

Comparison of Leaching of PAHs from two Contaminated Soils under Varying Hydraulic Retention Time

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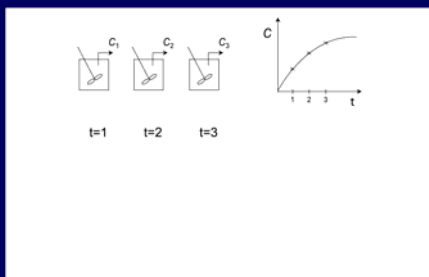


Background

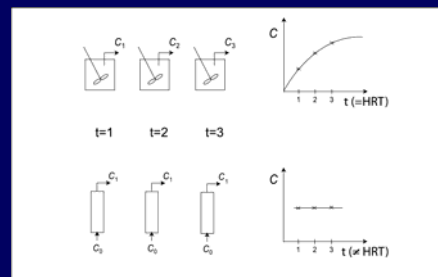
- Hydraulic retention time, HRT, is an important parameter when conducting leaching experiments
- HRT = the contact time between the liquid and the solid phase



Background

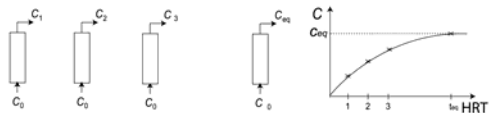


Background



Background

$$\text{HRT}_1 < \text{HRT}_2 < \text{HRT}_3 \dots < \text{HRT}_{\text{eq}}$$



Objectives

- To investigate which processes control leaching of PAHs from aged contaminated soil
- To investigate if leached concentrations from field contaminated soil, obtained by dynamic column leaching, can reach equilibrium concentrations at very short contact times
- To demonstrate that leaching of PAHs from aged contaminated soils can show significant differences in leaching behaviour, leachable concentrations and available amounts

Soil samples



1. Husarviken, Stockholm, Sweden
 - former gasworks plant (1893-1972)
 - **coal tar**, heavy metals, cyanide

2. Holmsund, Umeå, Sweden
 - former impregnation facility (1943-1983)
 - **creosote**, As, Cu, Cr

Soil data

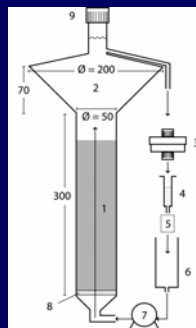
Parameters	Coal tar cont. soil	Creosote cont. soil
TOC %	16	3
pH water (1:10)	7.62	5.64
Density kg/L (d.w.)	1.36	1.06
Composition of soil (%)		
Coarse sand (>0.2 mm)	54	62
Fine sand (0.02-0.2 mm)	20	23
Silt (0.002-0.02 mm)	21	10
Clay (<0.002 mm)	5	5

Initial amounts

PAH	Abbreviation	Coal tar cont. soil (mg/kg soil)	Creosote cont. soil
Fluorene	FLU	7 ± 1	110 ± 12
Phenanthrene	PHE	61 ± 3	240 ± 43
Anthracene	ANT	60 ± 3	226 ± 101
Fluoranthene	FLU	221 ± 43	1066 ± 36
Pyrene	PYR	177 ± 34	521 ± 24



Design of the leaching experiment



1. Glass column
2. Sedimentation chamber
3. Filter
4. SPE cartridge
5. Drop counting detector
6. Reservoir
7. Peristaltic pump



Experimental set up



- Glassware and stainless steel
- Sterilization, HgCl_2
- 0.7 μm filter
- 0.5 kg soil



Experimental flow rates and HRTs

Husarviken (coal tar contaminated soil)

- Elevation difference of 38 m!
- Large proportion of medium to coarse material



High groundwater velocities



Experimental flow rates and HRTs

Flow rates chosen for the experiments: 0.1 – 0.5 L h⁻¹
(Darcy velocities: 450 – 2200 m year⁻¹)

$$HRT = \frac{m_{d.w.} \cdot \varepsilon}{Q \cdot \rho_{d.w.}}$$

Q = flow rate
m_{d.w.} = amount of sample material
ε = porosity
ρ_{d.w.} = density

