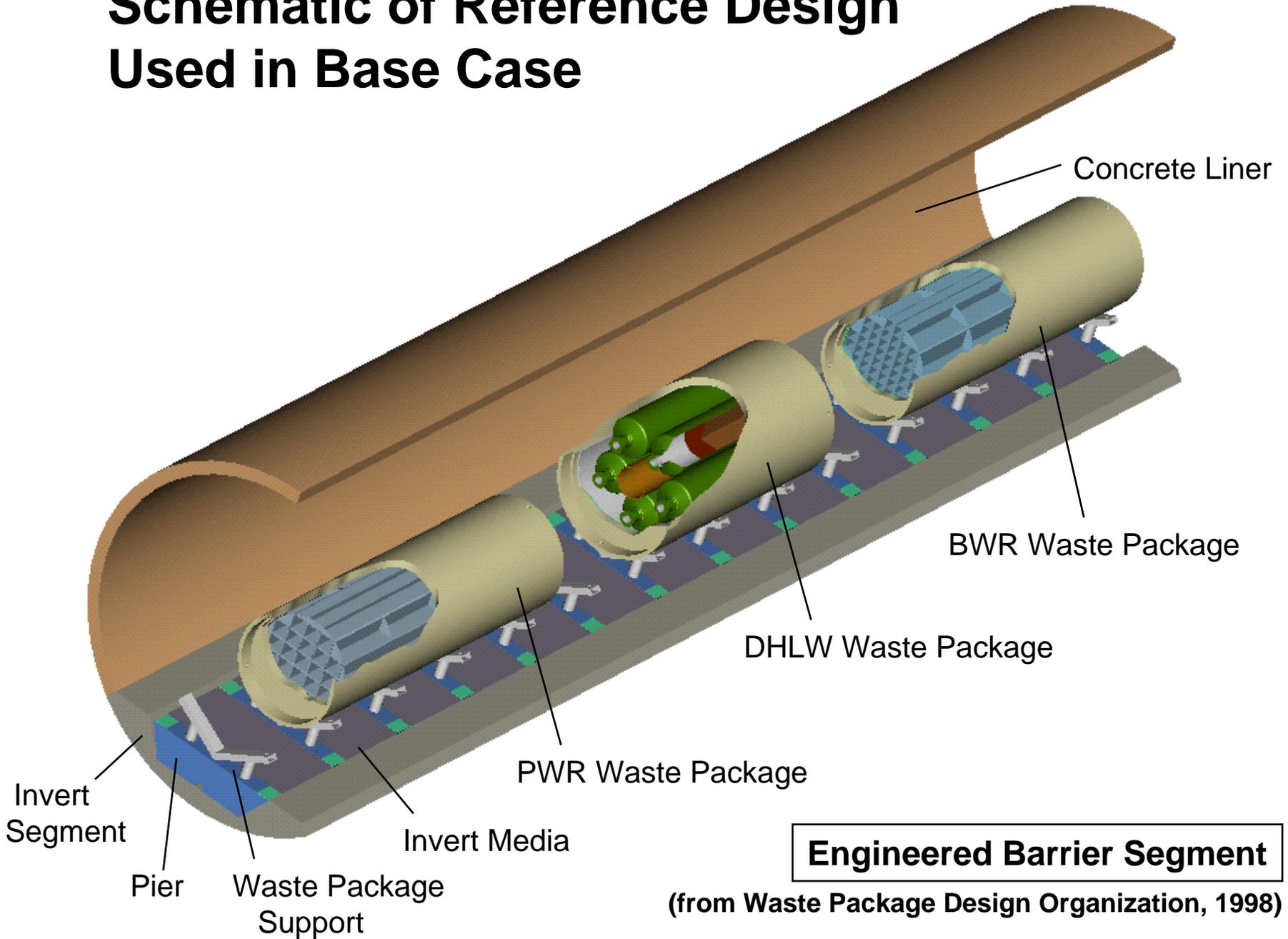
- **Near Field Geochemical Environment**
  - **Conceptual Models**
  - **Bases for NFGE Models**
  - **Results from NFGE Models**
- **Waste Form Degradation**
  - **Conceptual Models**
  - **Bases for WF Degradation Models**
  - **Results from WF Degradation Models**

# Outline

(Continued)

- **Radionuclide Mobilization**
  - **Conceptual Models**
  - **Bases for WF Mobilization Models**
  - **Results from WF Mobilization Models**
- **Engineered Barrier System Transport**
  - **Conceptual Models**
  - **Bases for EBS Models**
  - **Results from EBS Models**

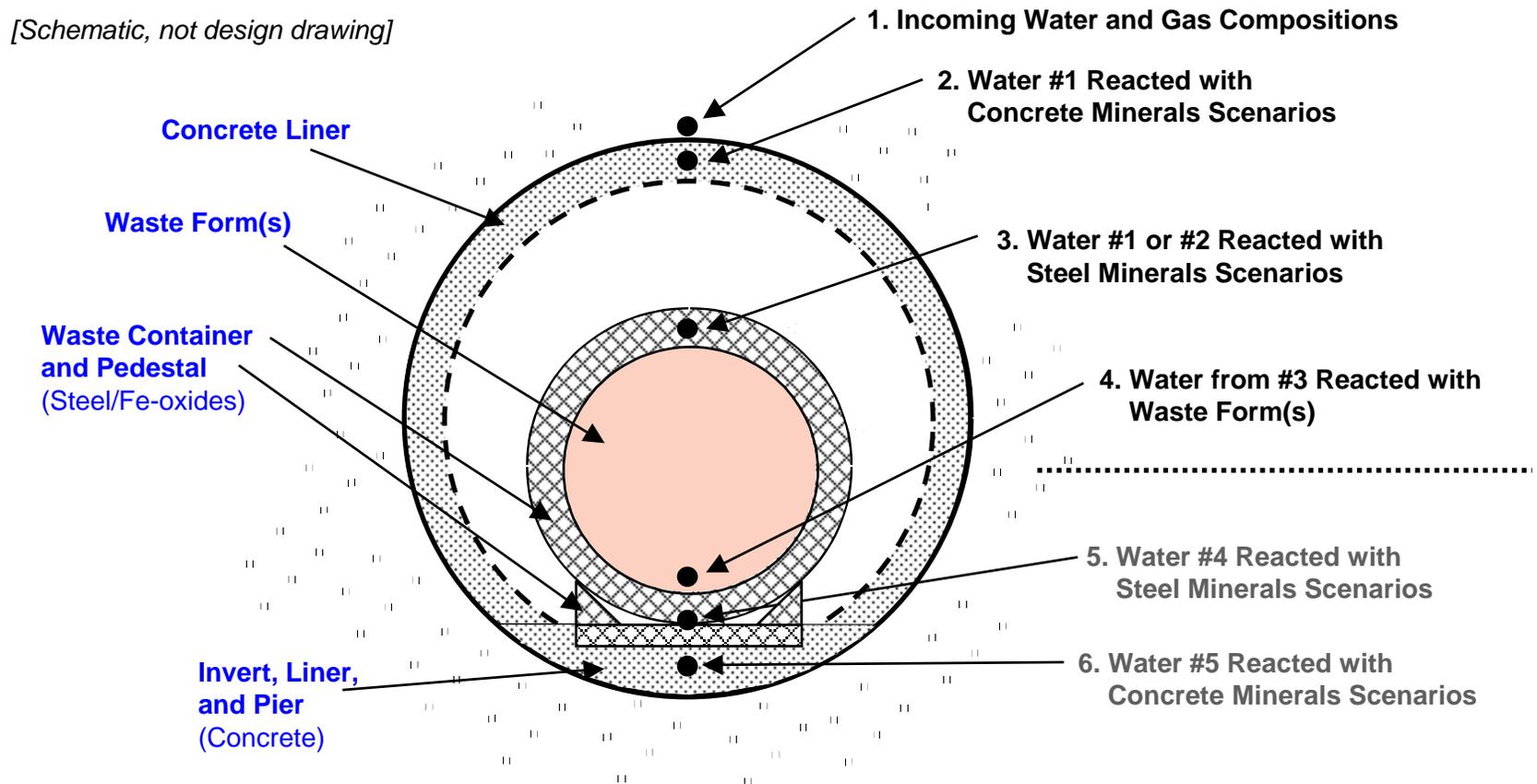
# Schematic of Reference Design Used in Base Case



# NFGE Conceptual Model

- **Discretize the EBS to evaluate**
- **Scenarios defined based on thermal conditions**
- **Locations defined based on discrete locations within the EBS**
- **Evaluate gas and water compositions at the various locations within the EBS**

