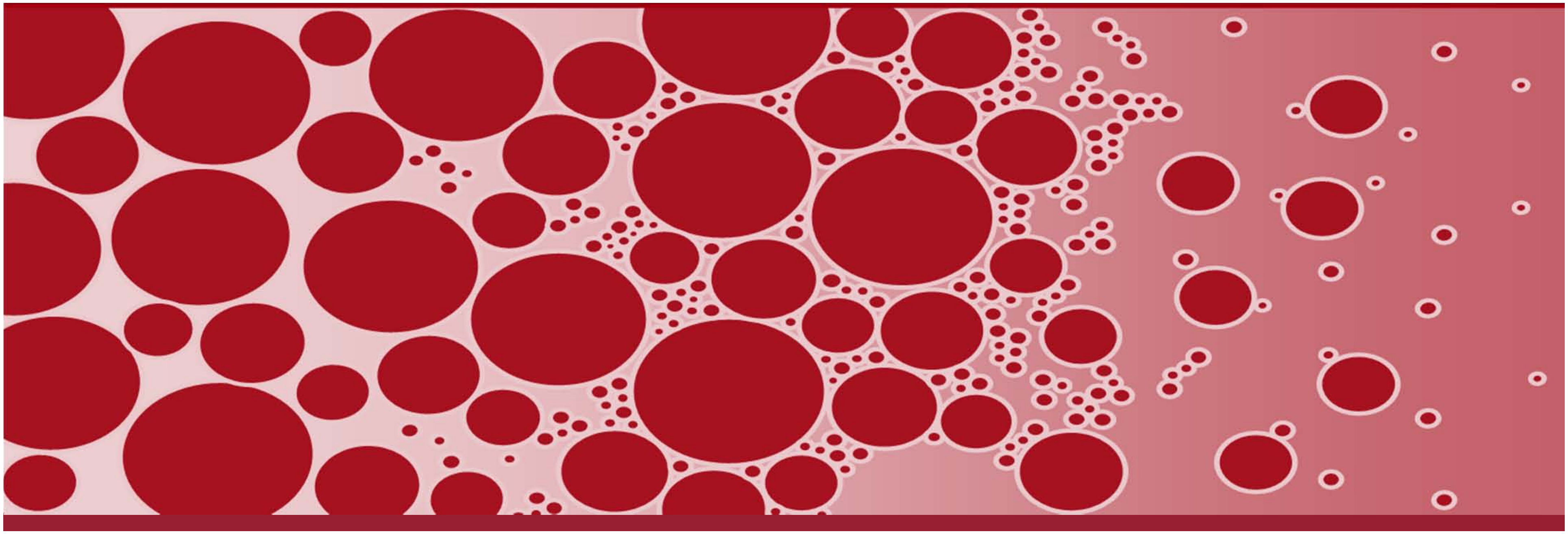

The Importance of Particle Size Distributions to The Characterization of Soils

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Acknowledgments

- The U.S. Department of Energy for funding through the Remediation and Closure Science and Science Focus Area Projects.
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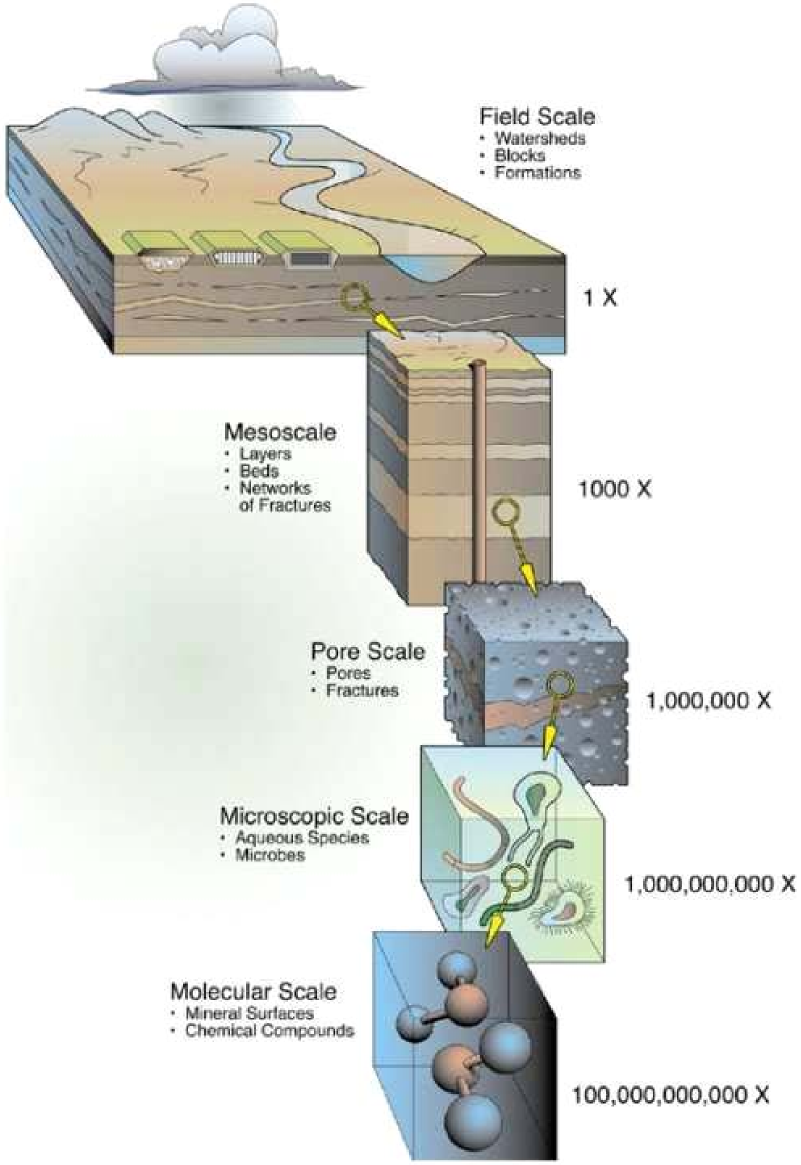
- Gary Rawson, Technologies North America Inc, perhaps the only sales person I have ever trusted

Background

- Reliable prediction of multi-scale transport behavior needed to support:
  - Environmental remediation
  - Engineered waste repositories
  - Geologic sequestration
  - Oil and gas production
  - Water resources management
- A critical need for all application areas is reliable estimation of model parameters, particularly flow and transport properties



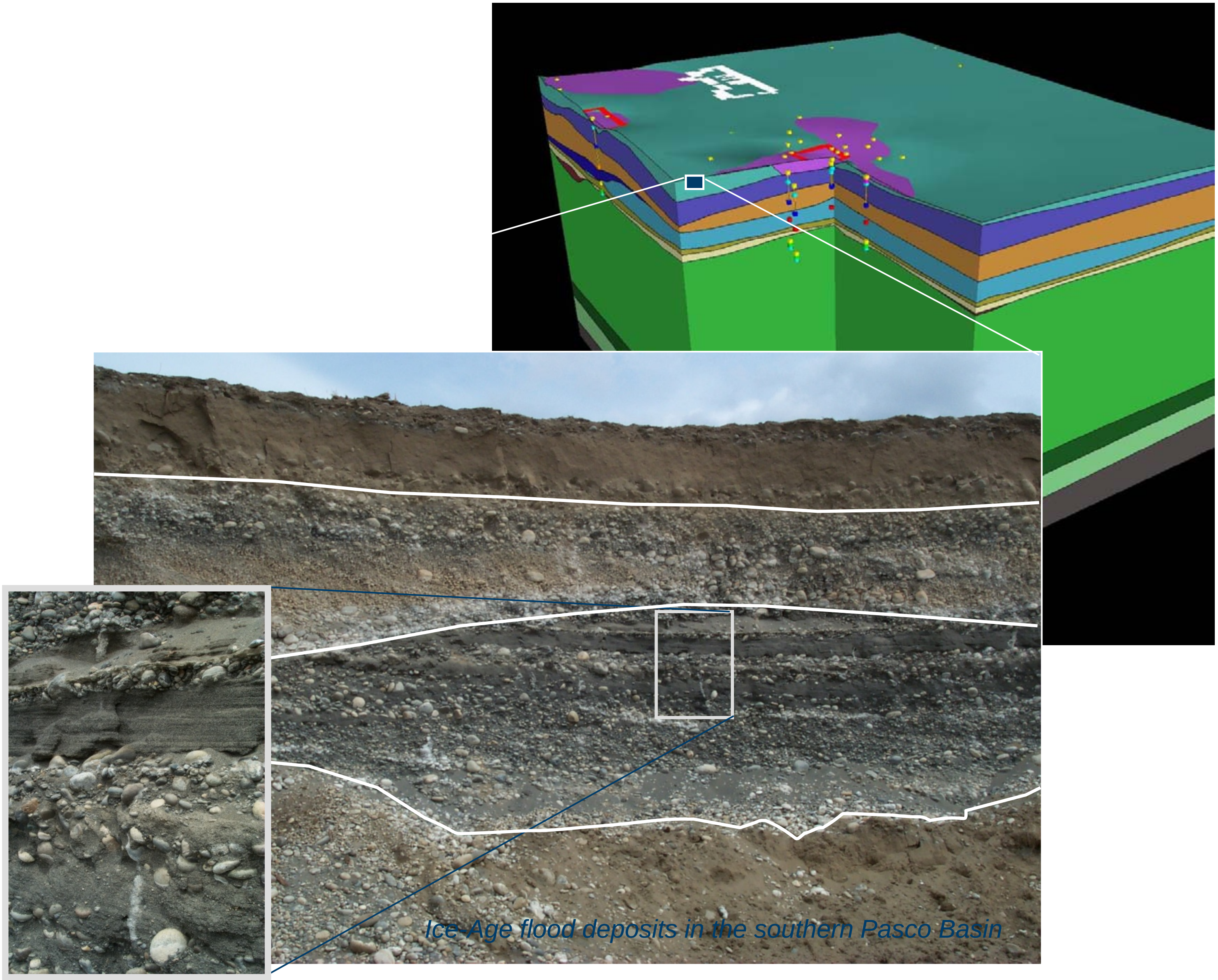
Major Challenge

- Rocks, soils and sediments are naturally heterogeneous
- Known to control near-surface and subsurface contaminant distributions
- Knowledge of flow and transport (energy, mass) properties and how they vary in space (and time) to:
  - Interpret current contaminant distributions
  - predict future contaminant migration
  - Manage soil and water resources under changing climate

