NAPLs, VOCs, Partitioning of contaminant

NAPLs: Non-Aqueous Phase Liquids

NAPLs: -organic liquids that do not mix freely with water
(sparingly soluble：難溶性).

-the most common contaminants:

-traveling large distances in subsurface

-difficulty in the ground water clean-up

intensive researches in the last two decades
regulations with respect to site remediation

Two classes of NAPLs

•LNAPLs (Lighter (than water) NAPLs)　　$\rho < \rho_w$ at 20- 25 °C typical soil temp.

 - immiscible petroleum hydrocarbon liquids

 - gasoline, fuel oils

 - Benzene, Toluene, Ethyl Benzene, Xylene (BTEX)　detection limits
　　　　　　　　　　　　　　　　　　　　　　2μg/L, *5μg/kg*

•DNAPLs (Denser (than water) NAPLs)　$\rho > \rho_w$ at 20- 25 °C

 - chlorinated solvents, trichloroethene (TCE),tetrachloroethane (PCE),
　(有機塩素系溶剤)　　　　etc.　　*for high-tech industry, cleaning*
　　　　　　　　　　　　　　　　　　　　　carcinogen　(発癌物質)

 - wood preserving process wastes (creosote)

 - coal tars

 - pesticides

Properties of NAPLs

　The distribution of NAPLs in the subsurface is the net results of coupled chemical and physical interaction between the NAPL, pore water, pore gases and porous medium.

　The chemical properties of NAPL, pore phase and porous medium, together with chemical equilibria determine the partitioning of the compounds between the various phase.
　(分配)

　The physical properties of the pore fluid and porous media determine the mobility of each phase.

Properties

　-specific gravity　(TCE:1.47)
　-aqueous solubility (TCE:0.1%)
　　(水溶性)　　　*immiscible, sparingly soluble*
　-viscosity (TCE:0.566cP)
　　　　　　more permeable than water
　-vapor pressure *(63kPa at 25ºC) cf. water 3.2kPa*
　　(蒸気圧)　　　*highly volatile same as Benzene*
　　　　　　VOCs (Volatile Organic Compounds)
　　　　　　　　(揮発性有機化合物)
　　　　　SVOCs(Semi-volatile Organic Compound)

p1-2

vapor pressure

The pressure exerted by vapor=the pressure of **the gas in equilibrium** (saturated condition) with **respective to the liquid or solid** at a given **temperature**. It represents a compound's **tendency to evaporate** and is essentially **the solubility of the gas**.

Raoult's law(ラウールの法則) giving the equilibrium partial pressure of a component of a NAPL mixture in the atmosphere above the mixture.

$$P_a = x_a P^n_a \quad (1)$$

P_a:the partial vapor of the NAPL component in the gas phase
x_a: the mole fraction of the NAPL component in the mixture
P^n_a:the vapor pressure of the NAPL component in pure-phase form

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Four distinct phases

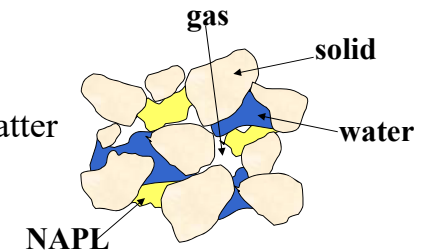- **gas phase in vadose zone**
- **solid phase:**
  soil grains and organic matter
- **aqueous phase:** *polar*
- **NAPL phase**: *nonpolar*

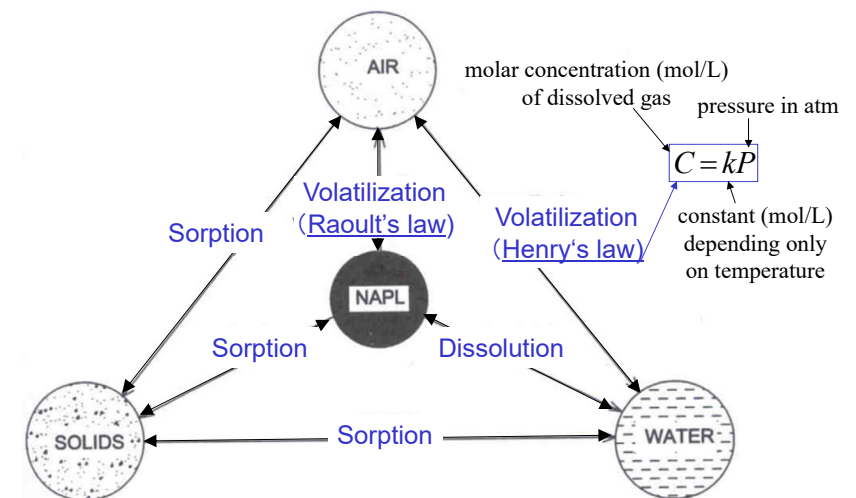## Existence in subsurface

- dissolution into aqueous phase
  （溶解）
- volatilization into gas phase
  （揮発、気体）
- NAPL phase as entrapped residual
  （液体)
- partition (sorption) onto solid phase
  （吸着)

Mass transport process involved in partitioning of NAPLs into solid, water, and gas phases

molar concentration (mol/L) of dissolved gas

pressure in atm

$$C = kP$$

constant (mol/L) depending only on temperature

Volatilization (Raoult's law)

Volatilization (Henry's law)

Sorption

Dissolution

Sorption

Mechanism of migration by DNAPLs

In vadose zone

 - NAPL plume move downward by **gravity force**.

 - **volatilized DNAPLs** move more laterally.

In saturated zone

 - moving rate slows due to **the resistance of water** (displacing water) and **buoyancy force**

- some chemical will dissolve in the **flowing ground water**, forming **contaminant plume**

- remainder will move downward and create **residual NAPL zone**

- Mobil DNAPL will reach to **impervious layer**, spreading laterally as **liquid pool**.

