

CArbon-14 Source Term CAST

Name: Erika Neeft

Organisation: COVRA (WMO)

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Content

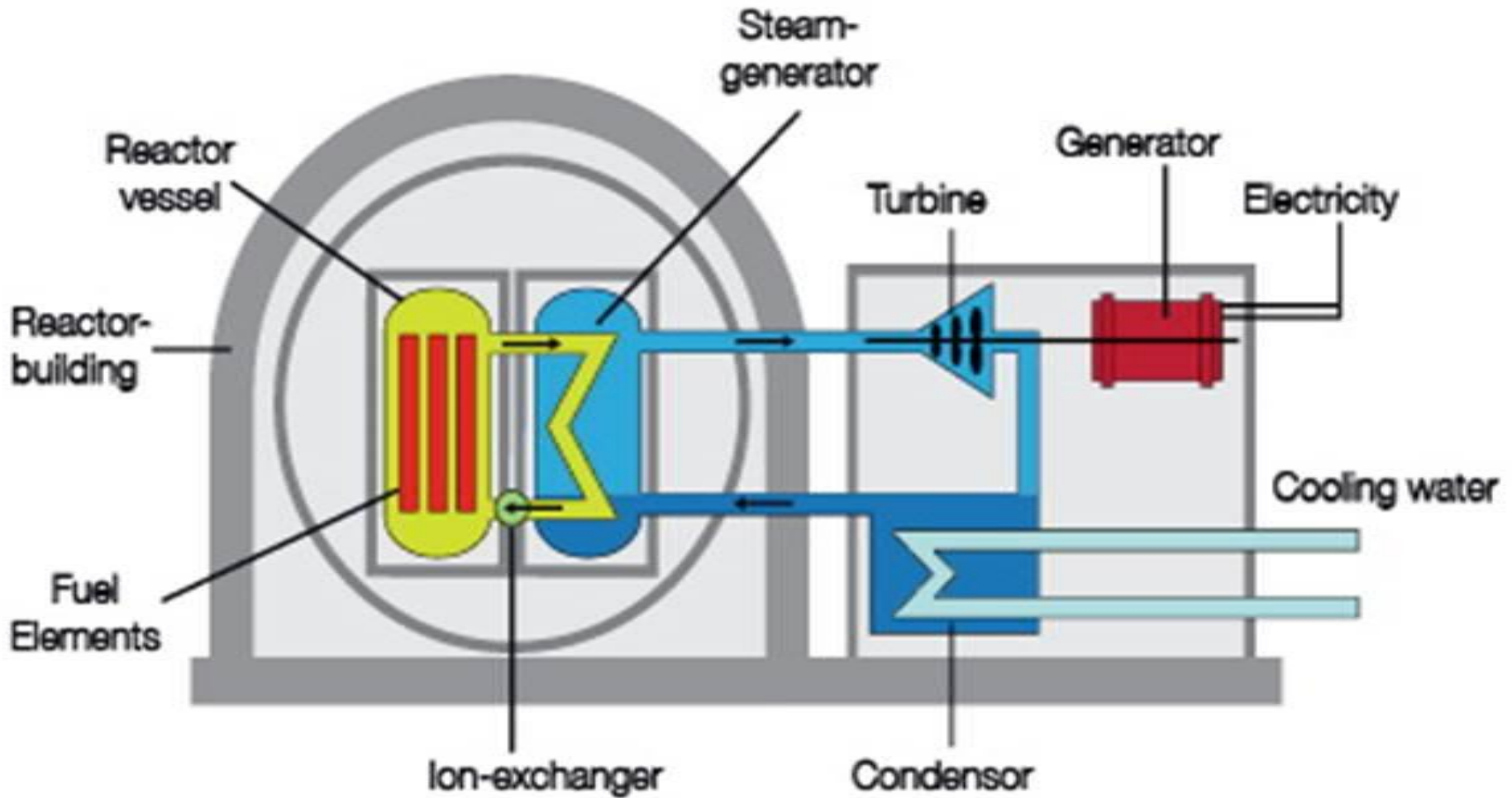


- Radionuclides
 - Generation carbon-14 in a nuclear plant and by cosmic radiation
 - Neutron activation of nitrogen-14
 - Different neutron source
 - Left for (geological) disposal
 - ETM and DTM
 - carbon-14
 - » Clearance level carbon-14 in waste in EU
- Disposal of waste
 - Potential migration of released radionuclides
 - Gas, dissolved, retarded
 - Natural carbon-14
 - Exposure
 - Potential exposure mechanism artificial carbon-14 if released as gas
 - Carbon-14 Source Term
 - Types of waste investigated
 - Carbon speciation from deep sea hydrothermal vents
 - Potential release mechanisms at (geological) disposal conditions
 - Cementitious materials

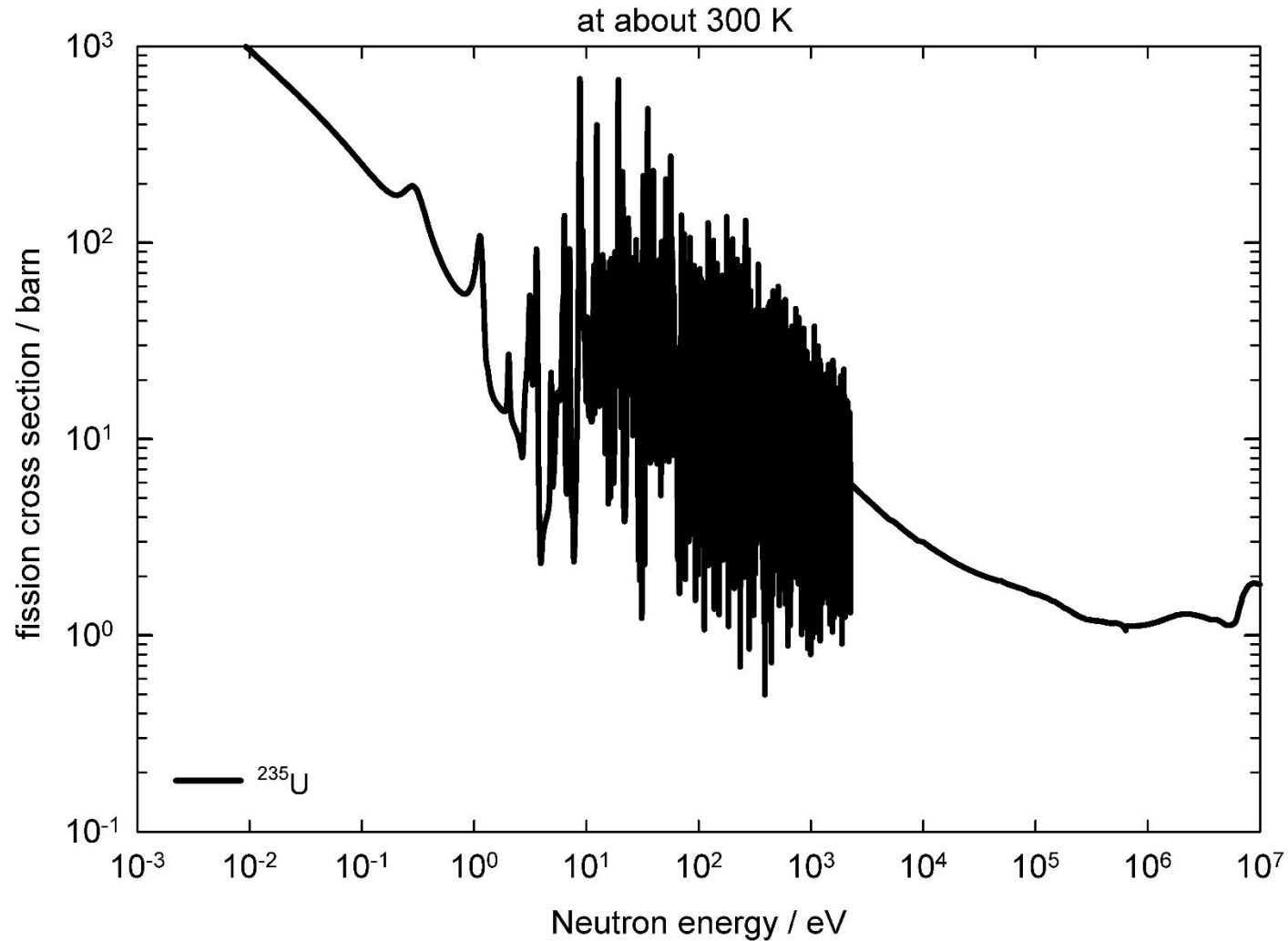
Generation



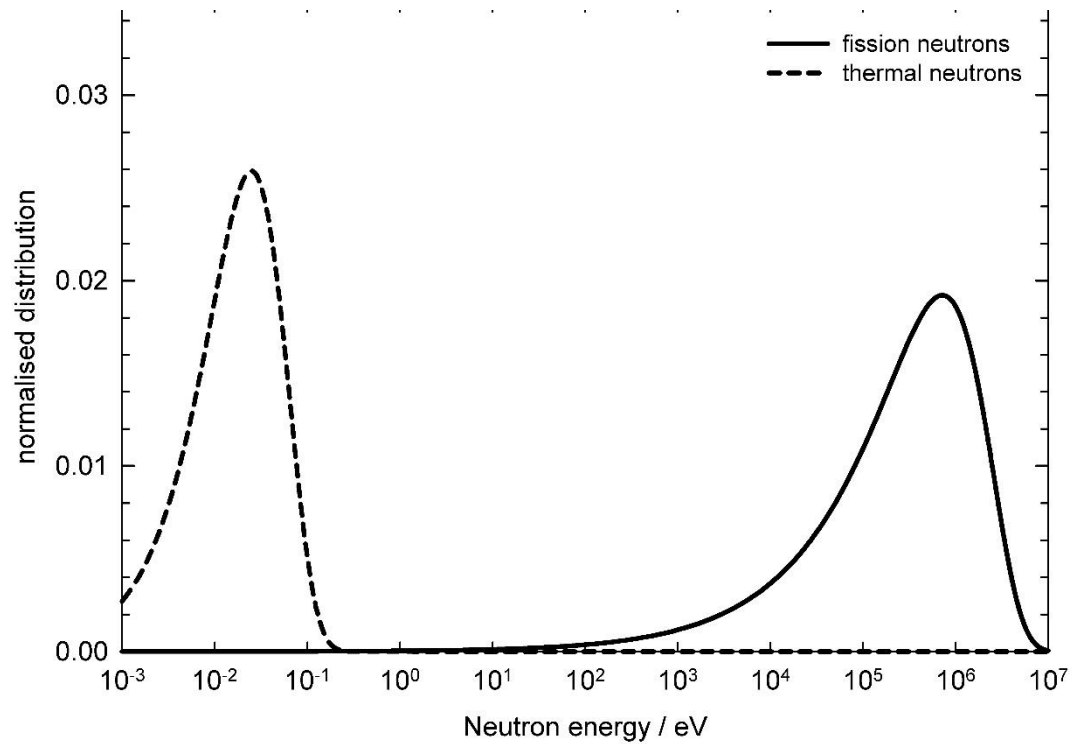
PWR



Generation

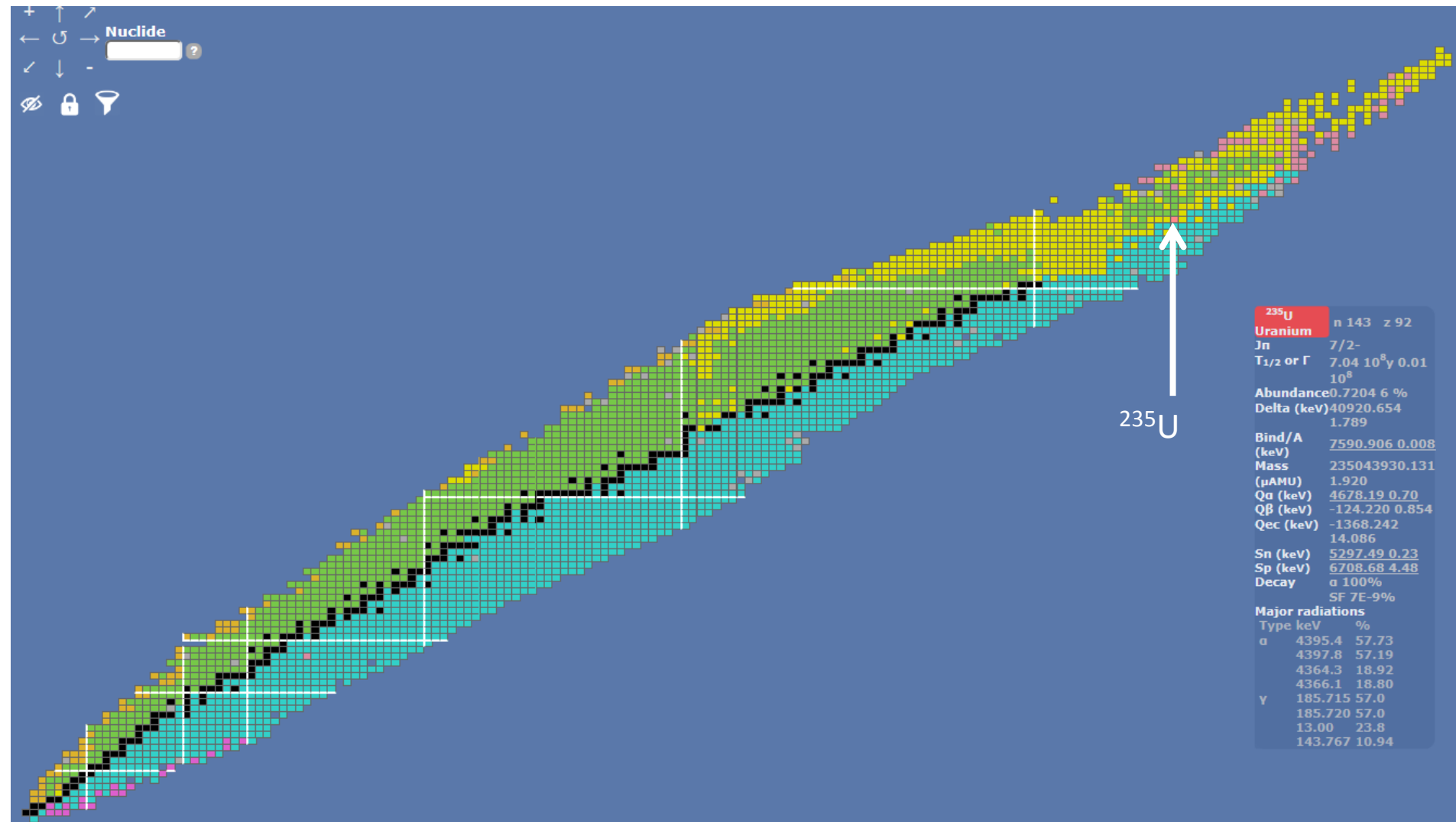


Generation



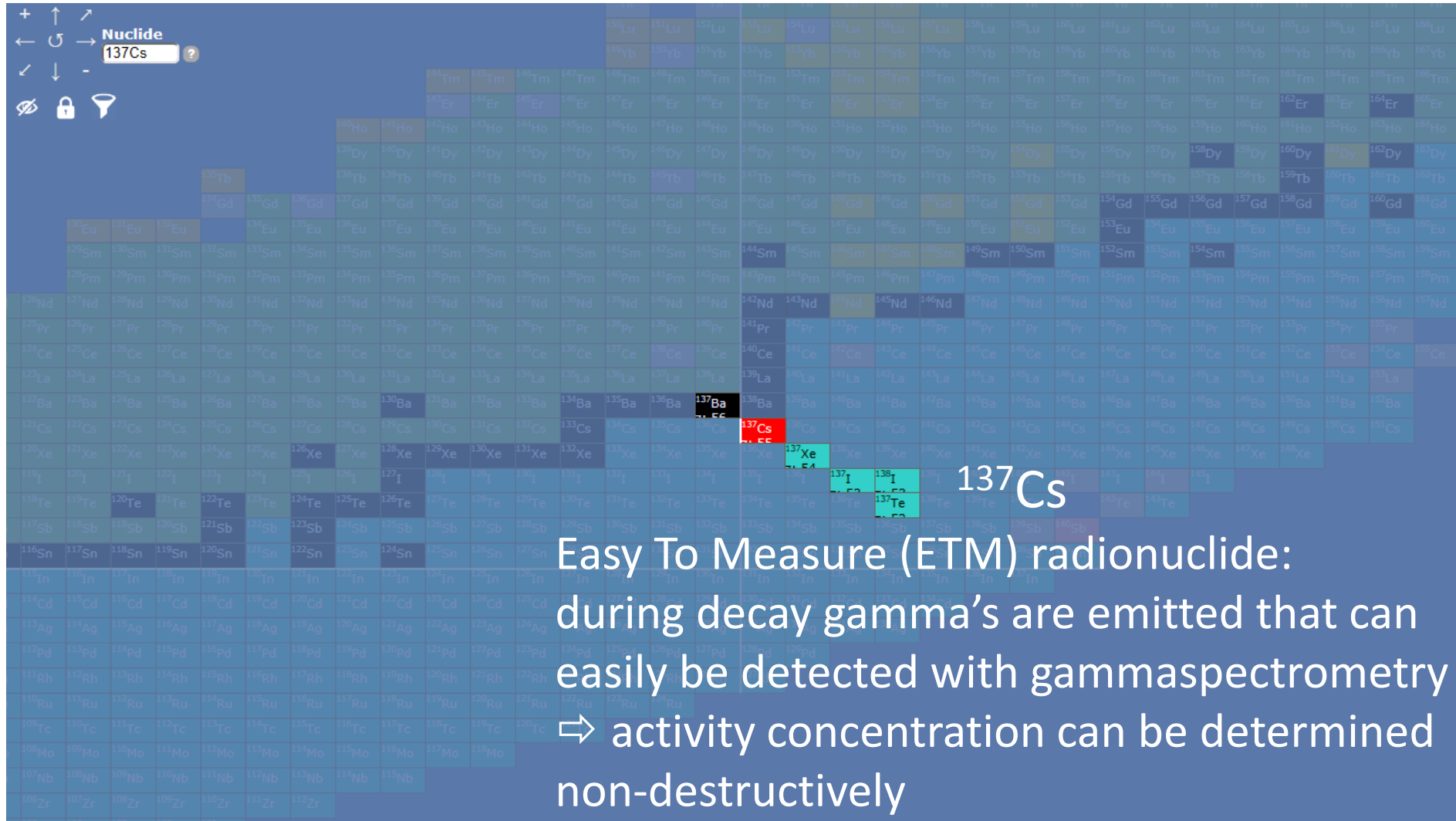


Generation

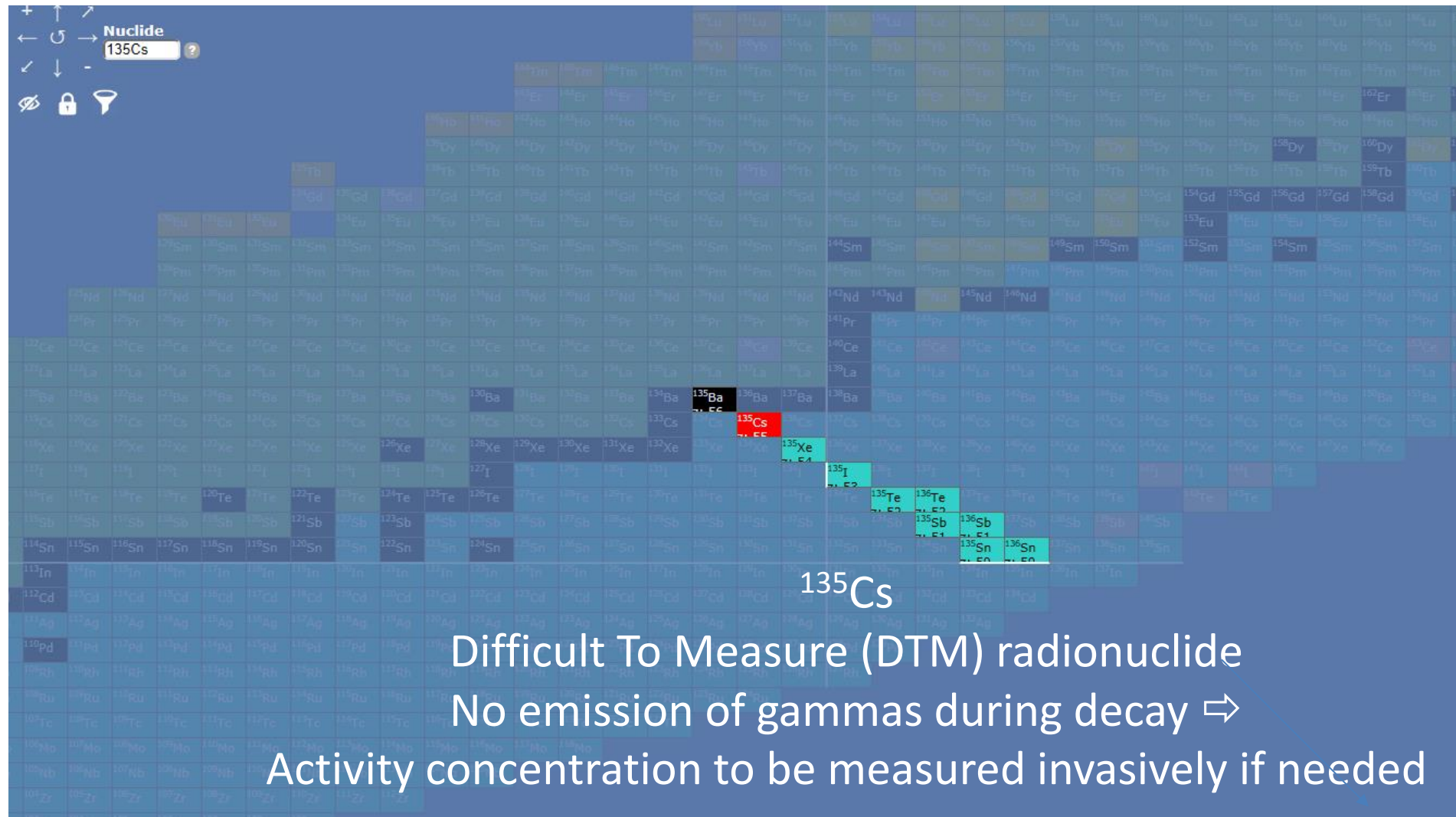


Live chart from IAEA, free online, also mobile phone **IAEA Isotope Browser**

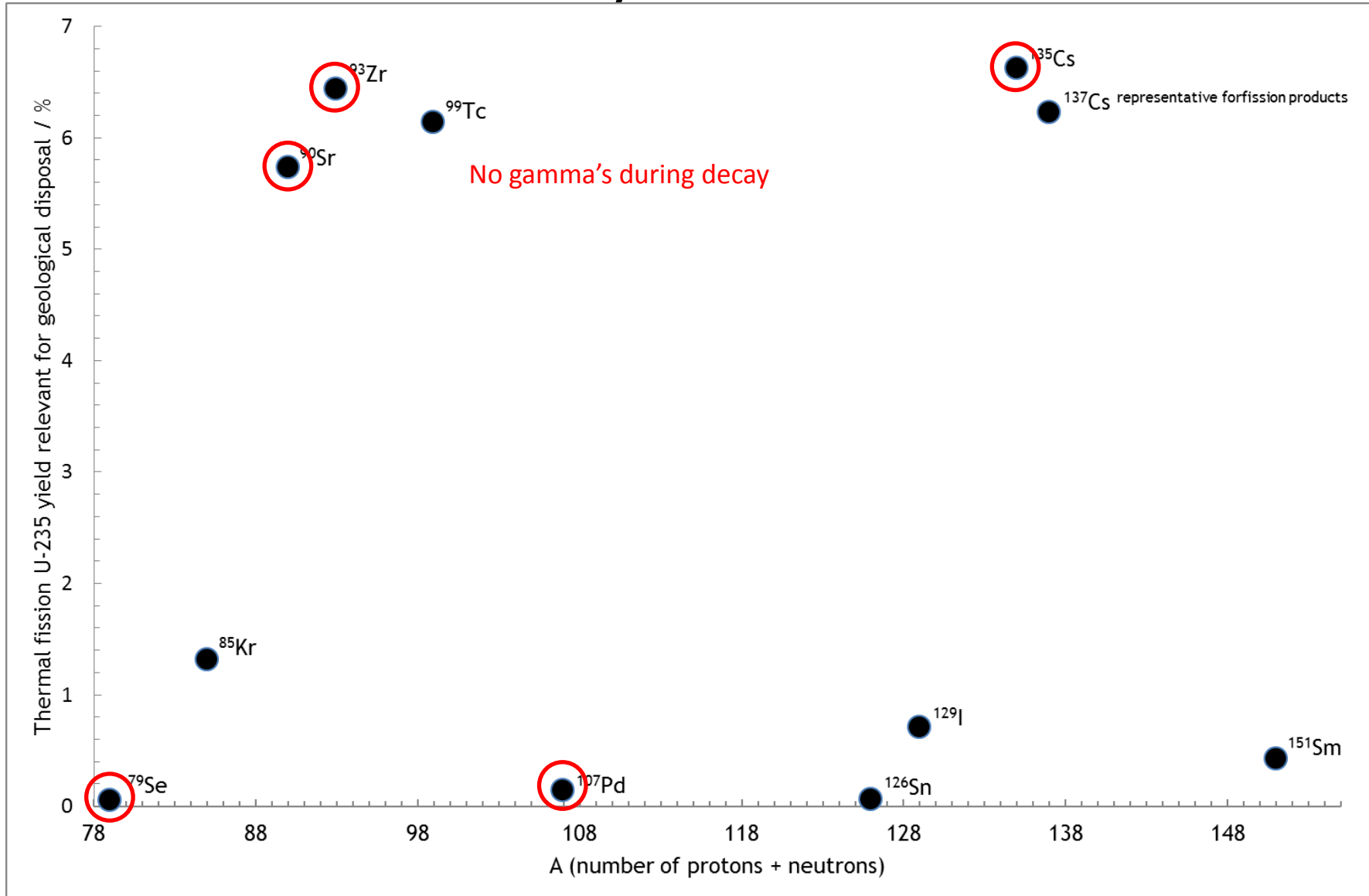
Example RN left for disposal from decay and fission



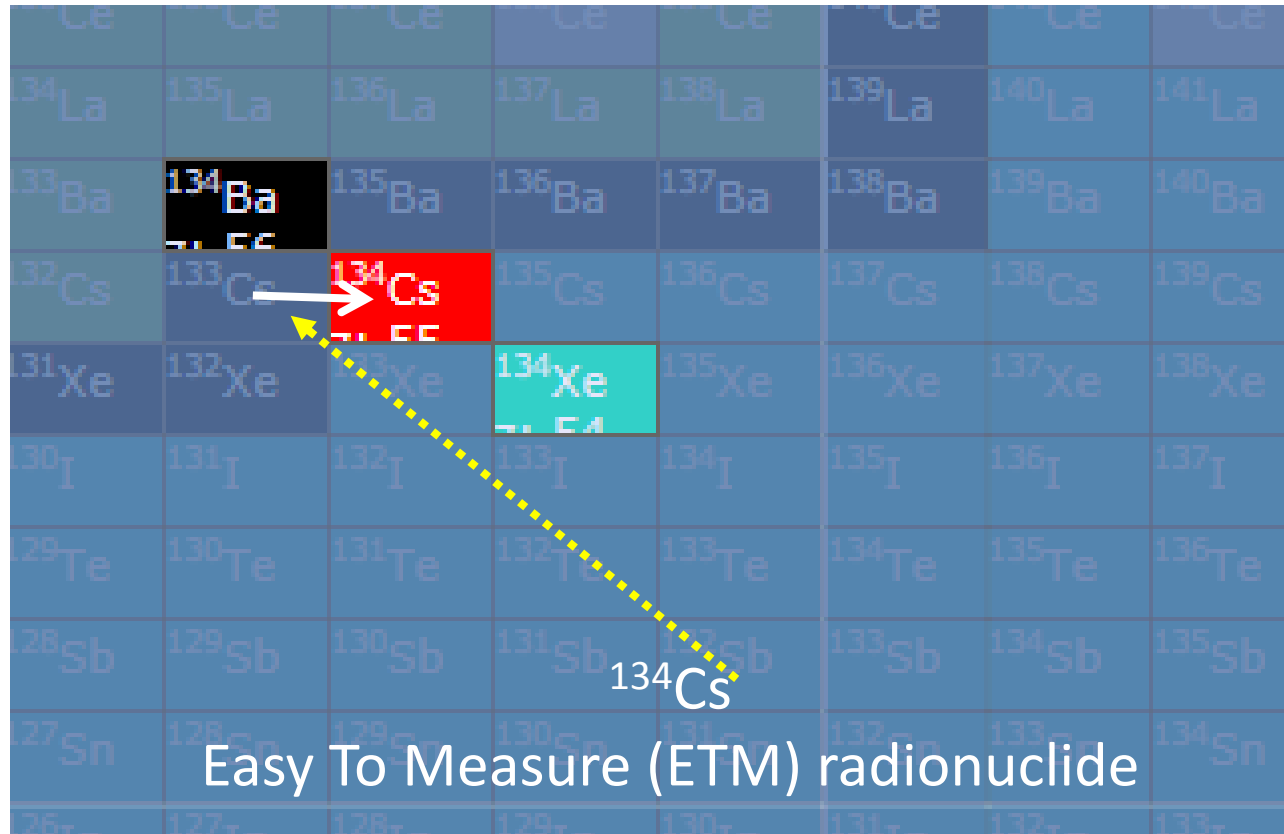
Example RN left for disposal from decay and fission



Examples RN left for disposal from decay and fission

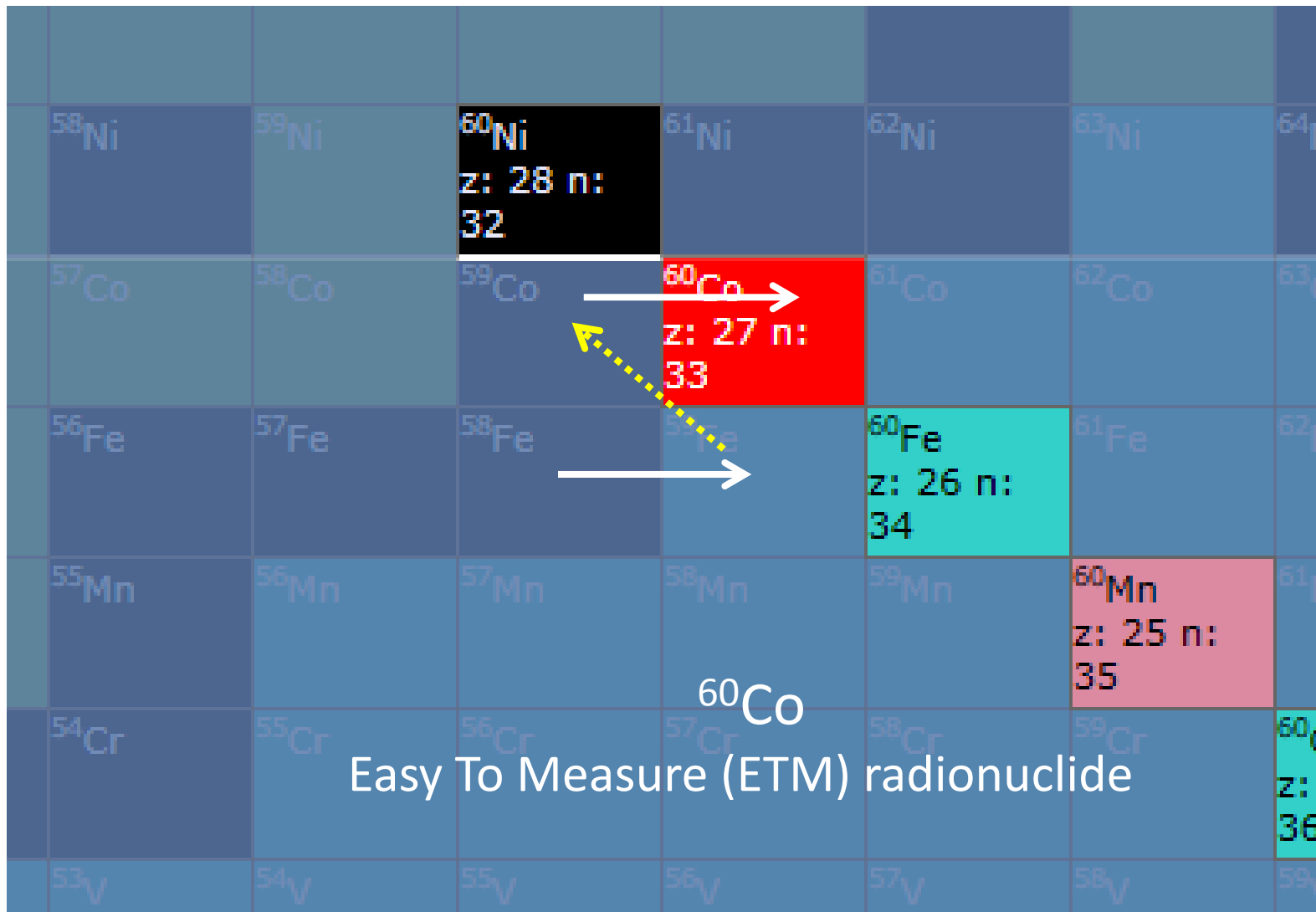


Example RN left from fission, decay and activation





Example RN left from activation for storage, processing, surface disposal





COVRA's storage period at least 100 years:
Fraction in activity left: $\left\{\frac{1}{2}\right\}^{100/t_{0,5}}$ for $^{60}\text{Co}=0,0000019$
i.e. reduction of a million



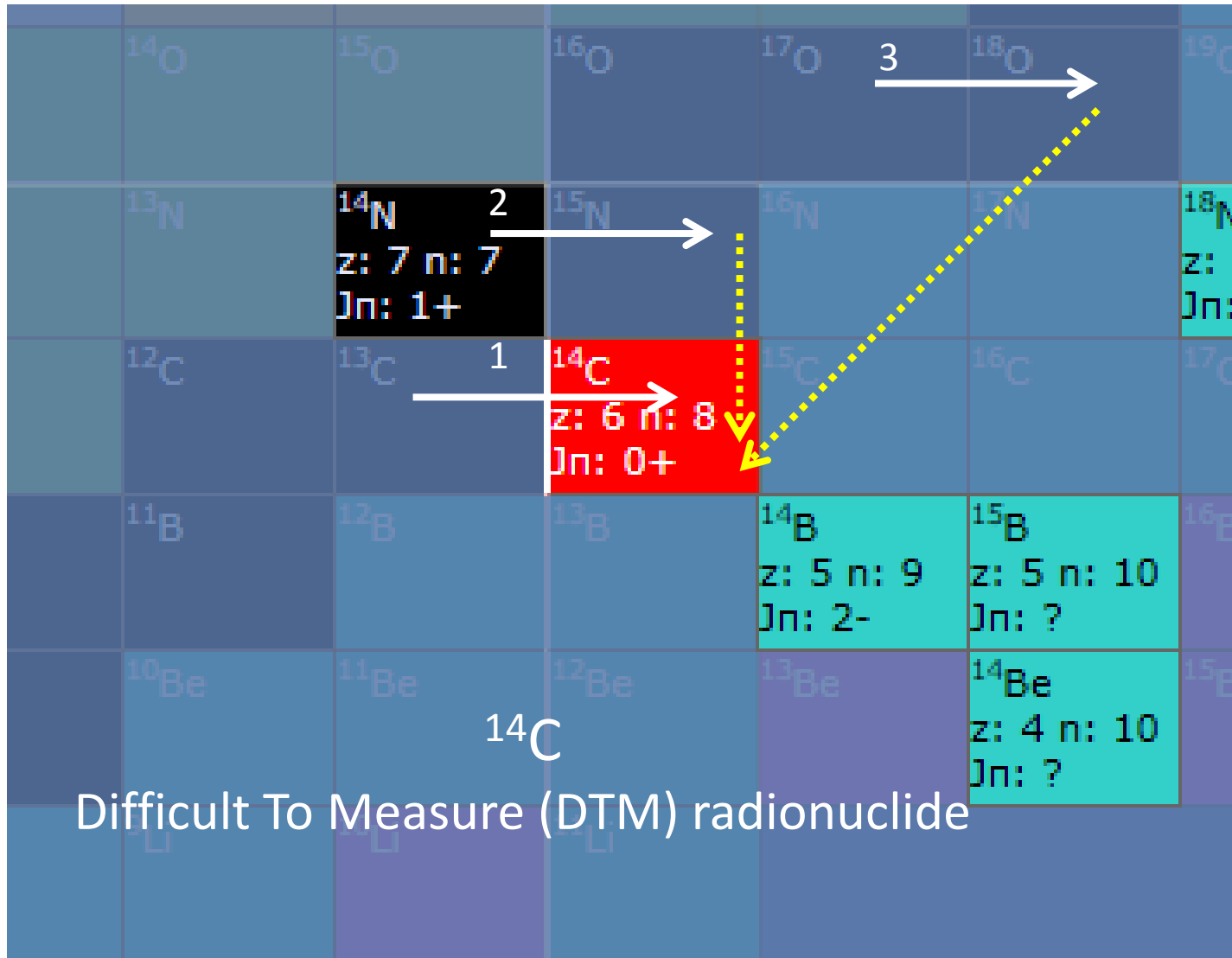
Neutron activation



- Identification activation path to obtain the precursors
 - Size of (thermal) neutron reaction cross section
- Knowledge of the chemical content of precursors
 - Can be impurities



Example RN left for disposal from activation





Is RN-conc. relevant for disposal?



- Clearance levels in EU:
 - Council Directive 2013/59/EURATOM of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, and repealing Directives...
 - ^{14}C : 1 Bq per gram solid matter for example 0.000024 ppm in iron

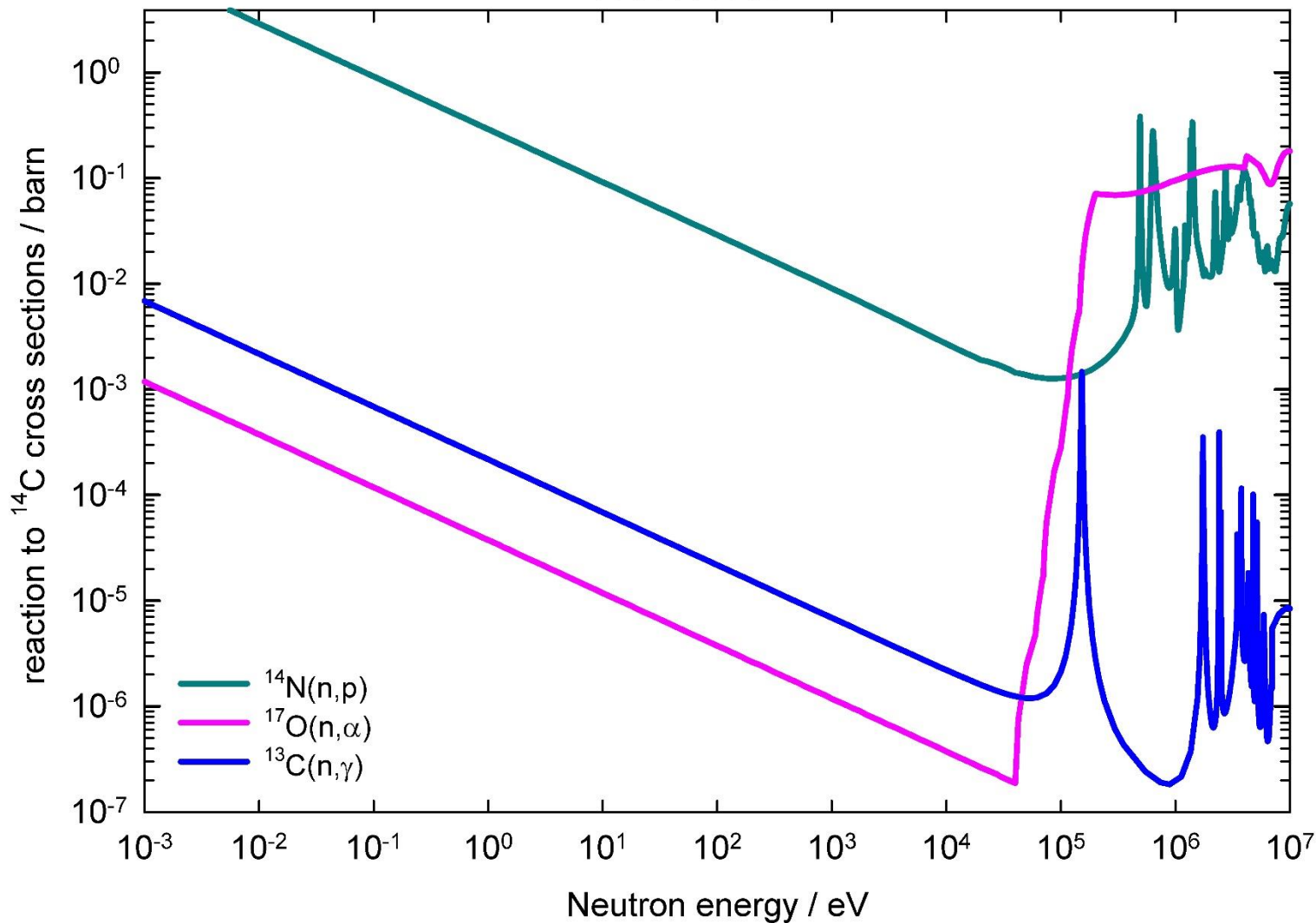


COVRA's storage period at least 100 years:
Fraction in activity left: $\left\{\frac{1}{2}\right\}^{100/t_{0,5}}$ for $^{14}\text{C}=0,99$
i.e. reduction after this storage period is negligible



Neutron activation

at about 300 K





Neutron activation



- Natural abundances
 - Nitrogen-14 : 99.636%
 - Oxygen-17 : 0.038%
 - Carbon-13 : 1.07%
- Natural abundance + thermal cross sections for the same carbon-14 contribution:
 - Chemical content carbon $\gg 10^5$ chemical nitrogen content
 - Chemical content oxygen $\gg 10^7$ chemical nitrogen content

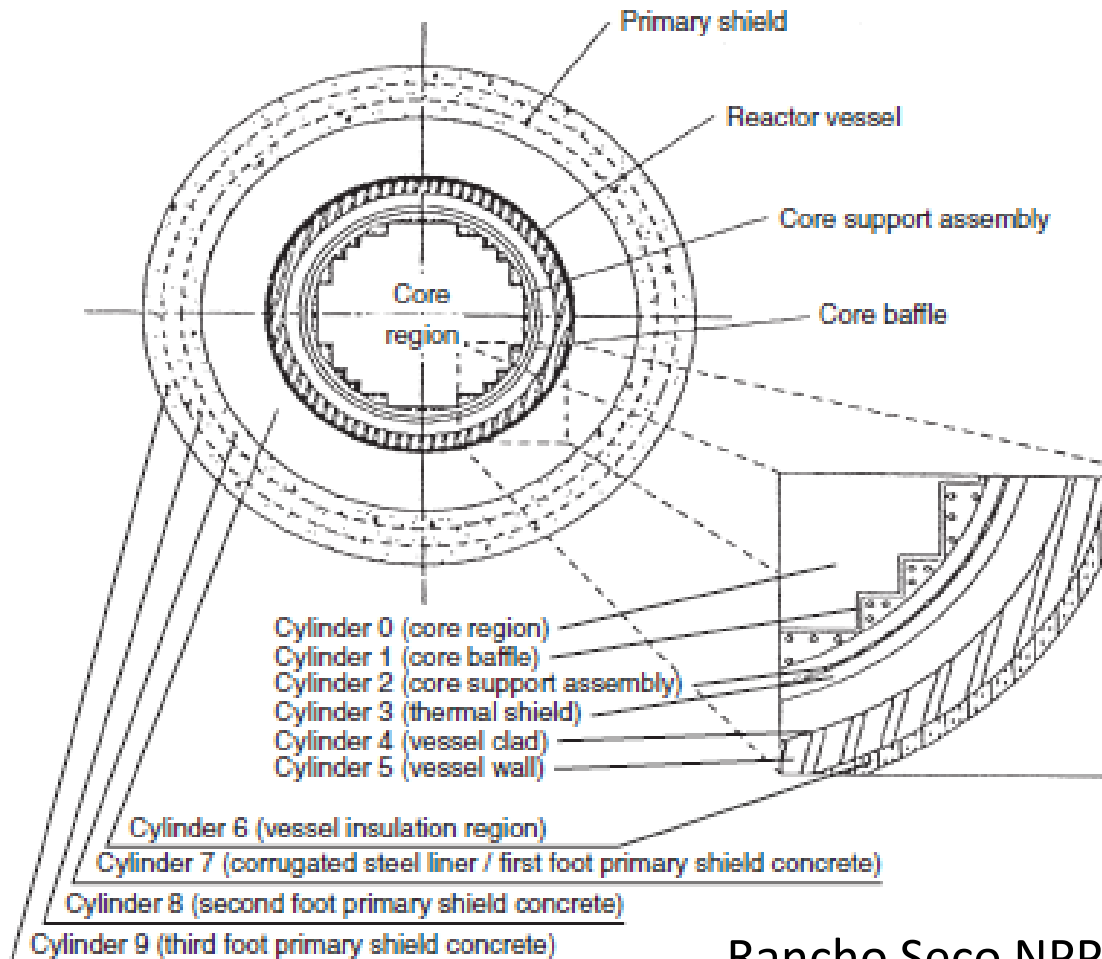
Carbon-14 inventory (scoping)

- $T_{1/2} = 5730$ years, no decay during neutron irradiation period in nuclear power plant
- Activated core negligible cross section
- Nitrogen content due to natural abundance ^{14}N 99.64% and high thermal cross section

$$C_{14C} = C_{\text{Nitrogen}} \sigma_{14\text{N,thermal}} \phi_{\text{thermal}} \Delta t_{\text{irradiation}}$$

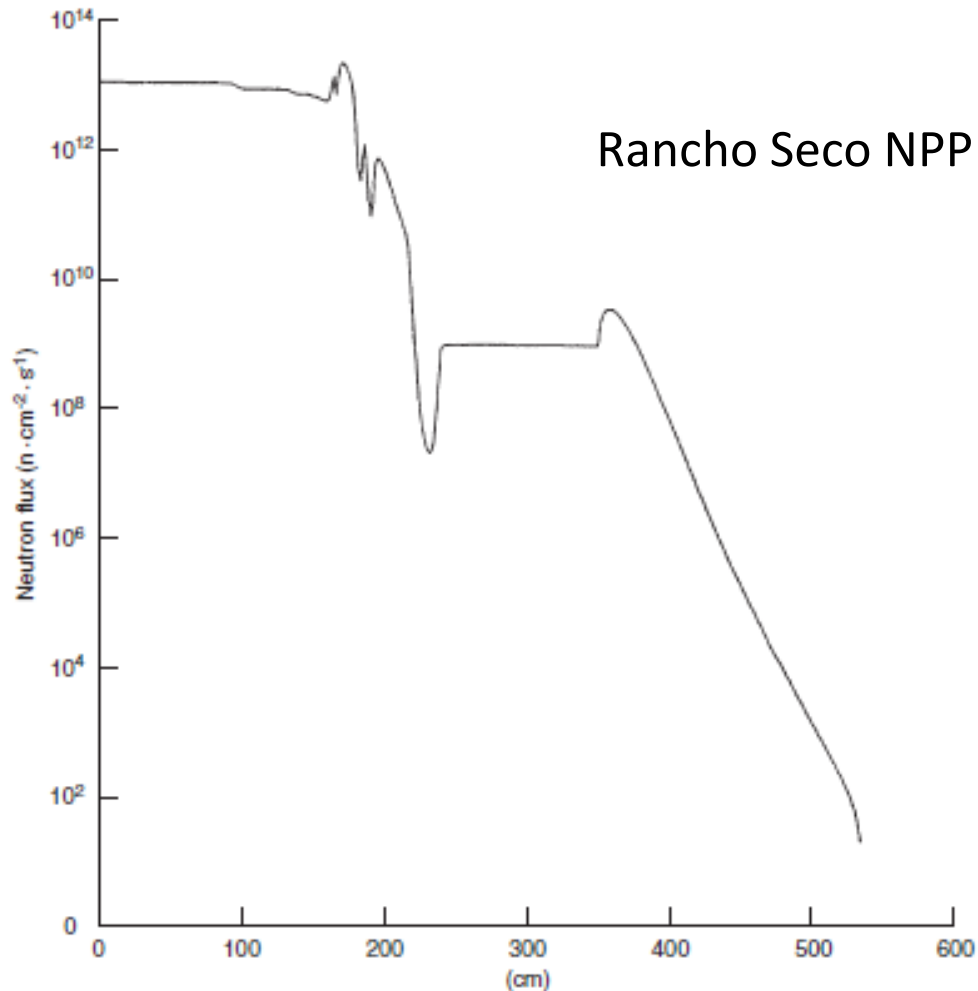
$$\text{Act.}C_{14C} = \lambda_{14C} C_{\text{Nitrogen}} \sigma_{14\text{N,thermal}} \phi_{\text{thermal}} \Delta t_{\text{irradiation}}$$

Neutron flux



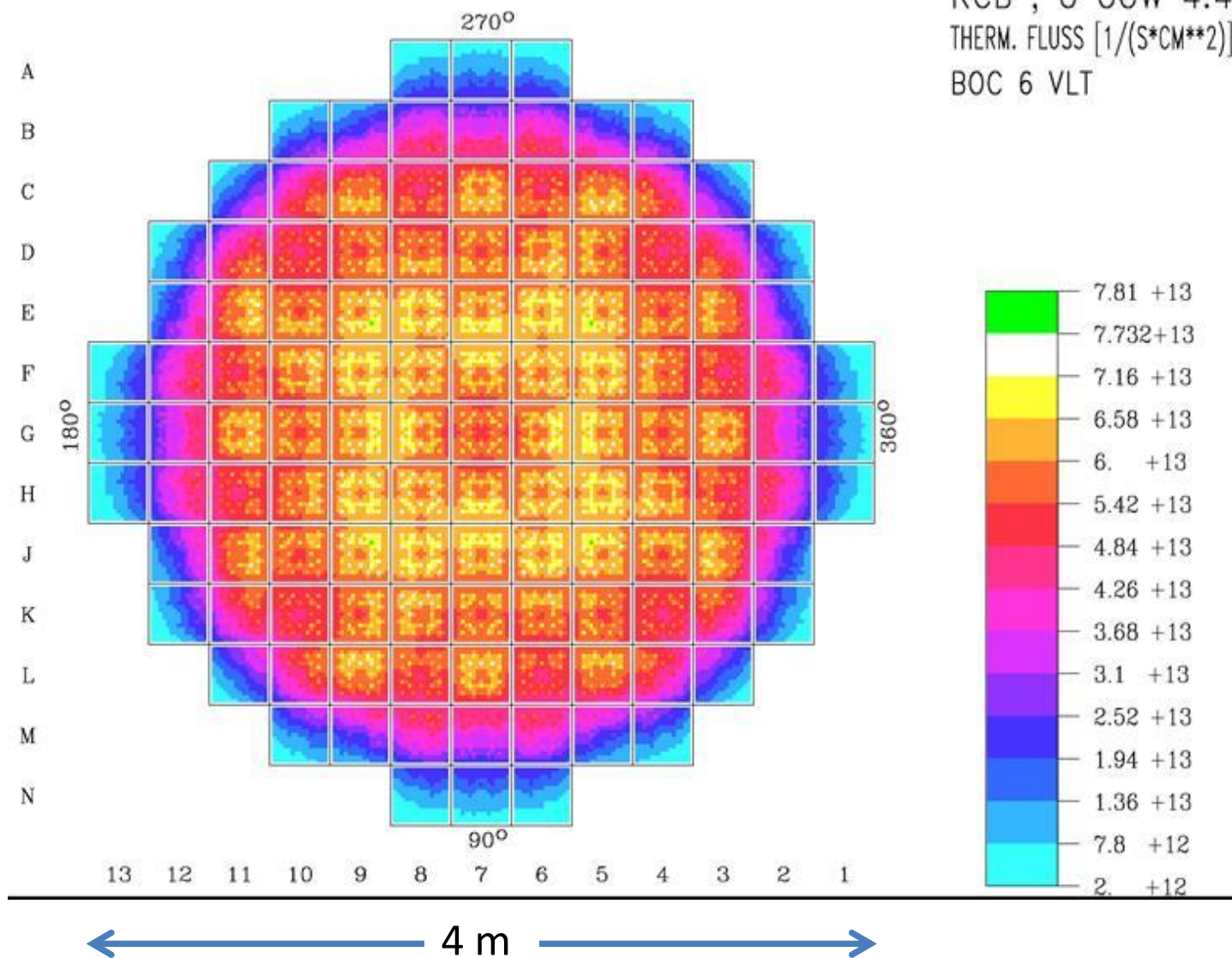
Rancho Seco NPP (PWR) 913 MW(e)

Thermal neutron flux

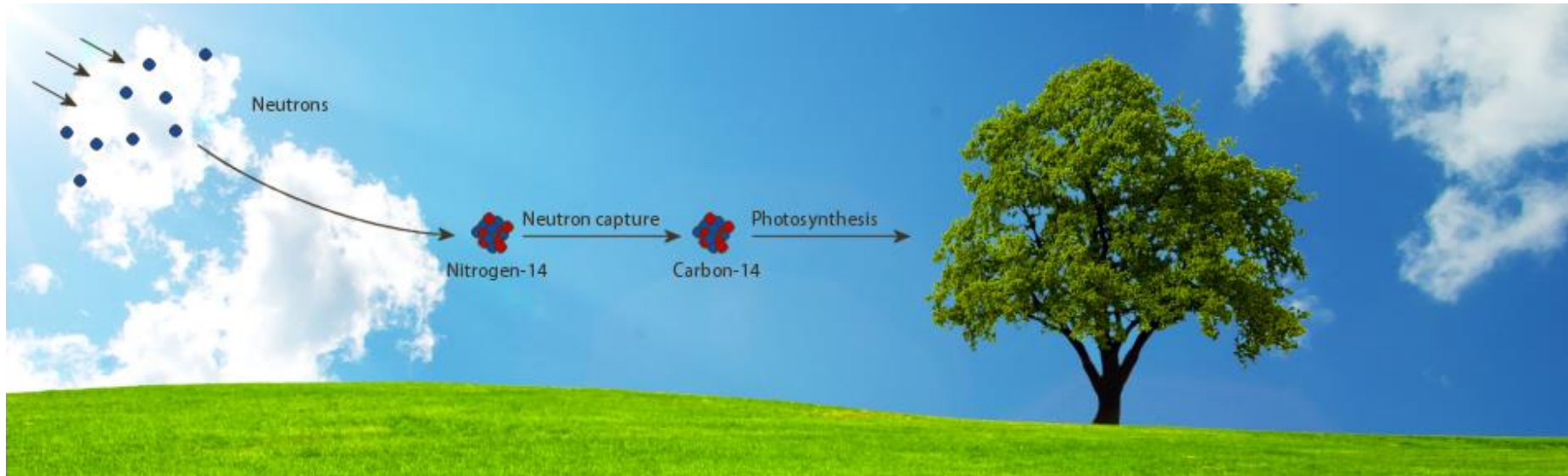


Thermal neutron flux

KCB , U GGW 4.4
THERM. FLUSS [1/(S*CM**2)]
BOC 6 VLT



Natural carbon-14



Generation of a.o. neutrons

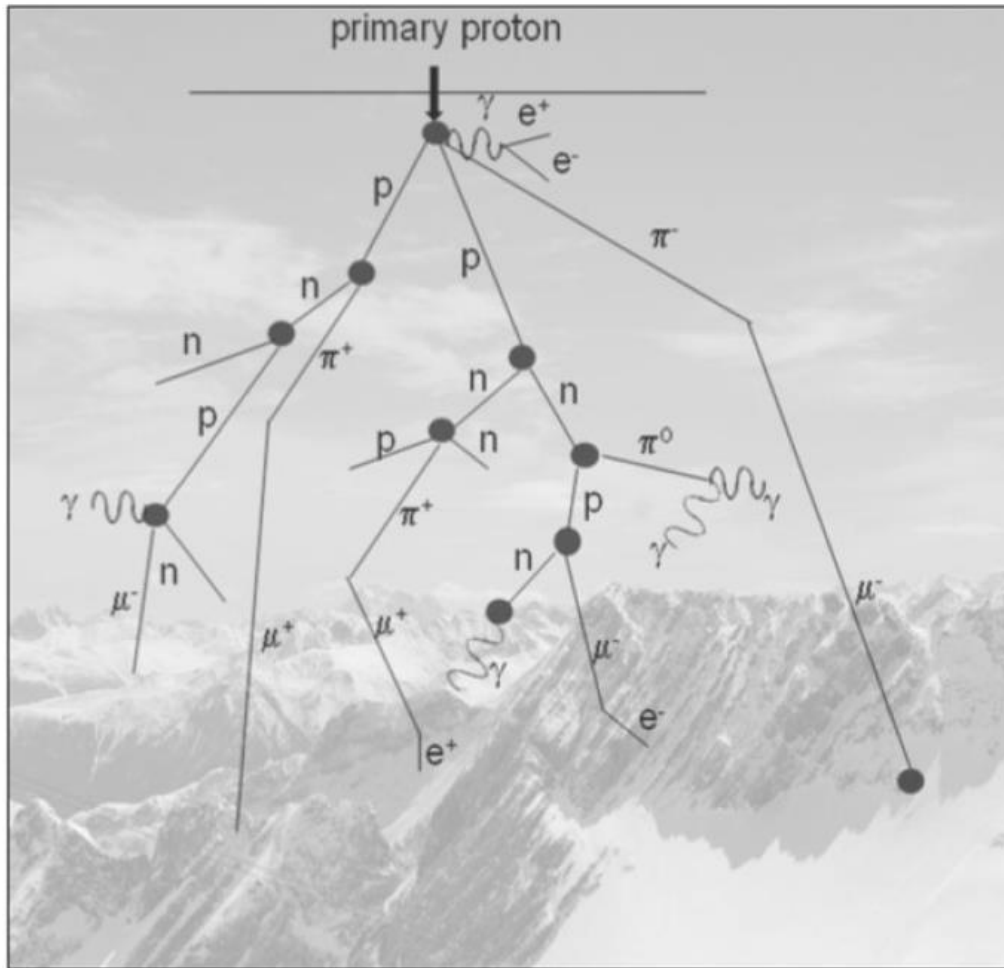
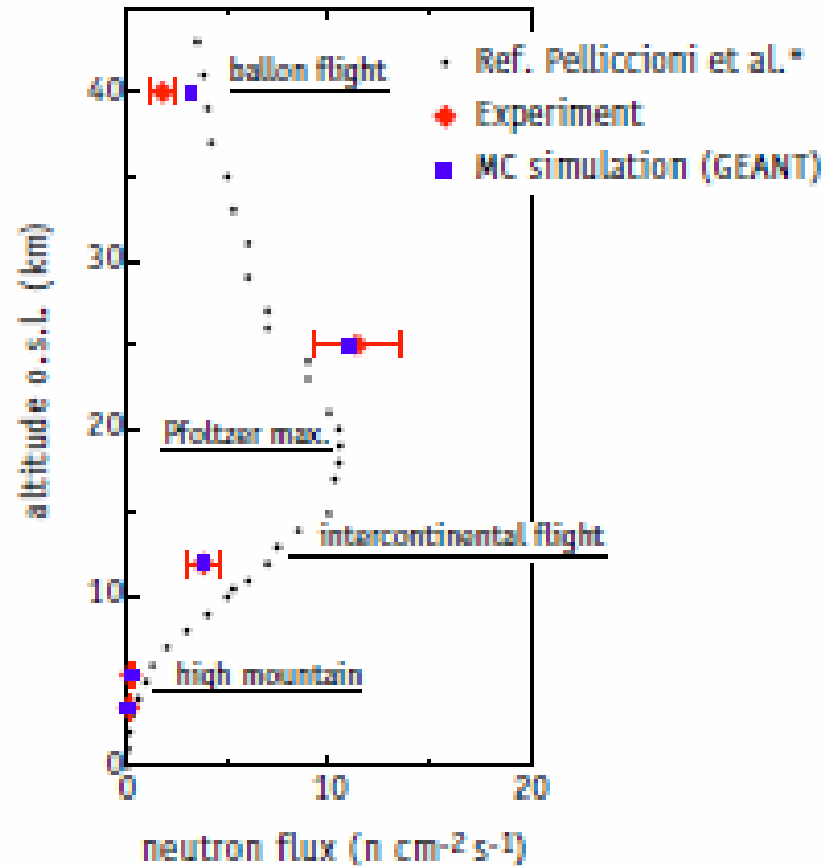


Image: ICRP 132 (2016)

Fig. 2.1. Sketch of cascade of secondary cosmic radiation: μ , muon; e^- , electron; e^+ , positron; γ , gamma rays; n , neutron; p , proton; π , pion (picture from W. Rühm).

Environmental neutron flux



*min. solar activity, max. latitude

Thermal neutron flux

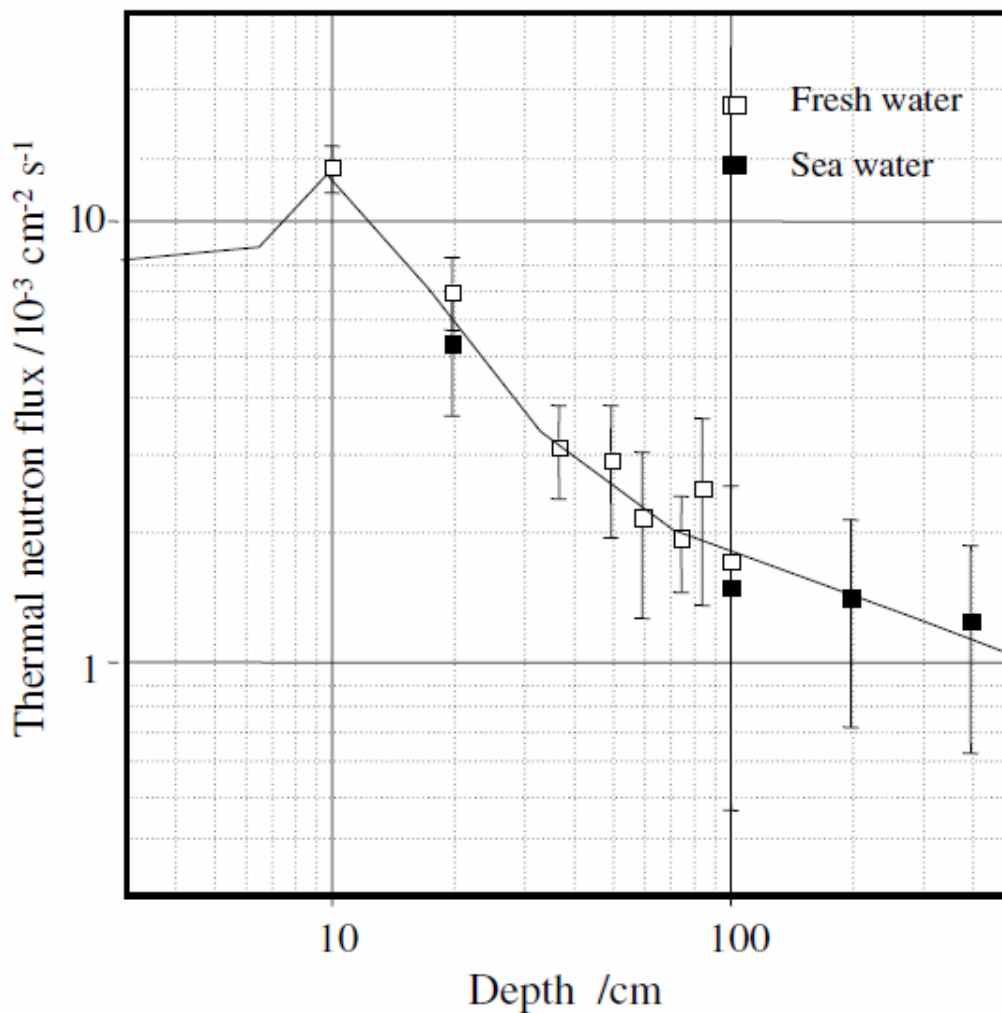


Figure 1. Depth profile of thermal neutron flux in the freshwater and seawater.