# TSPA-VA Base-Case Near-Field Geochemical Environment

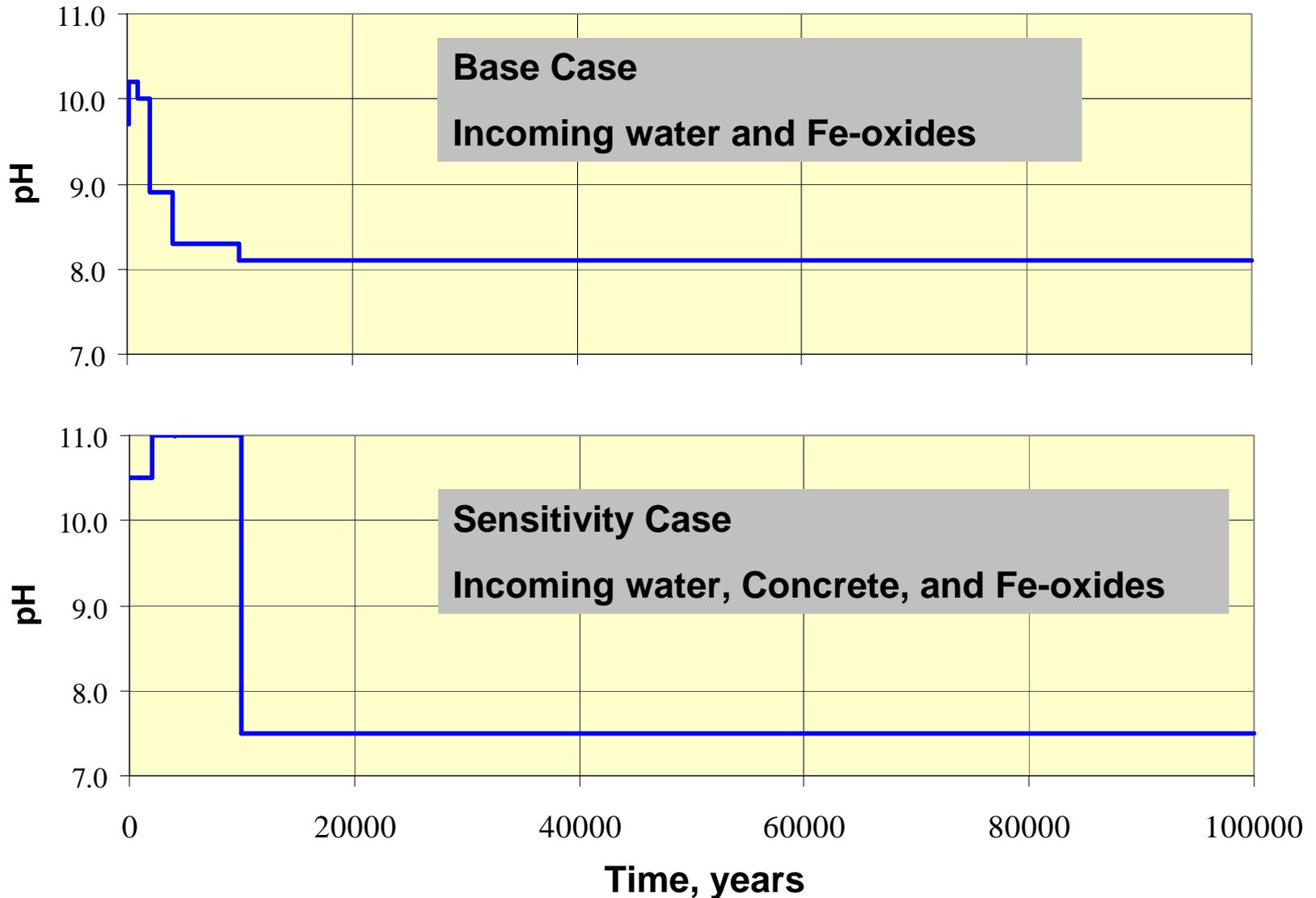


Schematic Representation of Materials Included for Base-Case NFGE and Locations along a Conceptual Pathway for Evaluation of Water Compositions [modified from M&O, 1998 B00000000-01717-2200-00200].

# NFGE Abstraction Summary

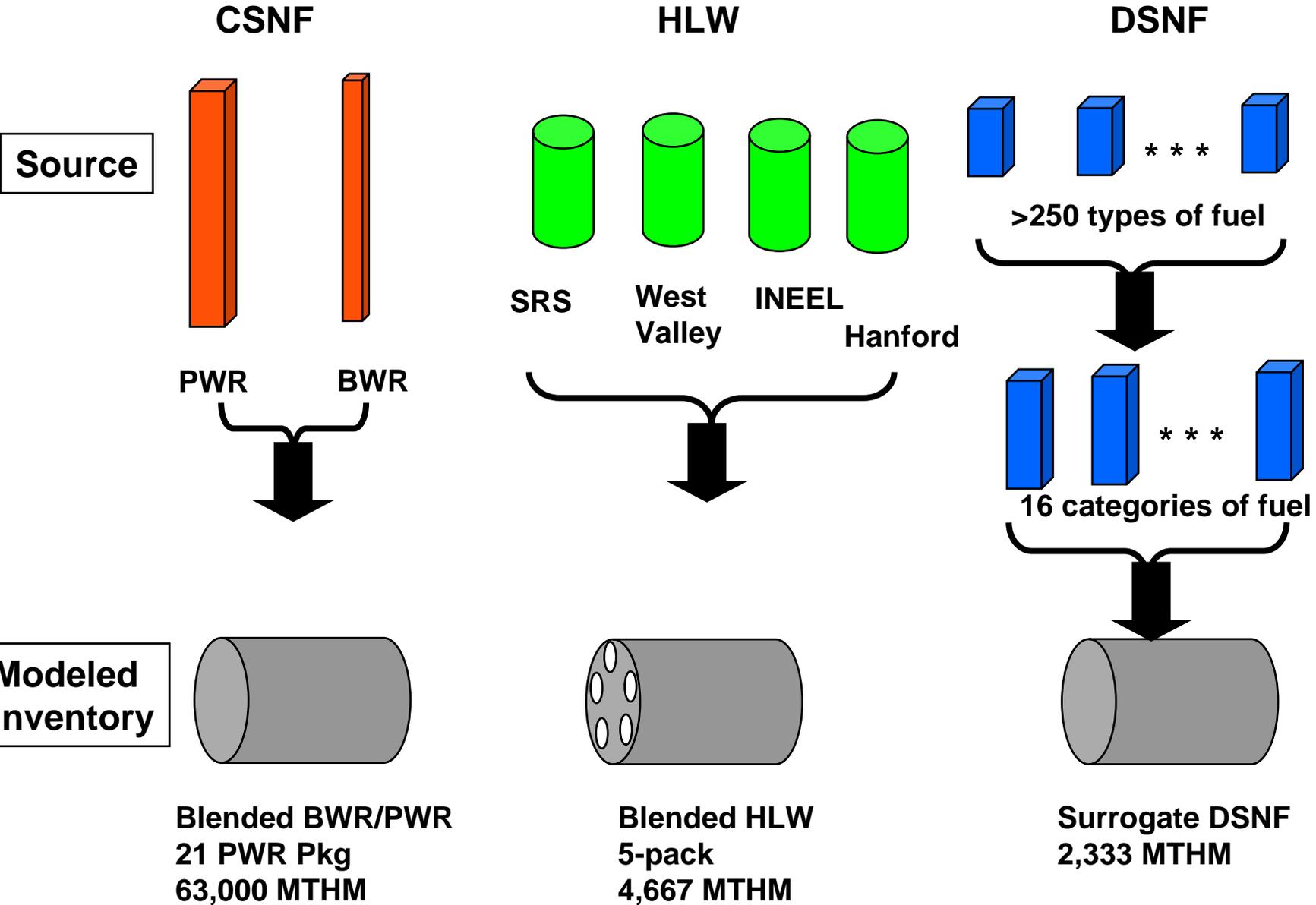
- **Develop NFGE Gas and Water Compositions as  $f(t)$** 
  - Input: gas flux & air-mass fraction from 2-D Mtn Scale TH results
  - Input: air composition ( $pO_2$  and  $pCO_2$ ) from pore-gas and single heater test data
- **Calculate NFGE Water Composition as  $f(\text{location}, t)$** 
  - include thermal effects on incoming water (boiling,  $pCO_2$ )
  - include in-drift reactions
  - include in-package reactions with spent fuel
- **Output: pH,  $\Sigma CO_3^{-2}$ , and I (ionic strength) as  $f(t)$**

# pH of water flowing into waste package



[Revised 20-Apr-98, NFGerev01reslt.ppt]