Conceptual model of point source contamination
- DNAPL case -

by gravity

volatilization

GWF

sorption

dissolution
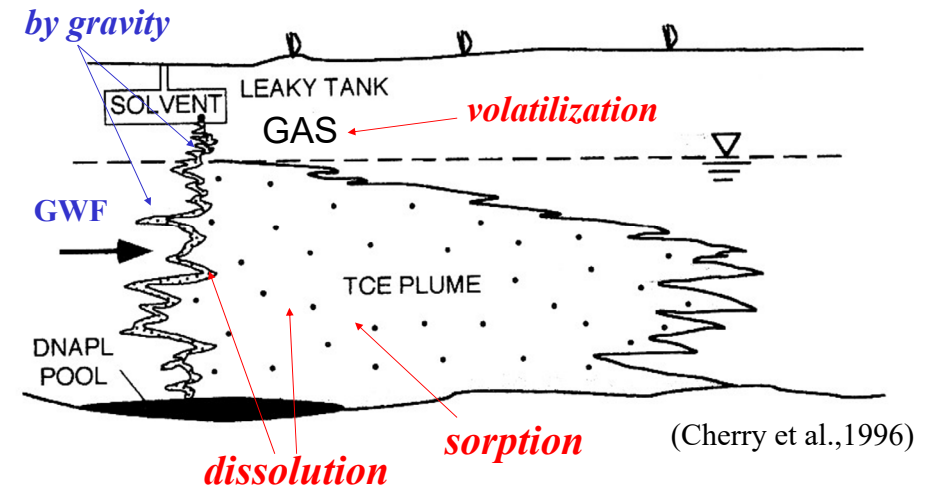
(Cherry et al.,1996)

Conceptual model of point source contamination
- Non-NAPL case -

GWF

dissolution   sorption
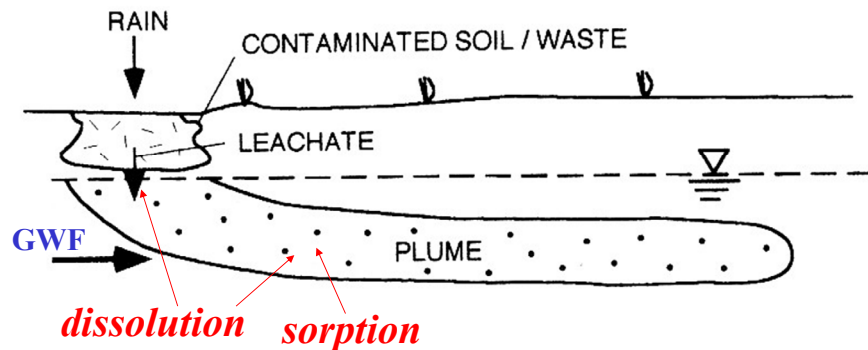
(Cherry et al.,1996)

Mechanism of migration by LNAPLs

In vadose zone

 Similar to DNAPLs

In saturated zone

 - spreads out laterally **by gravity**, forming a "**pancake" or lens**" on the **top of water table**.

- after formation, the lens will migrates primarily in the direction of **natural ground flow**.

- **Change of ground water** table due to seasonal fluctuations or pumping may enhance the spread vertically and area of contamination.

Conceptual model of point source contamination
- LNAPL case -
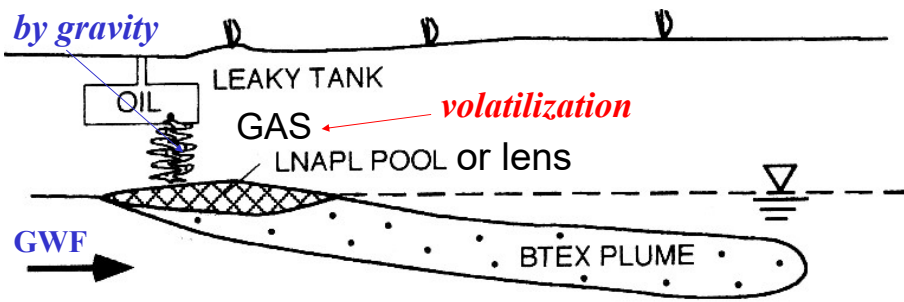
(Cherry et al.,1996)

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3 important migration pathway of NAPLs

- Vapor-phase transport
  wide spreading and diluting

- Aqueous-phase transport as solute with in contaminant plume
  faster and spreading
- NAPL phase transport
  slower but high concentration, source of CM

Vapor phase transport

The vapor transport depends on vapor pressure of chemical.

gas law $$\frac{n}{V} = \frac{p}{RT} \quad (2)$$

n: mole number
V: volume
p: pressure
R: universal gas constant
T: absolute temperature

Volatilization:
  volatile: vapor pressure more than 1.3×10^{-3} atm
  semi volatile: $1.3 \times 10^{-3} – 1.3 \times 10^{-13}$ atm

*Note: accuracy in the concentration measurement in dissolved phase

  Volatilization can be related to Henry's constant.

Migration of gas: advection and dispersion in **vadose zone**, larger than saturated zone.

vapor extraction method

Henry's law

Volatilization of dissolved organic solutes from water depends:
Henry's law, relating the aqueous-phase concentration of a chemical to its partial pressure in the gas phase.

$$P_x = H_L C_x \quad (3)$$

P_x: partial pressure of gas (atms at a given temperature)
C_x: equilibrium concentration of the gas in solution (mol/m^3,mol/L)
H_L: Henry's law constant in atm/(mol/m^3 , mol/L)

The larger Henry's law constant is, the greater volatilization from the soil or water.

  H_L less than about 3×10^{-7} atm/(mol/m^3 water): non-volatile
  For $H_L > 10^{-7}$ atm/(mol/m^3 water), volatilization might be a significant in mass-transfer mechanism.

Aqueous phase transport

The propensity of aqueous phase transport depends on aqueous (water) solubility and ground water flow.

Magnitude of aqueous solubility of NAPLs is a few order difference, depending on   *p1-2*
polarity, temperature, salinity, dissolved organic matter.
(極性)   (塩分濃度)   (溶解有機分)
Movement is caused by advection and dispersion.

dilution in case of enough travel distance
(希釈)

NAPL Phase transport
NAPL in water flow

When a wetting fluid (such as water) displaces a nonwetting fluid (such as oil) in a porous medium, a proportion of the nonwetting fluid will remain entrapped in the medium pores once the wetting front has passed. In general, the nonwetting fluid is trapped in the form of disconnected blobs or ganglia whose complicated morphology and distribution are dependent upon (i) the geometry of the pore space; (ii) fluid-fluid properties such as **interfacial tension**, **density contrast**, and **viscosity** ratio; (iii) the **fluid-solid interfacial** properties affecting wetting behavior; (iv) the applied pressure gradient; and (v) gravity.