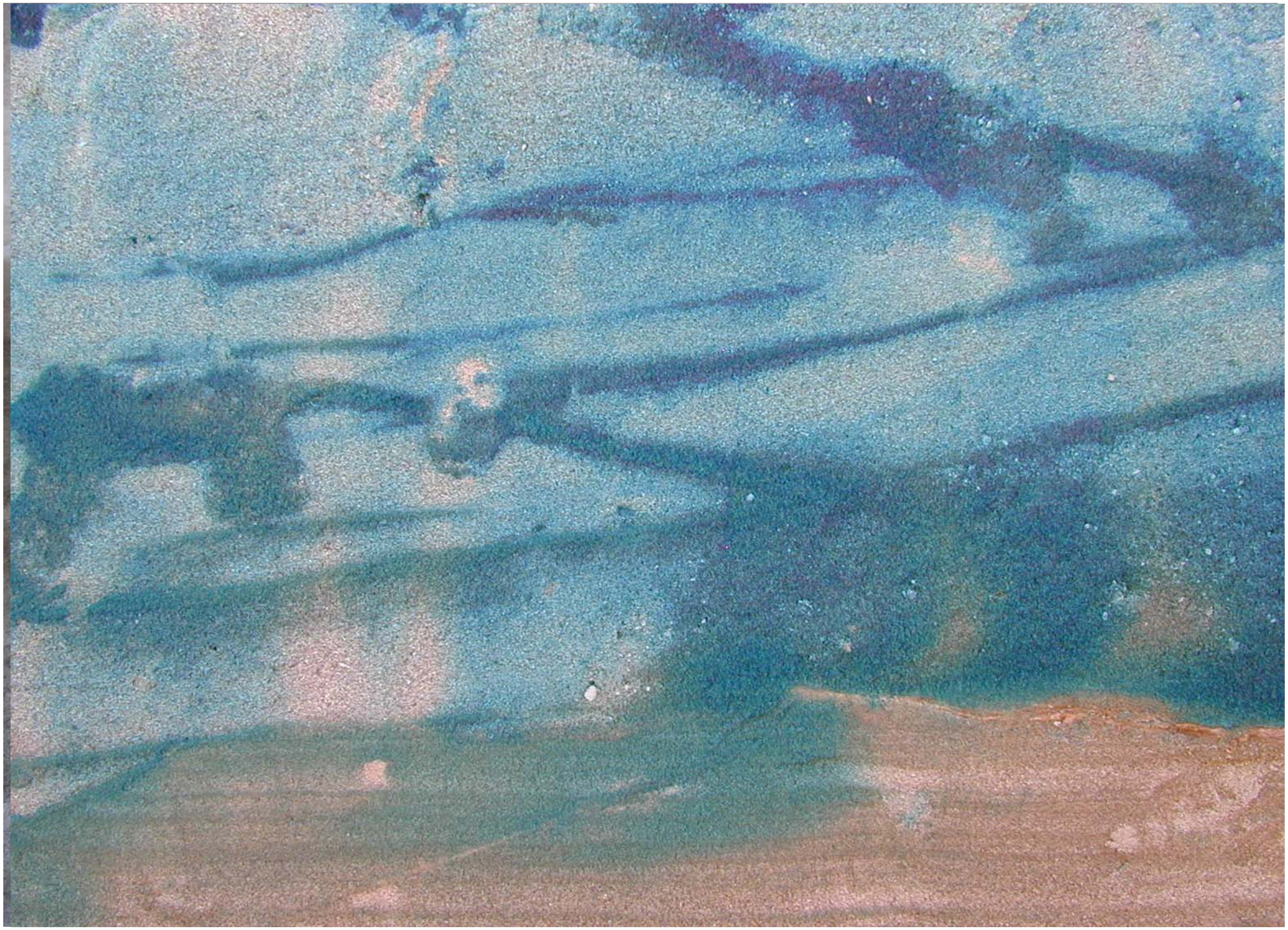
Typical Stratification

Atypical Stratification

Particle Size Distribution Transcends all Scales

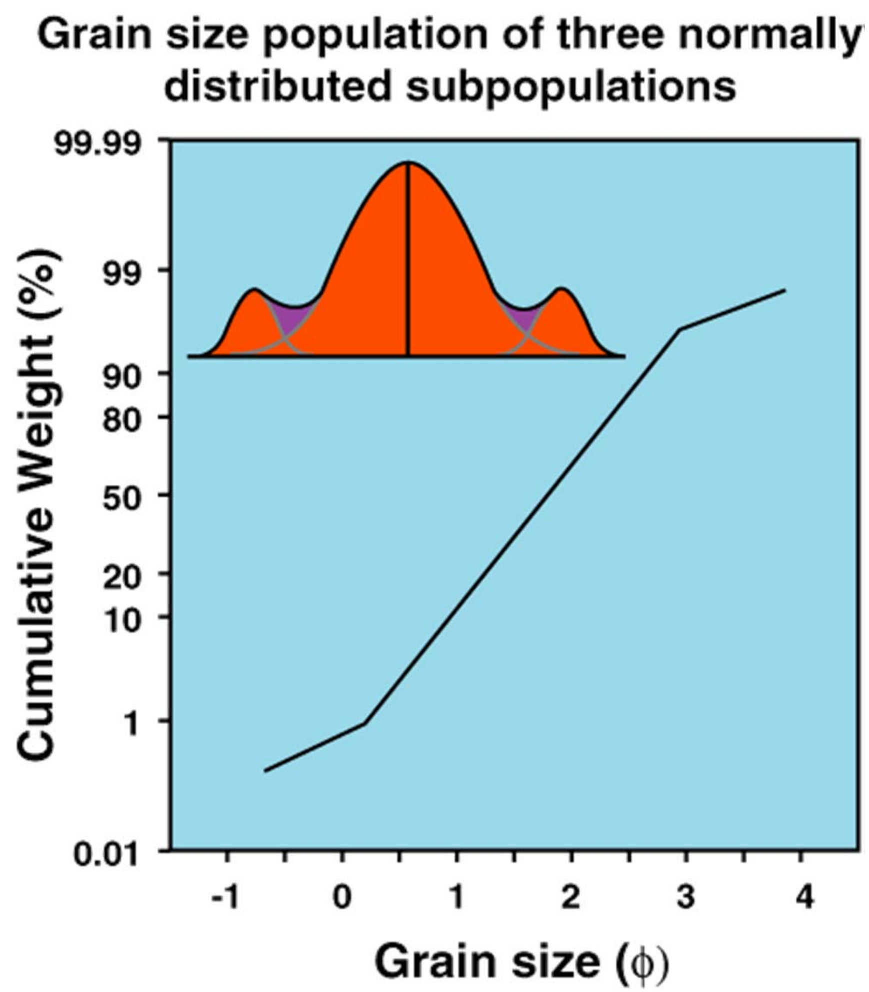


Effects Manifested at Multiple Scales



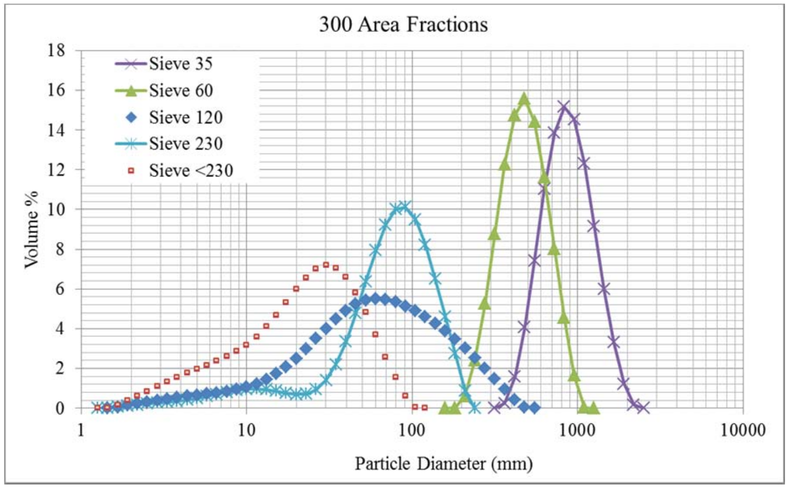
Why Measure Particle Size Distributions?

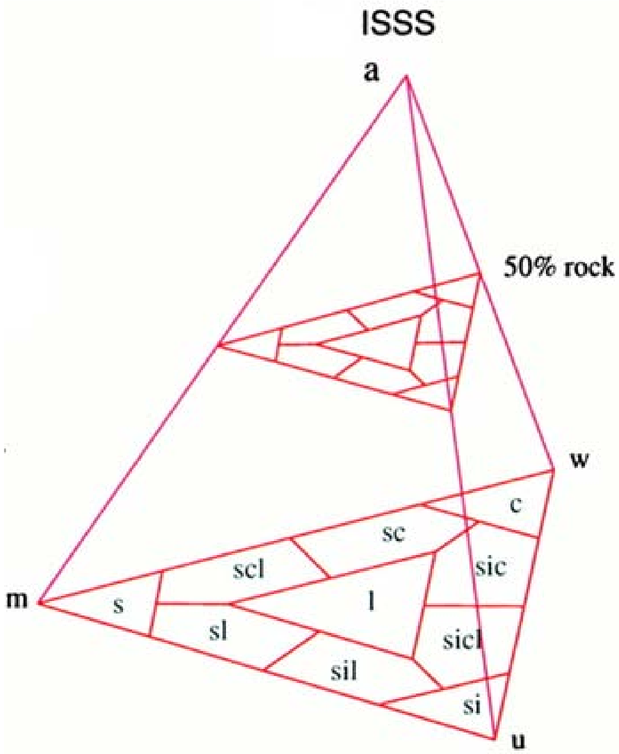
- Particle size is a fundamental property of any sediment, soil or dust deposit
  - can provide important clues to nature and provenance
- It influences a variety of other properties
- Can be defined across a hierarchy of scales
  - Stratigraphic Architecture
  - Sedimentary Sequences
  - Lithofacies
  - Small-scale heterogeneities



Particle Size Distributions

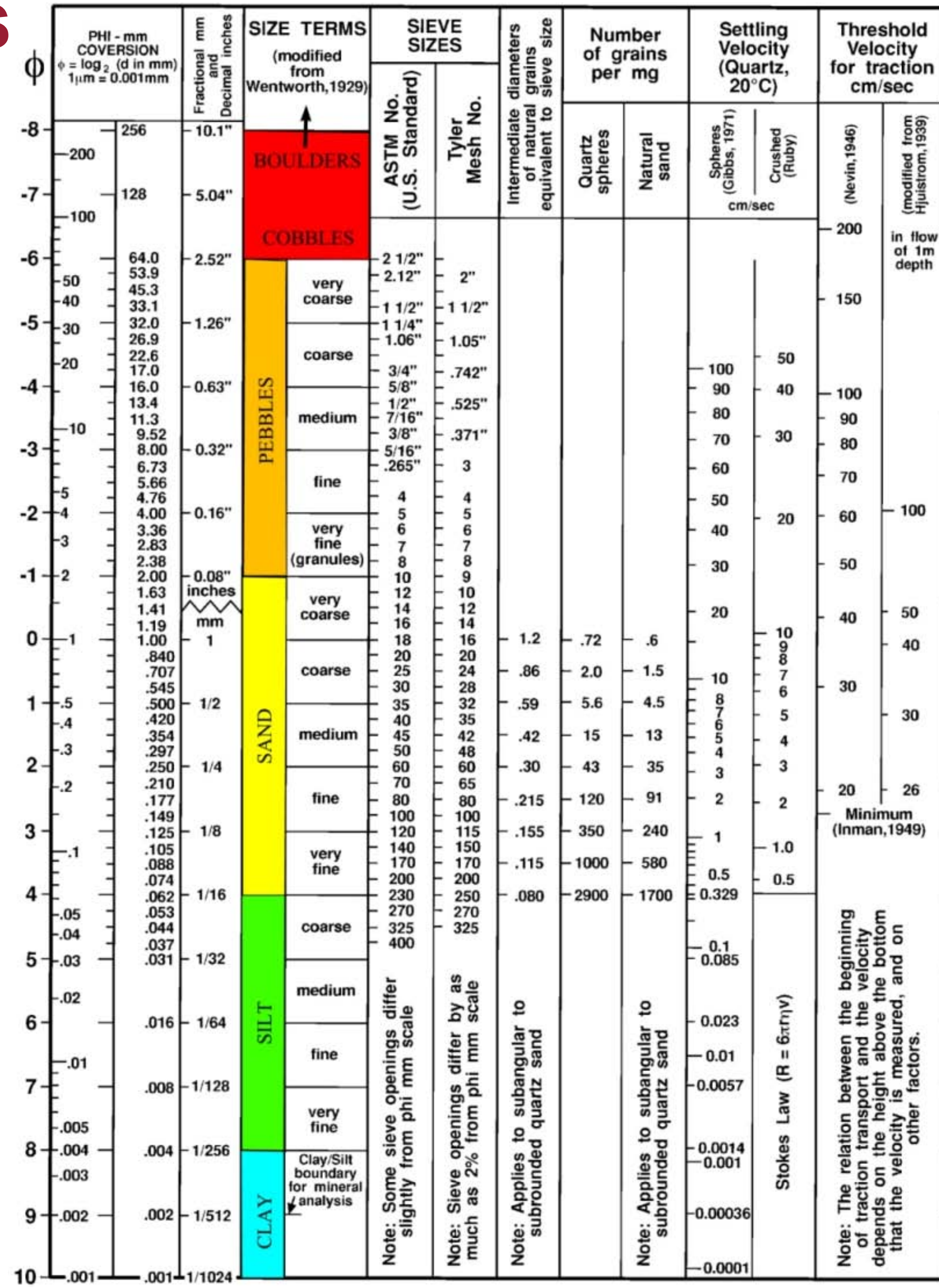
- Properties estimated from texture cannot explain transport behavior
- Measured PSDs mostly multi-modal
  - Size fractions
  - Gravel coatings
  - Rarely log normal
- More realistic and unique description using size statistics
  - mean diameter
  - sorting coefficient
  - must account for gravel





Particle Size Classes

- Boulders
- Pebbles
  - Very coarse
  - Coarse
  - Medium
  - Fine
  - Very fine
- Sand
  - Very coarse
  - Coarse
  - Medium
  - Fine
  - Very fine
- Silt
  - Very coarse
  - Coarse
  - Medium
  - Fine
  - Very fine
- Clay



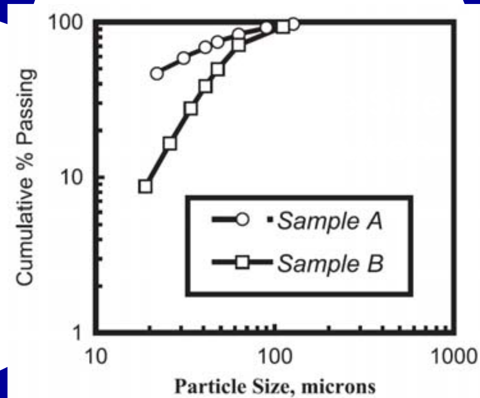
Properties Dependent on Particle Size

Primary sediment properties are controlled by facies distributions, which in turn are controlled by grain size distributions resulting from the depositional environment

Natural Isotope Abundance
- ^{40}K, ^{238}U, ^{232}Th
- Gross γ-ray response

Electrical Properties
- Dielectric Spectroscopy
- Surface conductivity
- Formation Factor
- Chargeability

Hydraulic Properties
- Bulk density
- Porosity
- Residual water content
- Water retention
- Relative Permeability



Reactive Properties
- Specific surface area
- CEC_{por}
- S_{por}
- Reaction Kinetics

Transport Properties
- Tortuosity
- Intrinsic Permeability
- Dispersivity
- Formation Factor

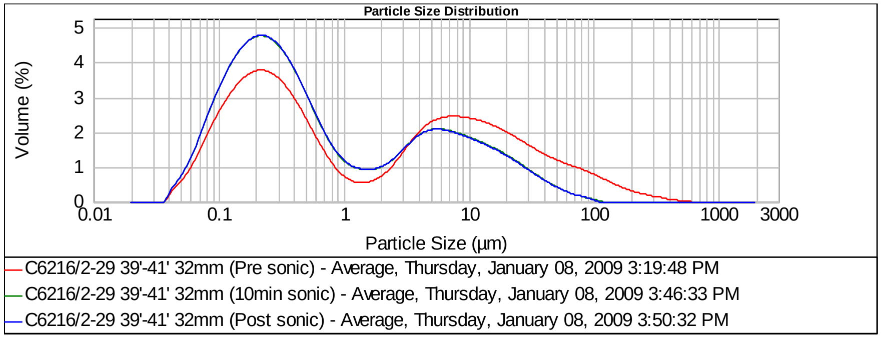
Thermal Properties
- Heat Capacity
- Thermal conductivity

Characterization of Primary Particles

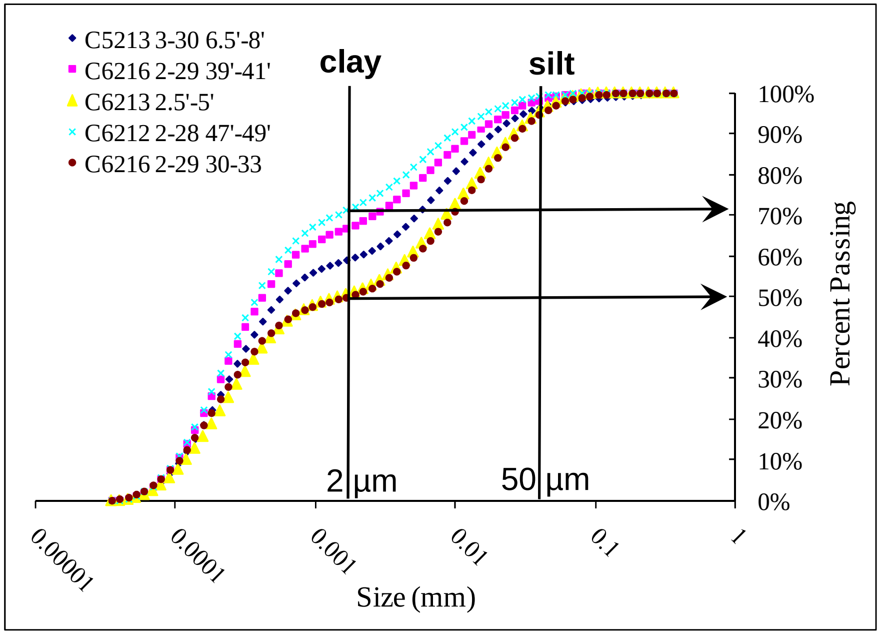
- Traditional characterization of size of "individual" particles by:
  - Sieving
  - Sedimentation
- Soil whose mineral phase is to be characterized is
  - Pretreated to remove organic matter
  - Treated to disperse aggregates
  - Passed through series of sieves with specified openings (smallest is 0.05 mm)
  - Sizes of remaining dispersed separates characterized indirectly by sedimentation (based on Stokes' Law)

Challenges in Estimating Properties

- Properties estimated from traditional PSDs often do not explain transport behavior
  - PSDs typically multi-modal
  - Fractions NOT log normal
  - Coatings that affect sorption
- Robust relationships demands a higher level of characterization
  - whole sediments
  - size fractions
  - coatings

