Potential for Colloidal Pu Transport:

Principles, monitoring, and challenges

George Redden (INL)
Jim Saiers (Yale)

Idaho National Laboratory

Brief Summary

- Pu is not a Contaminant of Concern, but requires attention
- Options for remediation have been identified, for example, capping to reduce infiltration
- However, near-term and long-term monitoring will be important
- Transport of Pu in colloidal form is one of the important factors regarding transport predictions (if transport can occur)

Pu at the INL Subsurface Disposal Area

- ~113 kg Pu in wastes from Rocky Flats buried at SDA
- ~42 kg (~4%) of Pu estimated to be in the form of particulates (e.g., in HEPA filters)
- Pu can be found in soluble form, but is found to adsorb strongly to mineral surfaces, including mobile particulates
- INL subsurface characterized by fractures and fast flow paths

- **→ Transport of Pu in colloidal form is a concern**
- **→ Transport models must consider colloids**
- **→ State of Pu found in field samples (if any) will impact transport interpretation**

Monitoring

- **Purpose**
  - Is current risk is unacceptable?
  - Will existing conditions prevent exposure in the future?
  - Do you find what you predict?
    - Concentrations
    - Distribution
    - State
  - What could be the consequences of new conditions?
  - Setting action levels: If unacceptable risk is predicted or observed, what are appropriate responses?
  - Will responses be feasible or should be developed?
- **Issues of uncertainty and scientific basis affect Public confidence and costs of response**

Facets of Monitoring

- Conceptual Model
  - Hydrology
  - Media properties
  - Contaminant properties
    - Chemistry, biochemistry
    - Colloid transport
      - Filtration
      - Rapid transport
- → Sampling
  - Methods and artifacts (e.g., preservation)
  - Sample types (solids, solutions, etc.)
  - Timing
  - Locations
- → Correspondence between contaminant distribution/speciation and model predictions for field experiments and real systems?

Questions:

- **Are the conceptual models adequate?**
- **Does sampling strategy and methods support the conceptual models?**

Current model:

- K_d = 0 in basalt
  - Fractures
  - Low surface area
  - Fast flow
- K_d in sedimentary interbeds = Kd for Pu sorption (2500 ml/g)
- Stratigraphy, flow paths, flow patterns