# Waste Form Inventory Abstraction



# **Waste Form Degradation/Radionuclide Mobilization Conceptual Model**

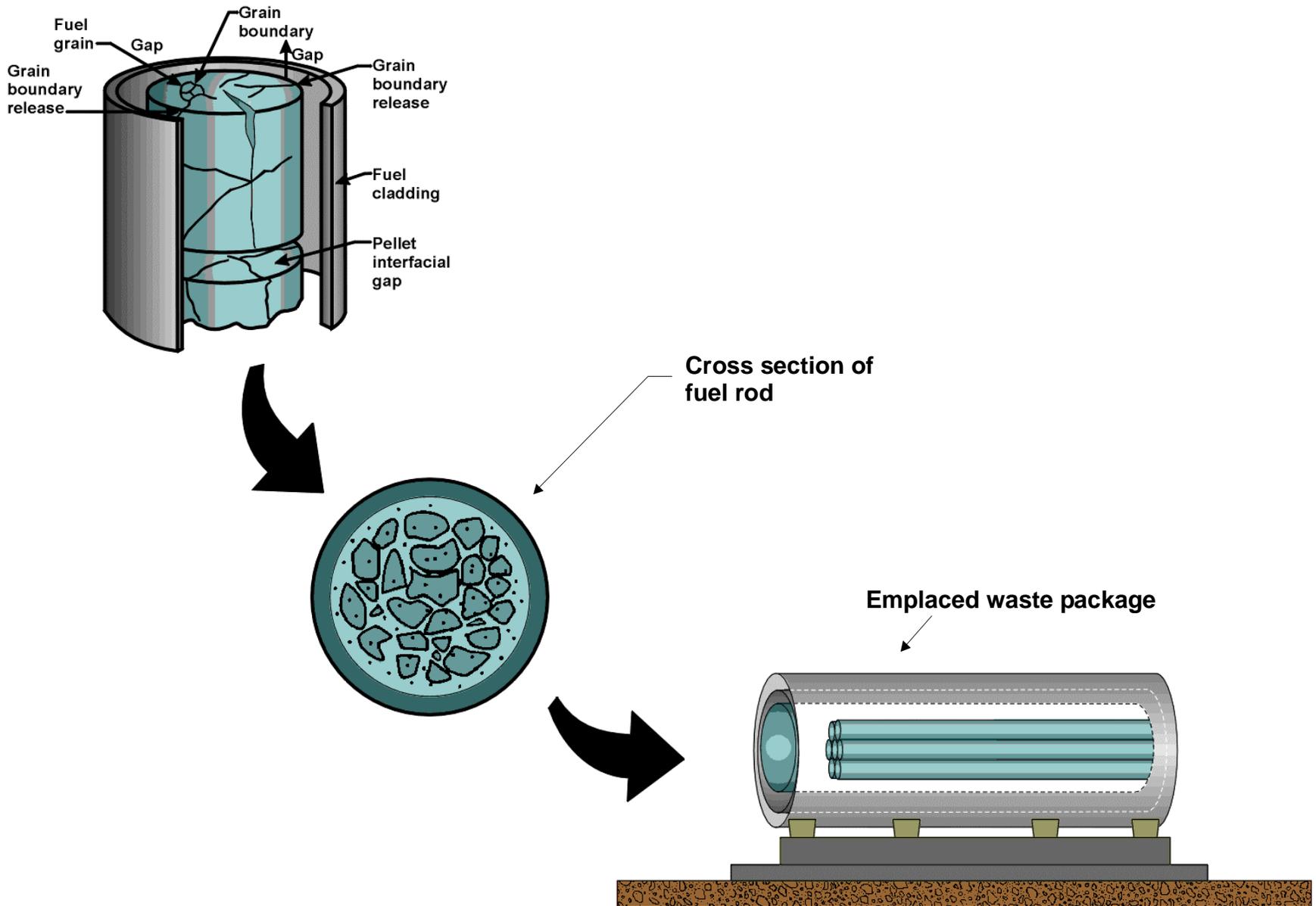
- **Assume waste forms exposed to the drift environment upon failure of the waste package and cladding.**
- **Assume water films adsorbed on porous alteration product layers provide aqueous conditions**
- **Waste form degradation is represented by an “Intrinsic Dissolution Rate” equation**
- **Radionuclides are considered potentially available for mobilization congruent with this dissolution**

# **Waste Form Degradation/Radionuclide Mobilization Conceptual Model**

**(Continued)**

- Mobilization of highly soluble radionuclides at this dissolution rate, into either diffusive or advective EBS transport**
- Most radionuclides are mobilized at aqueous solubility limits**
- A preliminary representation of aqueous concentrations limited by secondary phase formation has been prepared, but is not in the initial base-case**

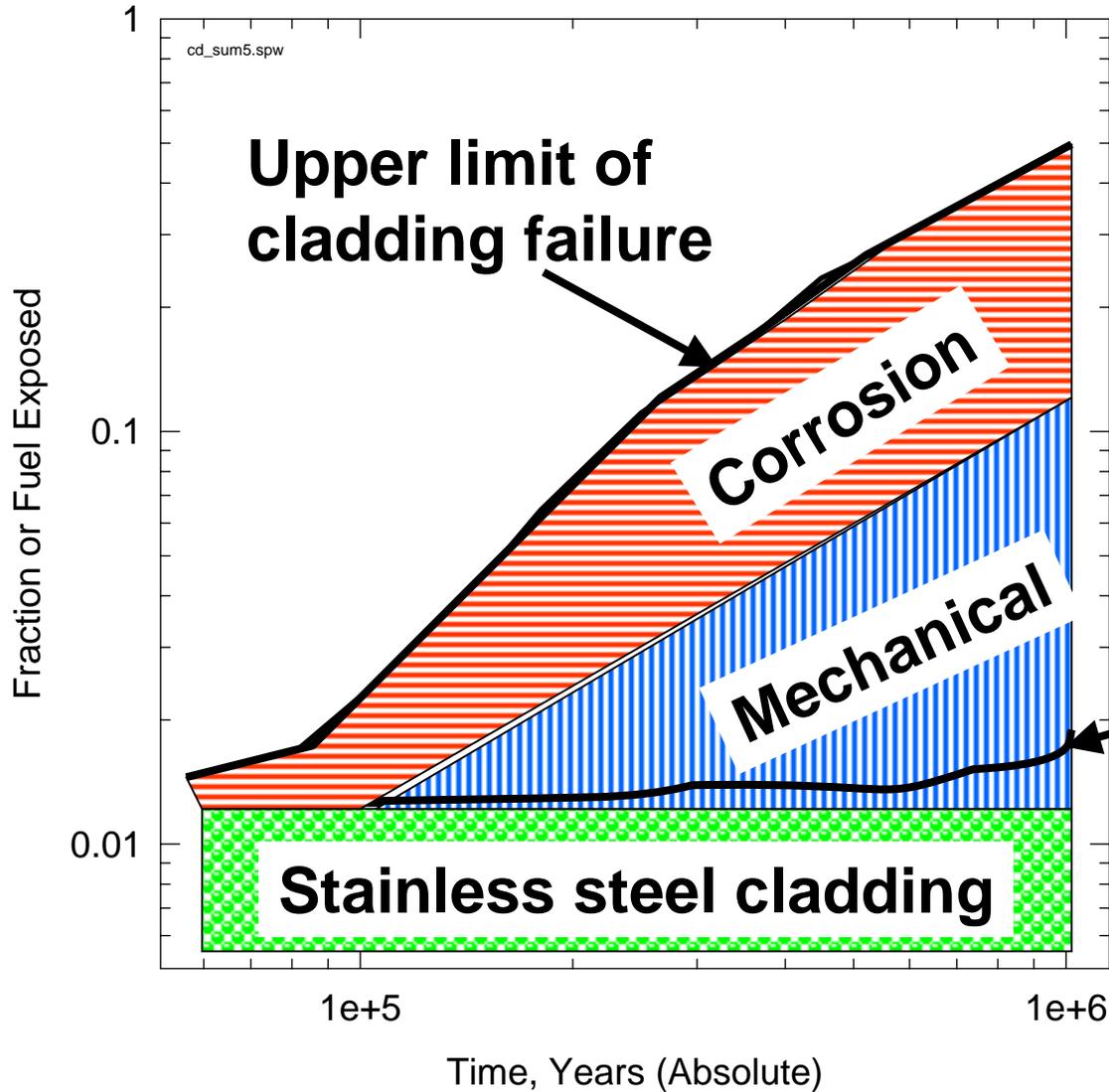
# Waste Form Schematic



# Failure Modes in Cladding Model

- **Juvenile cladding failure**
  - early time failure of cladding
- **Stainless Steel cladding failure**
  - assumed to fail at time of waste package failure
- **Creep (strain) cladding failure**
- **Mechanical failure**
  - due to rod breakage from rockfall
- **Corrosion of cladding**
  - corrosion model similar to C-22 corrosion

