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Modelling attenuation processes in the unsaturated zone beneath landfills

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CONTEXT

External drivers

- EU directives (Groundwater, Water Framework, Landfill)
- Regulator (EA)
- Waste Management Licensing Reg. 15

Problem of old 'dilute & disperse' sites

- Many sites still actively leaching
- Sited over important (major) aquifers
- Liability

Role of unsaturated zone

- Ability of unsaturated zone to attenuate
- Need for tools to model and predict effects

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Burntstump landfill site

Site History

- Situated on Triassic Sherwood Sandstone ~ 8 km north of Nottingham, UK
- Site used for sandstone quarrying
- Landfilling commenced in early 70s
- MSW with some industrial waste (Coal mining)
- Old cells without liners (dilute & disperse)
- Some of oldest cells now 'restored'

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Burntstump landfill site

Monitoring History

- Sequential unsaturated zone monitoring – cored borehole samples
- First sampled in 1978
- Periodic re-sampling (1981, 1985, 1987, 1991, 2000)
- Unique and highly important time series

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Burntstump landfill site

Modelling History

- EA/Golders, Landsim, 1996
  - Cl, NH4
- Erskin, QJEG&H, 2000
  - NH4
- Jones et al., QJEG&H, 2001
  - Cl

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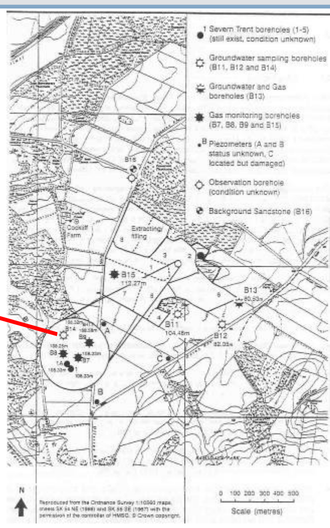
Burntstump landfill site



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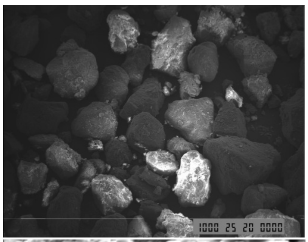
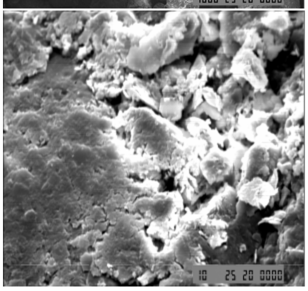
Sherwood sandstone

Grains of quartz and feldspar.
Magnification x50, Depth 8.85m.



Clay band. Magnification x1000.
Depth 27m.



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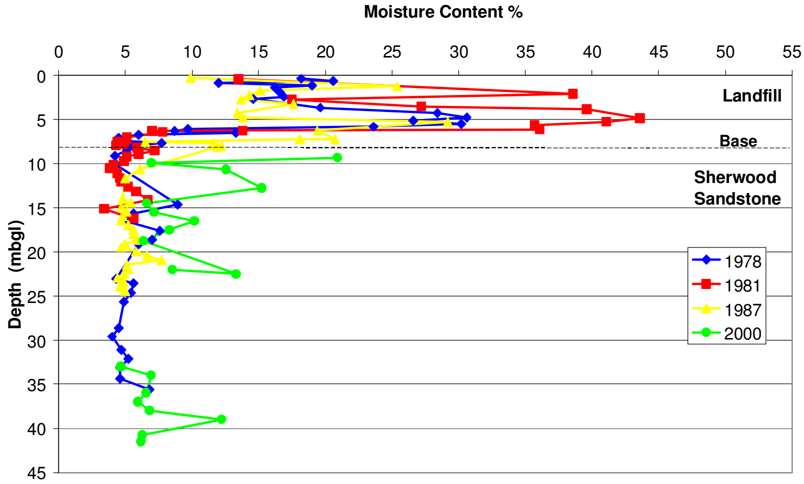
Moisture content



