Contaminant properties

Lecture 1

Learning outcomes

- Partitioning
- Dry/wet density
- Contaminant types and properties

Remediation

• What is remediation?
Process to remove contaminants or transform them into harmless compounds

•What is bioremediation?
Process relying on microbial activity to remove or transform contaminants

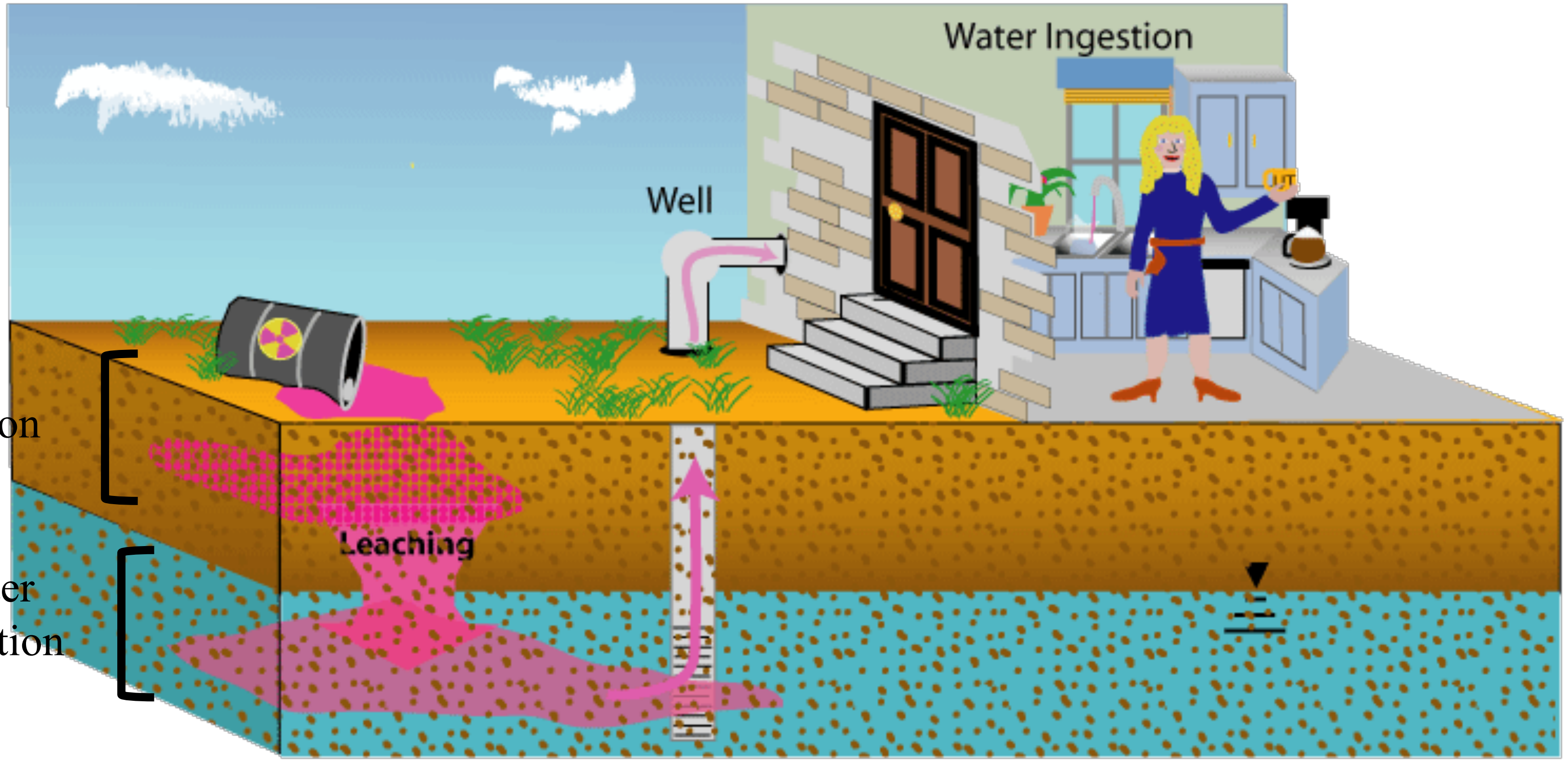
Major Concern:
Transport of contamination off site by ground and surface water flow

Challenges:
Unknowns in subsurface flow properties, contaminant behavior, response of microbial communities