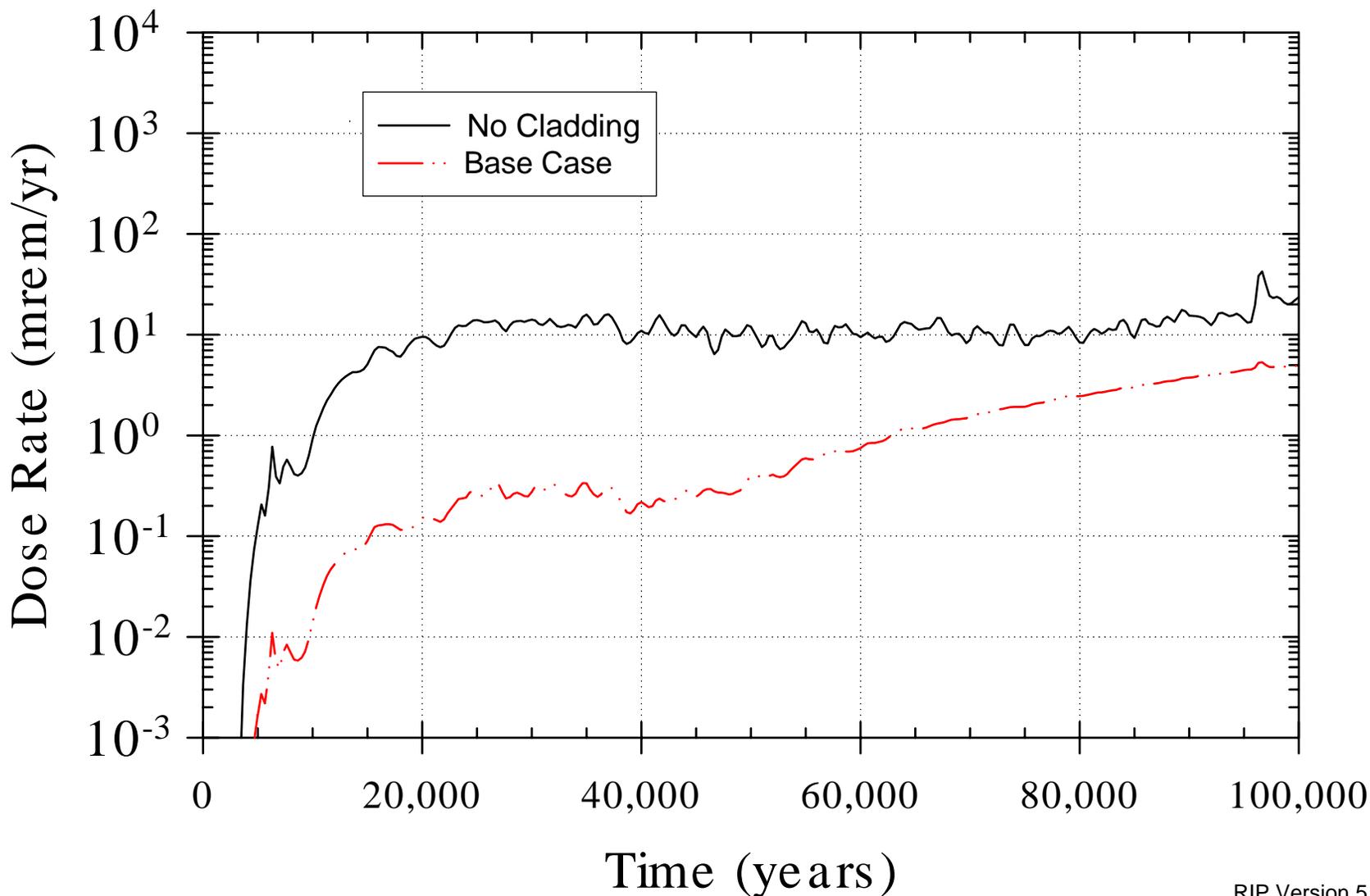
# Cladding Degradation



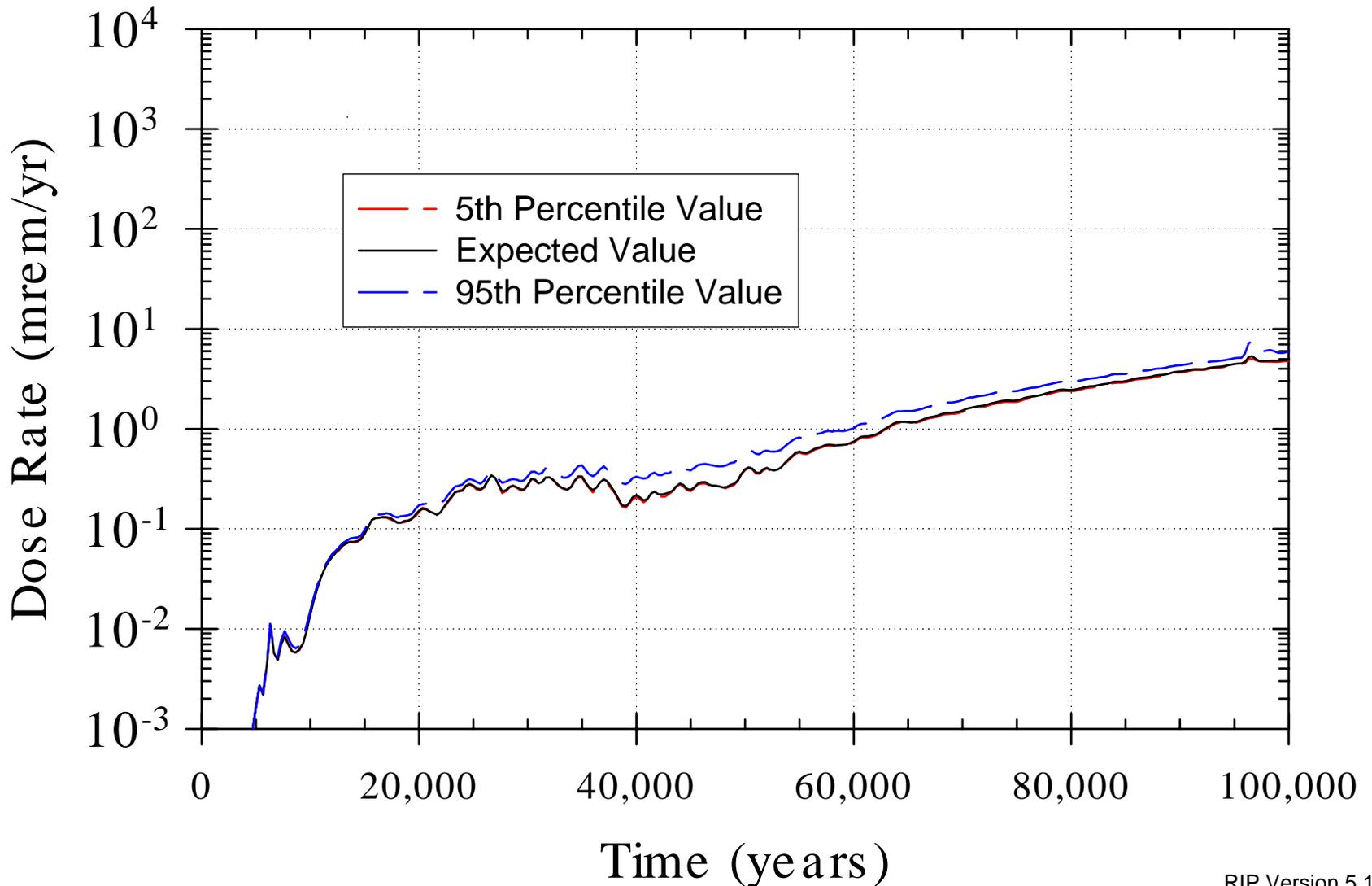
# Base Case vs No Cladding

## 100,000-yr Expected-Value Dose-Rate History

All Pathways, 20 km



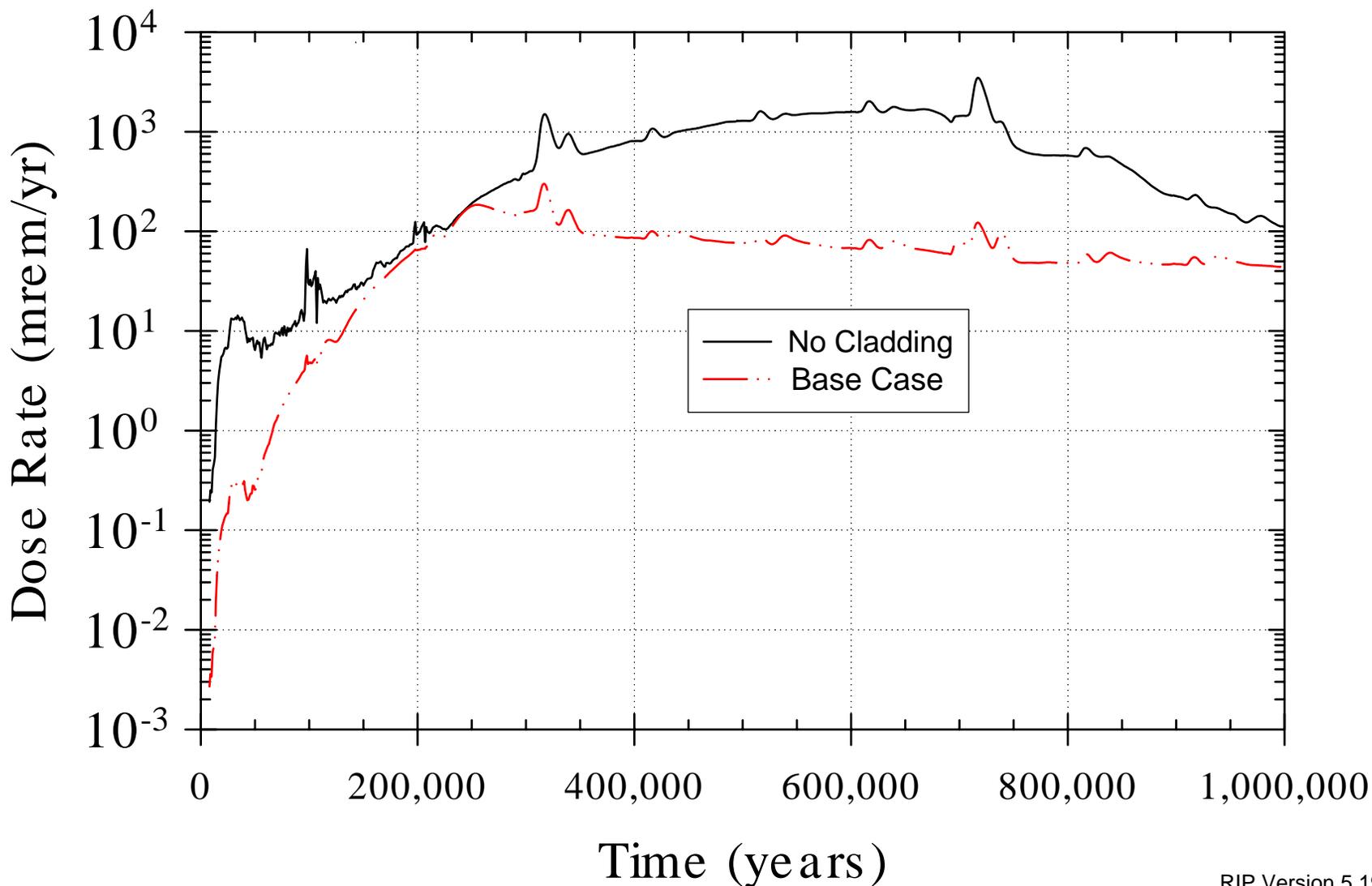
# Cladding 5th & 95th Percentile 100,000-yr Dose-Rate History All Pathways, 20 km