Natural versus artificial generation of carbon-14



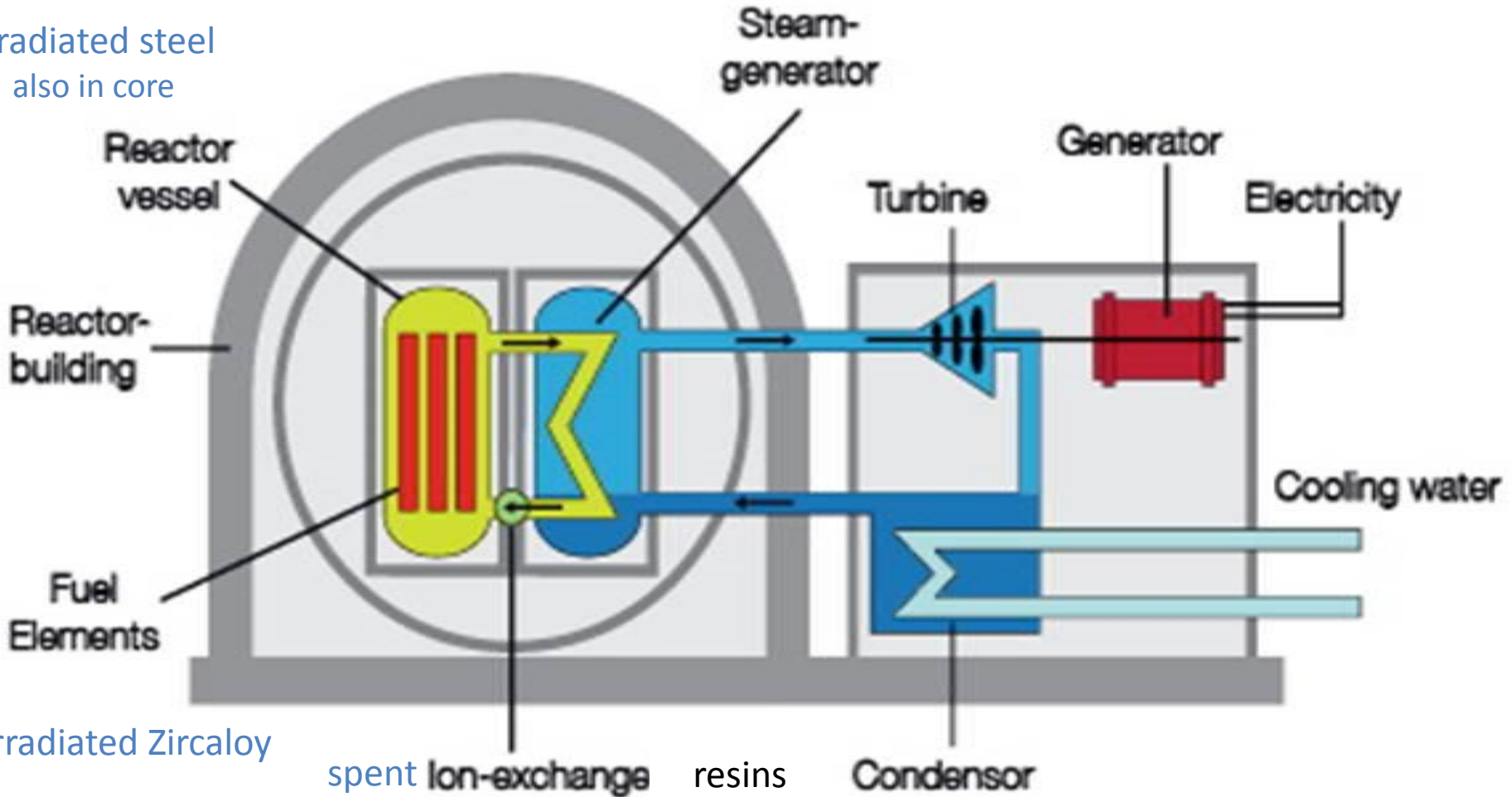
- Same generation process but different parameter values
- Nitrogen content
 - Nitrogen in air 80%
 - Nitrogen in reactor materials impurity level
- Thermal neutron flux
 - Artificial thermal neutron flux $10^{14} \text{ cm}^{-2}\text{s}^{-1}$ in NPP
 - Environmental thermal neutron flux at ground level at Earth's surface due to shielding (i.e. deflection magnetic field and collisions with atomic particles in our atmosphere) about $10^{-3} \text{ n cm}^{-2} \text{ s}^{-1}$ [Komura et al, 2008]

Difference in thermal neutron flux: carbon-14 containing radioactive waste although nitrogen content present in reactor materials as impurities

Types of waste investigated



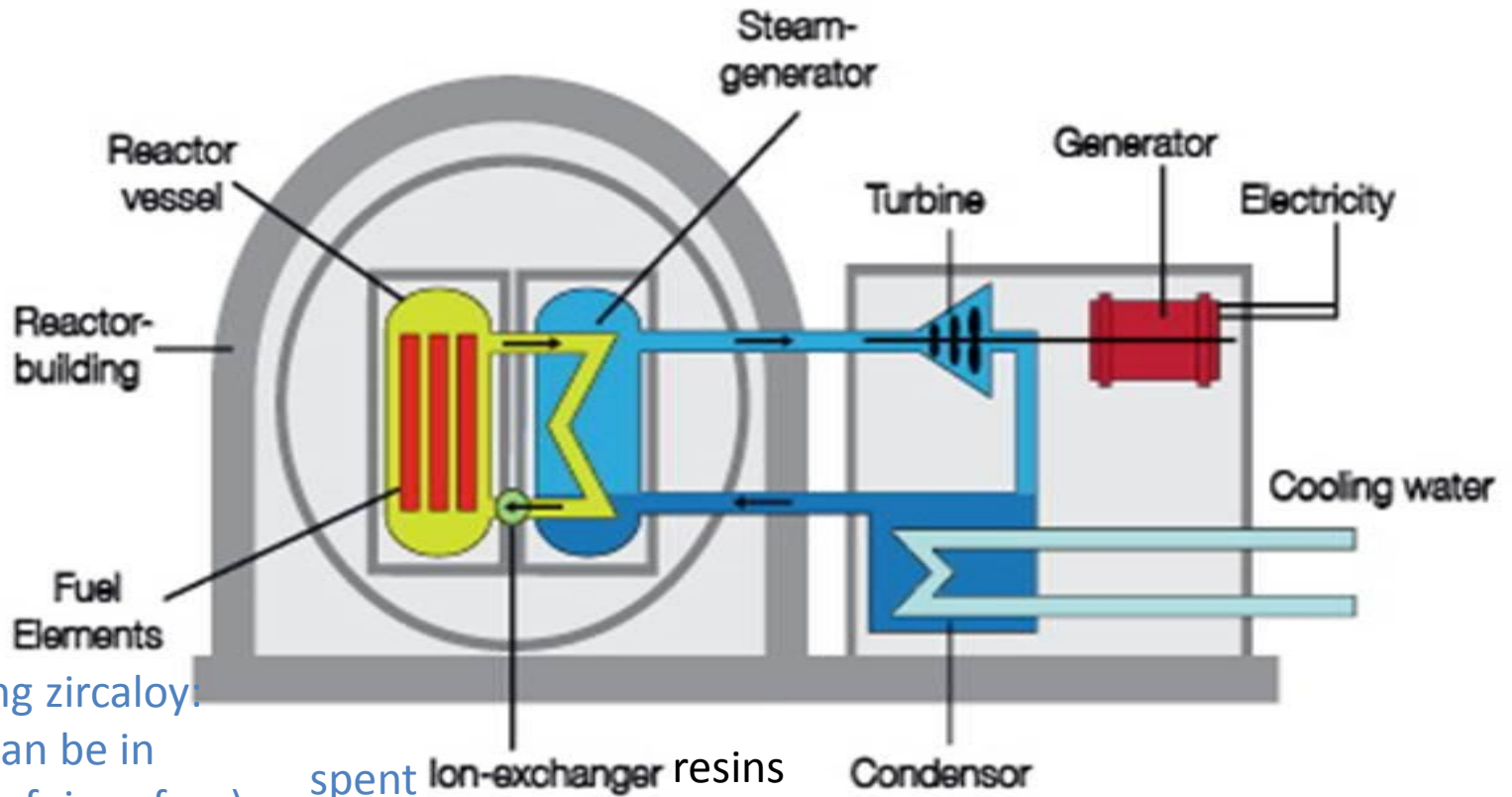
Irradiated steel
also in core



Origin nitrogen

Manufacturing steel: nitrogen can be in
pig iron,
Cokes
Stirring gas

Nitrogen content frequently not reported



Manufacturing zircaloy:
nitrogen can be in
Sponge ingot (Hafnium-free),
melt



Carbon-14 act.conc.

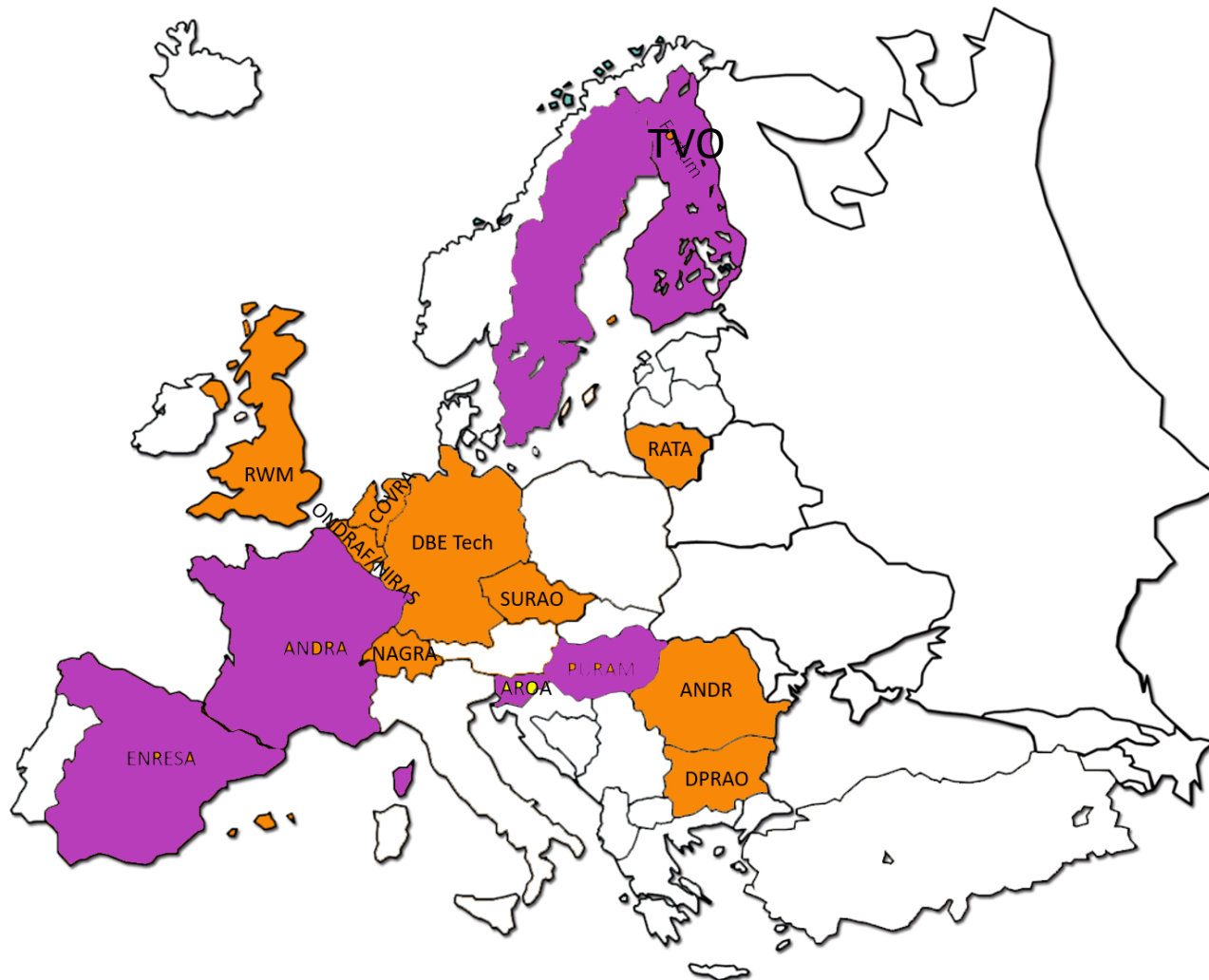


- Knowledge nitrogen impurities for many types of waste
 - EU study (1984) limit nitrogen impurities in steel, zircaloy and graphite when used in NPP
 - IAEA (2004) example limit nitrogen in neutron activation part of NPP
 - limit air ingress primary coolant
 - pH control primary coolant LiOH instead of hydrazine $\text{NH}_2\text{-NH}_2$
- Neutron thermal flux and irradiation period

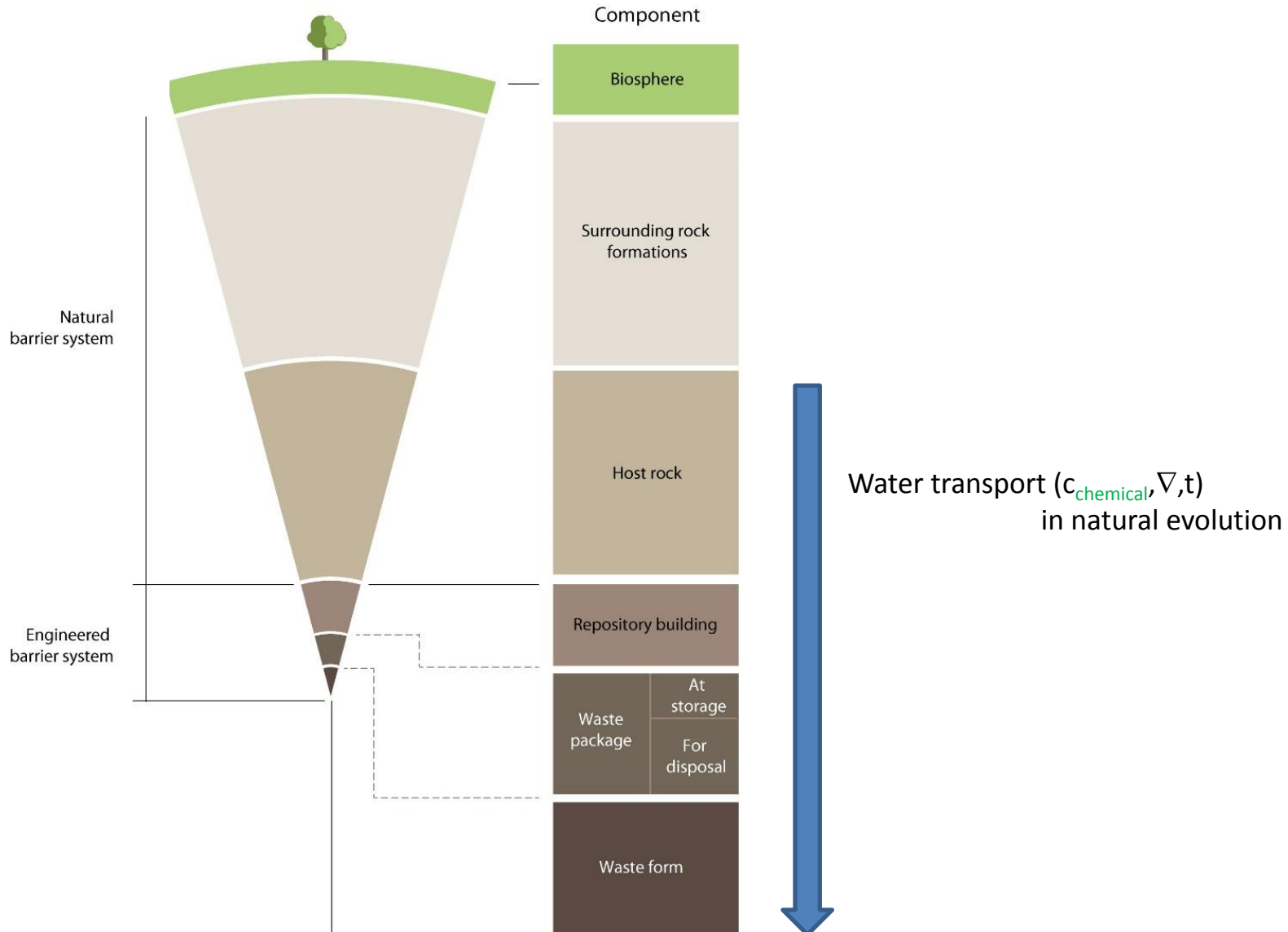
RP Bush, GM Smith, IF White, Carbon-14 waste management, EUR 8749 (1984).

IAEA, Management of waste containing tritium and carbon-14, Technical Reports Series No. 421 (2004)

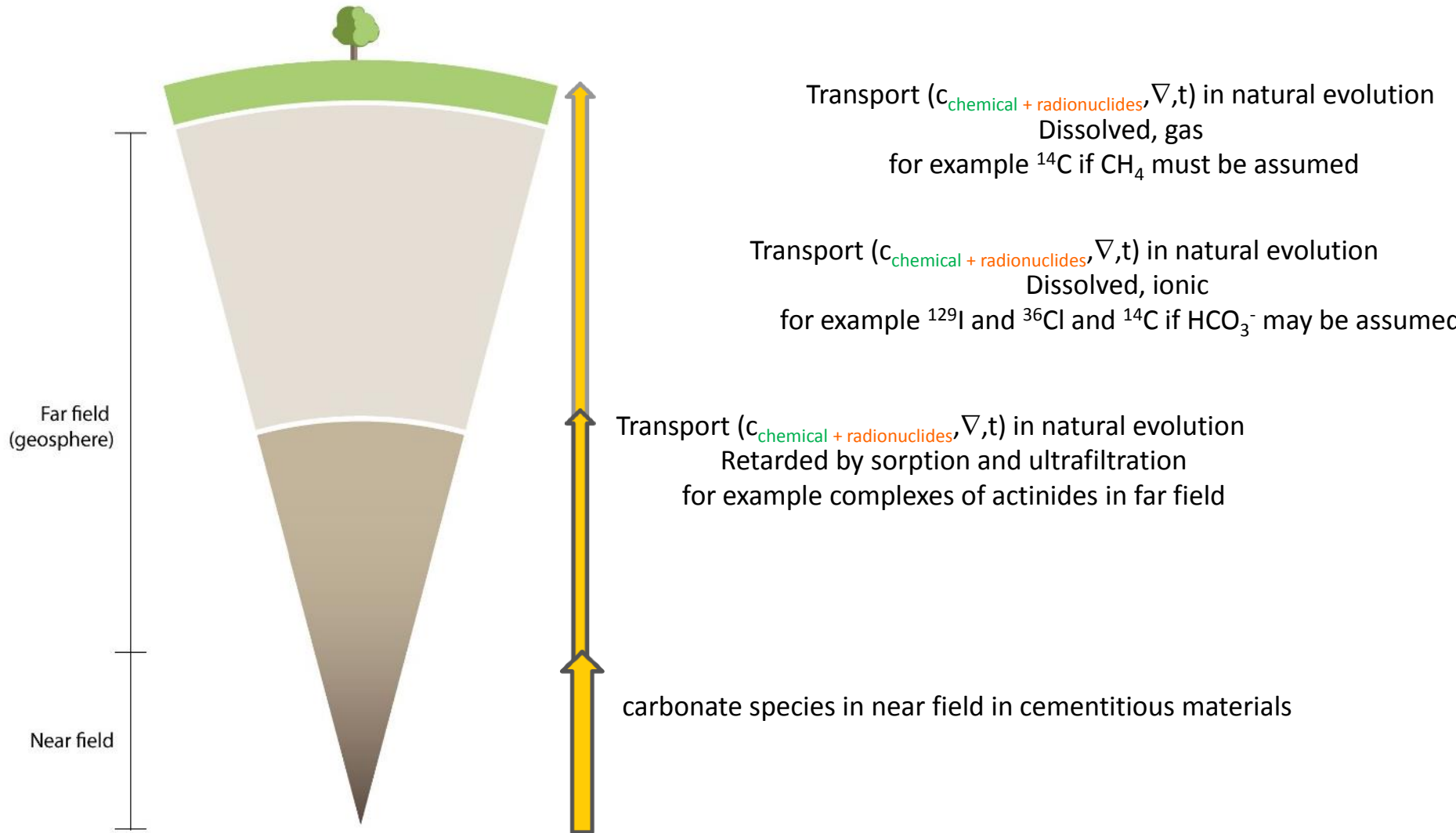
Designated end point **i.e. surface disposal**, of some waste investigated in CAST already implemented



Geological disposal of waste



Geological disposal of waste





Gas, dissolved

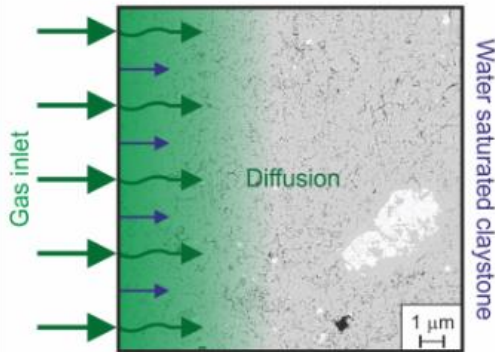


- Waste types investigated in CAST
 - Neutron irradiated metallic compounds
 - Degradation: anaerobic corrosion
 - Hydrogen generation rate
 - Non-metallic neutron irradiated compounds

Free gas, dissolved gas

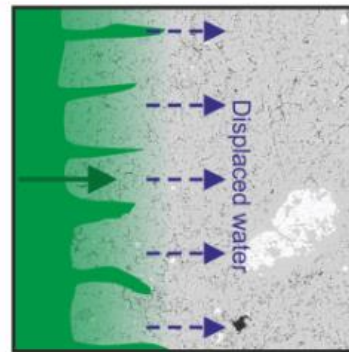
Identified in concrete?

yes



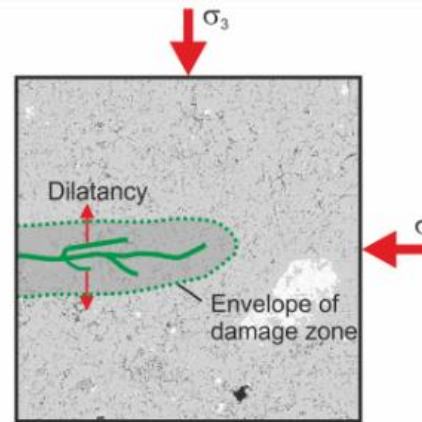
Advection and diffusion of dissolved gas

yes

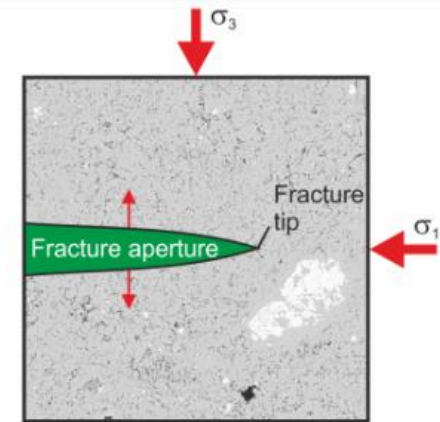


Visco-capillary flow of gas and water phase ("two-phase flow")

yes

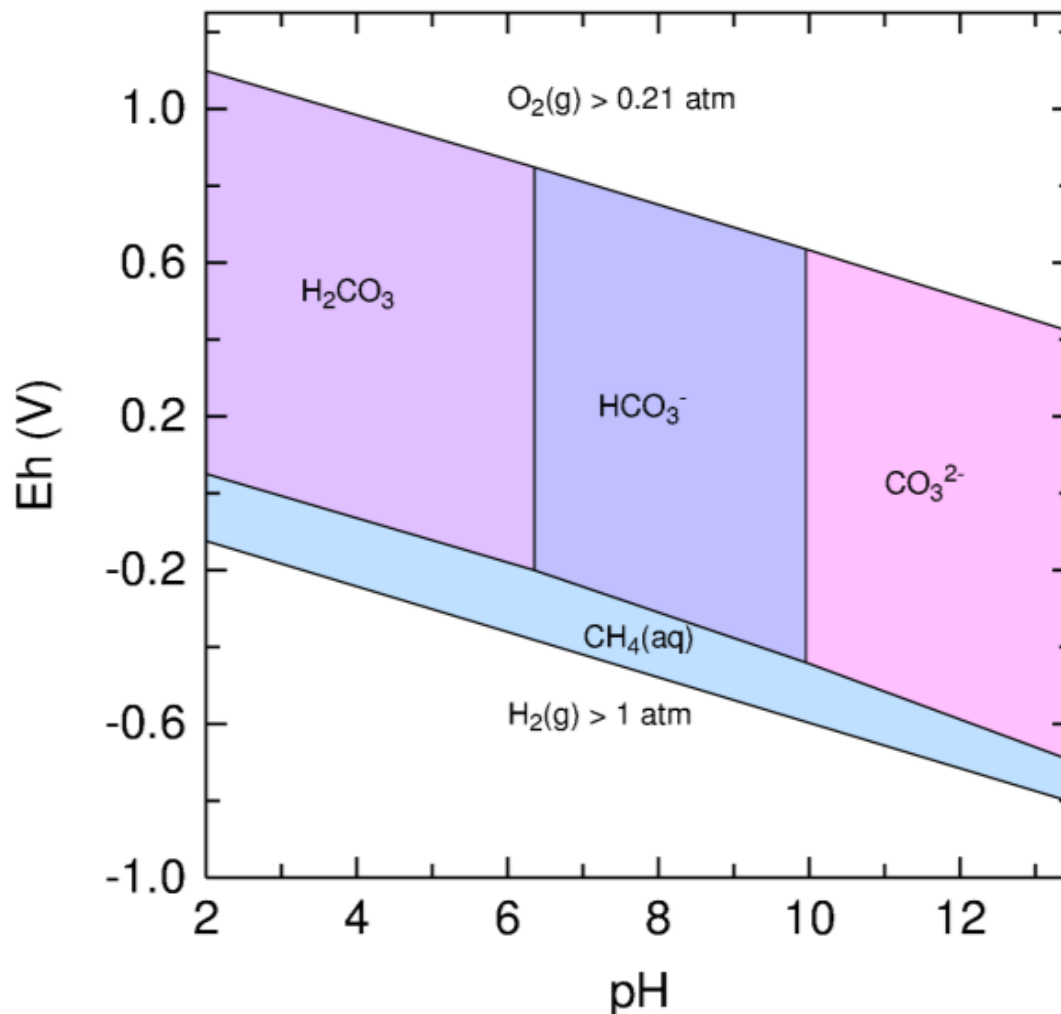


Dilatancy controlled gas flow ("pathway dilation")



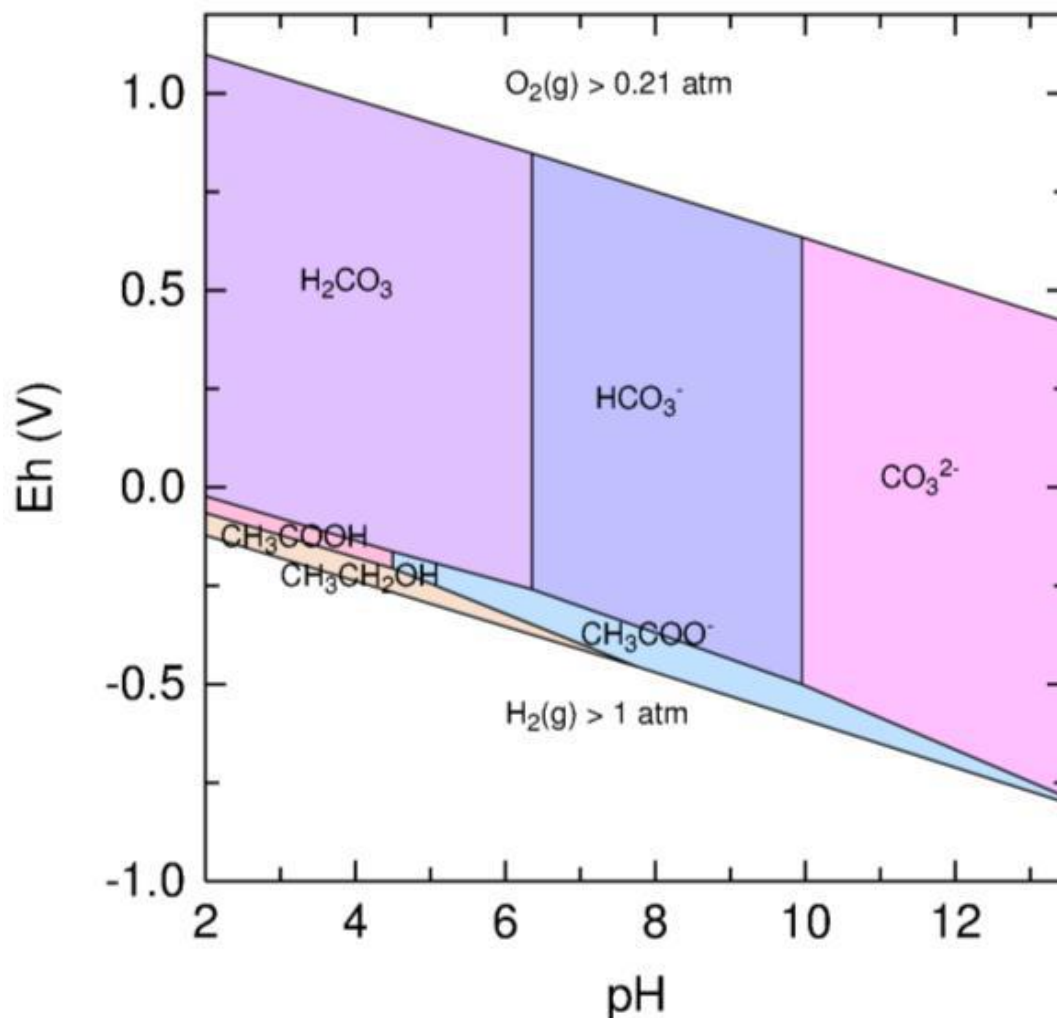
Gas transport in tensile fractures ("hydro-/gasfrac")

Gas, dissolved



Wieland E, Hummel W, Formation and stability of ^{14}C -containing organic compounds in alkaline iron-water systems: preliminary assessment based on a literature survey and thermodynamic modelling, Mineralogical Magazine Vol 79(2015) & Rizzato C, Rizzo A, Heisbourg G, Večerník P, Bucur C, Comte J, Lebeau D, Reiller PE, State of the art review on sample choice, analytical techniques and current knowledge of release from spent ion-exchange resins CAST report 4.1 (2015)

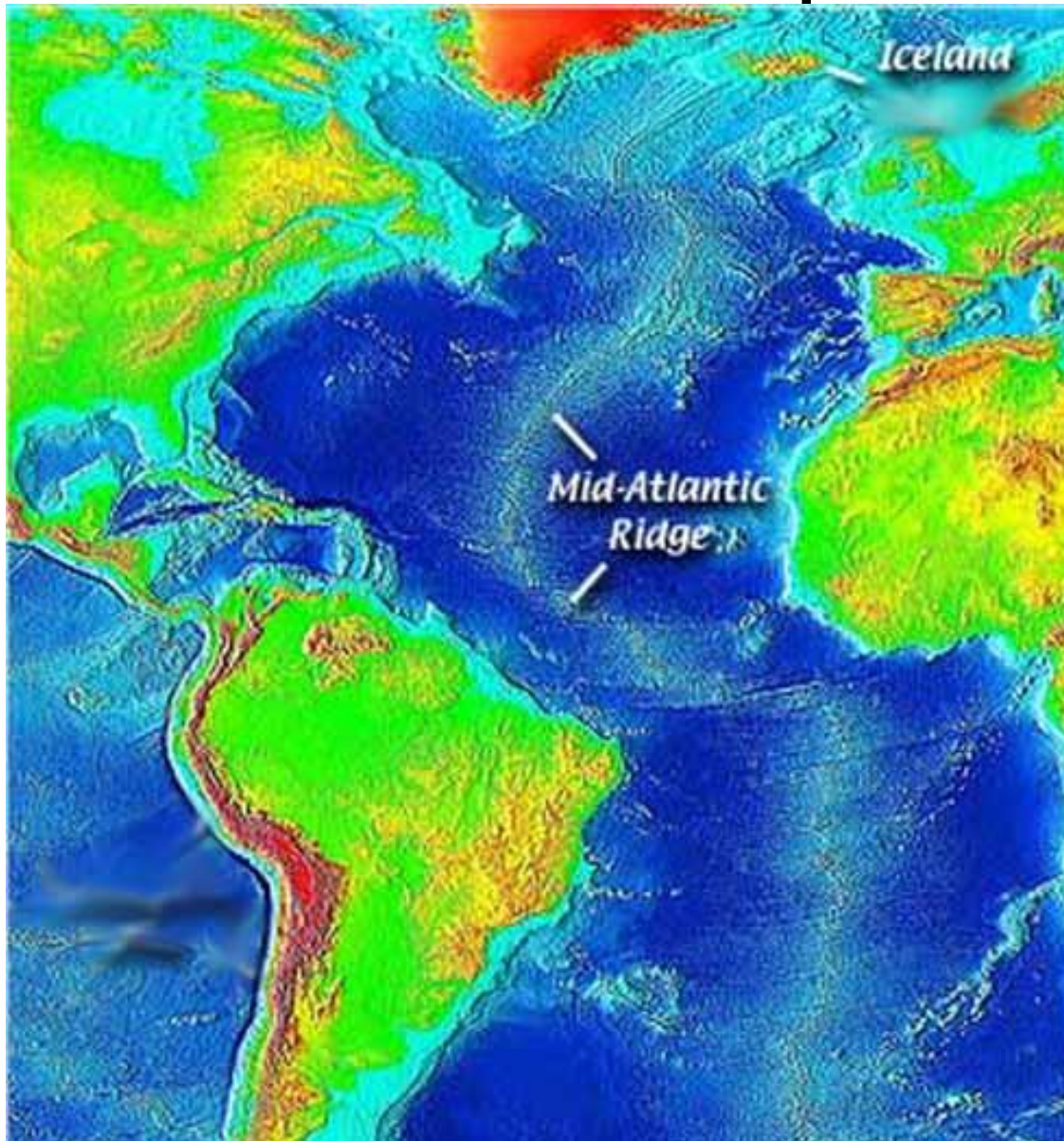
Dissolved



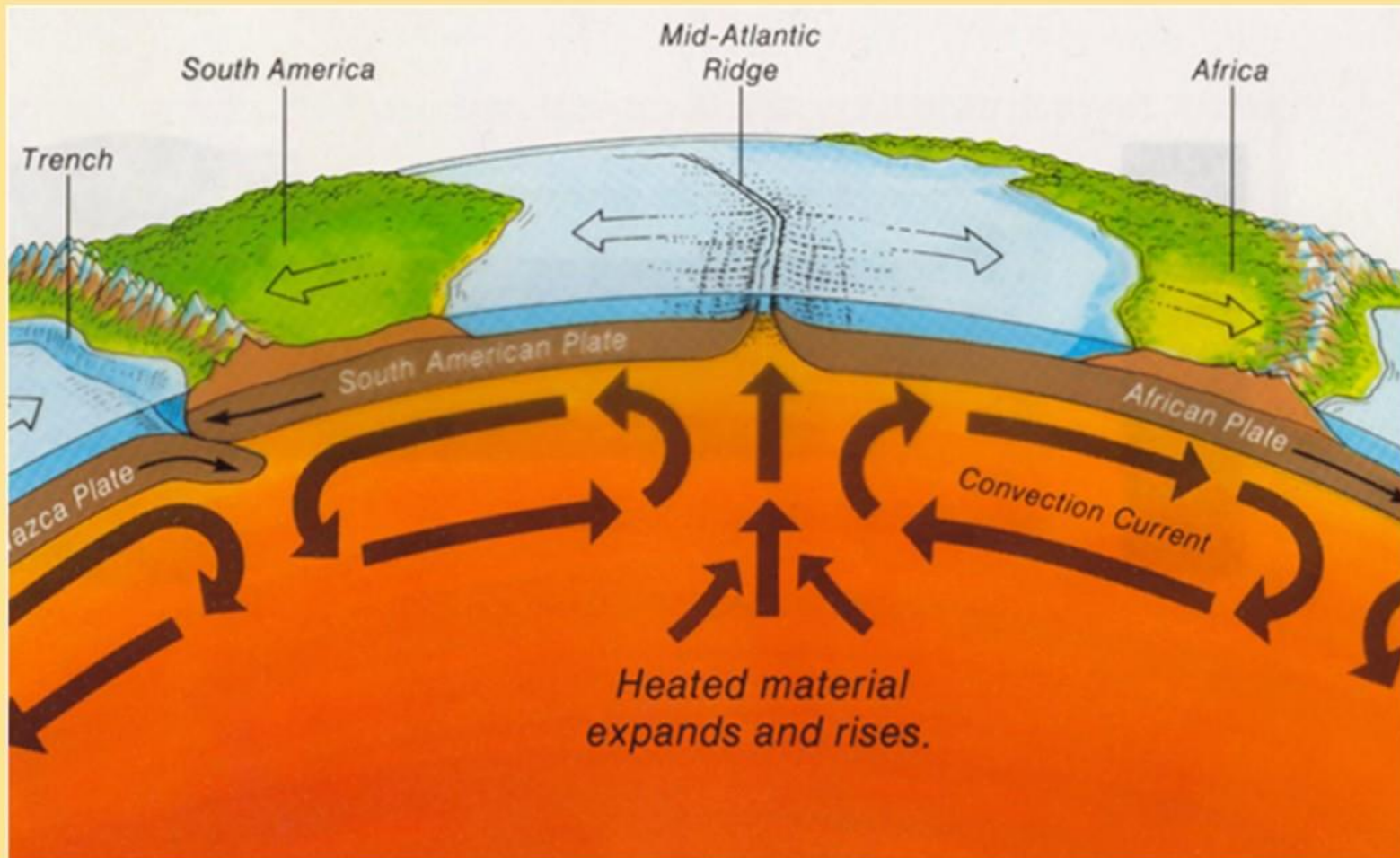
Wieland E, Hummel W, Formation and stability of ¹⁴C-containing organic compounds in alkaline iron-water systems: preliminary assessment based on a literature survey and thermodynamic modelling, Mineralogical Magazine Vol 79(2015) & Rizzato C, Rizzo A, Heisbourg G, Večerník P, Bucur C, Comte J, Lebeau D, Reiller PE, State of the art review on sample choice, analytical techniques and current knowledge of release from spent ion-exchange resins CAST report 4.1 (2015)



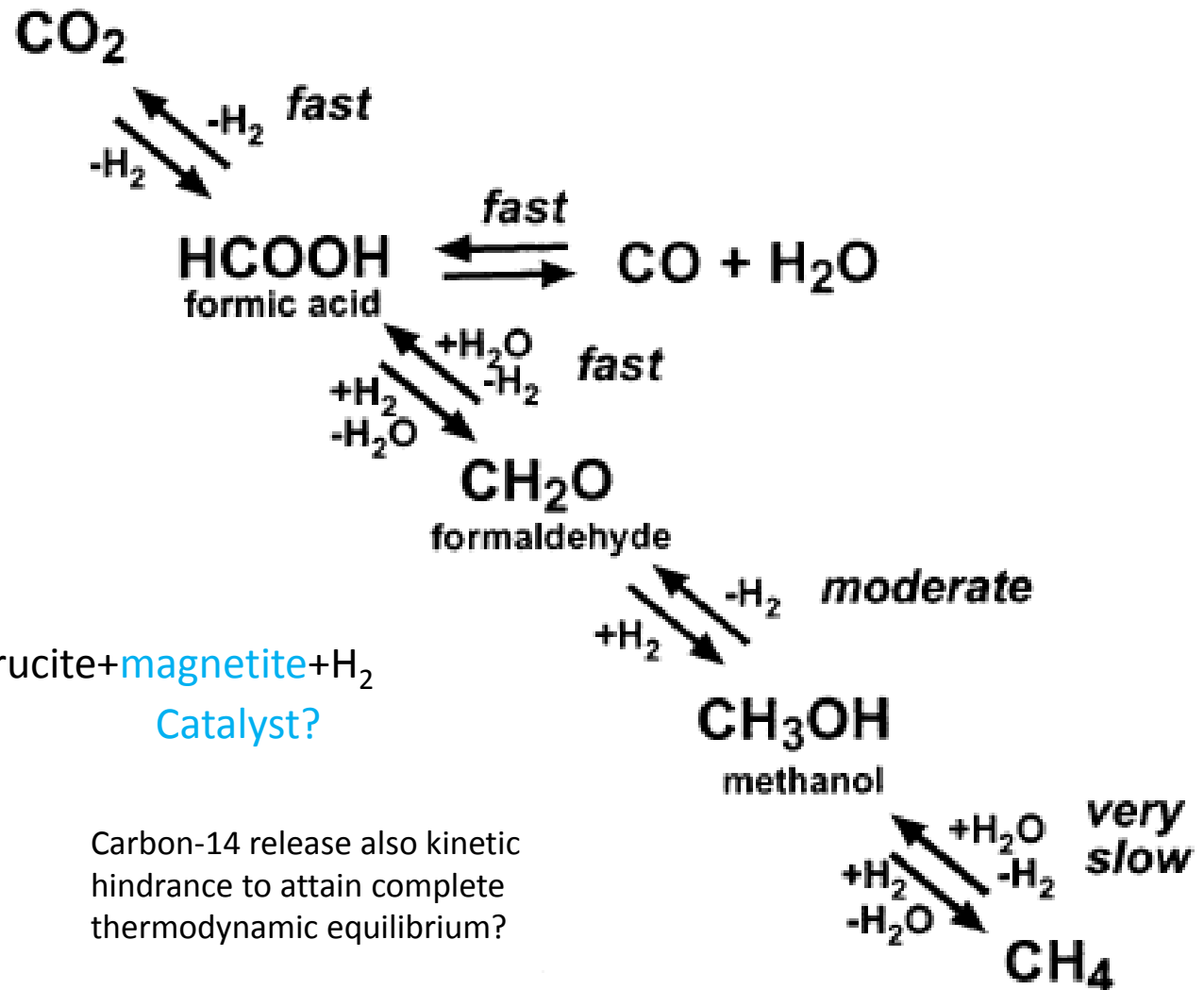
Potential carbon species



Potential carbon species



Potential carbon species

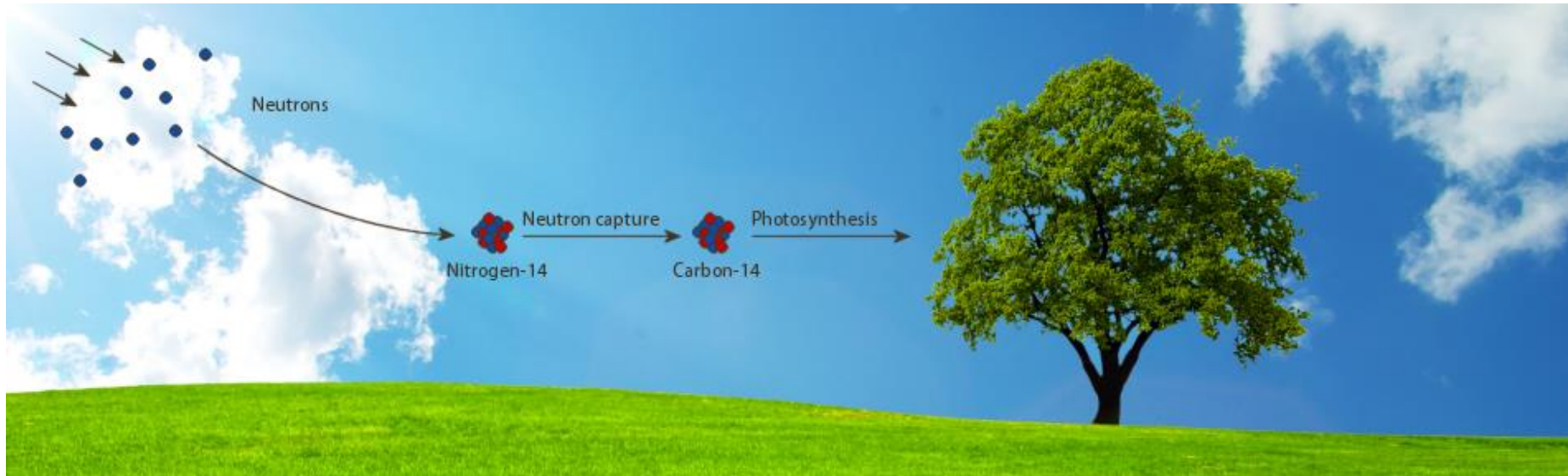


Olivine+water \Rightarrow serpentine+brucite+magnetite+H₂
Catalyst?

Carbon-14 release also kinetic hindrance to attain complete thermodynamic equilibrium?

McCullom & Seewald, 2007 Abiotic synthesis of organic compounds in deep-sea hydrothermal vents, Chemical Review
 Introduced by Wieland, 2015 for carbon-14 source term

Natural carbon-14



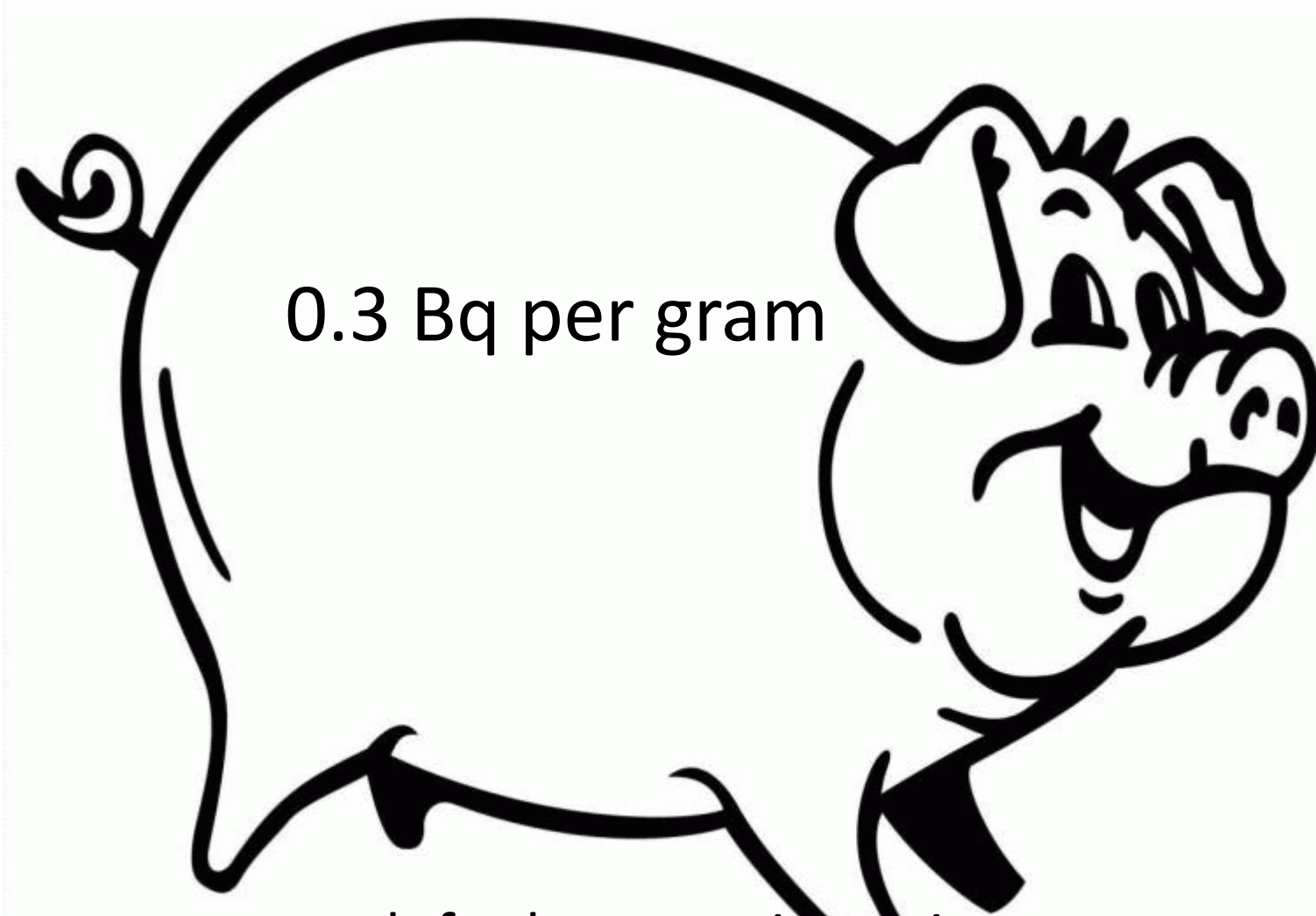


Exposure paths



- Inhalation
 - No concentration factor if not used by living matter for example noble gases
- Radiation exposure
 - For DTM radionuclides not likely
- Ingestion
 - Concentration factor if taken up by living matter for example carbon
 - Accumulation ^{14}C

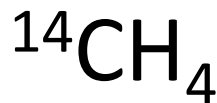
Natural carbon-14



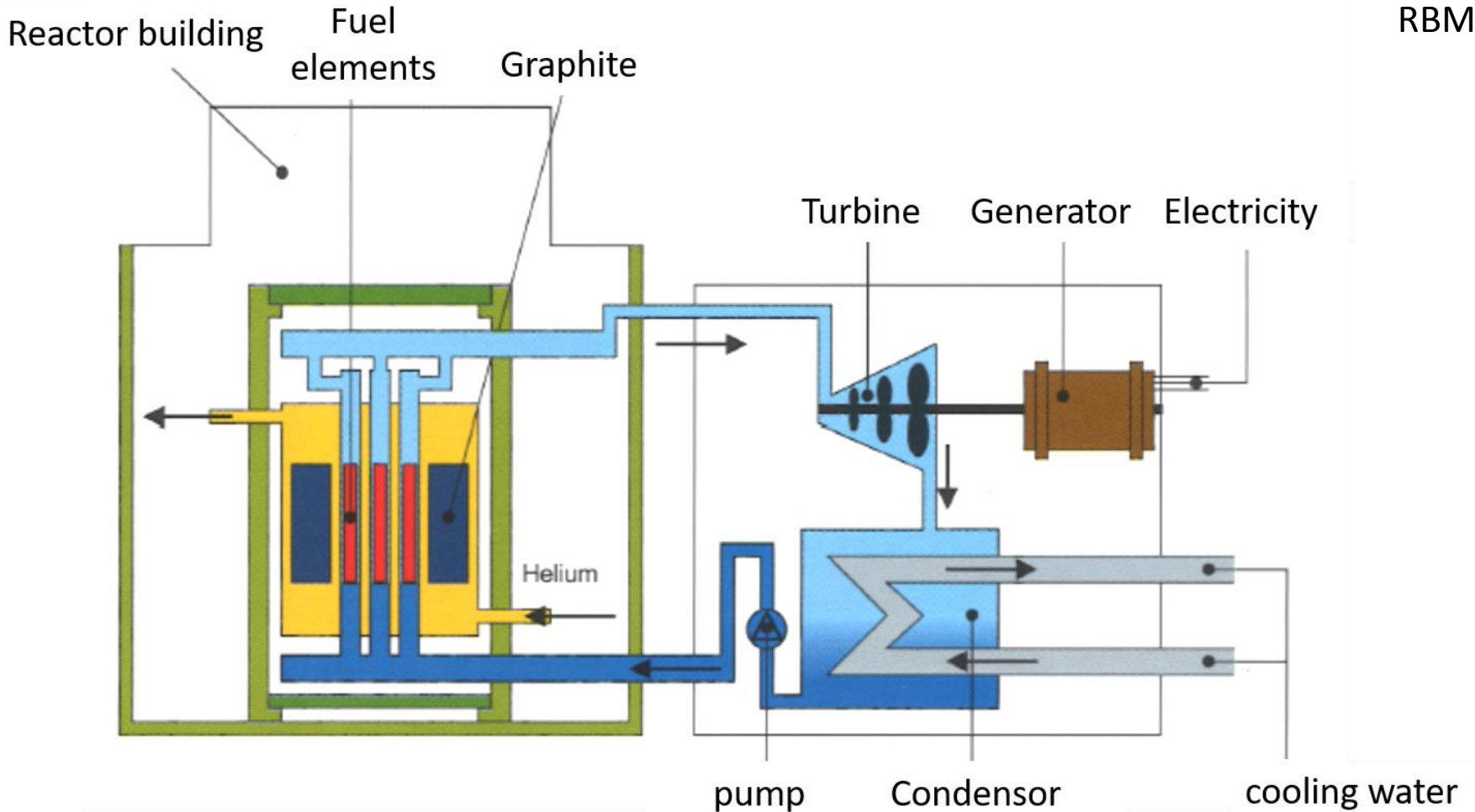
Main exposure path for humans: ingestion



root zone, microbial oxidation



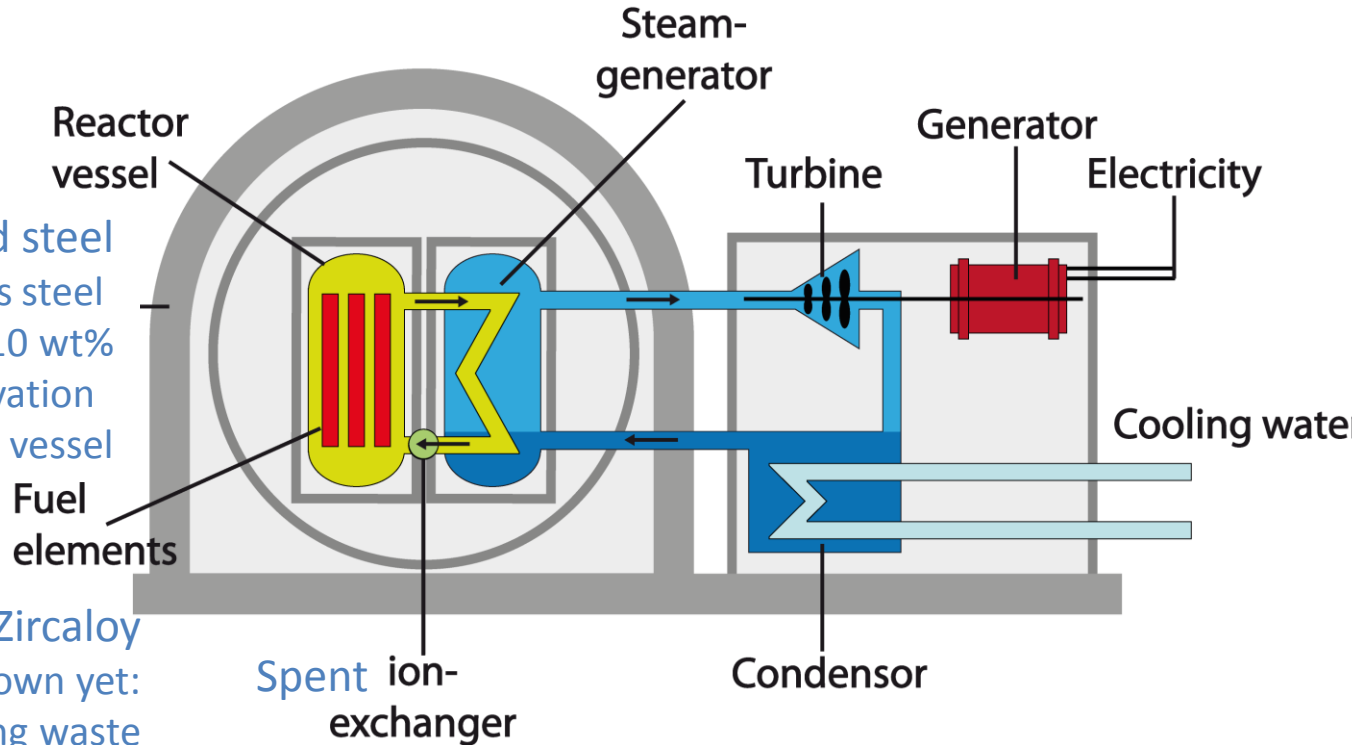
Main exposure path: ingestion



Irradiated graphite, in CAST mainly LILW: moderator graphite instead of water
 EU research project CarboWaste because HLW engineered barrier →
 expected containment period several decay times of carbon-14 ($t_{1/2}=5730$ years)



Radiological characterisation



Irradiated steel

Corrosion resistance: stainless steel

Nitrogen stainless steel max 0.10 wt%

Core parts mainly activation

Minor contribution inner part vessel

Irradiated Zircaloy

Nitrogen 34 ± 10 ppm but unknown yet:
if LILW \Rightarrow reprocessing waste

- 1) mixture with (foreign) wastes
- 2) Inconel ends
- 3) additional contribution capture gaseous carbon-14
- 4) Other neutron cross sections than JEFF 3.2

Irradiated graphite (in CAST mainly LILW):

if nitrogen content larger than 15 ppm

\Rightarrow main contribution to carbon-14

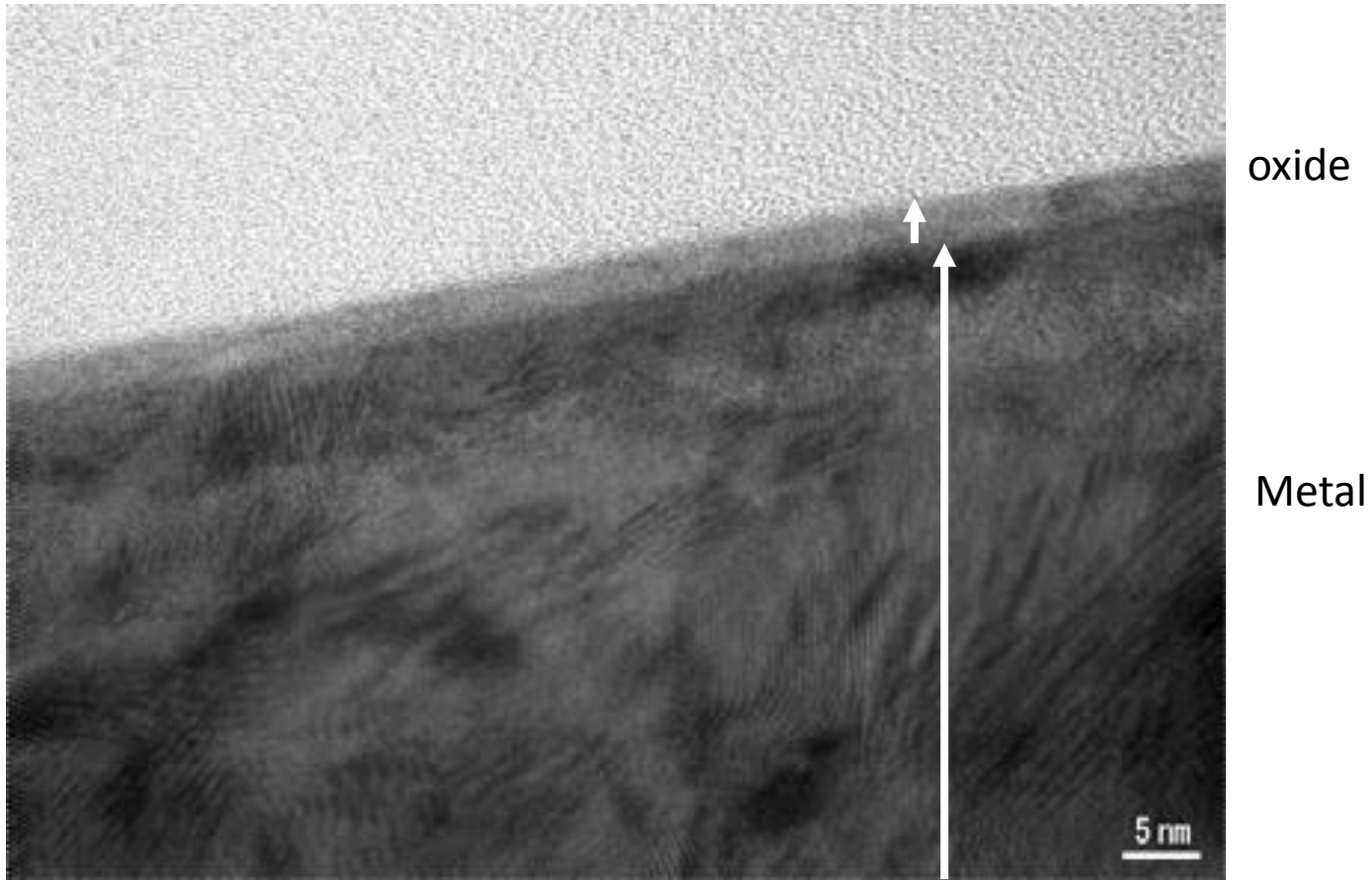


Neutron irradiated steel



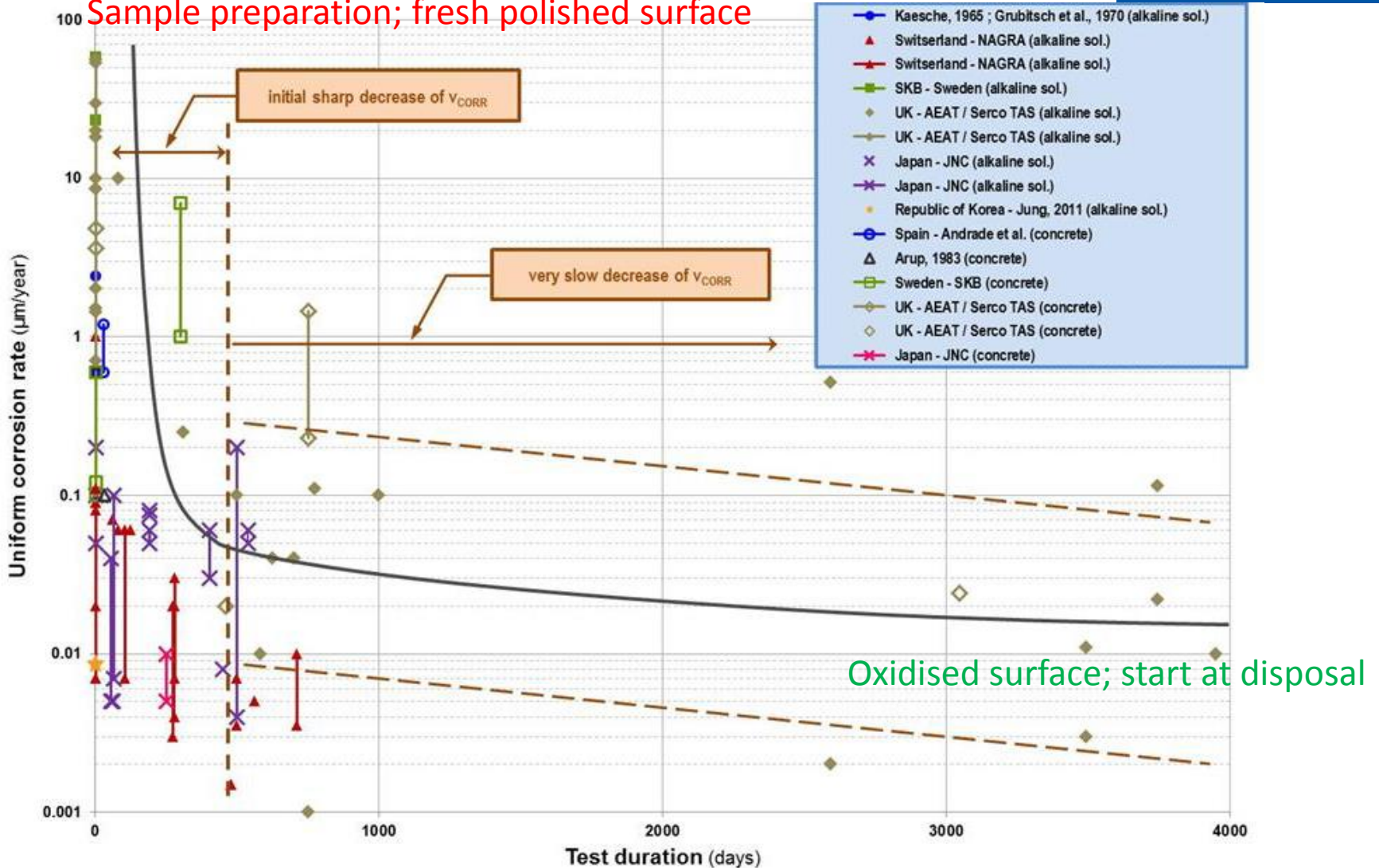
- Core assumed 10^5 Bq per gram steel
- Outer parts for example vessel assumed 10^3 Bq per gram
 - Sample vessel available in CAST 18 Bq per gram steel