*** No data for Pu plume that can be used for model calibration.**

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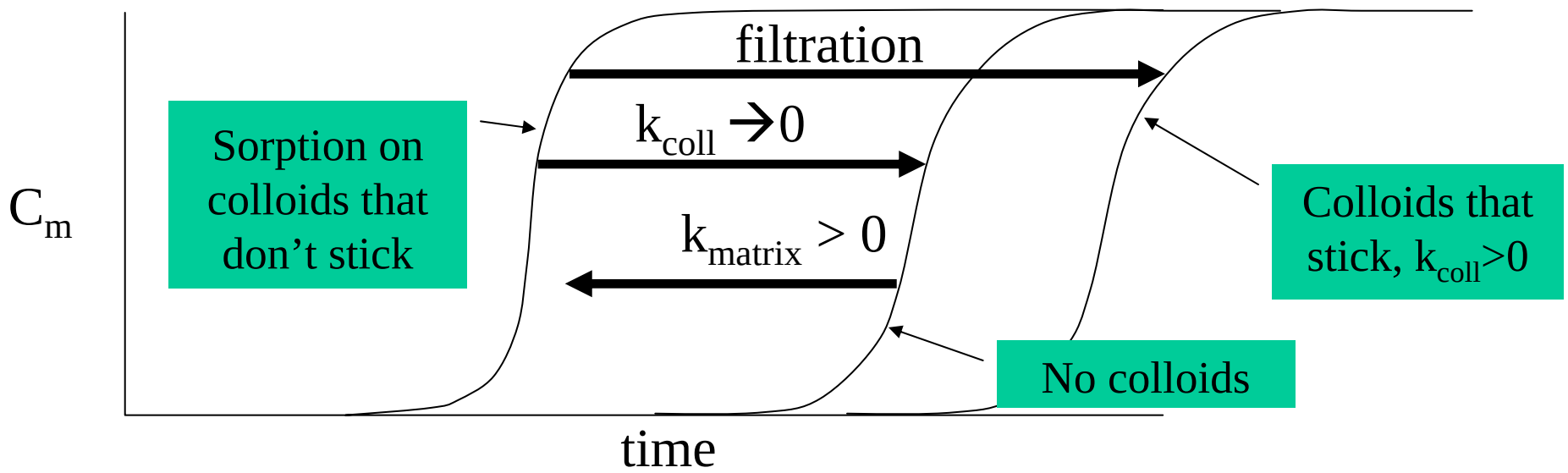
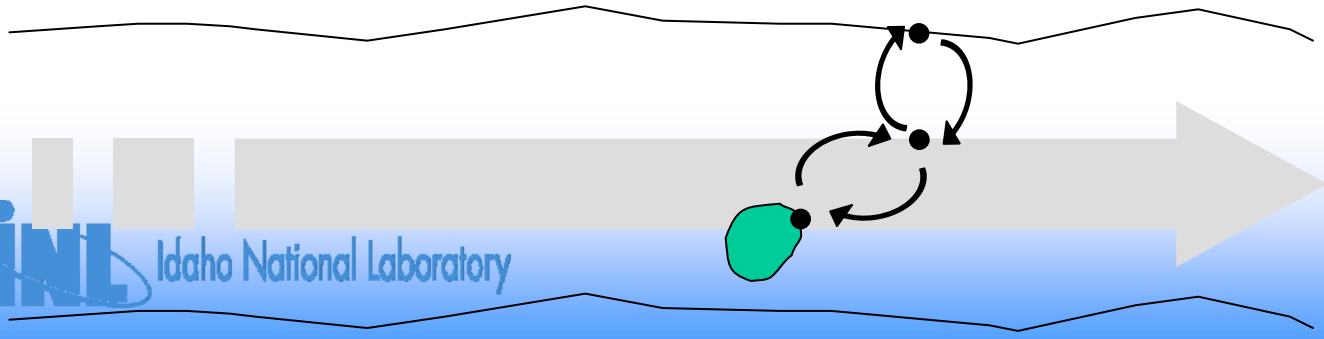
$$K_d = \frac{(mass\ adsorbed)}{(Concentration)(mass\ adsorbate)}$$

Diffusion Advection sorption

$$\frac{\partial C_m}{\partial t} = D\frac{\partial^2 C_m}{\partial x^2} - V_x\frac{\partial C_m}{\partial x} - \frac{B}{\theta}\frac{k_{matrix}}{(1+k_{coll})}\frac{\partial C_m}{\partial t}$$