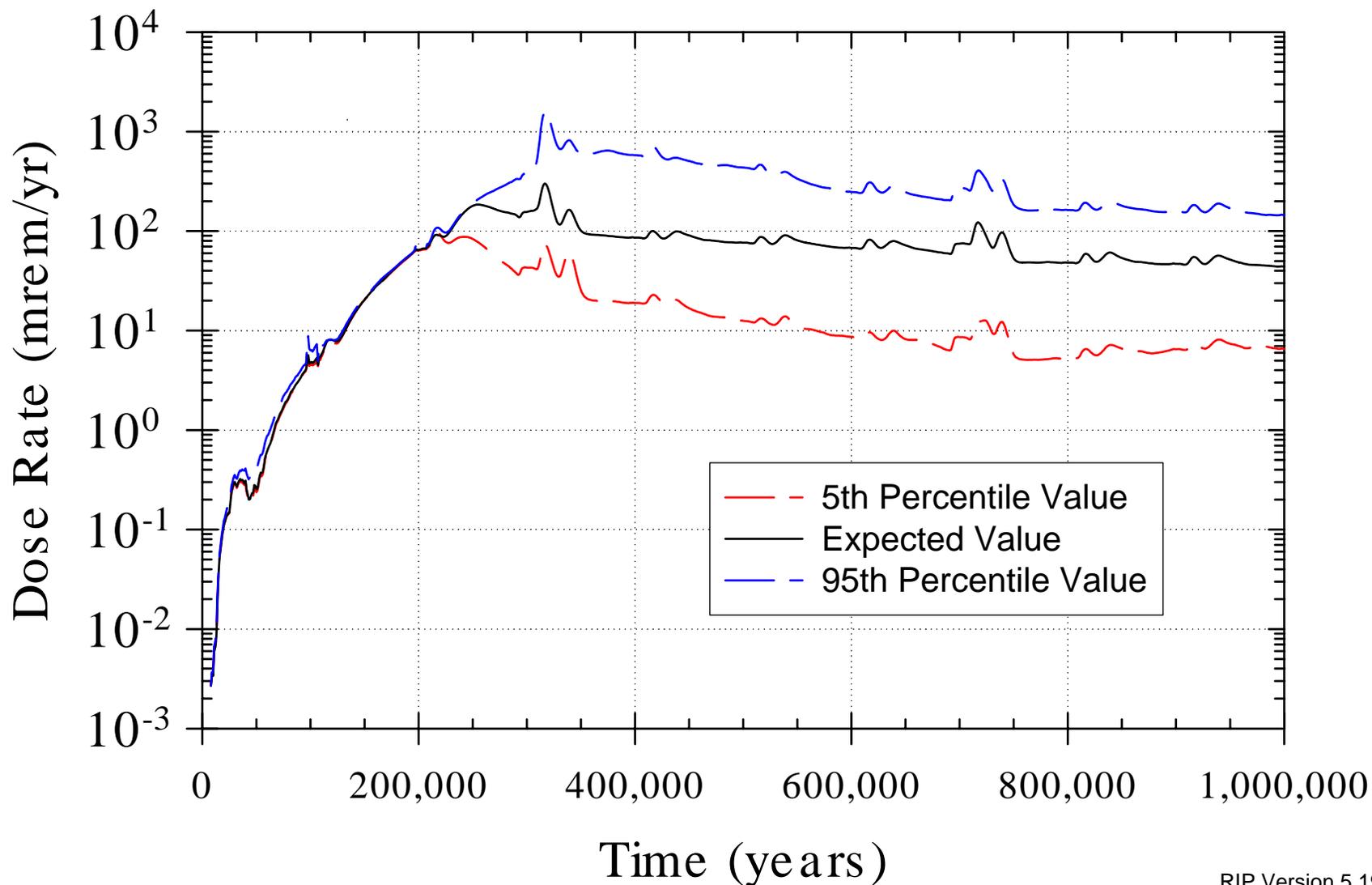
# Base Case vs. No Cladding

## 1,000,000-yr Expected-Value Dose-Rate History

### All Pathways, 20 km



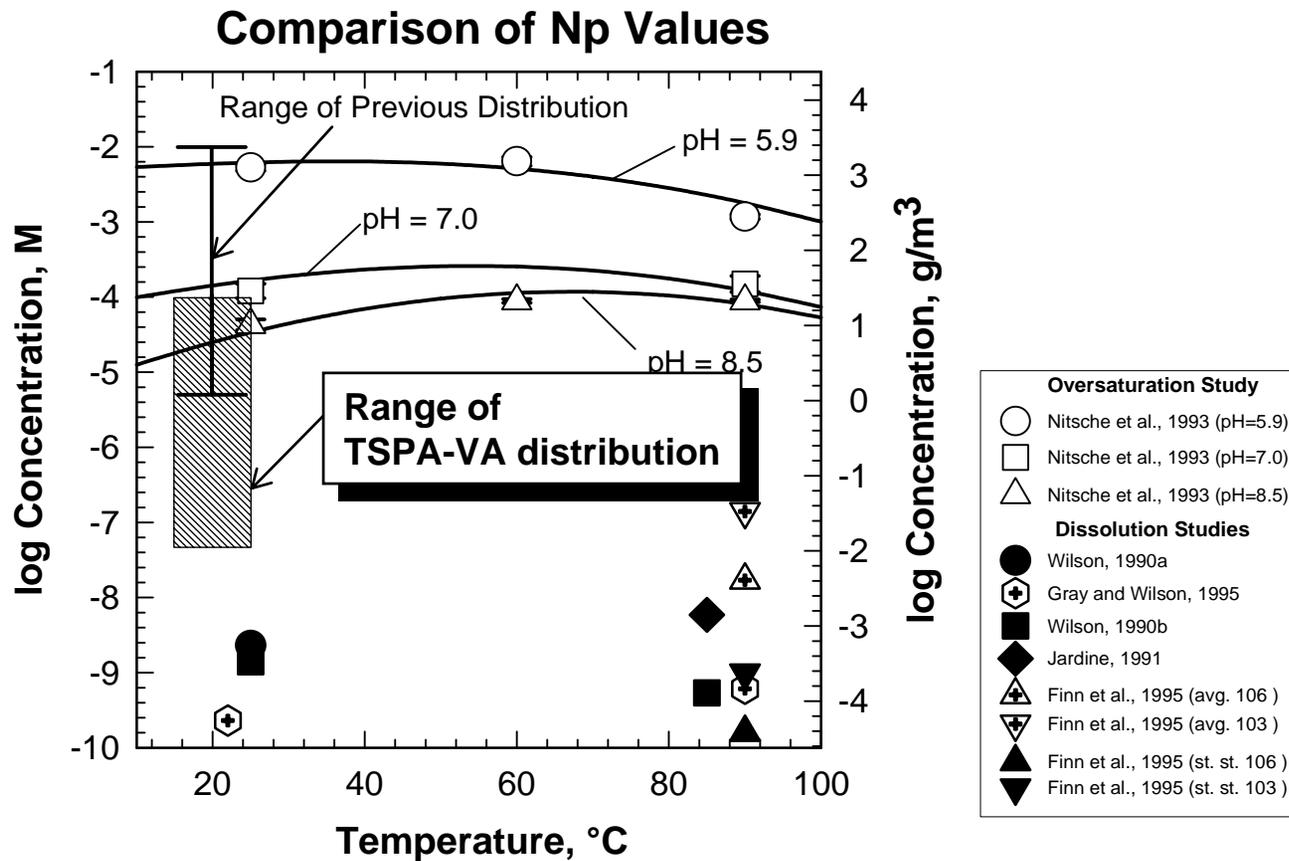
# Cladding 5th & 95th Percentile 1,000,000-yr Dose-Rate History All Pathways, 20 km