Neutron irradiated steel

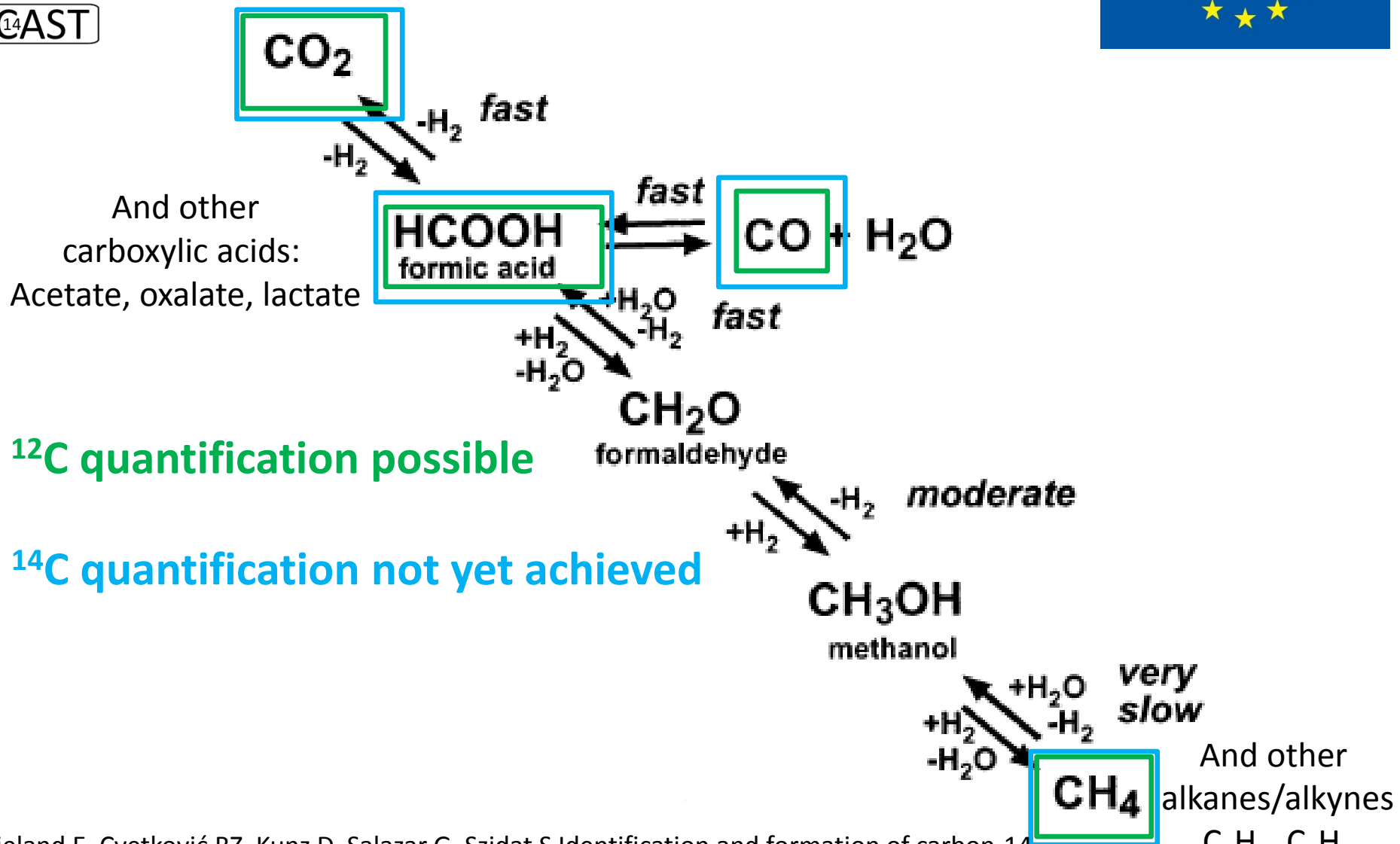


Steel

Sample preparation; fresh polished surface



Irradiated steel



Wieland E, Cvetković BZ, Kunz D, Salazar G, Szidat S Identification and formation of carbon-14 containing organic compounds during an anoxic corrosion of activated steel in alkaline conditions, IHLWRM-2017 proceedings

From Table in Mibus, 2018: CAST Final symposium WP2 outcomes, summary

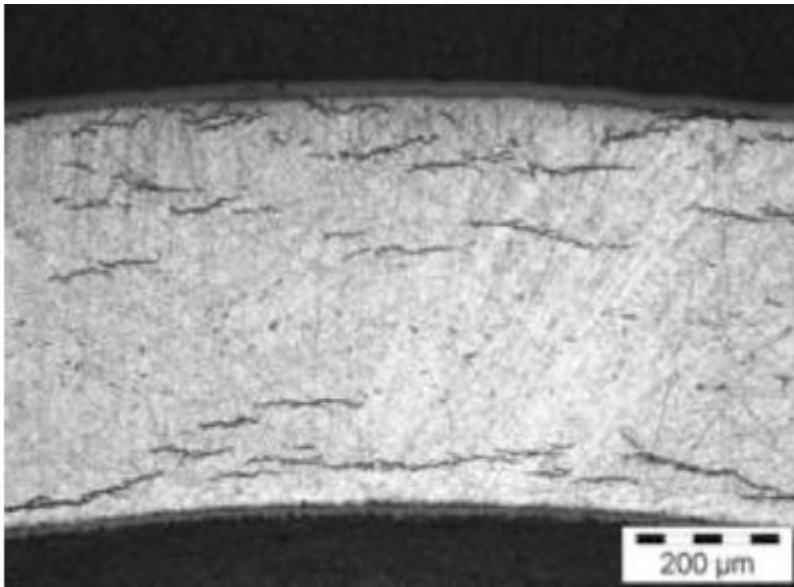


Steel

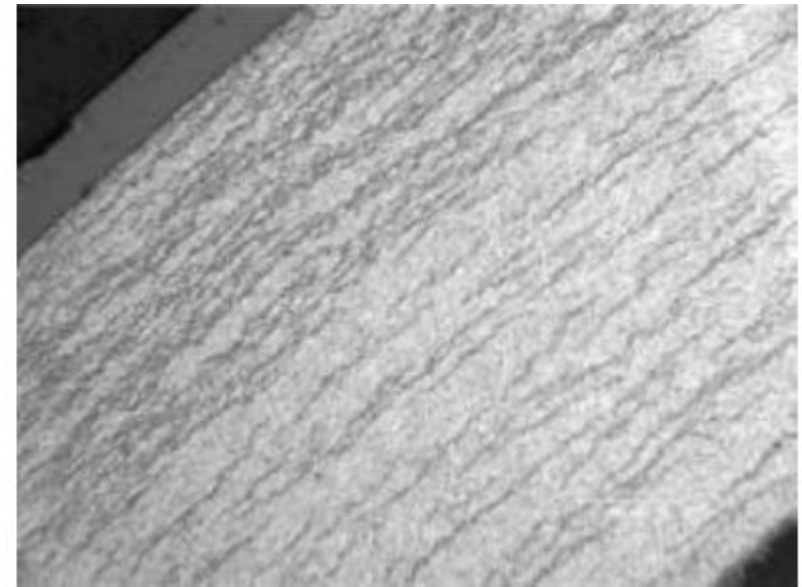


- Deep disposal reducing i.e. anoxic conditions
- Surface disposal
 - Potential ingress oxygen too small to prevent reducing conditions e.g. corrosion of metals
- During carbon-14 release at reducing conditions also hydrogen formation
 - $\text{Fe} + 2\text{H}_2\text{O} \rightleftharpoons \text{Fe}(\text{OH})_2 + \text{H}_2$

Hydride formation

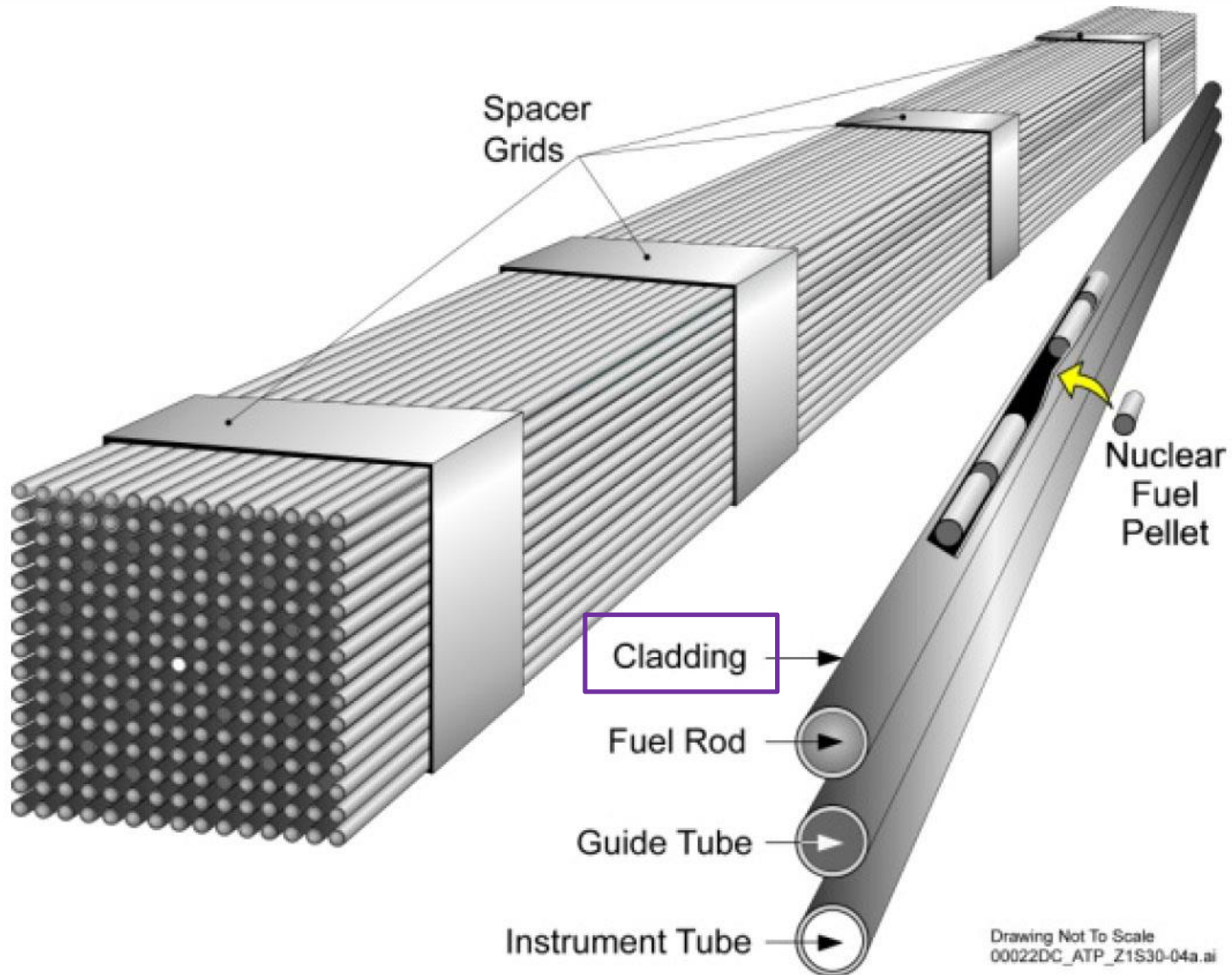


M5™, 5 cycles, stage 6
[AMB 2010]



Zirlo™, 67 GWd.t⁻¹
[AND 2012]

Neutron irradiated Zircaloy





Spent fuel



- High Level Waste
 - Half-life carbon-14 5730 years
- Deep disposal
 - Engineered containment copper canisters in Finland and Sweden expected to prevent contact with pore water many half lives of carbon-14
 - Negligible carbon-14 release from spent fuel



Neutron irradiated Zircaloy

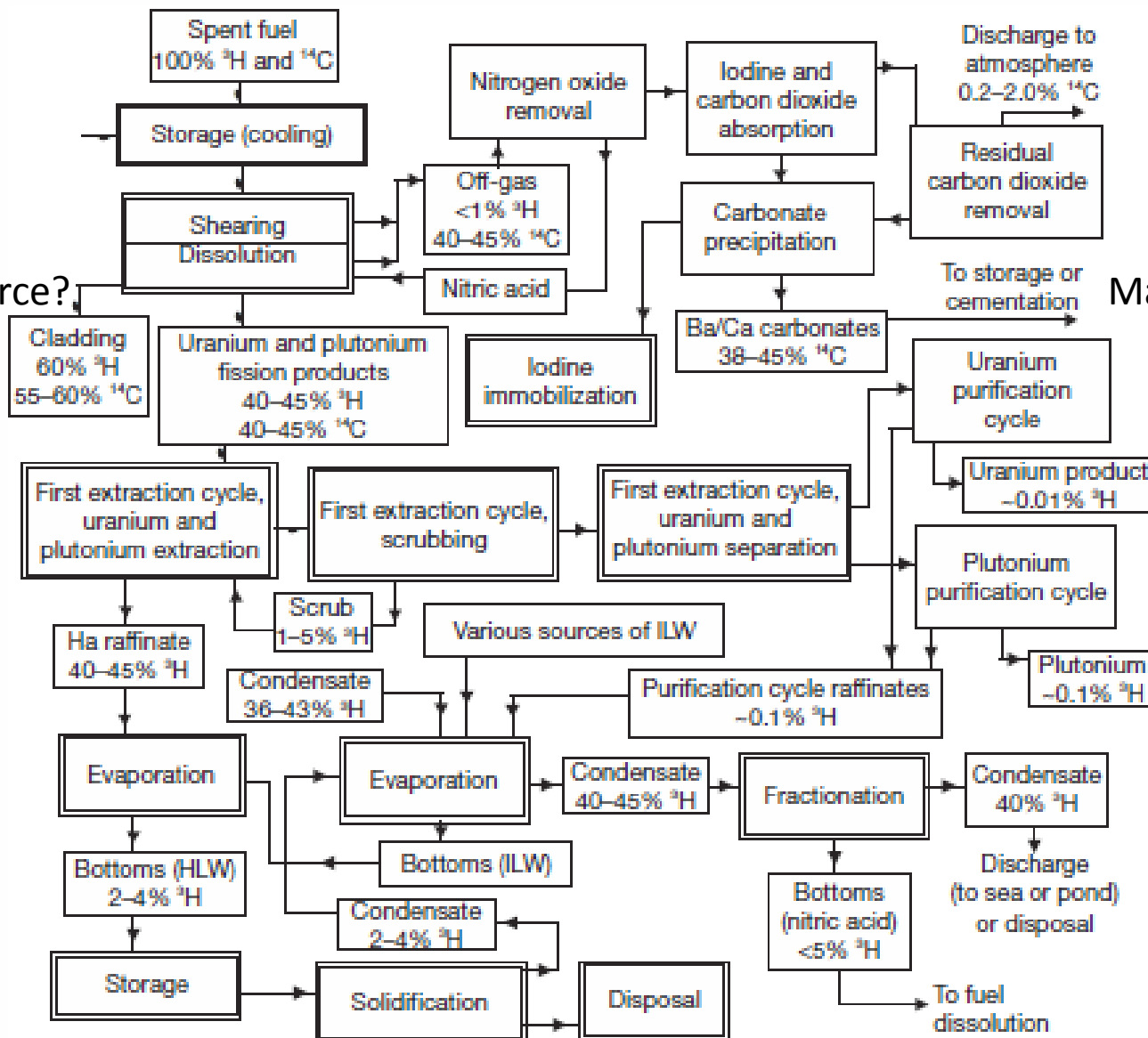


Caron, 2014: CAST report D3.2: Definition of operating conditions and presentation of the leaching experiments

Reprocessing

Main ¹⁴C source?

Main ¹⁴C source?





CSD-c as stored at COVRA's storage facility;

typical value for 900 MW NPP

1.4×10^{10} ^{14}C Bq per container; 'typical' value, best estimate

27000 Bq / gram solid waste

528 kg: 393 kg Zr (hulls) , 19 kg Inconel (ends) , 116 kg ss (technological waste)

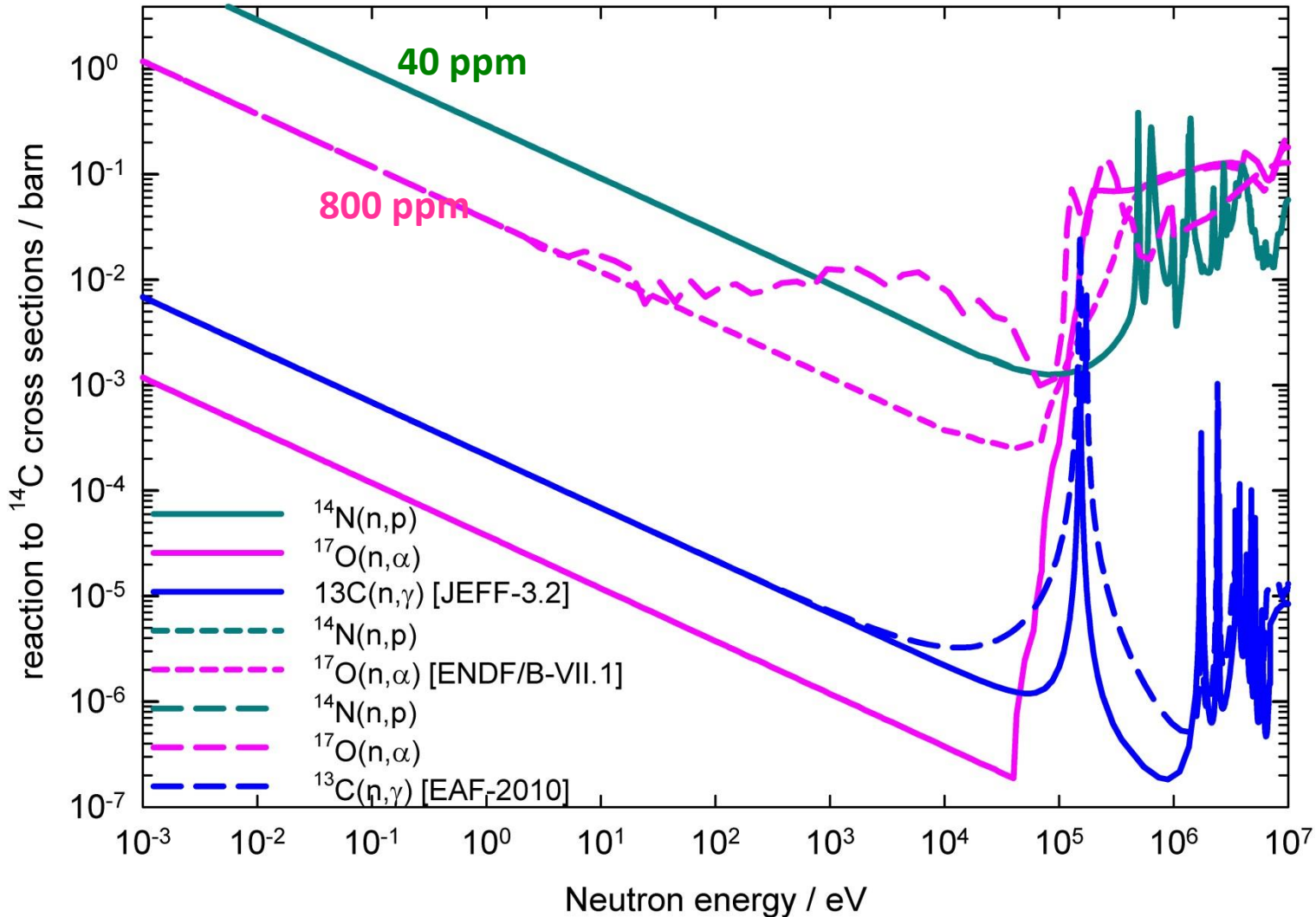
Reported by AREVA

GK280

07/R115

Neutron cross sections for determination typical value

at 300 K



NEA nuclear databank libraries Joint Evaluation Fission Fusion file (2014) JEFF-3.2,
Evaluated Nuclear Data File (2011) ENDF/B-VII.1, European Activation File EAF (2010)



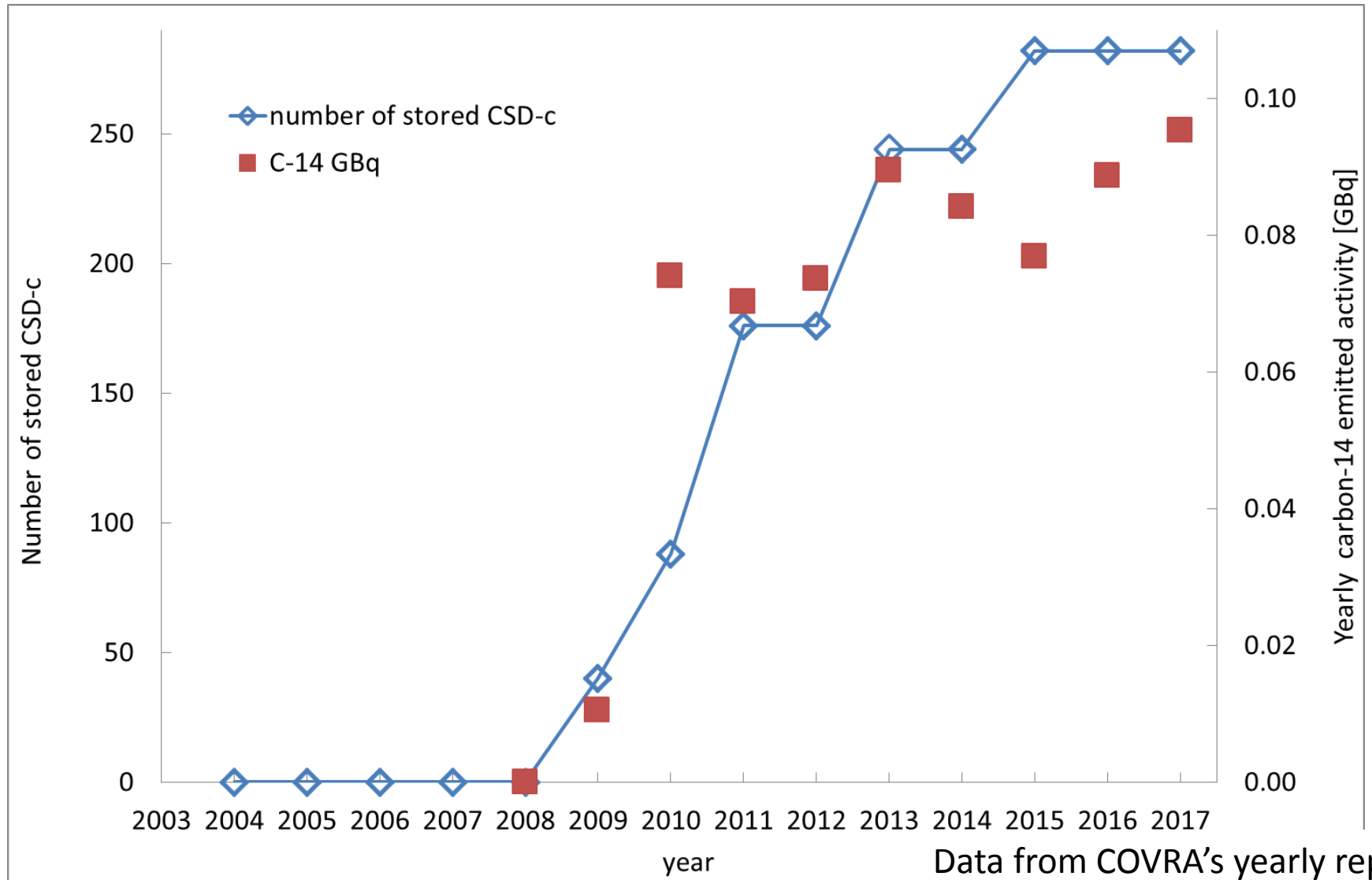
Neutron irradiated Zircaloy



- $\approx 10^4$ Bq ^{14}C per gram Zircaloy
 - Tenfold lower nitrogen content than steel
 - Operational waste not decommissioning waste consequently smaller neutron irradiation period
- Carbon solubility smaller than nitrogen solubility
 - Small precipitate 14-carbides



Gaseous carbon-14 release during storage





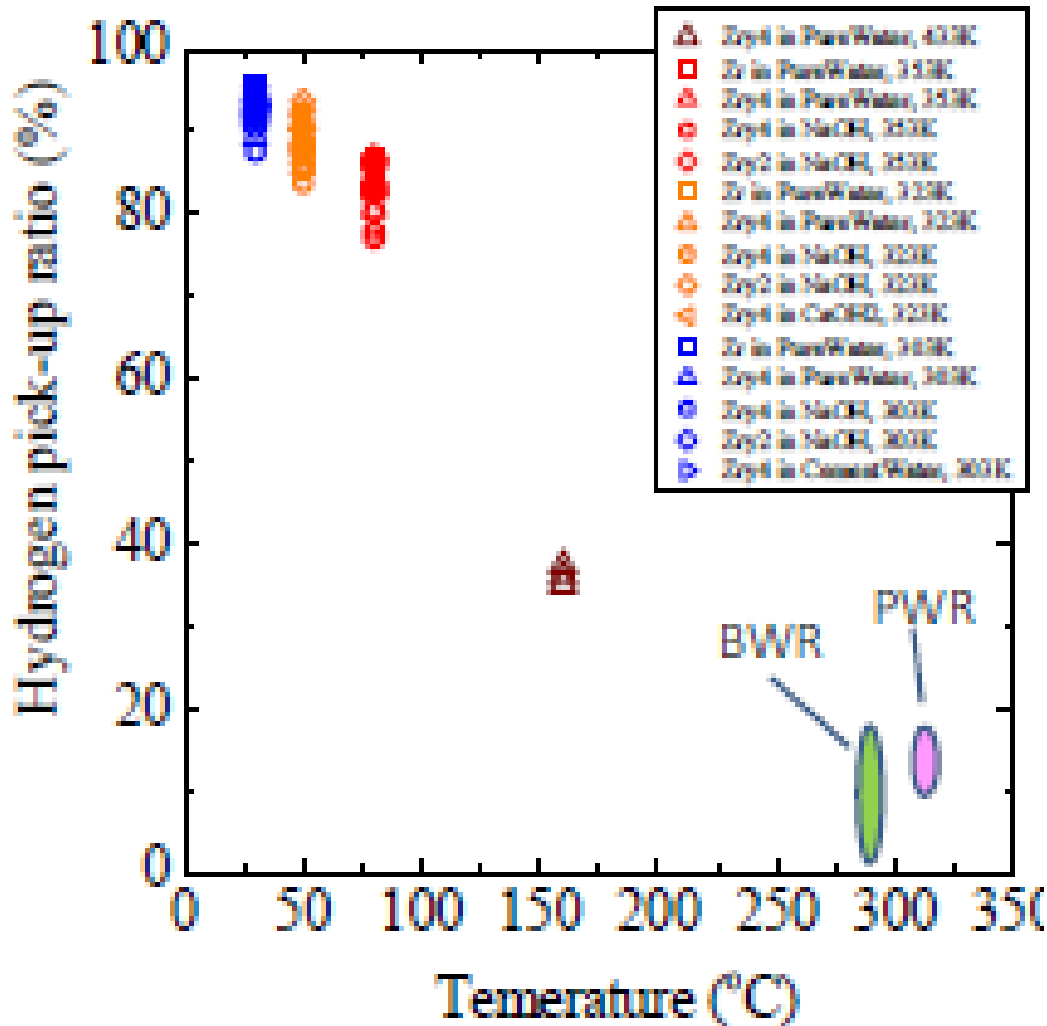
Neutron irradiated Zircaloy



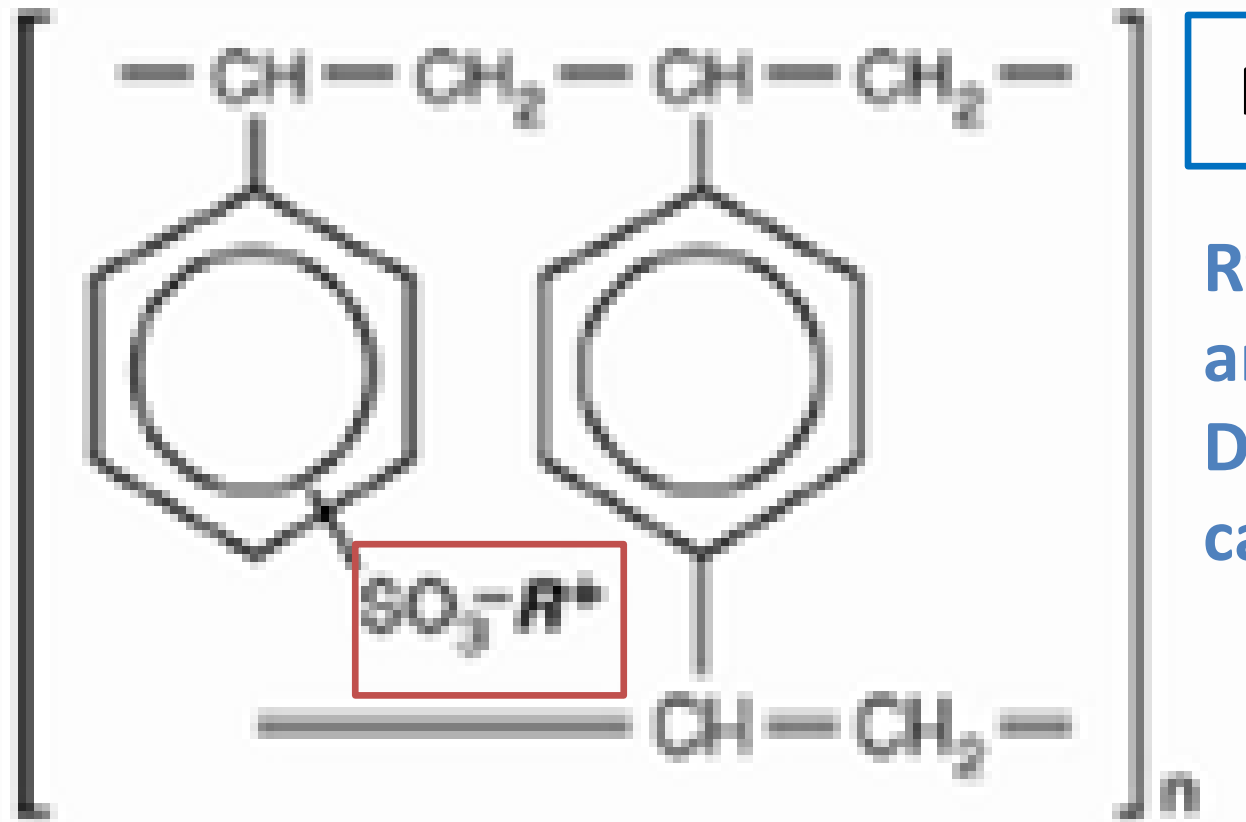
- Zirconium exothermic dissolution of hydrogen
 - Iron endothermic dissolution of hydrogen
 - Lacher, 1937: Zr-H phase diagrams, Iron and hydrogen Sievert's law
- Hydrogen pick-up
 - During neutron irradiation in reactor tritium containment
 - During disposal limited hydrogen inward flux into engineered and natural barriers

Neutron irradiated Zircaloy

Disposal conditions



Spent ion exchange resins

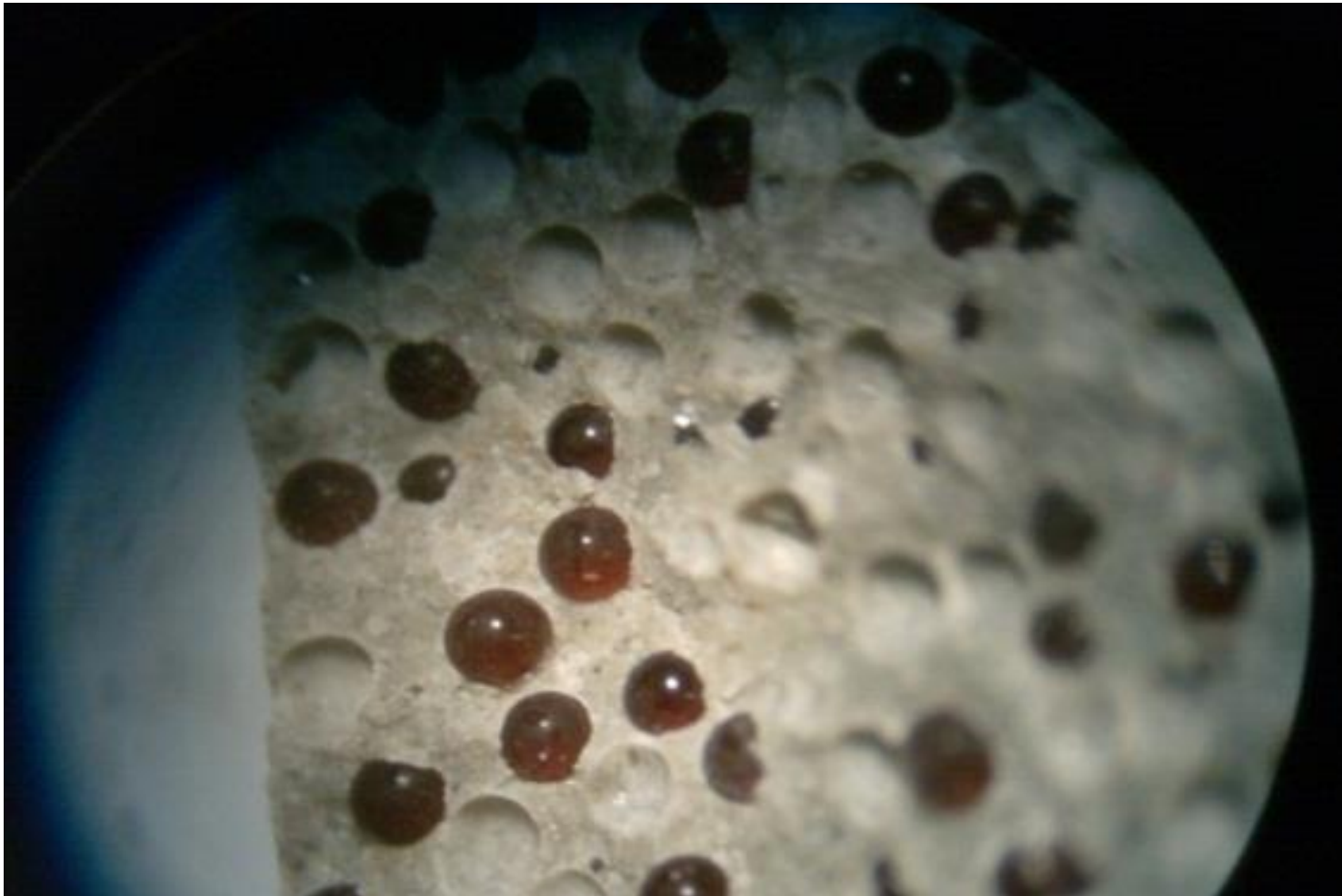


R^- e.g. OH^-
anion exchange
Dissolved
carbon species

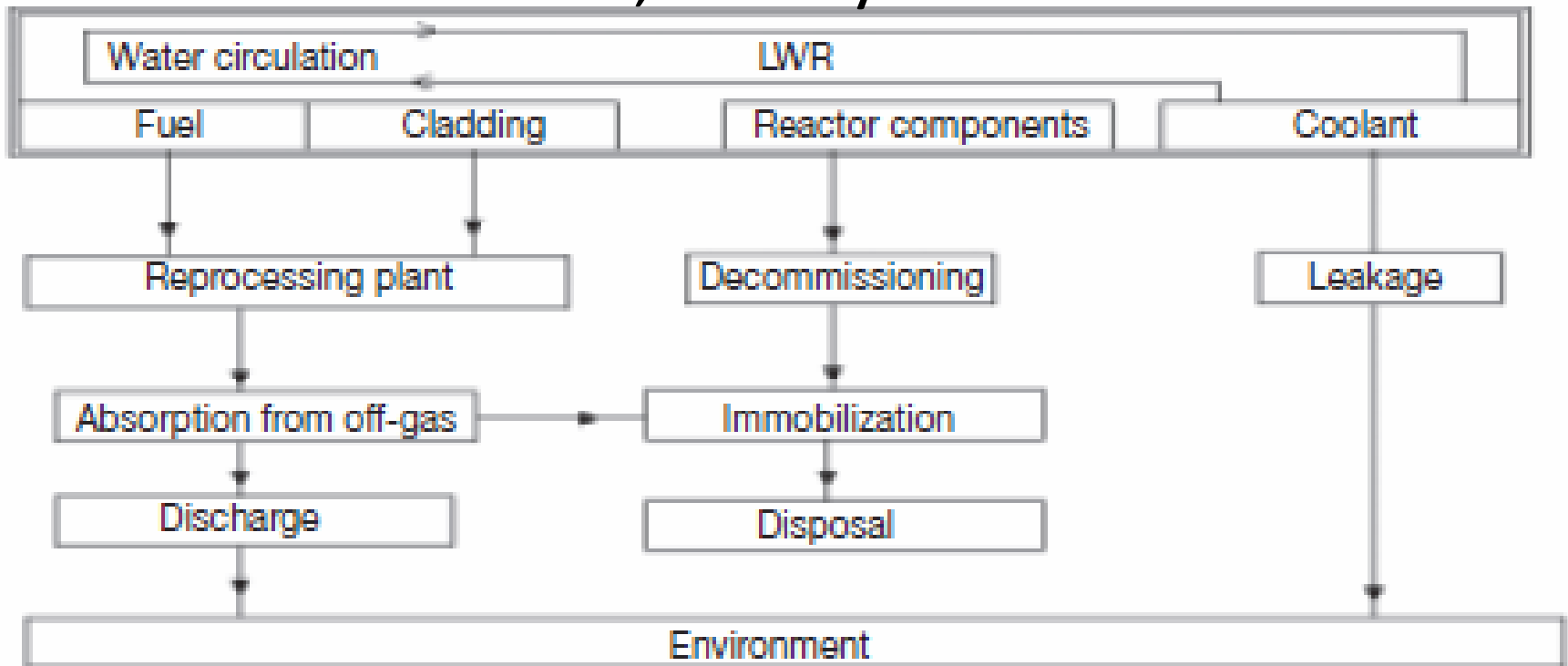
R^+ e.g. H^+ cation exchange



Spent ion exchange resins



- $\approx 10^3$ Bq ^{14}C per gram wet and dry resin measured in CAST, mainly beads



Neutron irradiated graphite

- Romania: contact-handled irradiated graphite research reactor



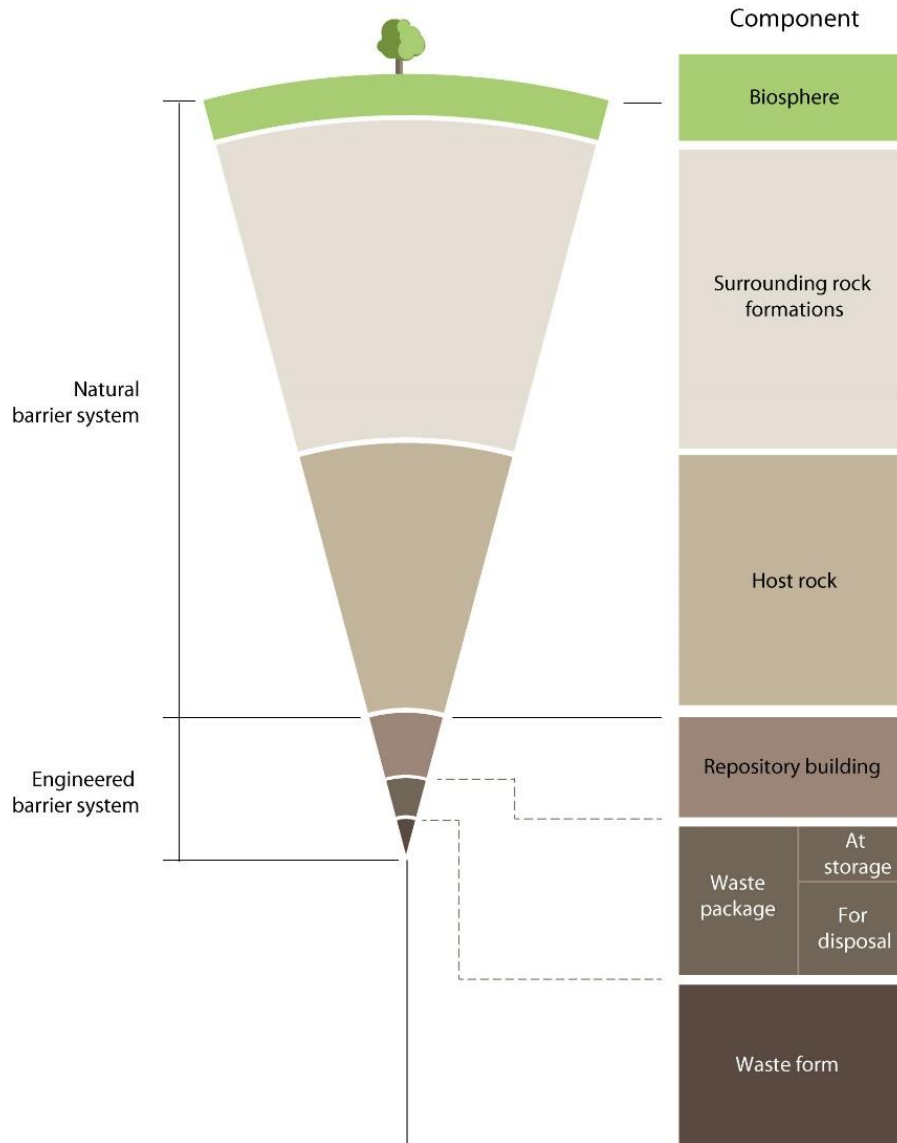
- Italy: remote-handled irradiated graphite
 - Canzone G et al (SOGIN) Dismantling of the graphite pile of Latina NPP: Characterization and handling/removal equipment for single brick or multi-bricks, Progress in Nuclear Energy 93 (2016) 146-154



Neutron irradiated graphite



Release mechanism



Cementitious materials



Release mechanism

- Source term: carbon-14 release rate or rates from waste
- Release under conditions relevant for waste packaging and disposal to underground facilities
 - Cementitious matrices, main waste packaging conditions considered in CAST
 - Cement alkaline conditions
 - Portland: initially slightly oxidising and largely unbuffered because of lack of electroactive species
 - corrosion of metals may reduce redox potential locally
 - Blast furnace slag: initially reducing due small amount of FeS_2 – blueish colour when not oxidised –
 - corrosion of metals may locally sustain reducing conditions
 - Underground
 - Near-surface disposal: aerobic exposure conditions
 - Deep geological disposal: anaerobic exposure conditions



Conclusions / highlights



- CAST finishes on 1 April 2018
 - Final General Assembly Meeting in January 2018 in France (Lyon)
- During running research programme CAST
 - State Of the Art reports at start of the programme
 - knowledge management
 - WMO view: what does experimental research contribute to what is already known?
 - Determination activity concentration of carbon-14 in waste appeared to become more important
 - Nitrogen impurities in steel and Zircaloy measured
 - Standard ASTM specification published in 1973 for quantitative determination of gaseous impurities in metal and alloy solid samples; in CAST performed by an external lab close to Toulouse, France: Evans Analytical Group
 - Unknown if nitrogen content has been reduced since 1984 ALARA
 - Focus on reliable determination of carbon-14 activity concentration in spent ion exchange resins and distinction between organic carbon i.e. functional groups exchanged carboxyl acids and inorganic i.e. functional groups exchanged with carbonate
 - Also in neutron irradiated graphite, nitrogen impurities can be main source of carbon-14
 - Obtaining representative samples and setting-up experiments takes time
 - Corrosion rates of steel at geological disposal, i.e. passivated surfaces in cementitious materials, possibly too hard to measure reliably DTM radionuclide such carbon-14, release rate. Experiments in the framework of the Swiss and UK programme continue after the end date of CAST.



Thank you
for attention. Any questions?

CAST reports and newsletters free online
available at www.projectcast.eu

***Radiocarbon dating,
monitoring activity around nuclear facilities
and modelling carbon-14 in the environment***

Mihály Molnár

Institute for Nuclear Research, Hungarian Academy of Sciences

Isotoptech Zrt, Hungary

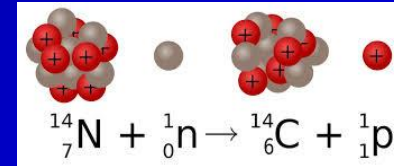
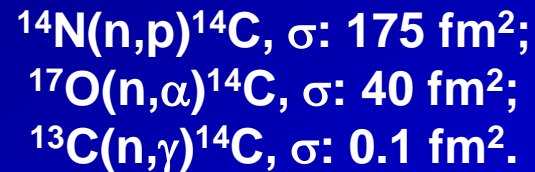
www.isotoptech.com

molnar.mihaly@atomki.mta.hu

**Enhanced atmospheric C-14 monitoring
around the Paks NPP of Hungary**

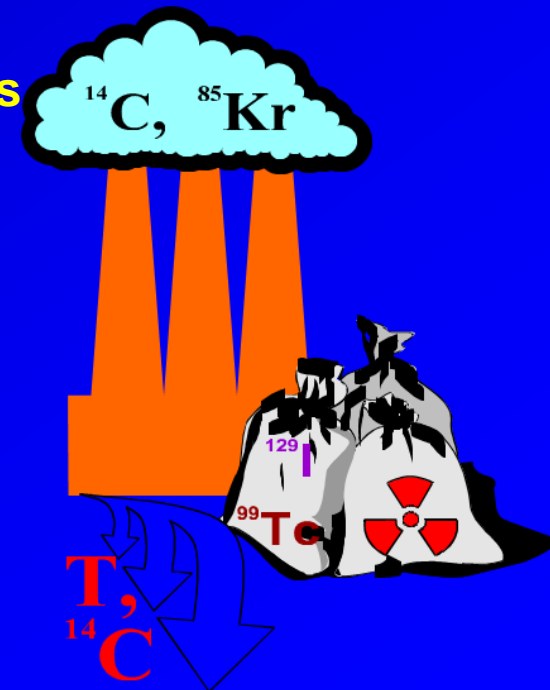
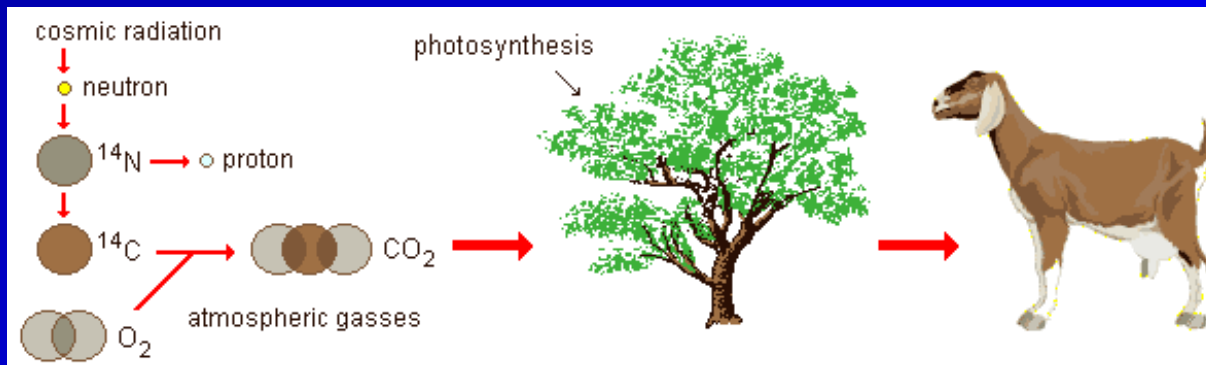
Importance of C-14 in NPP monitoring, motivation

In the light water nuclear reactors of VVER- 440 type several nuclear reactions are possible with different cross sections to produce ^{14}C (Chudy and Povinec, 1982):



The ^{14}C is released from nuclear reactors in different chemical forms. In the VVER-440 reactors ^{14}C is predominantly released in the form of hydrocarbons (70–95%) and rest in the form of CO_2 (Uchrin et al., 1998).

The total dose resulting from the release of all radionuclides from nuclear power reactors generally is dominated by the contribution from ^{14}C (see e.g. UNSCEAR 2000)



Paks VVER-400 type NPP and its tradition in C-14 monitoring



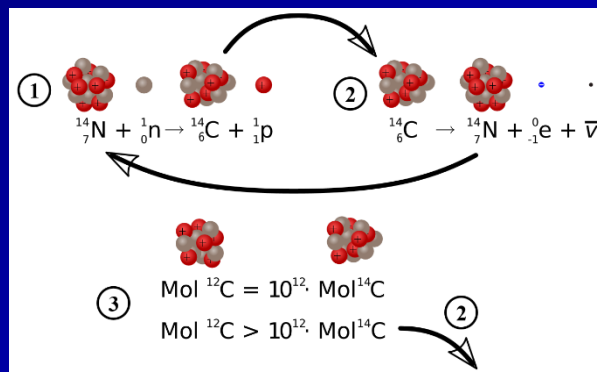
Since: 1982-1987
Units operational: 4 x 500 MW
Make and model: VVER-440
Units planned: 2 x 1,200 MW
Nameplate capacity: 2,000 MW
Capacity factor: 84.2%
Annual output: 14,749 GW·h



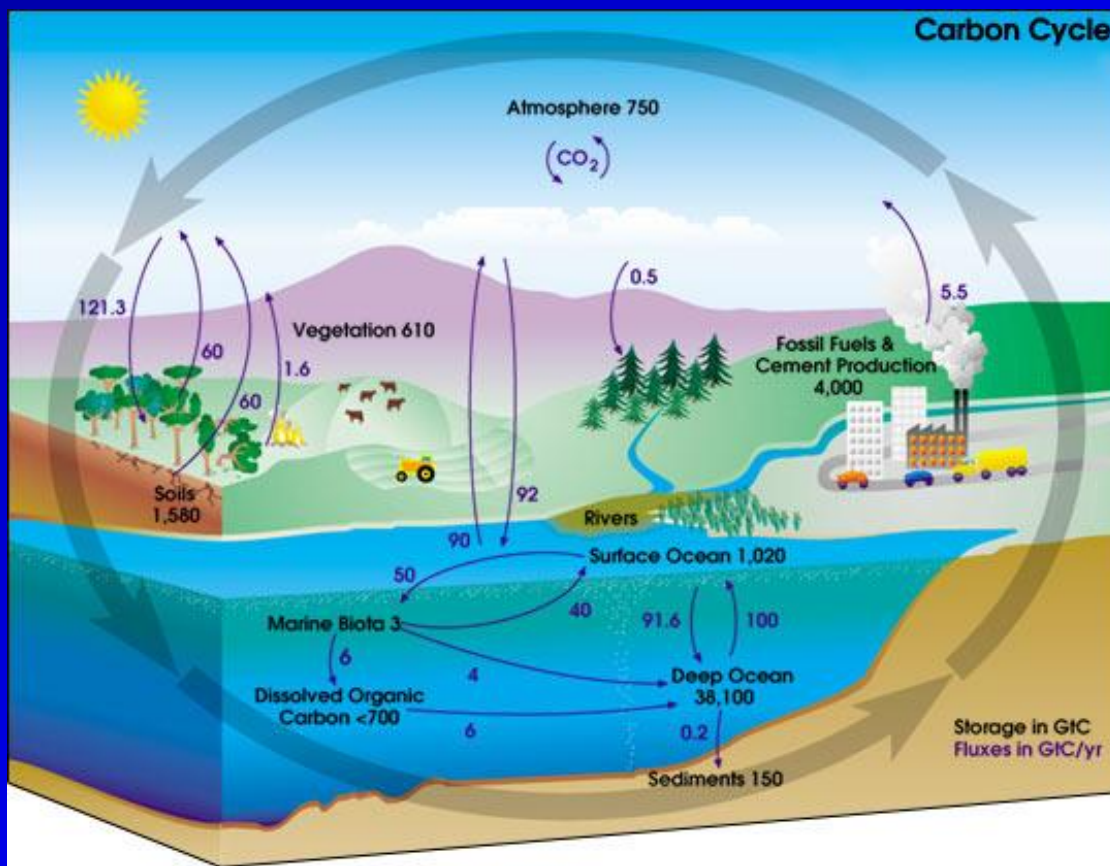
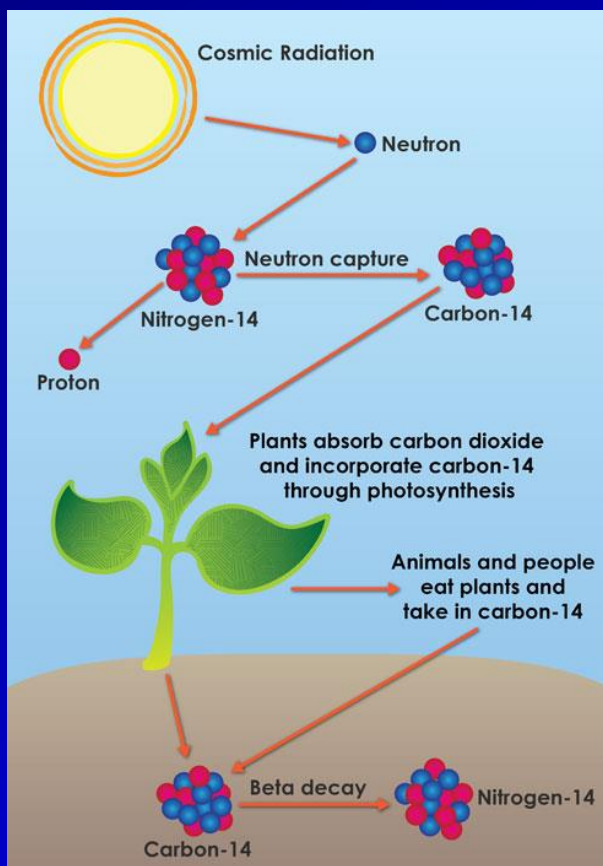
- *Uchrin et al. 1992. **^{14}C release** from a Soviet-designed pressurized water reactor nuclear power plant. Health Physics 63 (6), 651–655.*
- *Veres et al. 1995. Concentration of **radiocarbon and its chemical forms** in gaseous effluents, environmental air, nuclear waste and primary water of a pressurized water reactor power plant in Hungary. Radiocarbon 37 (2), 473–497.*
- *Uchrin et al. 1998. **^{14}C measurements at PWR-type nuclear power plants** in three middle European countries. Radiocarbon 40 (1), 439–446.*
- *Molnár M. et al. 2007. Monitoring of atmospheric **excess ^{14}C** around **Paks Nuclear Power Plant, Hungary.** Radiocarbon 49 (2007)1031-*
- *Now its is again 10 years left, so we are going to publish what is new at Paks NPP...*

Natural/National/Local background in C-14 monitoring

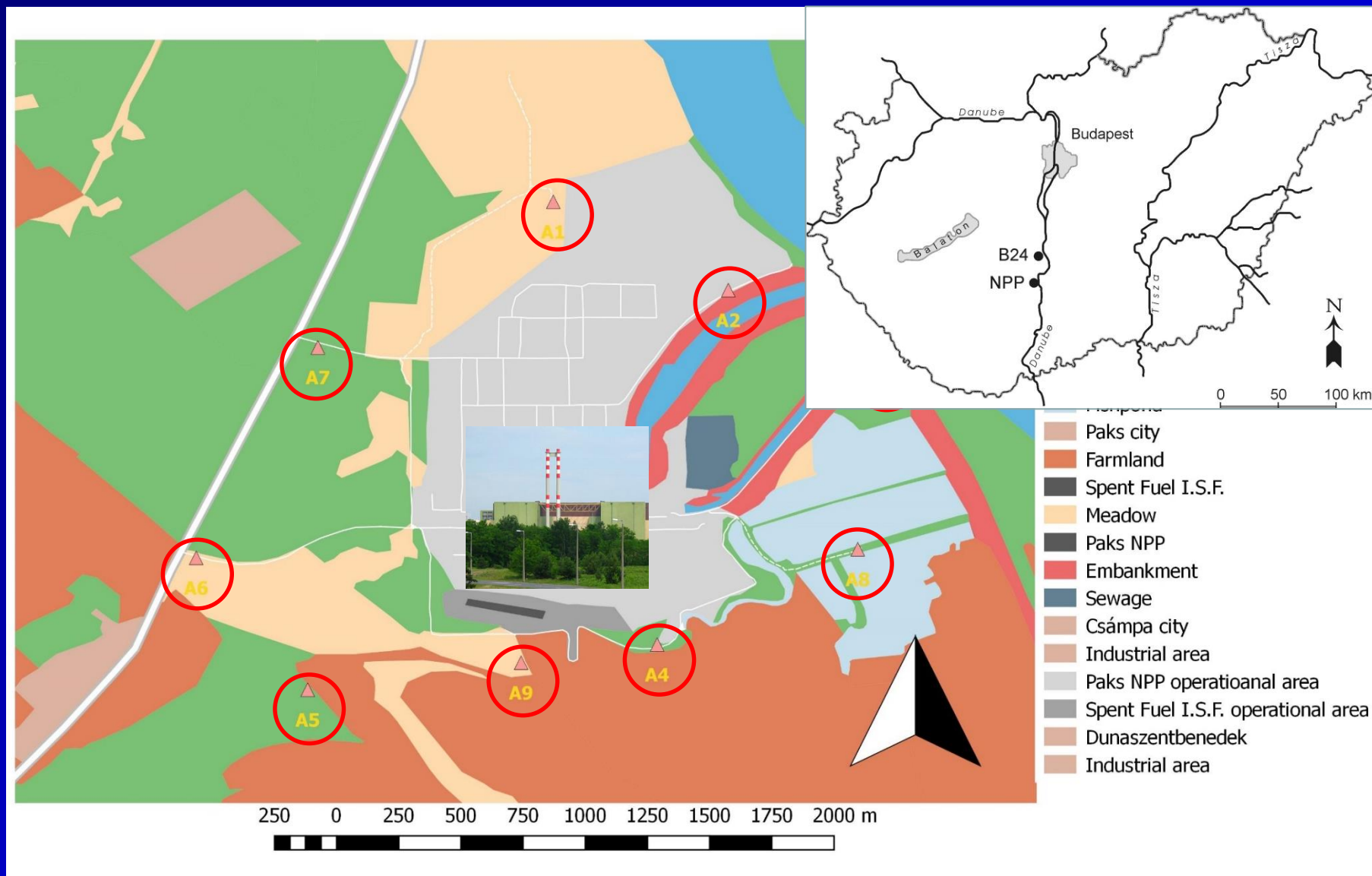
Natural production:
Cosmogenic isotope
 ~ 1500 TBq/ yr
 - Stratospheric origin



Beta decaying isotope
half-life: 5700 ± 40 yrs
Total amount: 51 t
 $^{14}\text{C}/^{12}\text{C}$ ratio: ~ 10^{-12}



Atmospheric C-14 monitoring network around Paks NPP



A local background monitoring station (B24) is running in 20 km distance

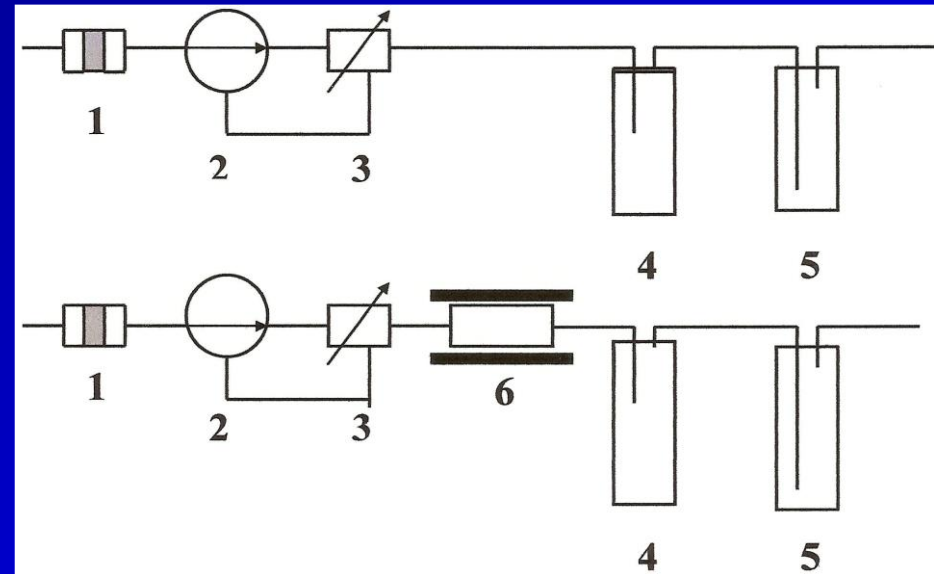
Atmospheric C-14 monitoring network: A-type station

Combined ^3H and ^{14}C sampling Unit
for H_2O , H_2 , CO_2 and C_nH_m forms



10 dm³ air/min sampling rate
for 1 months

Atmospheric C-14 monitoring network: Combined ^{14}C sampling Unit



Layout of ^{14}C sampler developed and used for monitoring of NPP ^{14}C discharges in the form of CO_2 and C_nH_m (separately):

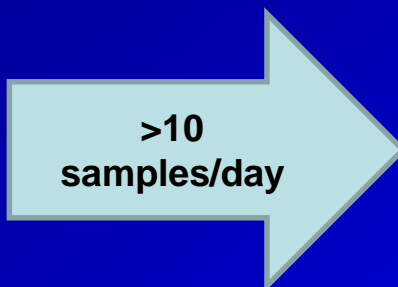
- 1) filter;
- 2) air pump;
- 3) flow controller;
- 4) puffer;
- 5) bubbler with 500 mL of 3M NaOH ;
- 6) converter (Pt-Pd catalyst at 450°C).