During stable fluid displacement, nonwetting phase (NAPL) entrapment is caused by **capillary forces** and overcome by **viscous pressure forces** and/or **buoyancy forces** (Mayer and Miller, 1992). The ratios of viscous pressure to capillary forces and gravity to capillary forces are usually expressed as dimensionless groups, known as the **capillary number** (C_a) and the **Bond number** (B_o), respectively

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## Capillary number: C_a and Bond number: B_o

$$C_a = \frac{v\mu}{\sigma}$$

$$B_o = \frac{\Delta\rho g D^2}{\sigma}$$

v: displacing (Darcy) fluid velocity,
μ: displacing fluid viscosity,
σ: interfacial tension,
$\Delta\rho$: fluid density contrast,
g: gravitational acceleration,
D: average particle radius of the porous medium

Above a certain upper, critical Capillary or Bond number, no capillary trapping occurs, and the residual nonwetting phase saturation is close to zero. Conversely, below a certain lower, critical capillary or Bond number, the trapping is dominated by capillary forces, and the residual nonwetting phase saturation reaches a constant maximum value.

Between these two extremes, the residual nonwetting phase saturation depends upon the combined effects of capillary, viscous pressure, and gravity forces.
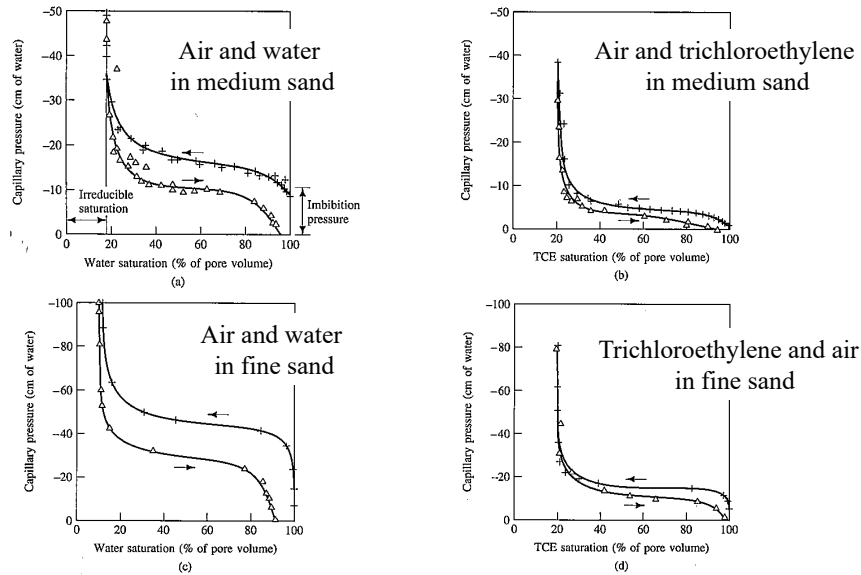
The mapping of these three domains in the plane C_a - B_o, constitutes the "phase-diagram" for a given system. The aim of this work was to establish experimental phase-diagrams for several uniform porous media systems, under vertical displacement conditions

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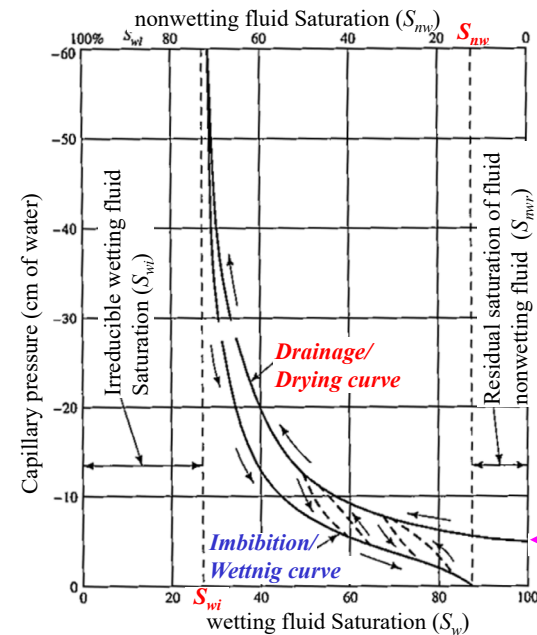
Capillary – pressure-wetting fluid saturation curves for two phase flow

"Contaminant Hydrology" Fetter (1999)

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Experimentally derived capillary –pressure-wetting fluid saturation curves

Air and water in medium sand (a)

Air and trichloroethylene in medium sand (b)

Air and water in fine sand (c)

Trichloroethylene and air in fine sand (d)

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Changes in the vertical distribution of oil（LNAPL）with time after a slug of oil is added to the top of a column of sand
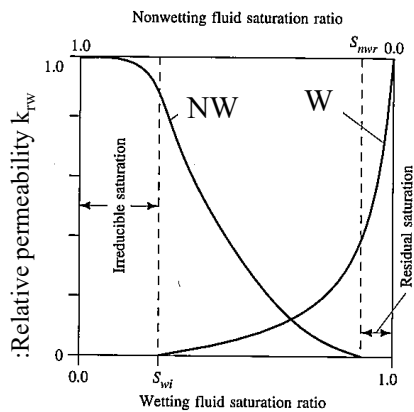
Contaminant Hydrology Fetter (1999)

Capillary fringe
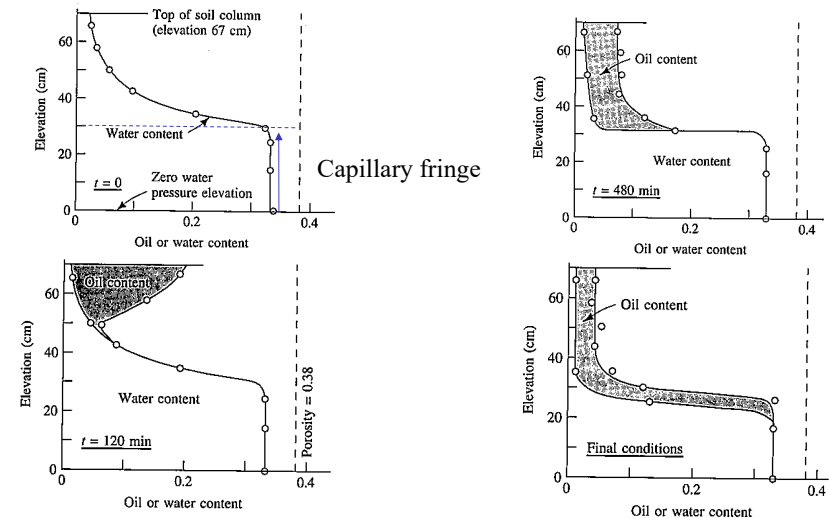
Relative permeability of two phase system