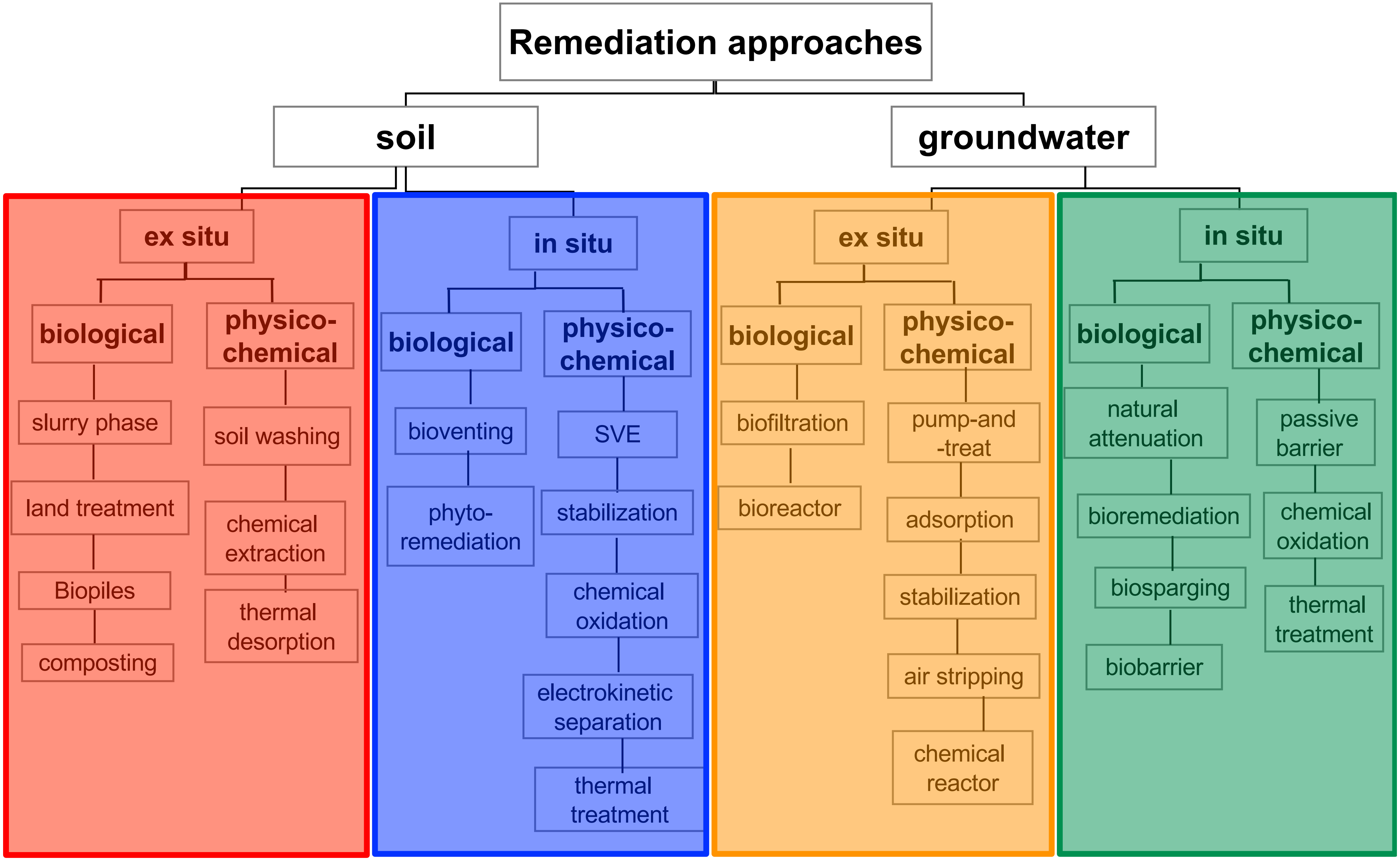
Types of contaminated environments

soil contamination

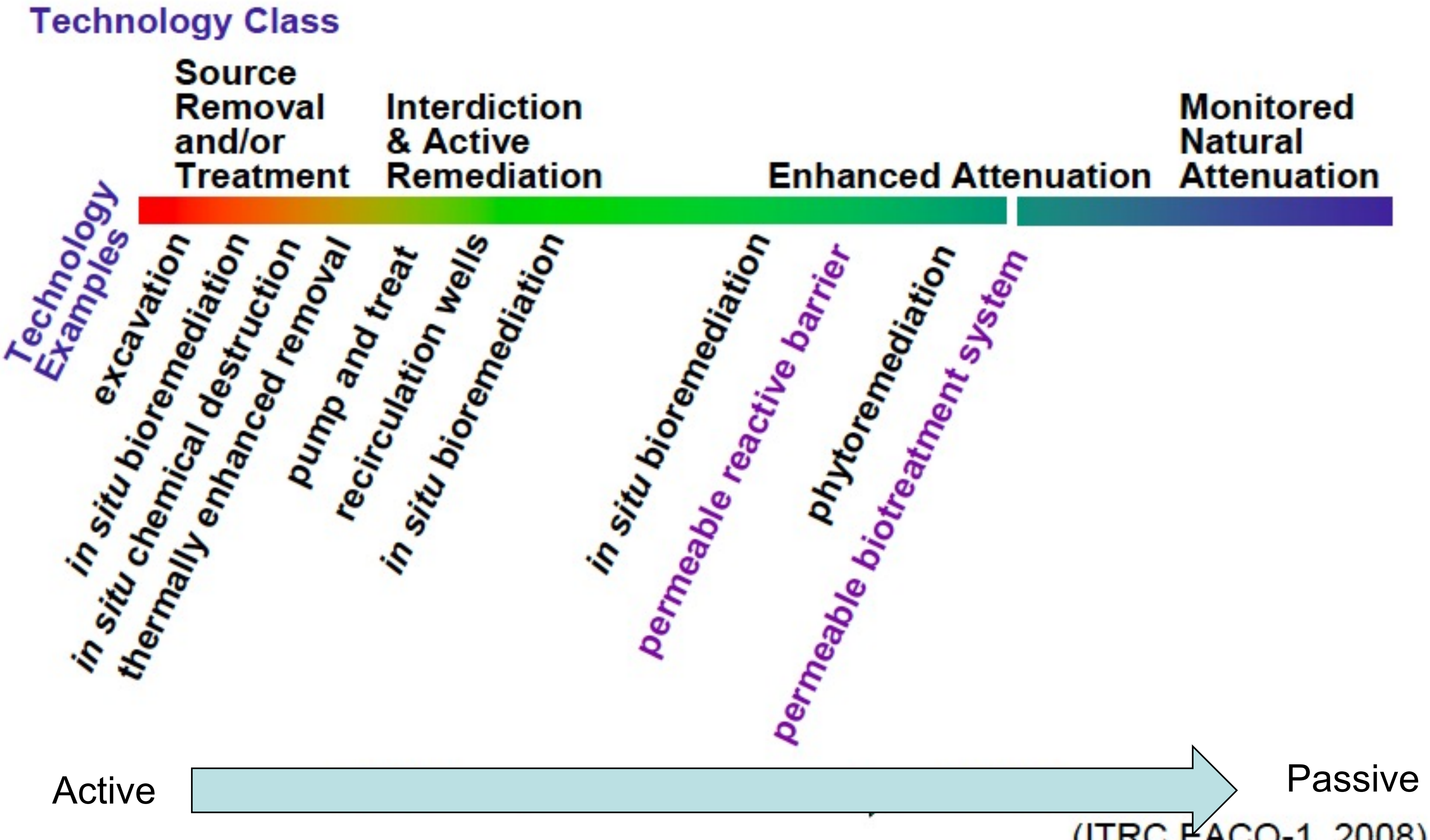
groundwater contamination



Remediation approaches



Continuum of Remediation Technologies: Active to Passive



(ITRC EACO-1, 2008)

Cost of soil remediation approaches (in US)

	Type	Cost	comment
In situ	Bioventing	79-970 US$/ m^3 soil	
	Enhanced biodegradation	30-100 US$/ m^3 soil	
	phytoremediation	147-2,322 US$/ m^3 soil	
	Chemical oxidation	No data	
	Electrokinetic separation	15-117 US$/ m^3 soil	No full-scale implementation
	Soil flushing	18-49 US$/ $yard^3$ soil	
	Soil vapor extraction	405-1,485 US$/$m^3$ soil	
	Solidification/stabilization		depends on mixing technology and amendment
	In situ vitrification	$375-425 per ton of soil	Few commercial applications
	In situ thermal treatment	29-62 US$ /$yard^3$ soil	
Ex situ	Biopiles	130-260 US$/ m^3 soil	
	Composting	481-578 US$ /$yard^3$ soil	
	Land-farming	100 US$/ m^3 soil	Add one-time laboratory and field test costs (125,000-150,000 US$)
	Slurry-phase biological treatment	100-160 US$/ m^3 soil	
	Soil washing	70-187 US$/ m^3 soil	
	Chemical extraction	358-1717 US$/ m^3 soil	
	Excavation and off-site disposal	300-510 US$/ m^3 soil	Off-site disposal can vary in cost

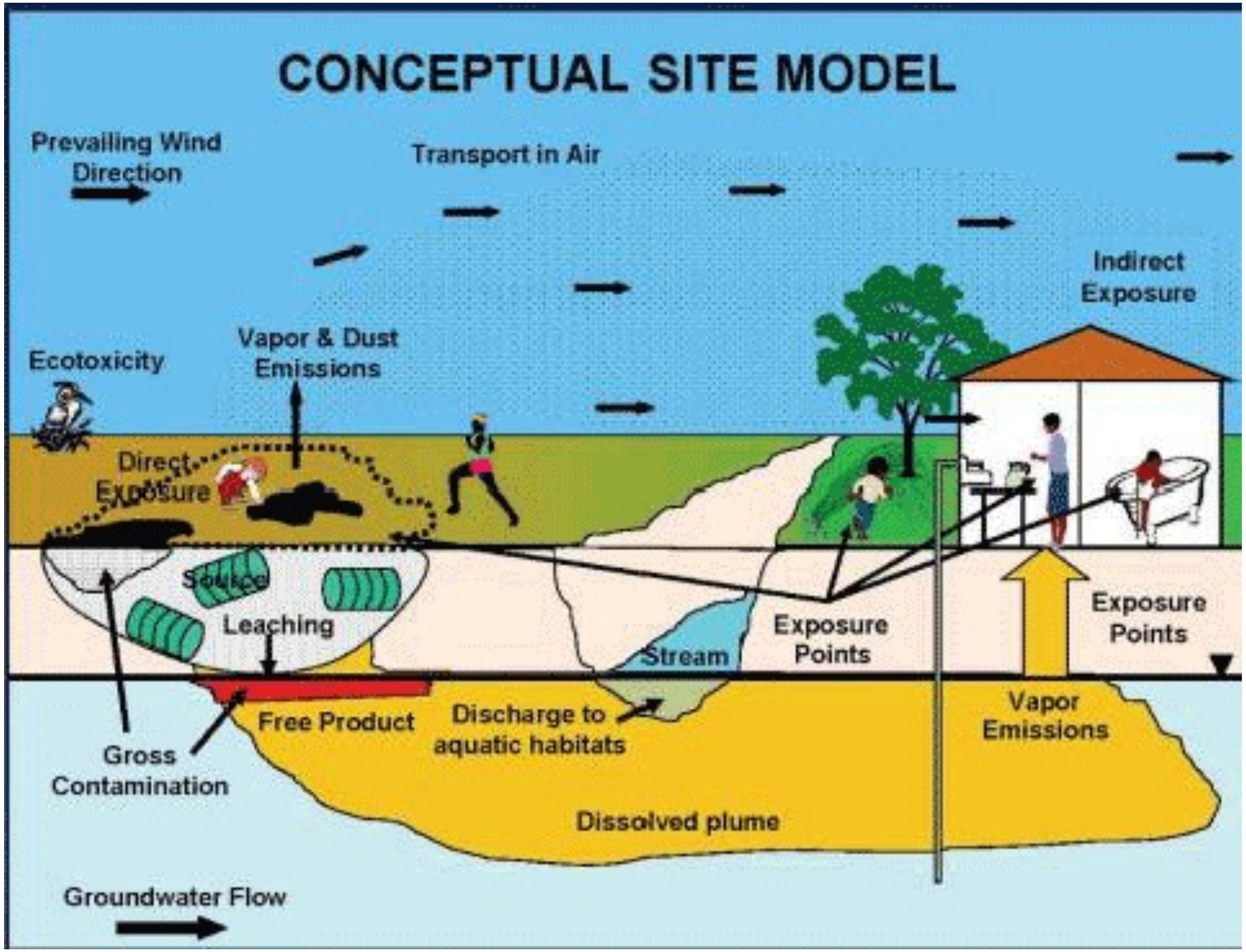
https://frtr.gov/matrix/default.cfm https://www.frtr.gov/matrix2/section1/toc.html

Cost of groundwater remediation approaches (in US)

	Type	Cost	comment
In situ	biosparging/nitrate enhancement	10-60 US$/ m^3 GW	
	biobarrier		
	Passive barrier	0.08-0.21 US$/cubic yard GW	
	Chemical oxidation	\$31-175 US$ /10,000 gal. GW	
	Thermal treatment	50-300 US$/cubic yard GW 963-1,961 US$/$m^3$ of barrier	
Ex situ	Biofiltration (gas phase)	5-10 US$/kg contaminant	
	Bioreactor	21-167 US$/1,000 gal. GW	
	Adsorption (gas phase)	\$1,000 for a 100-ft^3/min unit to \$40,000 for a 7,000-ft^3/min unit. Carbon costs 2 to 3 US$ per pound.	
	Adsorption (liquid phase)	0.32-1.70 US$ per 1,000 liters GW	
	Air stripping	4-21 US$/10,000 gal. GW	

https://frtr.gov/matrix/default.cfm https://www.frtr.gov/matrix2/section1/toc.html