+ the same sampling units are used for the stack air ^{14}C monitoring at Paks NPP

Sample preparation and AMS ^{14}C analyses of exposed NaOH samples



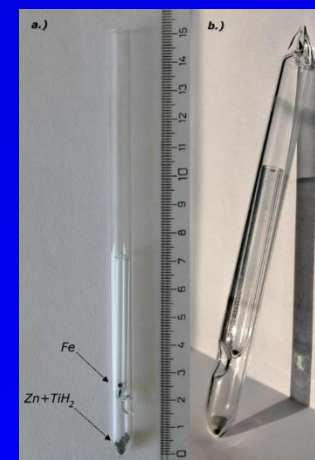
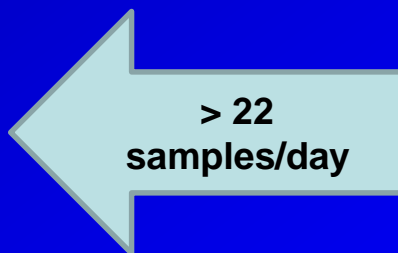
1-3 mL of NaOH prepared by acid in vacuum cell



2-4 cm³ of CO₂ extracted/cleaned

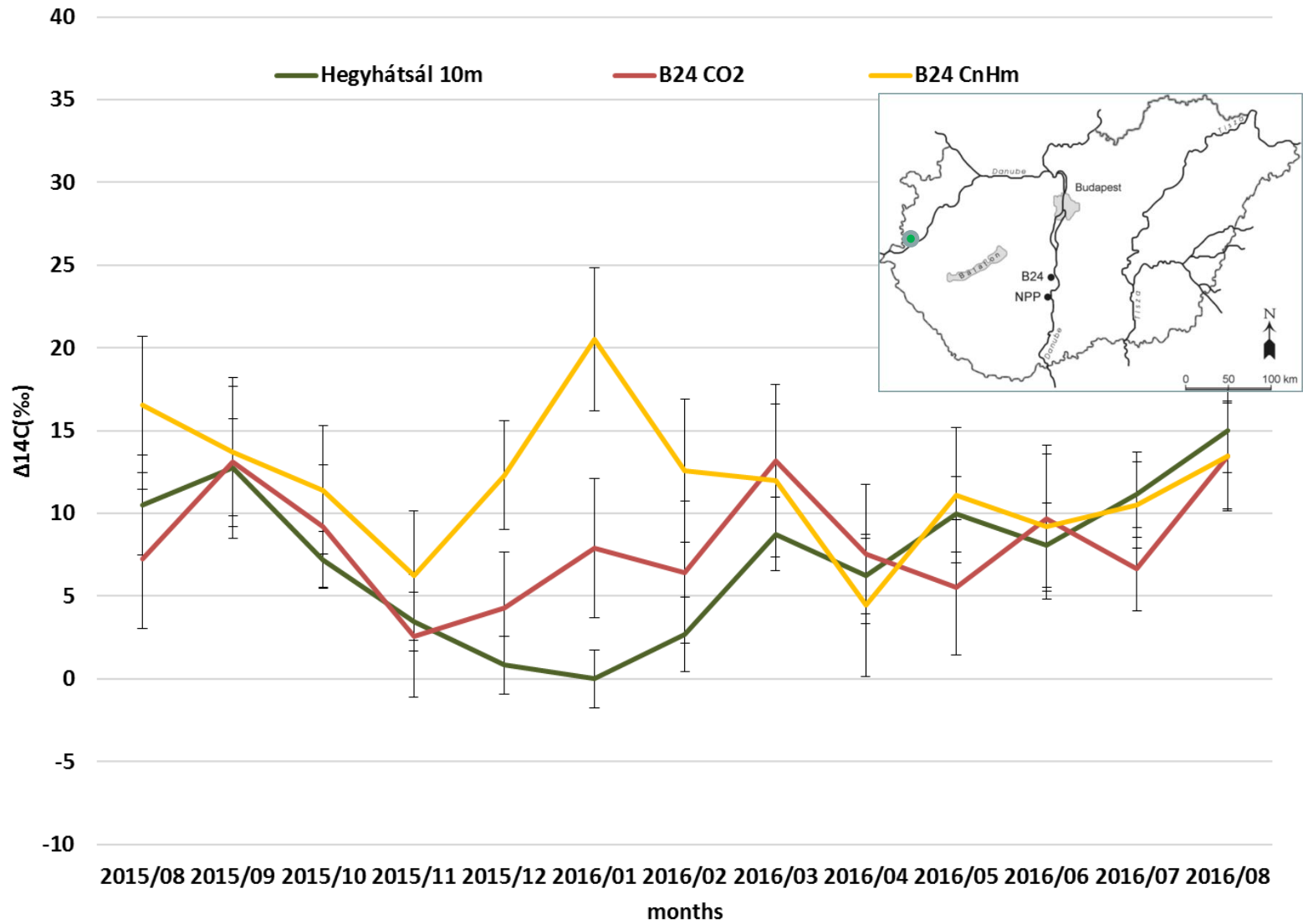


MICADAS ^{14}C AMS (ETHZ)
1s error < +/- 0.5%

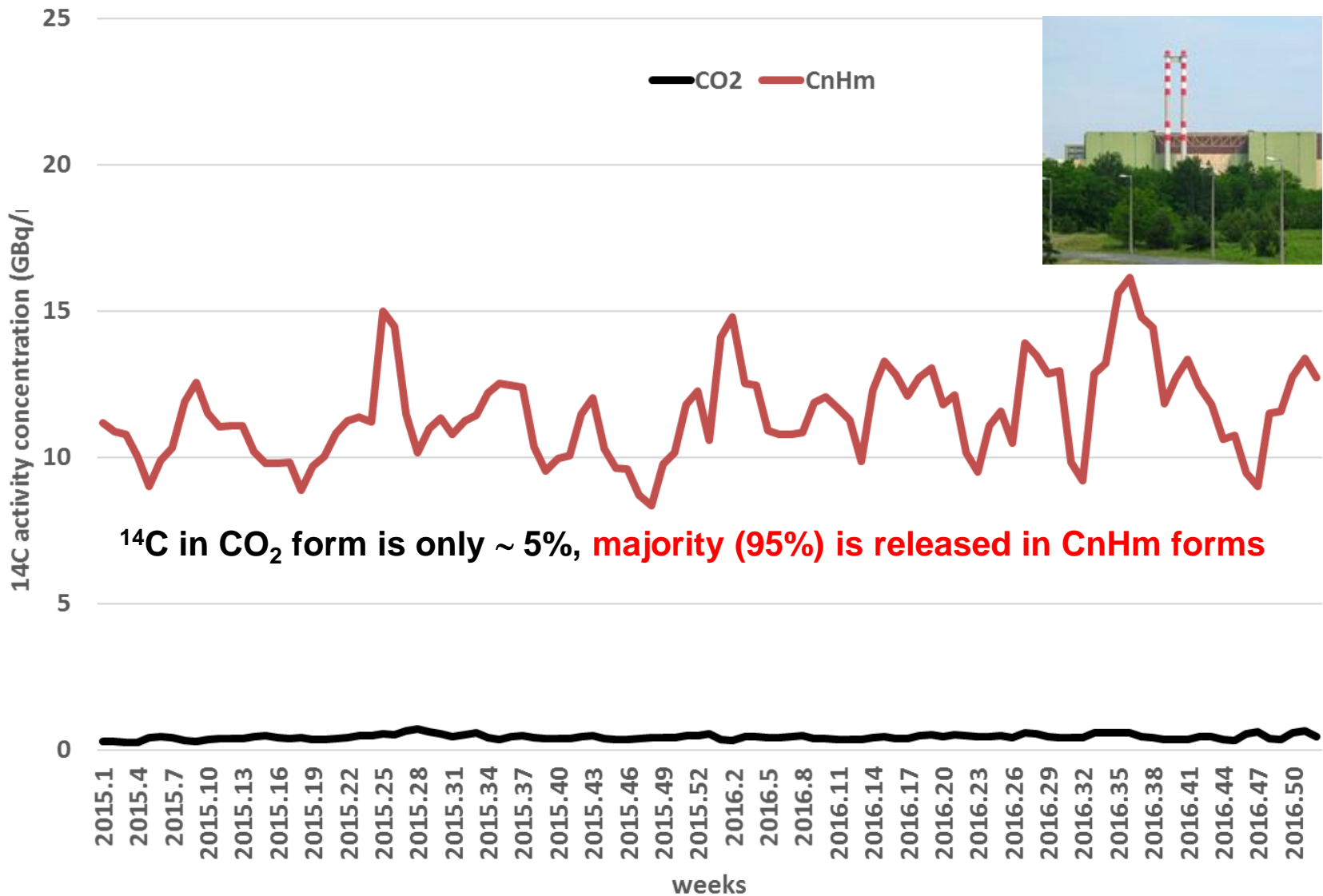


1-2 mg of C graphitized in sealed tubes

Results: Is the local (B24) ^{14}C background station enough clean?!

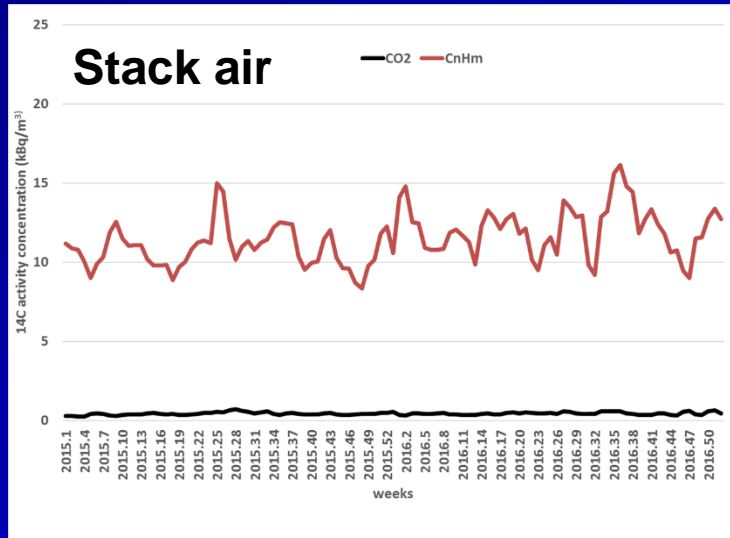


Results: C-14 release of Paks NPP (2015-2016): **stack air ^{14}C data (GBq/week)**

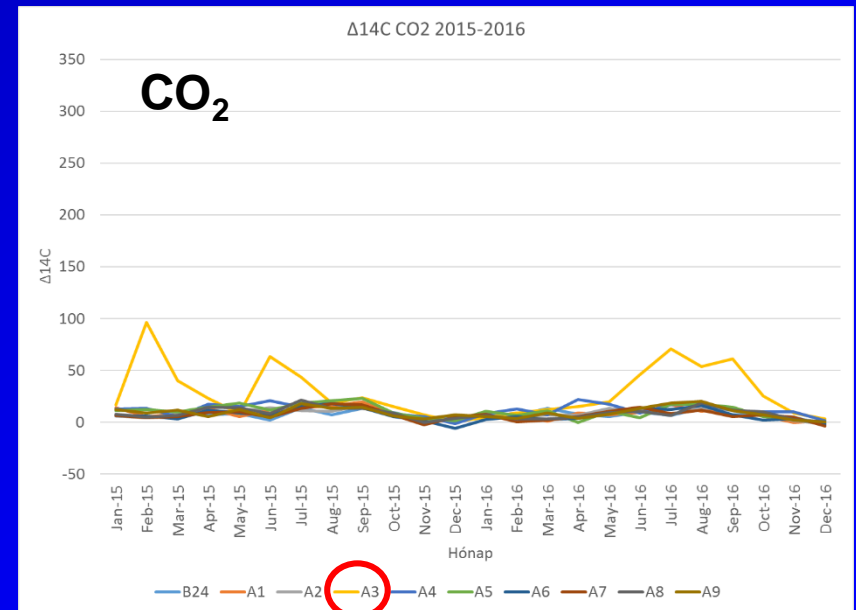
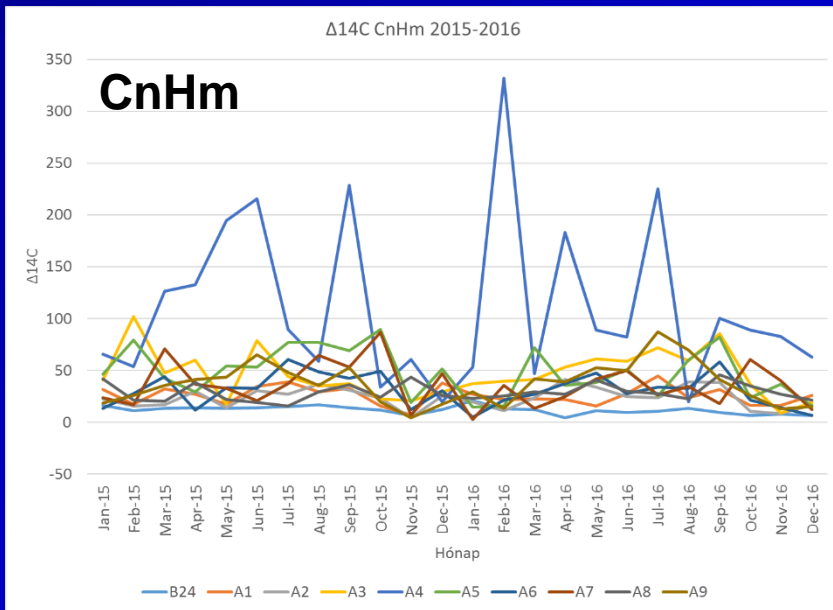


Results: 2 years (2015-2016) monthly atmospheric ^{14}C observations

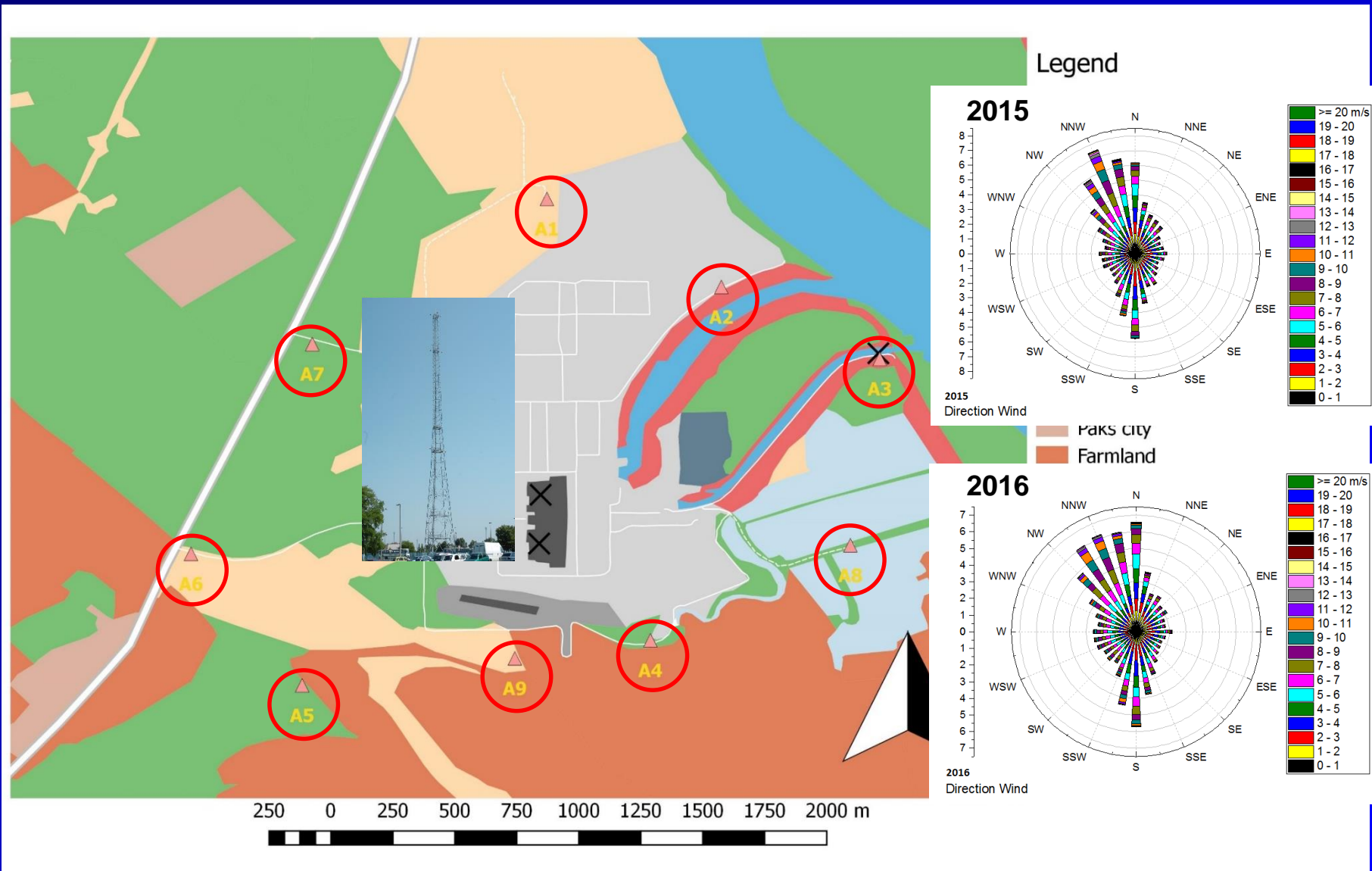
^{14}C in CnHm fraction:
is always higher,
at every station there
is some excess
max is around +35%
more fluctuations
than stack air ^{14}C



^{14}C in CO₂ fraction:
is always lower,
max is around +10%
less fluctuations
than stack air ^{14}C
different from stack air
and CnHm variations

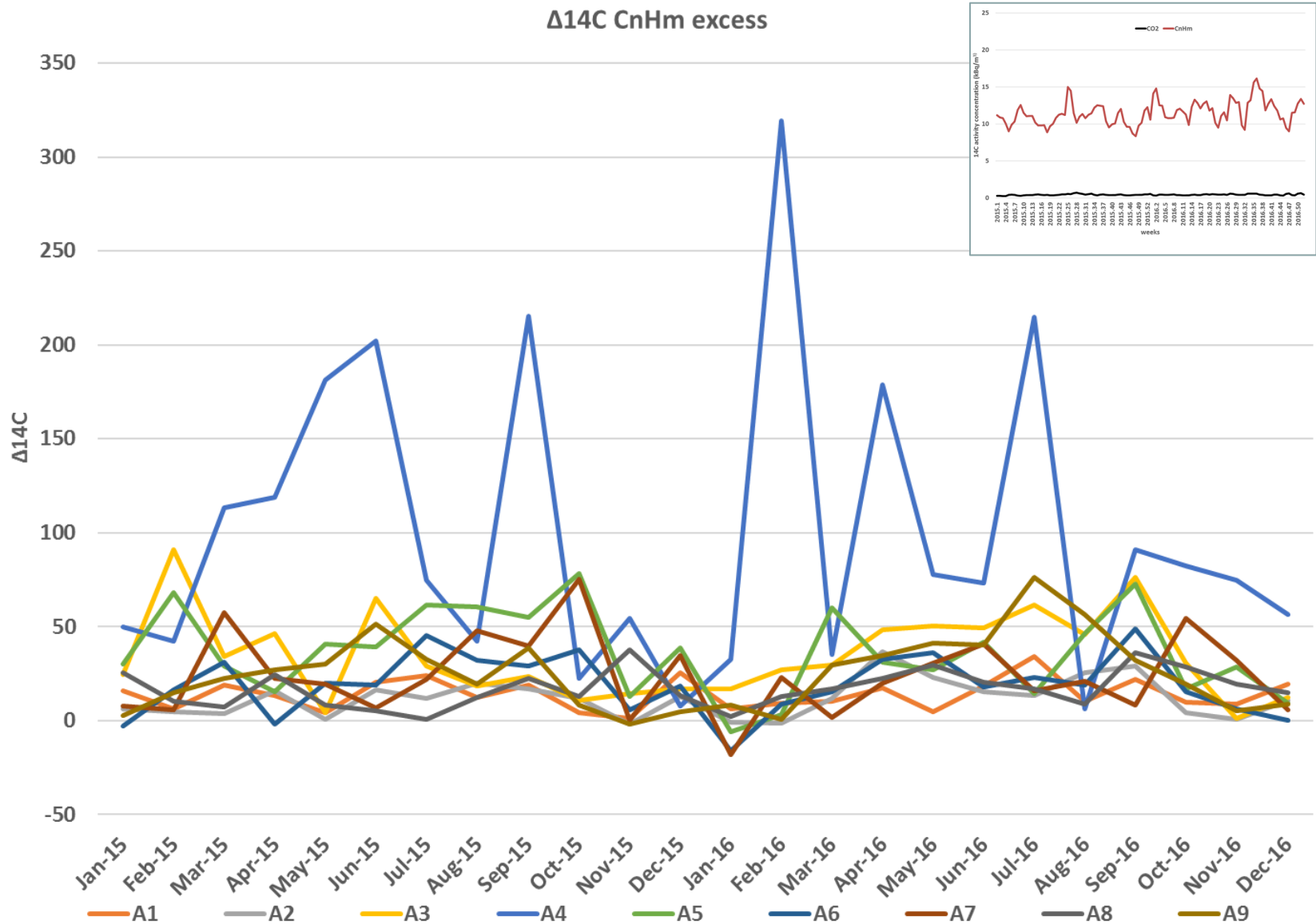


Atmospheric C-14 monitoring network around Paks NPP

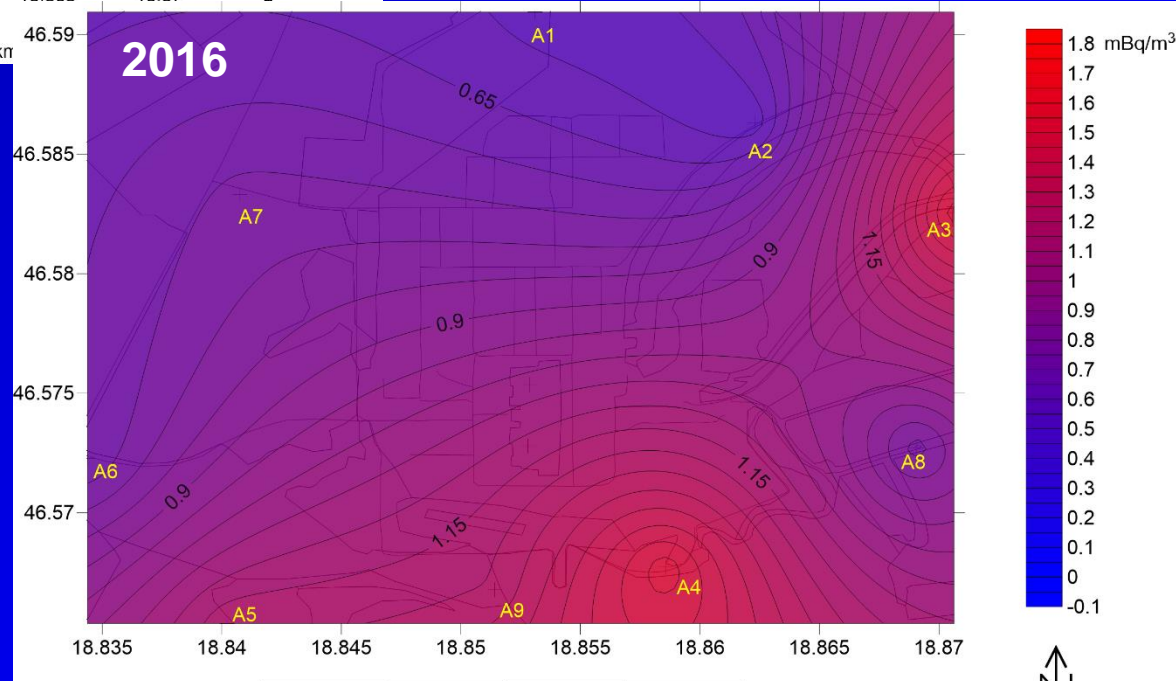
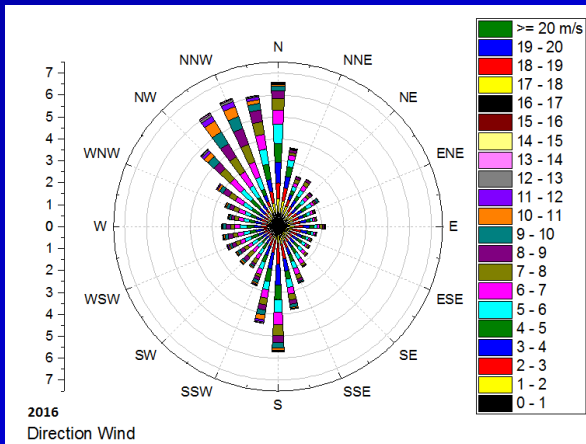
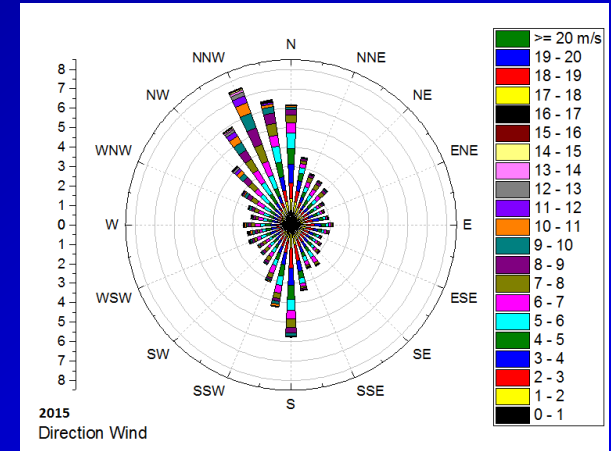
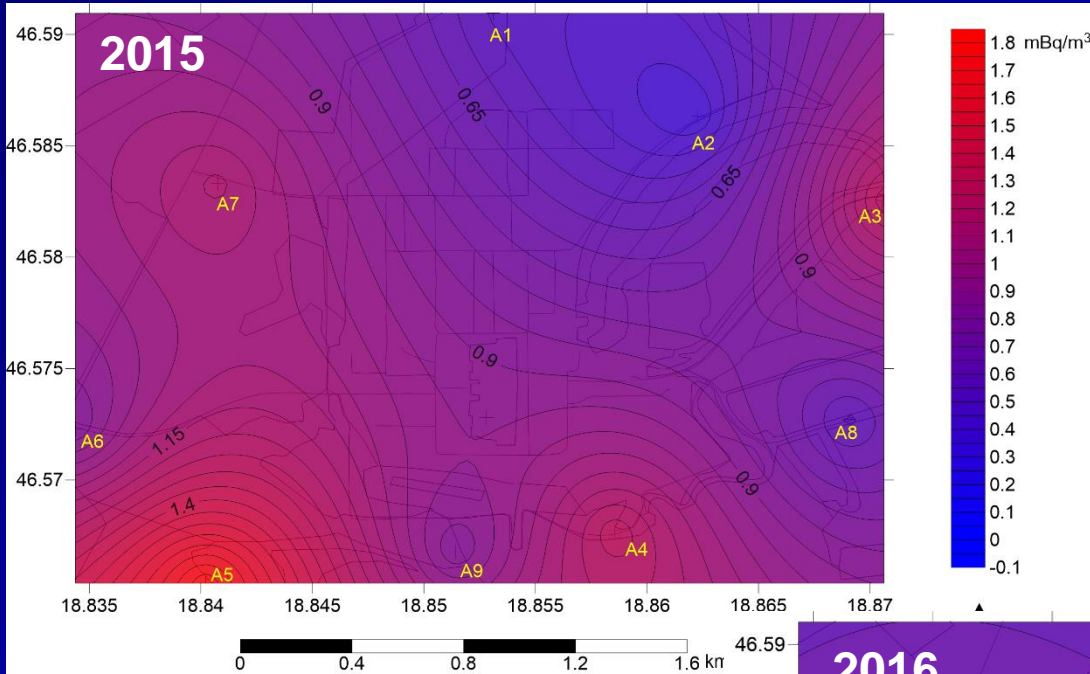


Detailed meteorological data are recorded on different elevations

Results: 2 years (2015-2016) monthly $^{14}\text{CnHm}$ excess observations



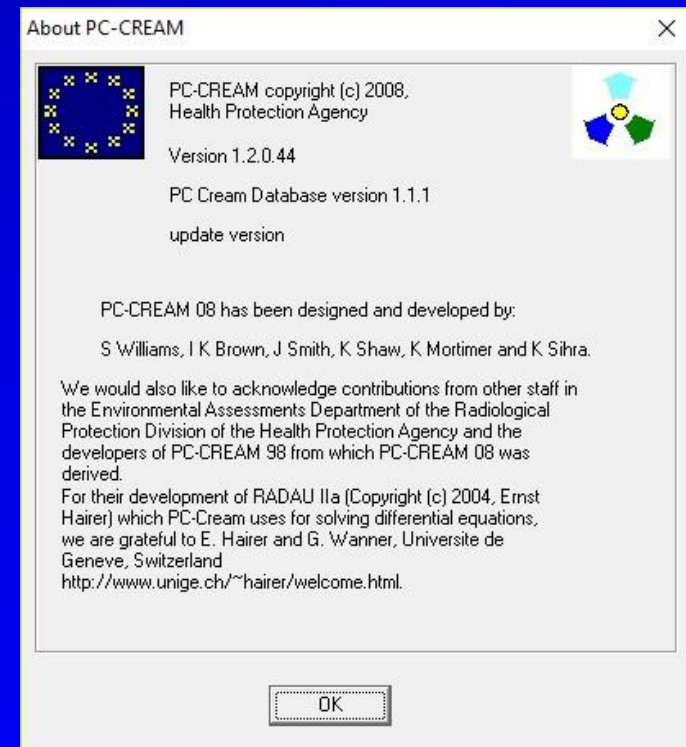
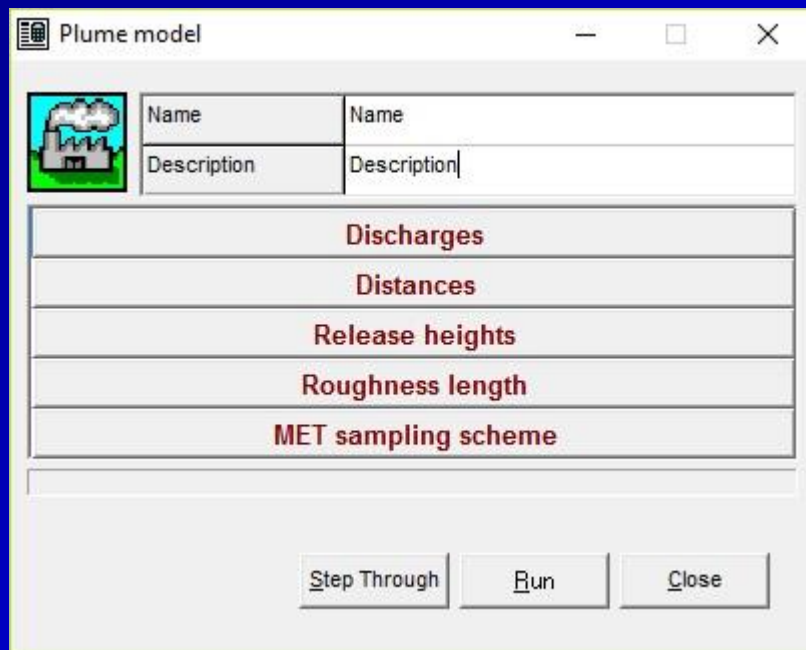
Results: 2 years (2015-2016) $^{14}\text{CnHm}$ fraction observations



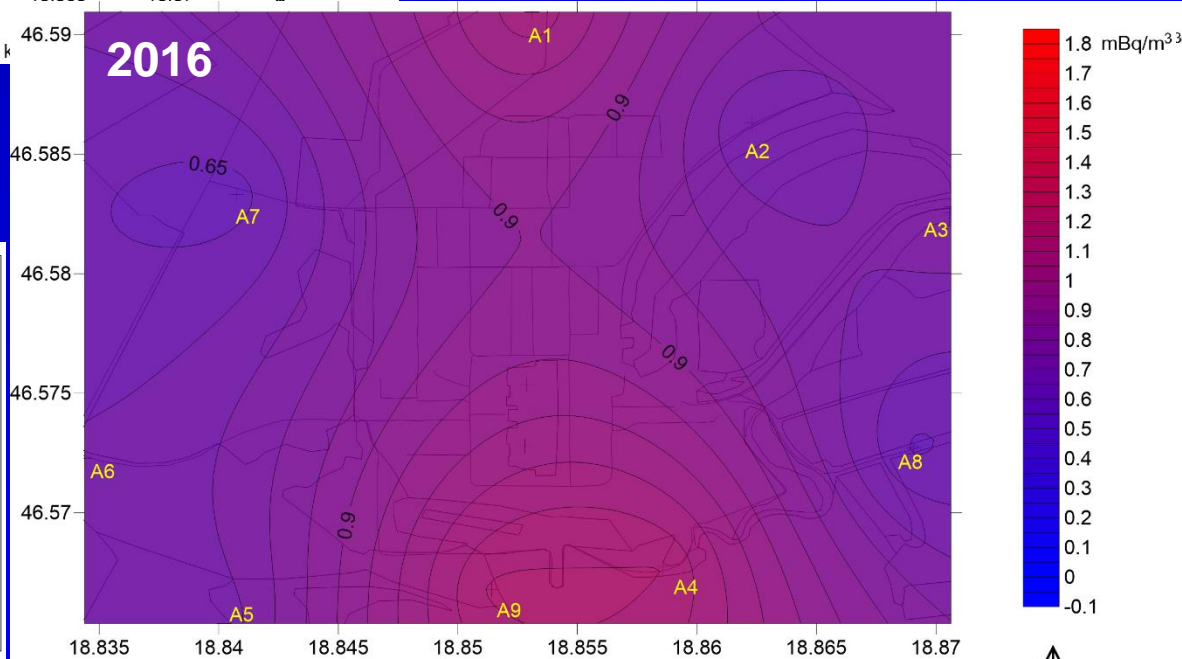
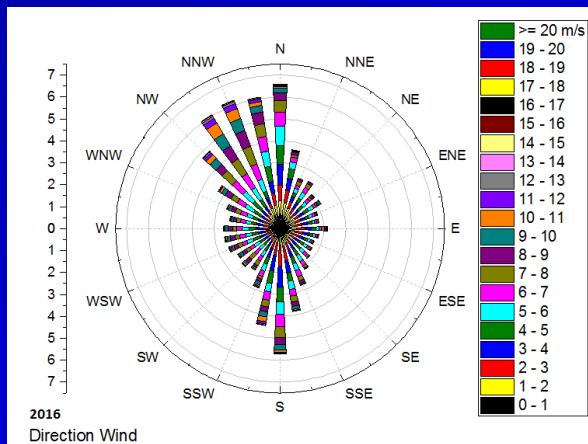
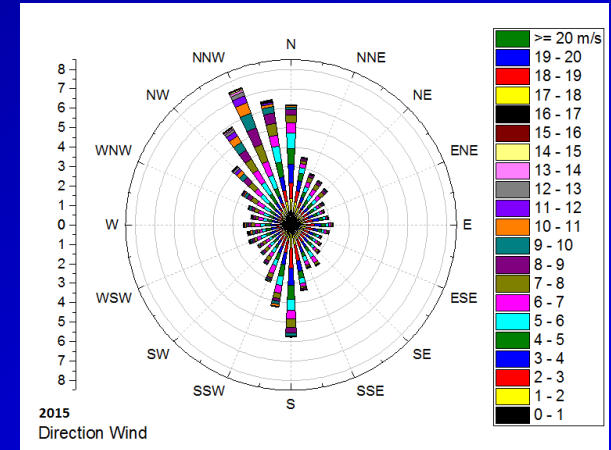
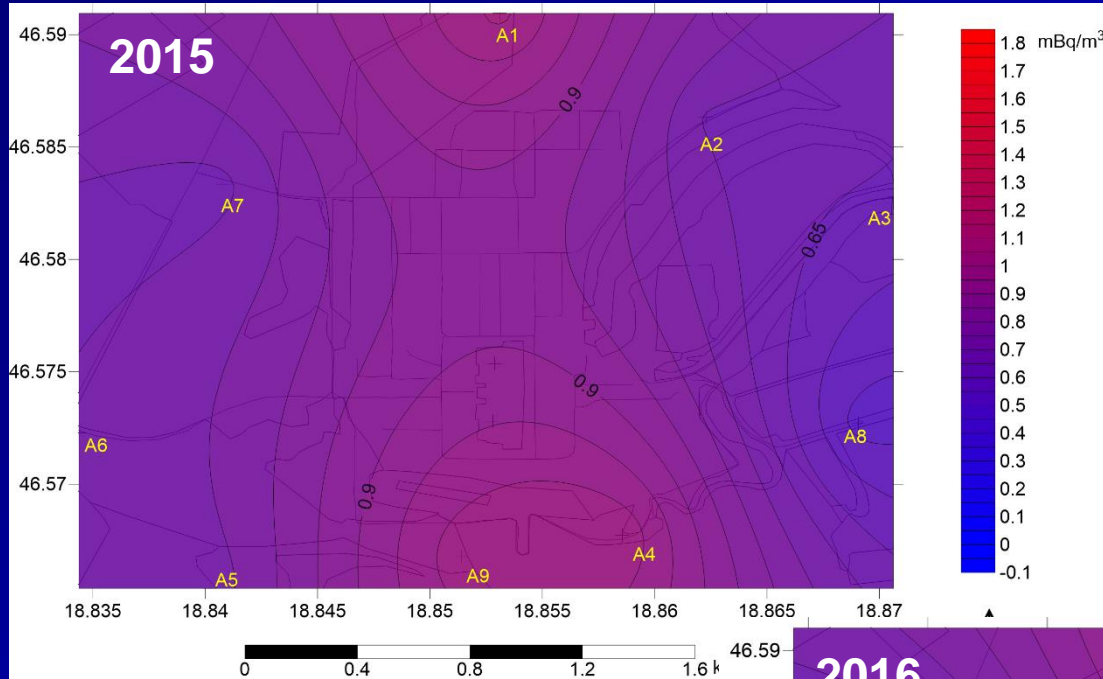
Results: 2 years (2015-2016) $^{14}\text{CnHm}$ fraction modelled

PC-CREAM 08® is an application for performing radiological impact assessments of routine, continuous discharges of radionuclides to the environment. It is used to estimate individual and collective doses arising from discharges of radionuclides to the atmosphere and aquatic environments. It is particularly useful for performing prospective assessments as a key input to discharge authorisations and waste management decisions.

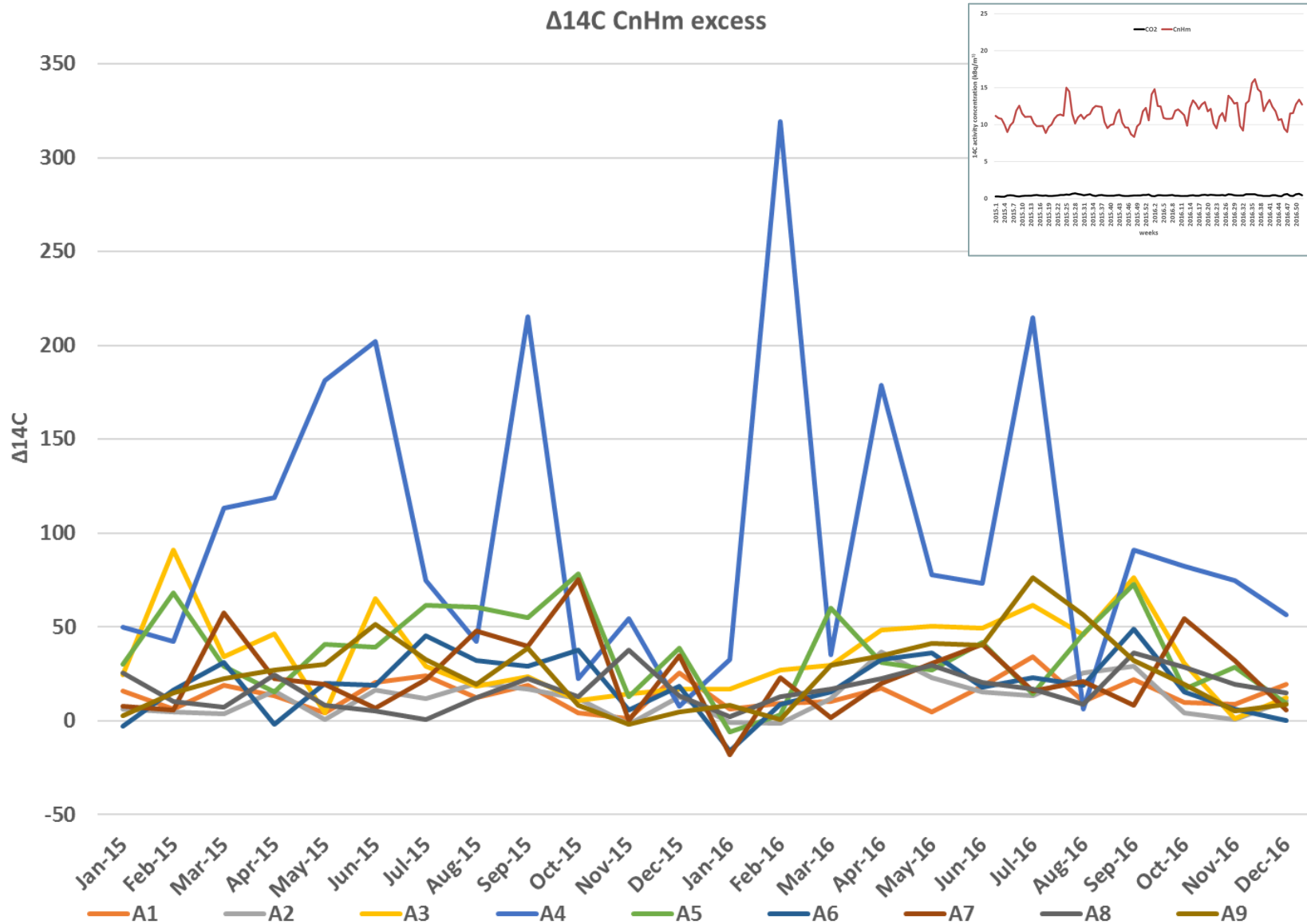
(<https://www.phe-protectionservices.org.uk/pccream/featureoverview/>)



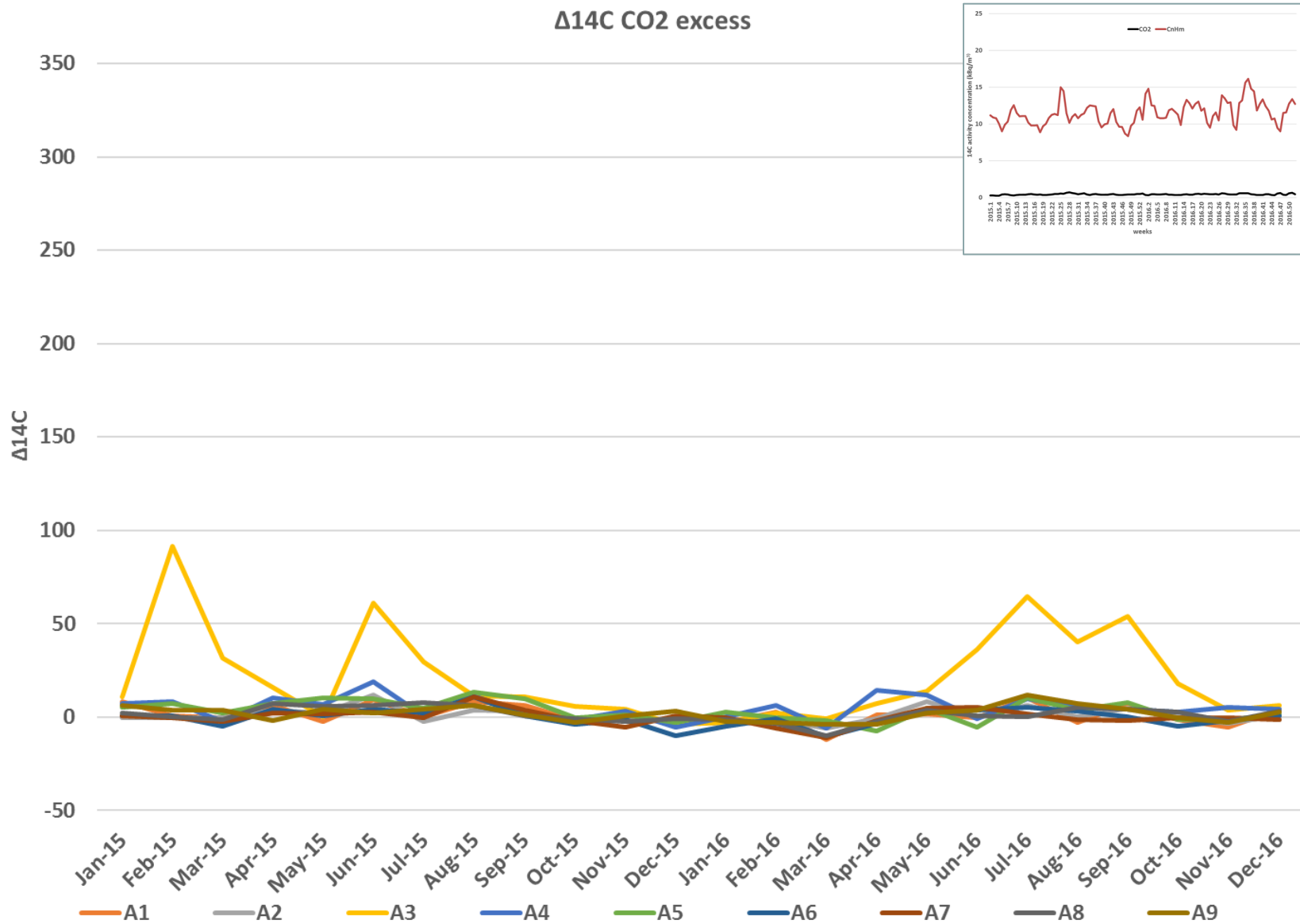
Results: 2 years (2015-2016) $^{14}\text{CnHm}$ fraction modelled



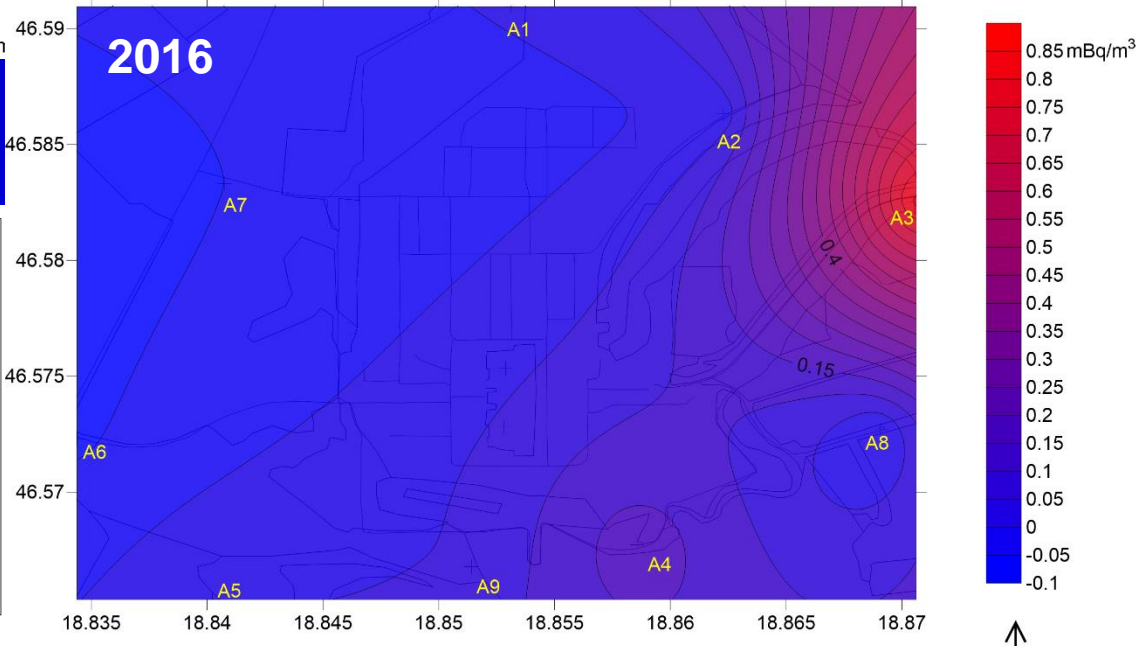
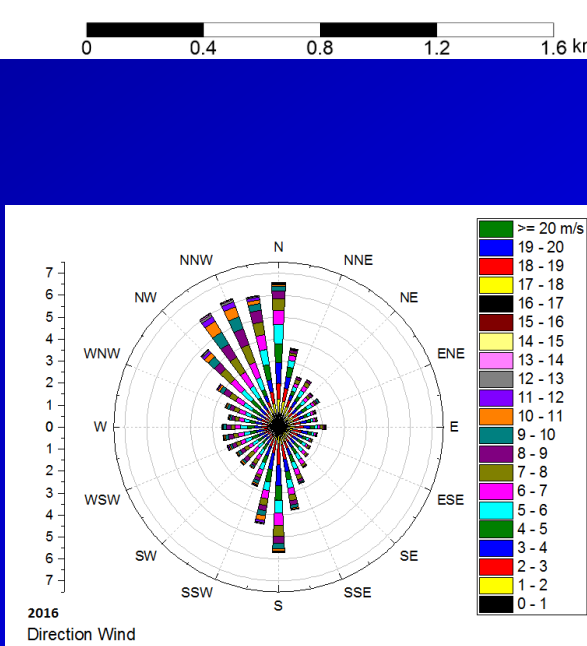
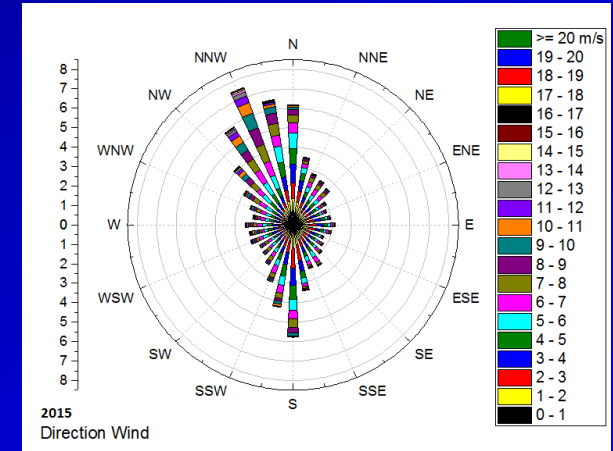
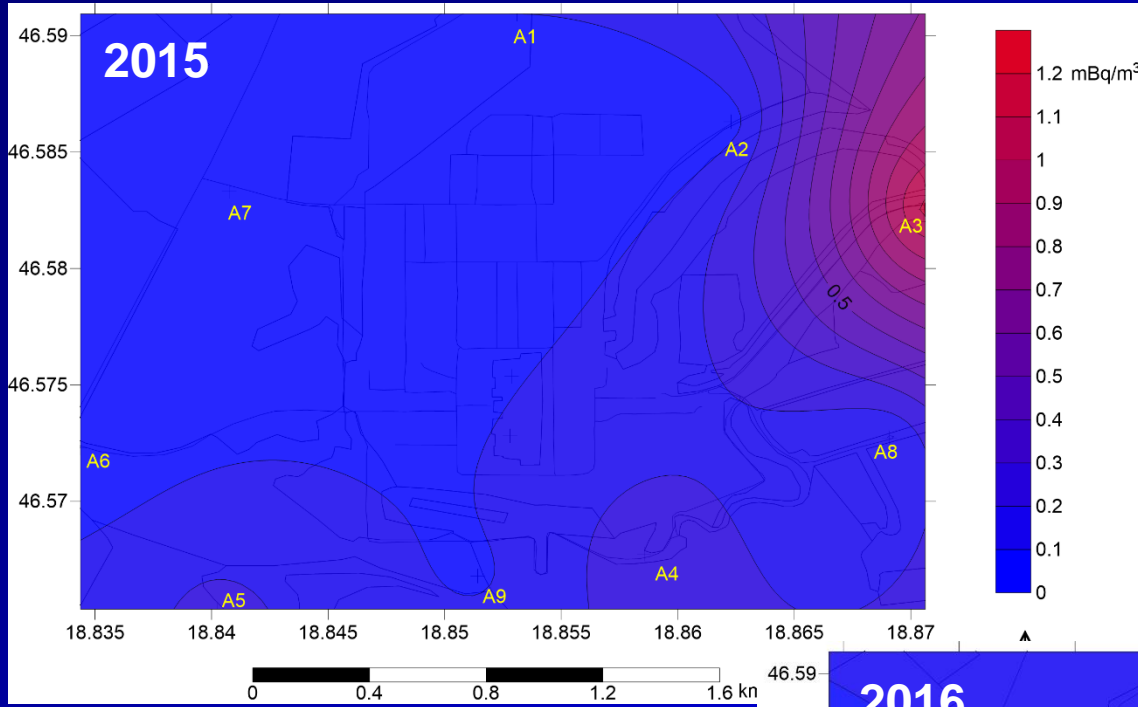
Results: 2 years (2015-2016) monthly $^{14}\text{CO}_2$ excess observations



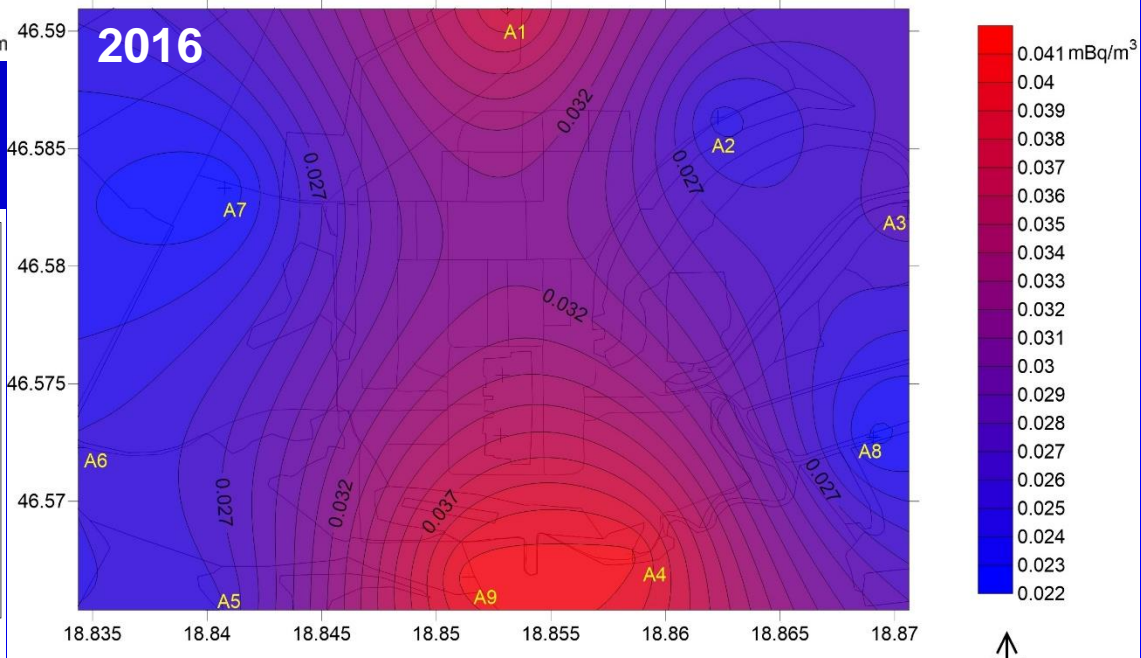
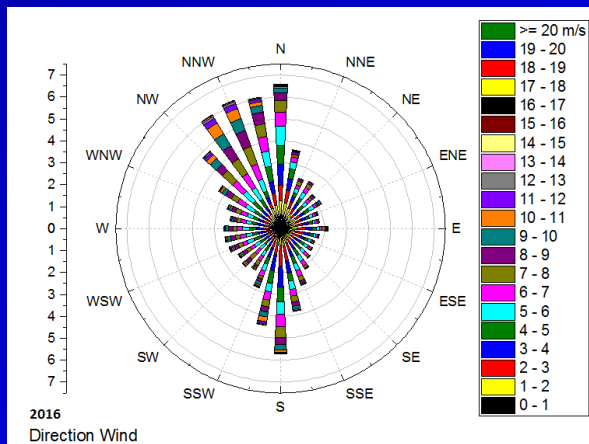
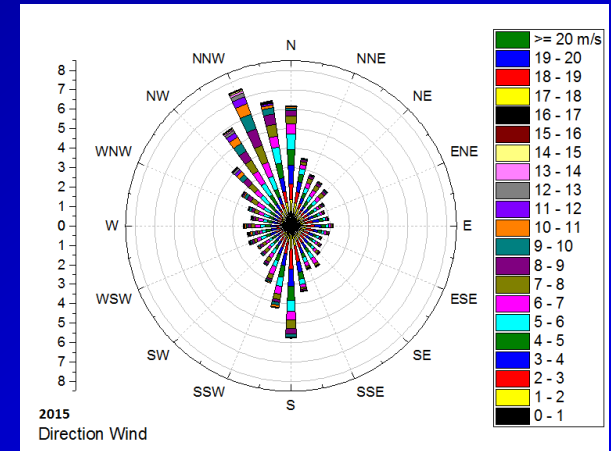
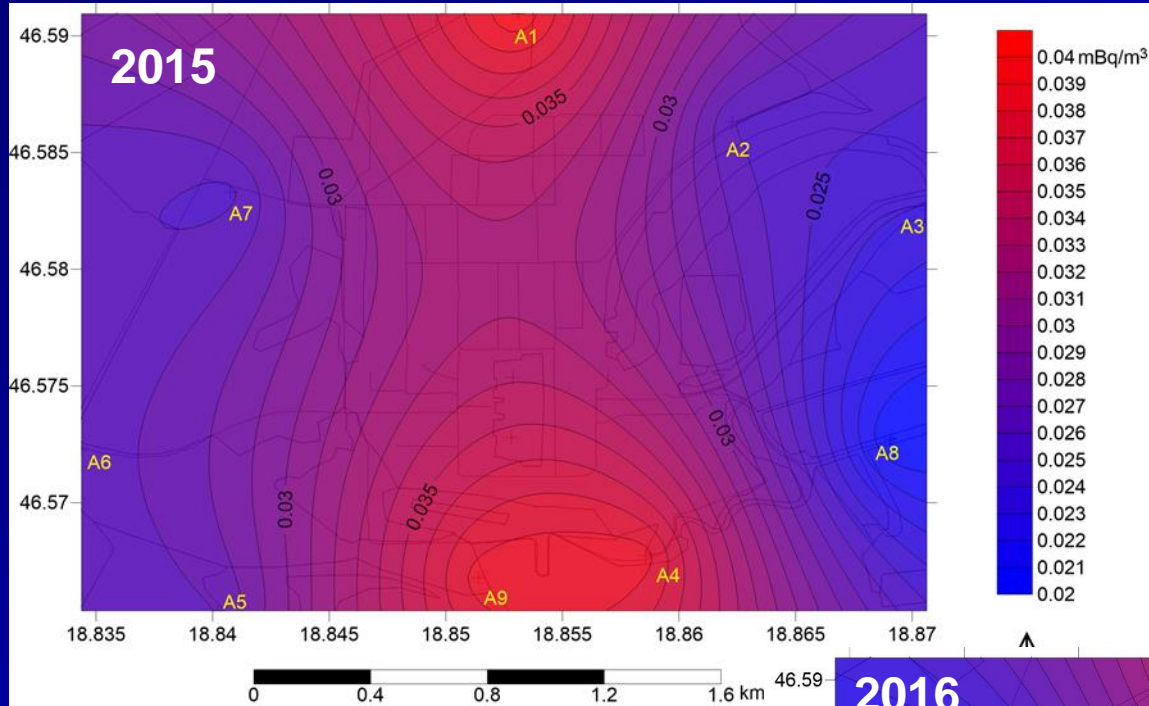
Results: 2 years (2015-2016) monthly $^{14}\text{CO}_2$ excess observations



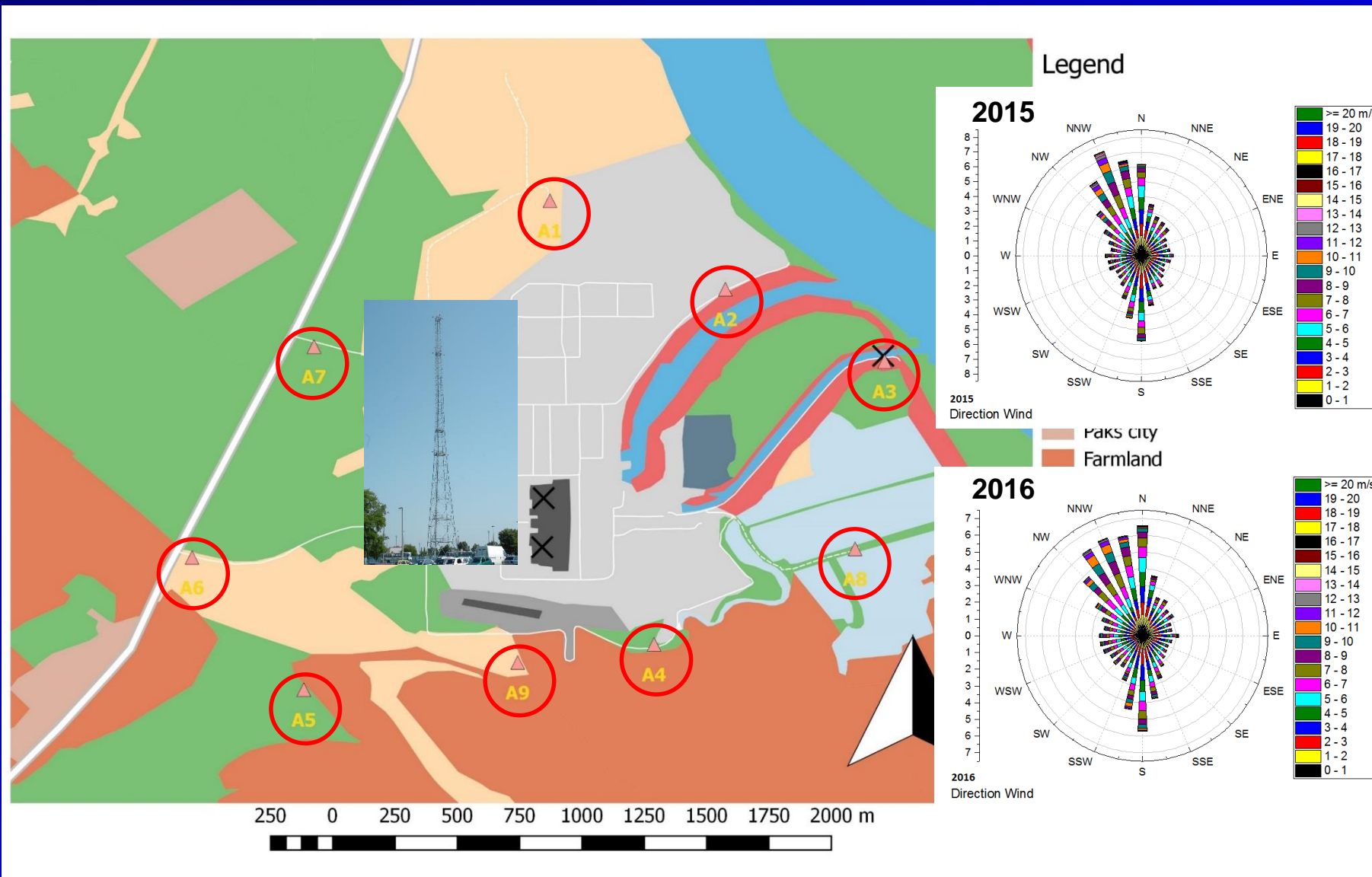
Results: 2 years (2015-2016) $^{14}\text{CO}_2$ fraction observations



Results: 2 years (2015-2016) $^{14}\text{CO}_2$ fraction observations



Results: Why 2 years (2015-2016) $^{14}\text{CO}_2$ observations so high at A3?