# Radionuclide Solubilities

- **Most radionuclides are released into the EBS transport process at their solubility limit**
- **Solubilities are sampled over a range with a minimum, maximum, average and probability distribution function**
- **In the current Base Case, solubilities (except Np) are the same as used in TSPA-95.**
- **After review, Np solubility has been reduced from TSPA-95 values by a factor of 100 (M&O, 1998)**

# Range of Solubility-Limited Np Concentrations



(from M&O, 1998)

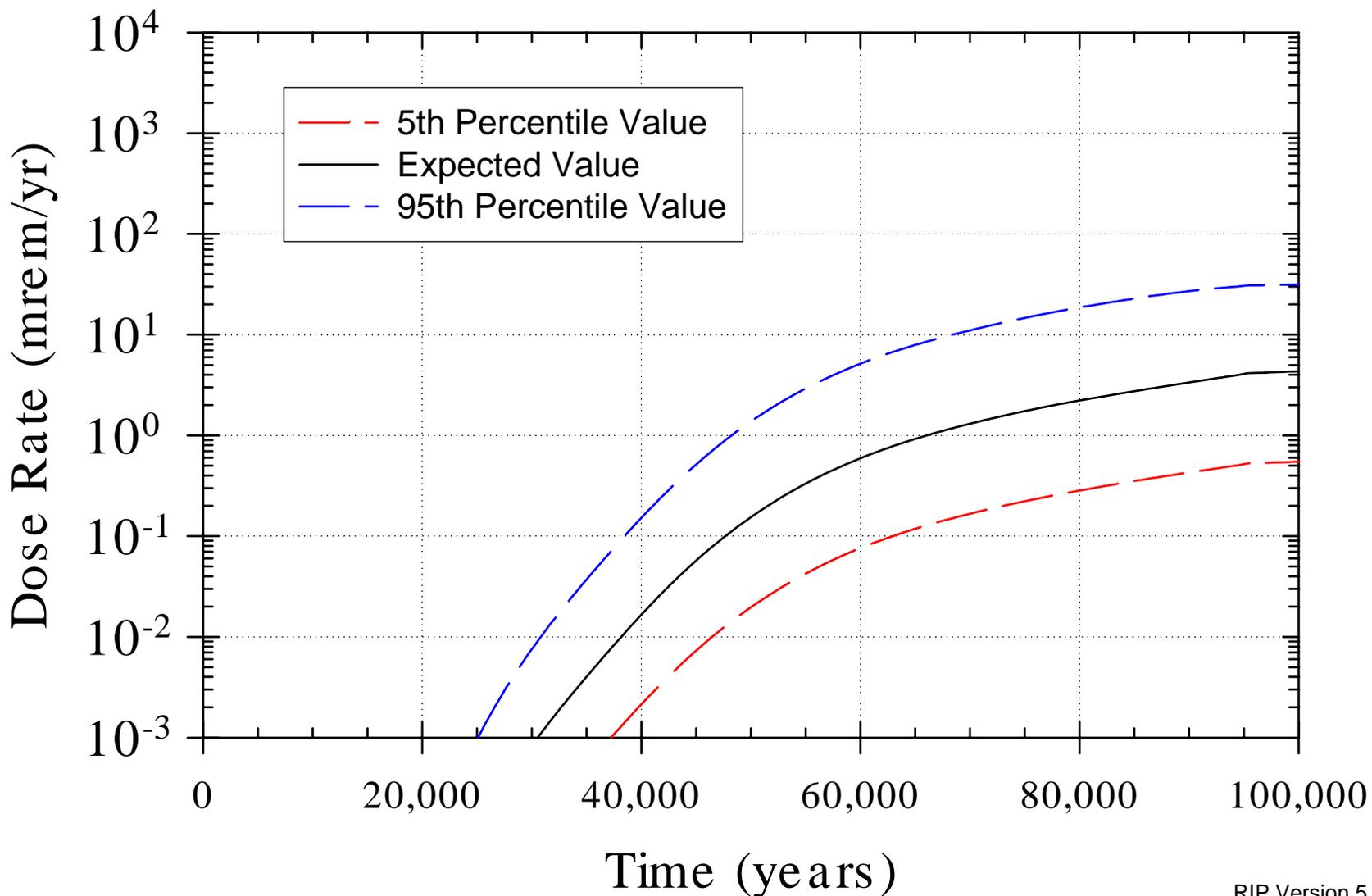
**Note: Np solubility 100 times less than TSPA-95**

# Summary of Change for Neptunium

- **Nitsche et al. Studies Used Concentrated Solutions from Np-salts to Approach from Oversaturation**
- **Thermochemical Data Suggest that Phases Formed in Studies Represent Metastable Solids**
- **Synthesized Results of Dissolution Studies**
  - **Does spent fuel in J-13-like fluids (starting with zero Np) reach such high values at steady state?**
    - » **flow-through tests**
    - » **drip tests**
    - » **batch studies**
  - **All Measured Np Concentrations Lower Than Needed to Saturate Phases in Nitsche et al. Studies**
    - » ***highest* time-averaged value is 1/37 of the *lowest* elicited value and steady-state values are even lower**
- **Metastable Phases not Expected to Apply, Stable Phases like  $\text{NpO}_2$  Should Keep Np Below about 1/100 of the Elicited Range**

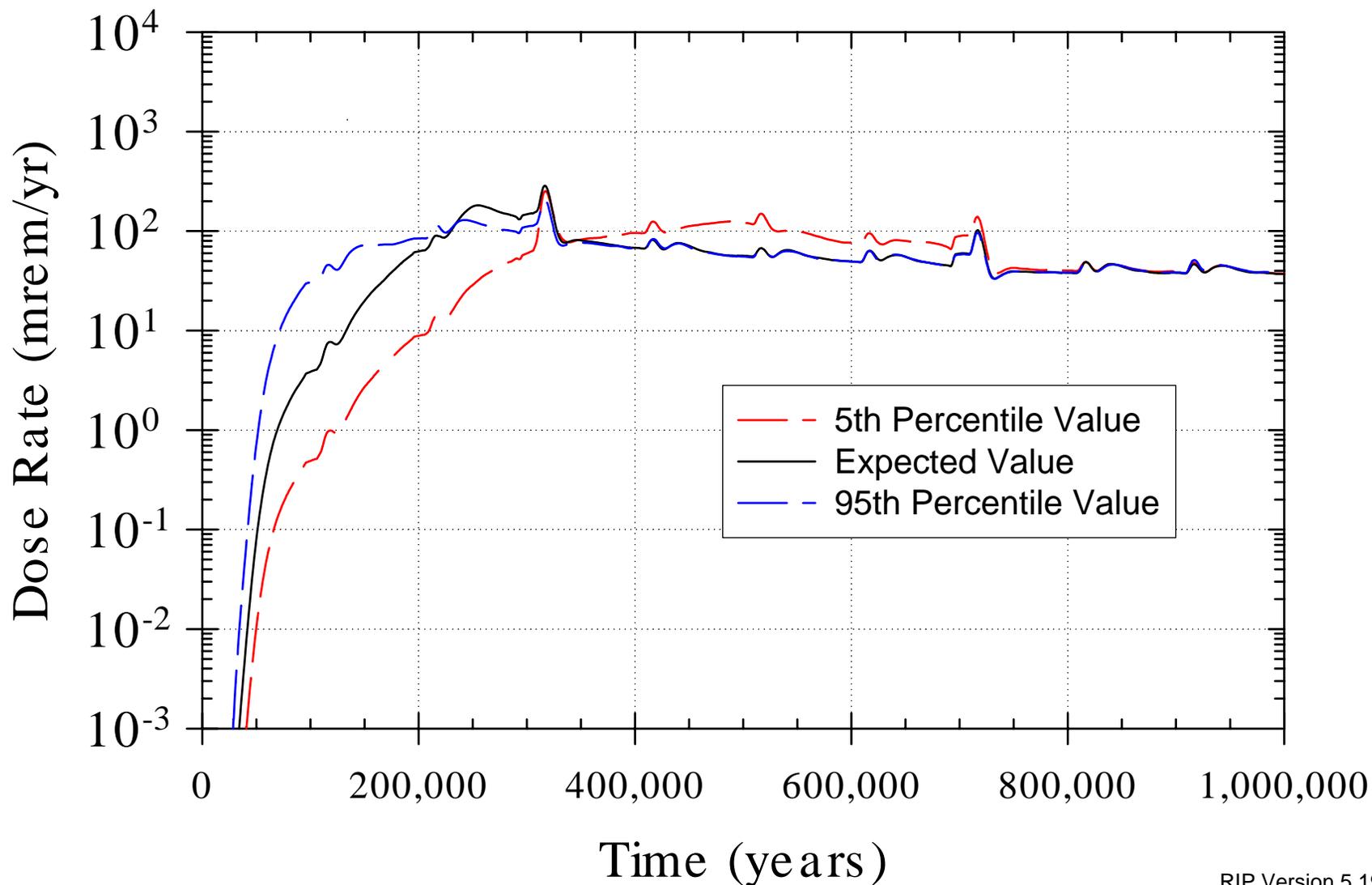
# Np Solubility 5th & 95th Percentile 100,000-yr Dose-Rate History