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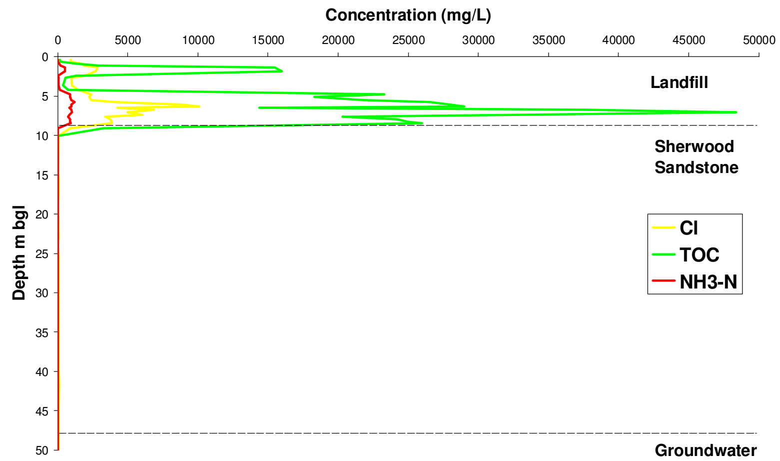
1978



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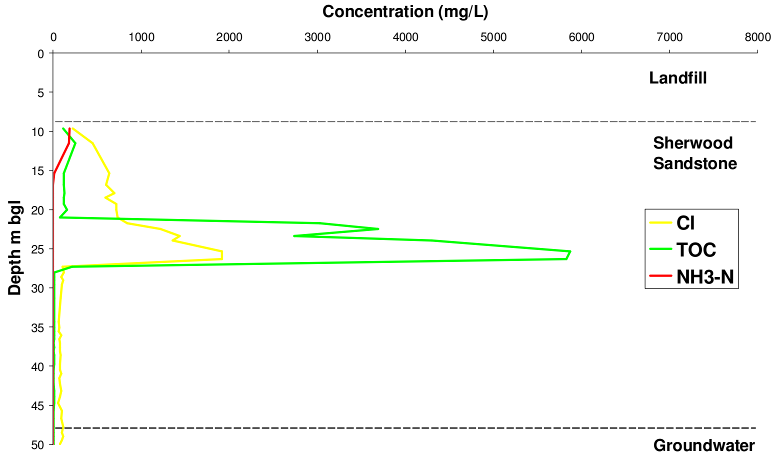
1991



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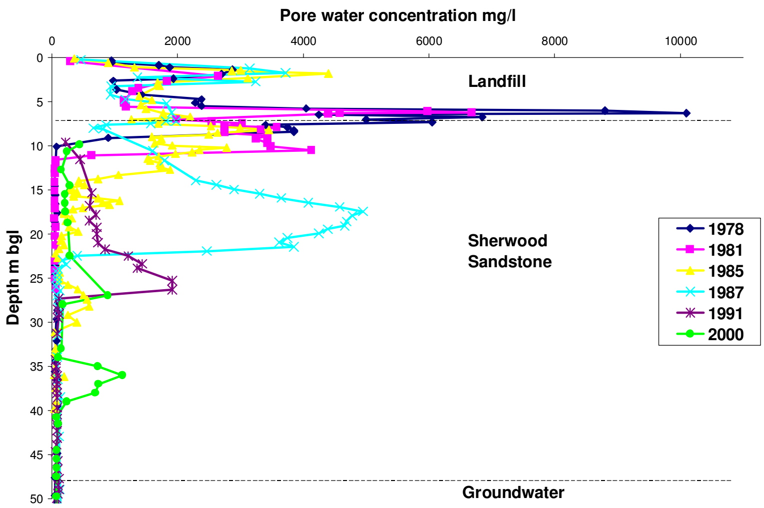
Chloride



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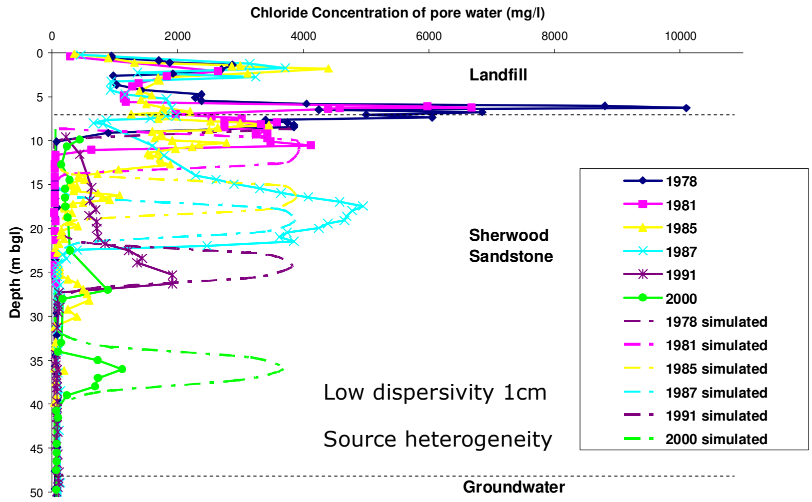
Cl modelling