⇒ HRT: 0.3 – 1 h



HRTs, Husarviken

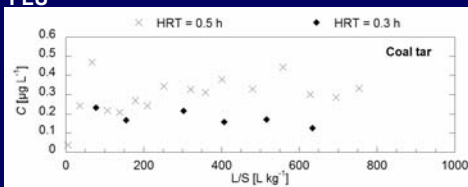
Husarviken_coal tar

HRT = 0.3 h x 2
0.5 h x 2
1.0 h x 1



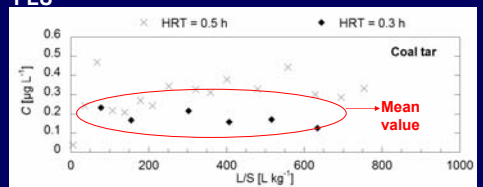
Leached concentrations, Husarviken

FLU

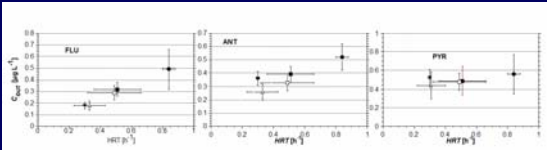


Leached concentrations, Husarviken

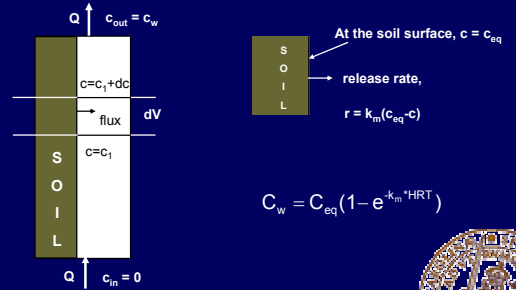
FLU



Leached concentrations, Husarviken



Modelling the column leaching



Modelling results

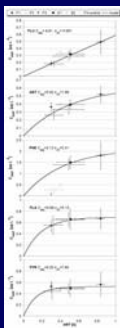
FLU $k_m = 0.006$

ANT $k_m = 1.99$

PHE $k_m = 2.31$

FLA $k_m = 5.13$

PYR $k_m = 7.60$



188 cm³ mol⁻¹

Le Bas molar volume

217 cm³ mol⁻¹



Interpretation of k_m

$$k_m \propto \frac{D \cdot A}{\delta \cdot V}$$

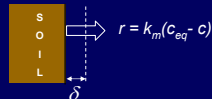
k_m = mass transfer coefficient (h⁻¹)

D = the diffusivity of the solute (cm² s⁻¹)

A = the sorbent surface area (cm²)

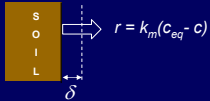
V = the pore water volume (cm³)

δ = the distance of the mass transfer (cm)



Interpretation of k_m

$$k_m \propto \frac{D \cdot A}{\delta \cdot V}$$



- k_m = mass transfer coefficient (h^{-1})
- D = the diffusivity of the solute ($\text{cm}^2 \text{s}^{-1}$)
- A = the sorbent surface area (cm^2)
- V = the pore water volume (cm^3)
- δ = the distance of the mass transfer (cm)

Conclusion: The leaching is not a result of dissolution of PAHs from a free phase of coal tar!



Interpretation of k_m

$$k_m \propto \frac{D \cdot A}{\delta \cdot V}$$



- k_m = mass transfer coefficient (h^{-1})
- D = the diffusivity of the solute ($\text{cm}^2 \text{s}^{-1}$)
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- V = the pore water volume (cm^3)
- δ = the distance of the mass transfer (cm)

D differ approx. 1.1 between the PAHs studied



Interpretation of k_m

$$k_m \propto \frac{D \cdot A}{\delta \cdot V}$$



- k_m = mass transfer coefficient (h^{-1})
- D = the diffusivity of the solute ($\text{cm}^2 \text{s}^{-1}$)
- A = the sorbent surface area (cm^2)
- V = the pore water volume (cm^3)
- δ = the distance of the mass transfer (cm)

Conclusion: The leaching may be a result of mass transfer resistance within the solid phase!



HRTs, Holmsund

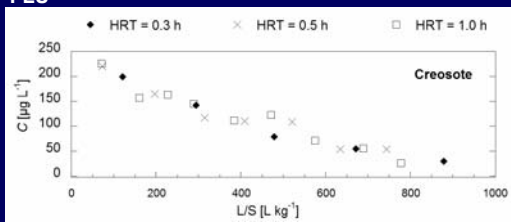
Holmsund, creosote

HRT = 0.3 h x 1
0.5 h x 1
1.0 h x 1



Leached concentrations, Holmsund

FLU



Leachable amounts

PAH	Creosote cont. soil		Coal tar cont. soil	
	% leached of total amount			
Fluorene	77		1.84	
Phenanthrene	32		0.46	
Anthracene	6		0.64	
Fluoranthene	4		0.18	
Pyrene	4		0.16	

Solubility limitations?

PAH	Creosote cont. soil	
	C _{max}	S
	[µg L ⁻¹]	
Fluorene	215 ± 14	440
Phenanthrene	166 ± 8	870
Anthracene	26 ± 2	51
Fluoranthene	85 ± 4	68
Pyrene	35 ± 2	47

Conclusion: The leaching may be a result of dissolution of PAHs from a free phase of creosote!

Solubility limitations?

PAH	Creosote cont. soil		Coal tar cont. soil	
	C _{max}	S	C _{mean}	S
	[µg L ⁻¹]		[µg L ⁻¹]	
Fluorene	215	440	0.31	220
Phenanthrene	166	870	1.36	190
Anthracene	26	51	0.35	53
Fluoranthene	85	68	0.59	18
Pyrene	35	47	0.42	15

Conclusions

- The leaching from the creosote contaminated soil, collected from Holmsund, was probably governed by dissolution.
- Desorption processes most likely controlled the release of PAHs from the coal tar contaminated soil from Husarviken.
 - Leached concentrations of the heavier PAHs seemed to be close to distribution equilibrium concentrations, despite the rather short HRT
- Significant differences in leaching behaviour, leached concentrations and available amounts –although initial concentrations were similar and both samples were aged
- Leaching tests for organic contaminants can provide valuable information for risk assessments!