Source control vs. plume remediation



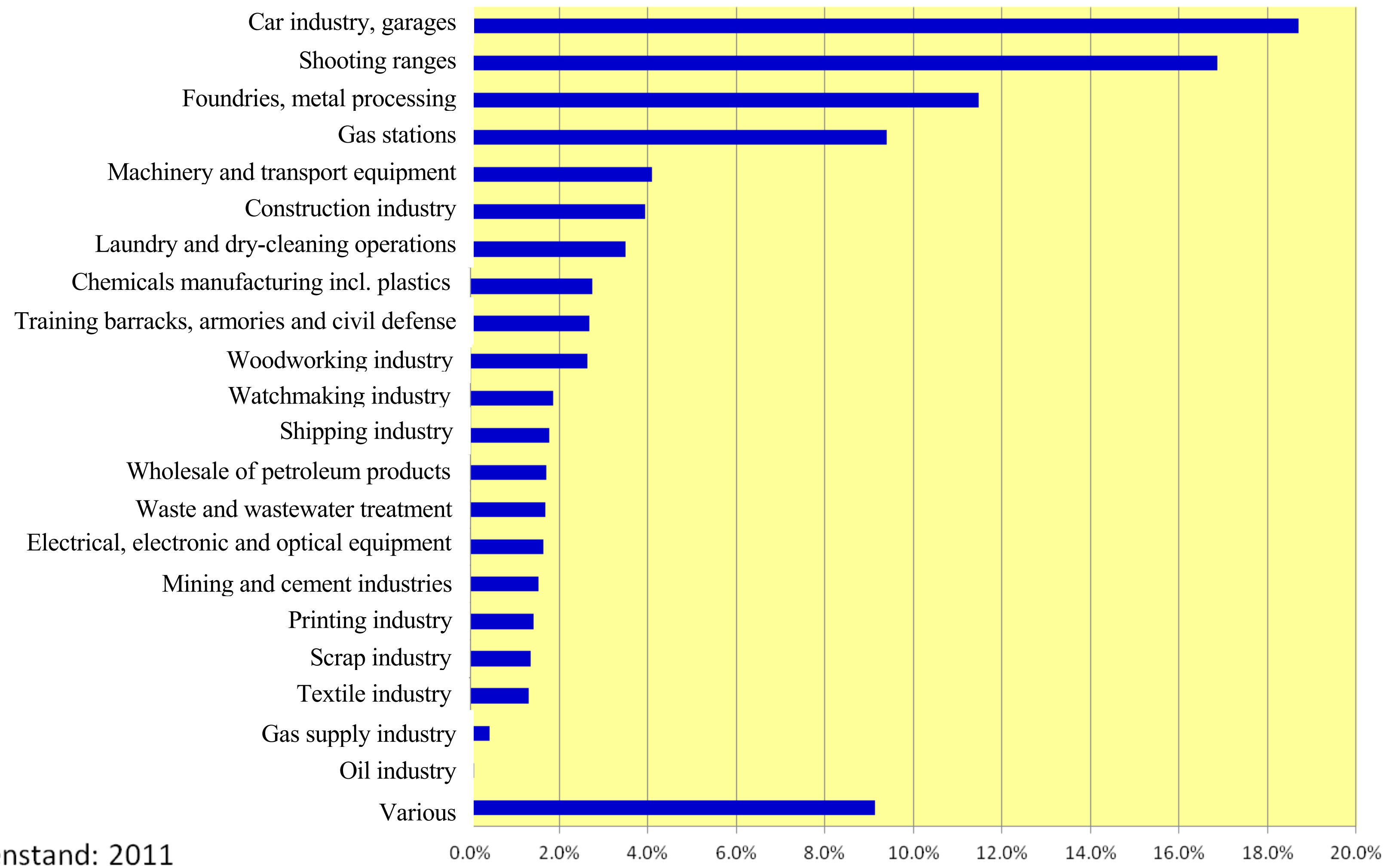
In Switzerland

• Of the approximately 700 soil remediation carried out to date- traditional methods are predominant
• Excavation of contaminated material and soil washing
• Securing of sites (barriers)
• *In situ* measures not fully accepted
• ‘The search for perfect solutions should be avoided’