Darcy's law for the steady state flow of saturated water in the presence of NAPL

$$Q_w = -\frac{k_{rw}k_i\rho_w}{\mu_w}A\frac{dh_w}{dl}$$

Q_w: volume of water flowing
k_{rw}: relative permeability of water in the presence of the nonwetting fluid
k_i: intrinsic permeability
ρ_w: density of the water
μ_w: dynamic viscosity of the water
A: cross-sectional area of the flow
d_{hw}/dl: gradient of the head of the water

For nonwetting fluid

$$Q_w = \frac{k_{rnw}k_i\rho_{nw}}{\mu_{nw}}A\frac{dh_{nw}}{dl}$$

Sorption of contaminant

Sorption causes contaminants to move slower than the flowing ground water (interstitial velocity), resulting **retardation**.

Effects of sorption should be considered both in

- evaluating the potential for movement of the contaminants in soils (aquifer, clay liner).

- designing remediation activities at hazardous-waste site.

Concept of sorption

Sorption can be defined as the interaction of contaminant with a solid. The process of sorption can be divided in **adsorption** and **absorption**.

Adsorption: an excess contaminant concentration at the surface of a （吸着）　　solid.

Absorption: penetration into the solid by a contaminant （吸収）

Sorbate and **sorbent** are the terms used in sorption behavior. Sorbate is the contaminant that adheres to the sorbent, that is, sorbing material.

sorbate: organic molecules; ions, sorbent: soils; aquifer matrix

Partitioning（分配）refers to the process by which a contaminant, originally in solution, distributes itself between the solution and the solid phase.

Concept of sorption, cont.

Sorption is determined experimentally by measuring the partitioning of a contaminant onto a particular sediment, soil or rock.

In **batch tests**, a number of solutions with various concentration are well mixed with the solid, and then the mount of contaminant removed (sorbed) is determined by measuring the concentration of the solution after mixing.

The results of batch tests are plotted on a graph, known as an **isotherm**, which shows the relationship between the concentration of contaminant of the solution and the amount sorbed on the solid.

Concept of sorption, cont.

If the sorption process is rapid compared with the flow velocity, the dissolved contaminant will reach an equilibrium condition with the sorbed phase and process can be described using **equilibrium isotherm**.

If the sorption process is slow compared with the rate of fluid flow in the porous media, a **kinetic** model is needed.

Batch test and isotherm

- sorbent
- sorbate

mixing 1 or 2 days measuring equilibrium concentration in the solution
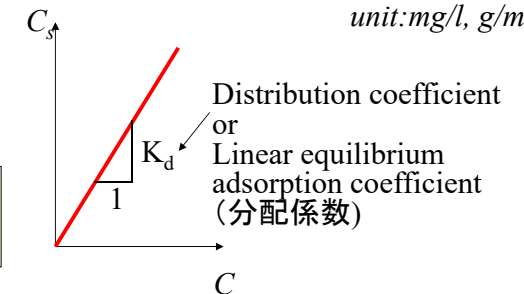
Liner equilibrium isotherm

$$C_s = K_d C \quad (4) \qquad R_d = 1 + \frac{(1-n)\rho_s K_d}{n}$$

C_s = mass of contaminant / unit mass of soil grain

unit: g/kg

C = mass of contaminant / unit volume of pore fluid

unit: mg/l, g/m³

Distribution coefficient or Linear equilibrium adsorption coefficient （分配係数）

Limitation of liner equilibrium isotherm

overestimation

Liner

C_s

C
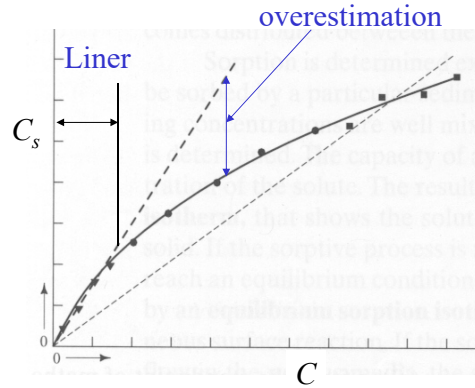
- upper limit of C_s
- nonlinear relation in wide range of C.

Freundlich sorption isotherm
- nonlinear -

$$C_s = KC^N \quad (5)$$

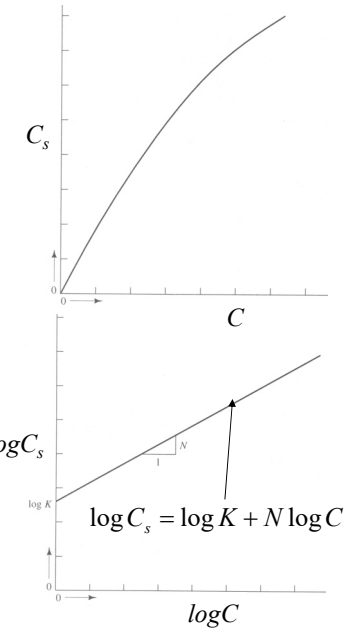K, N: constants

(5) => (14) and neglecting reactions ▶

$$\frac{\partial C}{\partial t} = D_{hl}\frac{\partial^2 C}{\partial x^2} - v_{int}\frac{\partial C}{\partial x} - \frac{(1-n)\rho_s}{n}\frac{\partial(KC^N)}{\partial t} \quad (6)$$

$$\frac{\partial C}{\partial t}\left(1 + \frac{1-n}{n}\rho_s KNC^{N-1}\right) = D_{hl}\frac{\partial^2 C}{\partial x^2} - v_{int}\frac{\partial C}{\partial x} \quad (7)$$

$$R_{df} = 1 + \frac{1-n}{n}\rho_s KNC^{N-1} \quad (8)$$

C_s

C

$logC_s$

$\log C_s = \log K + N\log C$

$\log K$

$logC$

Langmuir sorption isotherm
- nonliner -
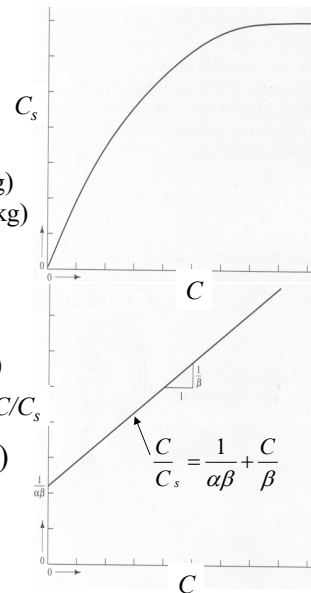
$$C_s = \frac{\alpha\beta C}{1+\alpha C} \quad (9)$$