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Cl migration

- Leachate front moves at ~ 1.7 m/yr
- Recharge rate of ~ 100-120 mm/yr
- Relatively low dispersion of leading edge
- Total Cl mass appears to be highly variable
- Effects of vertical heterogeneity (e.g. 1985, 2000)
- Effects of source heterogeneity

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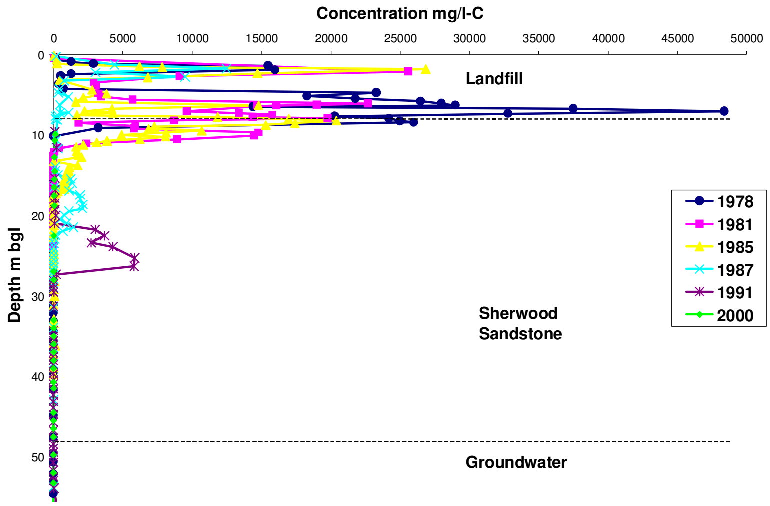
TOC



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Contaminant transport (which model?)

$$\frac{\partial(\theta + \rho S)}{\partial t} = \frac{\partial\left[\theta D_h \frac{\partial c}{\partial z} - qc\right]}{\partial z} - \lambda(\theta + \rho S)$$

$$\frac{\partial(\theta + \rho S)}{\partial t} = \frac{\partial\left[\theta D_h \frac{\partial c}{\partial z} - qc\right]}{\partial z} - \lambda c$$

c = soil water concentration (kg m^{-3})
S = sorbed soil concentration (kg kg^{-1})
D_h = hydrodynamic dispersion (m^2 s^{-1})
q = soil water flux (m s^{-1})
ρ_b = soil dry bulk density (kg m^{-3})
λ = decay constant (s^{-1})

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Attenuation coefficient

$$S = K_d c \longrightarrow R = 1 + \frac{\rho_b K_d}{\theta}$$

$$\frac{\partial c}{\partial t} = \frac{D_h}{R}\frac{\partial^2 c}{\partial x^2} - \frac{v}{R} - \frac{\lambda}{R} c$$

$$\frac{\partial c}{\partial t} = \frac{D_h}{R}\frac{\partial^2 c}{\partial x^2} - \frac{v}{R} - \lambda c$$

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Attenuation coefficient

$$S = K_d c \longrightarrow R = 1 + \frac{\rho_b K_d}{\theta}$$

Lagrangian perspective

$$\frac{\partial c}{\partial t} = -\frac{\lambda}{R} c$$

$$\frac{\partial c}{\partial t} = -\lambda c$$

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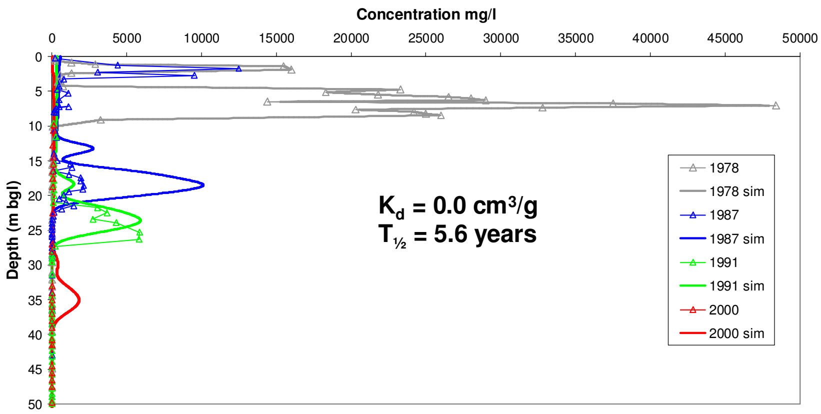
TOC

Simulated and observed TOC concentrations



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TOC migration

- Downward transport rate similar to Cl
- Marked attenuation of observed pore water concentrations
  - 1978 – 1991 T½ ≈ 6 yrs
  - more rapid between 1991-2000
  - microbial degradation (anaerobic/aerobic)
- Effects of heterogeneity (e.g. 1985,1991)?