In Switzerland

Contamination sources

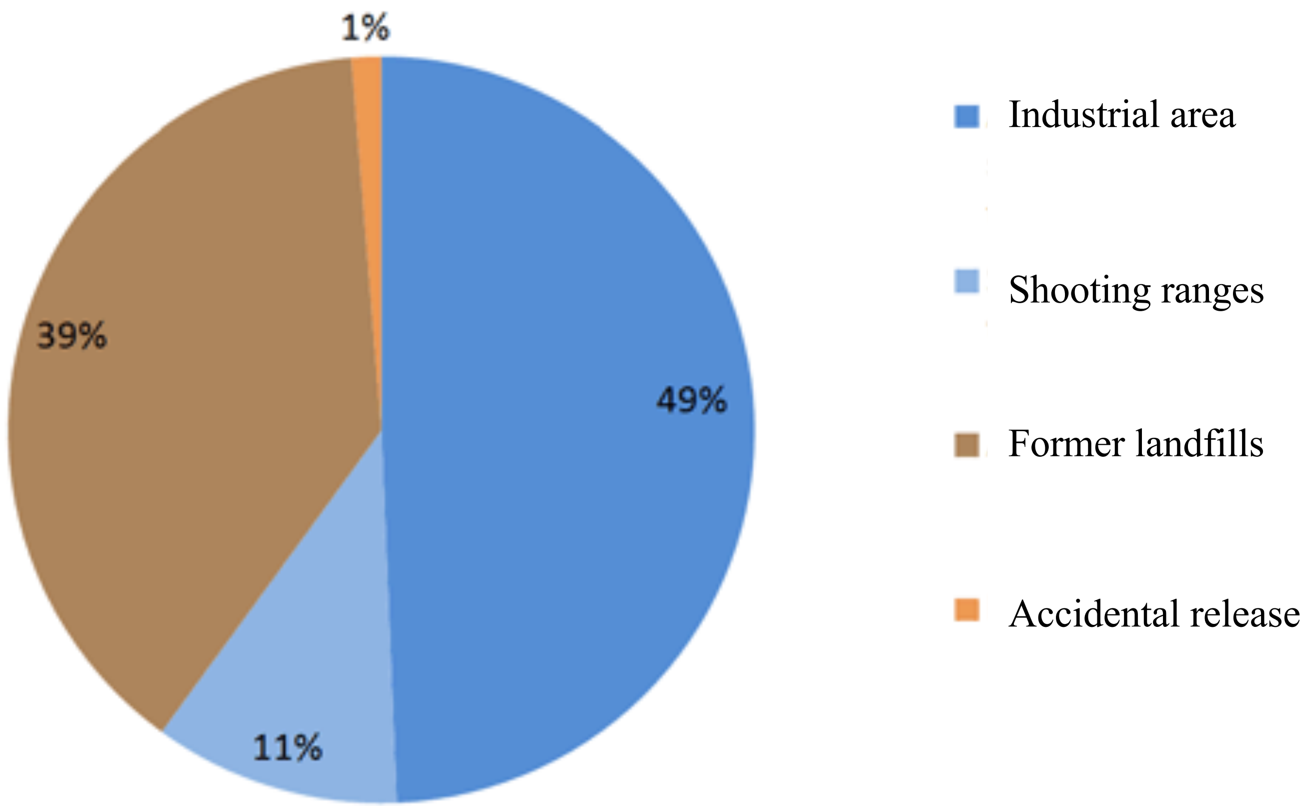
Operating business: Types



Datenstand: 2011

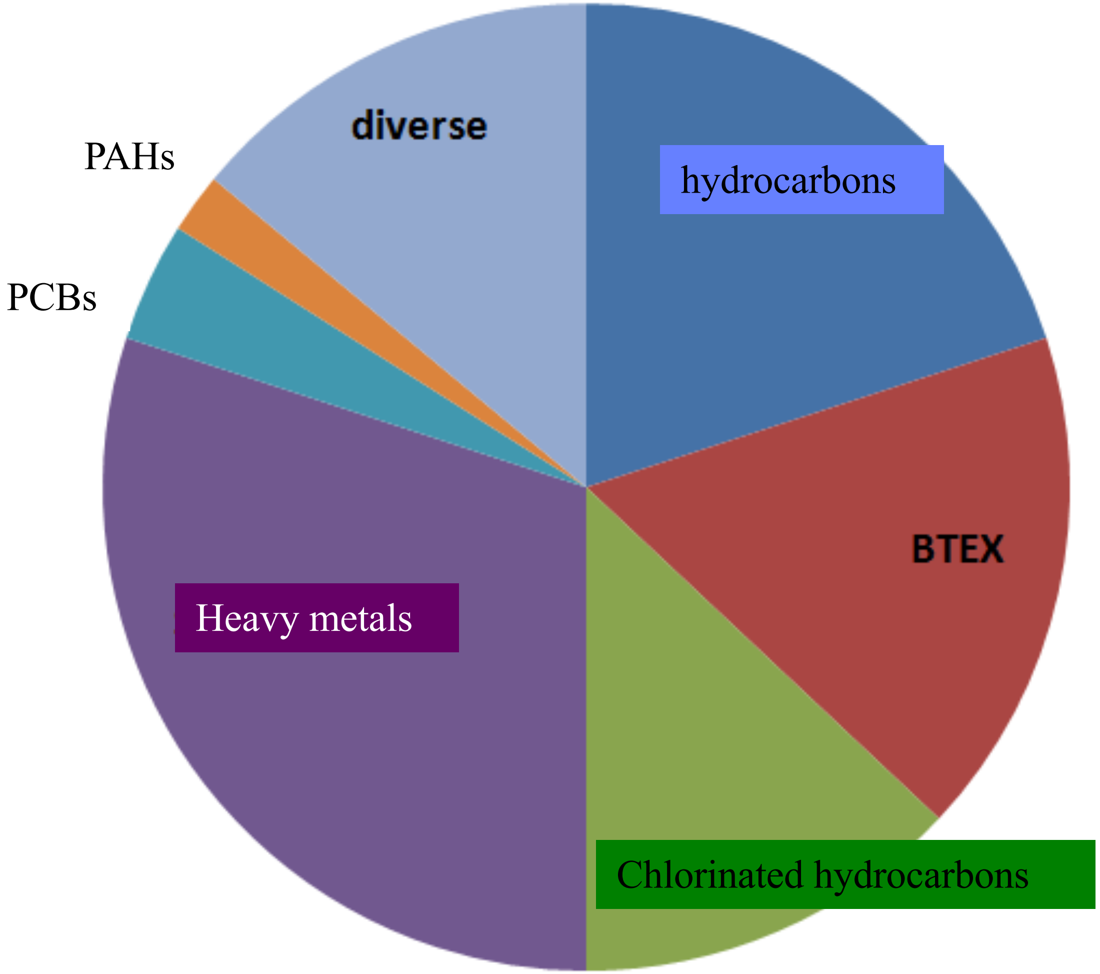
In Switzerland

Type of contamination area



In Switzerland

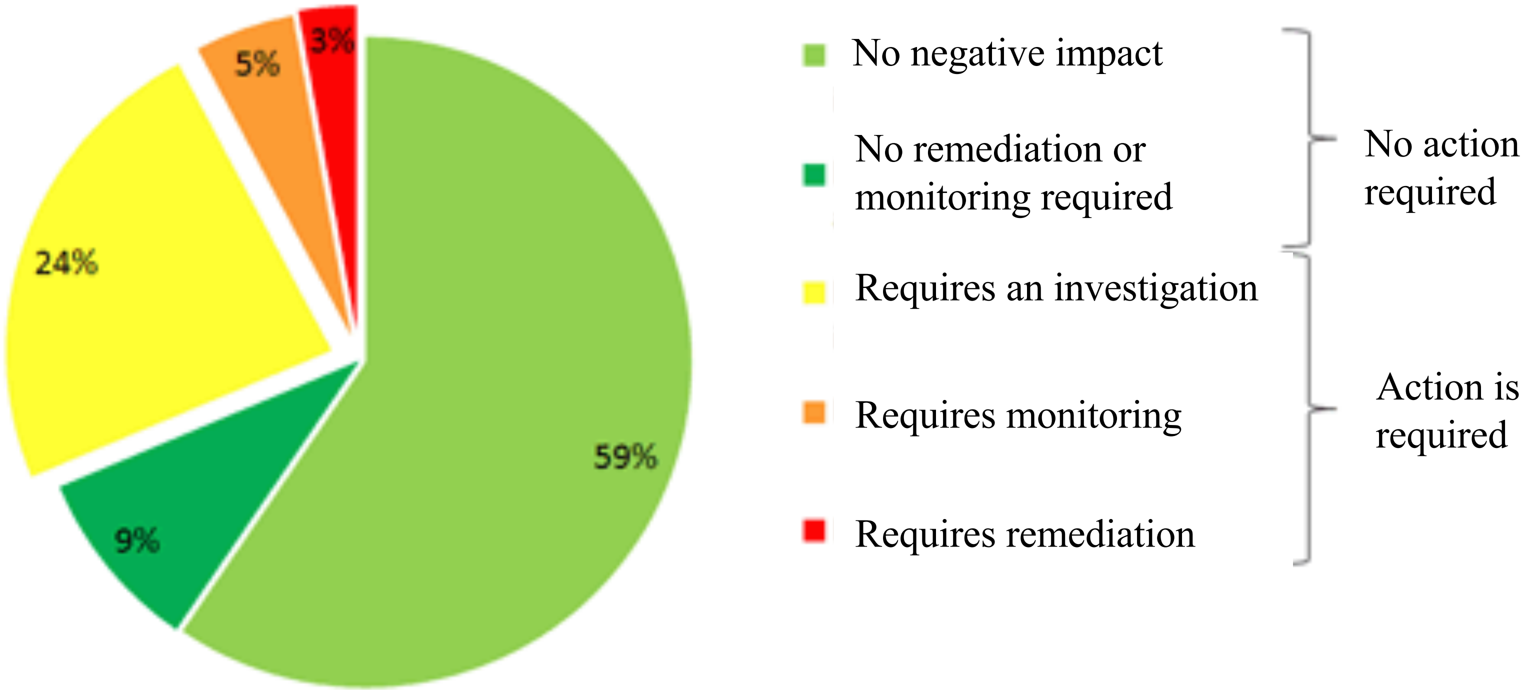
Contaminants



In Switzerland
Location of contaminated sites

In Switzerland

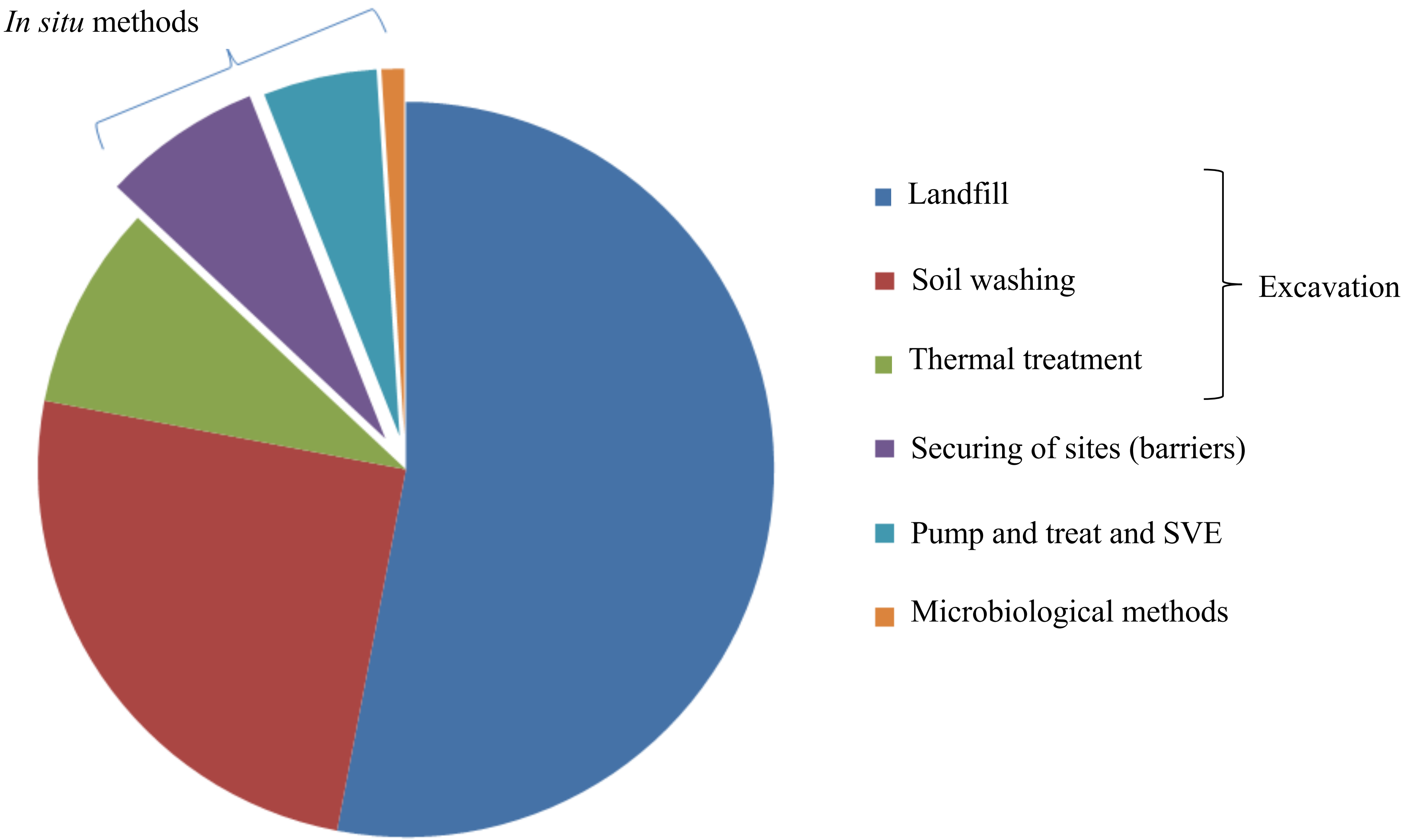
Sites to be remediated



- 38,000 known contaminated sites
- About 4,000 need to be remediated (700 already cleaned up)

