Cs and Sr transfers in Chernobyl Pilot Site soils (Chernobyl Exclusion Zone)

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Context

Site characterization

Modelling and system understanding

Summary & Perspectives for EPIC

Transfer to F-TRACE?
THE CHERNOBYL NPP ACCIDENT

- 04/26/1986: Explosion of Chernobyl NPP’s reactor n° 4
- during 10 days: $\sim 13650$ PBq of radionuclides (RN) rejected in atmosphere (till 10 km high)
- Volatile elements: $^{131}$I, $^{133}$I, $^{137}$Cs, $^{90}$Sr, $^{129}$Te, $^{132}$Te, $^{103}$Ru et $^{106}$Ru
- Inert gases: $^{85}$Kr, $^{133}$Xe
- Refractory elements: Pu isotopes

(adapted from Smith & Beresford, 2005).

THE PILOT SITE (EPIC)

Map indicating the location of different temporary waste deposit zones in Chernobyl exclusion zone and Sr-90 contaminated underground waters (Antropov et al., 2001)
RN dissolve and transfer from the trench to the unsaturated zone with percolating rainwater

The trench is located in the unsaturated zone

Contaminated sand with nuclear fuel particles and some radioactive organic material

Sources of radioactive contamination: dissolution of fuel particles & degradation of OM

Site instrumentation (UZ):
- Suction pressure,
- Moisture content and temperature captors
- Soil water samplers

Site instrumentation (SZ & background):
- Piezometers
- Weather station
- Laboratory
- Quartz (98-99%)
- Heavy minerals (1-2%)
- Organic carbon (<0.3%)

Clay fraction (<1%):
- illite, smectite, chlorite
- feldspaths

quartz (90-94%)
feldspar K-Na (5-9%)
heavy minerals (<0.5%)
hydromica,
montmorillonite,
crystalline calcite,
quartz, amorphic iron oxides (<0.5%)

DERIVATION OF Cs AND Sr DATABASE

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- Soil water samplers

Site instrumentation (SZ & background):
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Monitoring data

CHERNOBYL database
25 years after the accident:

* All the soluble particles should have been dissolved (UO$_{2+x}$ type), a priori
* All RN from dissolved FP should have been released into soil solution, a priori

→ Mainly the less degradable compounds are left (UO$_2$ & ZrU$_y$O$_x$ types)?
→ Decrease of available stocks and of Sr fluxes?

Specific activity in $^{137}$Cs & $^{90}$Sr in the trench 22 is $\sim 10^5$ to $10^6$ Bq/kg

(Guillou et al., 2000; Kashparov et al., 2004; Dewière et al., 2004)
THE SOURCE TERM: Cs and Sr distribution

Layout of the trench-3D model

Spatial distribution of $^{137}$Cs in the trench (3D model)

- Integration of $^{137}$Cs activity ponctual data
- Interpolation of these data by kriging method

Results:
- 2D and 3D spatial distribution of $^{137}$Cs in the trench
- A better definition of the trench layout

Parameter | Value
--- | ---
Groundwater level elevation (multi-year mean value), m a.s.l. | 111.5
Flow direction in eolian layer | North ($\pm 15^\circ$)
Horizontal hydraulic head gradient in eolian layer | 0.0015
Vertical hydraulic head gradient in alluvial layer | 0.03
Infiltration recharge rate, mm/y | 300
Hydraulic conductivity of eolian layer (isotropic), m/d | 3.6
Hydraulic conductivity of alluvial layer (anisotropic), Kx, m/d | 0.5
Kz, m/d | 0.0275

(Van Meir et al., 2009)
NUMERICAL SIMULATION OF Sr90-PLUME EVOLUTION

Stationary conditions - 2D model

Transport model in the groundwater:
- No chemical-physical variations
- No variations of groundwater flow
- No seasonality
- No OM as part of the source term

Global model
- Kinetic model of fuel particles dissolution (dynamic model)
- Stationary model for the saturated zone (plume)

Extension of the plume:
- 60 m length
- 4 m depth

Extension of the plume:
- 200 m length
- 10 m depth

(Bugai et al., 2011)
migration velocity $^{137}$Cs: ~0.16 cm/y (Szenknect, 2003; Guillou et al., 2000)

- Low content in $^{137}$Cs measured in the aquifer downgradient T22: <0,1 Bq/L
- 2011-12: above detection limit concentrations in the groundwater: >3 Bq/L
- $^{137}$Cs activities in T22 soil solution: 45-52 Bq/L >> 0,03 Bq/L in Pripyat river
- Strong affinity of Cs with OM (cf. fig)
- Cs is used by the plant (competing with nutrients like; Ca, K, etc.)

What about the Cs plume at T22?