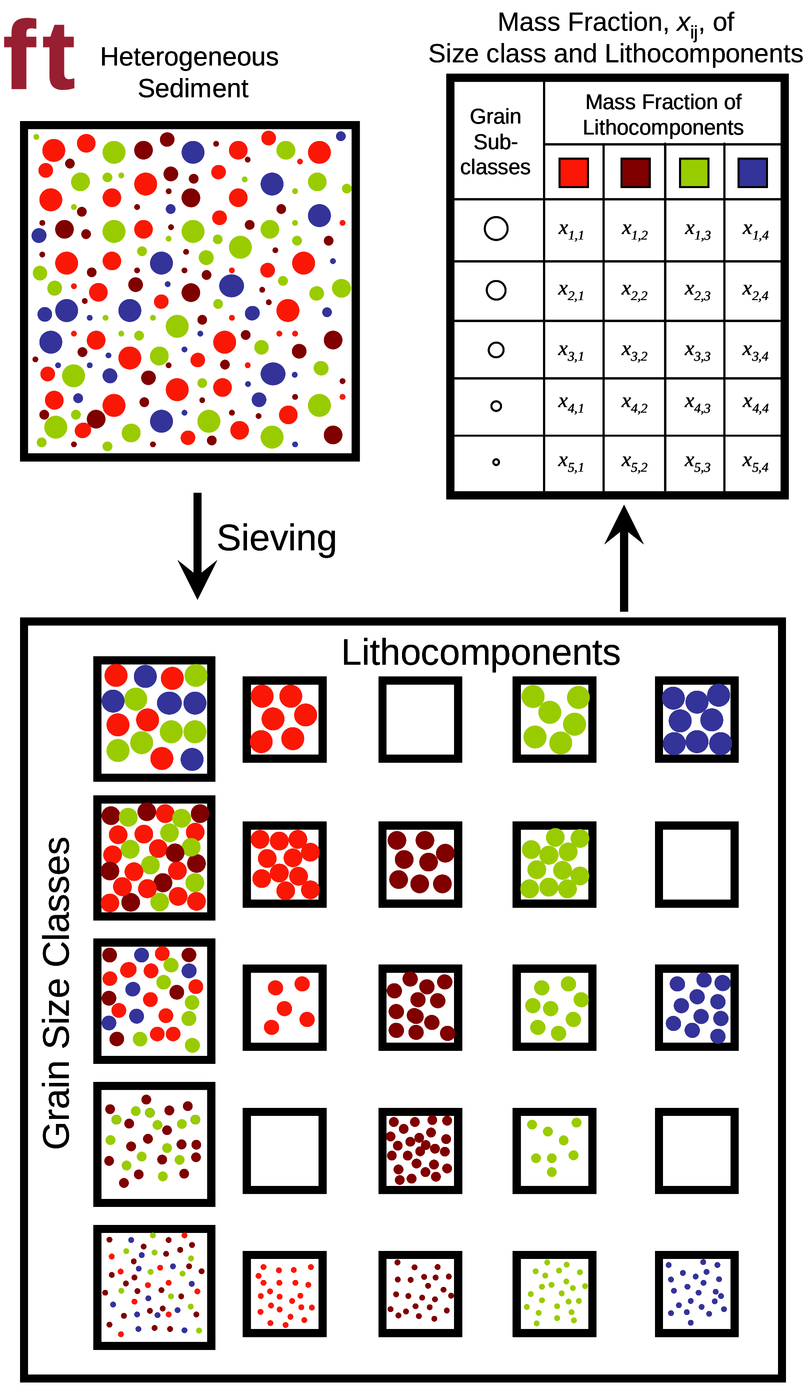


Particle Size of Coatings on 32 mm Gravel



Paradigm Shift

- Identifying such relationships requires a higher level of sediment characterization
  - Whole sediments
  - Size fractions
- Measure particle size distributions
- Measure Physico-chemical properties
  - CEC, SA, etc
- Characterize mineralogy



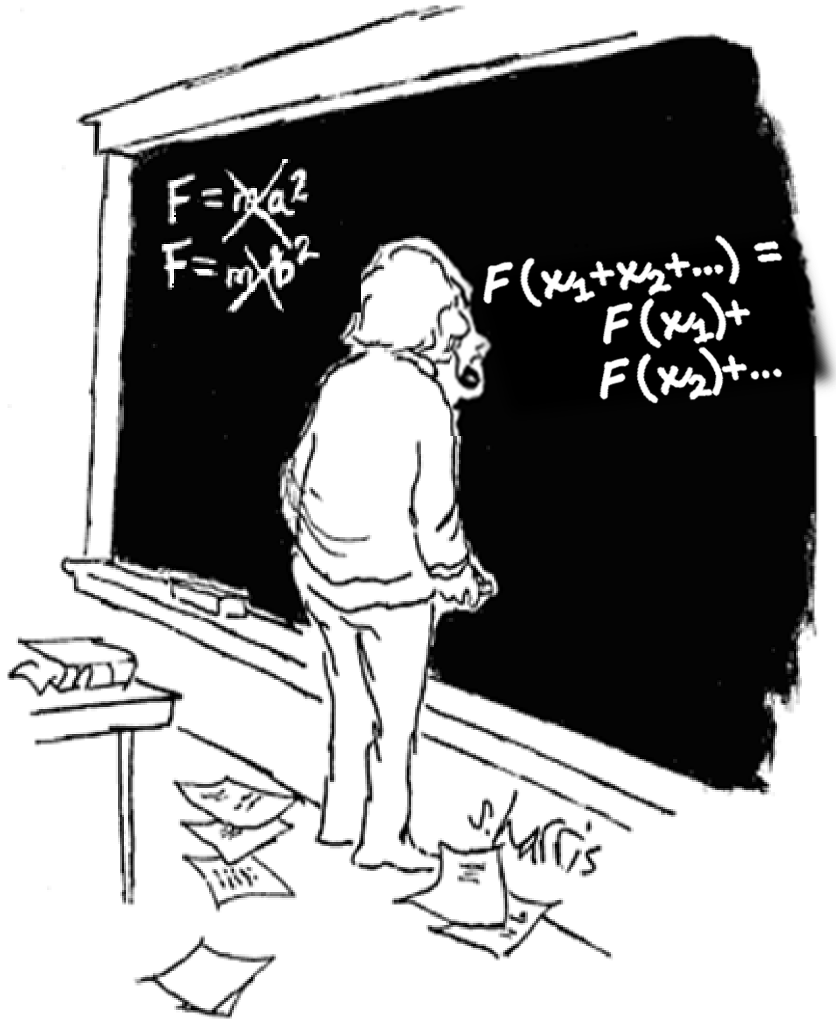
Conceptual Model for Polydiserse Materials

- Soils are linear systems that obey the additivity principle
- For all linear systems F(x) = y, where x is a stimulus and y is a response, the superposition of stimuli yields a superposition of the respective responses:

$$F(x_1 + x_2 + \ldots) = F(x_1) + F(x_2) + \ldots$$

- PSD of whole sample is then calculated from the distributions of, e.g., 2 components as:

$$f = p_1 f_1 + (1 - p_1) f_2$$



Challenges to Approach

- Particle Shape:
  - Assumption of spherical shape
  - Controls arrangement and packing thus mass-volume relationships
  - Individual property as fundamental as size
- Sample Size
  - Need PSD of very small samples
  - Requires precise determination using a rapid and reliable method with a high degree of precision
- Mineralogy
  - Affect geochemical properties
  - Transported aggregates are often polymineralic

Accounting for Mineralogy

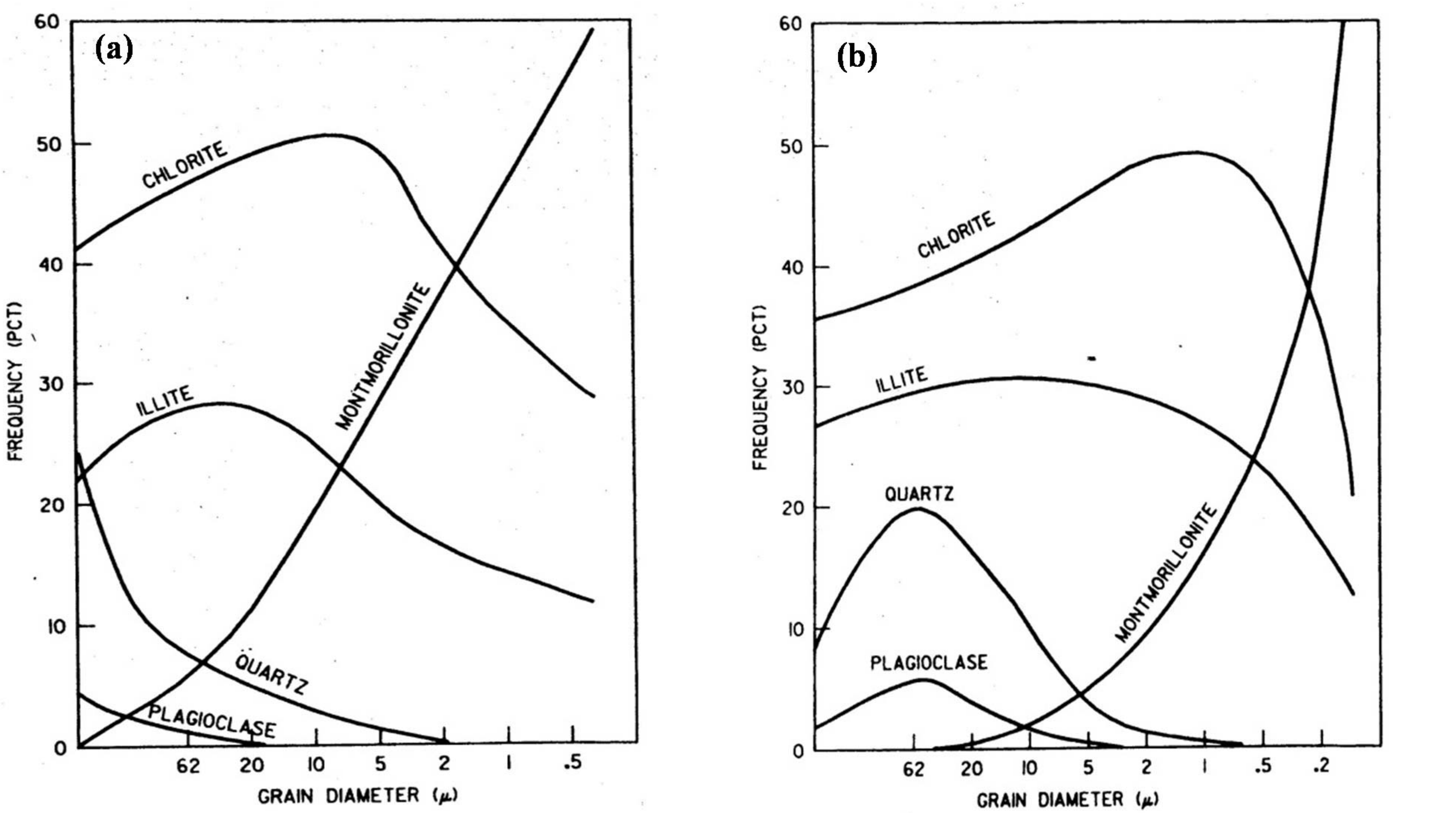


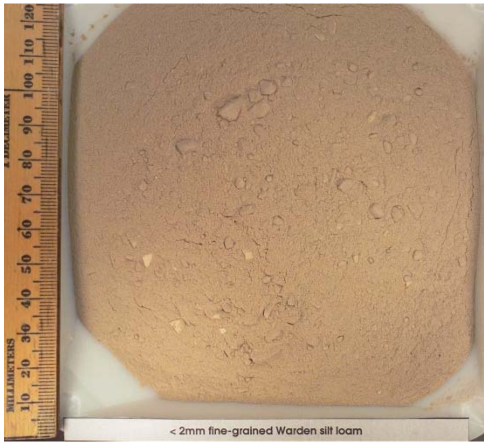
Figure 1.5: Mineralogy of Yukon River Sediment as a function of grain size for (a) fine material, and (b) coarse material (after Matthews, 2007).

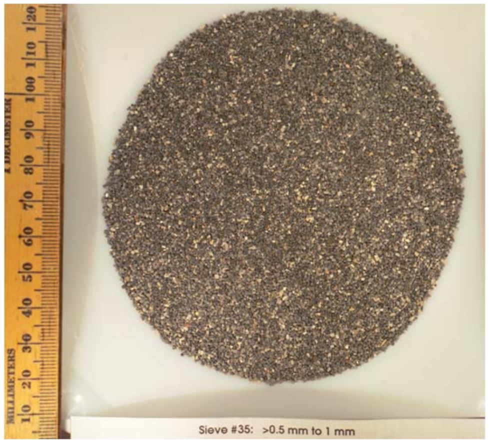
Solution to Most of My Problems

- Horiba LA 950 Particle Size Analyzer
  - Widest Range Available: 0.01-3000µm
  - Fastest sample analysis available
  - 60 seconds sample-to-sample
  - Rapid change from wet to dry analysis
  - Fully automated, modular sampling systems
  - Easy and cheap to repair even when no technician available provided Home Depot is open