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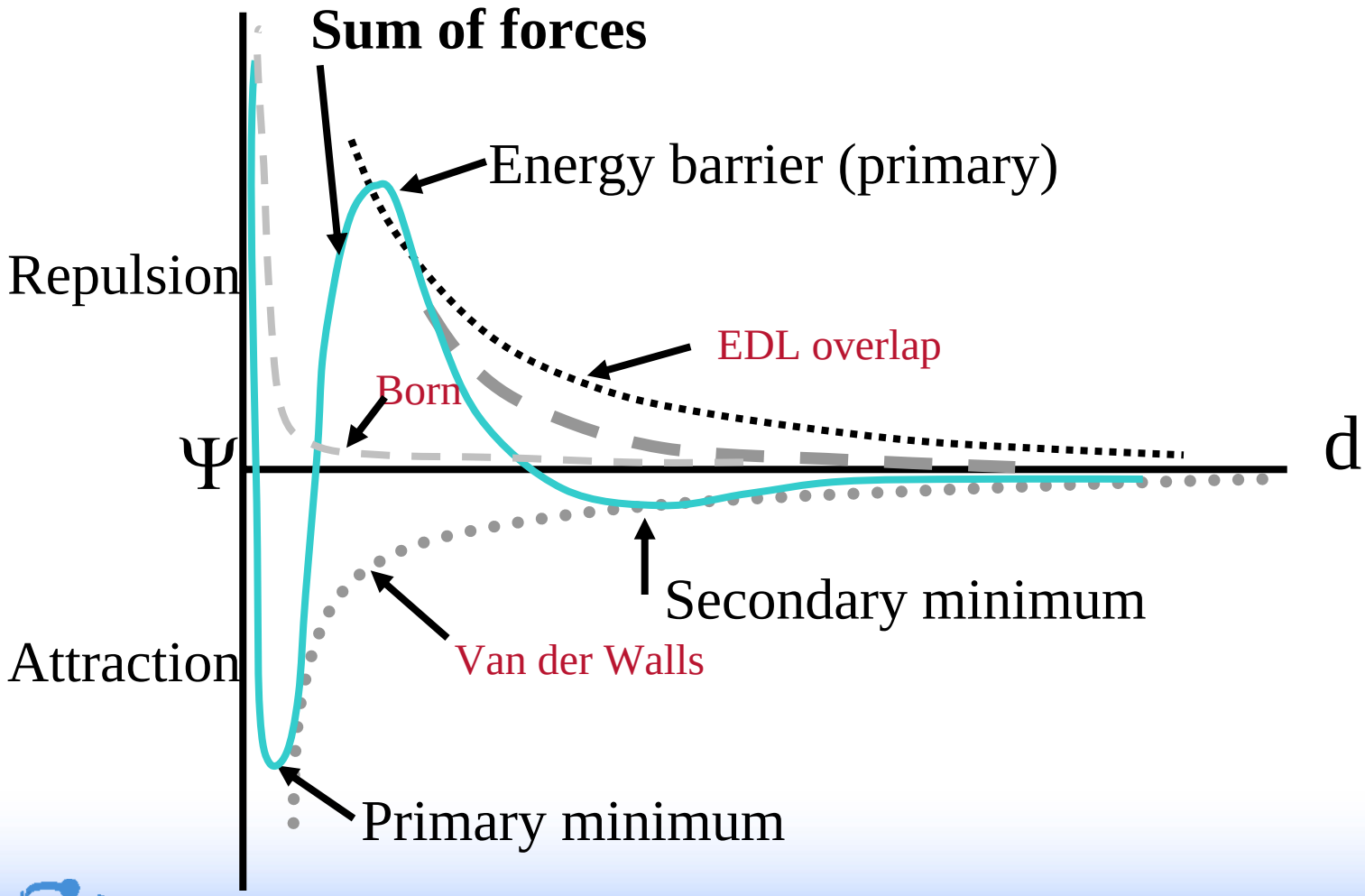
DLVO

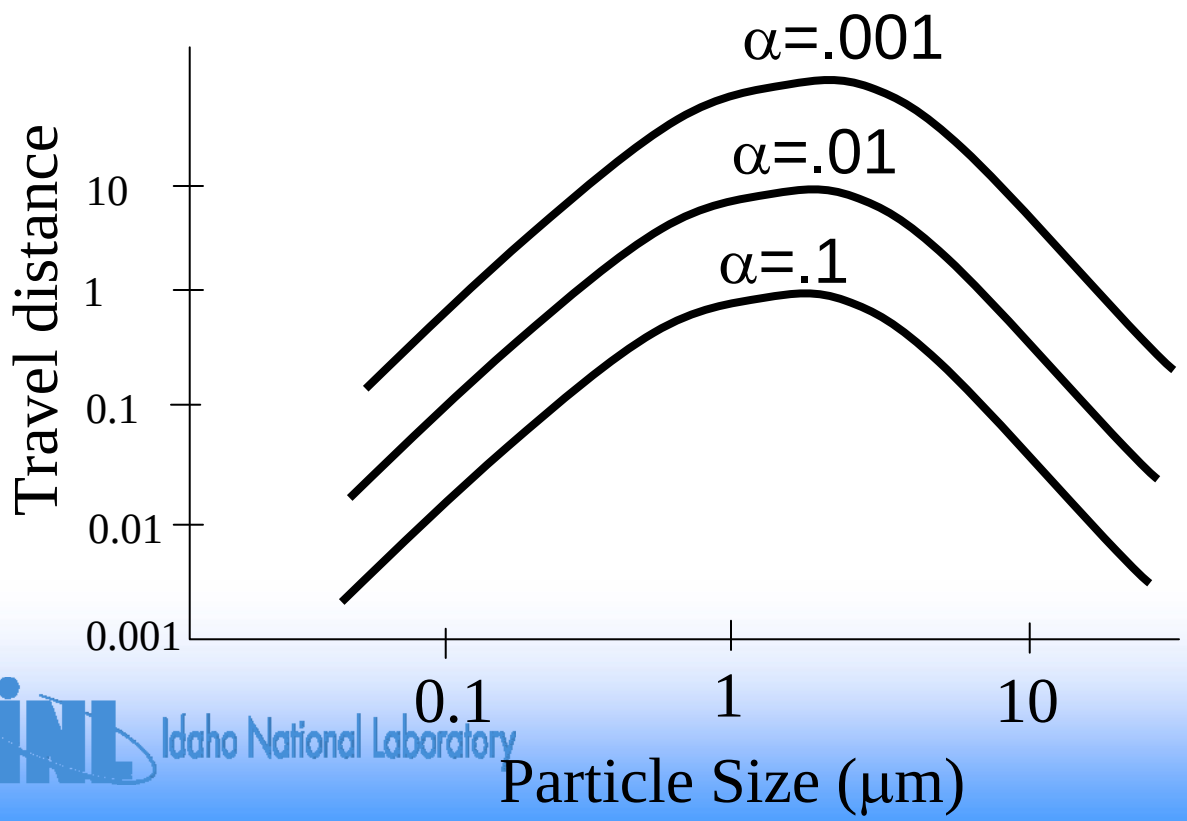
Sum of forces as surface approach (one example)