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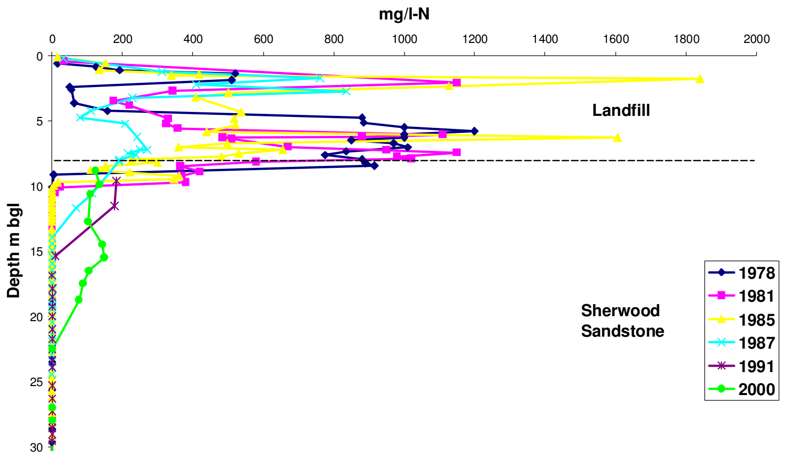
Amoniacal-N



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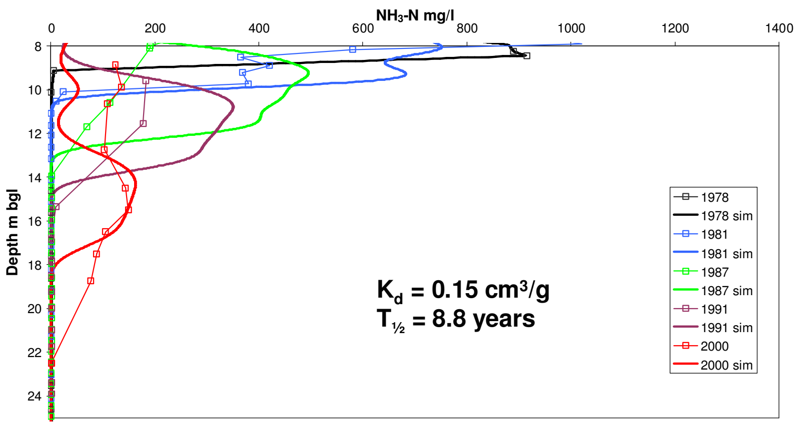
NH3 modelling



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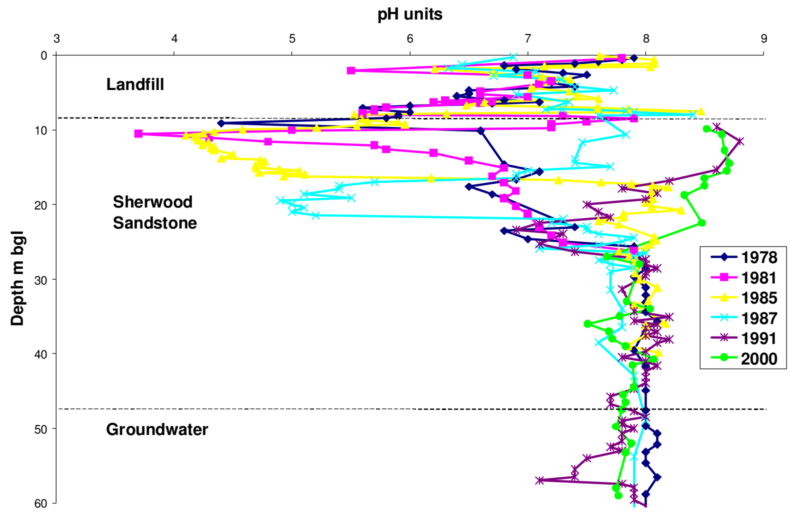
pH