In Switzerland

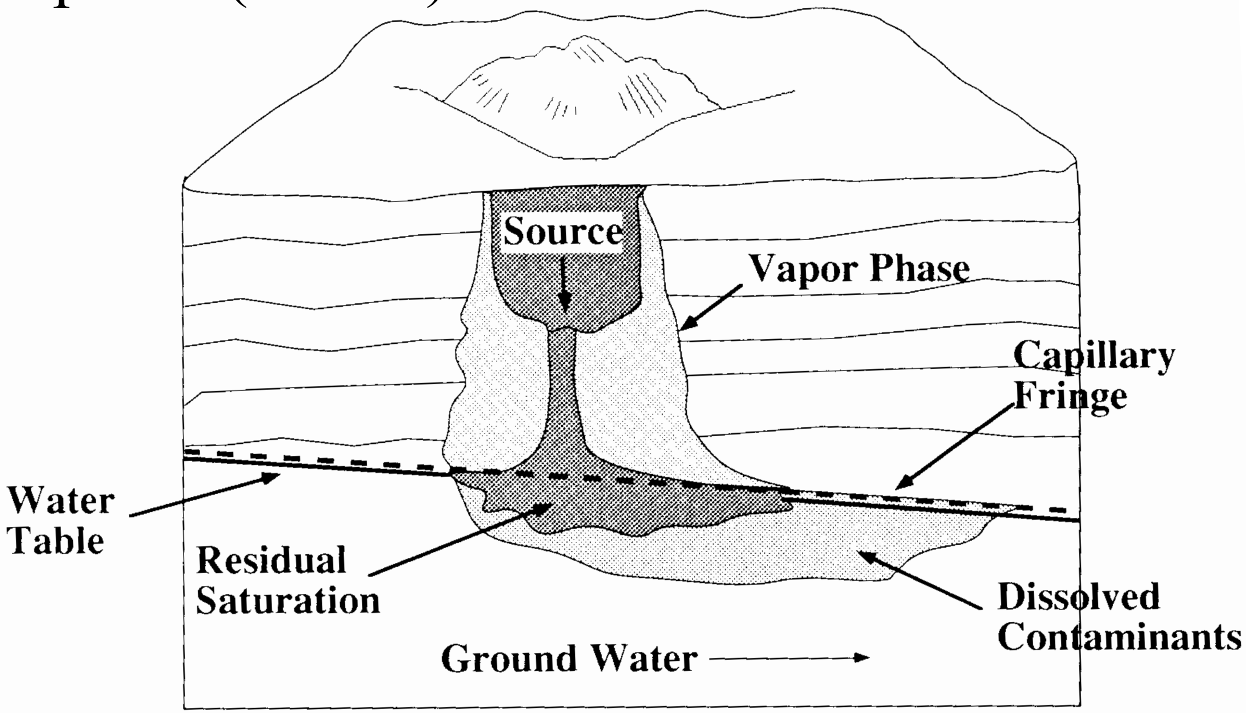
Remediation methods



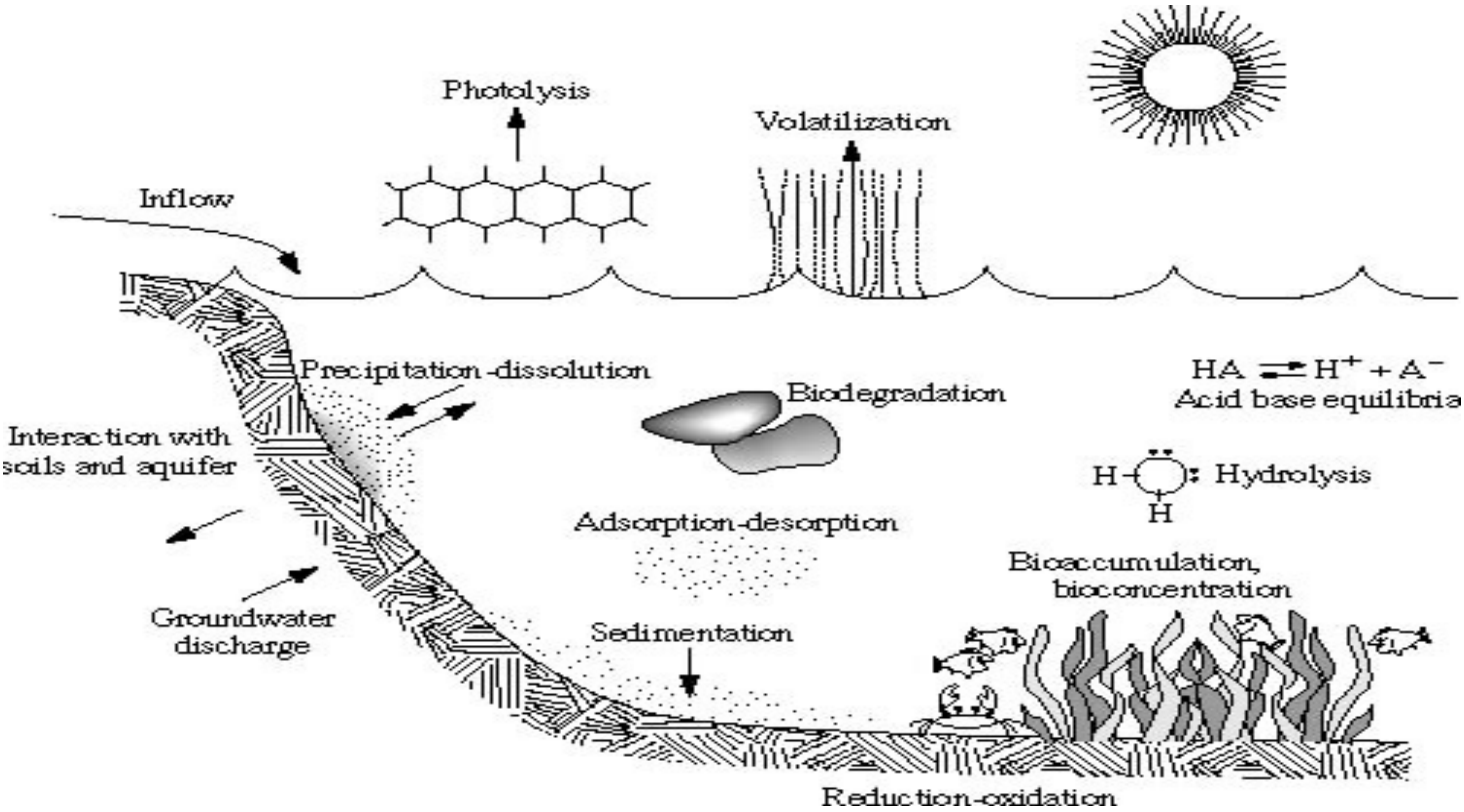
Contaminant properties

The contaminant is present in many different phases in the subsurface:

- vapor phase
- solid phase (sorbed)
- aqueous phase (dissolved)
- non-aqueous phase (NAPL)

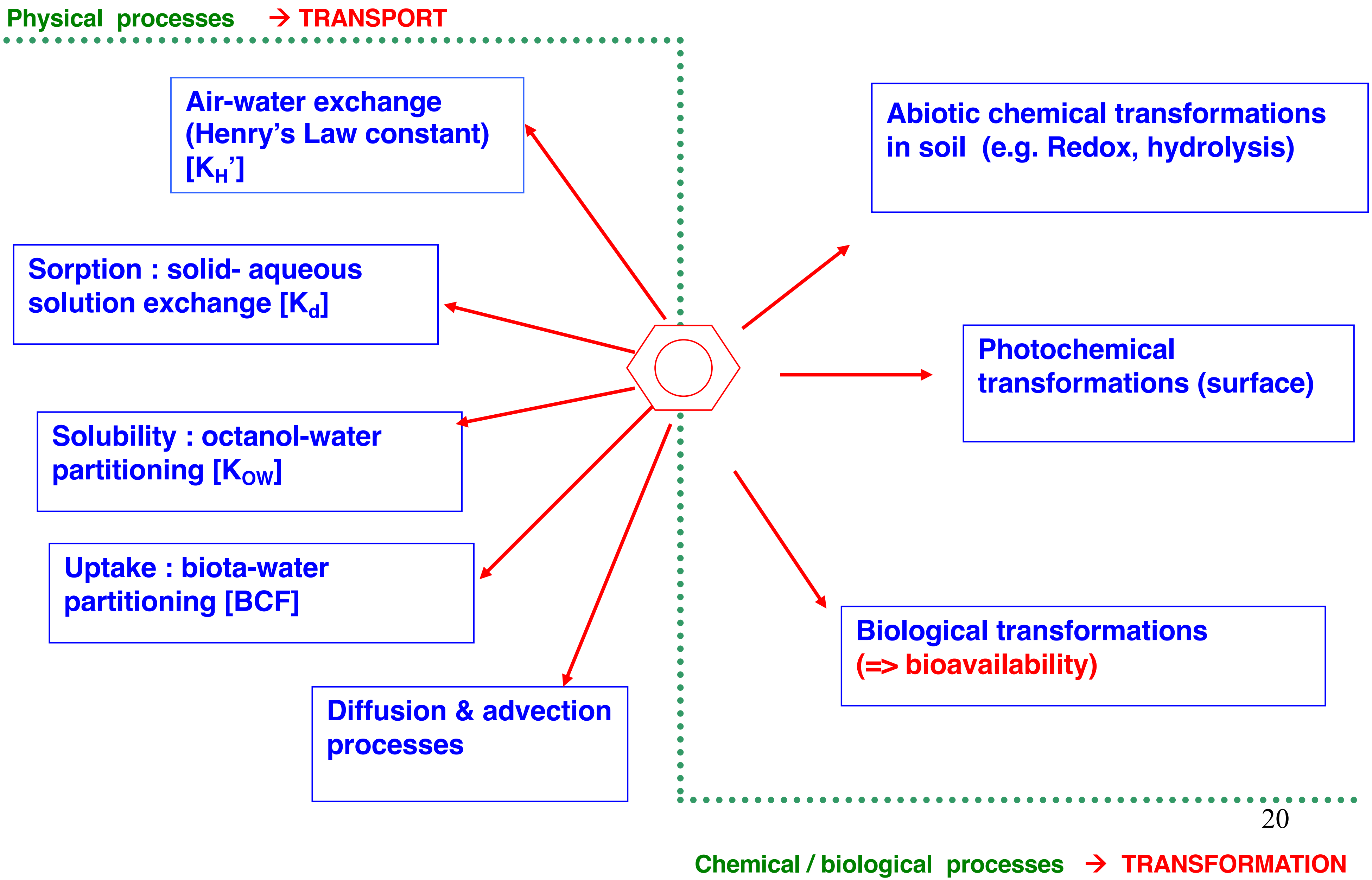


The challenge is to target all these phases

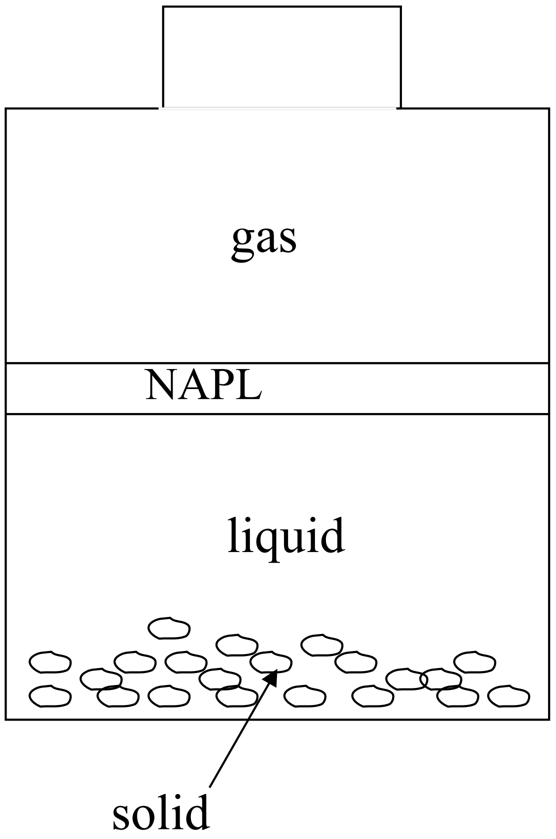


Contaminants can undergo many transformations

Environmental Fate of a Pollutant: Partitioning and transformations



Partitioning of a compound



V_L= volume of solution (L)
V_G= volume of gas (L)
M_S= mass of dry solid (g)
V_{NAPL}= volume of NAPL (L)