α: adsorption constant related to the binding energy (L/mg)
β: the maximum amount of solute adsorbed by solid (mg/kg)

(9) => (5.14) and neglecting reactions

$$\frac{\partial C}{\partial t} = D_{hl}\frac{\partial^2 C}{\partial x^2} - v_{int}\frac{\partial C}{\partial x} - \frac{(1-n)\rho_s}{n}\frac{\partial(\frac{\alpha\beta C}{1+\alpha C})}{\partial t} \quad (10)$$

$$\frac{\partial C}{\partial t}\left(1 + \frac{1-n}{n}\rho_s\left(\frac{\alpha\beta}{(1+\alpha C)^2}\right)\right) = D_{hl}\frac{\partial^2 C}{\partial x^2} - v_{int}\frac{\partial C}{\partial x} \quad (11)$$

$$R_{dl} = 1 + \frac{1-n}{n}\rho_s\left(\frac{\alpha\beta}{(1+\alpha C)^2}\right) \quad (12)$$

C_s

C

C/C_s

$$\frac{C}{C_s} = \frac{1}{\alpha\beta} + \frac{C}{\beta}$$

C

Effect of sorption
- results of numerical calculation on column test-

Distance (cm)	Description	BIO1D
8.00	Case 1, none	Version 1.2
	Case 2, linear	GeoTrans, Inc.

No retardation versus linear retardation

no retardation

$$C_s = K_d C \quad (4)$$

retardation
with K_d=0.476µg/g
lower peak value

v_{int}=0.1cm/s
D_{hl}=0.1cm²/s
n=0.37
C_i=0.0mg/L

16cm   8cm
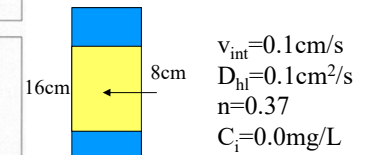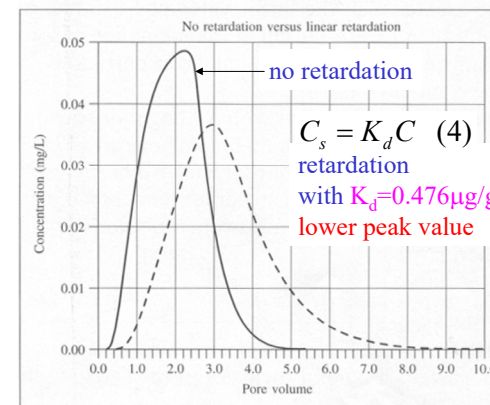
C=0.05mg/L is injected into the top of the column for 2mins as pulse injection. After 2 min injection, C is set back to 0.0mg/L.

"Contaminant Hydrology" Fetter, 1999

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Effect of sorption
- results of numerical calculation on column test-

Freundlich sorption isotherm
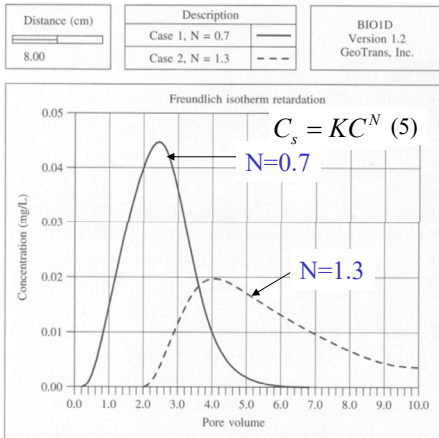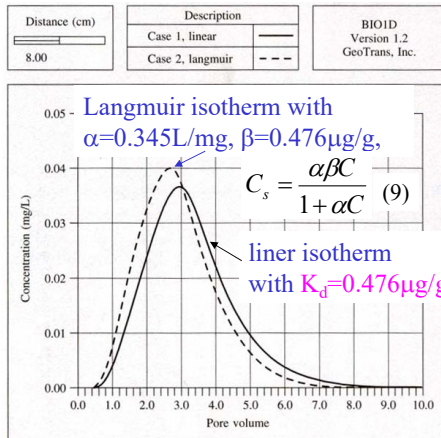
Distance (cm)	Description	BIO1D
8.00	Case 1, N = 0.7	Version 1.2
	Case 2, N = 1.3 ---	GeoTrans, Inc.

$C_s = KC^N$ (5)

N=0.7

N=1.3

Freundlich isotherm retardation (chart, Concentration (mg/L) vs Pore volume)

Langmuir sorption isotherm

Distance (cm)	Description	BIO1D
8.00	Case 1, linear	Version 1.2
	Case 2, langmuir ---	GeoTrans, Inc.

Langmuir isotherm with $\alpha=0.345$L/mg, $\beta=0.476\mu$g/g,

$C_s = \dfrac{\alpha\beta C}{1+\alpha C}$ (9)

liner isotherm with K_d=0.476μg/g

"Contaminant Hydrology" Fetter, 1999

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Isotherm for Contamination: *Batch tests*
Clay sand mixtures

$C_{solid} = \dfrac{C_i - C_{eql} \cdot V \,(mg/g)}{m_s}$

CC=100%
CC= 70%
CC= 50%
CC= 30%
CC= 10%   CC= 0%

C_{solid} increase with CC

The slope difference

C_{eql} (mg/L)

Volume of solution V (L)
Initial concentration C_i (mg/L)

Soil m_s (g)

Mixing

Concentration in liquid phase C_{eql} (mg/L)