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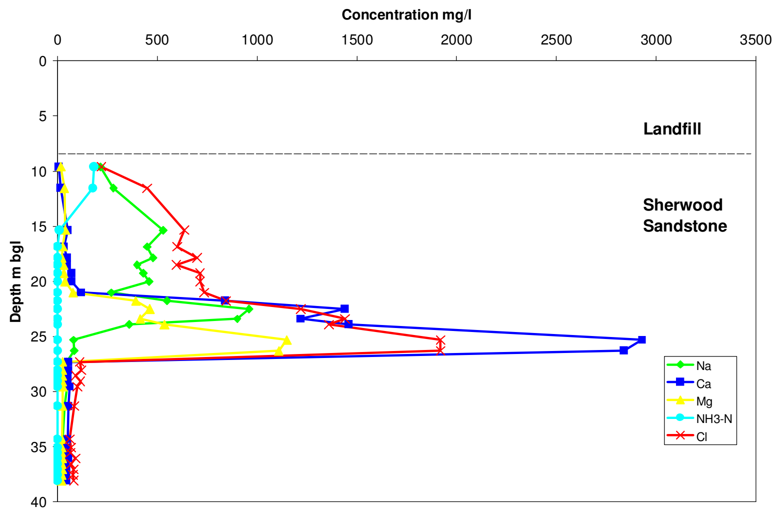
1991 Geochemical Profile



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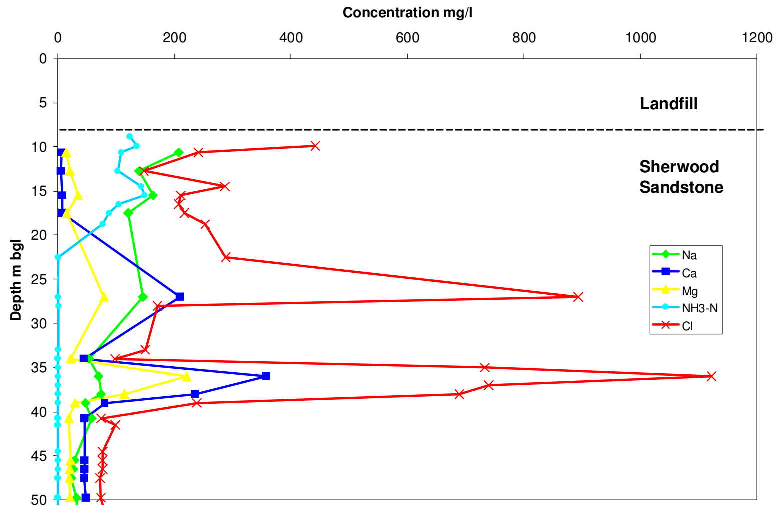
2000 Geochemical Profile



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Geochemical modelling (PHREEQC)

- PHREEQC-2 (Parkhurst and Appelo, 1999)
- Batch-reaction/ 1D transport
- Reversible reactions
  - Solution/dissolution
  - surface-complexation
  - ion-exchange
- Irreversible reactions
  - biodegradation
- Model application – challenging!

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Conceptual sandstone model

- 1-D 45m column of unsaturated sandstone
- Ambient conditions:
  - quartz matrix
  - dolomite cement
  - equilibrium chemistry (pH 7.8)
- Constant flow
- Time varying source term (based on observations)
  - organic acids + inorganic ions
  - initially pH 3
- Geochemical processes
  - dolomite dissolution (kinetic)
  - biodegradation (Michaelis-Menten rate kinetics)
  - cation exchange

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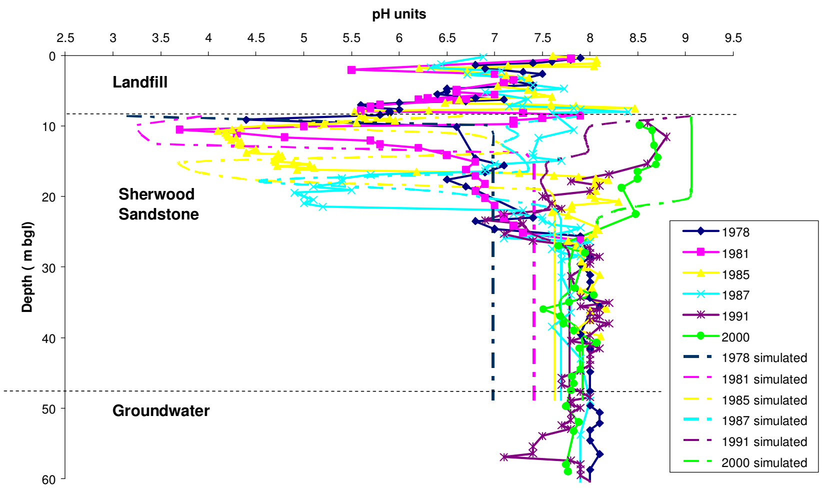
pH modelling