Materials

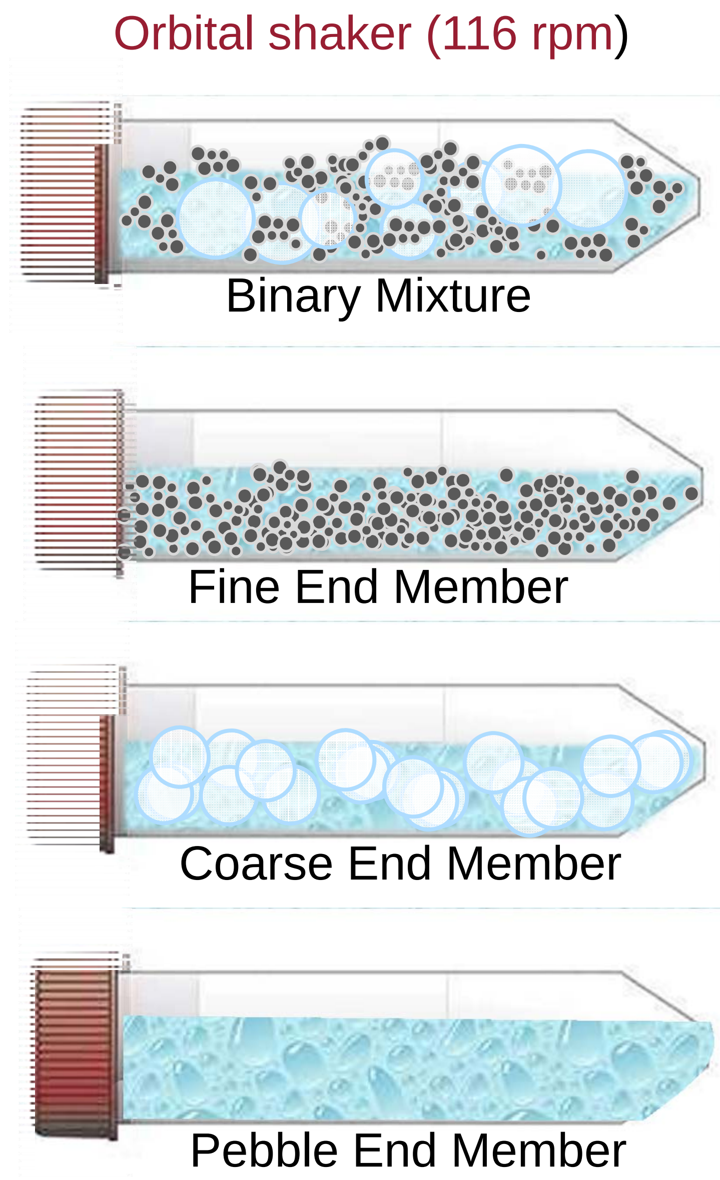
- Coarse and fine fractions
  - Silt loam
  - Accusand (.84-.54 mm)
  - Silica beads (4.95 mm)
  - Pebbles (4-5.6 mm)
- Binary mixtures
  - Triplicate samples
  - 10% increasing fines
  - Solution-Solid ratio 2:1
- Synthetic Groundwater
  - pH = 8.0
  - $[CO_3] = 1.05 \times 10^{-3}$ mol L^{-1}
  - 100 ppb U(VI)





Uranium Sorption Experiments

- Design
  - Contact times: 0.083, 0.167, 0.33, 0.5, 1, 2, 4, 8, 16, 32, 64, 128, 256 hrs
  - Supernatant separation using 15 minute centrifugation
  - Supernatant filtered (0.25 µm) and analyzed for U and pH
- Kinetics
  - 9% Silt + 91% Marbles
  - 51% Silt + 49% Marbles
  - Pebble and Silt end members
- Sorption on Binary Mixtures
  - Accusand, Marbles, Silt, Pebbles
  - Contact times: 24 hrs, 5 days

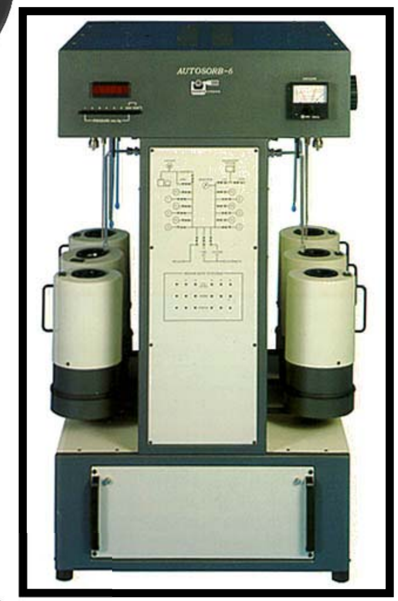


Analytical Methods

- **Solid Phase**
  - Continuous particle size distribution by laser diffraction
  - Surface area <u>measured</u> by N_2 gas adsorption
  - Surface area <u>calculated</u> by geometric method:

$$SA_{GEO} = \sum_{i=1}^{ns} \frac{\alpha_i p_i}{\psi_i \rho_i d_i} + \sum_{i=ns+1}^{n} \frac{\alpha_i p_i}{\psi_i \rho_i l_i}$$

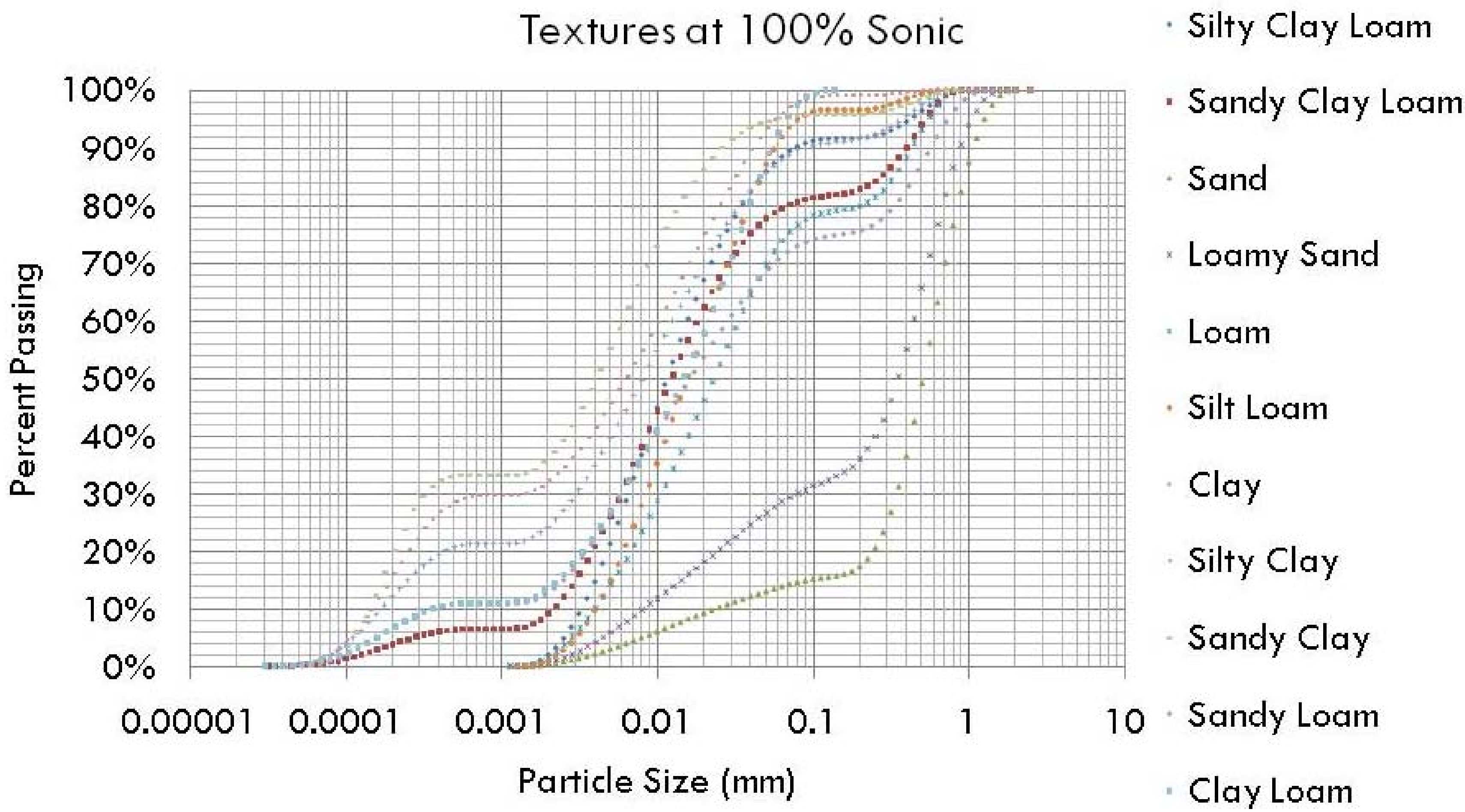
  - Surface topography and chemical composition by optical and scanning electron microscopy



Quantachrome Autosorb 6B Surface Area and Pore Size Analyzer

Horiba LA-950 laser diffraction analyzer

Laboratory Studies with Model Mixtures



How Do we Use these Data?

To Describe Particle Size Statistics

Folk and Ward (1957) introduced the Graphic Method to estimate the various statistical parameters describing a grain size distribution using only percentiles taken from cumulative frequency

Median

$$Md = \phi_{50}$$

Mean

$$M = \frac{\phi_{16} + \phi_{50} + \phi_{84}}{3}$$

Standard deviation

$$\sigma = \frac{\phi_{84} - \phi_{16}}{4} + \frac{\phi_{95} - \phi_{5}}{6.6}$$

Skewness

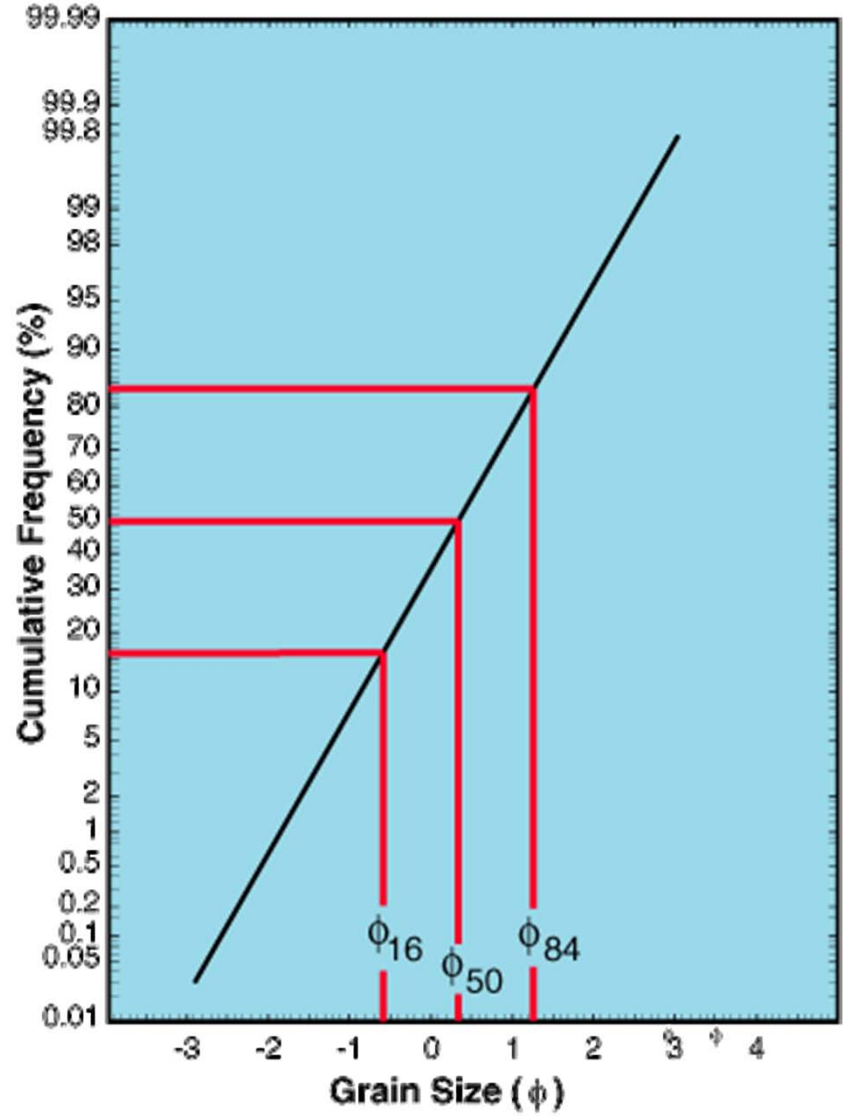
$$Sk = \frac{\phi_{84} + \phi_{16} - 2\phi_{50}}{2(\phi_{84} - \phi_{16})} + \frac{\phi_{95} + \phi_{5} - 2\phi_{50}}{2(\phi_{95} - \phi_{5})}$$