Detailed meteorological data are recorded on different elevations

Results: Why 2 years (2015-2016) $^{14}\text{CO}_2$ observations so high at A3?



V3 station: where NPP waste water released to the Danube, after final check

L/ILW waste gas analytical studies
Hertelendi Laboratory, Debrecen, Hungary

Facts & Problems with gas generation in LILW:

Gas is produced from LILW during storage

Produced gas could be combustible (H₂, CH₄)

Produced gas could be radioactive (³H, ¹⁴C etc.)

Chemical forms and radioactivity in the gas is poorly studied

LILW drums/vaults are not hermetically closed for gases

Gases could have a strong effect on the storage conditions (pressure, CO₂)

Inhalation of radioactive gases could be a problem

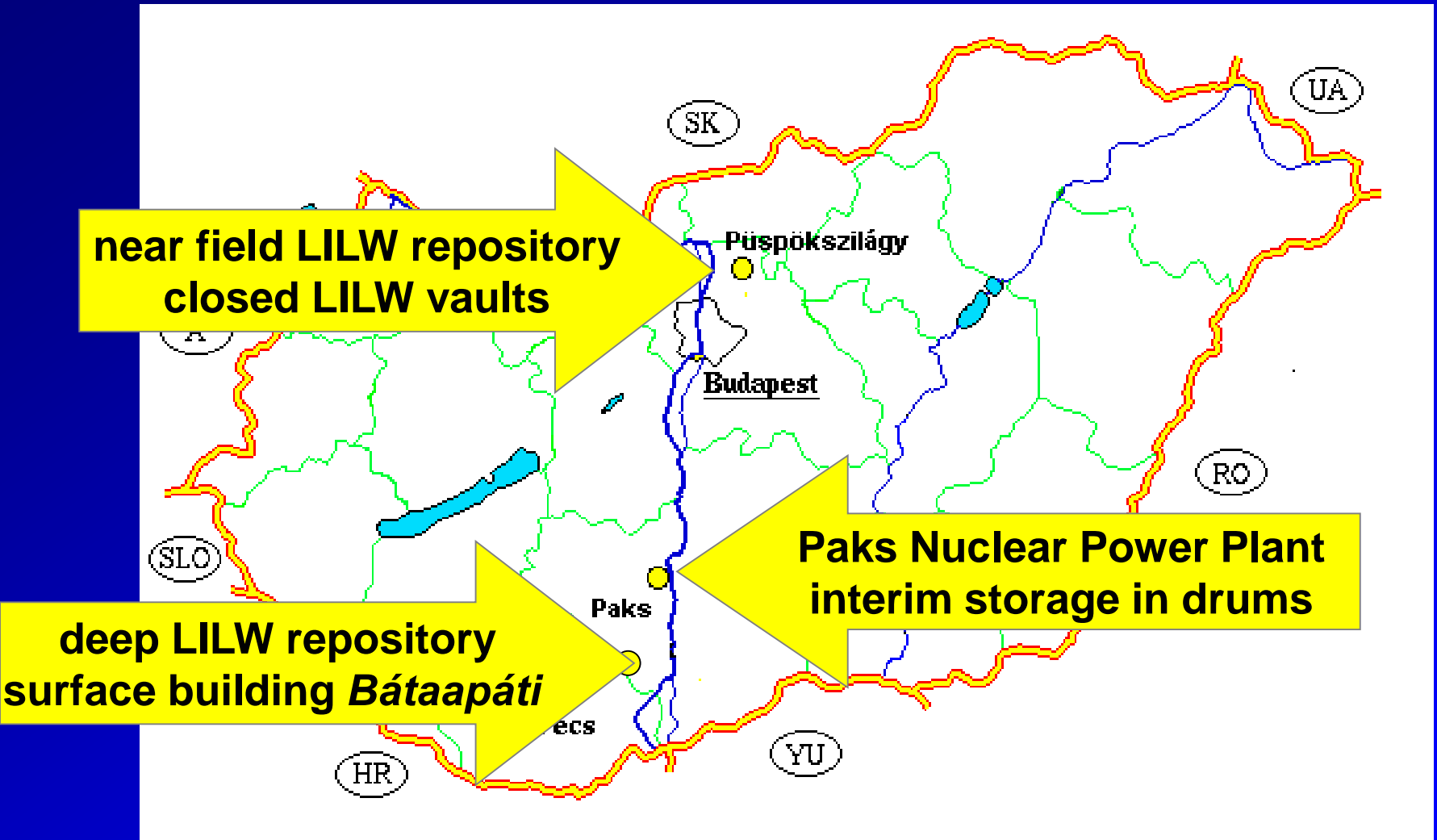
Study performed:

Study of gas generation in LILW drums (since 2000)

Monitoring of radioactive (³H and ¹⁴C) gases around LILW vaults

Gas measurements from vaults after closing (10-30 yrs closure)

LILW gas studies are running
at 3 different locations and 3 different dimensions in Hungary



Main concept of our LILW gas study:

Investigate first in smaller scale: **200 L individual drums**

Use **hermetic overpack** containers to avoid uncontrolled gas lost or air intrusion.

Measure the state parameters of the gas to calculate the produced gas amounts according the Ideal Gas Law: $pV=nRT$

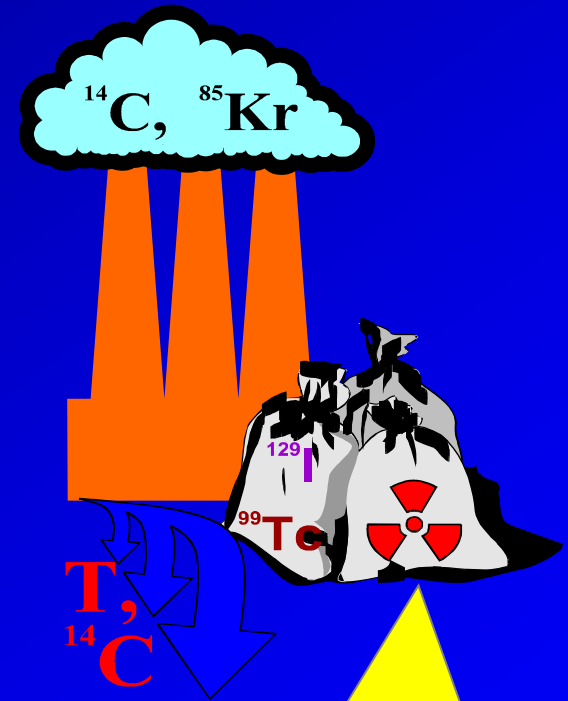
Separate main produced gas components and investigate their H-3 and C-14 contamination level. Calculate gas phase H-3 and C-14 production (release).

Investigate the gas headspace of old LILW **closed vaults** to compare drum results to bigger scale and **real storage conditions** (and help the safety assessment and repacking...)

Paks NPP (WWER-440), Hungary



Released materials



Solid waste of NPP

Not contaminated

L/ILW

HLW (Spent fuel)

**Low and intermediate level radioactive wastes (L/ILW)
must be stored in a repository
(temporarily stored in buildings of NPP)**



Studied LILW drums (from Paks NPP)

Non compacted (N)



debris of building material,
out-of-use tools,
mainly metals

Code **N**

Sludge (S)



liquids comes from
cleaning
(steam generators,
floor in labs and
workshops, etc.)

Code **S**

Compacted (T)

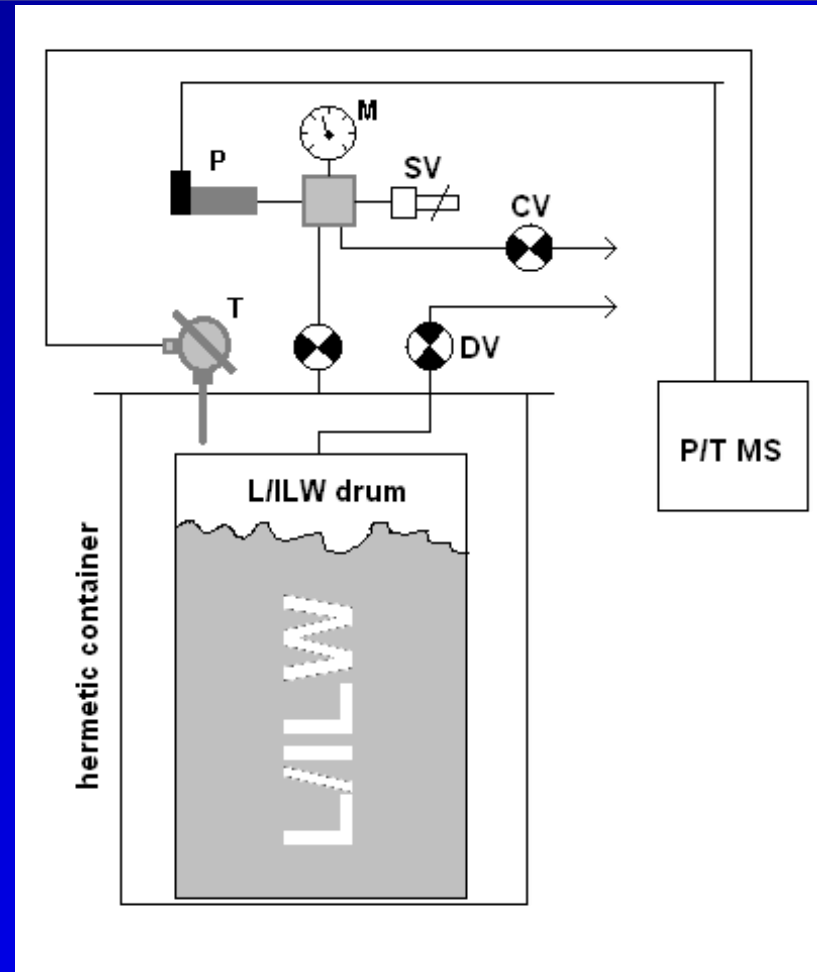


contaminated trash
and scrap, protective
clothes, gloves,
towels, mainly
plastics, textile, and
paper

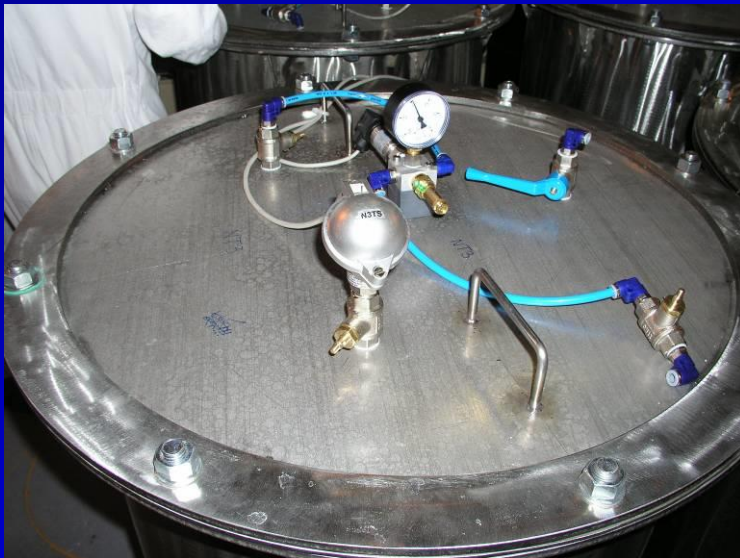
Code **T**

this type of drums are not enough gas tight to help gas studies...

> 50 different drums were studied (since 2000)



Drums closed into hermetic container in 2004



Recorded
pressure, temperature daily (P/T MS)
gas concentrations monthly (QMS)
isotopes in the gas field annually (Lab.)

Gas monitoring, sampling and component analyses

DMS (HEKAL)



p, T

QMS (Pfeiffer)



Gas MS



Gas prep.

Separation line (HEKAL)



$^{14}\text{C}/^3\text{H}$
in air

field sampling (HEKAL)

Isotope analytical measurements of gases in lab of HEKAL

AMS (MICADAS)

^{14}C



Delta XP^{plus} (CO₂) (Finnigan)

$\delta^{13}\text{C}$



^3H

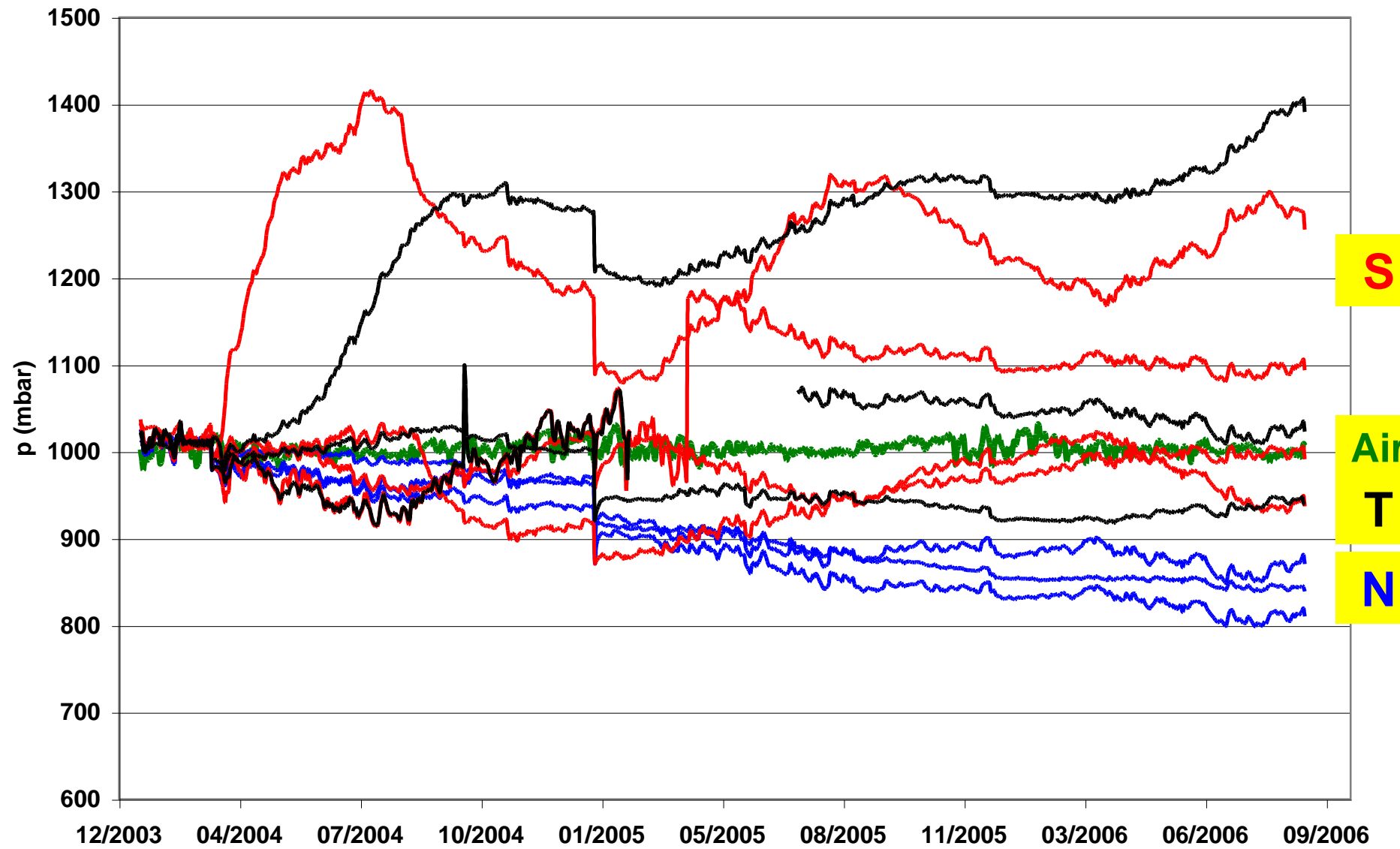
LSC (H₂O) (QUANTULUS1220)



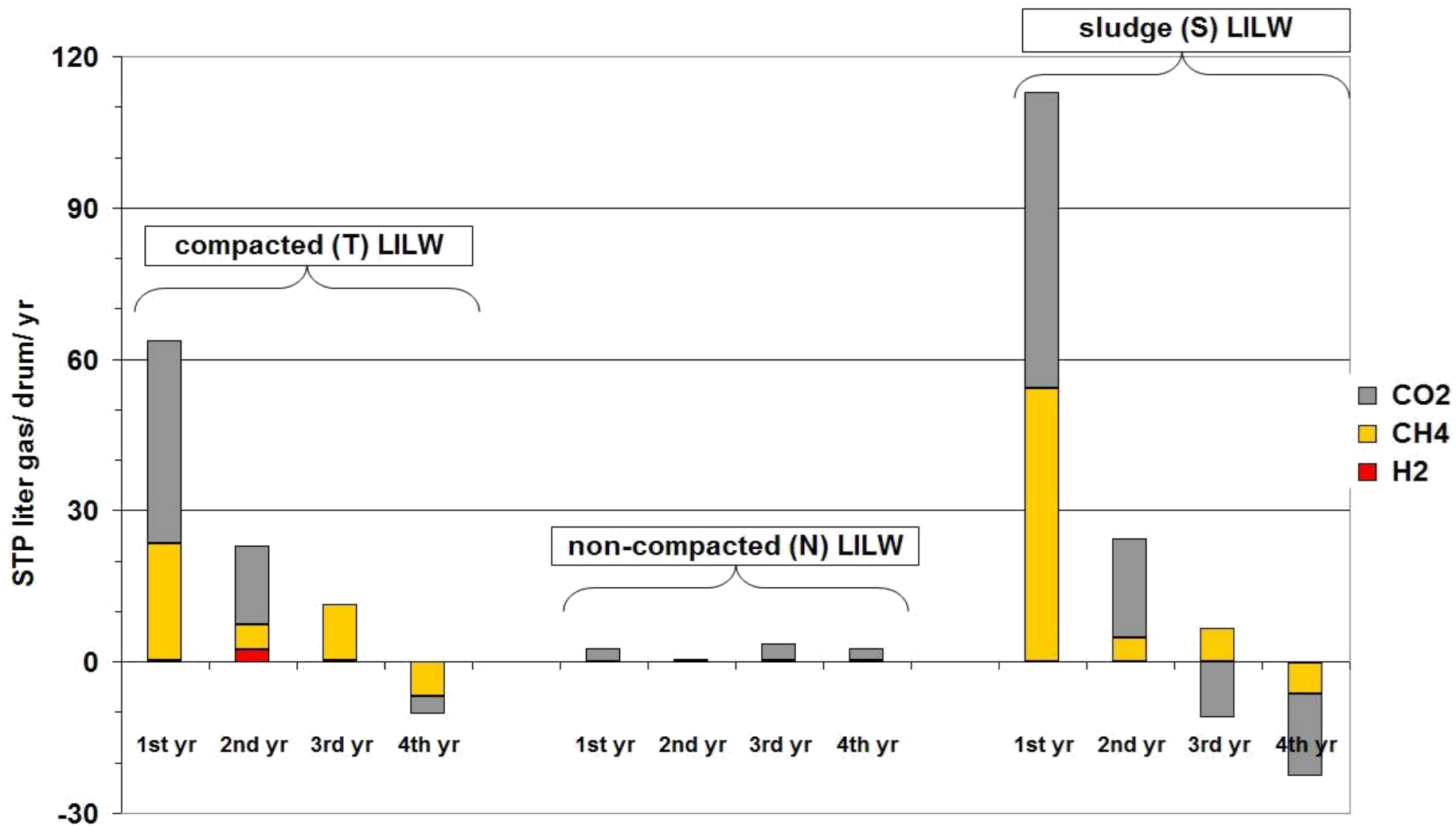
$\delta^3\text{He}$
He cc.

VG5400 (Fisons Instruments)

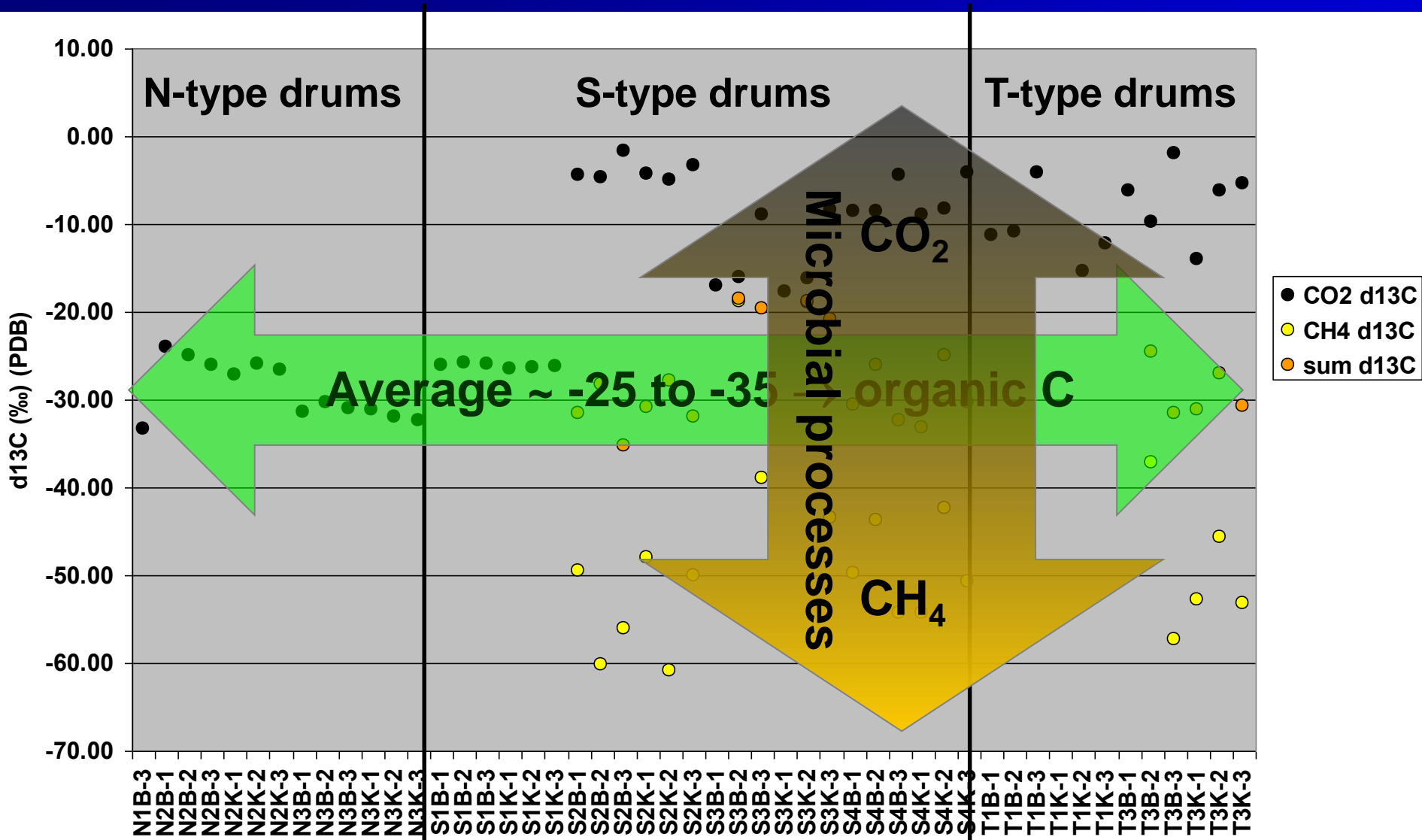
Normalised gas pressure in L/ILW drums (*T and vapor corrected*)



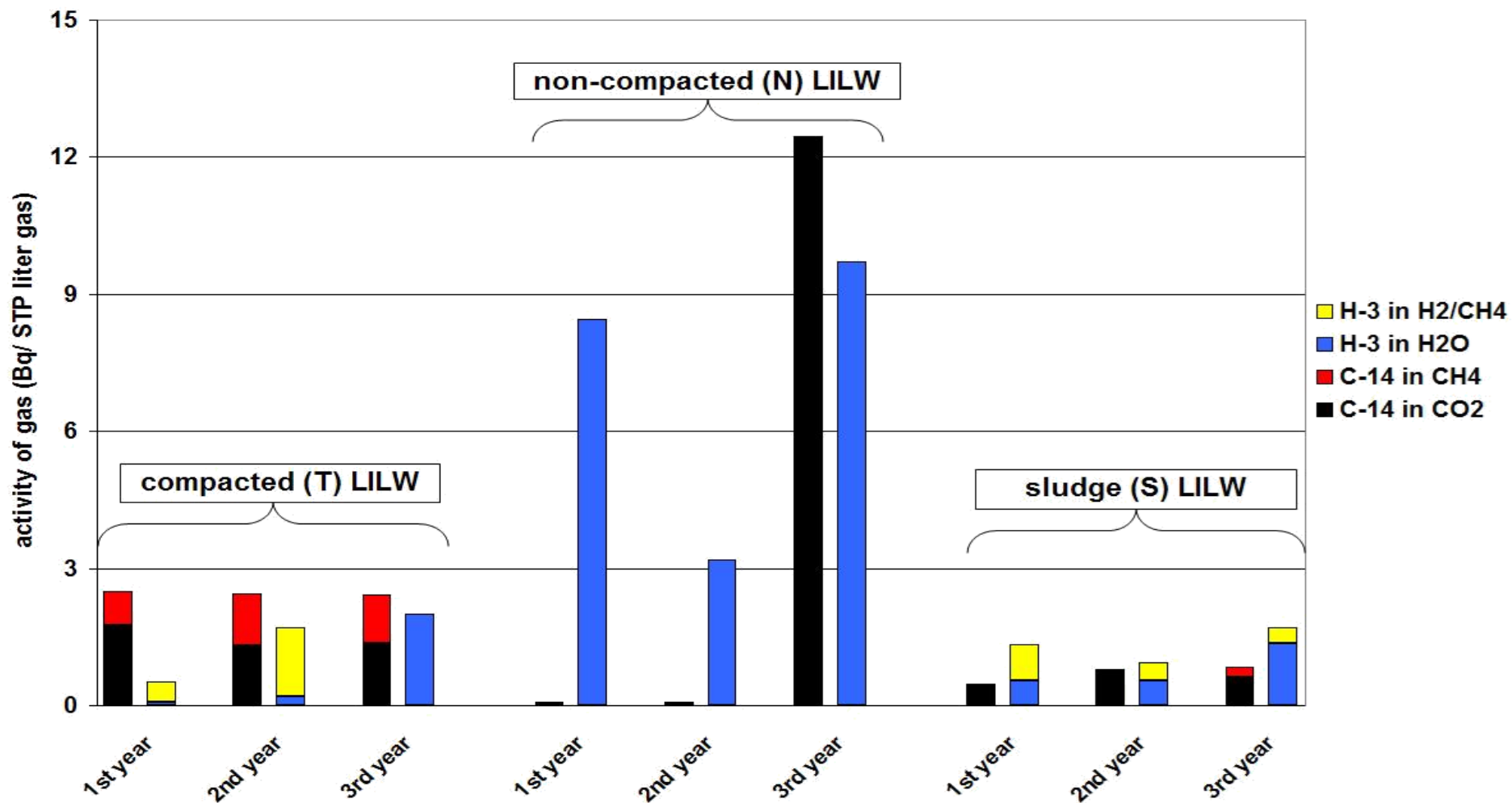
Main produced gases: H₂, CH₄ and CO₂



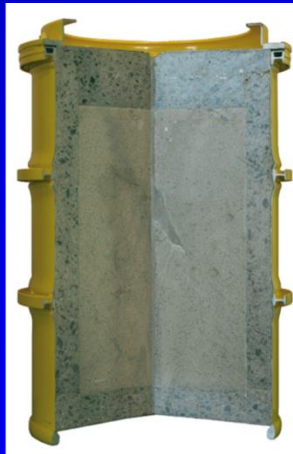
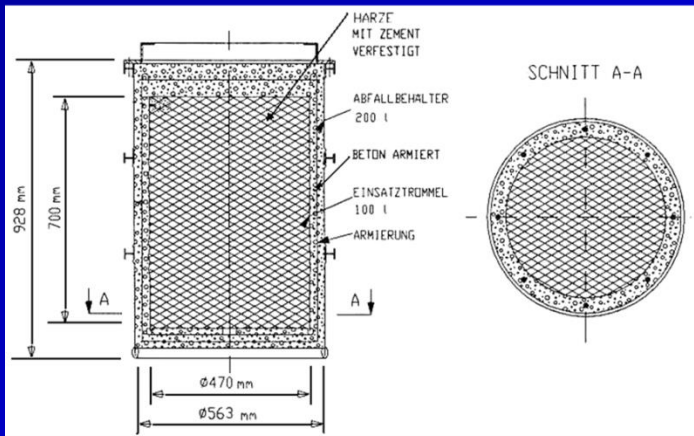
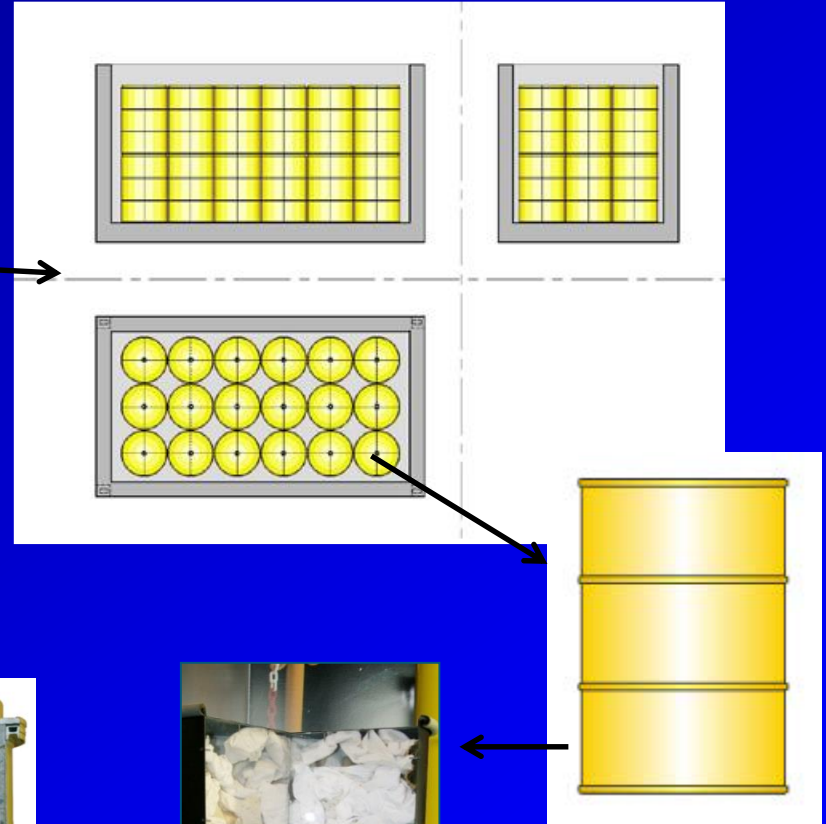
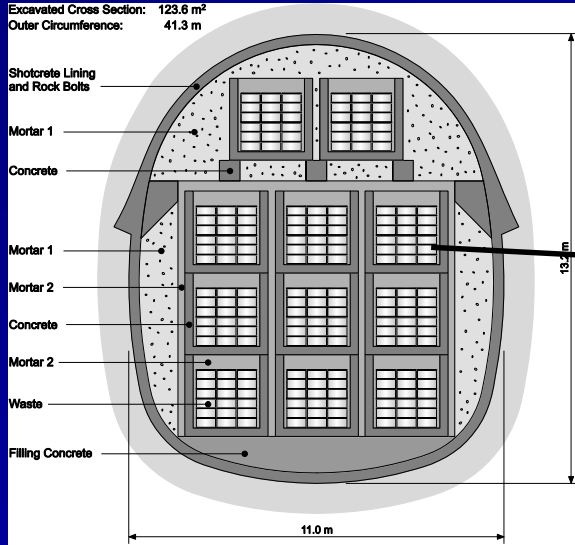
^{13}C isotope results from headspace-gases of LILW drums



^3H and ^{14}C activity conc. higher than in air by 5-6 orders of magnitude

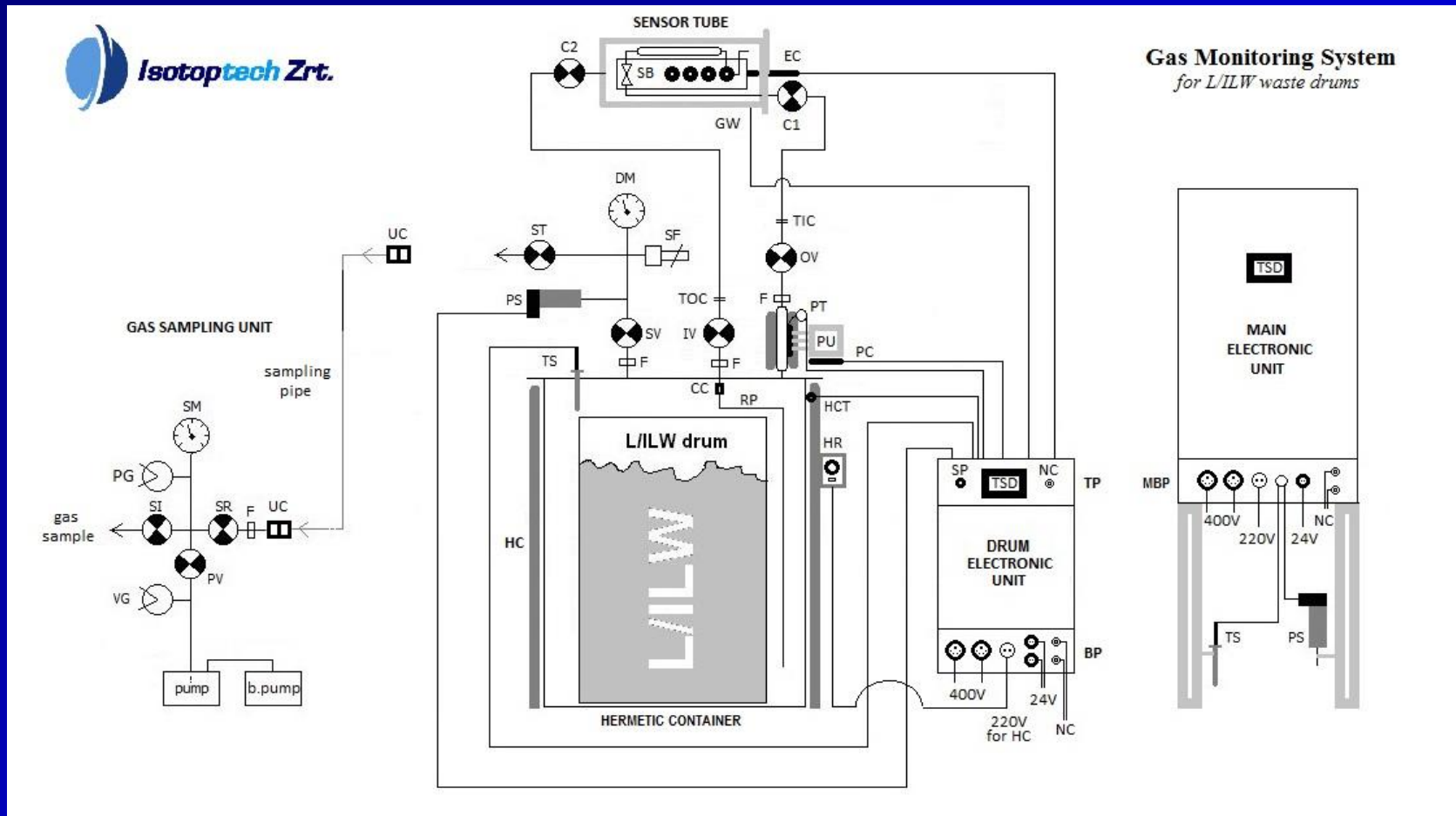


LILW management in Switzerland



Waste matrix where micro organisms live and produce gas emission

Gas Monitoring System (Isotoptech) - Layout



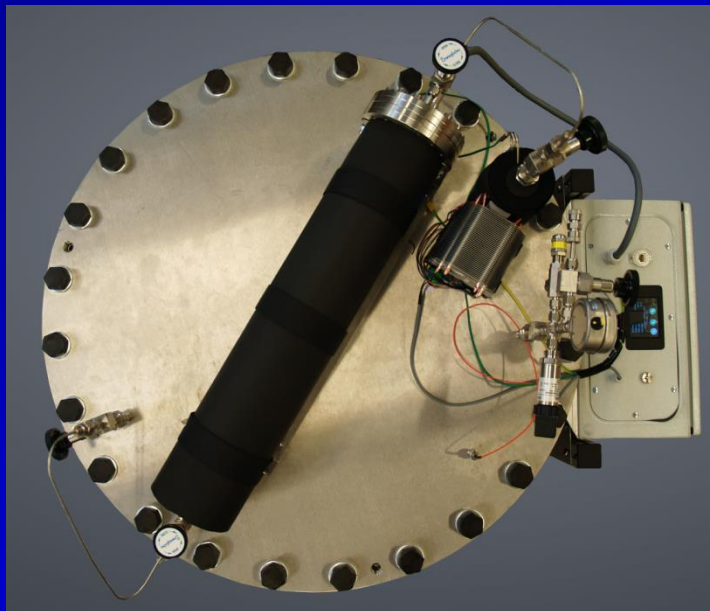
Requirements:

Inert/SS components, gastight, heating (up to 50 °C), gas sampling option, on-line gas temp/pressure/main components monitoring (H₂O, CO₂, CH₄, O₂), remote control and data transfer

Gas Monitoring System (Isotoptech) - Design

Elements:

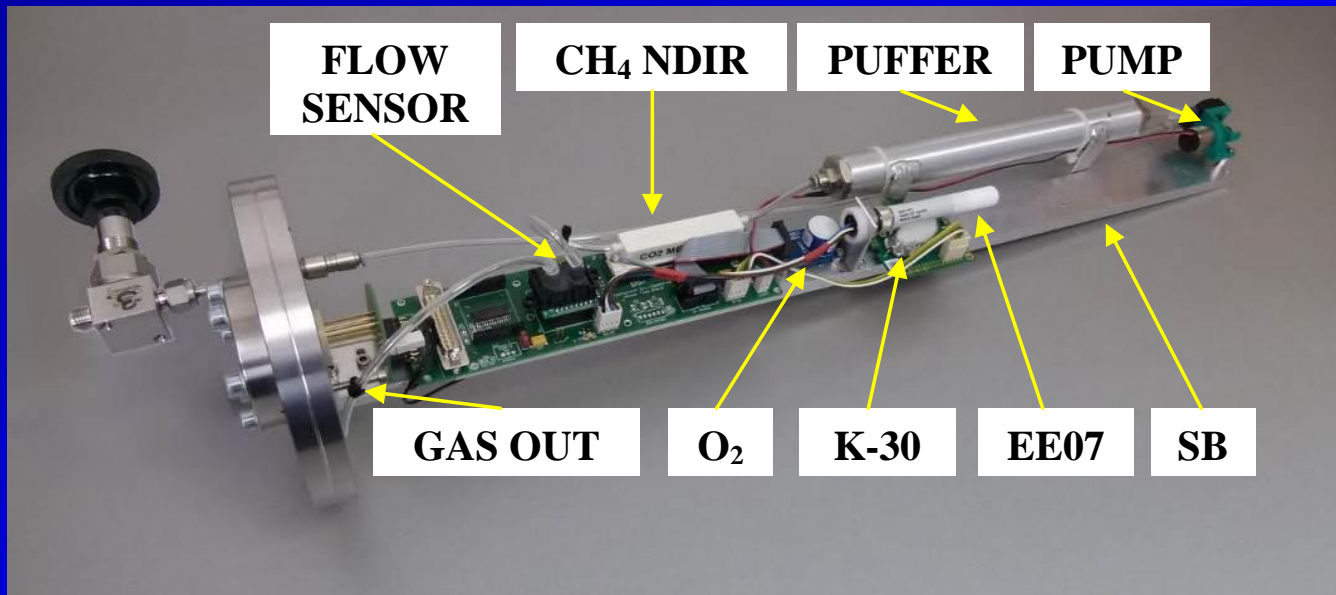
- Gastight tank
- Gas Sensor Tube
- Electronic Unit with Display
- Main Electronic Unit
- Gas Sampling Unit



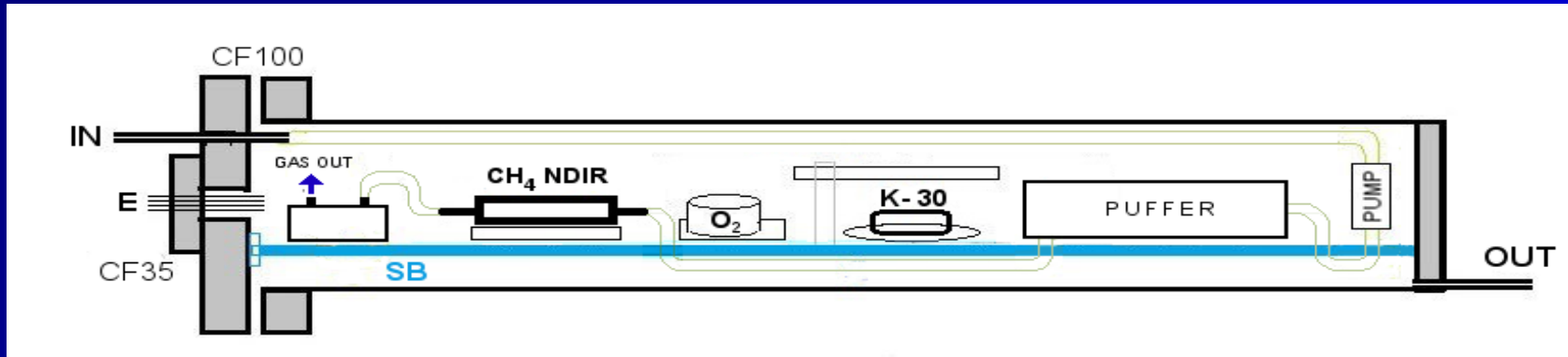
On-line in-situ gas analyses at NAGRA site

Gas specific sensors applied:

- K-30 sensor for CO₂ (0,01 – 1,0%)
- MH-Z92 CH₄ / CO₂ sensor (0,1%-100%)
- UV Flux Oxygen sensor (0-30 %)
- Temperature and humidity sensor
- Gas flow sensor (0-2000 ml/min)



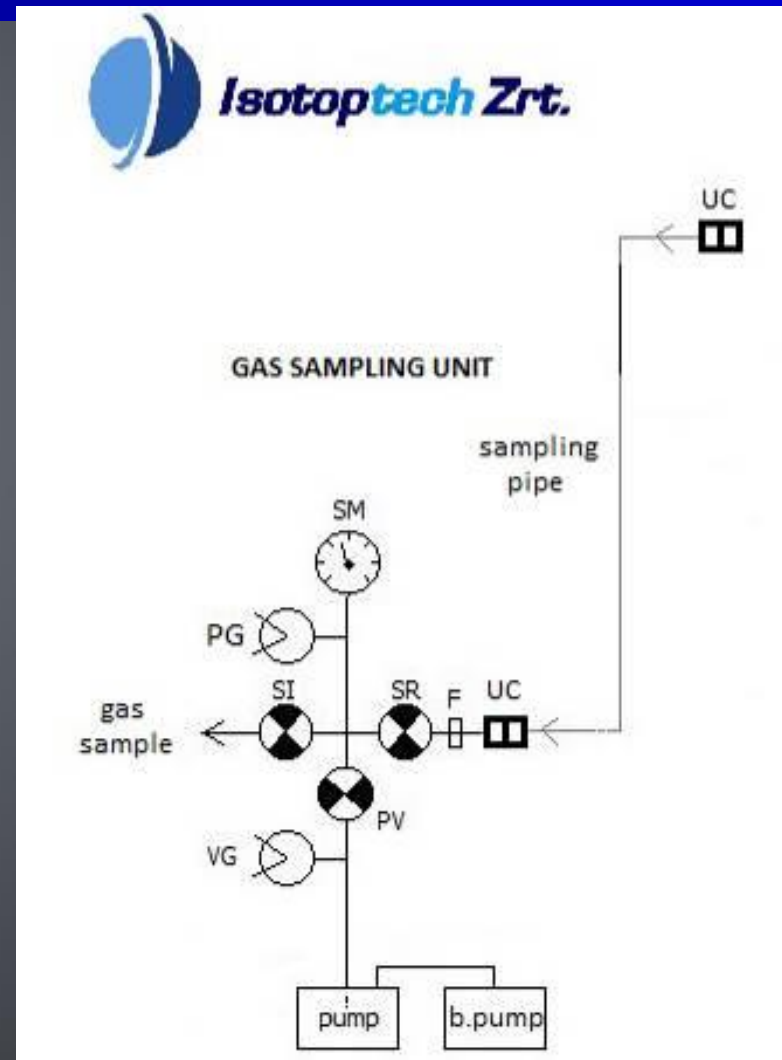
Concept



- Gas circulation between the tank and Gas Sensor Tube
- Gas flow is particle filtered, measured and returned back to the Tank
- Gas components are measured in the Sensor Tube and displayed/stored
- A Peltier Cooling Head is applied to keep humidity low for Gas Sensors



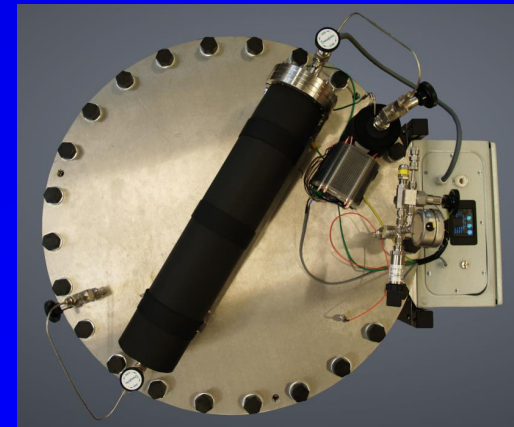
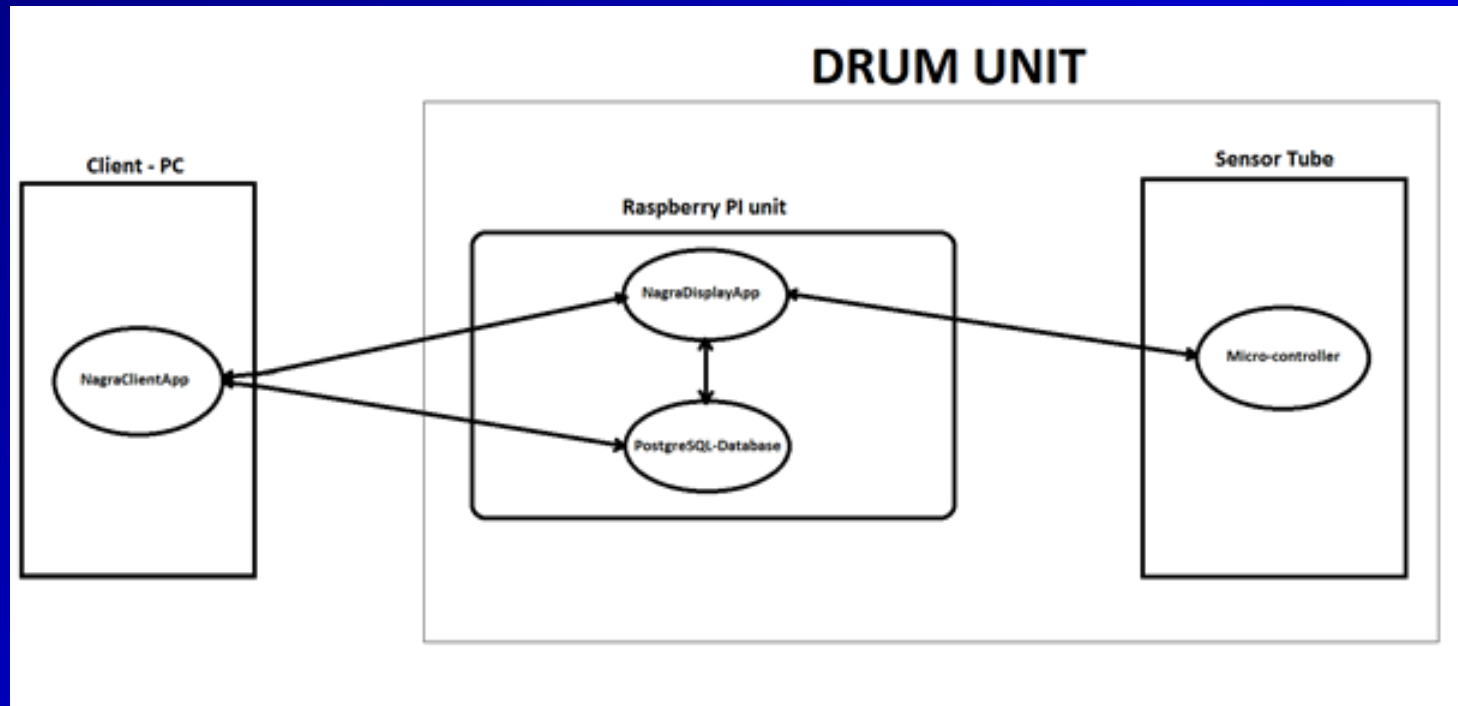
Gas Sampling Unit – 1L gas into SS bulbs



Drivers, Data Storage, Remote Control

Main tasks:

- Drivers for Sensor running/calibration / maintenance
- Database storage/handling
- On-line data / communication
- Data display/handling



Data base handling













12 different data/parameter is stored/handled for each Tank,
Calibration of sensors, storage of calibration data
Remote control and data transfer/ data visualization

NAGRA Drum Monitoring System --- Client Application

File Serial Operation Mode Calibration Database Help

Measurement protocols
Edit Actual Protocol List
Manage Measurement Protocols

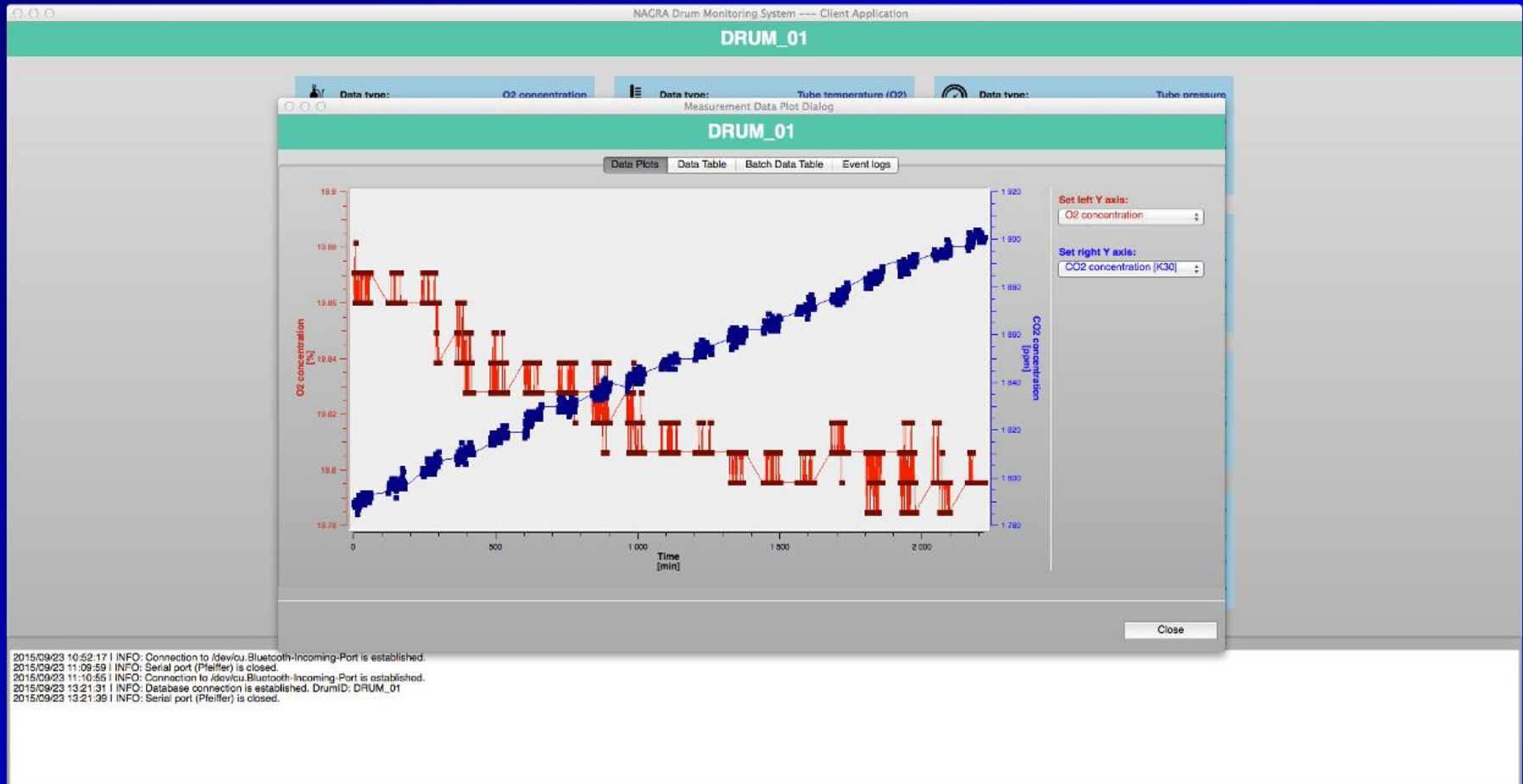
DRUM_01

 Data type: O2 concentration First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9353	 Data type: Tube temperature (O2) First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9353	 Data type: Tube pressure First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9353
 Data type: CO2 concentration (K30) First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9473	 Data type: Tube temperature (EE) First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9467	 Data type: Tube humidity First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9457
 Data type: CO2 concentration (MHZ) First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9468	 Data type: Drum temperature First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9480	 Data type: Tube flowrate First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9461
 Data type: CH4 concentration First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9464	 Data type: Heater temperature First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9463	 Data type: Drum pressure First data: 2015/09/13 12:00 Last data: 2015/09/29 12:59 Data number: 9480

Event log

Data base handling

12 different data/parameter is stored/handled for each Tank,
Calibration of sensors, storage of calibration data
Remote control and data transfer/ data visualization



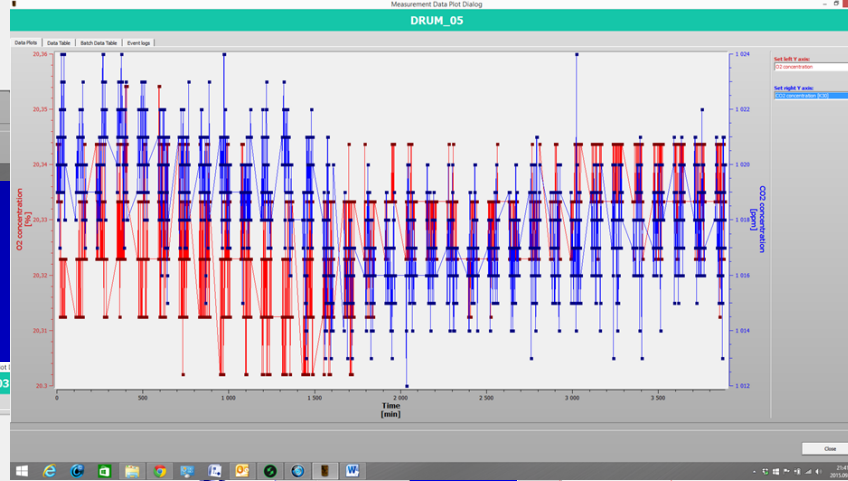
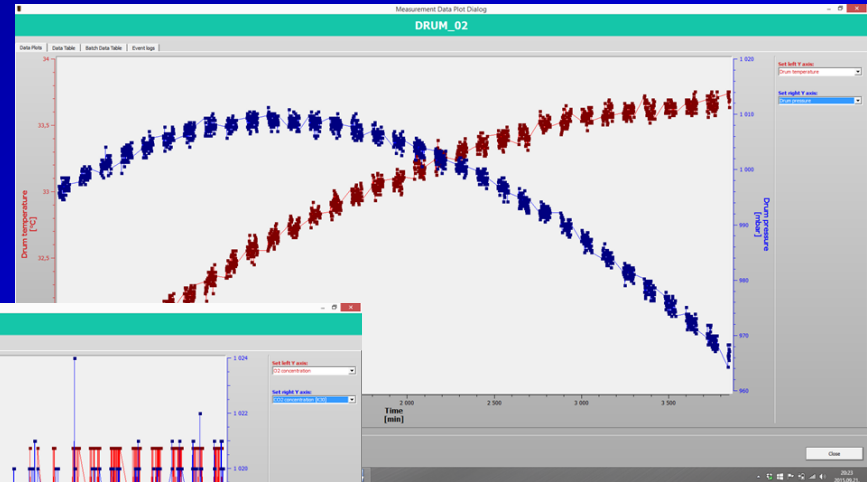
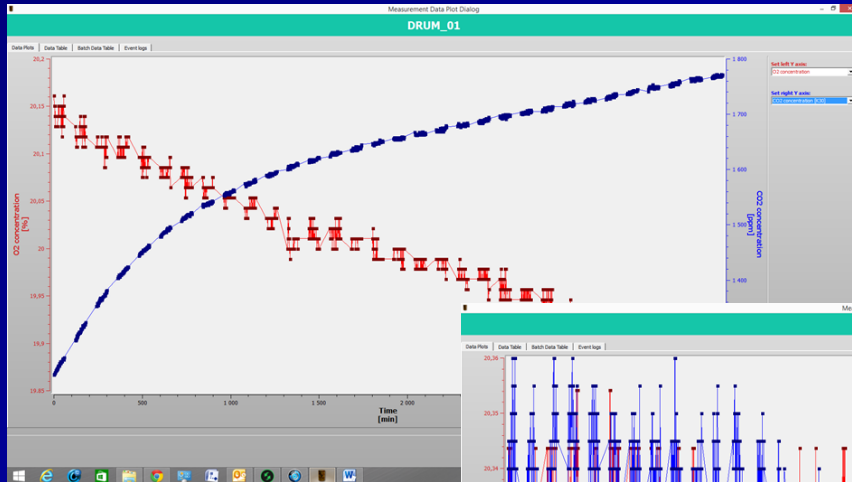
Installation

It took 3 days on site (2015.09.16-19.) Zwilag, Switzerland

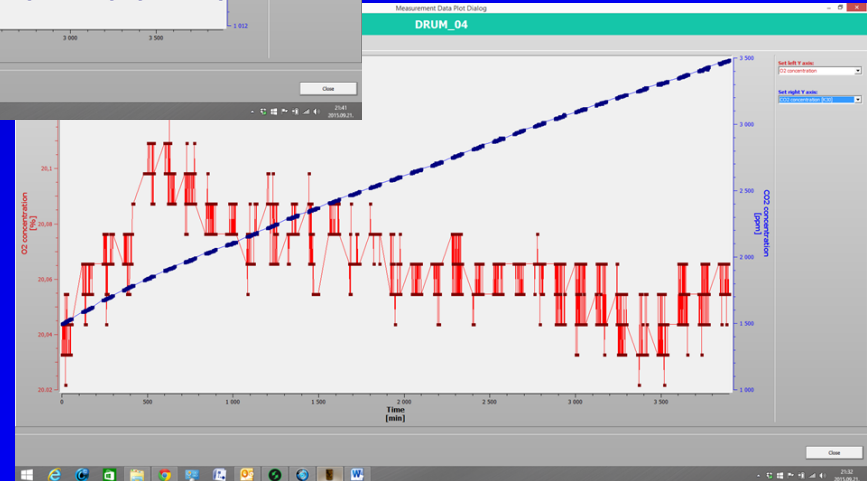
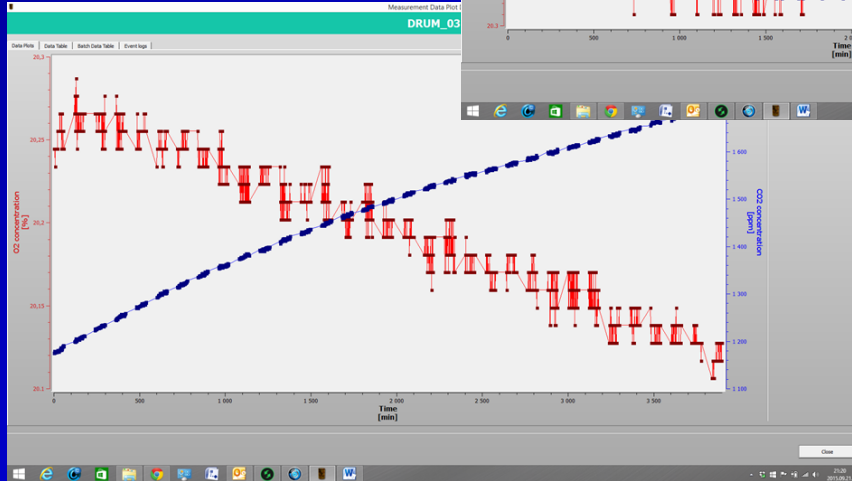


4 Tanks contain conditioned LILW-like waste and 1 Tank filled with air

During the first 3 days of storage/running: *...significant gas production was detected...*

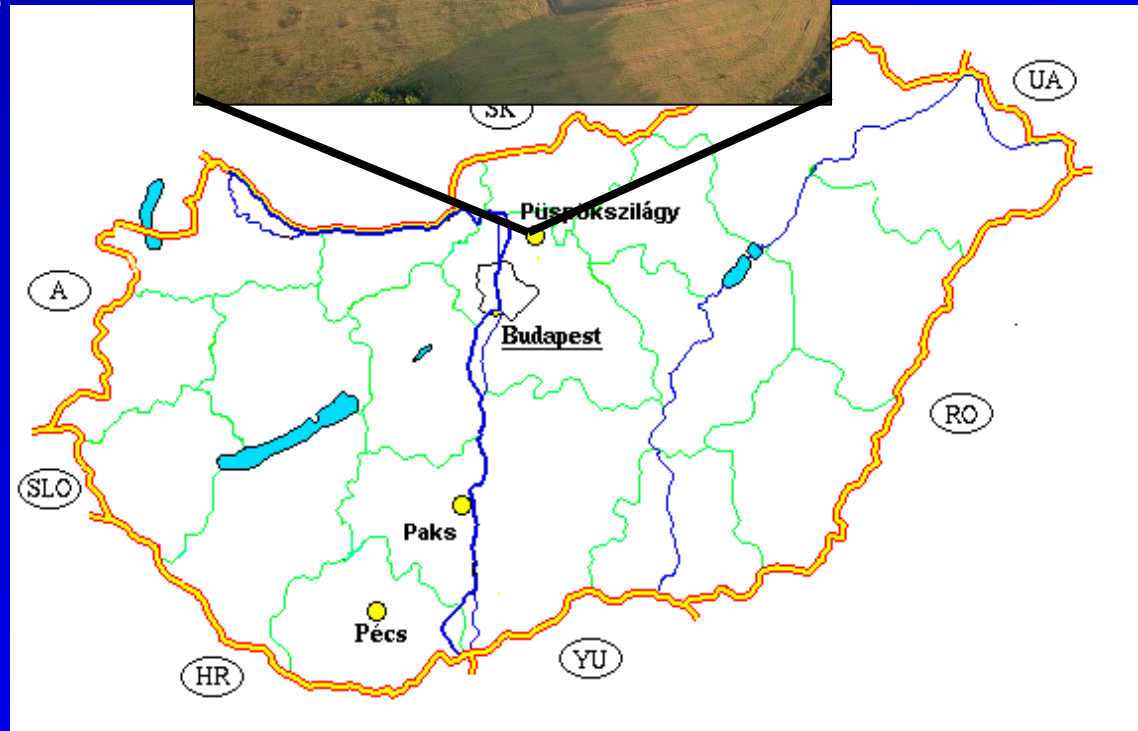


← Empty drum



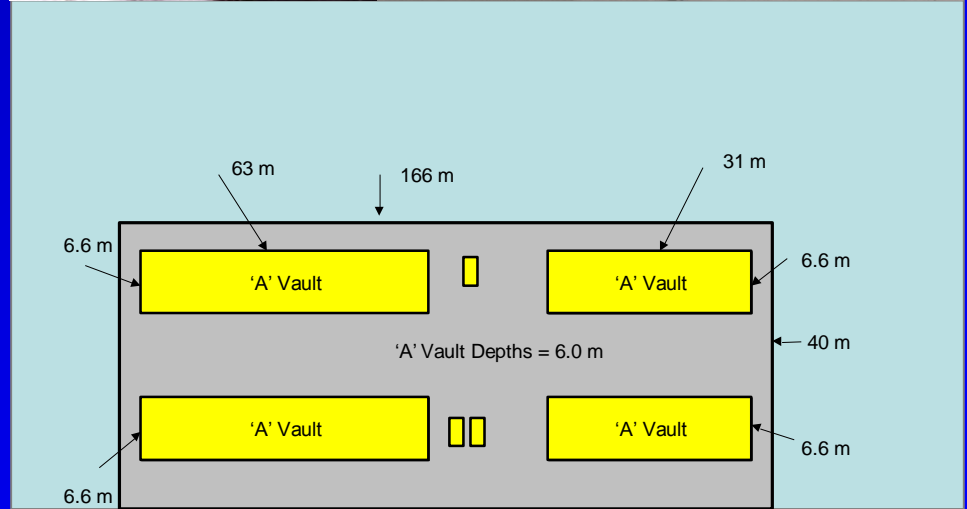
The Püspökszilágy Radioactive Waste (LILW) Disposal & Treatment Facility - Locality