Contaminant sorbed on soil C_{solid} (mg/g)
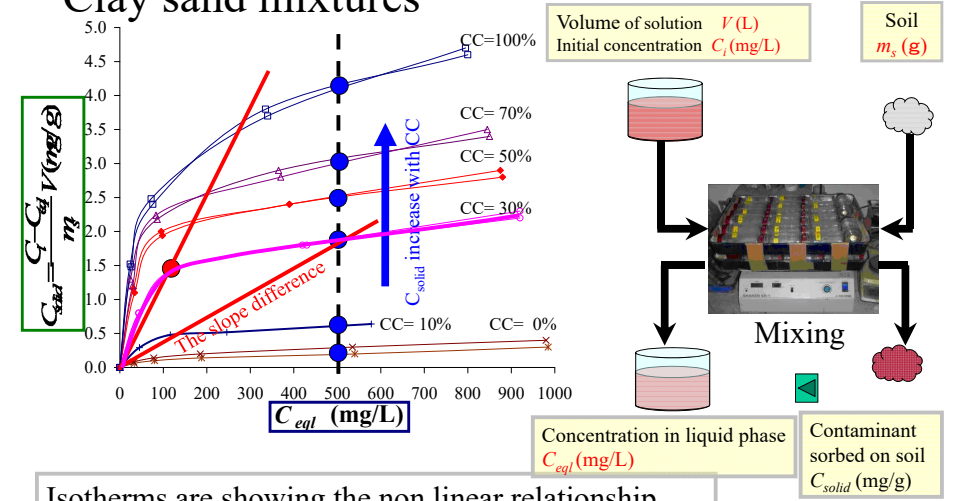
Isotherms are showing the non linear relationship
CC and C_{solid} are affected by C_{eql}

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Column test   Vertical stress

Clay –sand mixture

60mm
20mm

Hydraulic gradient
$i \sim 100$

Accelerometer — Loading yoke
Loading yoke rod
Displacement measurement board
Bearings
Fixing screws
Top chamber
O-Rings
Top lid
Air release/cleaning valves
Top porous metal
Bottom porous metal
Specimen ring
specimen
Inlet
C_0
C
Outlet
Base plate Outlet
Pressure sensors

Slide 36
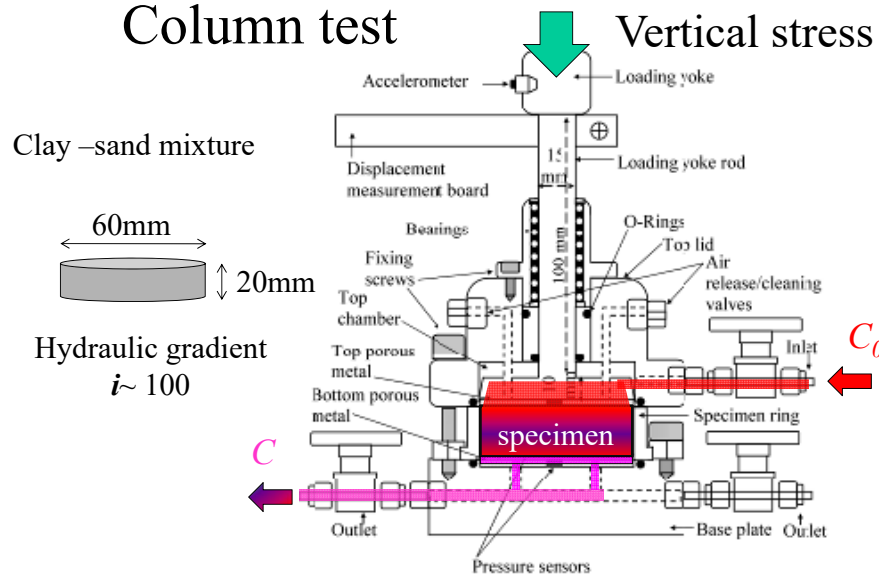
Koide's experimental results

CC= 10%
C_0=500mg/L
CC=100%
CC= 70%
CC= 50%
CC= 30%
C_0=50mg/L
CC= 30%

C/C_0

Nearly 6 times difference

10C1DNA2	10C2DAL2
30C4DAL2	30C5DNA2
30C6DNA2	30C7DAEL2
30C8DAEL2	30CL1DNA2
30CL2DAL2	50C1CO2
70C1CO2	100C1CO2

5 20 30   120   Pore Volume (PV) tv_{int}/H

One PV~0.3day
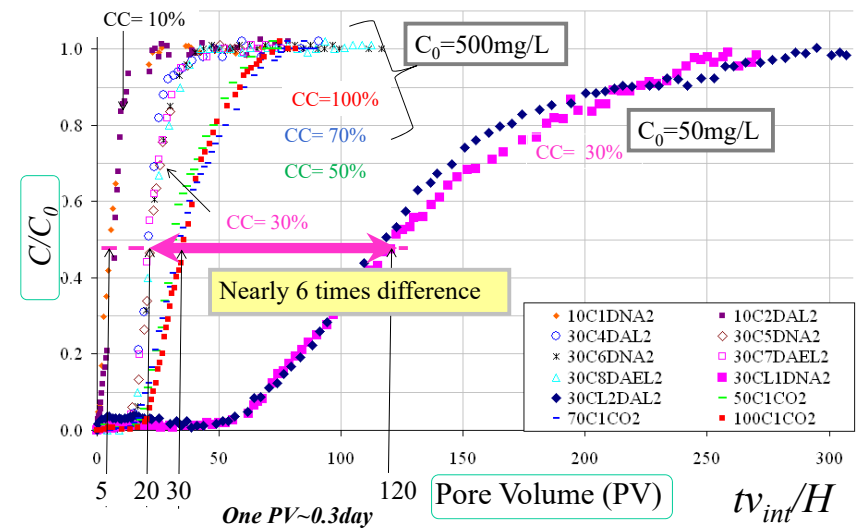
Examples of nonequilibrium (Kinetic) sorption model

Irreversible first order kinetic sorption model: rate of the sorption is a function of the solute remaining in solution and once it sorbed onto the solid, the solute cannot be desorbed.

$$\frac{\partial C_s}{\partial t} = k_1 C \qquad (13)$$

$$\frac{\partial C}{\partial t} = D_{hl}\frac{\partial^2 C}{\partial x^2} - v_{int}\frac{\partial C}{\partial x} - \frac{(1-n)\rho_s}{n}\frac{\partial C_s}{\partial t} \qquad (14)$$

k_1: a first order decay rate constant

Reversible liner kinetic sorption model: rate of the sorption is related to the amount that has already been sorbed and the reaction is reversible.

$$\frac{\partial C_s}{\partial t} = k_2 C - k_3 C_s \qquad (15)$$

k_2: forward rate constant, k_3: backward rate constant

Factors influencing sorption
- contaminant characteristics -

(1)water solubility

   The higher the solubility of chemical is, the easier or faster the chemical distributes by the hydraulic cycle, showing lower adsorption coefficient.