Kurtosis

$$K = \frac{\phi_{95} - \phi_{5}}{2.44(\phi_{75} - \phi_{25})}$$

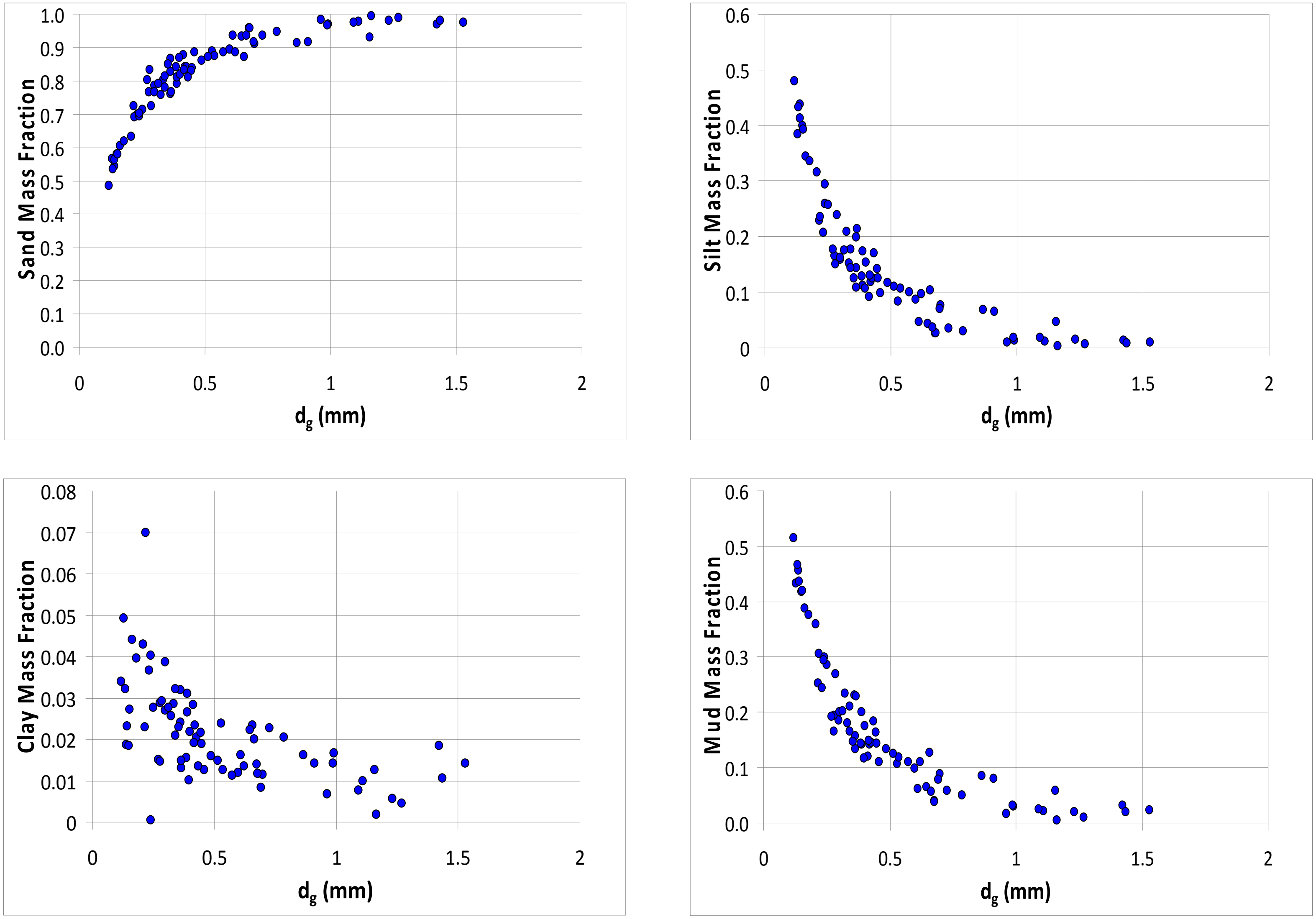
Example Calculation of the Mean

$$M = \frac{\phi_{16} + \phi_{50} + \phi_{84}}{3}$$



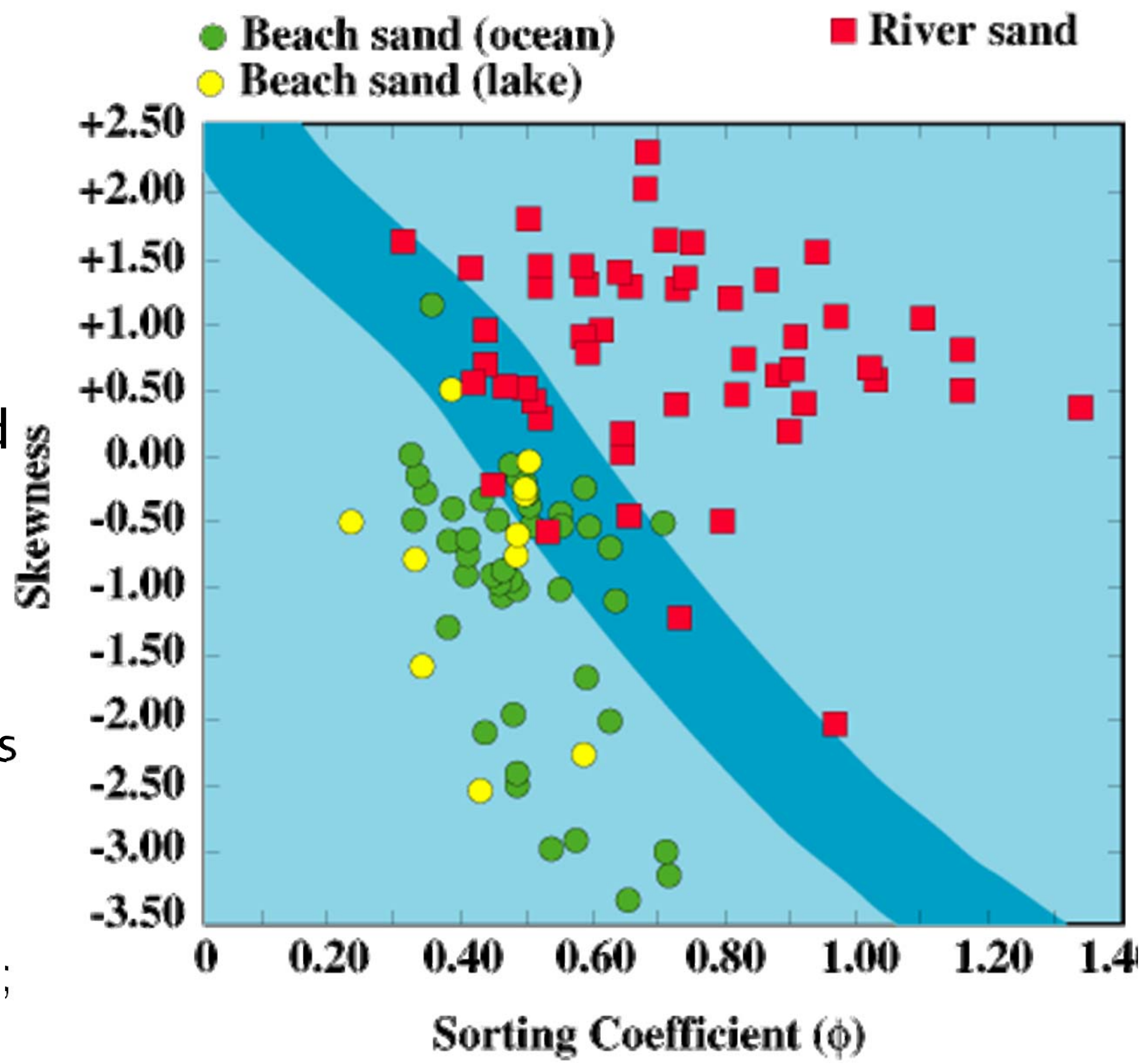
$$M = \frac{-0.59 + 0.35 + 1.27}{3}$$

Texture and Mean Diameter

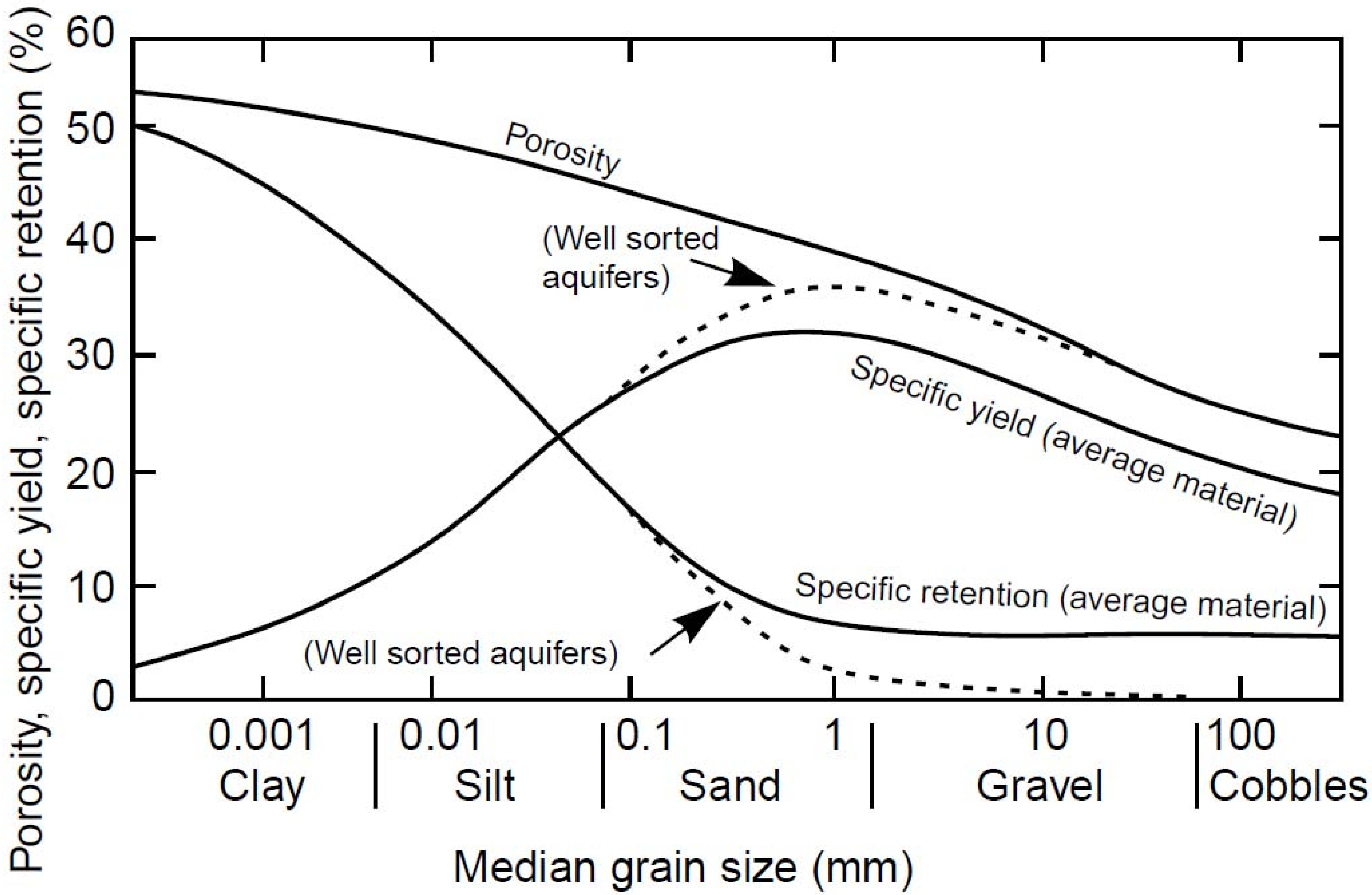


To Understand Depositional Environments

- Samples collected from rivers and beaches (lake and ocean)
- Skewness plotted against Sorting Coefficient
- Beach sands better sorted and with more common coarse tail skewness than river sands
  - Reflects difference in processes acting on rivers and beaches
  - Rivers carry wider range of sizes: large particles move in contact with bed; large volume of fine particles in suspension
  - Poorly sorted; rich in fine particles (+ve skewness).



Particle Size and Water-Storage in Alluvium

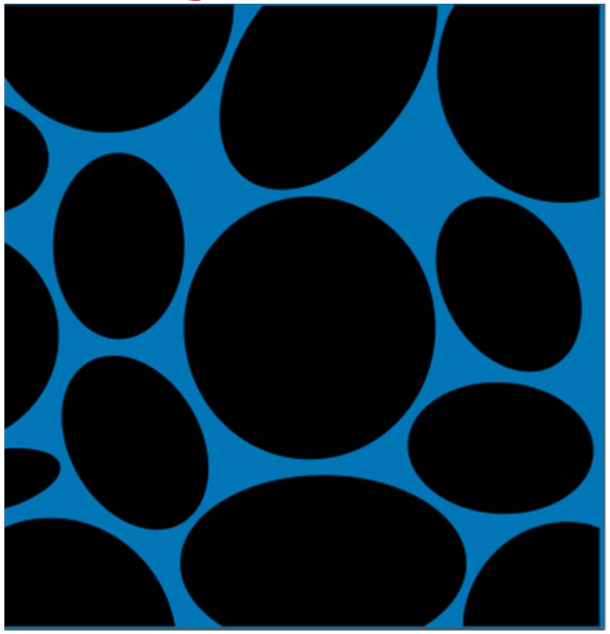


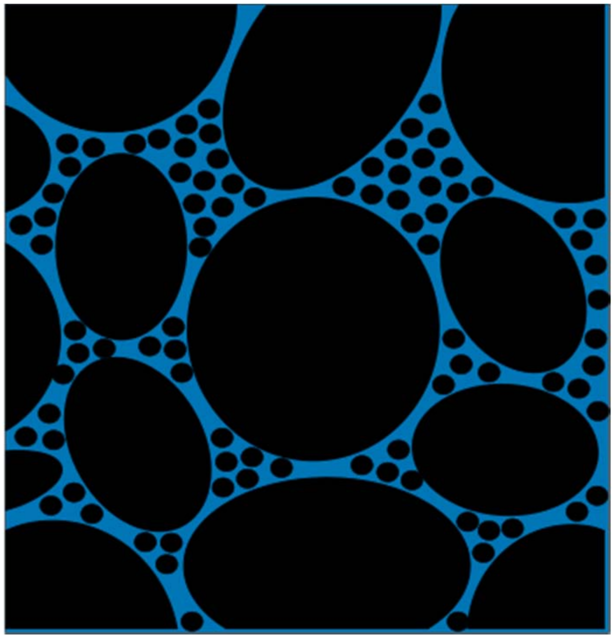
Particle Size and Porosity

- Typical sediment made up of Spheres of different sizes
  - Small spheres can fill in pore throats formed by larger spheres
  - Result is a lower porosity

$$n = 0.255(1 + 0.83^{C})$$

$$C = \frac{d_{60}}{d_{10}}$$



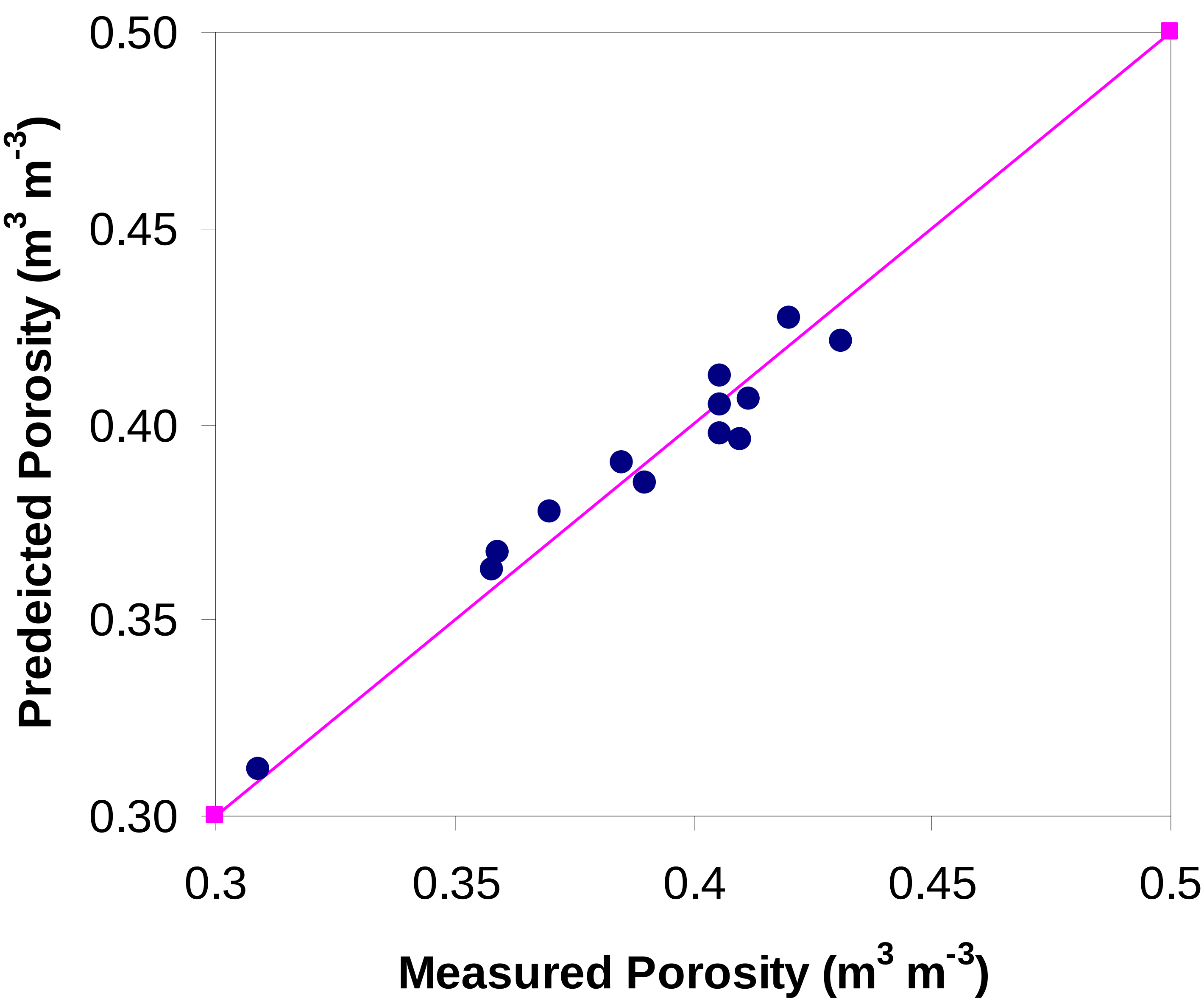


- The porosity, ϕ_b, of a multicomponent mixture may then be calculated as:

$$\phi_b = f(X_1, X_2, \ldots, X_n; d_{p_1}, d_{p_2}, \ldots, d_{p_n}; \phi_1, \phi_2, \ldots \phi_n)$$

where X_i is the fractional solid volume of the i^{th} component.