All Pathways, 20 km



# Np Solubility 5th & 95th Percentile 1,000,000-yr Dose-Rate History

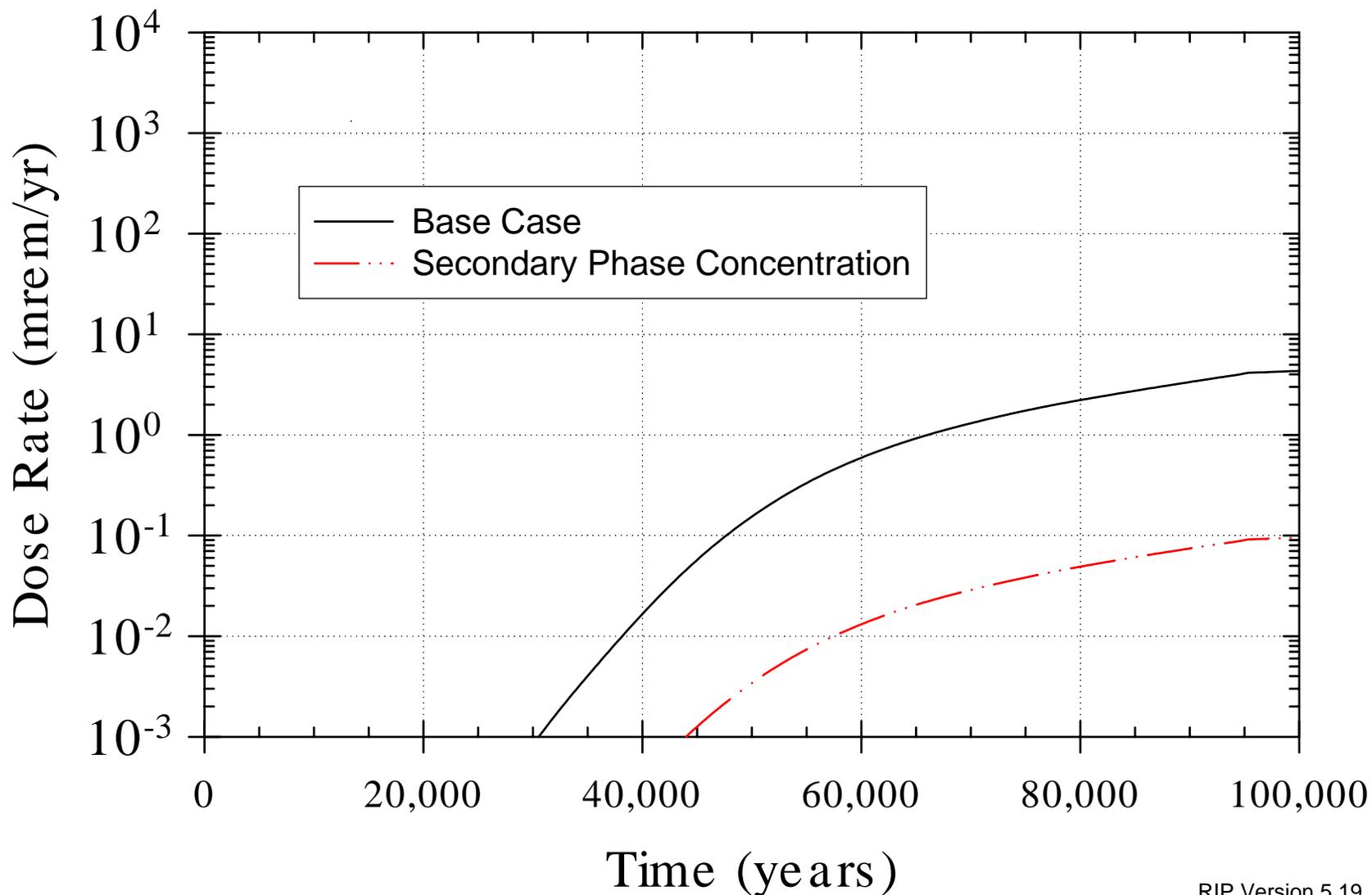
All Pathways, 20 km



# Sensitivity to Np Solubility Model

## 100,000-yr Expected-Value Dose-Rate History

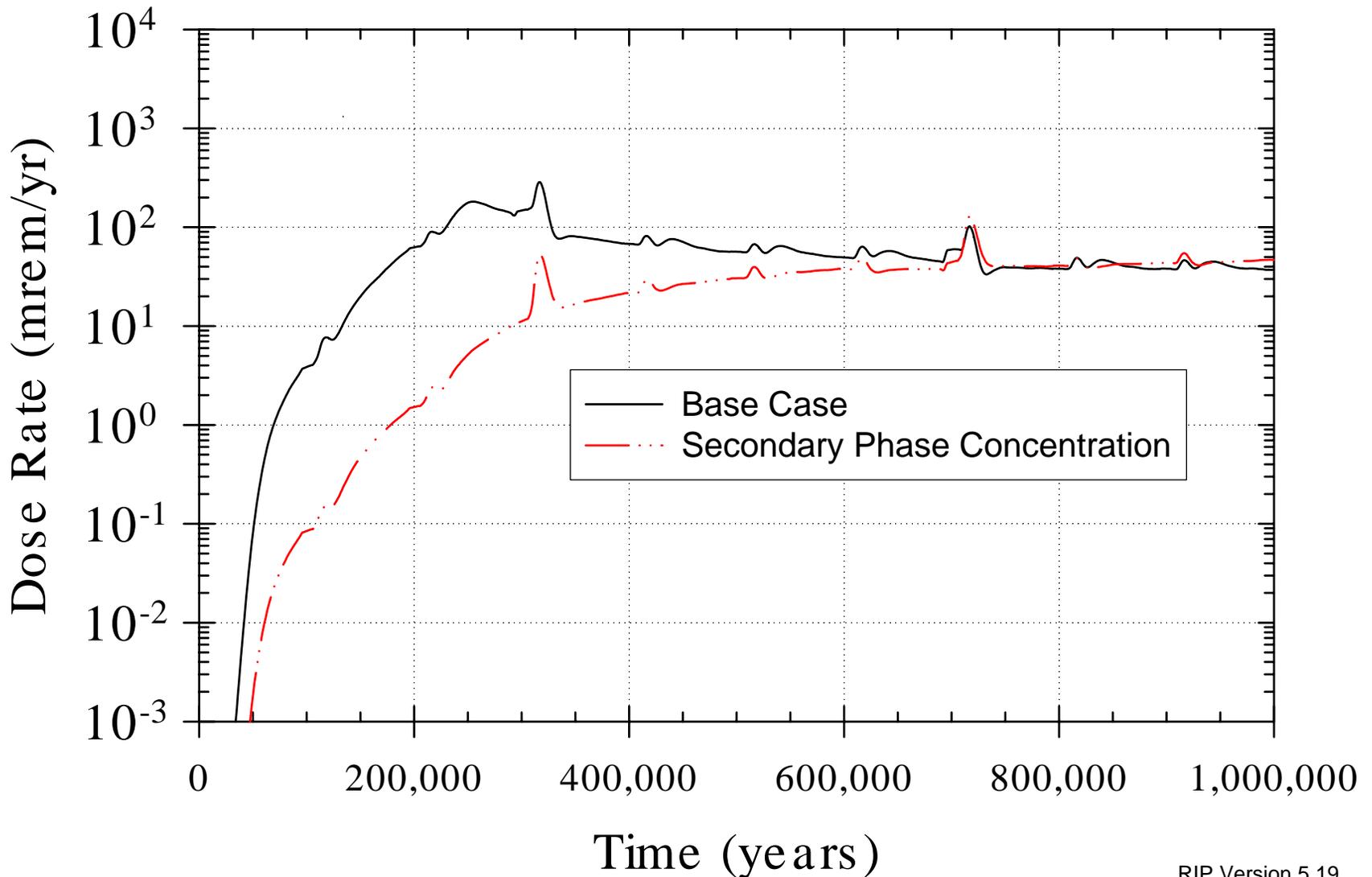
All Pathways, 20 km



# Sensitivity to Np Solubility Model

## 1,000,000-yr Expected-Value Dose-Rate History

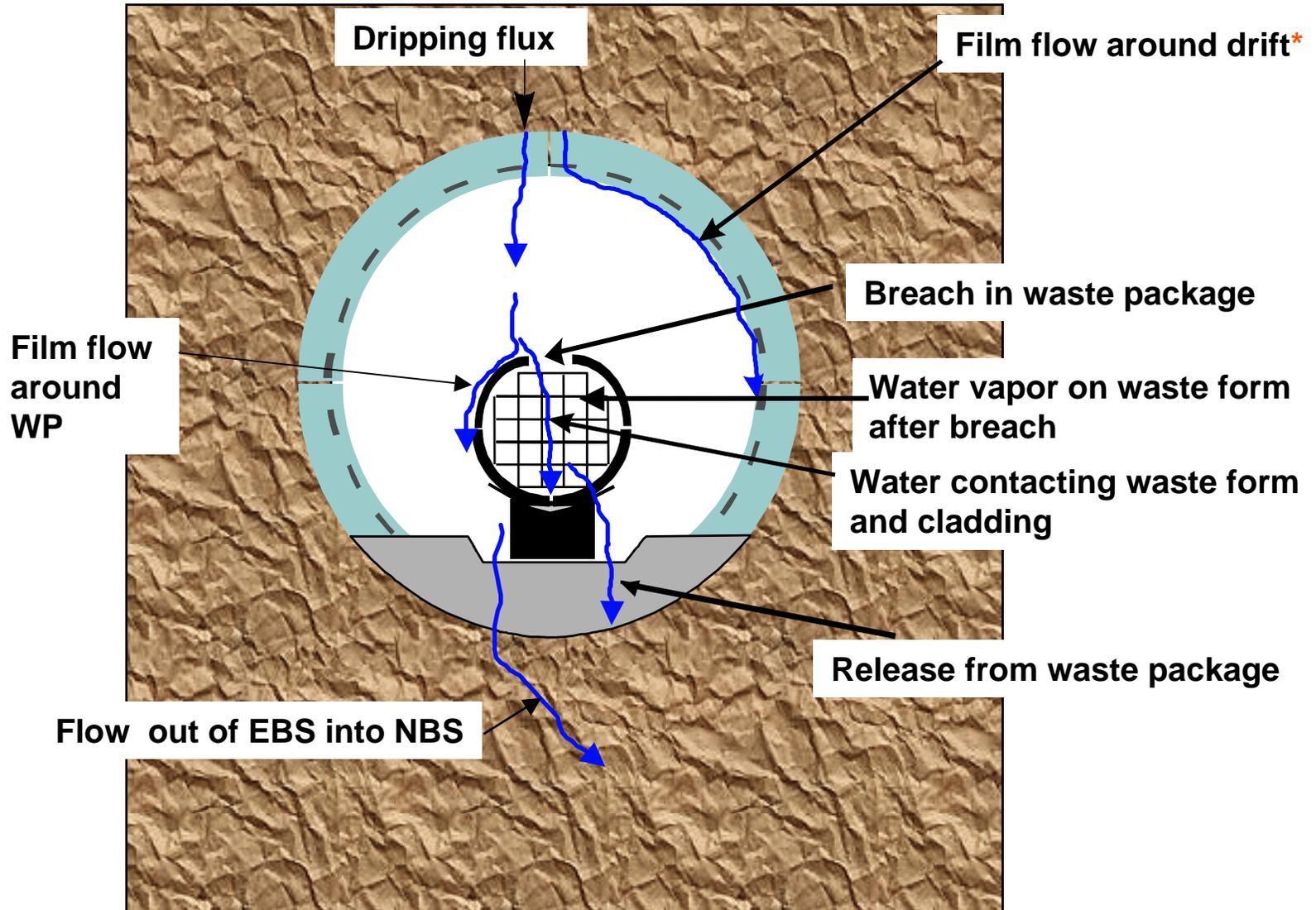
### All Pathways, 20 km



# Colloidal Plutonium Transport - Base Case

- **May increase release from waste package and decrease travel time in near field and far field**
- **Significance depends on stability and reversibility of RN attachment**
- **Four colloid types considered in TSPA-VA: clay, iron oxide, “spent-fuel waste-form” and “glass waste-form”**
- **Reversible sorption considered in TSPA-VA base case with ratio of amount mobilized on colloid to amount dissolved ( $= K_c$ ) ranging from  $10^{-5}$  to 10 based on laboratory data**

# Conceptual model of all potential water flow pathways through the EBS



**\*Not in base case**

# Engineered Barrier System Transport

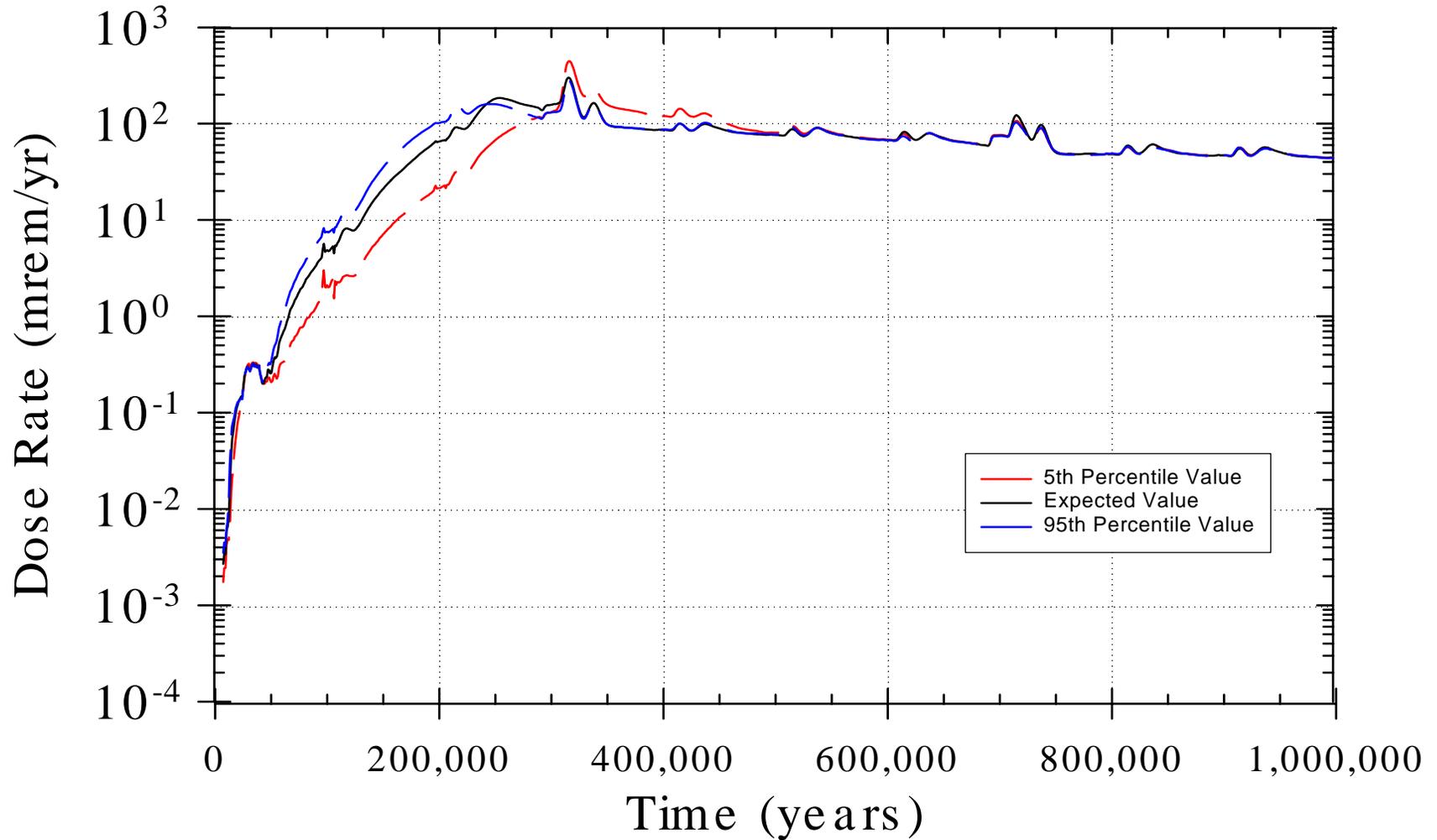
## EBS release occurs when:

- WP is breached allowing air and/or water into can
- Clad is breached allowing air and/or water into WF
- Waste form degrades
- RN's are mobilized (dissolved and colloidal)
- RN's transport through EBS by advection and diffusion

## Performance improves if:

- Protect WP from drips
- Clad remains substantially intact (protected from high heat and mechanical disruption)
- WF degradation very slow
- RN's less mobile (insoluble, little colloid mobilization)
- RN's transport slowly (advective and diffusive barriers, retardation)

# Sensitivity of dose to seepage into waste package



# Summary/Conclusions

- **NFGE information included in TSPA-VA**
- **Improvements in waste form degradation and radionuclide mobilization models**
- **CSNF dissolution model has been extended to consider temperature, burnup,  $\Sigma\text{CO}_3^{-2}$ , pH and  $\text{O}_2$**
- **HLW glass dissolution model has been updated**
- **Np elemental solubility updated**
- **Colloid mobilization has been added**

# Summary/Conclusions

(continued)

- **Significant effect on EBS transport performance**
  - Waste Package (and cladding) longevity
  - Np Solubility
  - Advection control
  - Colloid control (if necessary)
- **Additional data requirements**
  - Interaction of water with waste package and waste form
  - Nature of advective and diffusive flow paths in EBS
  - Geochemistry along flow paths in EBS