(2)polar-ionic character: three classes

  1) ionic or charged species: ex) pesticide and phenol

  2) uncharged polar species: benzene, toluene, xylene

  3) uncharged and nonpolar species: TCE,PCE, chlorinated benzene
    The higher polarity, the lower adsorption.

(3) octanol-water partition coefficient
（オクタノール/水分配係数）

Octanol-water partition coefficient: K_{ow}

オクタノール／水分配係数

   The octanol-water partition coefficient represents the distribution of a chemical between octanol($C_8H_{17}OH$) and water in contact with each other at equilibrium conditions. Key parameter in the studies of the environmental fate of organic chemicals

$$K_{ow} = \frac{\text{concentration in octanol phase}}{\text{concentration in aqueous phase}}$$

Range of K_{ow} : **$10^{-3} - 10^7$** => it is often given by **log K_{ow}**

-smaller molecular, higher polarity => lower K_{ow}

-larger molecular, lower polarity => more hydrophobic (low solubility)
（疎水性）

  => higher K_{ow} => larger partition on solid phase

Factors influencing sorption
- soil characteristics -

  •Mineralogy

  •Permeability – porosity

  •Texture

  •Homogeneity

  •**Organic carbon content**:
        particularly important NAPLs
（活性炭吸着）
  •Surface charge（表面電荷）   *ex: activate carbon sorption*
                                              to exclude NAPLs
  •Surface area(粒子表面積)：

clay and silt having higher specific surface associate with natural organic matter, creating a rich sorptive environment.
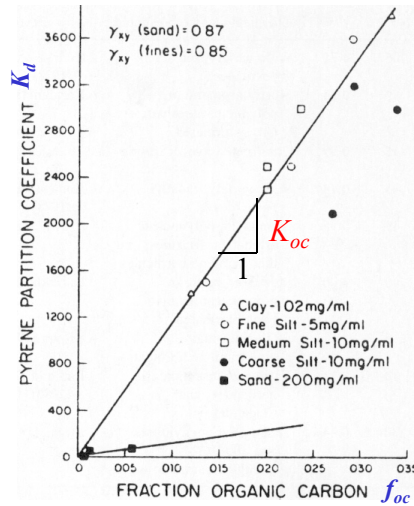
Sorption of hydrophobic compounds (NAPLs)

(Karichhoff et al. 1979)

The partitioning of a solute onto mineral surface or organic carbon contents of soil is almost exclusively on to the organic carbon fraction, f_{oc} if it constitutes at least 1% and strong correlation between K_d and organic carbon content of the sediment.

sorption constant

$$K_{oc} = \frac{K_d}{f_{oc}} \quad (16)$$

"Ground water contamination" by Bedient, Rifai & Newell

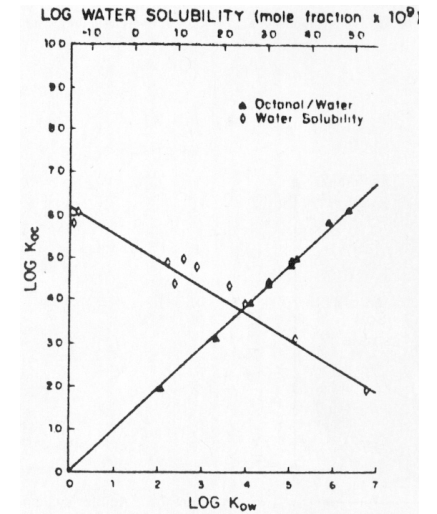## Sorption of hydrophobic compounds (NAPLs)

(Karichhoff et al. 1979)

strong correlation between K_{oc} and K_{ow},

$$K_d \approx 0.6 K_{ow} \quad (17)$$

especially for $\log K_{oc}$ and $\log K_{ow}$
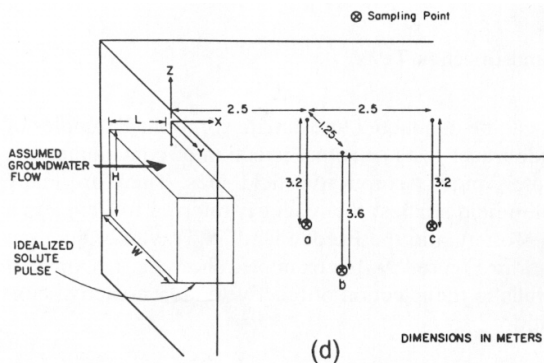
$$\log K_d = \log K_{ow} - 0.21$$

"Ground water contamination" by Bedient, Rifai & Newell

Natural gradient field test at the Borden landfill site

ave.v_{int}= 0.091m/d
two conservative tracers (Chloride, Bromide)
five VOCs (Bromoform CTET, PCE, DCB, HCE) were injected for one day and intensive monitoring was conducted for two years.
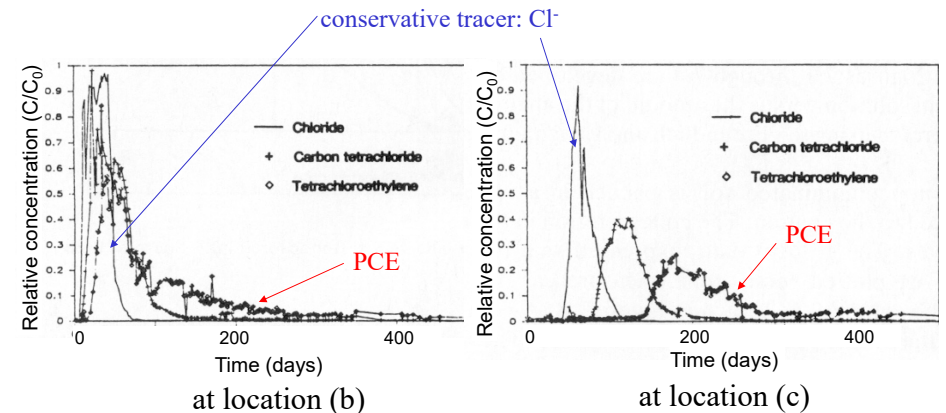
"Ground water contamination" by Bedient, Rifai & Newell

Field retardation data from the Borden landfill site

at location (b)   at location (c)