Porosity Predicted from Particle Size Distributions



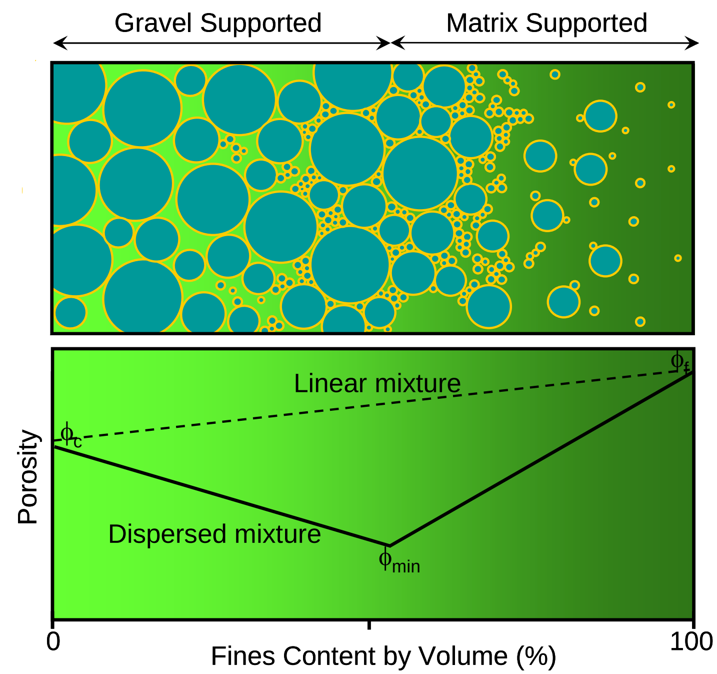
Particle Size and Permeability

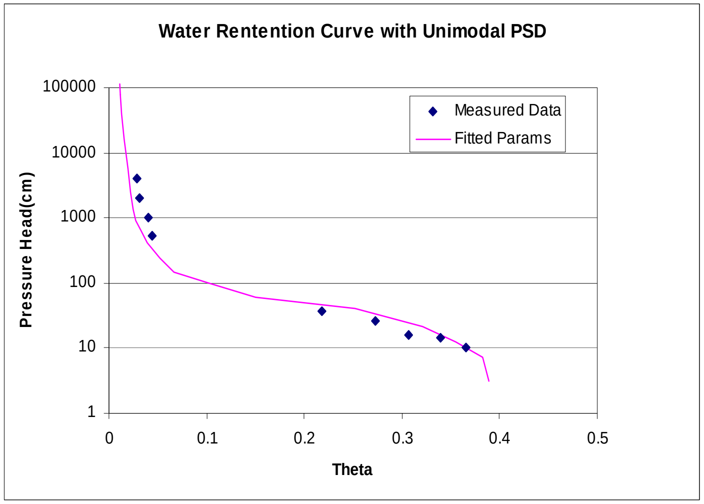
Equation	Reference
$K = C(d_{10})^2$	Hazen (1911)
$K = (9.66E-04)(760 d_g^{\ 2}) EXP(-1.31\sigma_g)$	Krumbein and Monk (1942)
$K = \left(\frac{\rho g}{\mu}\right)\frac{d^2\phi^3}{180(1-\phi)^2}$	Kozeny-Carman (in Bear, 1972)
$K = \left(\frac{\rho g}{\mu}\right)\frac{\phi^3}{C_0 S_{sa}^{\ 2}(1-\phi)^2}$	Kozeny-Carman (in de Marsily, 1986)
$K = \left(\frac{\rho g}{\mu}\right)\frac{\phi^3}{C_T T(S_{sa})^2}$	Kozeny Equation, modified by Collins (1961)

C_0 = factor reflecting pore shape and packing in the Kozeny-Carmen eqn. [-]
C_T = factor reflecting pore shape and packing in Kozeny eqn, mod. By Collins [-]
C = factor in the Hazen equation [T/L]
d_{10} = grain diameter for which 10% of particles are smaller [L]
d_g = geometric mean grain diameter [L]
d = representative grain diameter [L]
g = gravitational acceleration [L/T^2]
K = hydraulic conductivity [L/T]
ϕ = total porosity, accounting for compaction [-]
μ = dynamic viscosity [M/LT]
ρ = density [M/L^3]
σ_g = geometric mean standard deviation [L]
S_{sa} = surface area exposed to fluid per unit volume of solid medium [1/L]
T = tortuosity [-]

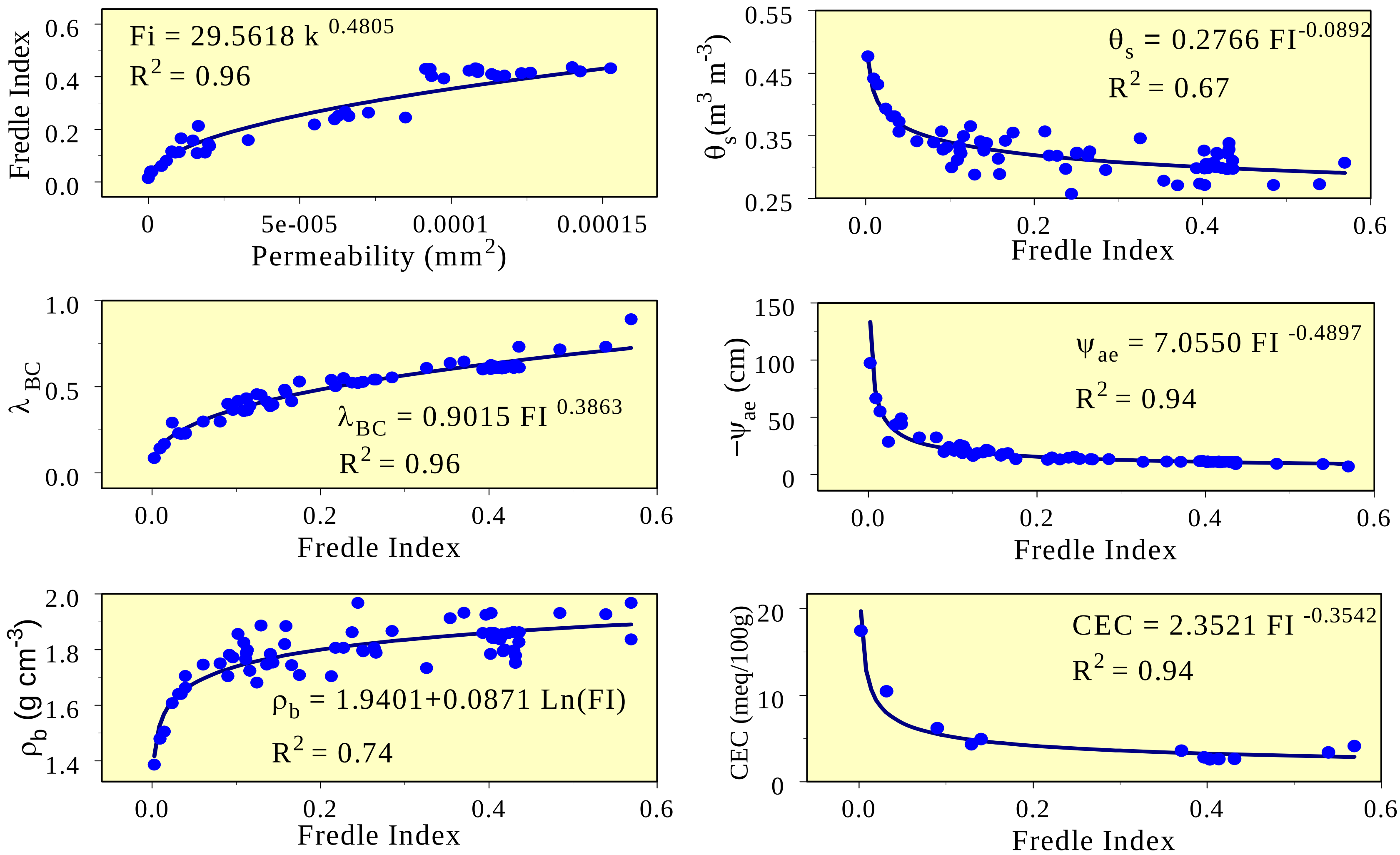
Hydraulic Properties From Particle Size Distributions

- Microstructure Characterization
  - grain parameters controlling particle arrangement and packing
- Pore Structure
  - Identify individual particles and arrangement
  - Simulate packing
- Feasibility established with simple case of binary mixture (coarse + fine)
  - Extend binary fractional packing concept to the *n* fractions of the Udden-Wentworth particle-size scale
  - Robust approach for upscaling basic parameters derived from grain size distributions
  - Allows correction for sizes > 2000 micron