- Some 40 km north-east of Budapest in a hilly area
- 1.5 km far from Püspökszilágy village
- Operated by PURAM
- Licensed by NHMOS
- (HAEA from 2015)



The -Facility - “A” type vaults

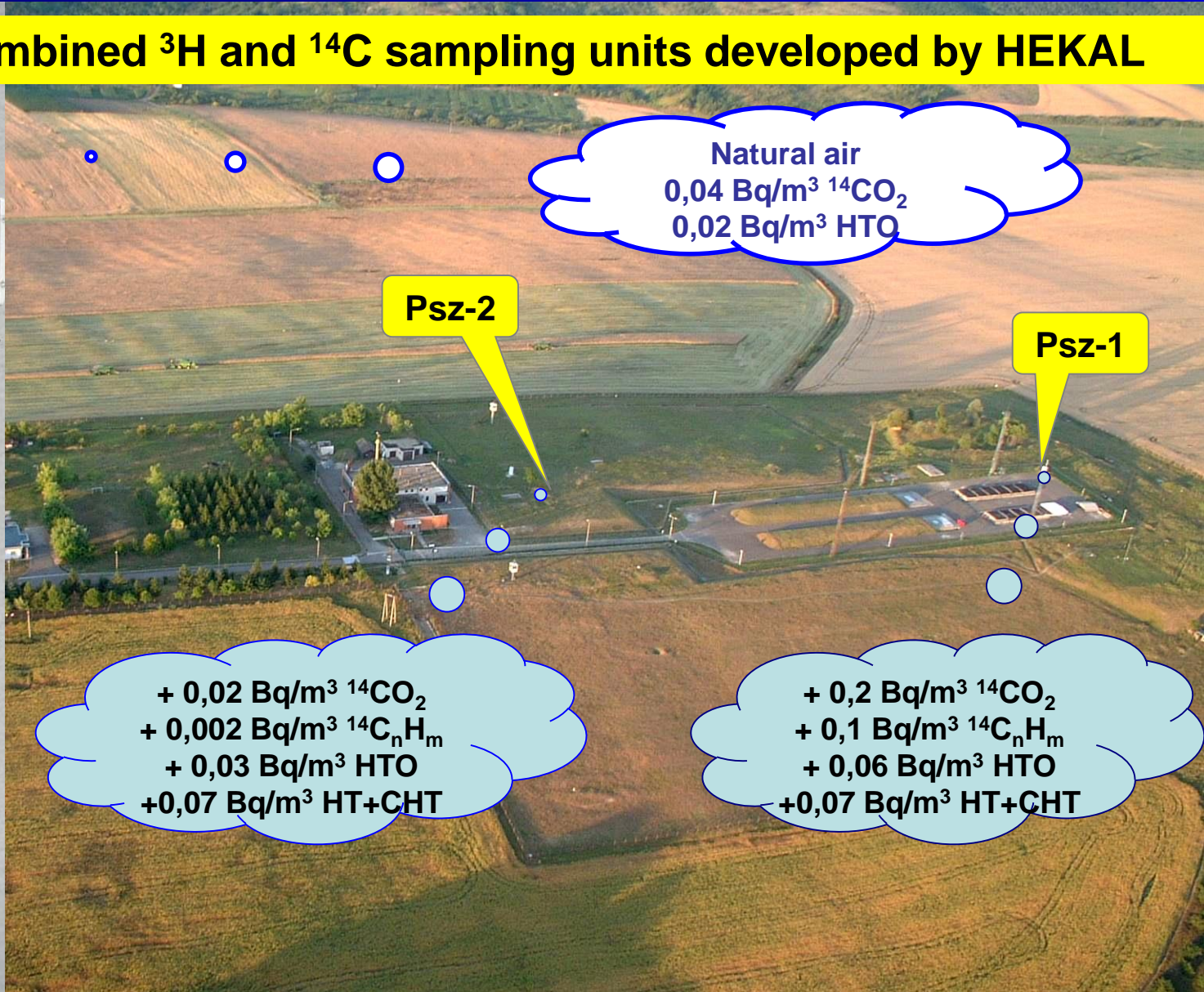
- solid radioactive waste in drums (and plastic bags)
- reinforced concrete structure (40 cm thick walls)
- 4 vaults: AI, AII: 24 cells, AIV: 12 cells of 70 m³, AIII: 6 cells of 140 m³
- total capacity 5040 m³
- covered by protective roof during the filling, then temporarily sealed by 2 m thick clay layer



Major dimensions above are ± 2 m

Atmospheric ^3H & ^{14}C monitoring at the LILW Repository

Combined ^3H and ^{14}C sampling units developed by HEKAL



Natural air
 $0,04 \text{ Bq/m}^3 \text{ }^{14}\text{CO}_2$
 $0,02 \text{ Bq/m}^3 \text{ HTO}$

Psz-2

+ $0,02 \text{ Bq/m}^3 \text{ }^{14}\text{CO}_2$
+ $0,002 \text{ Bq/m}^3 \text{ }^{14}\text{C}_n\text{H}_m$
+ $0,03 \text{ Bq/m}^3 \text{ HTO}$
+ $0,07 \text{ Bq/m}^3 \text{ HT+CHT}$

Psz-1

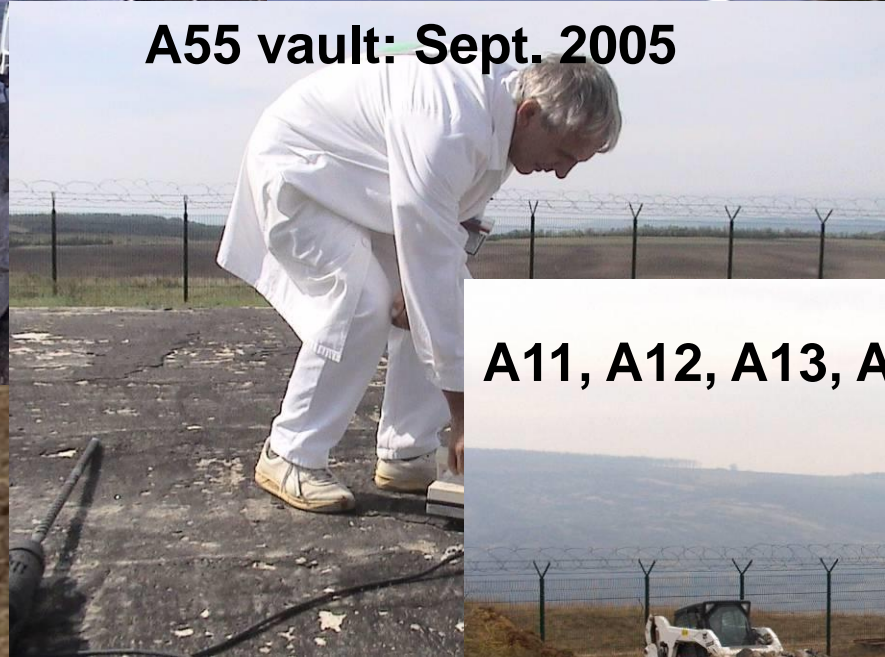
+ $0,2 \text{ Bq/m}^3 \text{ }^{14}\text{CO}_2$
+ $0,1 \text{ Bq/m}^3 \text{ }^{14}\text{C}_n\text{H}_m$
+ $0,06 \text{ Bq/m}^3 \text{ HTO}$
+ $0,07 \text{ Bq/m}^3 \text{ HT+CHT}$

Gas sampling from 7 different closed A-type vaults of Püspökszilágy Facility between 2000-2006

A5 and A6 vault: Marc 2000



A55 vault: Sept. 2005

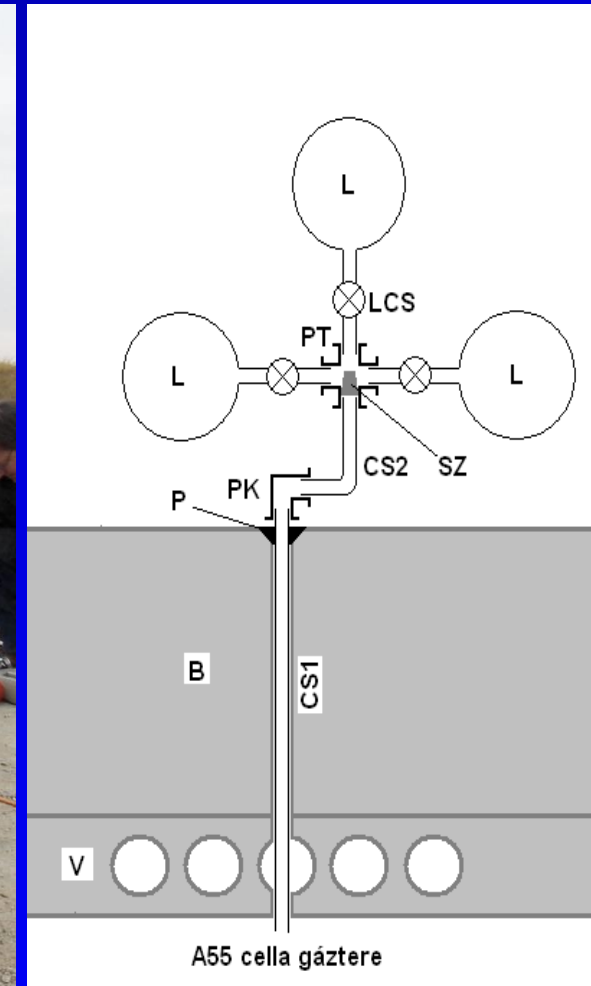
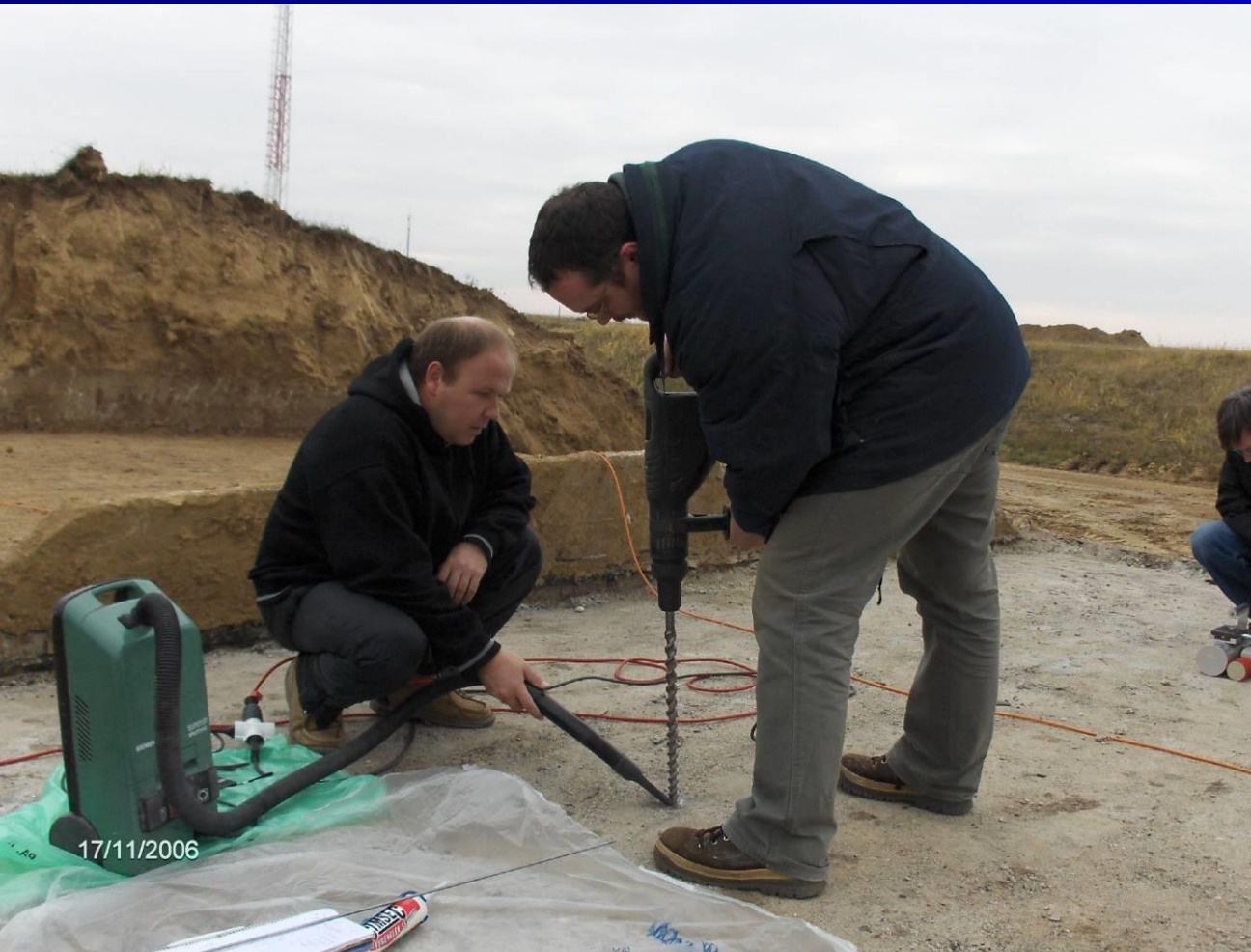


A11, A12, A13, A14 vaults: Nov. 2006



17/11/2006

Pipe-sampling through the vault's ceiling

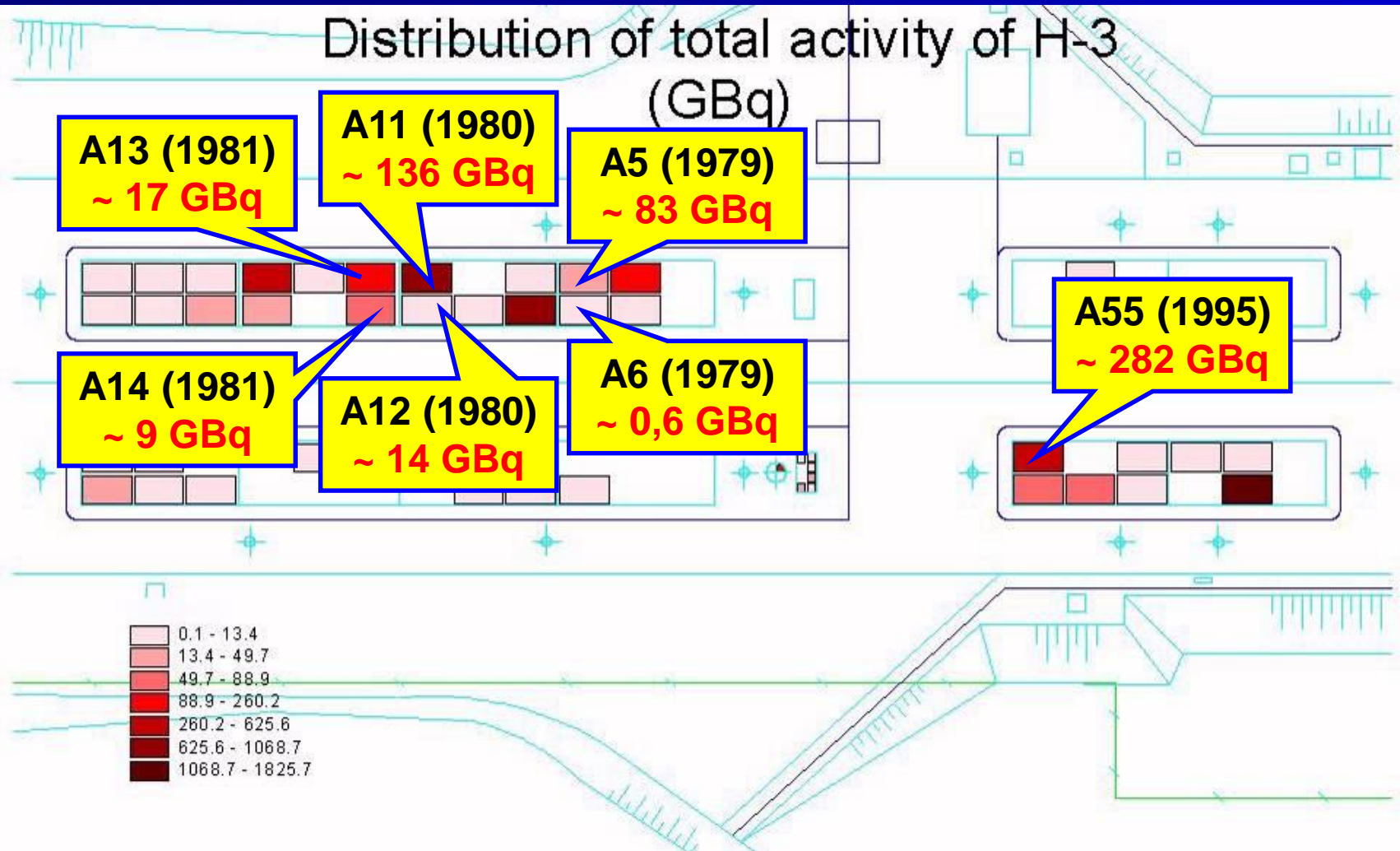


If you are lucky, you can make a proper hole for sampling...

Isotope analytical results of gases from A-type vaults

Vault Nr.	CO ₂ conc. (%)	CH ₄ conc. (cm ³ /l)	¹⁴ C act. (Bq/l gas)	δ ¹³ C (PDB) ‰	³ H act. (Bq/l gas)	³ He (ppm)
A5	1.8	-	61.8	-25.9	8.8	0.130
A6	0.1	-	2.88	-26.7	0.04	0.001
A55	0.5	12.9	88.0	-16.1	21.5	0.280
A11	16.7	-	814.2	-25.1	826.6	0.240
A12	16.8	-	1295.1	-25.4	295.0	0.026
A13	12.8	-	866.1	-26.4	32.2	0.029
A14	9.3	-	869.7	-27.3	110.6	0.016
Air	0.04	-	~ 5·10 ⁻⁵	-7 ~ -9	10 ⁻⁴ - 10 ⁻⁵	~ 7·10 ⁻⁶

Estimation of total restored ^3H activity by ^3He results



$^3\text{H} \longrightarrow ^3\text{He}$, number of produced ^3He atoms is equal with number of decayed ^3H atoms, if the vault is (enough) closed for gases...

Palcsu L. et al. Journal of Radioanalytical and Nucl. Chem. 286 (2010)483-487

Deep LILW Repository for Nuclear Power Plant waste at B́ataapati, opened in 2010.



Surface building with 3000 LILW drums controlled ventilation and C-14/H-3 sampling

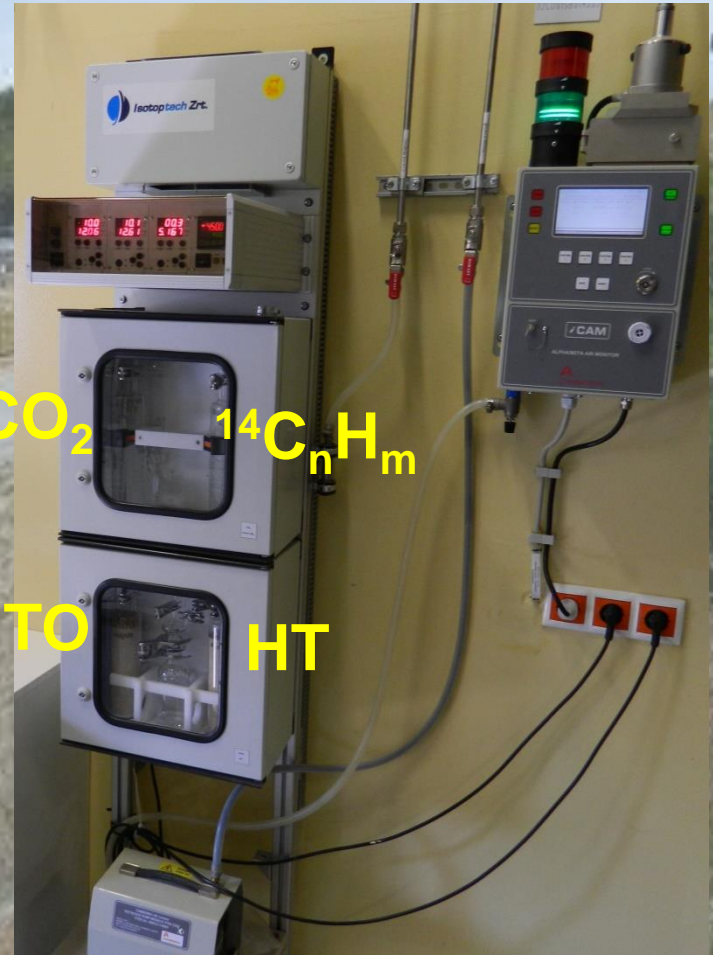
Used air

7500 m³ air/day

Fresh air

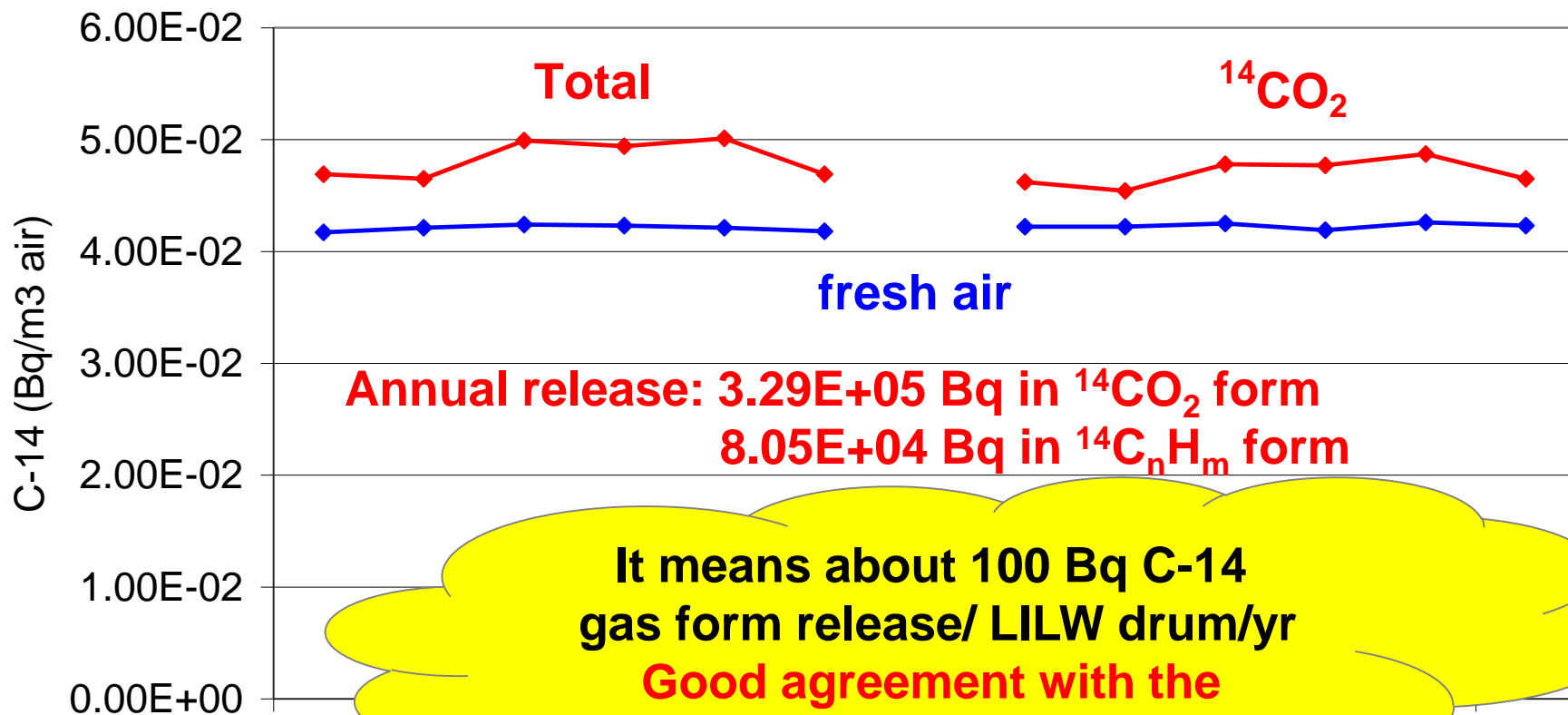
$^{14}\text{C}\text{O}_2$ $^{14}\text{C}_n\text{H}_m$

HTO HT



Surface building with 3000 LILW drums controlled ventilation and C-14/H-3 sampling

C-14 in ventilated air of LILW storage building,
2 months integrated samples, 2012



**It means about 100 Bq C-14
gas form release/ LILW drum/yr**

**Good agreement with the
individual LILW results!**

Summary

- ✓ **> 70 different real LILW drums and 7 different real LILW vaults were studied during last 15 years by AHEKAL.**
- ✓ **Special sampling methods and sample preparation techniques were developed.**
- ✓ **The main detected gases: H₂, CH₄ and CO₂.**
- ✓ **In LILW drums the ³H and ¹⁴C activity conc. of headspace gas was 5-6 orders of magnitude higher than in natural air.**
- ✓ **In LILW vaults the ³H and ¹⁴C activity conc. of headspace gas was 7-8 orders of magnitude higher than in natural air.**
- ✓ **Using ³He measurements we could make a realistic estimation of the total restored ³H activity in several LILW vaults.**
- ✓ **Ventillated air C-14 release is in agreement with LILW drum results: 100 Bq ¹⁴C /LILW drum/yr released!**

The background of the slide is a close-up photograph of a light-colored wood surface, showing distinct, wavy grain patterns and natural textures. The lighting is even, highlighting the fine details of the wood fibers.

**INVESTIGATION OF THE ^{14}C EMISSION OF A
RADIOACTIVE WASTE DISPOSAL FACILITY IN THE
ANNUAL RINGS OF THE NEARBY TREES**

Description of the Püspökszilágy LILW facility

Constructed in 1976 on the basis of the recommendation of the IAEA Safety Series No. 15 (1965)

The facility was built on the top of an elevation to drain the precipitation.

The geological environment is clay

Concrete storage cells were buried in the ground

Research, medical, industrial and agricultural LILW are stored

Previously sealed storage cells have been reopened since 2001



Monitoring of the ^{14}C content of the air at the disposal facility

Atmospheric ^{14}C sampling devices have been operating in the disposal facility. (Psz-1, Psz-2, Psz-3)

The sampler takes integrated CO_2 samples representing for two month periods

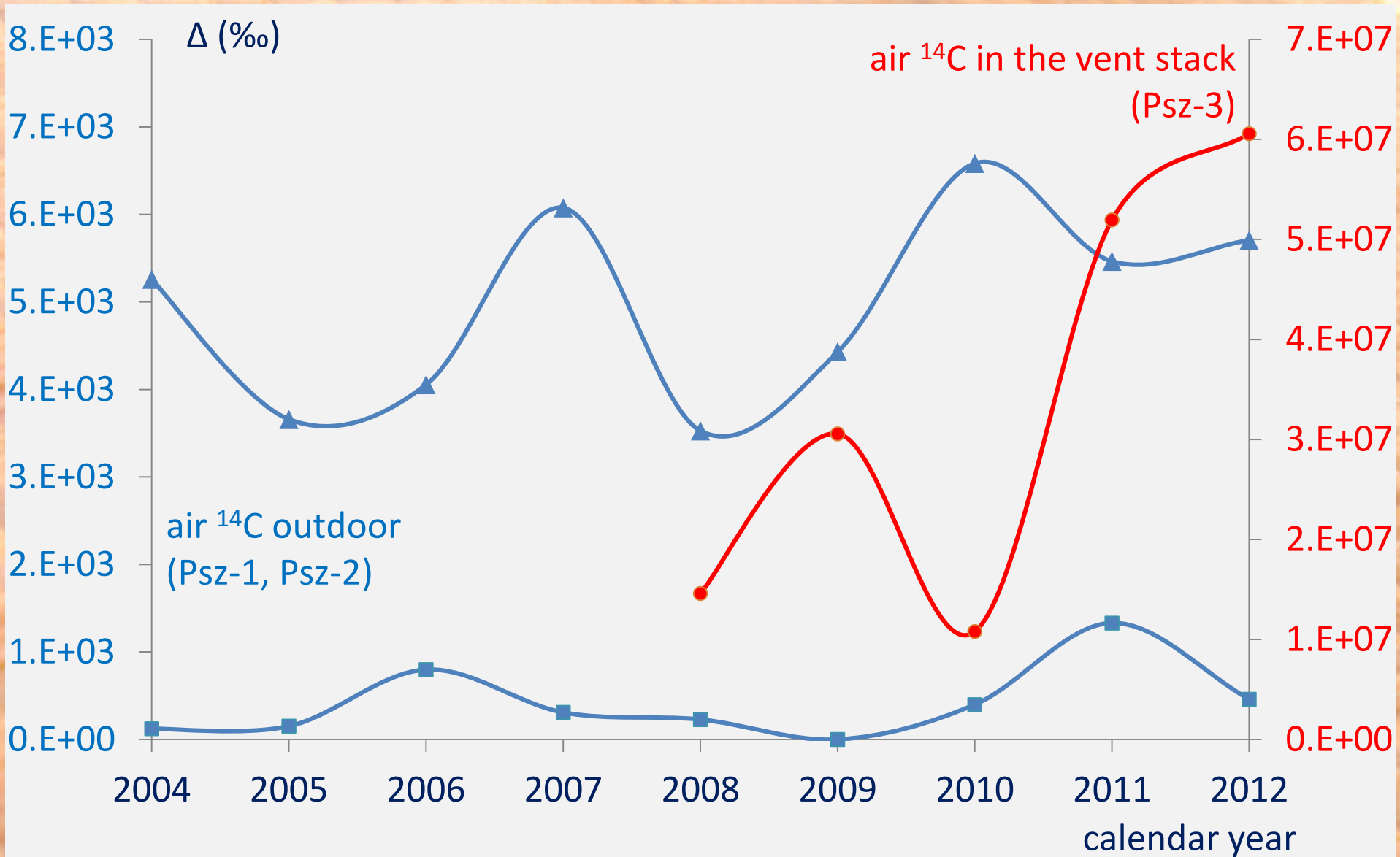
Two samplers are operating outside next to the storage cells (Psz-1, Psz-2)

and one is operating inside the vent stack (Psz-3)

atmospheric ^{14}C and ^3H monitoring unit developed by Isotoptech and Atomki



Monitoring of the ^{14}C content of the air at the facility



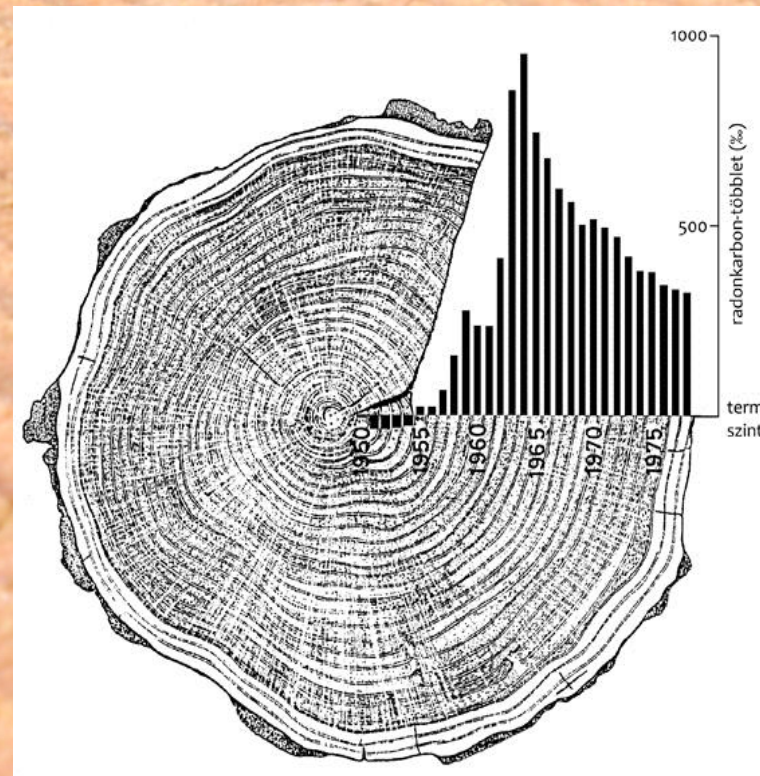
Footprint of the atmospheric ^{14}C in the trees

Plants build their organic materials from the atmospheric CO_2

The tree rings preserve the radiocarbon concentration of the air with the resolution of one year.

In several published cases, excess ^{14}C was measurable in the annual rings of the trees near the nuclear facilities.

^{14}C signal of the atom bomb peak in a Hungarian tree



(Hertelendi et al. 1982)

Sampling of tree rings

Background sample was taken upwind about 3 km from the facility

BKG tree

Sampling was performed in the facility, 50 m from the vaults in the wind direction.

DF tree



Poplar trees were sampled in May 2013

Multiple cores were extracted using increment borer.

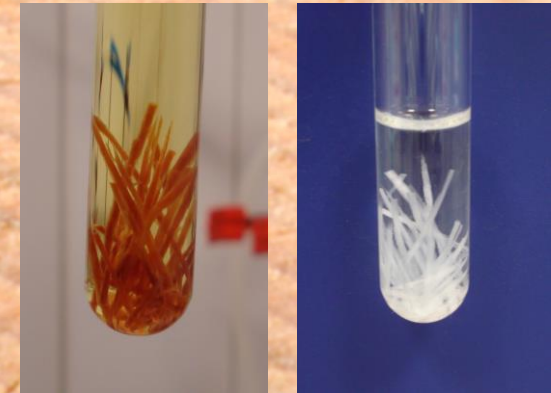


Sample preparation and AMS measurement

The cores were cross-checked and the rings were separated



In order to remove lignin and waxes cellulose was prepared from the tree rings by BABAB method (Němec, et al., 2010)



Cellulose was combusted to CO_2

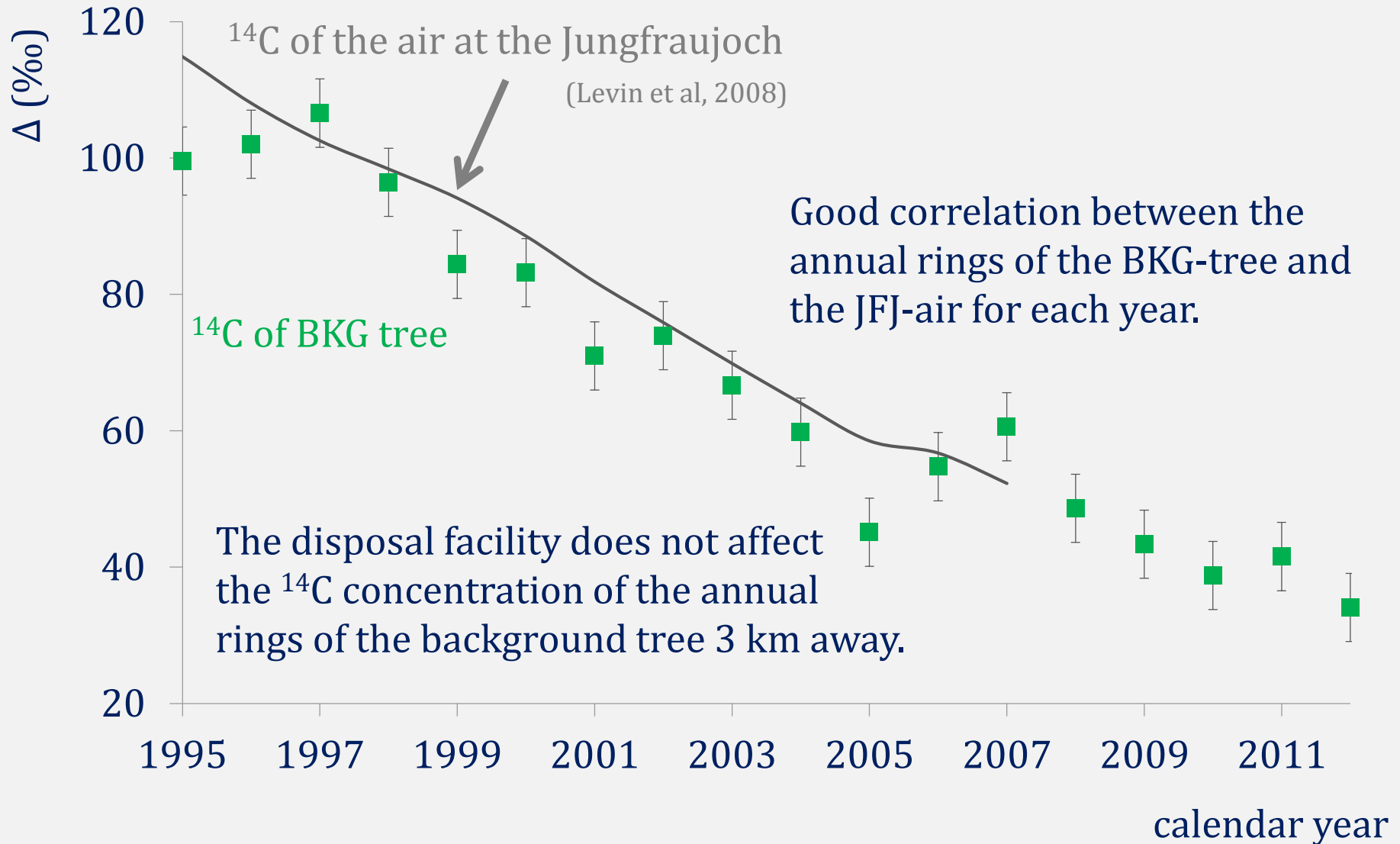
The CO_2 was converted to graphite by sealed tube graphitisation method.



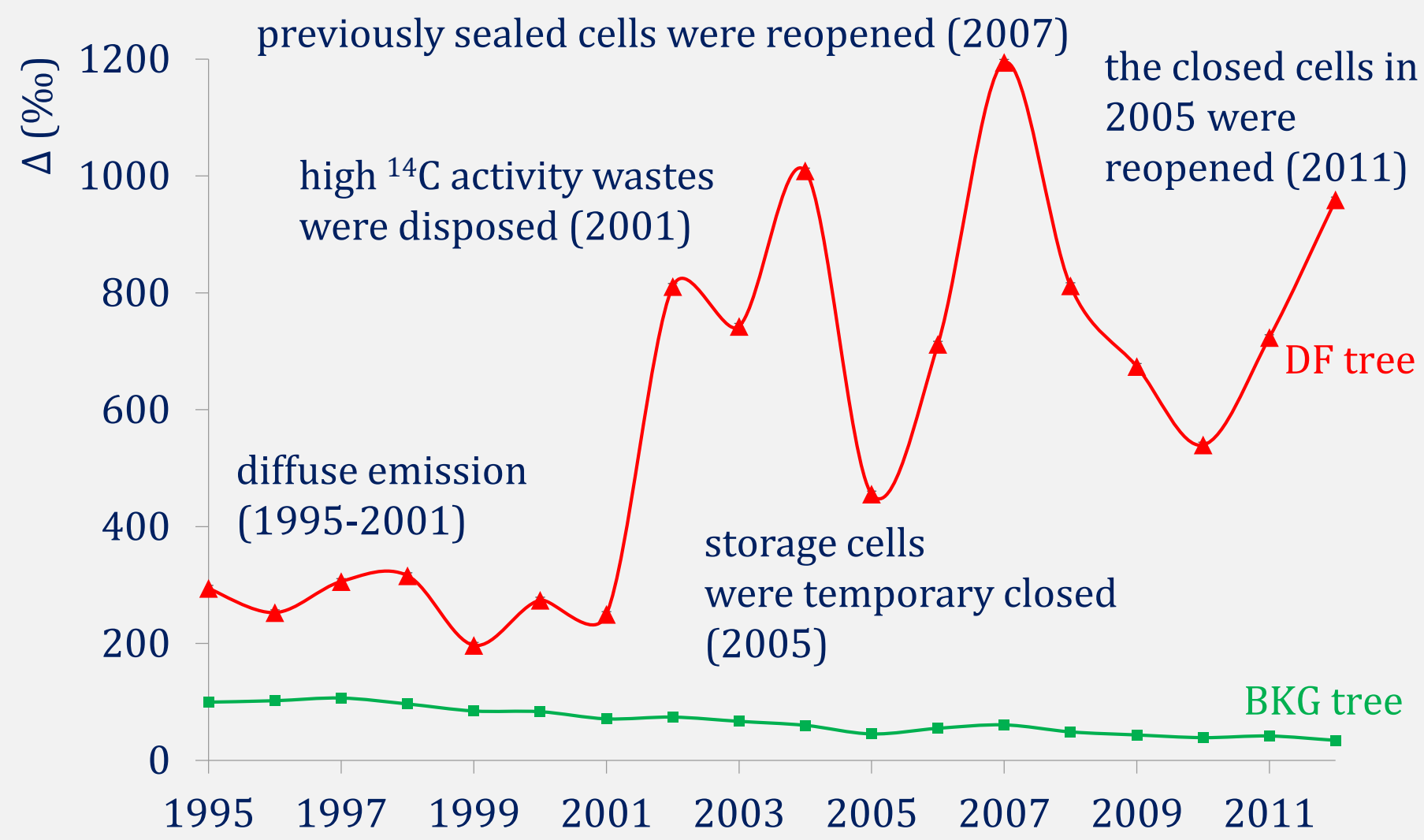
The ^{14}C measurements were performed with the MICADAS AMS in Debrecen.



Comparison of the ^{14}C content of the background tree and the Jungfraujoch air

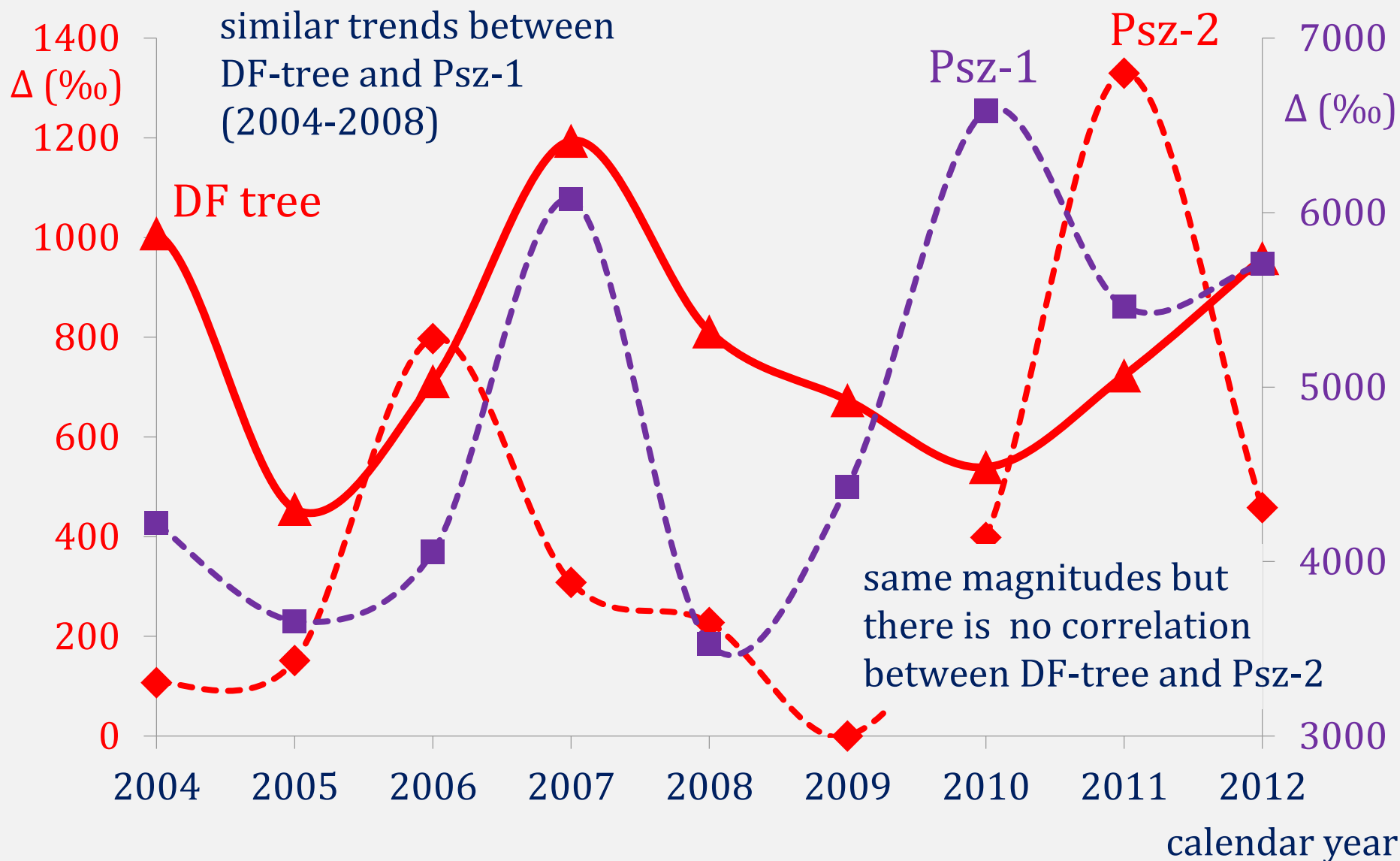


^{14}C content of the annual rings of the tree at the storage cells

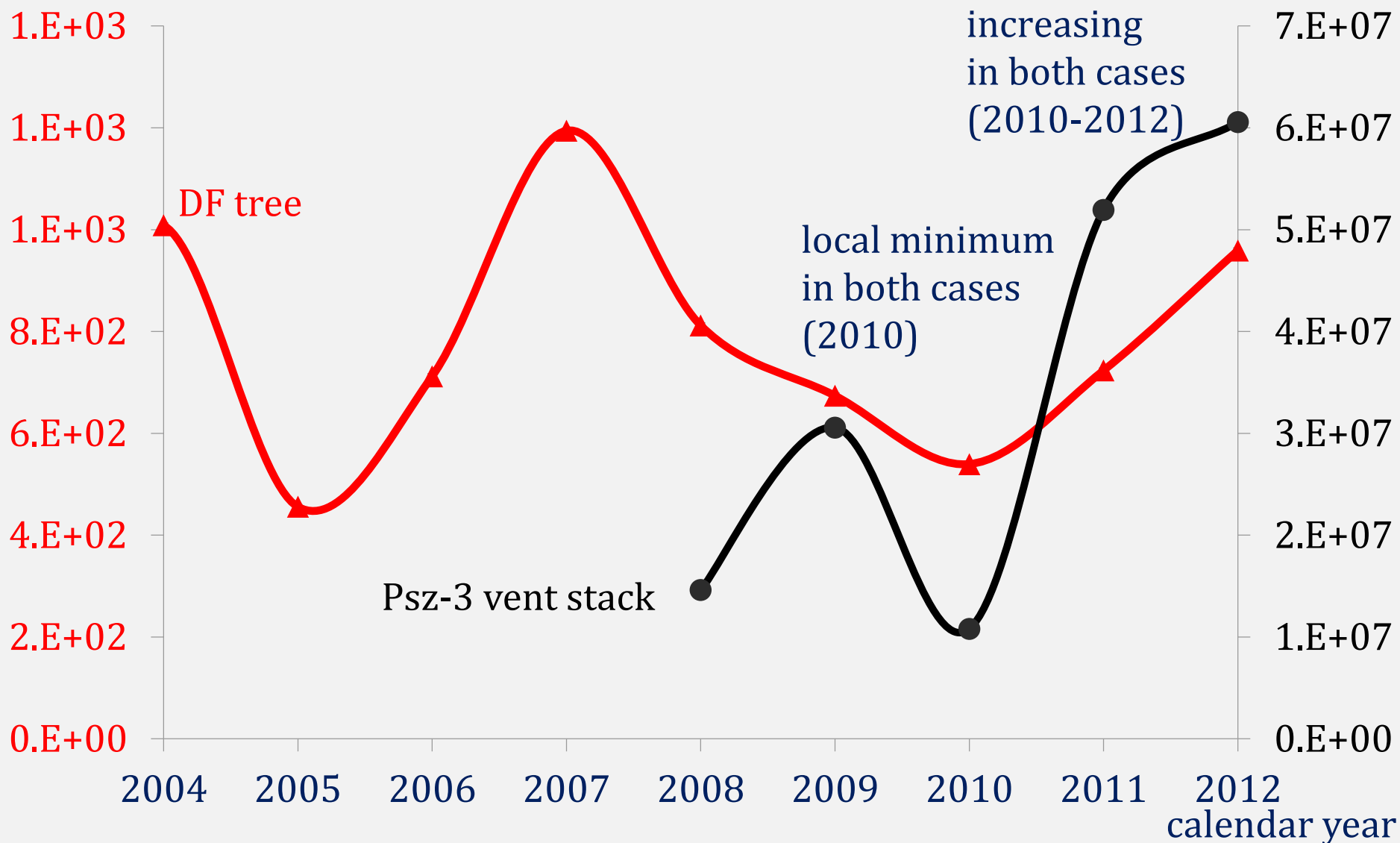


The technological activities can be traceable in the ^{14}C content of the tree rings

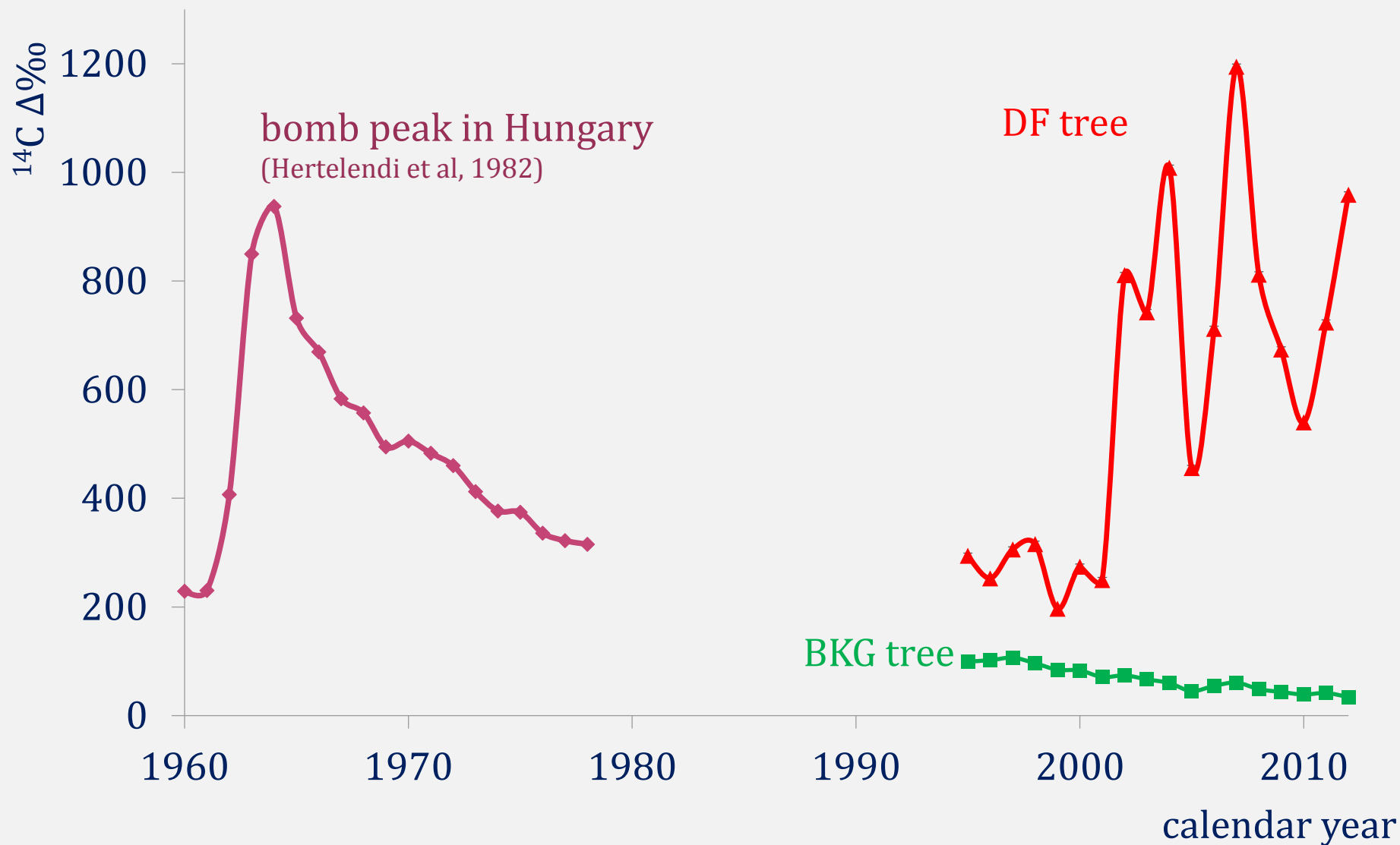
Comparison of the ^{14}C content of the DF-tree with the air sampling unit in outdoor (Psz-1, Psz-2)



Comparison of the ^{14}C content of the DF-tree with the air sampling unit in the vent stack (Psz-3)



Comparison of the ^{14}C content of DF tree rings to the tree rings in the 60th years in Hungary



Conclusions

The disposal facility does not affect the ^{14}C concentration of the annual rings of the background tree 3 km away.

The ^{14}C concentration of DF tree is significantly higher than the background tree. Each jump and decrease can be attributed to a technological process performed during the development and the processing work.

It can be concluded that the emissions of the storage cells and the technological building affect more or less the ^{14}C content of the trees nearby.

The Püspökszilágy LILW disposal facility constructed on the basis of the IAEA Safety Series No. 15. (1965) only locally affects the environment regarding the atmospheric ^{14}C emission.

Thank you for your attention!



Isotoptech Zrt.



**Research was financed by the Hungarian Public Limited Company
for Radioactive Waste Management (PURAM)
special thanks for I. Barnabás, P. Ormai and S. Kapitány**

Great honor for Dr. Ede Hertelendi[†] and Károly Bérci[†]

**molnar.mihaly@atomki.mta.hu
www.isotoptech.com**

Carbon-14 Source Term CAST

Name: Erika Neeft

Organisation: COVRA

Date: 20-Feb-2018



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.



Purpose of this day



- Lot of knowledge available
- What is relevant knowledge?
- Integration of knowledge
 - For this day \Rightarrow post-closure safety assessment
 - Implementation transport model for ^{14}C
 - Focus on justification for
 - Model validation
 - Traceability of parameter values for assumed model
 - Have you experienced that getting calculated results may be the most easy part of an assessment

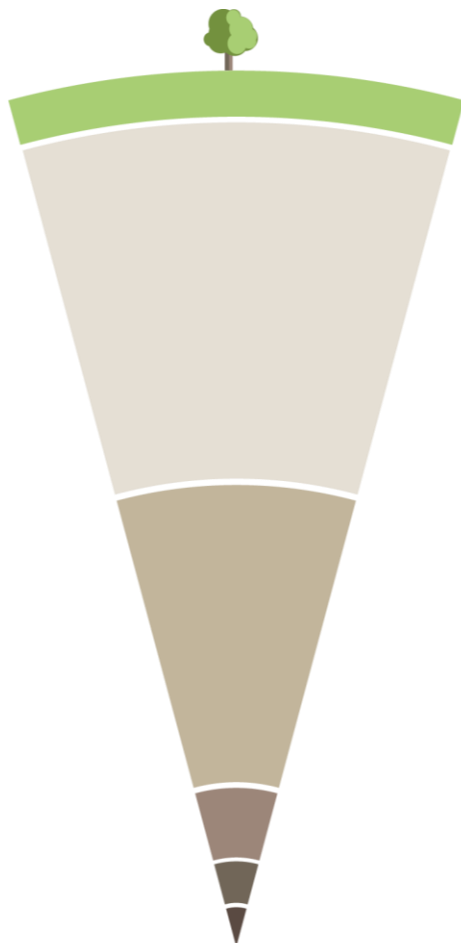


MeSA - initiative



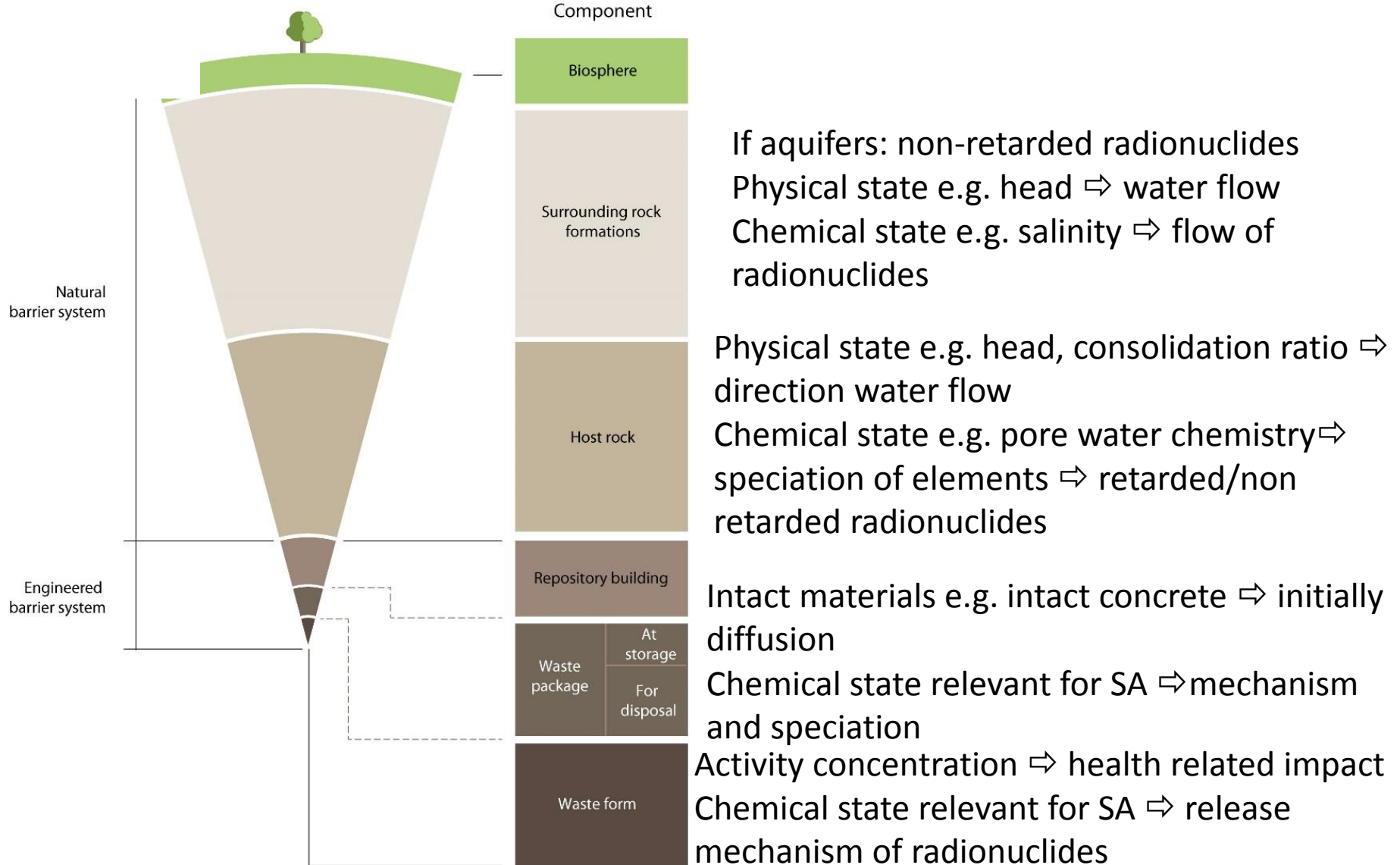
- NEA, 2012
- Safety concept
 - description of roles of natural and engineered barriers for different time frames
 - Evaluation of implication of uncertainties in the fulfilment of the safety functions over time
 - Formulation of scenarios: specific description of a potential evolution of the disposal system from a given initial state

Disposal system



Component		High-Level Waste	Low- and Intermediate-Level Waste
Biosphere		Physical media: soil, atmosphere, climate, water bodies et cetera Living organisms: humans, animals and bacteria, interacting with physical media	
Surrounding rock formations		Formations in (extended) the Netherlands Hydrological Instrument: aquifers and aquitards	
Host rock		Poorly indurated clay or salt formation unaffected by the presence of excavations	
Repository building & affected materials		Backfill can be composed of materials such as grout, crushed salt and bentonite. Concrete support is required for poorly indurated clay such as Boom and Yperian Clay. Affected materials include the host rock disturbed by the presence of excavations (clay or salt formation)	
Waste package	At storage	Canister	Concrete and galvanized, painted steel or steel container
	For disposal	Overpack, concrete buffer, steel envelope for clay overpack, for salt	No additional packaging considered for disposal
Waste matrix		Matrices can consist of different materials including glass, UAl _x , zircalloy and concrete	Waste matrix can be grout and can consist of a variety of materials including glass, metal, ash, textile and plastic.
			U ₃ O ₈ conditioned with concrete

Initial state, MBS relevant for SA



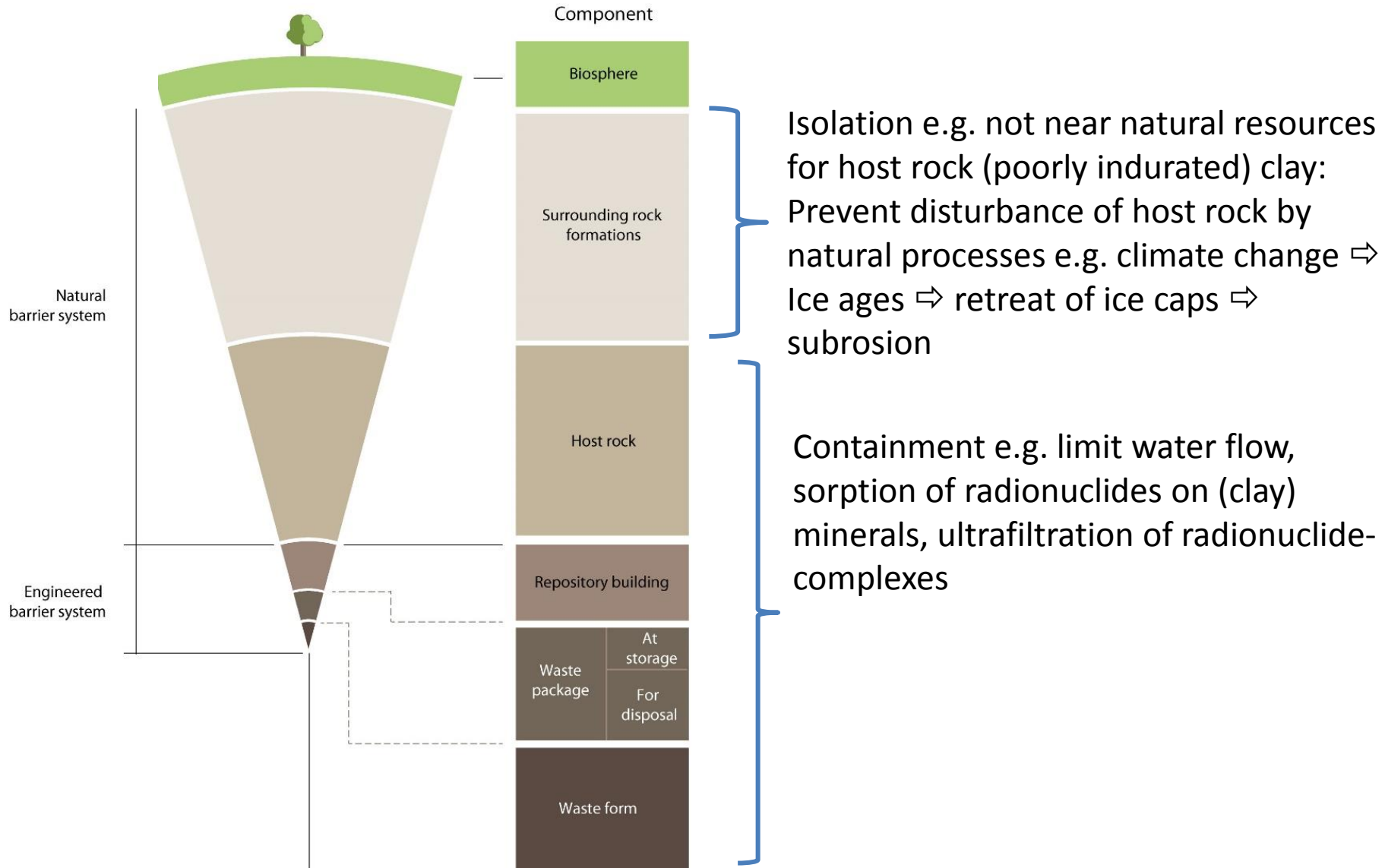


Safety concept

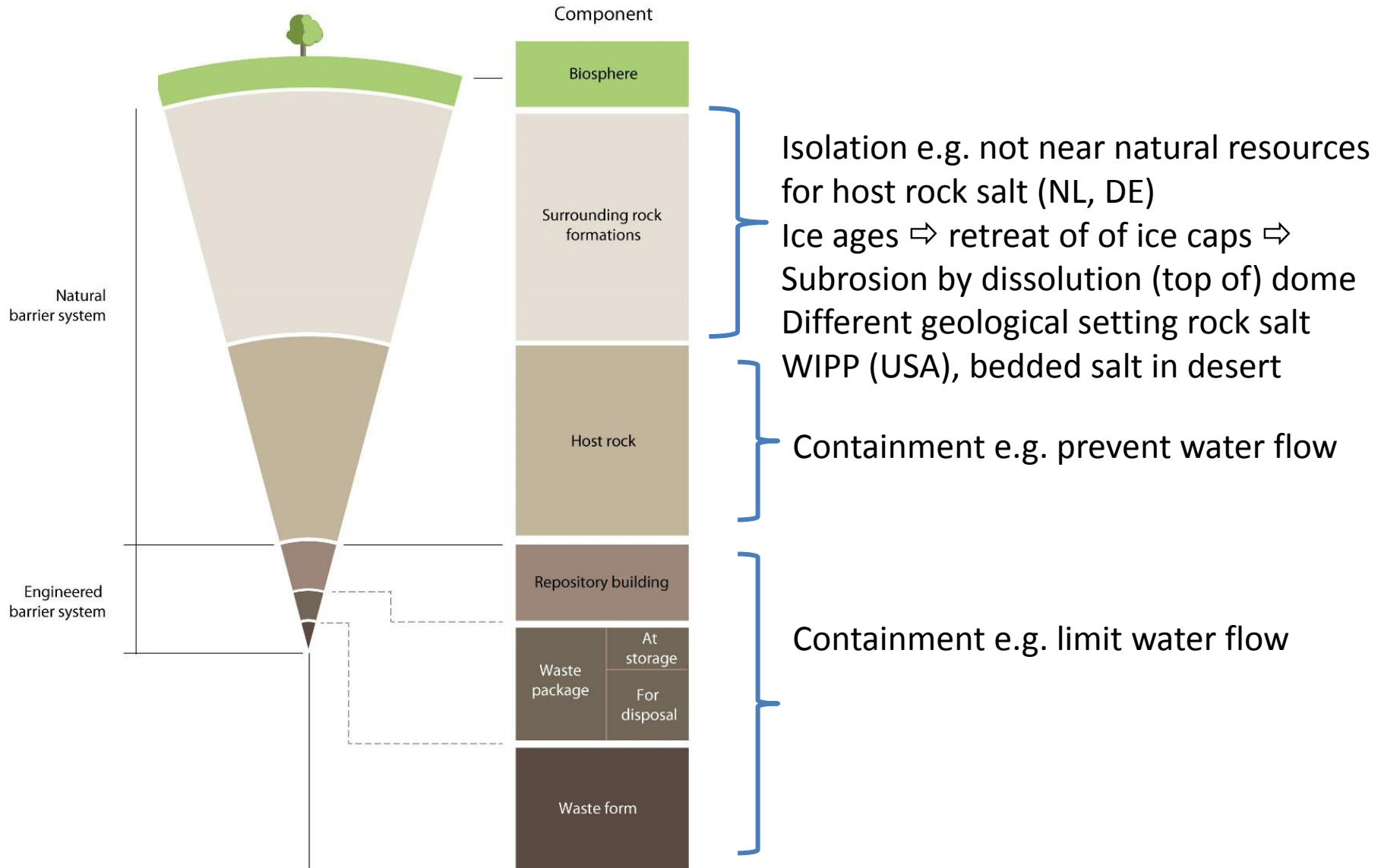


- Safety functions for what period
 - Isolation
 - Removal of waste safely from direct interaction with people and environment
 - Containment
 - Retaining radionuclides within the multi barrier system (MBS) until radioactive decay has reduced the radiation hazard of waste.