"Ground water contamination" by Bedient, Rifai & Newell

Slide 45

Determination of retardation factor from the field data

$$R_d = \frac{\text{distance from source of nonreactive plume } (x_n)}{\text{distance from source of reactive plume } (x_r)}$$
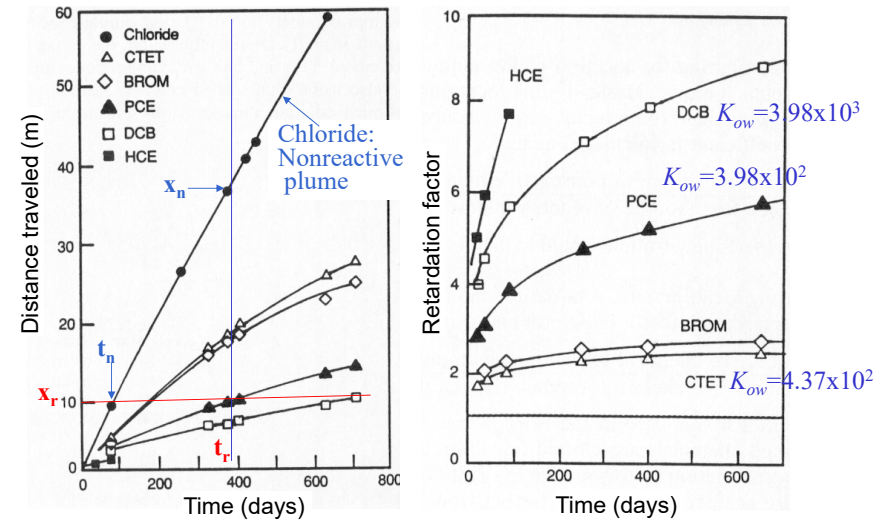
$$R_d = \frac{\text{time for reactive chemical to reach a given point } (t_r)}{\text{time for nonreactive chemical to reach a given point} (t_n)}$$

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Determination of retardation factor from the field data

Left graph: Distance traveled (m) vs Time (days). Legend: Chloride, CTET, BROM, PCE, DCB, HCE. Chloride: Nonreactive plume, labeled x_n, t_n, x_r, t_r.

Right graph: Retardation factor vs Time (days). HCE, DCB $K_{ow}=3.98\times10^3$, $K_{ow}=3.98\times10^2$, PCE, BROM, CTET $K_{ow}=4.37\times10^2$.

"Ground water contamination" by Bedient, Rifai & Newell

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Example of exposure pathway

GAS — Prevailing wind direction, Transport medium (air), Exposure point, Inhalation exposure route, Ingestion exposure route, Release source (volatilization), Release source (site leaching), liquid, Solute, Groundwater exposure pathway, Air exposure pathway, Transport medium (groundwater), Water table, Groundwater flow

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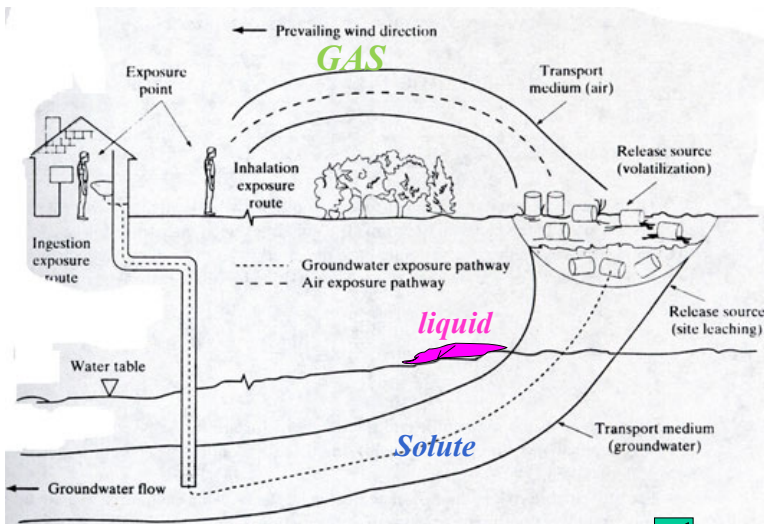
home work 5
due July 12

Investigate the environmental standards of HMs, VOCs , DIOXINs and others (Ground W, Surface W, soils, water supply, effluent) in your country
Compare the standards and discuss the similarity and differences

Contaminant transport equation cont.

total mass(adsorbed + fluid phase)

general mass balance eq.

$$\frac{\partial m}{\partial t} = -\nabla J \pm R \pm \lambda m \qquad (13)$$

$\partial/\partial x + \partial/\partial y + \partial/\partial z$

chemical reaction

a general rate constant in radio active and/or biological decay

For 1-D flow

$$\frac{\partial(nC)}{\partial t} + \frac{\partial((1-n)\rho_s C_s)}{\partial t} = -\frac{\partial}{\partial x}\left(nv_{int}C - D_{hl}n\frac{\partial C}{\partial x}\right) \pm R \pm \lambda\left(nC + (1-n)\rho_s C_s\right)$$

$$(14)$$

Adsorption（on solid phase)

In fluid phase

Working Groups

Group	student ID	last name	first name	Country	Field of research
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1	17M58316	FENG	Yi	China	Structure
1	17M58322	GUO	Yuliang	China	Geotech
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1	18M51205	Kawakami	Jyun	Japan	Geotech
1	18M51429	GUNAWAN	DEVIN	Indonesia	Concrete Structure
1	18R51503	DURATORRE	Tommaso	Italy	Hydrology
1	18R57014	IDHAR	Ridha Amalia	Indonesia	Planning
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2	18M51257	Koizumi	Daiya	Japan	Planning
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3	15_12682	Fujimoto	Shouta	Japan	Geotech
3	17M58374	LYU	Hang	China	Geotech
3	18M51197	Kawai	Tomohiro	Japan	Planning
3	18M51263	Kobayashi	Kenta	Japan	Concrete Structure
3	18M53032	HONG-IN	Phadungsak	Thailand	Geotech
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4	18M51286	Sato	Michitaka	Japan	Water Env
4	18M51300	Shiroma	Hiroya	Japan	Planning
4	18M51352	Noda	Shouta	Japan	Geotech
4	18M51398	Muro	Shoutaro	Japan	Planning
4	18M51458	XIAO	TENGXIANG	China	Geotech
4	18R51005	Esponazs	Jasson	Mexico	
4	18R57013	NURSHAFIYANA	Safyra	Indonesia	Planning