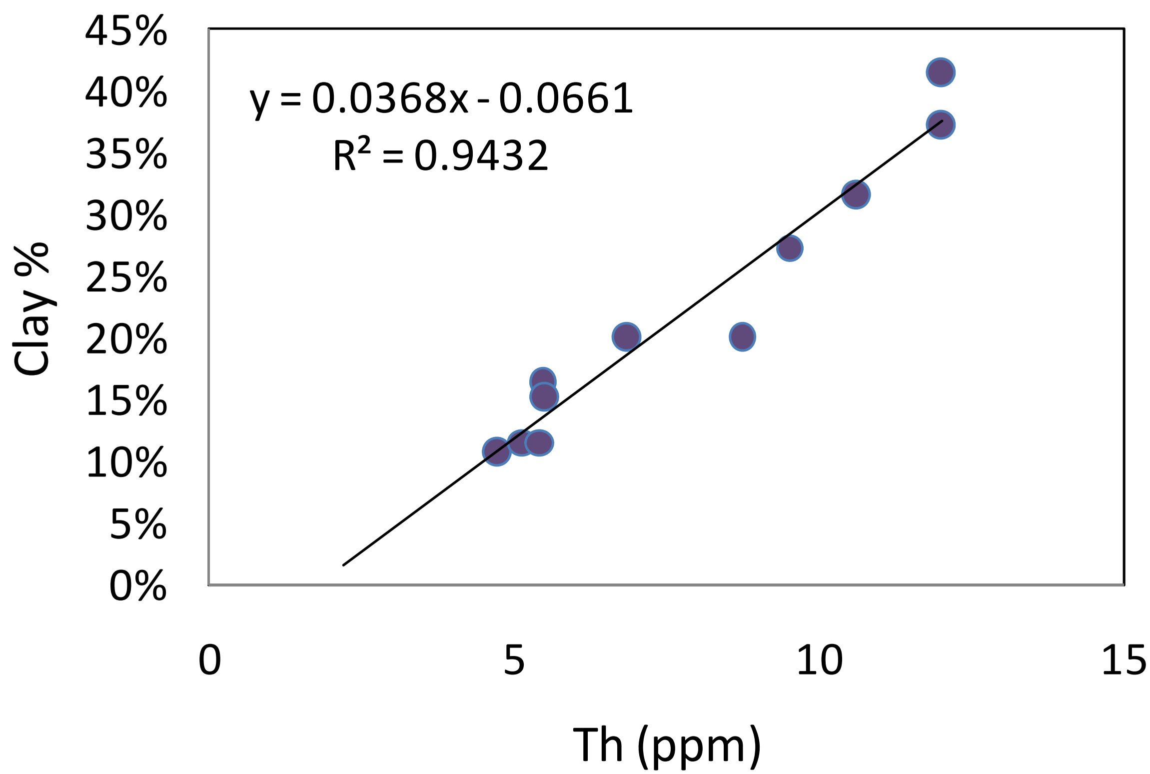


Hydraulic Properties and Texture

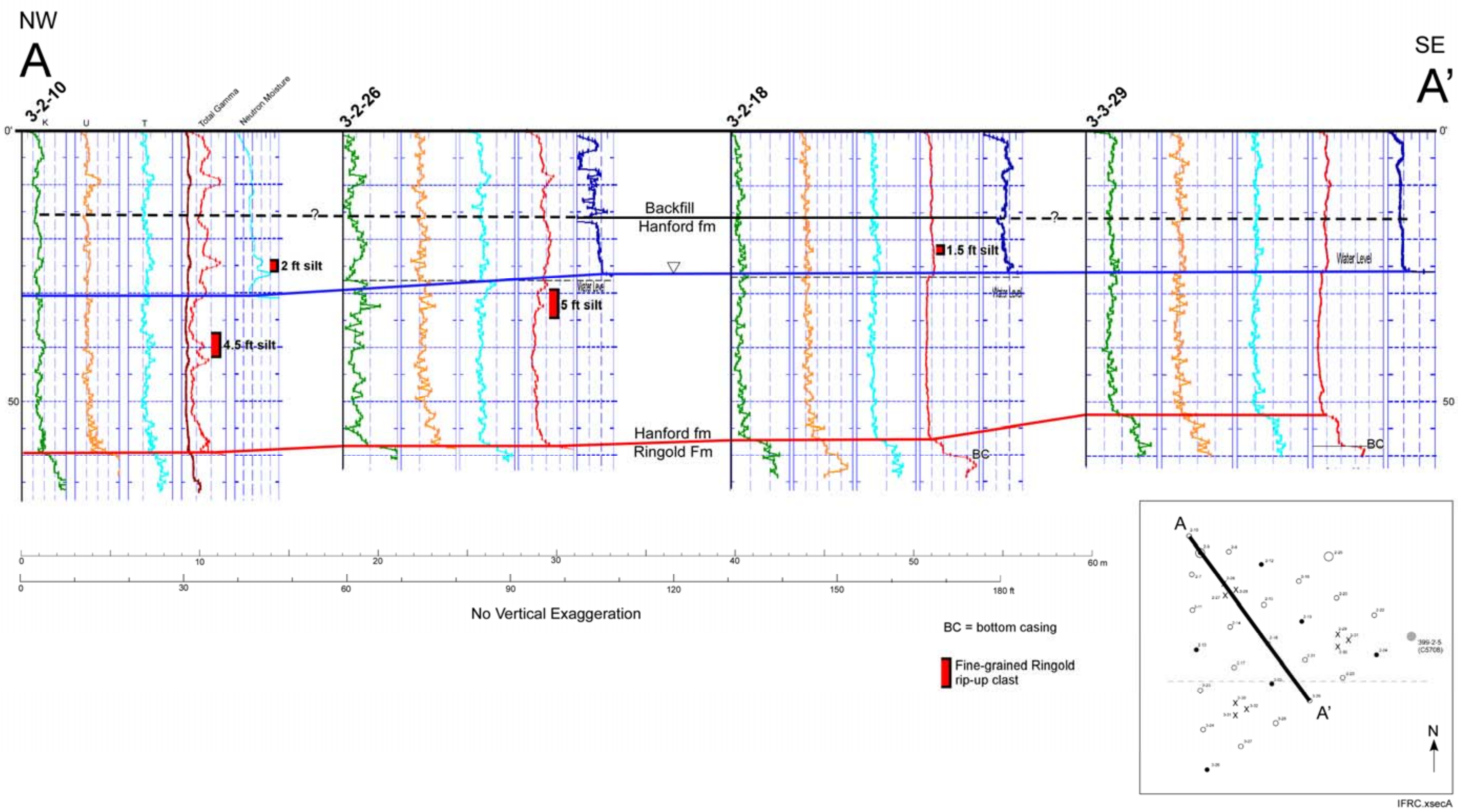


Facies Identifcation Particle Size Distributions

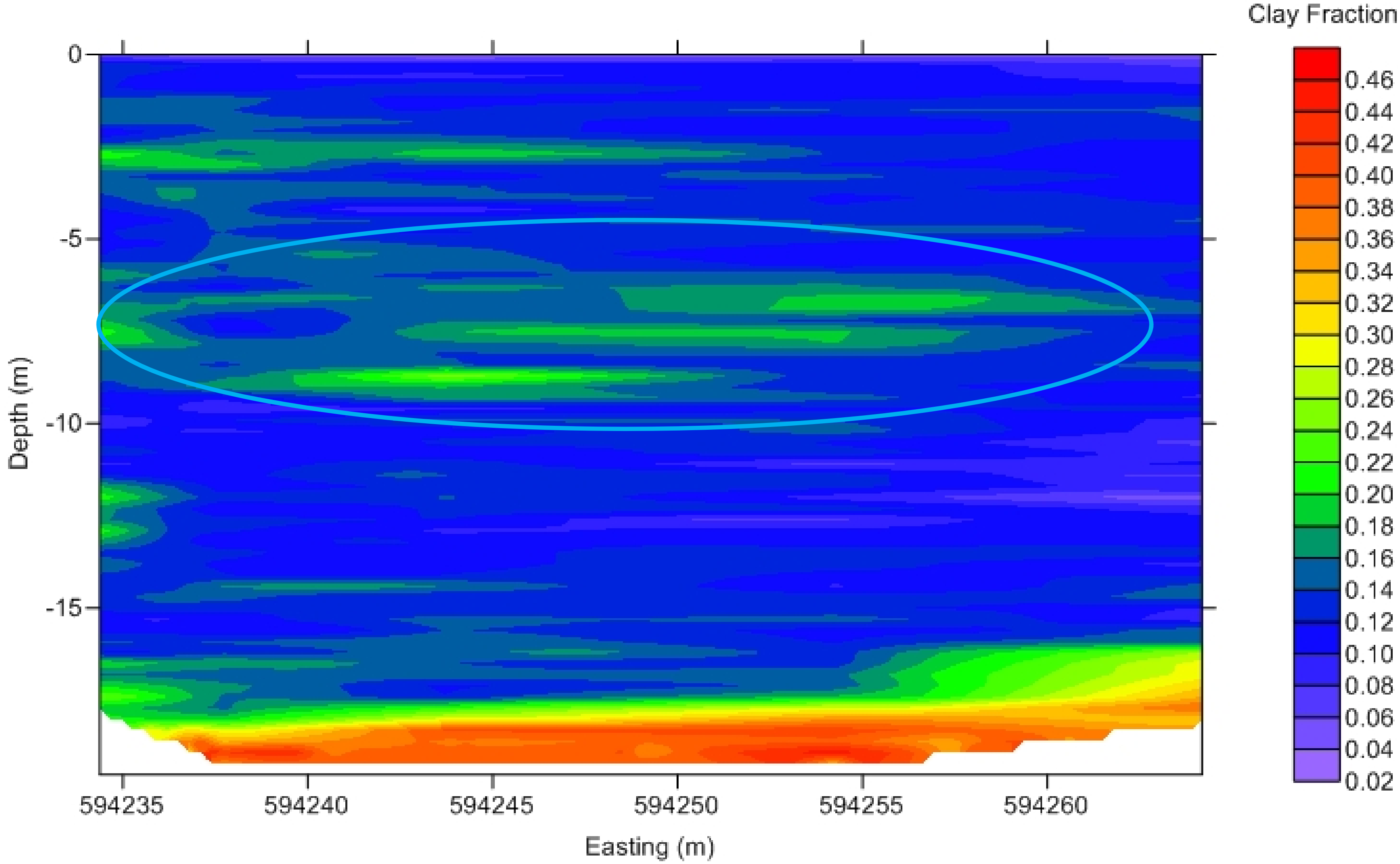
- Identification of Lithofacies
  - Th/K expresses relative K enrichment as indicator of clay mineral species and useful for distinguishing architectural elements (e.g. Coarse vs. fine) grain parameters controlling particle arrangement and packing



Multi-scale Heterogeneity

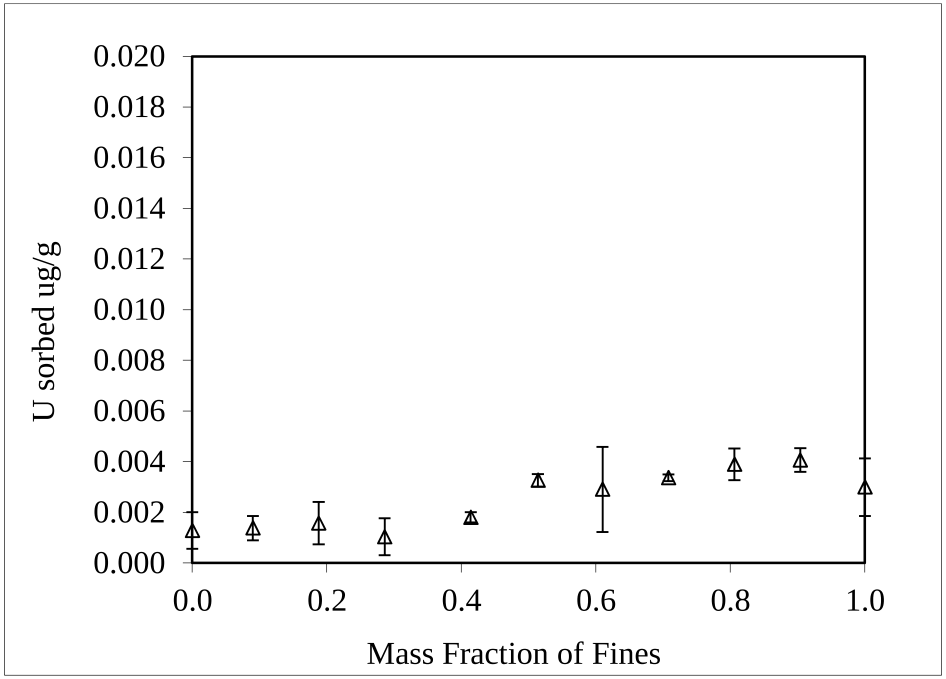


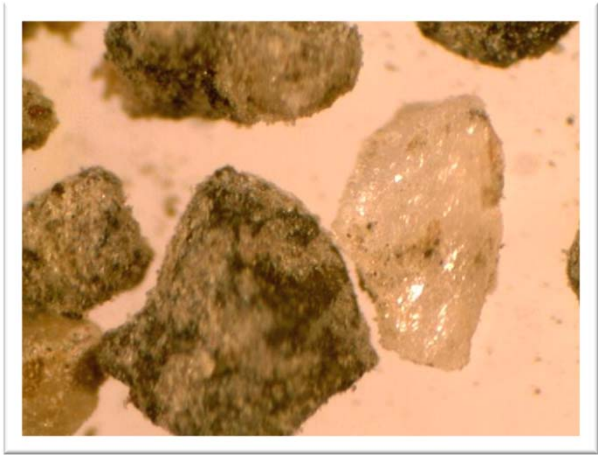
Transect A-A' Clay Content



Sorption of Marbles – Accusand

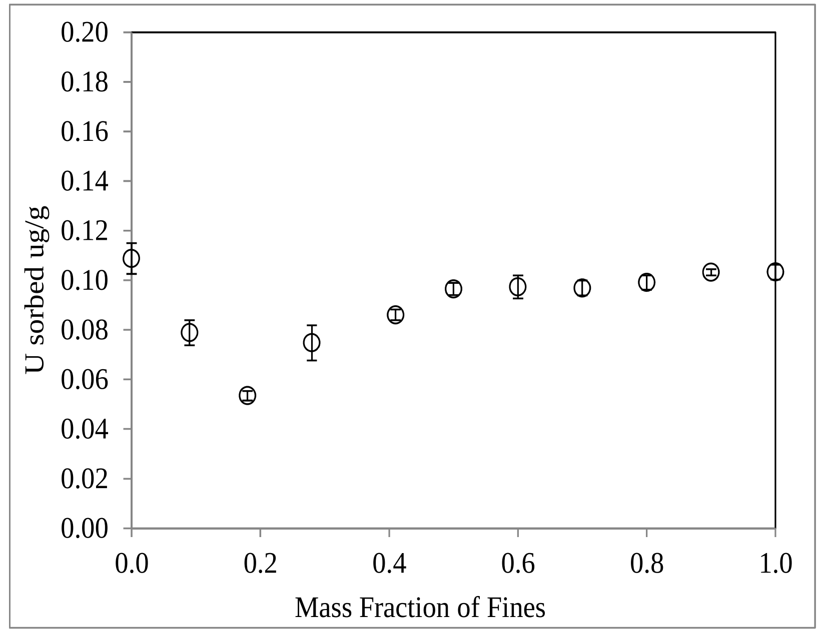
- Accusand and marbles are primarily silica
  - No sorption expected
- Low but non-zero sorption with standard high SE
  - no change for fines < 40%
  - Nonlinear after 40%
- Higher sorption in accusand due to:
  - rough surfaces
  - metal-oxide coatings
  - organic matter

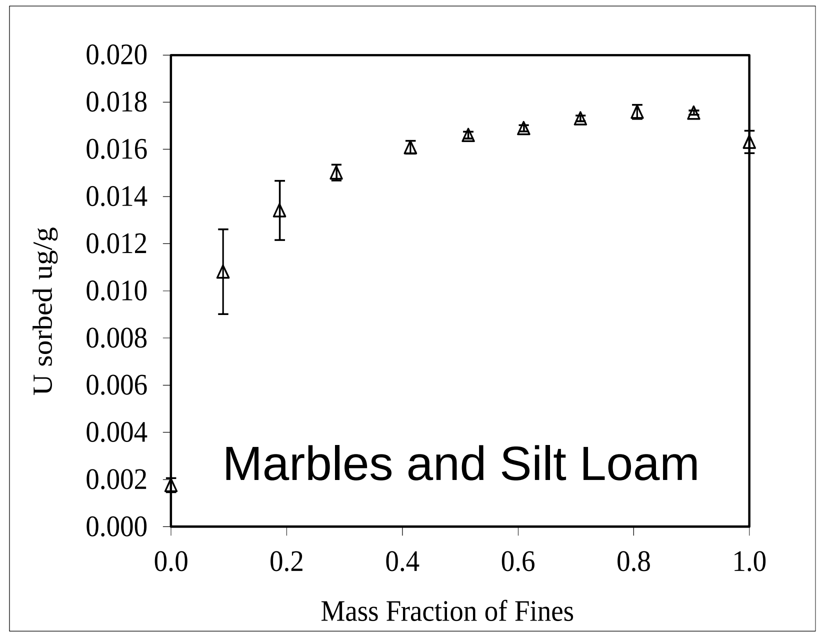




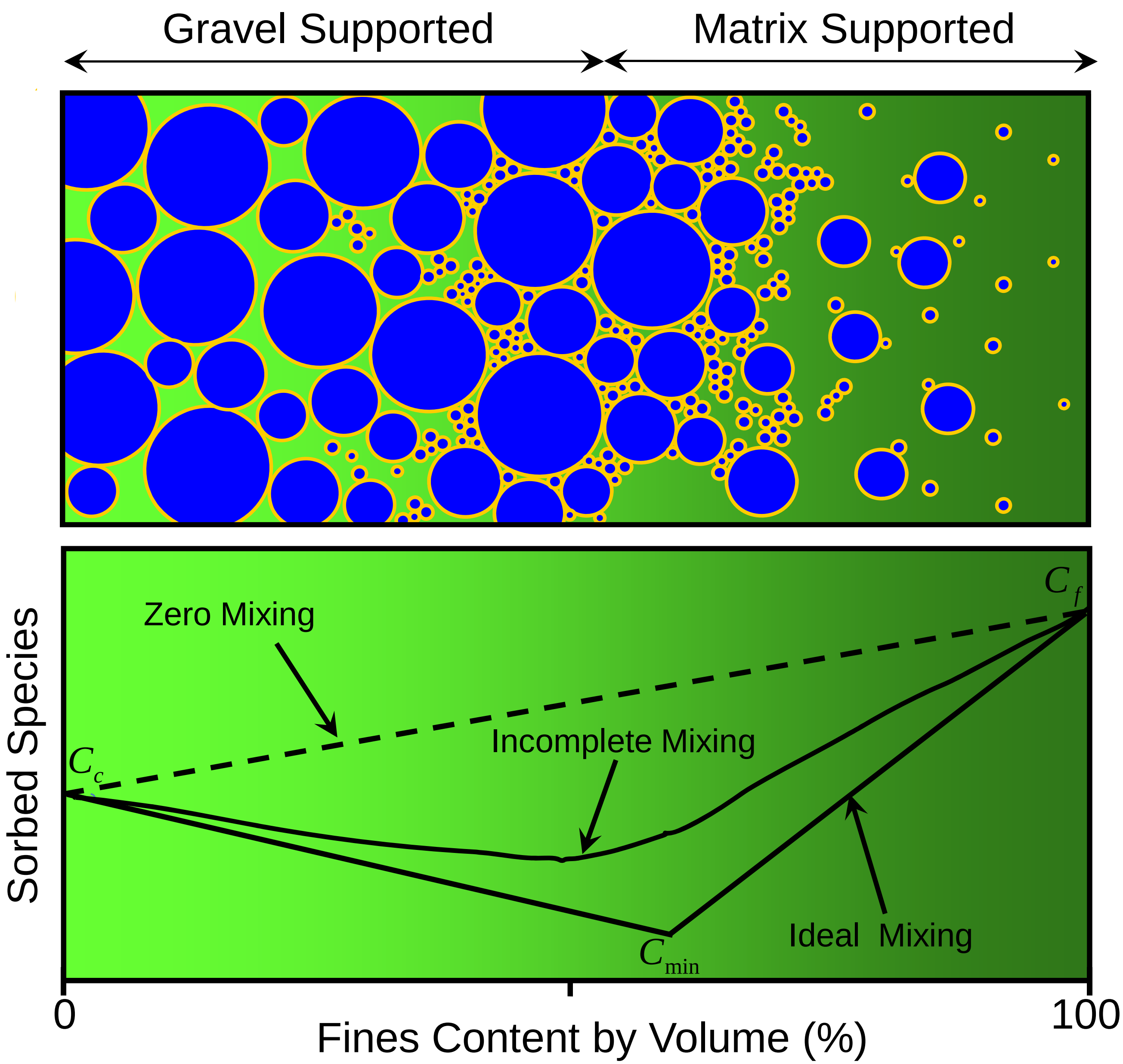
Sorption of Pebbles - Silt Loam

- Large amount of U(VI) sorbed by pebbles
- Initial decrease in sorption on the addition of silt loam
  - Likely blocks access to fractures on pebbles
- Classic v-shaped curve indicative of incomplete mixing
- Pebbles sorption inconsistent with current conceptual models
  - negligible surface area
  - no contribution to sorption
  - gravel correction based on linear dilution (zero mixing)