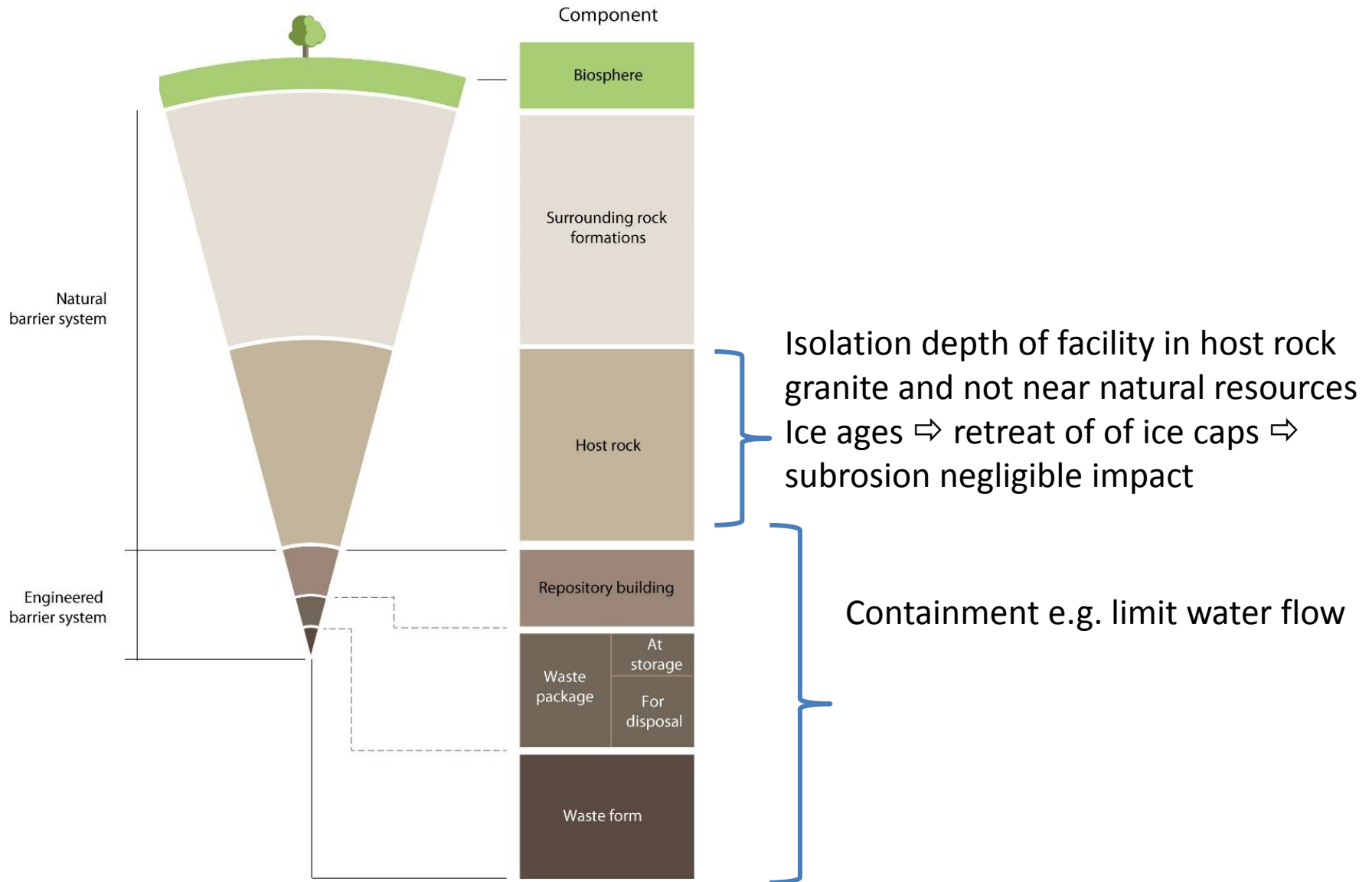
Safety concept clay



Safety concept salt

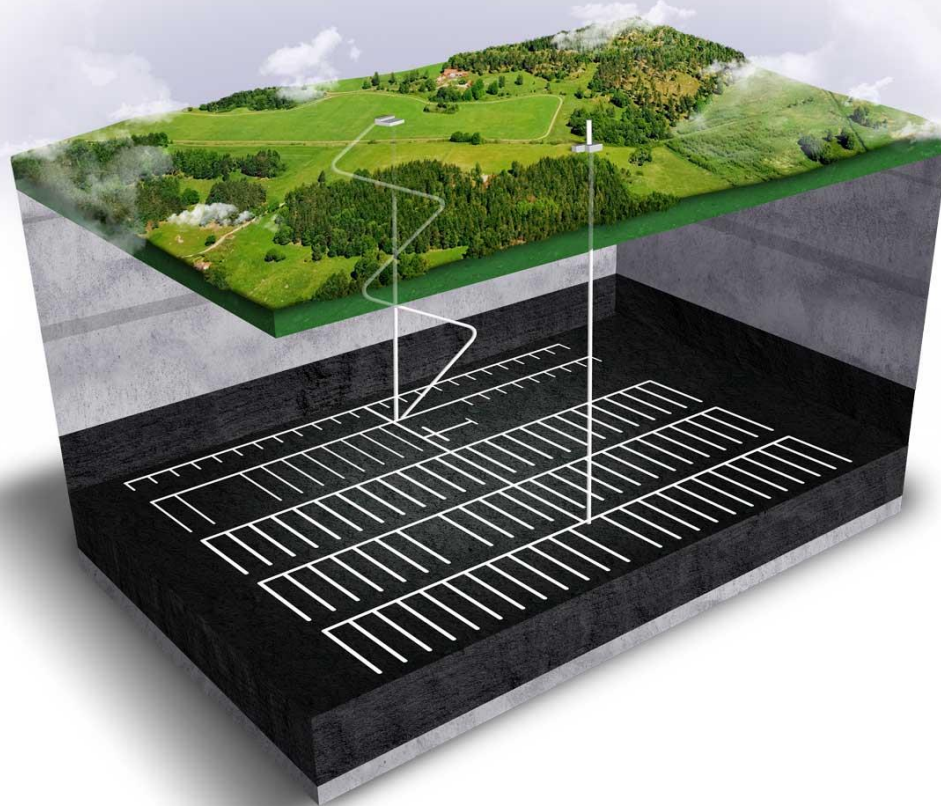


Safety concept granite

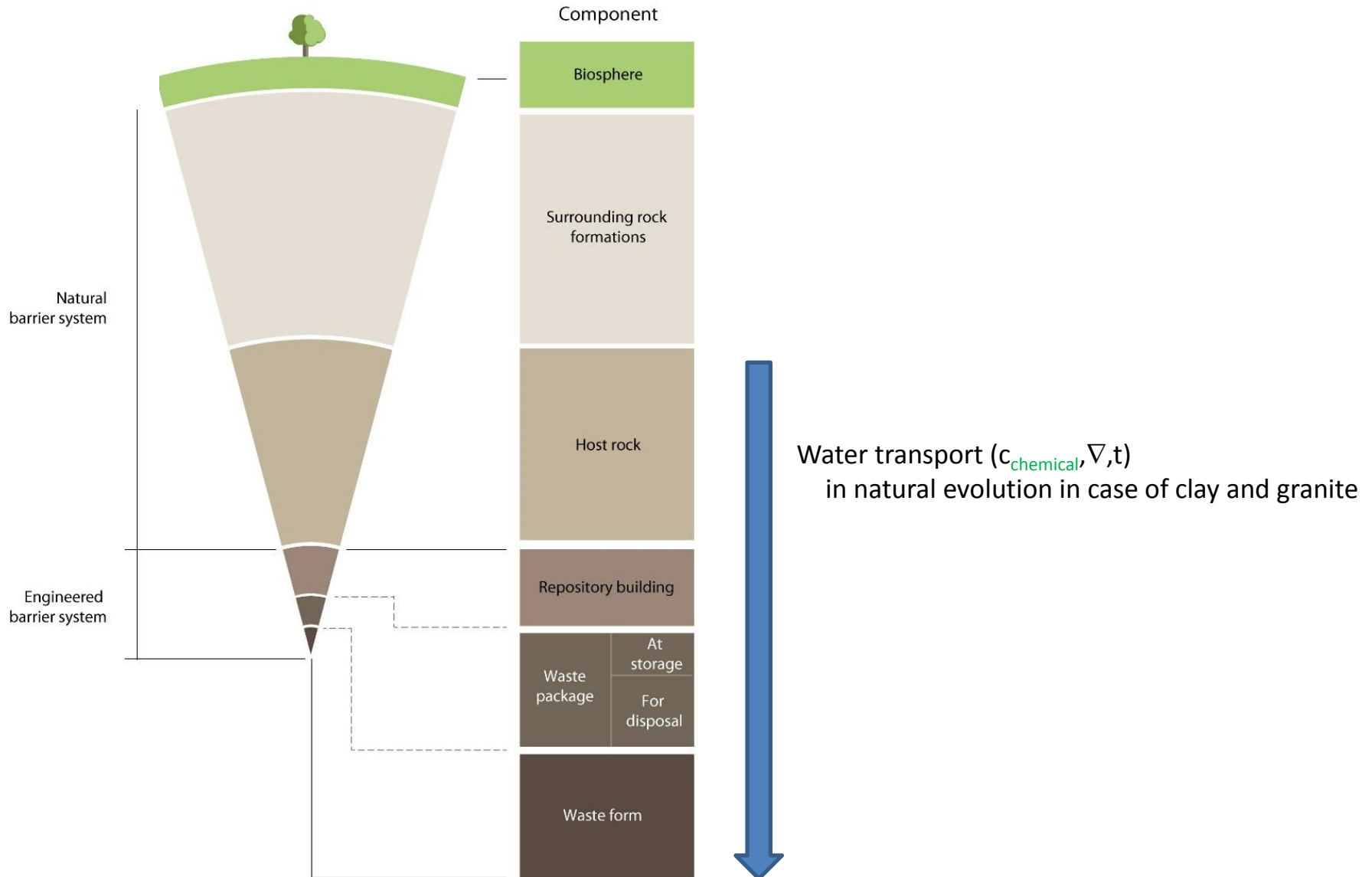


- Evaluation of the implication of uncertainties in the fulfilment of safety functions
 - Normal evolution i.e. most expected evolution
 - If calculations of models showed high health related impact then
 - update disposal concept (how (processed) waste is suggested to be disposed) and/or change processing of waste e.g.
 - » Disruption MBS by criticality, gas generation
 - » Different geological setting of host rock, deeper disposal depth
 - Altered evolutions
 - Human intrusion

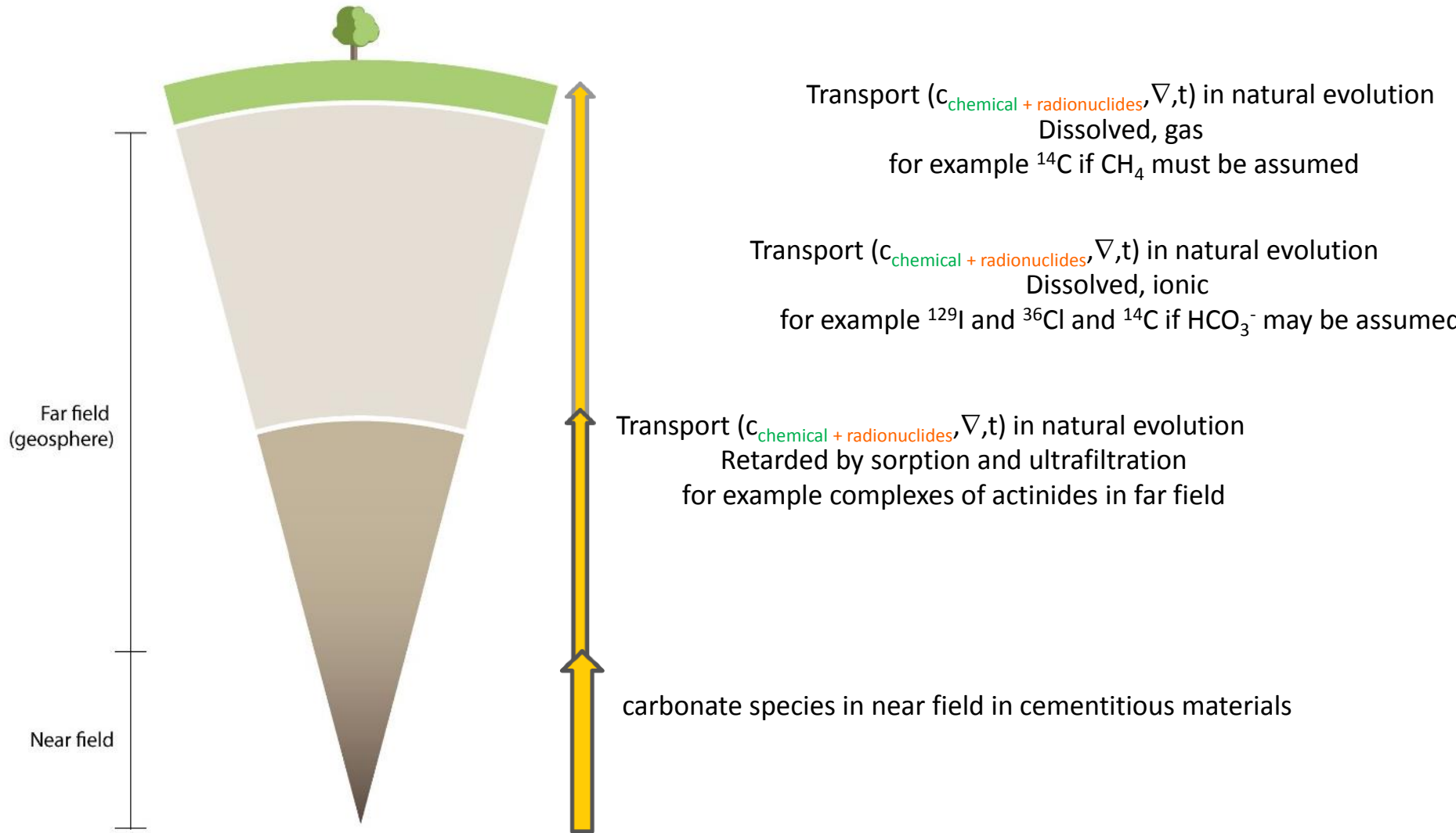
Example disposal concept

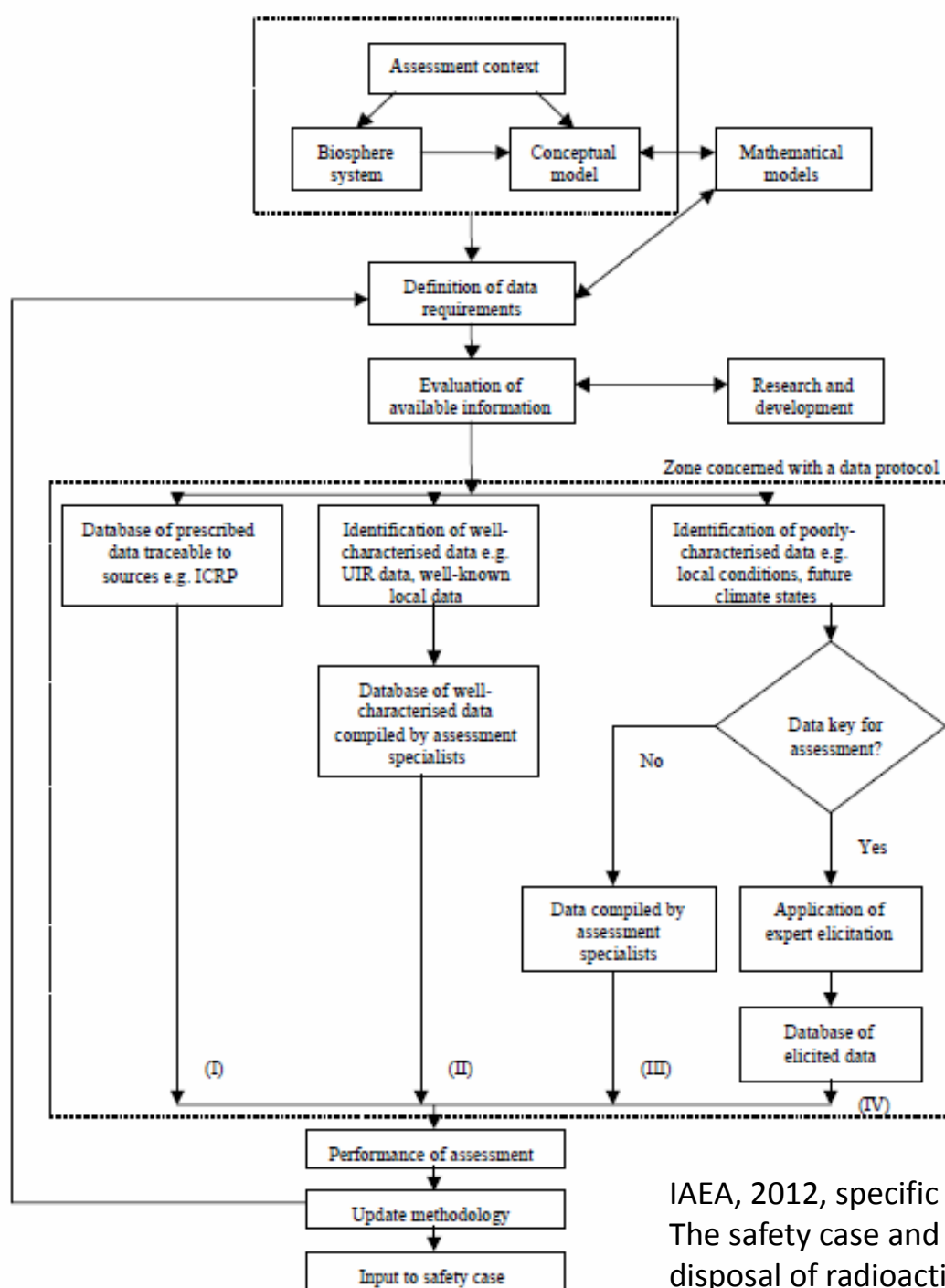


Geological disposal of waste



Geological disposal of waste





IAEA, 2012, specific safety guide 223
 The safety case and safety assessment for the disposal of radioactive waste



Traceability



- If the safety assessment is undertaken iteratively, there may be a tendency for references simply to refer to decisions made in a prior iteration of the safety assessment ('self-citations'). The reviewer may need to trace through a chain of documents before finding the origin of an assumption, parameter value or decision, which may be time consuming. Further, caveats and limitations to the work included in the primary references may become lost or diluted with subsequent repetition. This can lead to a reduction in confidence in the operator i.e. organization that executed the SA that and, consequently, confidence in the safety of the facility by the reviewer. As such, primary references should be cited directly, and each iteration of the documentation should permit straightforward evaluation of its traceability.

Traceability

- In this presentation, sedimentary i.e. not primary references are indicated with *
- Primary references preferred for SA except for reviewed data e.g. databases from organisations/individuals with a high international established confidence e.g.
 - Dose conversion coefficients
 - Values for half-lives of radionuclides
 - Thermodynamic data

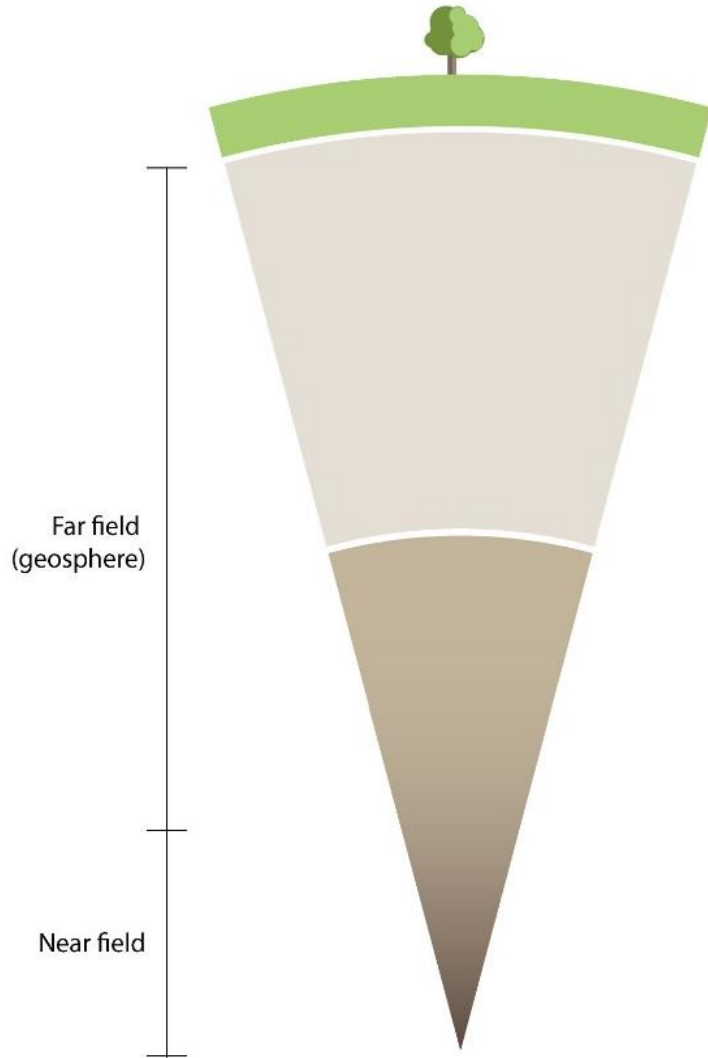


MeSA - initiative



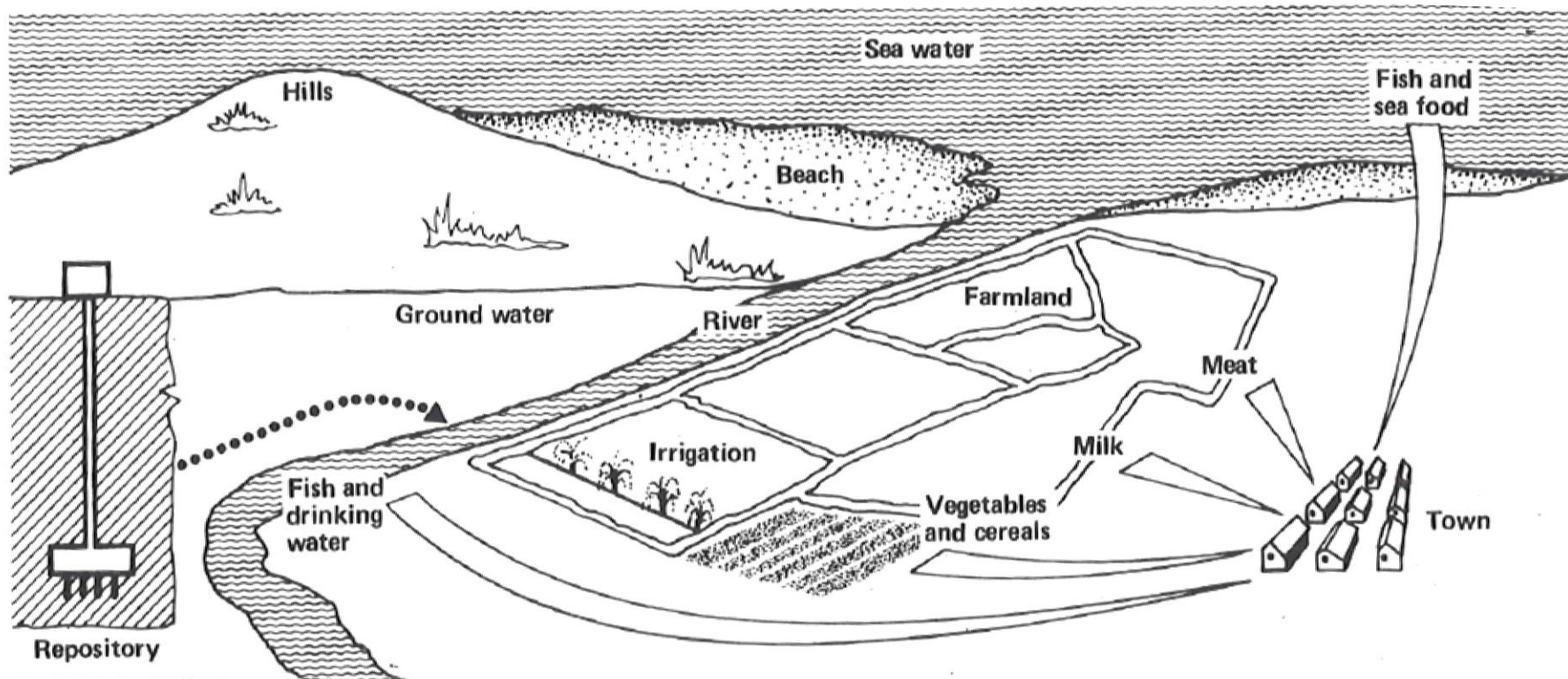
- NEA, 2012
- Safety assessment : justification of assumptions as important as calculated results
 - Validity of assumptions for carbon-14 containing waste investigated in CAST

Disposal



Biosphere: receptor for any radioactivity that moves upwards from the geosphere.
SA model biosphere processes that control how people might be exposed to radionuclides transported from disposal facility

Biosphere





IAEA biosphere



- Assumptions
 - Ingestion of food and drinking water: m^3 /year intake
 - Inhalation rate: m^3 /h
 - Exposure time / External radiation

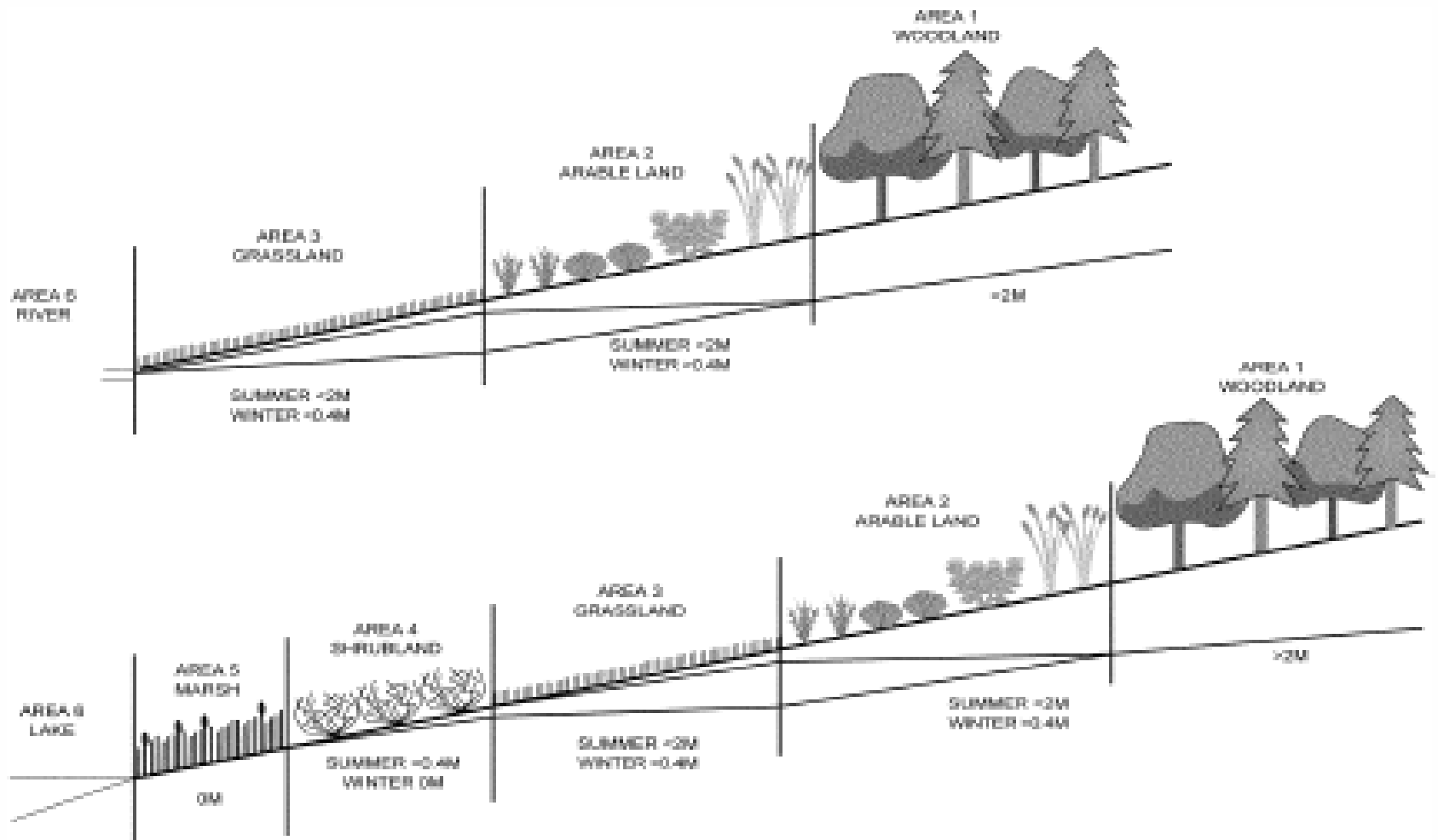


IAEA biosphere

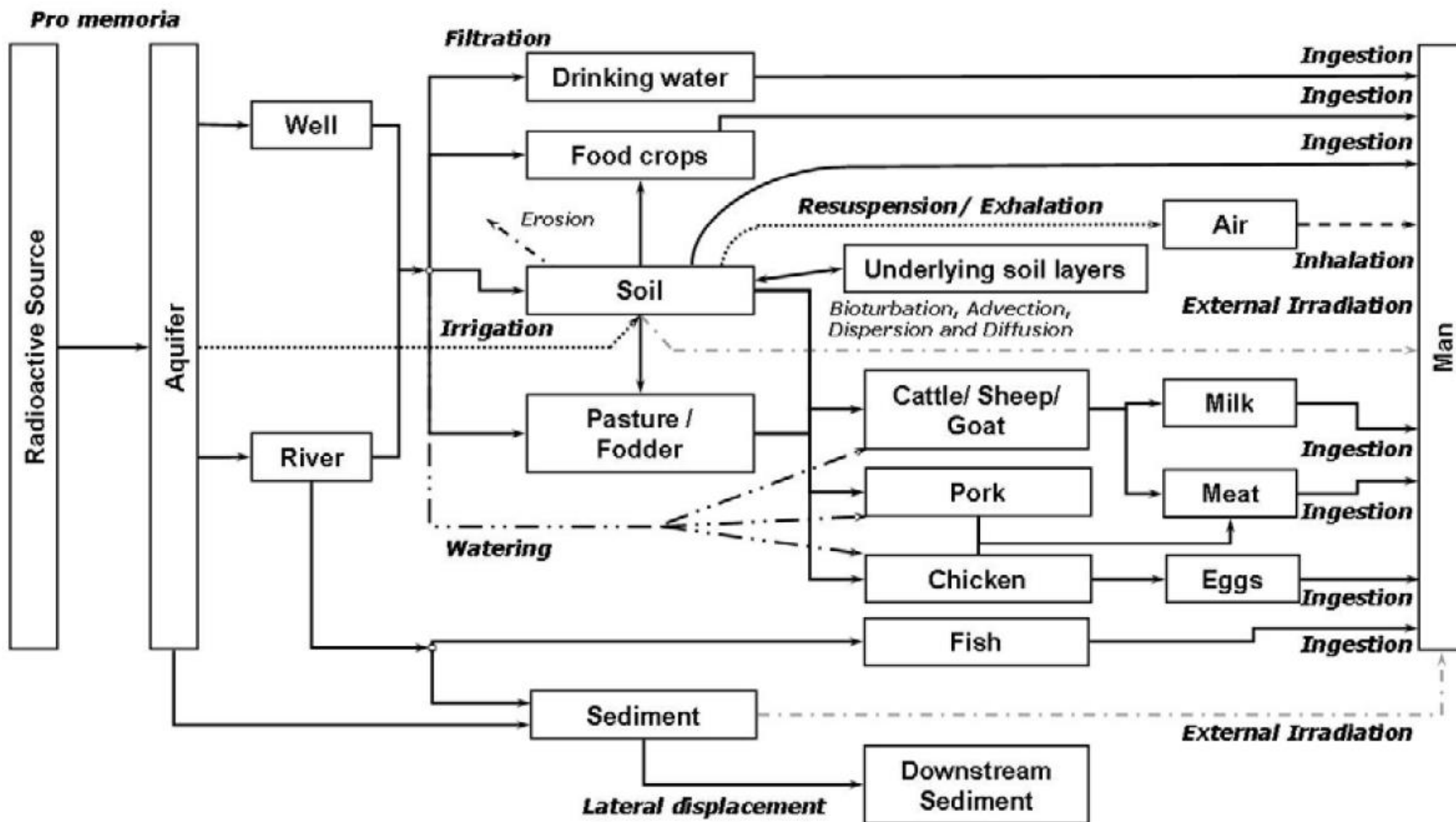


- Assumptions
 - In the time frame of 10^4 to 10^6 years after closure disposal facility, significant changes in climate, human behaviour highly speculative \Rightarrow Stylized biosphere
 - Deep geological disposal
 - In a shorter time frame than 10^4 years \Rightarrow habits in particular region
 - Surface disposal

Stylised biospheres



Biosphere





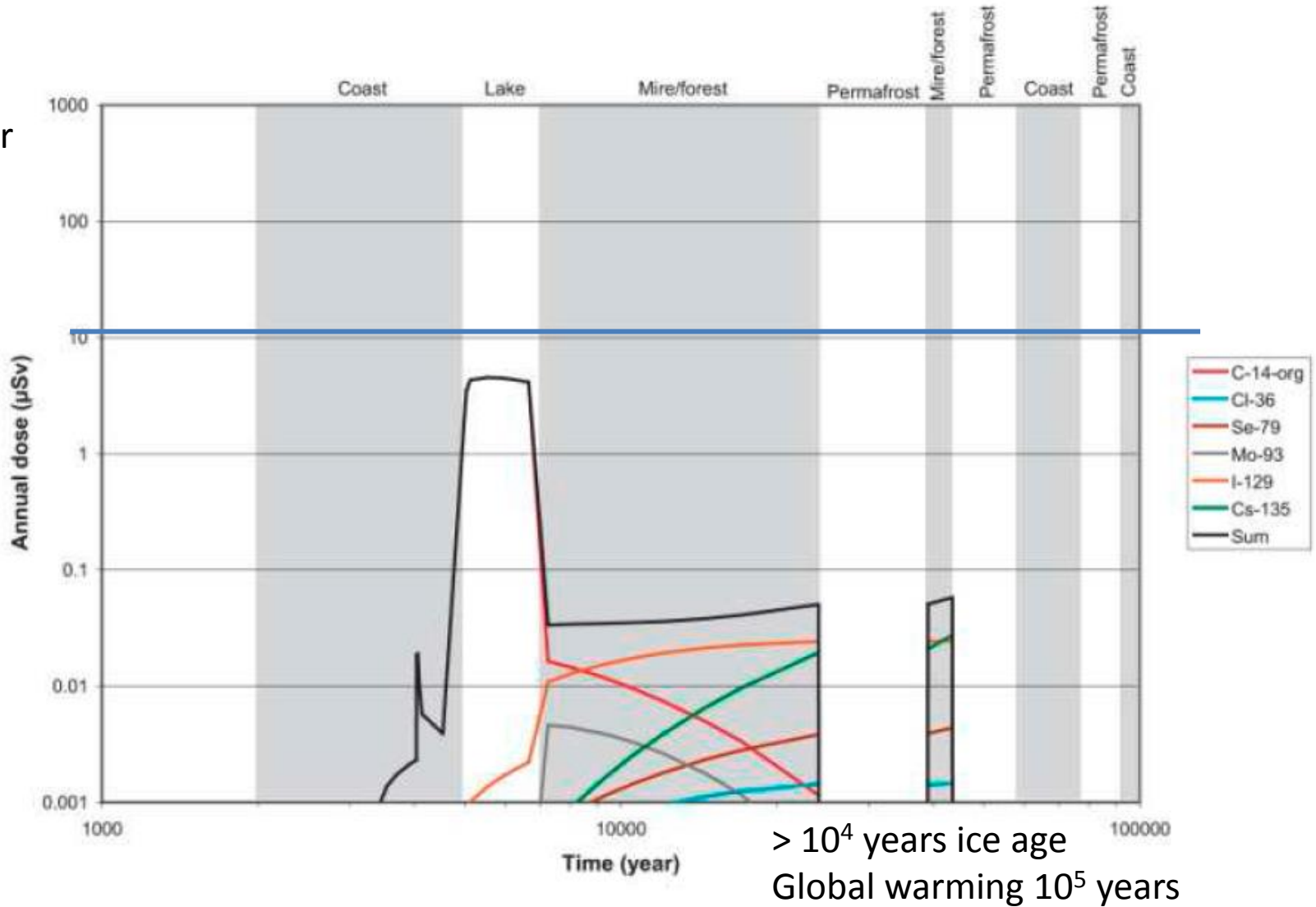
ICRP



- Up to date dose conversion coefficients i.e. Sv/Bq for every radionuclide for infants, adult members of the public and workers
 - Ingestion
 - Inhalation
 - Size of particles 1 μm or 5 μm
 - For carbon-14, soluble or reactive gas
 - External radiation
 - not ICRP but
 - To be determined from gamma-emitted radionuclides that reached the soil

Calculated results

14 μSv /year
to meet
Risk
Constraint
 10^{-6}





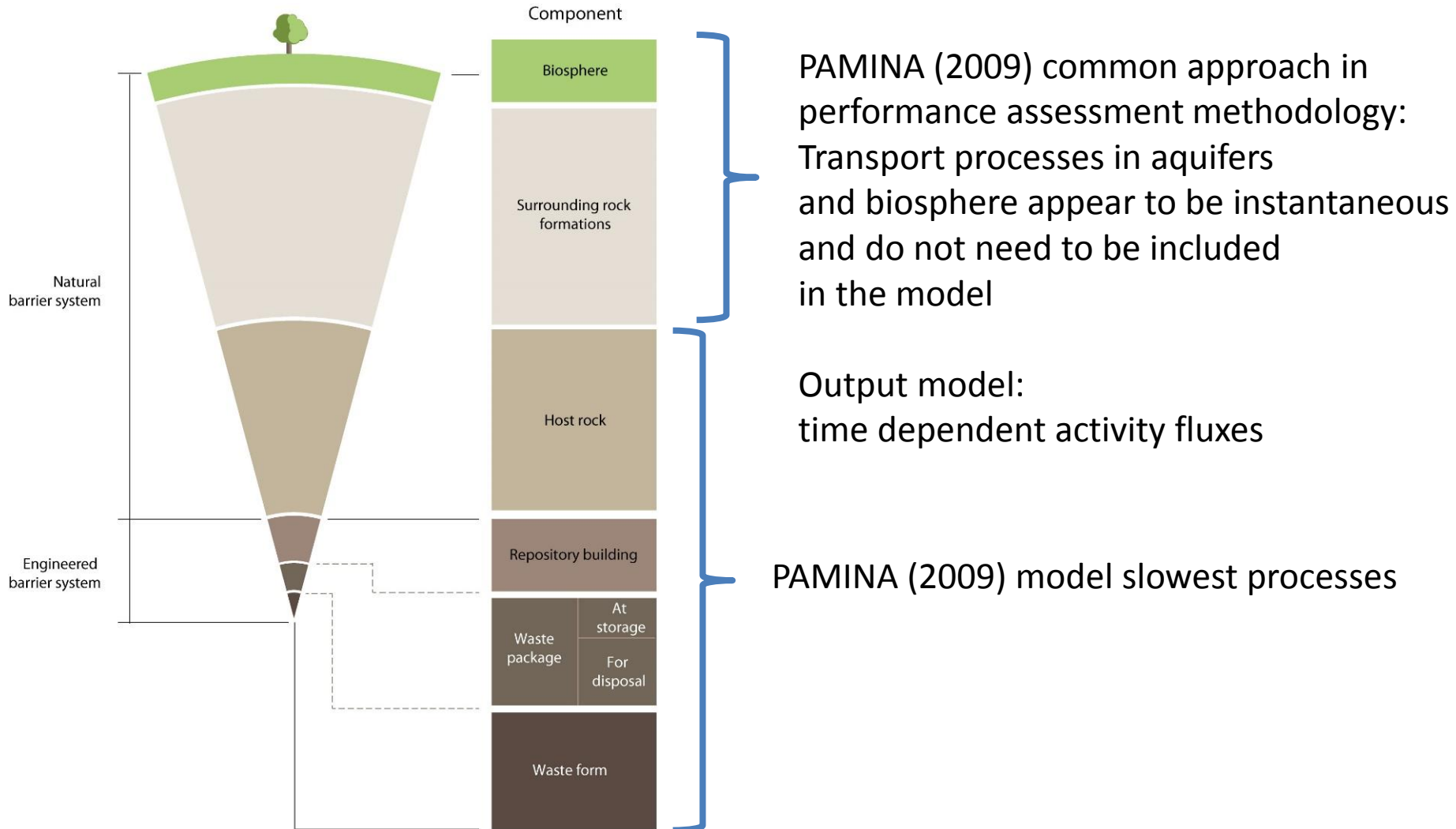
Safety assessment

- Comparison calculated exposure with yardstick to optimise Radiological Protection; ICRP 0.3 mSv per year for a GDF
- Carbon-14 yardstick?
 - Disposal facility contributes a fraction to radiological exposure from natural sources
 - $1/10$
 - In case of natural carbon-14
 - 2 atoms $\text{cm}^{-2}\text{s}^{-1}$ flux into biosphere from cosmic origin
 - 0.2 atoms $\text{cm}^{-2}\text{s}^{-1}$ to compare calculated carbon-14 release from waste

ICRP, 2013: Radiological protection in geological disposal of long-lived solid radioactive waste, ICRP 122

Kovaltsov GA, Mishey A, Usoskin IG, A new model of cosmogenic production of ^{14}C in the atmosphere, Earth Planetary Sciences Letter 337 (2012) 144

Geological disposal



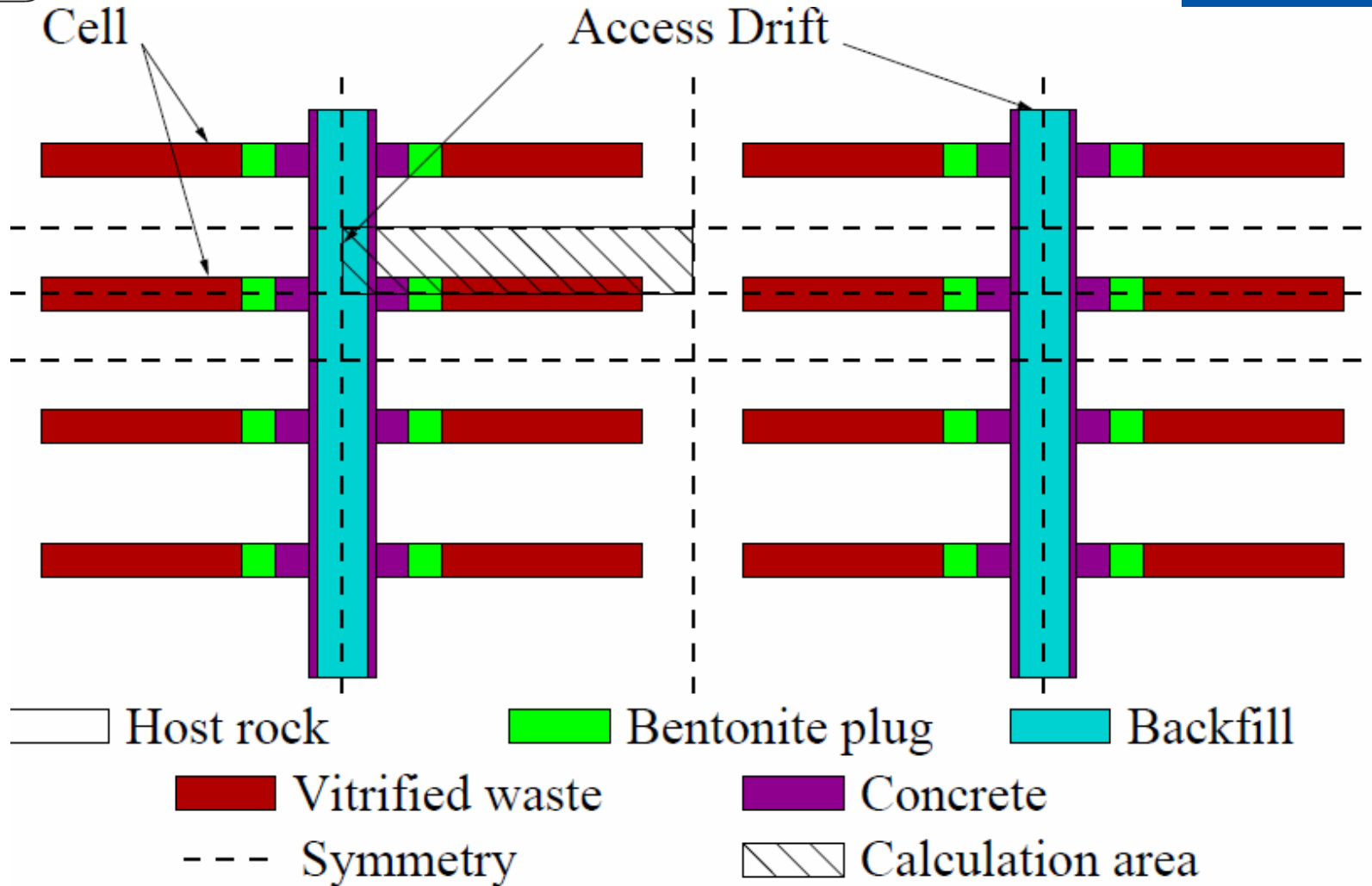


Geological disposal



- Biosphere
 - Receptor for activity fluxes
 - Local well
- Surrounding rock formations as aquifers
 - Deep local well
 - Near surface aquifer for local well
 - Travel time radionuclides to reach biosphere
 - Dilution
 - Benchmark: 10^4

Model - input



Parameters



*

*

*

Name	Radionuclide type	Molecular diffusion coefficient (m ² /s)	half-life (years)	Solubility limit (mol/l)
¹²⁹ I	Non-sorbed	1.08 10 ⁻⁹	1.57 10 ⁷	-
¹³⁵ Cs	Sorbed	0.72 10 ⁻⁹	2.3 10 ⁶	-
⁷⁹ Se	Solubility controlled	1.13 10 ⁻⁹	3.56 10 ⁵	5.5 10 ⁻⁸ x 0.085*
²⁴⁵ Cm	Decay chain	1.08 10 ⁻⁹	8500	-
²⁴¹ Pu		1.08 10 ⁻⁹	14.4	5.0 10 ⁻⁷
²⁴¹ Am		1.08 10 ⁻⁹	433	-
²³⁷ Np		1.08 10 ⁻⁹	2.14 10 ⁶	1.0 10 ⁻⁶
²³³ U		1.08 10 ⁻⁹	1.59 10 ⁵	3.2 10 ⁻⁸
²²⁹ Th		1.08 10 ⁻⁹	7340	5.0 10 ⁻⁷

Performance Assessment Methodologies in Application to Guide the Development of the Safety Case PAMINA Final report on the benchmark in clay D-N° 4.2.4 (2009) Genty, Mathieu, Weetjens

*** Lack of traceability (IAEA SSG-23) but in this case, benchmark in modelling software**

Comparison in software



- Meshing issues
- Calculated results the same

Computation time (min.)	PORFLOW 3.07	COMSOL Multiphysics 3.2
¹²⁹ I	194	3
⁷⁹ Se	195	3
¹³⁷ Cs	198	2
4N+1 actinide chain	215	12

Table 10: Indicative computation times.



Model qualification



- NEA, 2012: MeSA initiative
 - Model verification
 - Show that computer code, via numerical code, correctly implements the intended mathematical model
 - Analytical solution for ‘simple’ problems
 - Check source codes Fortran or C++
 - Software platform user defines more directly in terms of mathematical formula e.g. COMSOL is easier to verify
 - Model validation
 - Demonstrate that model correctly represents reality
 - More difficult than model verification



Model validation



- NEA, 2012: MeSA initiative e.g.
 - Is model consistent with scientific understanding?
 - Difficulties associated with model validation have contributed to the development of safety case concept, with its emphasis on multiple lines of reasoning
 - Does the model consider phenomena and interactions relevant for the assessment?



Sources for values for half-lives

- There are many sources in which the half-lives of radionuclides can be found. Experts' decision which half-lives are correct
 - the authoritative Karlsruhe Nuclide Chart, which is periodically updated by Nucleonica and the JRC for the European Atomic Energy Community.
 - compare with the Isotope Browser from the IAEA Nuclear Data Section.
- Assumed half-life can be good reason not to use primary reference



Sources for data to calculate solubility limit



- There are many sources in which the solubility limits can be found but
 - Solubility limits highly depend on pore water chemistry
 - Cementitious pore water
 - Geological formations
 - Clay pore water
 - » More details necessary than fresh or saline
 - Granitic pore water
 - » More details necessary than fresh or saline
 - Thermodynamic data updated by Nuclear Energy Agency Thermodynamic Database
 - To calculate solubility limit with assumed pore water chemistry (measurements + geochemical modelling)



Model validation



- Does gas enhanced transport of radionuclides need to be taken into account?

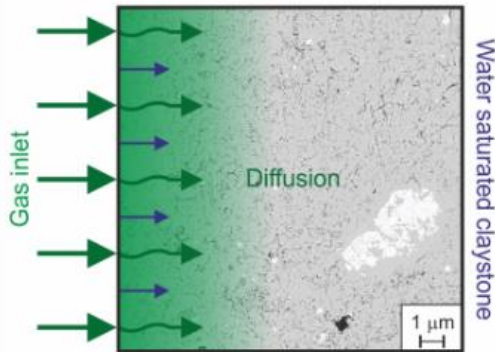


Types of waste investigated



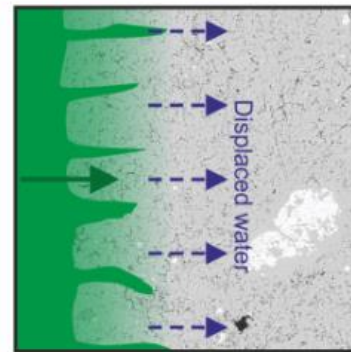
- Irradiated Steel
- Irradiated Zircaloy
- Spent ion exchange resins
- Irradiated Graphite

No gas enhanced transport of RN



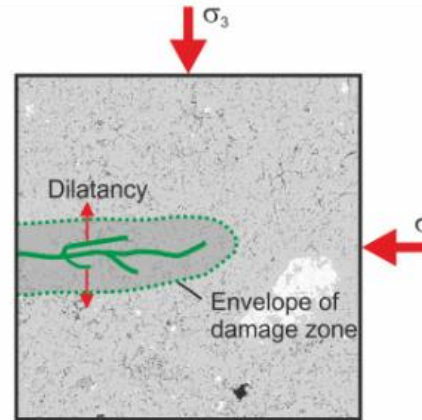
Advection and diffusion of dissolved gas

Gas enhanced transport of RN
But clay fabric intact

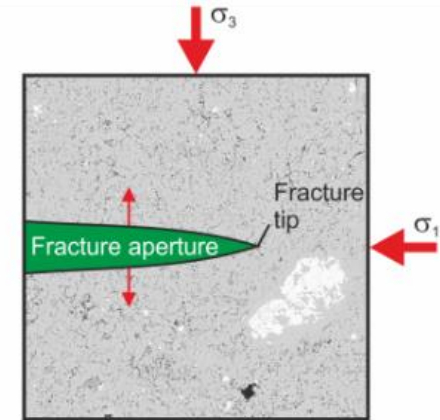


Visco-capillary flow of gas and water phase ("two-phase flow")

Gas enhanced transport of RN and clay fabric damage



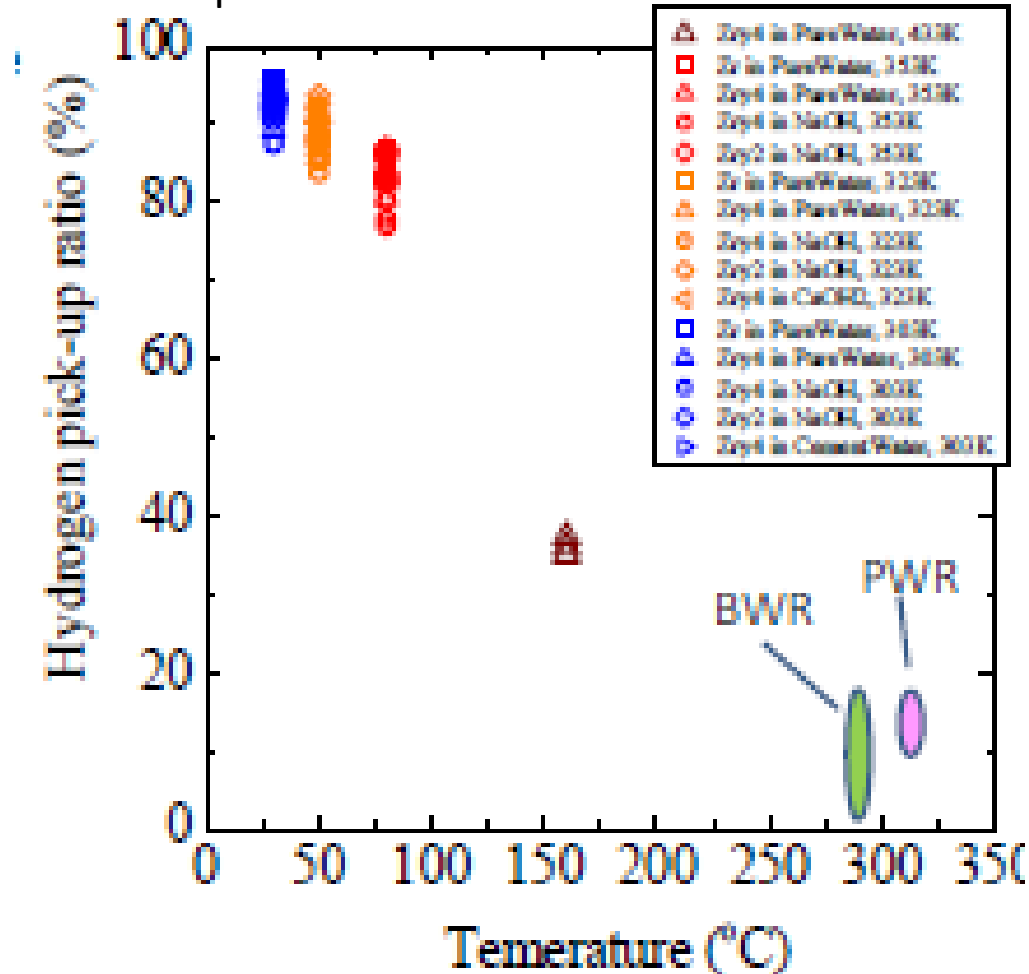
Dilatancy controlled gas flow ("pathway dilation")



Gas transport in tensile fractures ("hydro-/gasfrac")

Neutron irradiated Zircaloy

Disposal conditions



SAKURAGI, T , et al. 2013. Long-term corrosion of Zircaloy-4 and Zircaloy-2 by continuous hydrogen measurement under repository condition, Mater. Res. Soc. Symp. Proc. 1518, 173-178.