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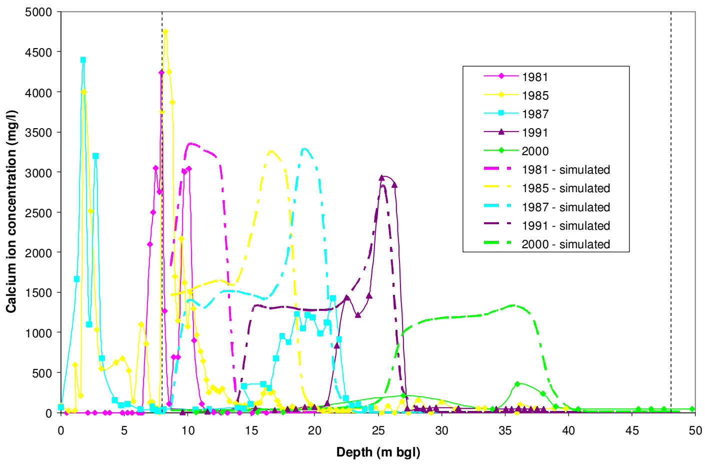
Ca



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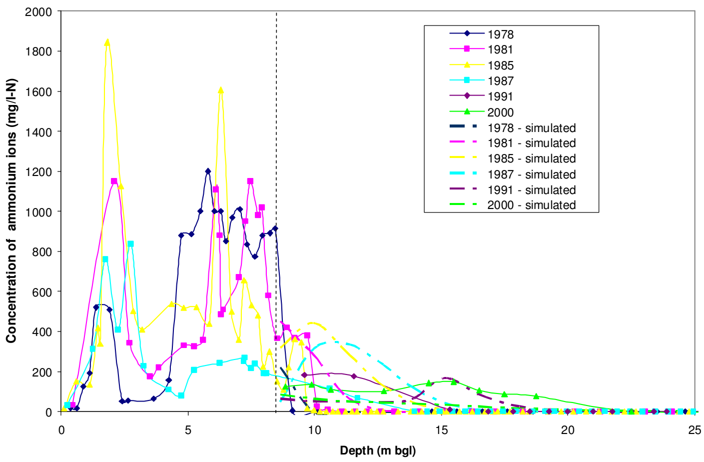
NH3-N



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Geochemical modelling insights

- Highly acidic leachate enters sandstone during ~ first 5 years of operation
- Results in dissolution of the sandstone Ca, Mg cementation material (similar molar concentrations of Ca/Mg indicates possible presence of Mg-calcite/dolomite) – also generates HCO3-
- Reaction results in pH buffering
- Ca/Mg act as ion balance for Cl- and HCO3- and, along with organic components, migrate with the flowing recharge water
- Microbial degradation results in reduction of organic compounds (esp. OAs), related to changes in pH and possibly enhanced by changes in redox related to methane extraction
- Dissolution/transport processes followed by mineral precipitation – resulting in changes in rock mineral structure
- NH3-N sorption occurs on the amended rock structure and is likely to be enhanced by the raised pH levels

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Conclusions

- Sharp chloride/TOC front → very little dispersion
- Apparent reduction in chloride mass
- Similar observations with TOC concentrations (despite overall attenuation)
- Likely cause is ***small-scale heterogeneity***
- Ammoniacal-N sorption on the amended rock structure
- Likely to be enhanced by the raised pH levels ($R = 4 \rightarrow K_d \approx 0.12$ cm^3/g)
- Possible attenuation but difficult to quantify

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Implications

- Major potential for attenuation of leachate contaminants in unsaturated Sandstone
- Geochemistry of rock material key component
- Large depth of unsaturated zone allows substantial degradation prior to impacting water table
- Ammoniacal-N retarded – possible degradation, potential for further retardation on clay bands

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