A compound (i) added to a closed bottle containing soil and water and a gas phase will partition into all three phases:

- $C_{i,\,aq}$ (mg/L water)
- $C_{i,\,g}$ (mg/L gas)
- $C_{i,\,s}$ (mg/g dry solid)
- $C_{i,\,NAPL}$ (mg/L NAPL)

- The concentration of i in aqueous, gaseous, sorbed and non-aqueous phase will depend on:
- K_H' = gas-water partitioning
- K_D= sorbed- water partitioning
- K_{NW}= NAPL-water partitioning

- The amount of i in each phase (including the non-aqueous phase) is:
- $m_{i,\,aq} = C_{i,\,aq} * V_L$
- $m_{i,\,g} = C_{i,\,g} * V_G$
- $m_{i,\,s} = C_{i,\,s} * M_s$
- $m_{i,\,NAPL} = C_{i,\,NAPL} * V_{NAPL}$

Partitioning relationships

Air-water= Henry's Law constant

$$K_H'\ [-] = \frac{\text{molar conc. in gas}}{\text{molar conc. in aq. sol' n}} = \frac{C_{i,g}\ \text{(mol in gas phase/L gas phase)}}{C_{i,aq}\ \text{(mol in solution /L liquid)}} = \frac{P_i^{sat}\ /\ RT\ \ \text{(mol/L)}}{C_{i,aq}\ \ \text{(mol/L)}}$$

R= 82.06* 10^{-3}
atm.L/mol.K; T in K

Soil-water= soil-water partitioning coefficient

$$K_D\ [m^3/g] = \frac{\text{conc. sorbed (g/g)}}{\text{conc. in aq. sol' n }(g/m^3)} = \frac{C_{i,s}\ \text{(g sorbed /g dry solid)}}{C_{i,aq}\ (\text{g in solution }/m^3\text{ liquid})}$$

NAPL-water= NAPL-water partitioning coefficient

$$K_{NW}\ [-] = \frac{C_{i,NAPL}\ \text{(mol in NAPL phase/L NAPL phase)}}{C_{i,aq}\ \text{(mol in solution /L liquid)}}$$

Note that all these partitioning coefficients include the aqueous phase

Henry's Law

Air-water partition= Henry's Law constant

$$K_H{}' \,[\text{-}] = \frac{\text{molar conc. in gas}}{\text{molar conc. in aq. sol'n}} = \frac{C_{i,g}\ (\text{mol in gas phase/L gas phase})}{C_{i,aq}\ (\text{mol in solution /L liquid})} = \frac{P_i^{sat} / RT\ \ (\text{mol/L})}{C_{i,aq}\ \ (\text{mol/L})}$$

R= 82.06* 10^{-3} atm.L/mol.K; T in K

$$K_H{}^v\ \frac{\text{L.atm}}{\text{mol}} = \frac{\text{partial pressure}}{\text{molar conc. in aq. sol'n}} = \frac{P\ (\text{atm})}{C_{i,aq}\ (\text{mol in solution /L liquid})} = \frac{P_i^{sat}\ \ (\text{atm})}{C_{i,aq}\ \ (\text{mol/L})}$$

Where does K_D (soil-water partitioning) come from?

K_D is derived from K_{OC}

K_{OC}= organic carbon/liquid partitioning coefficient (m^3/g)

$\boxed{K_D\ (m^3/g) = K_{OC} * f_{oc}}$

f_{oc} = organic carbon fraction in soil [g OC/g soil]

K_{OC} is derived from K_{ow} through an empirical relationship

$\log K_{OC} = a * \log K_{ow} + c$ where a and c are empirical constants

$$\boxed{K_{OW}\ [-] = \frac{\text{conc. in octanol (mol/L)}}{\text{conc. in aq. sol'n (mol/L)}} = \frac{C_{i,oct}\ \text{(mol in octanol/L octanol)}}{C_{i,aq}\ \text{(mol in solution/L water)}}}$$

K_{OW} can be directly measured (like K_H' and K_{NW}) or computed

Mass and volume relationships in the soil matrix

- The soil (or sediment) is composed of solids, water and air.
- The solid portion is made of minerals, organic matter and microorganisms.
- The void volume in soil is filled with air and water. (mass of air is negligible)
- Due to density differences between air and water, dry and wet soil densities are different.

M_S= mass of dry solid (g)

V_S= volume of dry solid (m^3) V_L= volume of water in soil (m^3) V_G= volume of gas in soil (m^3)

V_T= total volume of soil (m^3) = $V_S + V_L + V_G$

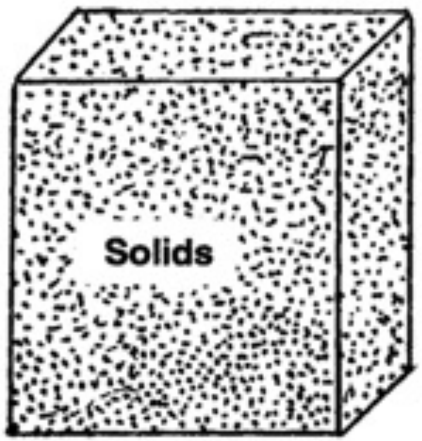
M_L= mass of water (g) = $\rho_W * V_L$
with ρ_W water density (g/m^3)

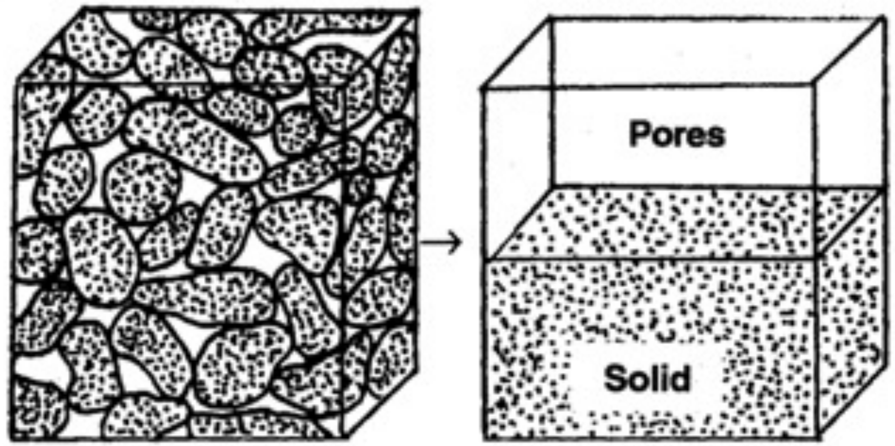
$$\rho_S = \text{(dry) solids density } (g/m^3) = \frac{\text{mass of dry solid (g)}}{\text{volume of dry solid } (m^3)} = \frac{M_S\,(g)}{V_S\,(m^3)}$$

$$\rho_b = \text{dry bulk density } (g/m^3) = \frac{\text{mass of dry solid (g)}}{\text{Total soil volume } (m^3)} = \frac{M_S\,(g)}{V_T\,(m^3)}$$

$$\rho_{wb} = \text{wet bulk density } (g/m^3) = \frac{\text{mass of wet soil (g)}}{\text{Total soil volume } (m^3)} = \frac{M_S + M_L\,(g)}{V_T\,(m^3)}$$

$$\rho_b < \rho_{wb}$$





solids density

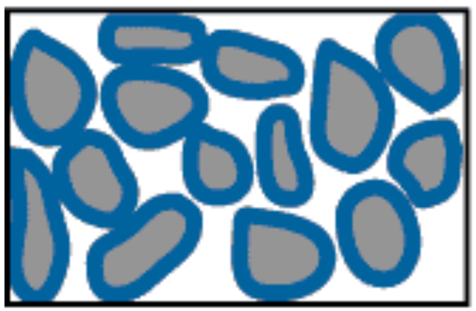
100% solid
M_S = Weight = 2.66 g
V_S = Volume = 1 cm³
ρ_s= 2.66 g/cm³