Detachment occurs when particle $E > E_{binding}$

Colloid Transport: Size and Ionic Strength Effects

$$\frac{\partial C}{\partial x} = \frac{3}{4}\alpha\eta_o \frac{(1-\Theta)}{r_{media}} C$$



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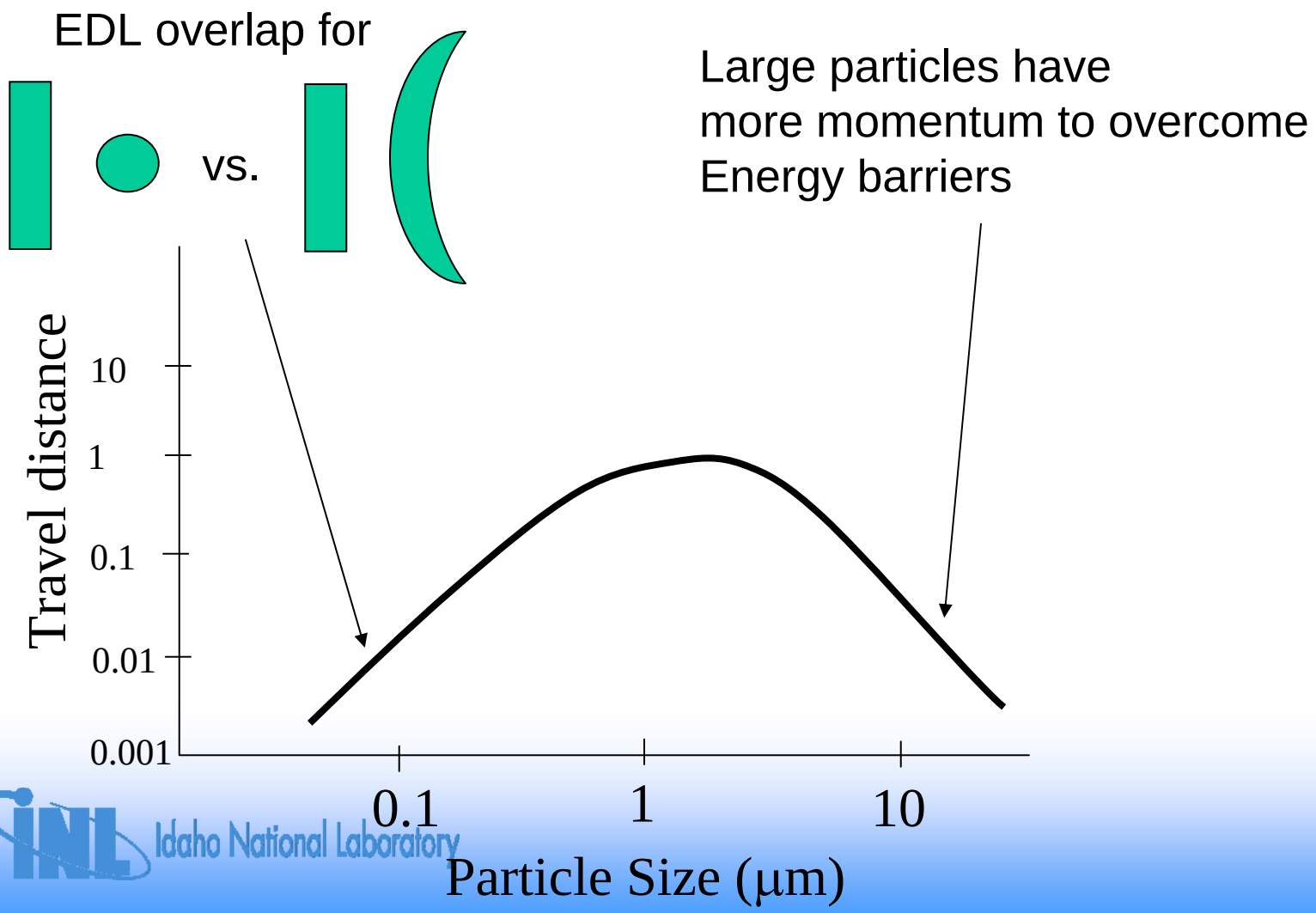
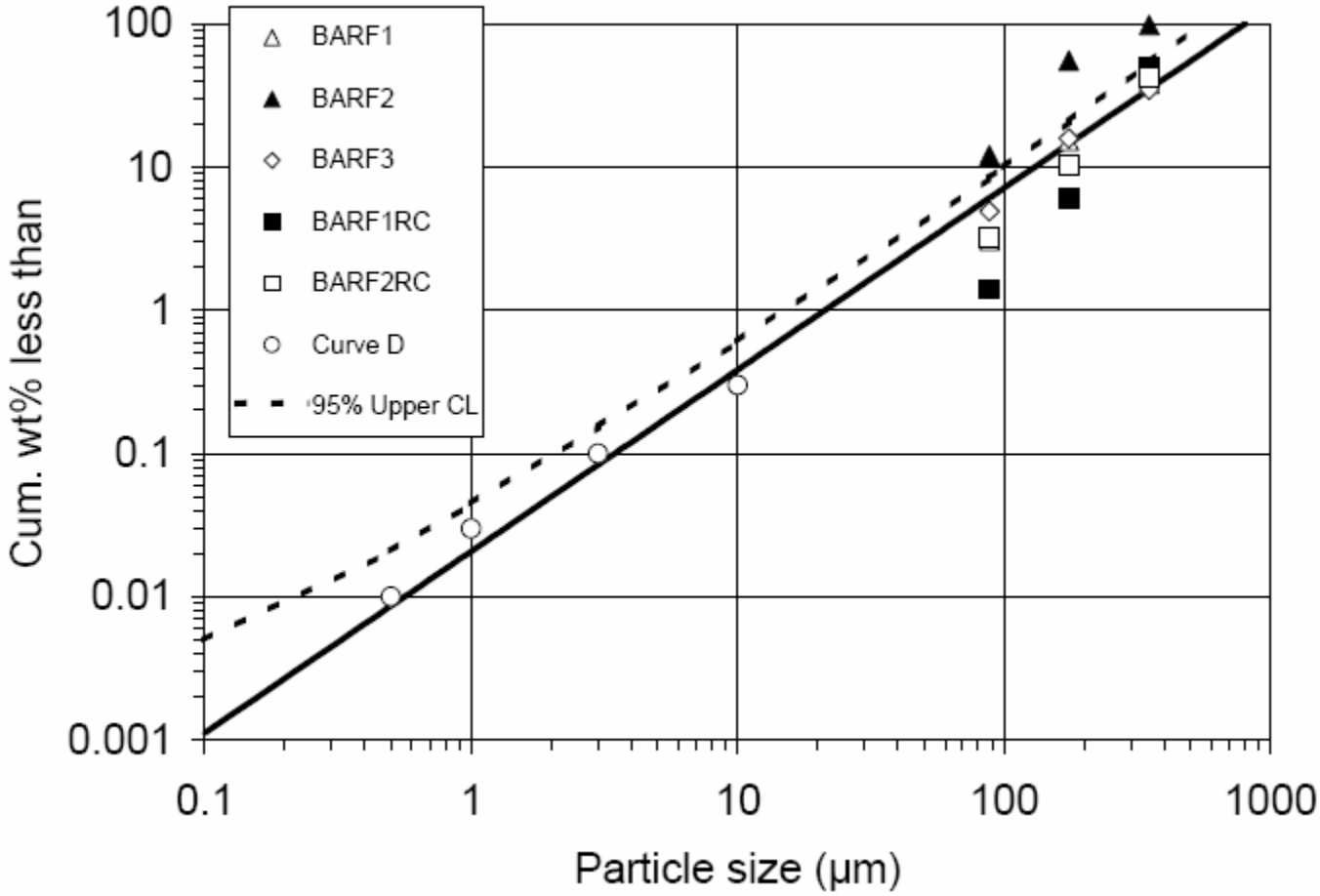
Colloid Transport:

Why does the curve have a maximum?

What decreases η or α ?



Estimated particle size distribution for Pu particles at SDA



Colloid migration lengths

$$L_T = -\ln\left({C_L}/{Co} \right)\left(\frac{4r_c}{3(1-f)\alpha\eta_o} \right)$$

Fraction removed
along flow path

99.9999% removal distance in sediments

- **Colloid diameter = 1 micron (optimum transport)**
- **Colloid density = 11.4 gm/cm3**
- **Case 1:**
  - Porosity = 0.4
  - Flow = 10cm/yr
  - Mean grain size = 0.5mm
- **Case 2:**
  - Porosity = 0.5
  - Flow rate = 100 cm/year
  - Grain size = 2mm

- **Case 1: 4.7 cm**
- **Case 2: 205 cm**

However

- **Kersting et al., Pu transport at NTS**
- **Fjeld, Coates and Elzerman, 2000 (column studies)**
- **Penrose et al., 1990 (LANL)**
  - Conservative tracer (tritium): 3.4 km/yr
  - Pu transport with sorption: 4.2 cm/yr
  - Pu transport with sorption and colloids: 4.7 cm/yr
  - Actual Pu migration: ~500 m/yr

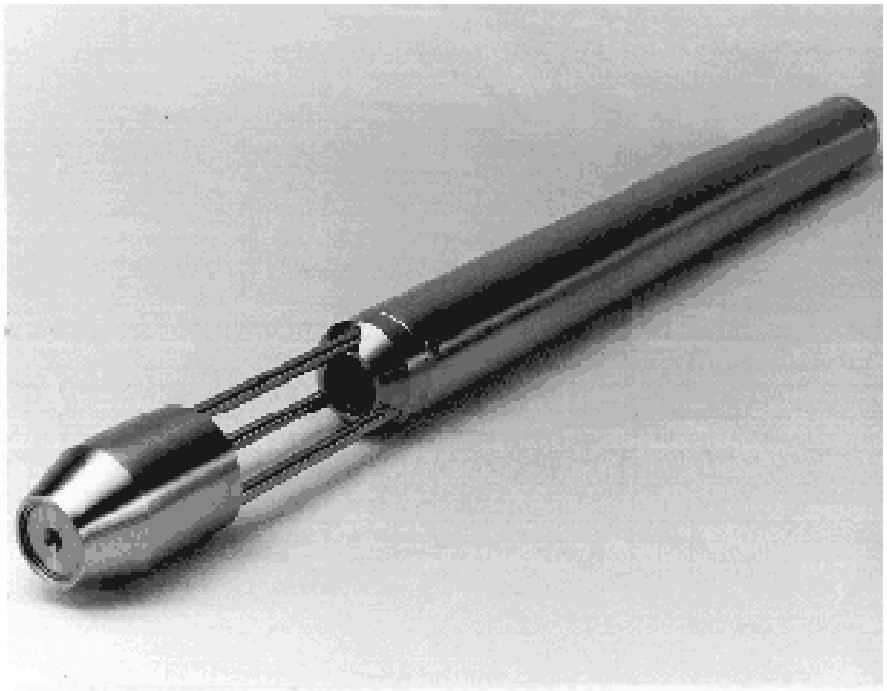
- **Also, Perturbations?**
  - All fluids intercepted by sediments
  - Constant, “low” flow
  - Constant chemical conditions
- **Also, how good are colloid filtration models?**