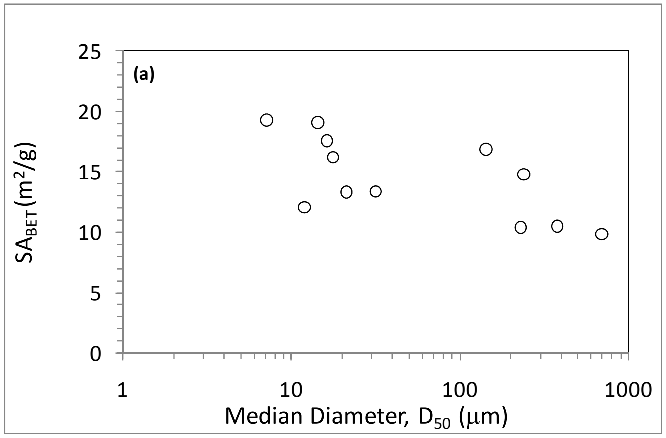


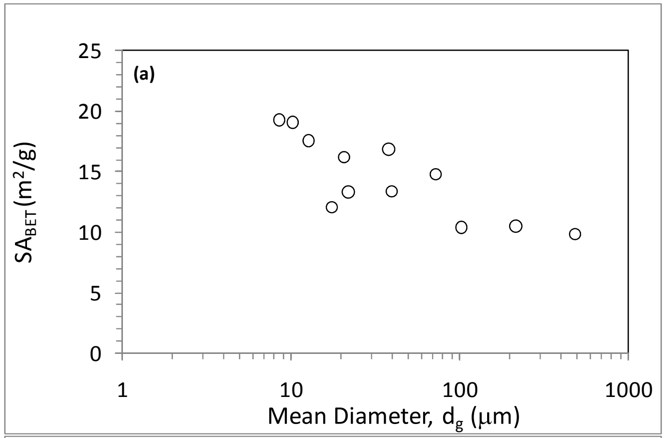
Partial Mixing Model

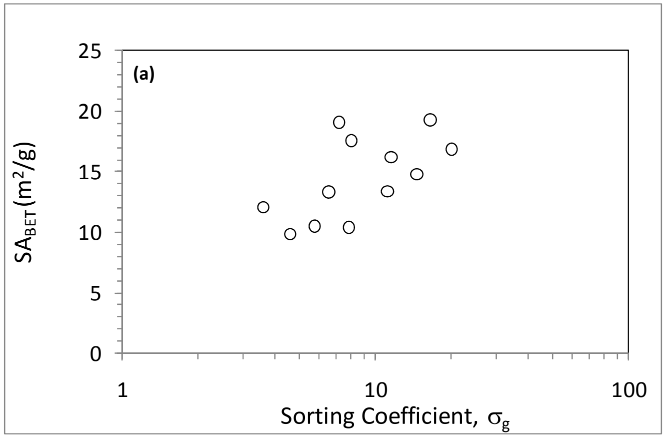


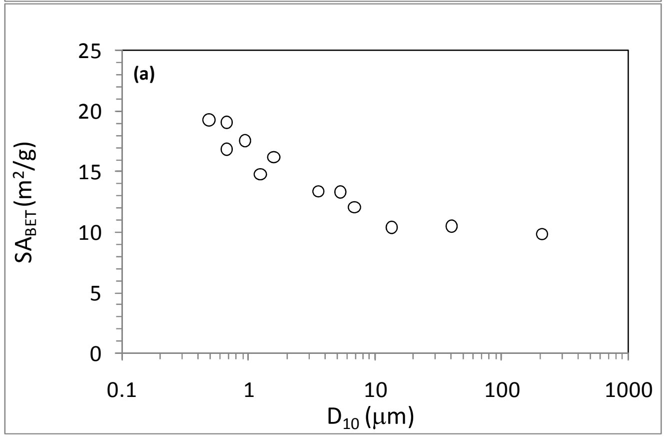
Surface Area vs. Size Statistics

- Surface area measurements in mixtures show:
  - nonmonotonic decrease with increasing D_{50}
  - decrease with geometric mean diameter, d_g
  - Well-behaved decrease as D_{10} (measure of fines) increases
  - Increase with sorting coefficient
- Geometric method assumes smooth spherical particles and not applicable to natural materials



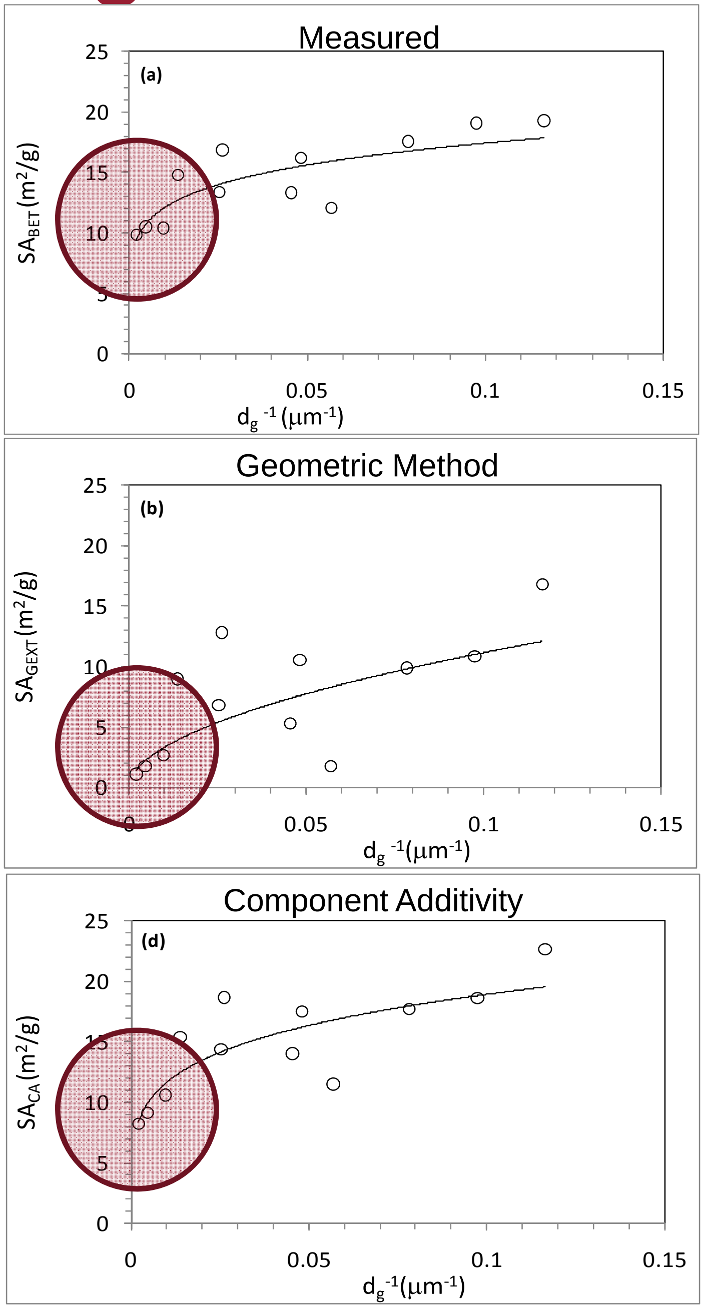






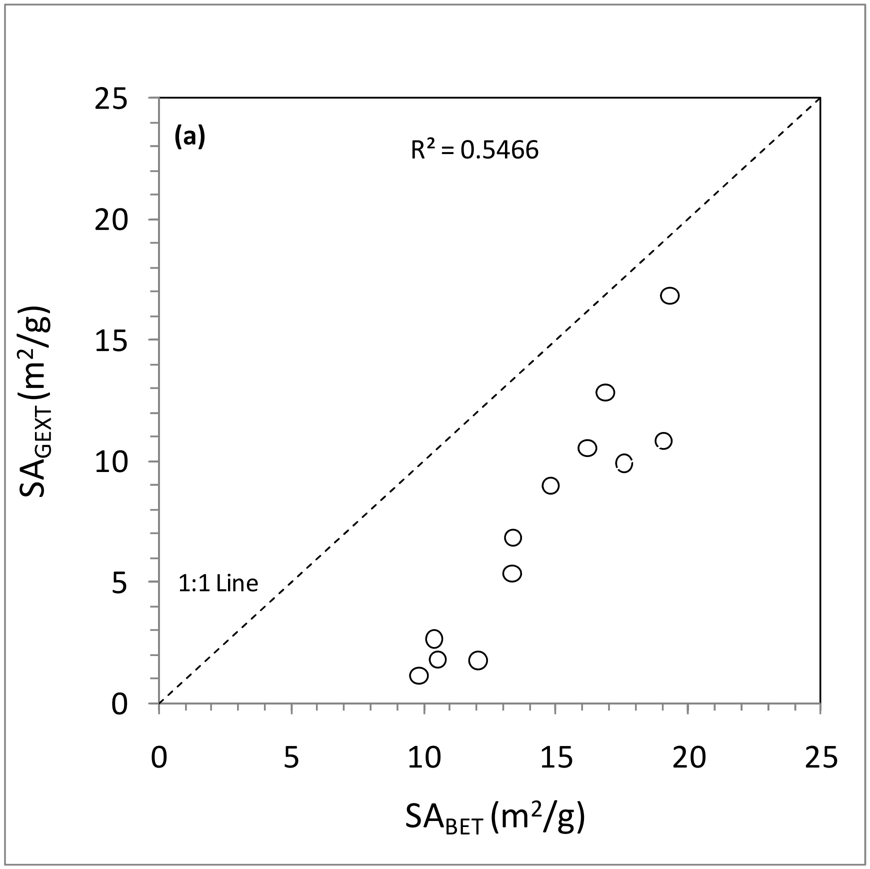
Effects of Surface Roughness

- A plot of SA(d_g^{-1}) should be linear
  - intercept = internal SA, SA_{int}
  - Slope dependent on roughness, i.e., $\lambda_{ext} a/\rho$
- Non-zero Intercept
  - indicates $SA_{int} > 0$
  - inconsistent with the smooth, nonporous spherical particle assumption
- Nonlinear relationship
  - Suggest that SA_{int} and λ_{ext} both dependent on d_g

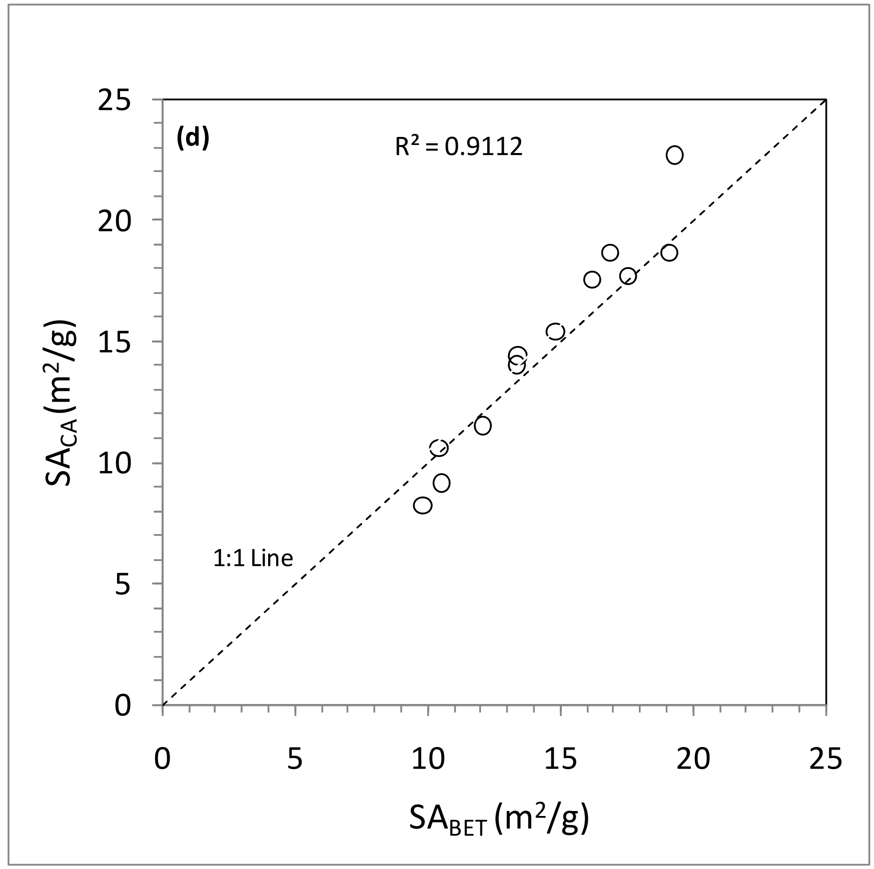


Comparison of PSD-based SA Methods

Geometric Method



Component Additivity Method



Conclusions

- Primary properties of sedimentary structures are largely controlled by the distribution of facies, which is in turn controlled by the depositional environment and grain size distributions
- Particle size is a fundamental property of any sediment, soil or dust deposit
- Shape and mineralogy can be assumed fixed for a depositional environment
- High resolution particle size distributions of < 3000 micron sediments and application of the principle of superposition allows accurate estimation of critical properties
- Data most easily obtained with the Horiba LA-950

For More Details

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Thank-you