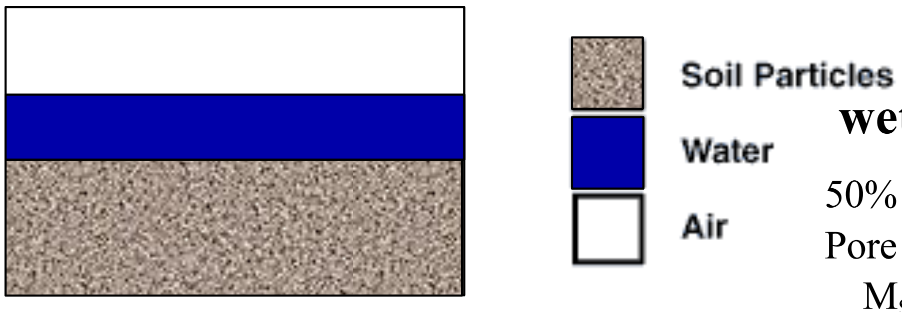
$$\rho_S = \frac{M_S\,(g)}{V_S\,(m^3)}$$

dry bulk density

50% solid, 50% pore space
M_S = Weight = 1.33 g
V_T = Volume = 1 cm³
ρ_b= 1.33 g/cm³

$$\rho_b = \frac{M_S\,(g)}{V_T\,(m^3)}$$





wet bulk density

50% solid, 50% pore space
Pore space: 40% water, 60% air
M_S = 1.33g
M_L = 0.2 g
V_T = 1 cm³
ρ_{wb}= 1.53 g/cm³

$$\rho_{wb} = \frac{M_T\,(g)}{V_T\,(m^3)} = \frac{M_{L+}M_S\,(g)}{V_T\,(m^3)}$$

Dimensionless sorption coefficient K_p

Convert K_D to a dimensionless ratio of mass concentration (K_p)

$$K_p \ [-] = \frac{\text{conc. sorbed (g/m}^3\text{)}}{\text{conc. in aq. sol'n (g/m}^3\text{)}} = \frac{C_{i,s,\,vol} \ \text{(g sorbed /m}^3\text{ solid)}}{C_{i,aq} \ \text{(g in solution/m}^3\text{ liquid)}}$$

$$= \frac{C_{i,s} \ \text{(g sorbed /g dry solid)}}{C_{i,aq} \ \text{(g in solution /m}^3\text{ liquid)}} * \frac{\rho_s \ \text{(g dry solid)}}{\text{(m}^3\text{ solid)}} = K_D \, \rho_s$$

$$\rho_s \ \text{solid density (g/m}^3\text{)} = \frac{\text{g dry solid}}{\text{m}^3\text{ dry solid}}$$

$$\boxed{K_p = \frac{C_{i,s,\,vol}}{C_{i,aq}} = K_D \, \rho_s}$$

Where $C_{i,s,\,vol}$ is the volume-based concentration of sorbed compound i on the solid phase

$C_{i,s,\,vol}$ (g sorbed/m^3 solid)= $C_{i,s}$ (g sorbed/g solid) * ρ_s (g solid/m^3 solid)

Mass distribution coefficient

If solids are suspended in water or an aquifer is being considered, the mass distribution of a substance i between solid and liquid is:

$$\frac{m_{i,s}}{m_{i.aq}} = \frac{mass\ i\ on\ dry\ solid}{mass\ i\ in\ solution} = mass\ distribution\ coefficient$$

$$\boxed{\frac{m_{i,s}}{m_{i.aq}} = \frac{C_{i,s} * V_S * \rho_S}{C_{i,aq} * V_L} = K_p * \frac{V_S}{V_L} = K_D * \rho_S * \frac{V_S}{V_L} = K_D * \left(\frac{M_S}{V_L}\right) = K_D * C_{ss}}$$

C_{SS} is concentration of suspended solids (g dry solids/m^3 solution)= M_S/V_L

M_S is the total mass of dry solids (g solids)= ρ_s (g solid/m^3 solid) * V_S (m^3 solid)

$f_{i,s}$ is the fraction of total mass i in solids (assuming no gas phase)

$$\boxed{f_{i,s} = \frac{mass\ of\ i\ on\ solids}{total\ mass\ of\ i} = \frac{m_{i,s}}{m_{i,s} + m_{i,aq}} = \frac{\left(m_{i,s}/m_{i,aq}\right)}{\left(m_{is} + m_{i,aq}\right)/m_{i,aq}} = \frac{\left(m_{i,s}/m_{i,aq}\right)}{\left(m_{i,s}/m_{i,aq}\right) + 1}}$$

$$\boxed{f_{i,s} = \frac{K_D * C_{ss}}{K_D * C_{ss} + 1}}$$

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$$\rho_b = \text{dry bulk density } (g/m^3) = \frac{\text{mass of dry solid (g)}}{\text{Total soil volume } (m^3)} = \frac{M_S\ (g)}{V_T\ (m^3)}$$

$$\rho_{wb} = \text{wet bulk density } (g/m^3) = \frac{\text{mass of wet soil (g)}}{\text{Total soil volume } (m^3)} = \frac{M_S + M_L\ (g)}{V_T\ (m^3)}$$

$$\varepsilon = \text{porosity [-]} = \frac{\text{volume of void } (m^3)}{\text{total volume of soil } (m^3)} = \frac{V_L + V_G\ (m^3)}{V_T\ (m^3)}$$

$$\omega = \text{soil moisture content (mass basis) [-]} = \frac{\text{mass of water (g)}}{\text{mass of dry solids (g)}} = \frac{M_L\ (g)}{M_S\ (g)}$$

$$\theta = \text{soil moisture content (volume basis) [-]} = \frac{\text{volume of water } (m^3)}{\text{Total volume of soil } (m^3)} = \frac{V_L\ (m^3)}{V_T\ (m^3)}$$

Dry bulk density of soils

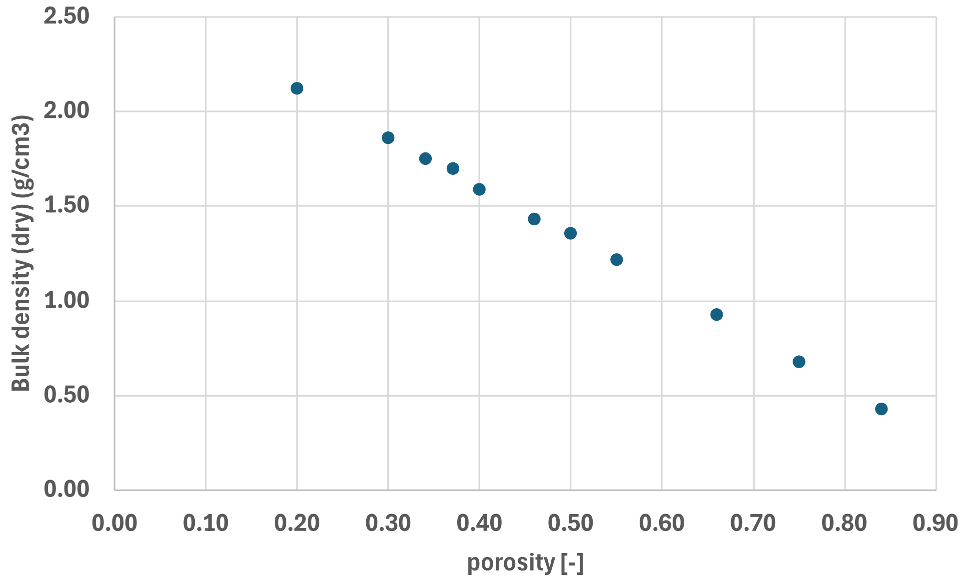
Soil Description	Porosity	Soil Bulk Density ρ_K (dry g/cm^3)
Uniform sand, loose	0.46	1.43
Uniform sand, dense	0.34	1.75
Mixed-grain sand, loose	0.40	1.59
Mixed-grain sand, dense	0.30	1.86
Windblown silt (loess)	0.50	1.36
Glacial till, very mixed-grained	0.20	2.12
Soft glacial clay	0.55	1.22
Stiff glacial clay	0.37	1.70
Soft slightly organic clay	0.66	0.93
Soft very organic clay	0.75	0.68
Soft montmorillonitic clay (calcium bentonite)	0.84	0.43

[a] From Peck et al. (1962).

$$\rho_b = \frac{M_S\,(g)}{V_T\,(m^3)}$$

High porosity typically results in **low ρ_b**.

This is because the solid phase takes up less volume (meaning less mass) than a low porosity material.



Henry’s law temperature effect

- Henry’s law constant increases by about 60% for each 10ºC rise in temperature

T (ºC)	$K'_{H,TCE}$
10	0.24
20	0.38
30	0.61

$$K'_{H,T} = K'_{H,20^{o}C} * (\Theta_H)^{(T-20)}$$

Θ_H=1.048

In-class problem 1:

Part 1. A groundwater is impacted by gasoline, and the average dissolved gasoline concentration is 20 mg/L. In situ bioremediation is being considered for the aquifer with the following characteristics:

Porosity= 0.35; Organic content= 0.02; **Dry bulk density** of aquifer materials= 1.6 g/cm$^{3;}$ Toluene (C_7H_8): K_{oc}= 182 L/kg

What is the concentration of toluene adsorbed to the solid? How much toluene in present in the solid phase of 1 m^3 of aquifer sediment?

In-class problem 1:

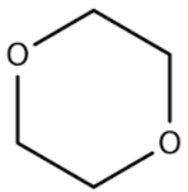
Part 2. A groundwater is impacted by gasoline, and the average dissolved gasoline concentration is 20 mg/L. In situ bioremediation is being considered for the aquifer with the following characteristics:

Porosity= 0.35; Organic content= 0.02; **Wet bulk density** of aquifer materials= 1.6 g/cm$^{3;}$ Toluene (C_7H_8): K_{oc}= 182 L/kg

What is the concentration of toluene adsorbed to the solid? How much toluene in present in the solid phase 1 m^3 of aquifer sediment?