$^{137}$Cs is mainly located in the trench and use by the vegetation

### THE SECONDARY SOURCE TERM

- tree-trunks
- forest contaminated soils + liter (OM 3-6%)
- vegetable debris: needles, branches, …
- other sup. plants: herbs, shrubs, …

**Kinetic** of organic decaying:
- fast decaying of easily degradable compounds ($T_{1/2}= 3-4$ years) for fine liter (Pausas, 1997)
- slow decaying of less degradable compounds ($T_{1/2}= 7-42$ years) for coarser materials (Currie et al., 2002)

25 years after the accident:

Most of the organic compounds easily degradable have been transformed → *Mainly the less degradable compounds are left (ex.: trunks)*?
**BIOTIC MIGRATION OF RADIONUCLIDES**

1998

Vegetation on the top of the trench: 47 pines, 14 birch trees, 49 bushes (at approx. 400 m² area)

**RN activity in trees (2001)**
- $^{90}$Sr - 0.6 – 6.7 MBq/kg
- $^{137}$Cs - 0.01 – 2.1 MBq/kg
(Kashparov et al., 2002)

2008

**Chronicle of $^{90}$Sr fluxes evolution in the aquifer vs. water-table fluctuations**

![Chronicle of $^{90}$Sr fluxes evolution in the aquifer vs. water-table fluctuations](chart.png)
Chronicle of $^{90}$Sr fluxes evolution in the aquifer vs. water-table fluctuations

Evolution of $^{90}$Sr fluxes coming out of the trench and to the aquifer

Explicative processes:
- $\downarrow$ stock of soluble fuel particles
- $\downarrow$ stock of easily degradable organic matter
- modification of the physical-chemical environment
- $\uparrow$ of transfers (cations & RN) to the superior plants with $\uparrow$ of their biomass

Role of vegetation: - direct RN uptake; - nutrient element (Ca,K,...) uptake, which influences GW geochemistry and hence - RN mobility
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90Sr fluxes from the trench may be controlled by:
- Dynamic of bio-physical-chemical modifications linked with stocks of buried OM and with their kinetic of degradation
- Transfers towards plants which lead to an increase with time of plant biomass
- Seasonality
WHAT DID WE LEARN?

- Complex relationships between hydrogeological, geochemical and biological processes observed in the “real world” of a contaminated site such as the Red Forest (EPIC site) require the achievement of interdisciplinary researches.
- The realization of relevant predictive calculation requires the use of a GLOBAL MODEL coupling transfers and RN migration in every compartment of interests (atmosphere, UZ, SZ, vegetable cover), and to consider the influence of the main geochemical and biological factors.

WHAT IS NEXT?

- Global numerical modeling coupling biogeochemistry-transport for Sr, Cs.
- Evolution of the buried source term 25 years after: reevaluation of inventories for FP and OM. What is the main process that rule the dynamic of their evolution?; What is the role of microorganisms and their influence?; Water-table fluctuation influence (flood of the bottom of the trench) vs. precipitation influence on the dynamic of RN release?
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- Modeling of the most complex cases (Pu, U, etc.)? These last will need the use of other coupling (e.g. colloids role; interaction with microorganisms; factors controlling speciation variations, ...)
- Dynamic of the RN uptake by plants (bioavailability, translocation, bioaccumulation)
- The importance and the impact of a new source term, more diffuse on soil surface (contaminated liters); modification of RN speciation and their reactivity?
- Scale changing: from the pilot site to the exclusion zone (water-basin scale)

Post-accidental situation: What can we learn? Contribution to F-TRACE

How to:

- Manage soil decontamination in a post-accidental situation?
- Remove radionuclides from soils?
- Manage remediation wastes?
- Organize the return of population and under what exposure conditions?
- Manage land reuse when population returns?
- Manage/organize monitoring after the return of population?

Map of Cs-137 (Bq/m²)
Which tools can we use?

Laboratory Studies & Field Observations → Integrated Modeling

How geochemistry and hydrology can help?
- Long-term predictions of RN transfers
- Impact assessment
- Upscaling
- Remediation solutions
- Population return and use of land

How geostatistics can help?

How biochemistry can help?

How modeling can help?
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DGT can be used for many different purposes, including:
- In situ measurements
- Monitoring (time averaged concentrations)
- Speciation (labile inorganic and/or organic species)
- Bioavailability (effective concentration)
- Trace metals, phosphate, sulphide and RN
- Fluxes & conc. in sediments/soils & fresw./seaw.
- Kinetic and thermodynamic constants
- High spatial resolution measurements (sub-mm)
- 2D concentration images

DET can be used for in situ measurements of solute concentrations at high spatial resolution

DET: Diffusive Equilibration in Thin films
DGT: Diffusive Gradient in Thin-films

DGT & DET were invented by Bill Davison and Hao Zhang
How geostatistics can help?

- How to improve RN mass estimate from an optimized sampling plan?
- How to assess uncertainty on contaminated volumes of soils?

Which tools can we use?

How biochemistry can help?

Which biochemical functions are lost in a contaminated soil?

Metagenomic analysis (i.e. analysis of the DNA of all the soil microorganisms) could help to:
- assess the contamination impact
- better identify biochemical reactions occurring in the contaminated soils and whether they impact RN fate

What about Fukushima Contaminated Soils?

Delmont et al. 2011
THANK YOU VERY MUCH FOR YOUR ATTENTION!