For breakout sessions

- **Can we prioritize the science leading to the development of scientifically based, integrated* monitoring strategies?**
- *** Integrated:**
  - **Conceptual models**
  - **Sampling methods and strategies**
  - **Lab and field studies**

- **Important issues: Scaling, simplifications**

Colloidal Borescope



- ***A CCD (charged-couple device) camera***
- ***Flux-gate compass***
- ***Optical magnification lens (140X)***
- ***Illumination source***
- ***Stainless steel housing***
- ***24 inches long by 1.7 inches in Diameter***

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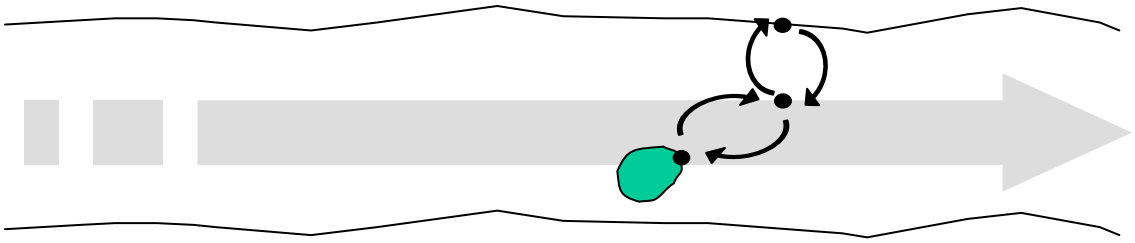
(ICP, 2003)

Colloid Facilitated Transport



$$\frac{\partial C_m}{\partial t} = D\frac{\partial^2 C_m}{\partial x^2} - V_x\frac{\partial C_m}{\partial x} - \frac{B}{\theta}\frac{\partial C_{sorb}}{\partial t}$$

Diffusion Advection sorption

C_m = total conc. in mobile phase at point x
(includes dissolved and colloid-bound)
C_{sorb}= total conc. Sorbed on immobile phase
D = diffusion coef. (or dispersion)
V_x= velocity in x direction
B = bulk density of matrix
Θ = porosity

$L_T = -\ln\left(C_L / C_o\right)\left(\frac{4r_c}{3(1-f)\alpha\eta_o}\right)$, distance in porous media where colloid concentration is reduced from C_o to C_L

$\eta_o = 4A_s^{1/3}\left(\frac{D_\infty}{Ud_c}\right) + A_s N_{LO}^{1/8} R^{15/8} + 3.38\times10^{-3} A_s N_G^{1.2} R^{-0.4}$, collection efficiency based on the number of colloids that impact a collector grain impacting and retained by a collector

$D_\infty = kT / 6\pi r_p \mu$, spherical particle diffusion coefficient (Stokes-Einstein equation), (m^2/s)

$N_{LO} = 4H / 9\pi\mu d_p^2 U$, van der Waals number

$N_G = (\rho_p - \rho) g d_p^2 / 18\mu U$, gravitational force number

$A_S = 2(1-p^5) / (2 - 3p + 3p^5 - 2p^6)$, porosity-dependent flow parameter

$p = (1-f)^{1/3}$

$R = d_p / d_c$, aspect ratio.

μ	Fluid viscosity	kg/(m s) (0.001)
ρ	Fluid density	kg/m^3 (1,000)
ρ_p	Colloid density	kg/m^3 (11,400 for PuO_2)
C_L	Colloid concentration at distance L	kg/m^3
Co	Initial particle concentration	kg/m^3
D_∞	Diffusion coefficient	m^2/s
d_c	Collector (sediment grain) diameter	m (see text)
r_c	Collector (sediment grain) radius	m (see text)
d_p	Colloid diameter	m (see text)
r_d	Colloid radius	m (see text)
f	Porosity	unitless
g	Gravitational constant	m/s^2 (9.8)
H	Hamaker constant	$kg\ m^2/s^2$ (1×10^{-20})
k	Boltzmann's constant	$kg\ m^2/(s^2\ K)$ (1.381×10^{-23})
T	Temperature	kelvin, K (see text)
U	Velocity	m/s (see text)