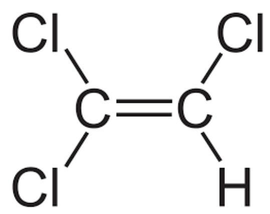
Partitioning of compounds among phases

	unit	1,4-dioxane	benzene	TCE	1,1,1-TCA	1,1-DCE
Water solubility	g/L (25 ºC)	1,000	1.79	1.28	0.91	2.42
Henry's Law	atm.L/mol at 25 °C	4.8×10^{-3}	5.55	9.85	16	26.1
$\mathrm{LogK_{OC}}$	-	1.23	1.75	2.0	1.95	1.85
Log Kow	-	−0.27	2.13	2.42	2.49	2.13

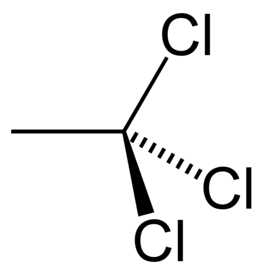


1,4-dioxane

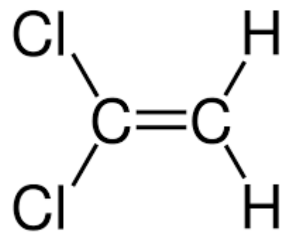
benzene



trichloroethylene (TCE)



1,1,1-TCA
1,1,1-trichloroethane



1,1-DCE
1,1-dichloroethylene

Other useful parameters

- Solubility product:

Solubility is the degree to which a solute with dissolve into a solvent. The solubility of chemicals in water is a function of the chemical species and temperature. Solubility is usually expressed in g/L.

$Ca^{2+} + 2\ OH^{-} \implies Ca(OH)_2\ (s)$

$K_{sp} = [Ca^{2+}][OH^{-}]^2 = 6.5 * 10^{-6}$

- In-class problem 2:

Calculate the solubility of $Ca(OH)_2$ in g/L at pH 12

MW of $Ca(OH)_2$ is 74 g/mol

Other useful parameters

- Vapor pressure:
  When a liquid is in contact with air, molecules leave the liquid as a vapor, via evaporation.
- Vapor pressure of a pure liquid is the pressure exerted by the vapor on the liquid at equilibrium

Raoult's law:

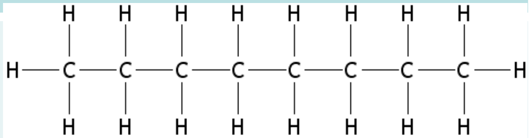
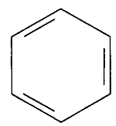
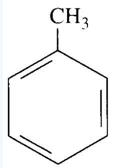
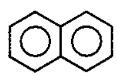
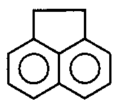
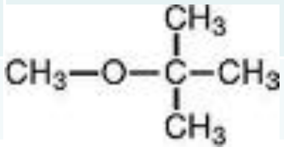
- $p_a = p_{vp,\,a} * x_a$ and $p_b = p_{vp,\,b} * x_b$
- p_a= partial vapor pressure of component a over the solution
- $p_{vp,\,a}$= vapor pressure of pure component a
- x_a= mole fraction of component a in solution= mol a/(mol a + mol b)
- $p_{total} = p_a + p_b$

Major classes of contaminants

- Petroleum hydrocarbons (including PAHs= polyaromatic hydrocarbons)
- Halogenated hydrocarbons (including PCBs= polychlorinated biphenyls)
- Pesticides
- Explosives
- PFAS
- Inorganic

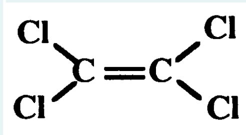
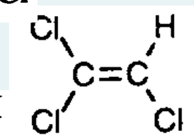
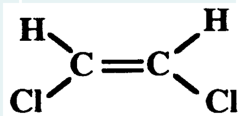
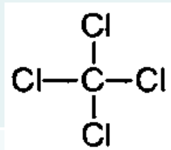
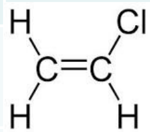
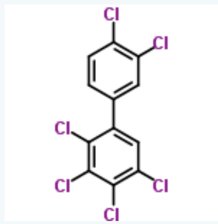
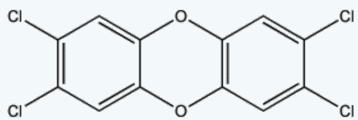
Petroleum hydrocarbons

- Aliphatic hydrocarbons
- Aromatic hydrocarbons (BTEX)
- Polycyclic aromatic hydrocarbons (PAHs)
- Oxygenates (added as fuel oxygenates for more efficient combustion)

	examples	structure	Water solubility (mg/L)	MW (g/mol)	source
aliphatic	octane hexane		0.78	114.23	fuel
aromatic	Benzene Toluene Ethylbenzene Xylene	 Benzene  Toluene	1,800 470	78.1 92.14	fuel
Polycyclic aromatic hydrocarbons (PAHs)	Naphthalene Acenapthalene	 Naphthalene  Acenaphthene	31.7 3.9	128.2 154.2	Burning byproduct, coal gas manufacturing
oxygenates	MTBE		50,000	88.15	additives

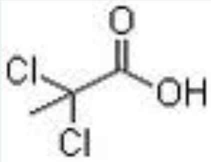
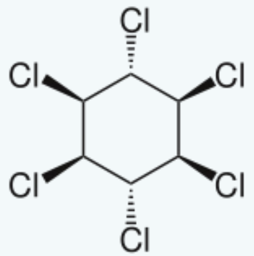
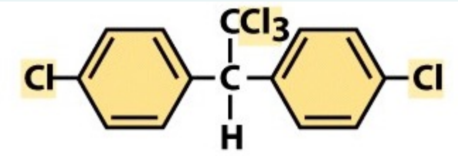
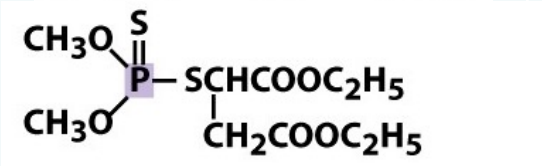
Halogenated hydrocarbons

- Halogenated aliphatics
- Halogenated aromatics

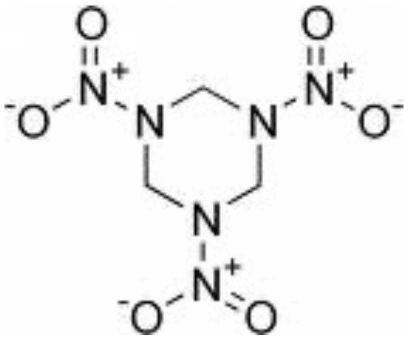
	examples	structure	Water solubility (mg/L)	MW (g/mol)	source
aliphatic	Tetrachloroethylene (PCE)		150	155.8	Dry cleaning, electronics, garages, military
	Trichloroethylene (TCE)		1,280	131.4	
	Dichloroethylene (DCE)		400	96.9	
	Carbon tetrachloride (CT)		785	153.8	
	Vinyl chloride (VC)				
Aromatic (dioxins and PCBs) often mixtures	2,3,3',4,4',5-Hexachlorobiphenyl		< 40 µg/L	360.8	Wood treatment, insulators
	2,3,7,8,-tetrachlorodibenzo-p-dioxin (2,3,7,8,-TCDD)		19.3 ng/L	321.9	Combustion, exhaust

Pesticides

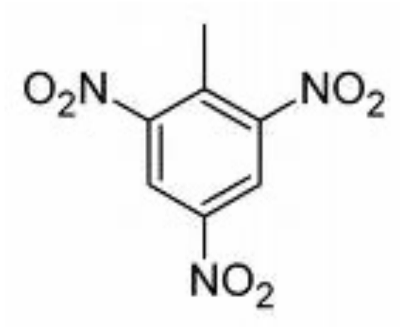
Large number of pesticides with many variations.

	examples	structure	Water solubility (mg/L)	MW (g/mol)	use
Halogenated aliphatic	2,2-dichloropropionic acid (Dalapon)		500 g/L	143	herbicide
Chlorinated cyclic aliphatics	Lindane		7.3	290.8	insecticide (not in use)
Halogenated aromatics	DDT		0.025	354.5	insecticide (not in use)
Organophosp -hates	Malathion		130	330.4	insecticide

Explosives



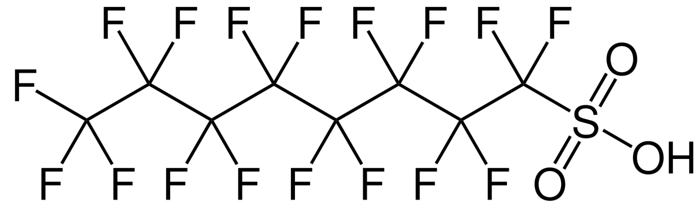
1,3,5-Trinitroperhydro-1,3,5-triazine
(RDX or T4)



Trinitrotoluene (TNT)

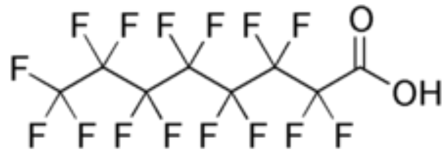
Emerging contaminants

Per and Poly-fluoroalkyl substances (PFAS)



- A large and complex family of compounds, all containing several F atoms
- Chemical designed to resist heat, oil, stains, and water
- Many are persistent due to the C-F bond (“forever chemicals”)
- Evidence for bio-accumulation
- Present everywhere at low concentrations (< European limit for drinking water- 0.5 μg/L)

- PFOA: perfluoro-octanoic acid
  (8-carbon chain)



- PFOS: perfluoro-octanesulfonic acid
  (8-carbon chain and sulfonate group)

<u>Example</u>:
In les Vernets (new development in Geneva on former military site), very high concentrations of PFAS in soil and GW (>1,000 ug/L). No legal basis in Switzerland for GW. Soil separation and washing was used to reduce the volume of contaminated material (by 75%). Gravel was recycled into cement, and the contaminated material was landfilled in type B (low-contamination) landfill in Geneva and in type E landfill (contaminated materials) in Bern.

Metals

1 H																1 H	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110	111	112		114		116		118

58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

- Cr(VI)= CrO_4^{2-}
- U(VI)= UO_2^{2+},
- Se(VI)= SeO_4^{2-}
- Zn^{2+}, Cd^{2+}, Pb^{2+}