* Sakuragi T , et al. Corrosion behaviour of irradiated and non-irradiated zirconium alloys: Investigations of corrosion rate, released ¹⁴C species, and IRF (2018) CAST Final symposium; **the one used yesterday**



Neutron irradiated steel



- Corrosion rate μm per year
- Exposed surface area
- During carbon-14 release at reducing conditions also hydrogen formation





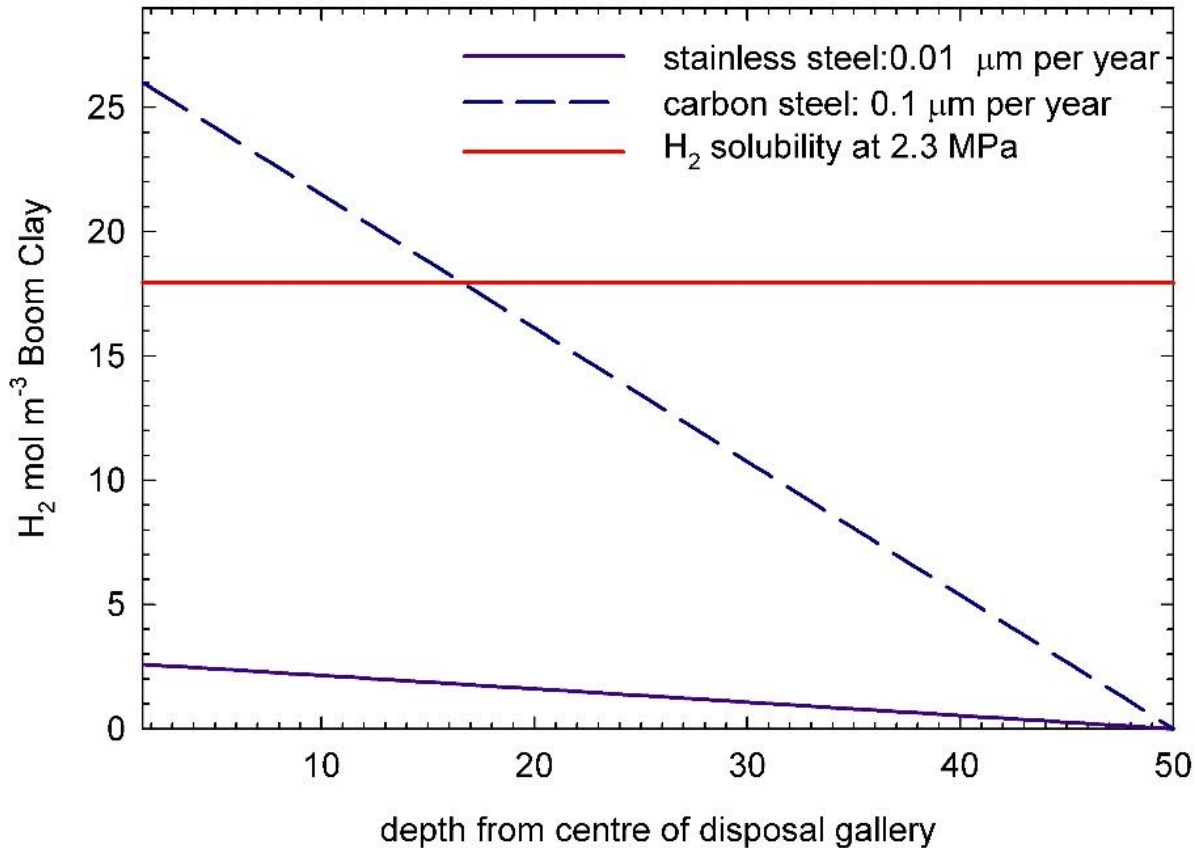
Modelling exercise



Neutron irradiated steel



1 m² reactive surface area per 1 m² interface gallery-clay
 1D linear flow; $D_{H_2} 1.1 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$



Hydrogen in Boom Clay data from Belgium programme

Yu L, Weetjens E, Estimation of the gas source term for spent fuel, vitrified high-level waste, compacted waste and MOSAIK waste, SCK•CEN ER-162 (2012) 1-59.

Neft EAC, Grigaliuniene D, Overview of achievements for regulators for workshop 2 (D7.16) (2017)

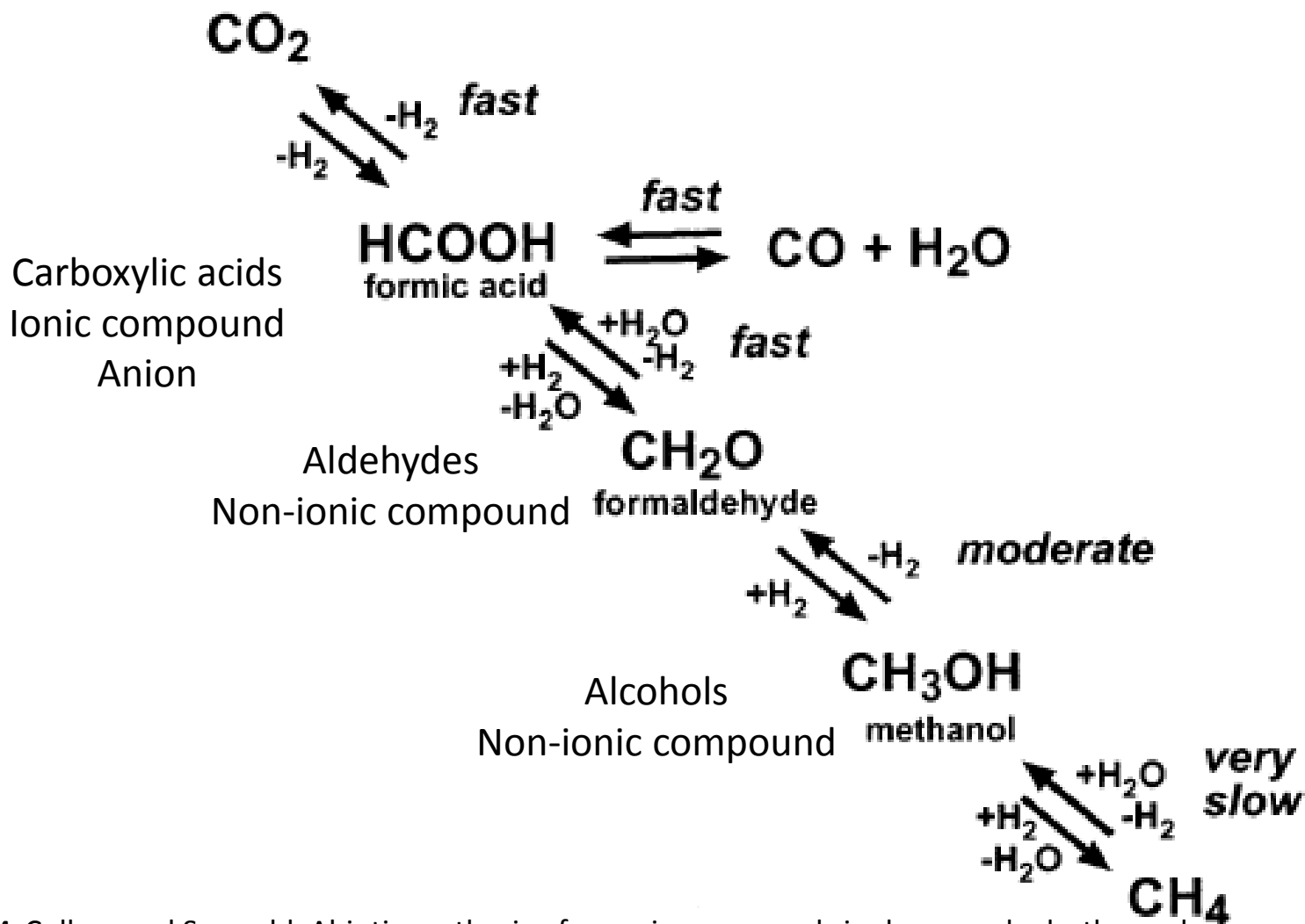


Carbon-14 species

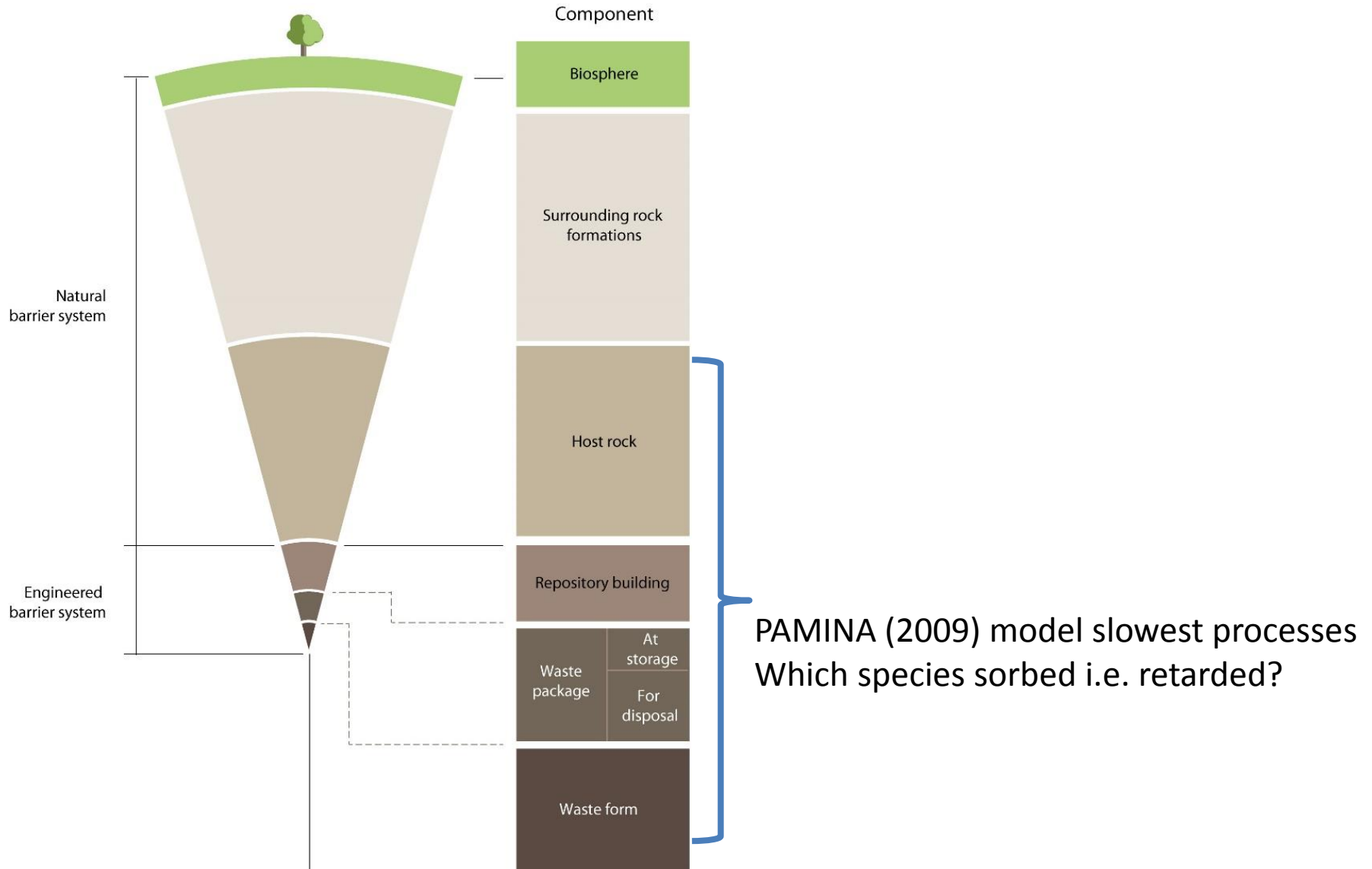


- As contained by waste form
- Released at (geological) disposal conditions

Carbon-14 species



Compartments



Retardation

Dry bulk density [kg/m³] Solid liquid distribution coefficient [l/kg]

$$R = 1 + \frac{\rho_b K_d}{\eta}$$

η Diffusion accessible porosity

$$D^i_{app} = \frac{D^i_{pore}}{R}$$

Cementitious materials



K_d [l/kg]

Table 4: Selected best estimate R_d (l/kg) values as function of cement degradation state.
[§] inorganic carbon. i.d. = insufficient data. For Ni site-specific cement data were used to calculate R_d for States I, II, and III (bold face). Values between parentheses are for high chloride background concentrations

Element	NaOH	Ca(OH) ₂	CSH	Alteration products
	State I	State II	State III	State IV
Cl	2×10^1 (1)	5×10^1 (1)	2×10^1 (1)	0 (0)
I	1×10^0 (1)	1×10^1 (1)	1×10^0 (1)	4×10^{-1} (0)
Nb	5×10^4	5×10^4	5×10^4	5×10^2
Ni	6.5×10^1	4×10^2	4×10^2	5×10^0
Am	1×10^1	1×10^1	1×10^1	1×10^1
C [§]	2×10^3	5×10^3	2×10^3	i.d.
H	0	0	0	0
Tc(IV)	2×10^3	2×10^3	2×10^3	i.d.



Cementitious materials compared to clay



- Non-retarded species
- Portland based concrete
 - Kursten, 2015 * Cl^- : 2.03×10^{-10} till 3.83×10^{-11} m²/s
 - porosity
- Boom Clay
 - D_{CH_4} 2.42×10^{-10} m²/s Jacobs, Applied Science
 - $D_{\text{HCO}_3^-}$ 6×10^{-11} m²/s Aertens, 2010, SCK external report
 - D_{I^-} 1.4×10^{-10} m²/s Bruggeman, 2010 *
- Bentonite for non-retarded species
 - Van Loon, 2007 Cl^- : D_e 10^{-11} till 3×10^{-14} m²/s
 - Density, porosity (compaction)

Carbon-14 species

- Carbonates: 'Inorganic' carbon or mineral / oxidised form
 - As dissolved species HCO_3^- , CO_3^{2-}
 - In clay as non-sorbed i.e. non-retarded species
 - In cementitious material as retarded species
 - As gaseous species CO_2
 - In clay as non-retarded species
 - In cementitious material as retarded species
- Organic carbon also called reduced form
 - As dissolved species
 - Carboxylic acids e.g. CH_3COO^- (acetate), $\text{C}_2\text{O}_4^{2-}$ (oxalate)
 - Alcohols e.g. CH_3OH (methanol)
 - Alkanes/alkynes
 - Assumed as non-retarded species for clay and cement
 - As gaseous species e.g. alkane CH_4 (methane)
 - Clay as non-retarded species
 - Cementitious material as non-retarded species

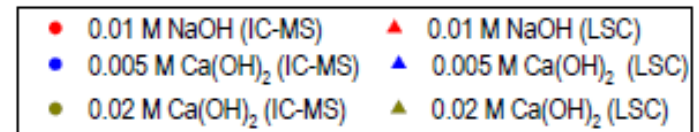
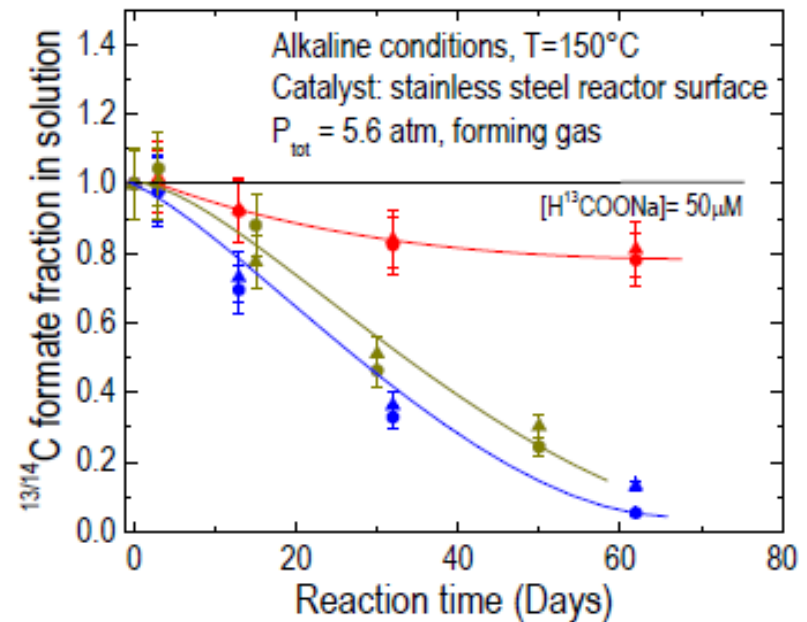


Modelling exercise



- Speciation has an impact on values for diffusion to be assumed
 - $D_{\text{HCO}_3^-}$ in clay pore water
 - D_{CH_4} as dissolved species in clay pore water

- Organic carbon-species: carboxylic acids
 - Van Loon, 1995 (PSI)
 - Ca-oxalate precipitation
 - Wieland, 2018 (PSI)
 - poster CAST Final Symposium



Van Loon, 1995: The radiolytic and chemical degradation of organic ion exchange resins under alkaline conditions: effect of radionuclide speciation, NAGRA, Technical report 95-08

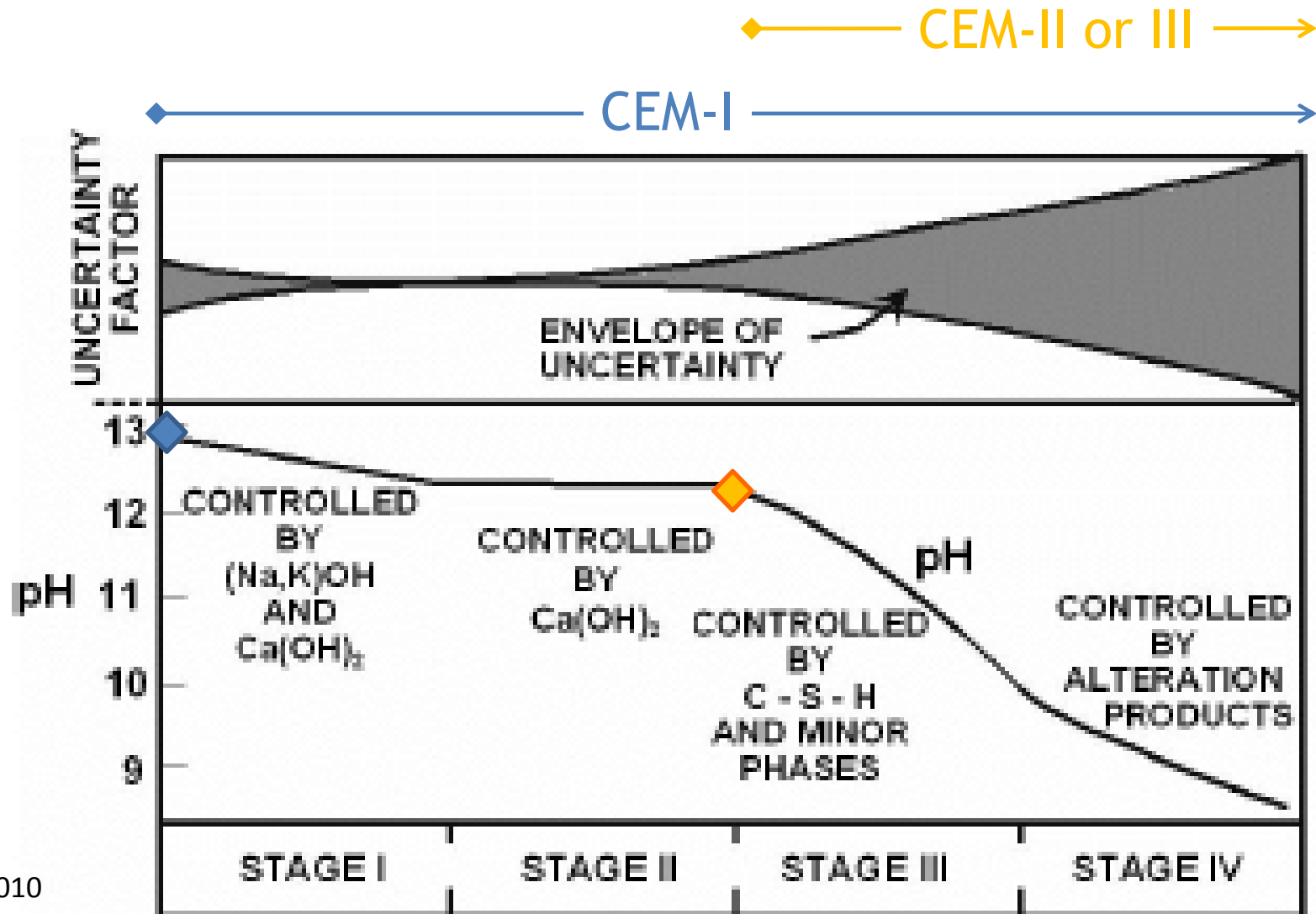


Cementitious materials



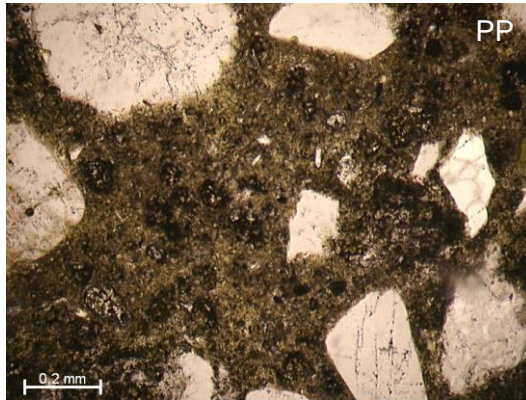
- For a long term
 - In pore water: high dissolved calcium content
 - Cementitious minerals: adsorption
 - Portlandite
 - CSH phases

Cementitious materials

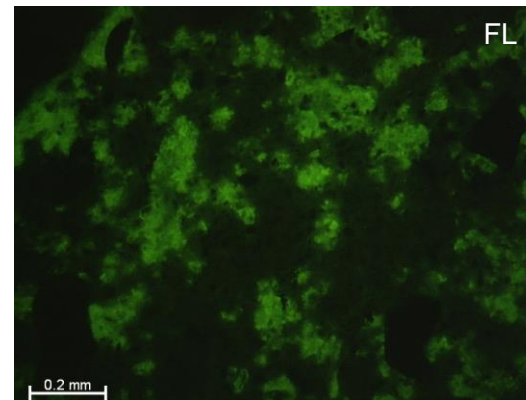
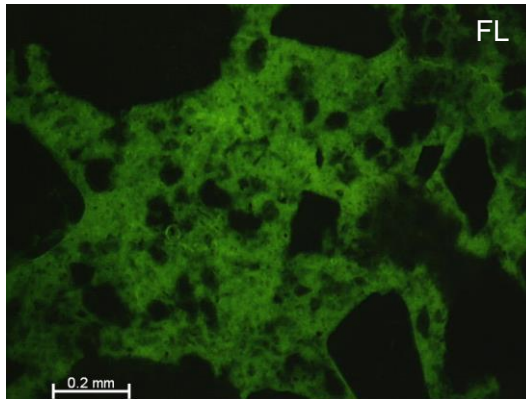
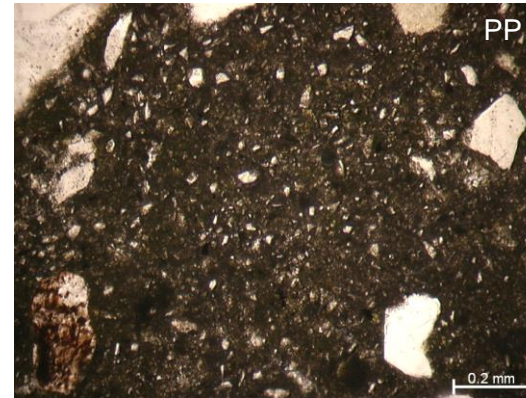


Cementitious materials

28 days old



28 days old



CEM-I

CEM-III



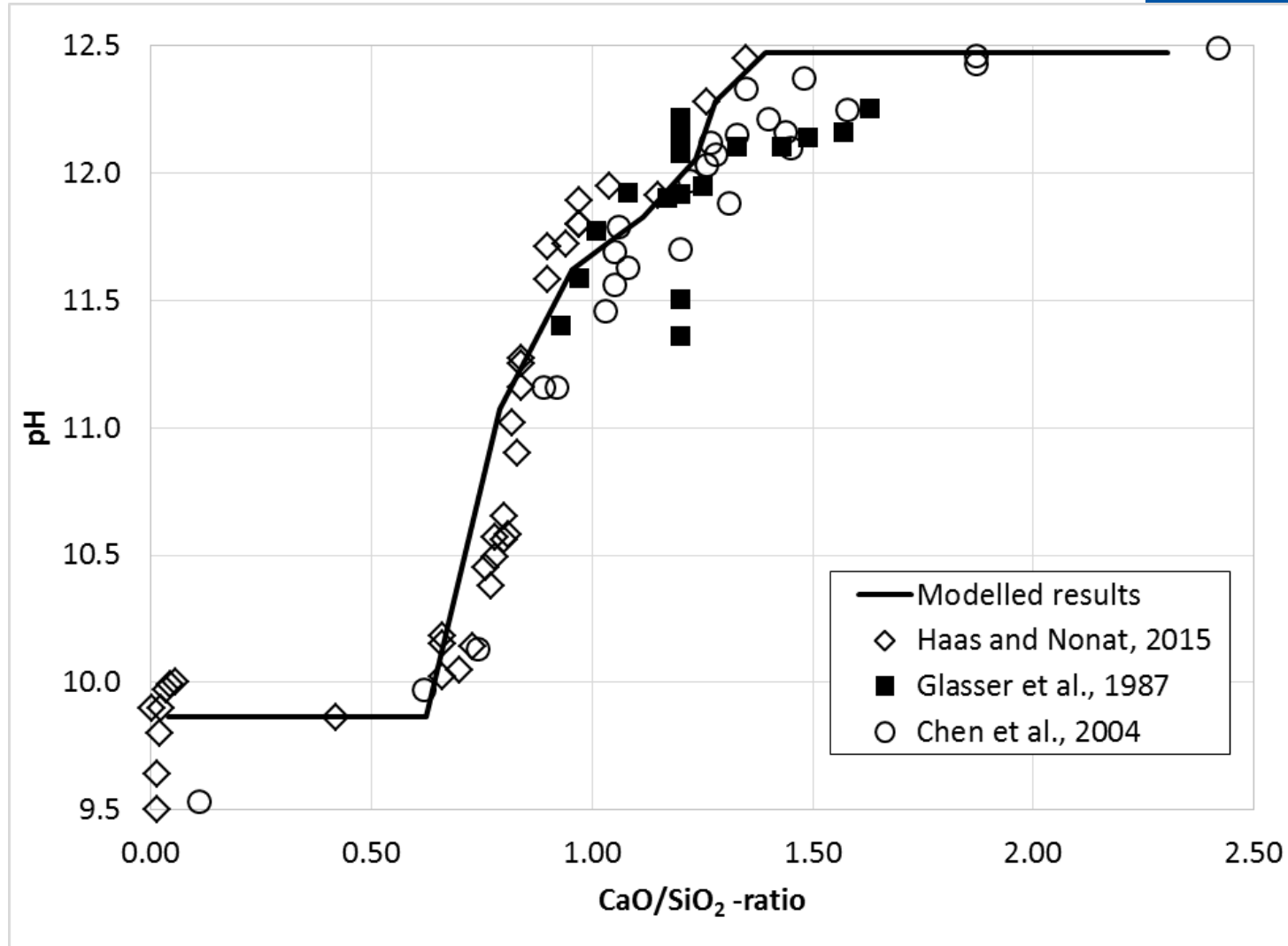
Cementitious materials

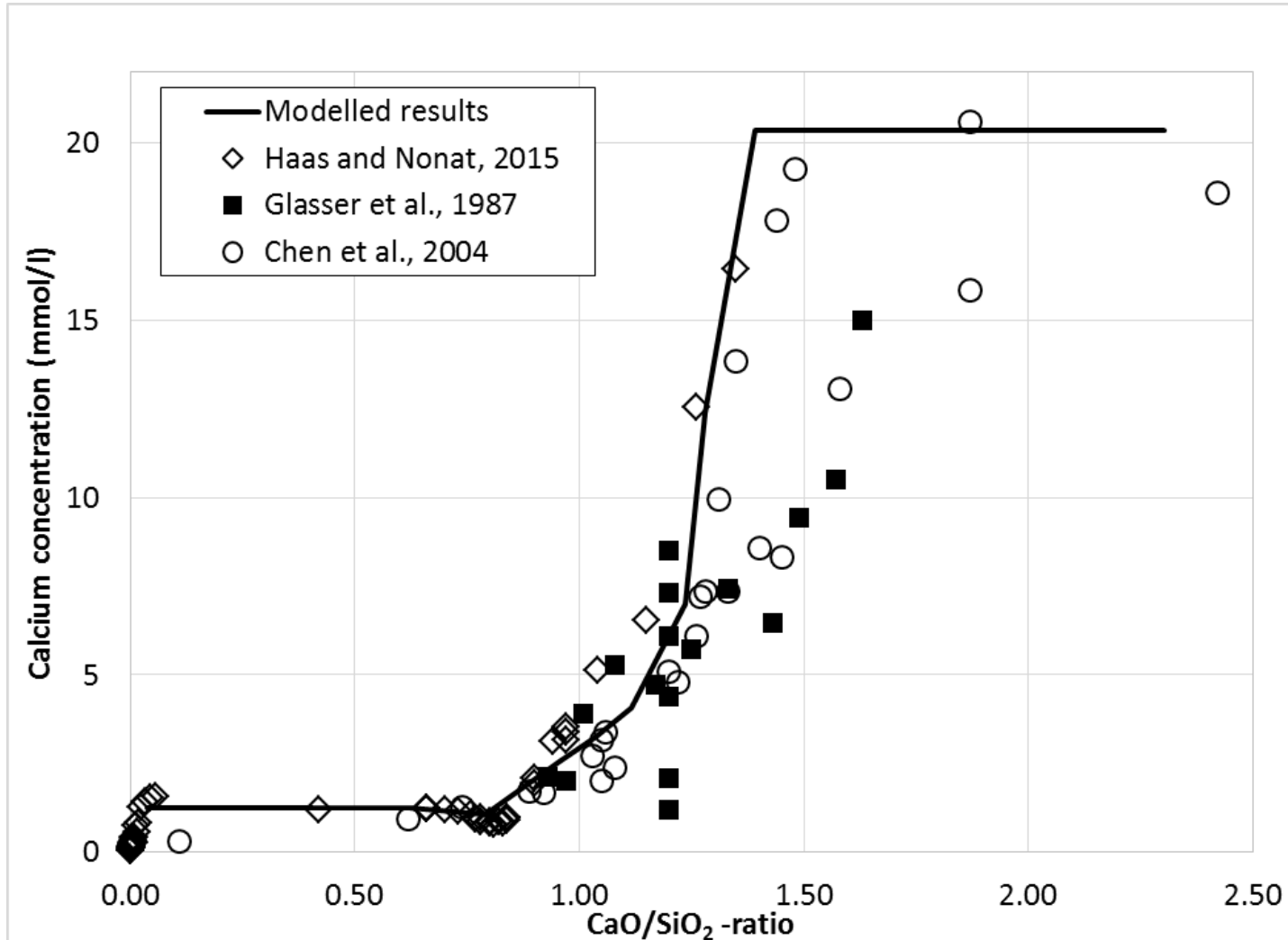


- Long term
 - Initial permeability values differ
 - Some pore water conditions may ‘last’ longer than others
 - Superplasticisers to have the smallest permeability value
 - ⇒ achieving impermeability in engineering terms i.e. the European standard EN 12390-8 e.g.
 - » One of COVRA’s concrete requirements for storage
 - » One of Posiva’s concrete requirements for disposal

Verhoef EV, de Bruin AMG, Wiegiers RB, Neeft EAC, Deissmann G, Cementitious materials in OPERA disposal concept in Boom Clay, (2014) OPERA-PG-COV023

Vehmas T*, Schnidler A, Lõija M, Leivo M, Holt E, Reference mix design and castings for low-pH concrete for nuclear waste repositories, EU Research project Cebama, First Annual Workshop (2016) Proceedings





Cementitious materials

- Near field conditions in many countries
- Microbial conditions
 - Viable microbial size: 0.2 μm
 - Size pore throat in undisturbed clay e.g. Boom Clay: 10 to 50 nm \Rightarrow space restriction
 - Microbes present but in a dormant phase
 - In cementitious materials depends on cement type in Portland based concrete 10 to 50 nm (well hydrated) but in blended cements such as cement mixed with fly ash and blast furnace slag smaller water permeability i.e. due to smaller pore throat; with superplasticifiers smaller permeability possible



BREAK





Carbon-14 species



- As contained by waste form
- Released at (geological) disposal conditions



Spent ion exchange resins



- Which type of reactor?
 - Control chemistry reactor coolant?
- Processing details?
 - E.g. drying, heating

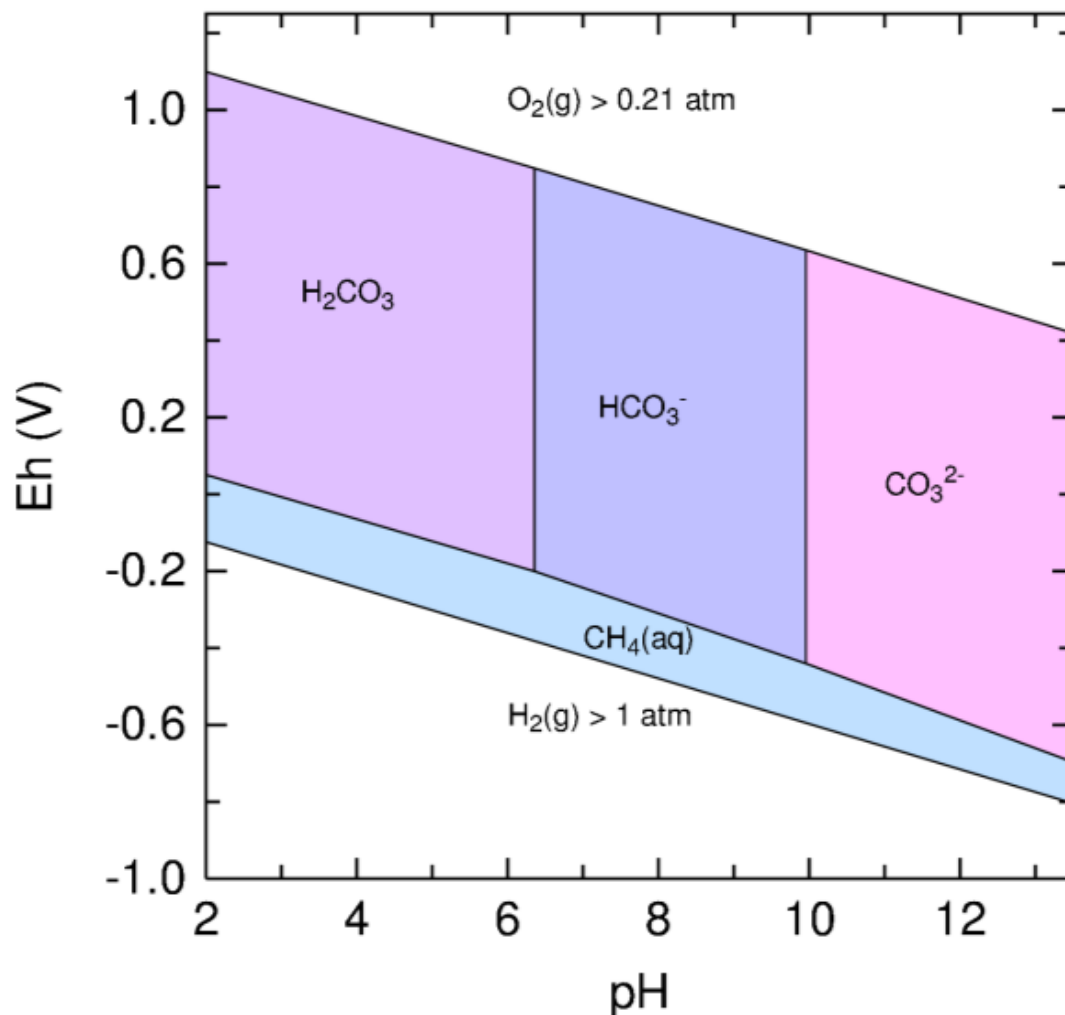


Spent ion exchange resins



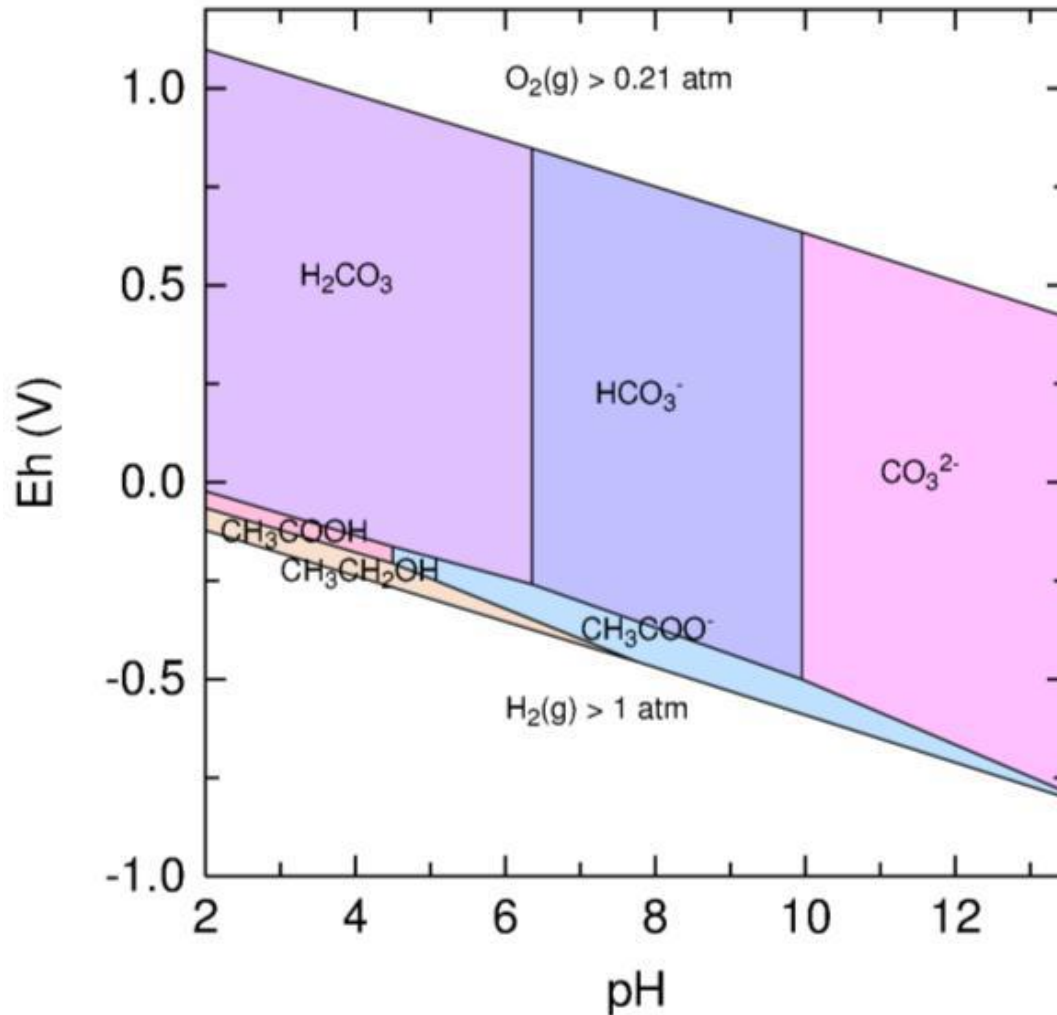
- Carbon-14 exchanged with a functional group as an anion
 - Organic: carboxyl acid e.g. oxalate, acetate, formate
 - Inorganic: carbonate, bicarbonate

Gas, dissolved



Wieland E, Hummel W, Formation and stability of ¹⁴C-containing organic compounds in alkaline iron-water systems: preliminary assessment based on a literature survey and thermodynamic modelling, Mineralogical Magazine Vol 79(2015) & Rizzato C, Rizzo A, Heisbourg G, Večerník P, Bucur C, Comte J, Lebeau D, Reiller PE, State of the art review on sample choice, analytical techniques and current knowledge of release from spent ion-exchange resins CAST report 4.1 (2015)

Dissolved



Wieland E, Hummel W, Formation and stability of ^{14}C -containing organic compounds in alkaline iron-water systems: preliminary assessment based on a literature survey and thermodynamic modelling, Mineralogical Magazine Vol 79(2015) & Rizzato C, Rizzo A, Heisbourg G, Večerník P, Bucur C, Comte J, Lebeau D, Reiller PE, State of the art review on sample choice, analytical techniques and current knowledge of release from spent ion-exchange resins CAST report 4.1 (2015)



Spent ion exchange resins



- Carbon-14 exchanged with a functional group as an anion
 - Only inorganic carbon measured from cleaning coolant and other liquids from BWR
 - Also organic carbon measured from cleaning coolant and other liquids PWR

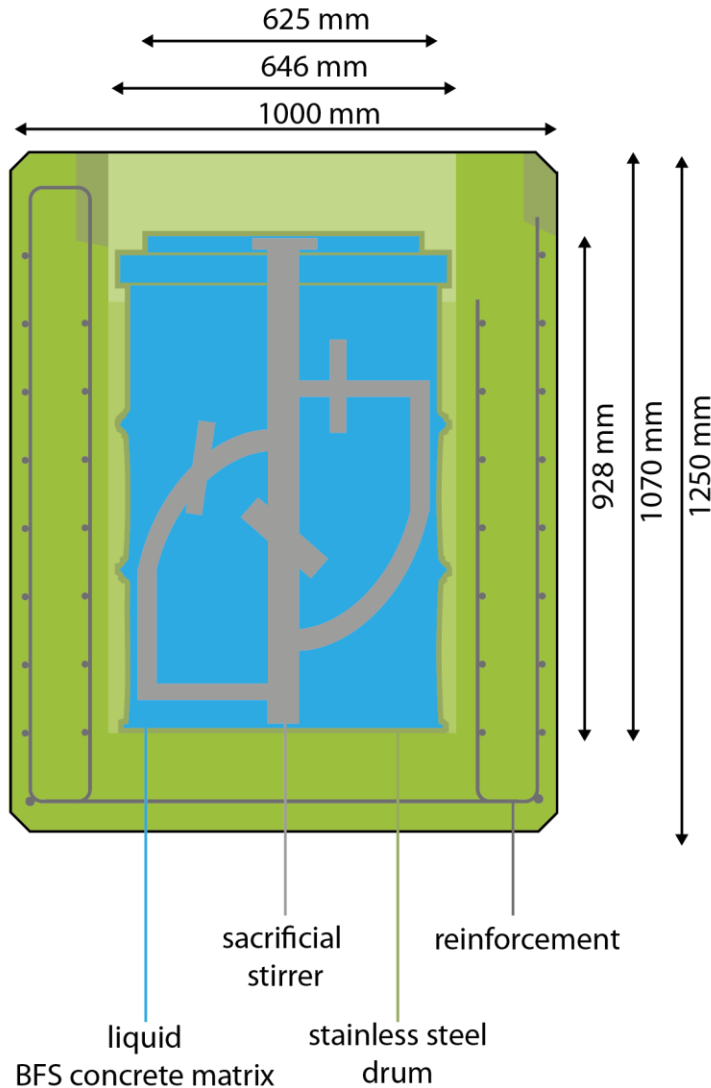


Sludge & SIER waste container

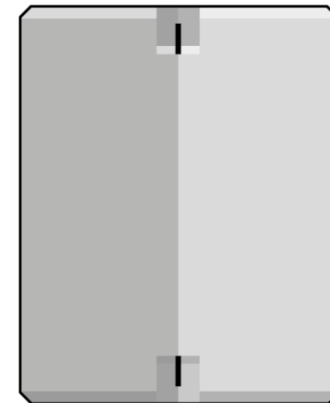
low- and intermediate-level waste



cross section



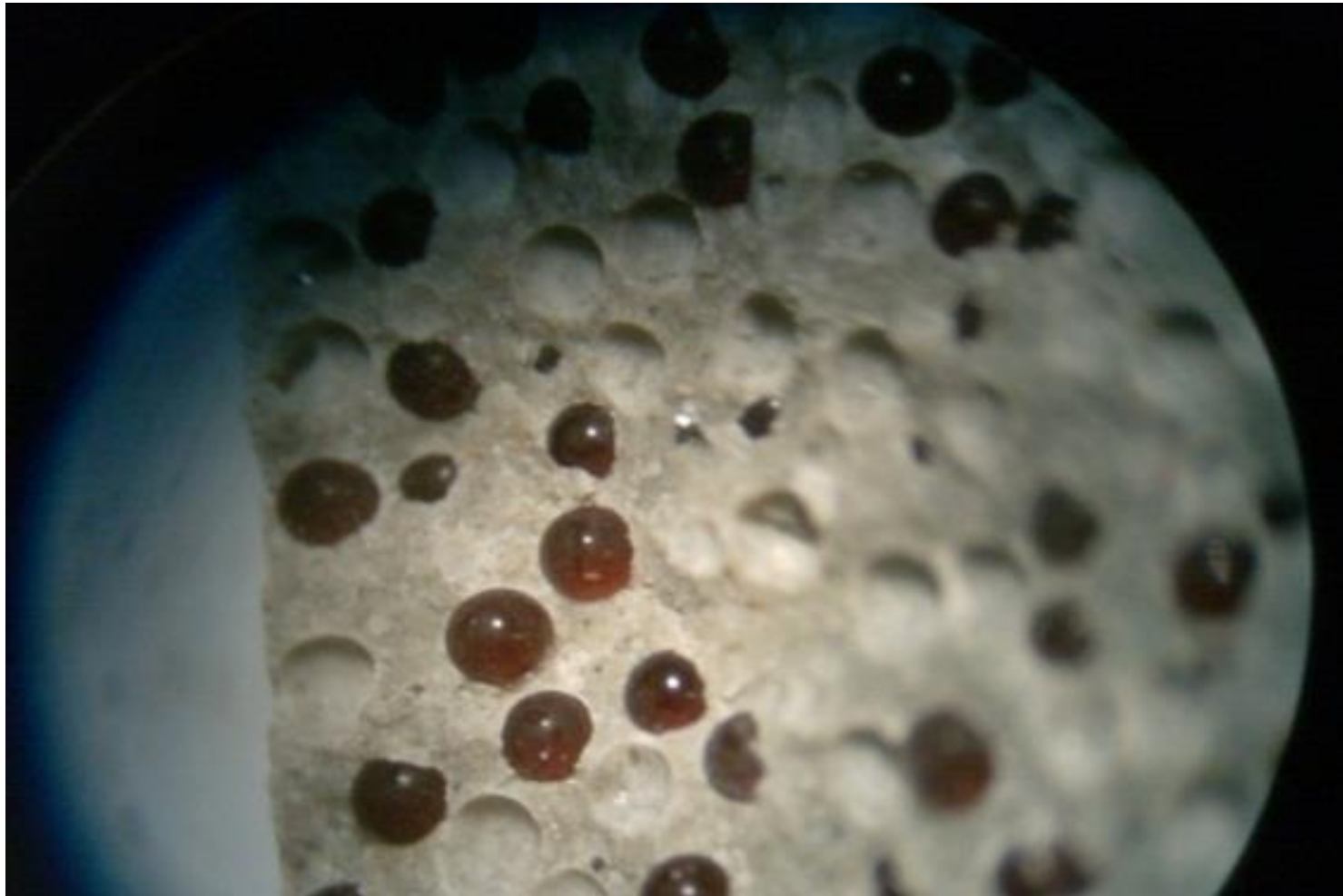
inside diameter 740 mm
inside height 940 mm



liquid I: 1,000 l magnetite
concrete container



Spent ion exchange resins



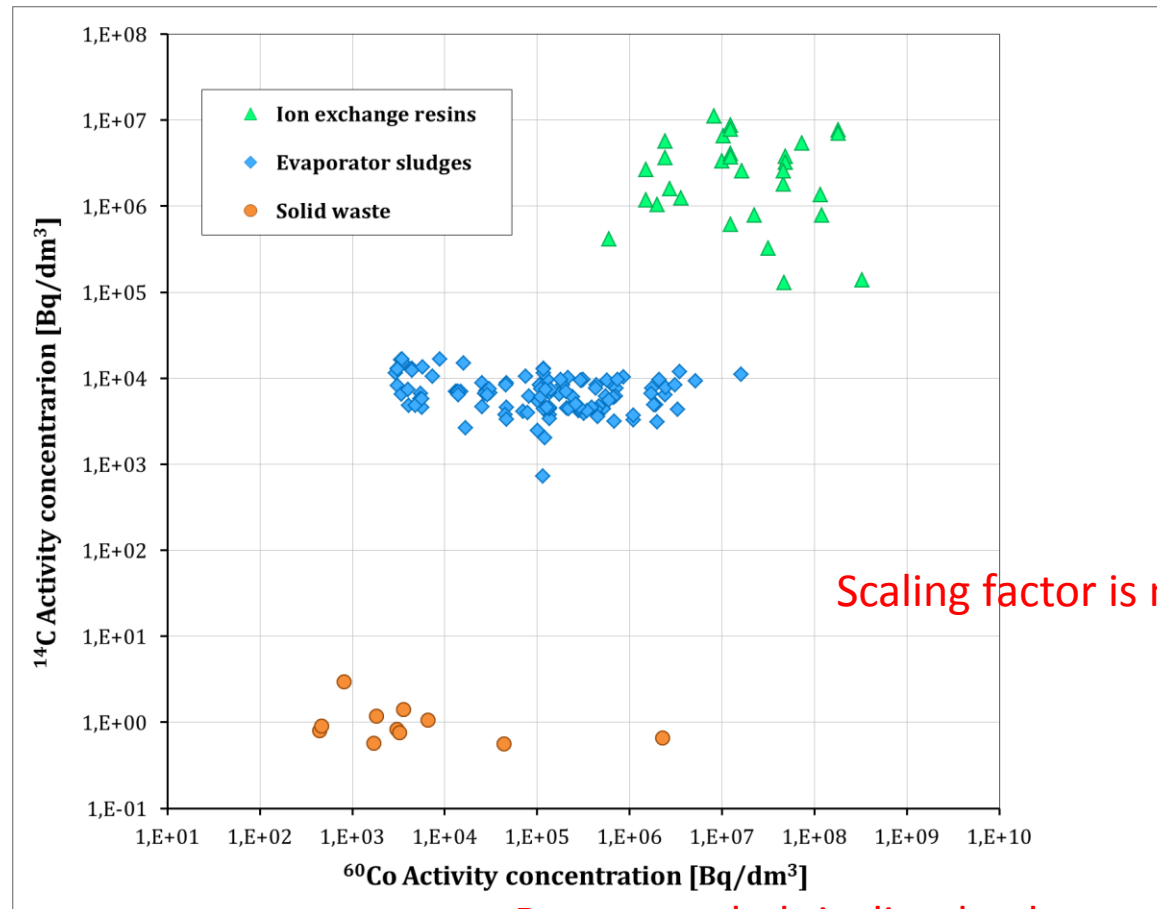


Spent ion exchange resins



- Dutch SIER : Carbon-14 content not measured
 - Scaling factor method not applicable
 - CAST workshop 1 (2016; Hungarian WMO)
 - CAST Final symposium (2018; Swedish WMO)

^{14}C and ^{60}Co activity concentration in different RW



Scaling factor is not applicable!

Because cobalt is dissolved as a cation in aqueous environments, different sorption than carbon species



Spent ion exchange resins



- Dutch SIER : Carbon-14 content not measured
 - Assumption 10^4 Bq per gram (more than maximum measured in CAST, beads)
 - Maximum resin content in Dutch processing with cementitious materials 16.8 kg for a 200 litre drum



Spent ion exchange resins



- Borssele PWR
 - Also organic carbon-14 expected
 - Sorption in cementitious materials not investigated in sufficient detail \Rightarrow no sorption
 - In CAST (Fortum Power Oy) assumed
 - Both organic (carboxylic acid) and inorganic carbon (carbonate species) can be sorbed

COMSOL

$$\frac{\partial c}{\partial t} = -\nabla(D\nabla c) + \textit{Reaction}$$

$$\frac{\partial c}{\partial t} = -\nabla(D\nabla c) - c\lambda$$

$$c(n, t) = c(n, 0)e^{-\lambda t}$$

$$c(n, t) = 0$$

Simplification , usually D expressed with porosity e.g. Weetjens, 2012

Here calculated without porosity changes and therefore not explicitly included

Carbon-14 Source Term

CAST: TRAINING COURSE 2

Name: **Jose Luis Leganes Nieto**

Organisation: **ENRESA**

Date: **22th Feb 2017**



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.



NPP Decommissioning:

Characterization and infer into a suitable Waste Form



- 1- Introduction
- 2.- Characterization stages
- 3.- Site&Facility Characterization
- 4.- Materials classification in dismantling
- 5.- Radioactive waste conditioning
- 6.- Material release
- 7.- Surface release
- 8.- Site release



NPP Decommissioning:

Enresa introduction



- State-owned company
- Responsible, by law, of
 - a) The management of all the radioactive waste produced in Spain (NPP, hospital, research centres, ...)
 - b) The decommissioning of Nuclear Installations
(after post-operational activities performed by the former operator)
- Owns and operates a LILW disposal facility (El Cabril)
- Obtains funds from Waste producers / NPP owners
- Responsible to manage funds and liabilities, in accordance with a periodical 'Activity Plan' approved by the government.



NPP Decommissioning:

Enresa introduction



High Level Waste (HLW)



Dismantling of Nuclear Facilities



Very Low Level Wastes (VLLW) and Low and Intermediate Level Wastes (LILW)



NPP Decommissioning:

Enresa introduction



EI CABRIL

It's the disposal center of radioactive wastes of low, very low and intermediate level that Enresa operates in Cordoba.

In this facility are disposed of wastes proceeding from hospitals, research centers and nuclear facilities.



NPP Decommissioning:

Enresa dismantling projects



VANDELLÓS 1

1998 / 2003



PIMIC

2006 / 2015



Jose Cabrera

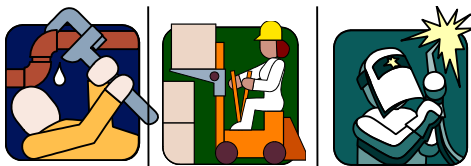
2010 / 2018

NPP Decommissioning:

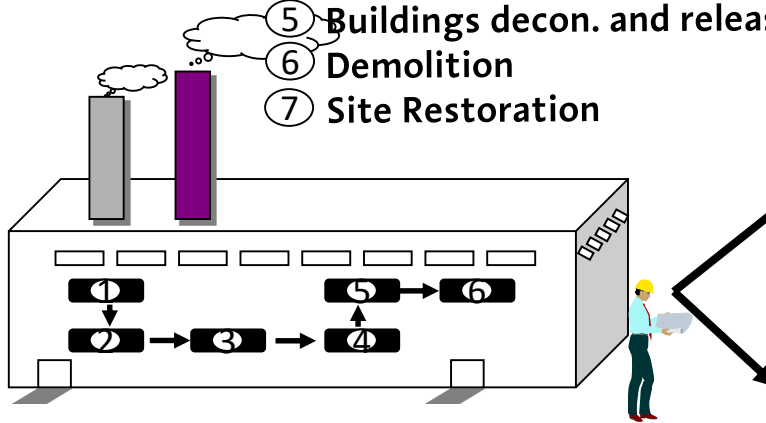
Dismantling as an Industrial Process



RESOURCES



- ① Facility Characterization
- ② Equipment Retrieval
- ③ Materials declassification
- ④ Dispatching
- ⑤ Buildings decon. and release
- ⑥ Demolition
- ⑦ Site Restoration



MATERIALS



DISMANTLING is not only DEMOLITION, but an INDUSTRIAL PROCESS



NPP Decommissioning:

Three Main Characterization Stages



Site&Facility Characterization: the objective is to obtain a radiological image of the whole Installation (site and facility) or at least the locations where it is possible to be accessed for characterization.

In situ Characterization for Classification: it can be considered as an operational, and sometimes rough, characterization for disassembles, cleaning or remediation activities that allow a quick and therefore an operative way of classifying the materials.

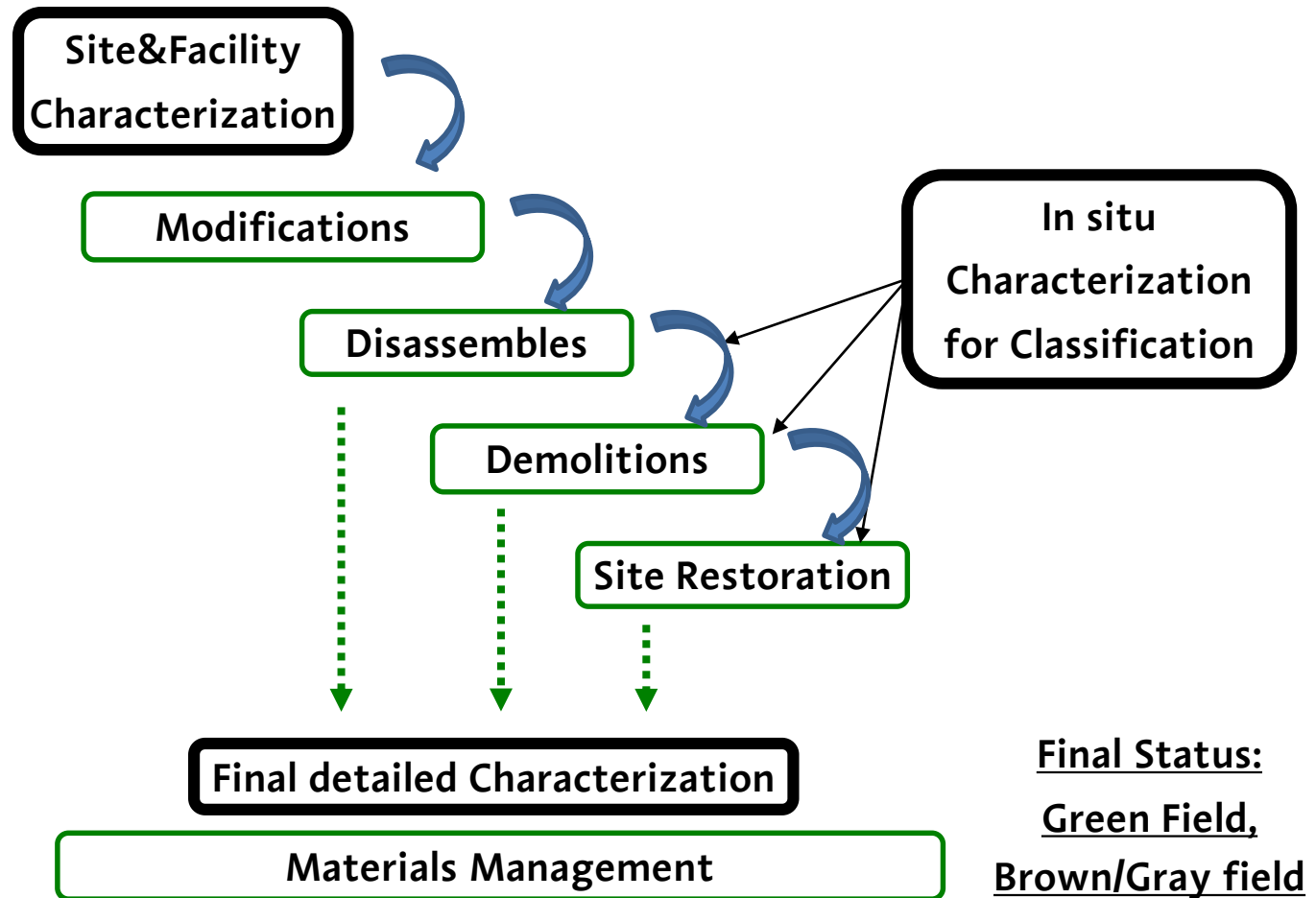
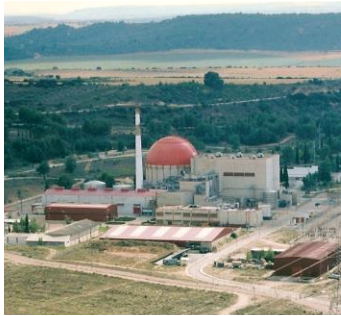
Characterization for Final Assignment/Assessment: it is an as thorough as possible characterization that would define the final destination of the materials, or the release objective fulfillment.

NPP Decommissioning:

Characterization Stages Outlook



Transfer of Ownership





NPP Decommissioning:

Site&Facility Characterization



- **HISTORICAL DATA**

Old nuclear installations do not usually have enough radiological data, if any, they are lack of useful information.

Operational data should be also used to develop isotopic vectors.

- **PREVIOUS CHARACTERIZATIONS BEFORE DISMANTLING STAGE**

Taken during or after operational stage.

Site specific measurement, different from those which were taken during operational life.

- **ESTIMATION OF VECTORS PRIOR FINAL CHARACTERIZATION**

Based both on previous measurements and operational features of the nuclear installation.

Source term of the installation should be developed, in order to know which isotopes have to be measured.

- **ISOTOPIC VECTORS (I.V.)**

What do we mean by Isotopic Vector?

- Same source of contamination?
- Same origin?
- Same waste stream?

ALL MATERIAL WITH THE SAME ISOTOPIC COMPOSITION

- Depends on the source of contamination
- Depends on the physical-chemical processes
- Depends on the nature of the material



NPP Decommissioning:

Site&Facility Characterization: Isotopic Vector



- **ISOTOPIC VECTORS**

What do we mean by same isotopic composition:

- ❑ No significant differences from qualitative point of view, supported by graphical tools.
- ❑ No significant differences from statistical point of view.
- ❑ No significant differences from others under your technical considerations.

**ISOTOPIC VECTORS OBTAINED FROM SEVERAL MEASUREMENTS
INSTEAD OF FROM ONE ONLY MEASUREMENT.**



NPP Decommissioning:

Site&Facility Characterization: Isotopic Vector



- **ISOTOPIC VECTORS PRIOR FINAL CHARACTERIZATION**

It is better to first hypothesize more I.V. than finally expected, as a function of

- ❑ Operational features
- ❑ Nature of the material
- ❑ Source of contamination
- ❑ Physical-chemical mechanisms

Once the I.V. has been hypothesized, the next step is to collect samples from every I.V. location in order to find out the actual and final I.V.

DESIGN OF FINAL CHARACTERIZATION



NPP Decommissioning:

Site&Facility Characterization: Sampling Process



- **PERFORM A COMPLETE RADIOLOGICAL MAP OF THE INSTALLATION**
Using mobile instrumentation
 - ❑ Contact dose rate measurement
 - ❑ Contact Beta
 - ❑ Contact Alfa
 - ❑ Others in situ devices
- **COLLECT SAMPLES (FOR RADIOCHEMICAL ANALYSIS) AS A FUCTION OF**
OF
 - ❑ Lower or higher values of dose rate?
 - ❑ Lower or higher values of Alfa, Beta?

• BRIEF SCALING FACTORS (S.F.) SUMMARY

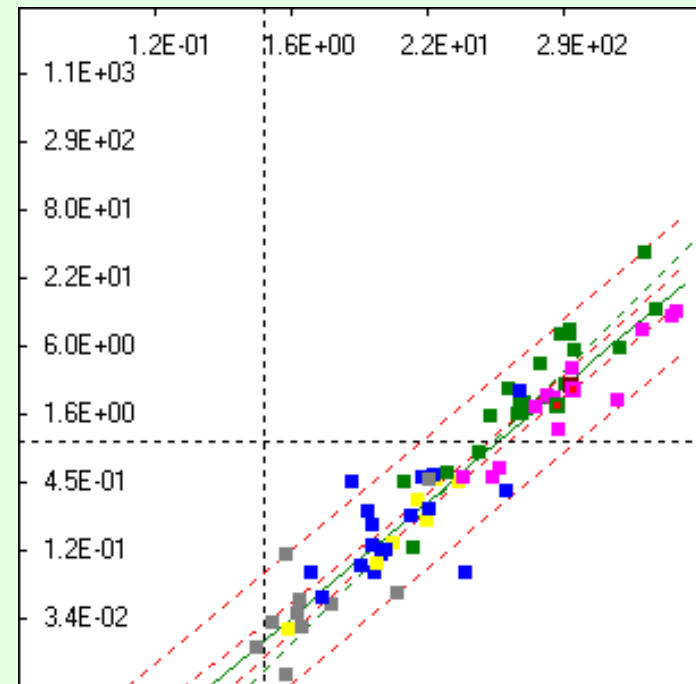
Finding correlation between difficult and easy to measure isotopes (Key Nuclides).

key Nuclides (K.N.):

- Gamma emitter easily detected for any gamma spectrometry.
- Relatively high half-lives (Co-60 and Cs-137).

Difficult to measure Isotopes us K.N.:

- Activation Products (AP), or Fission Products (FP) .
- Similar solubility.
- Similar transport process.



NPP Decommissioning:

Site&Facility Characterization: Scaling Factors



- **SCALING FACTORS, REPRESENTATIVE OF GREAT ACTIVITY RANGE**

Applicable to:

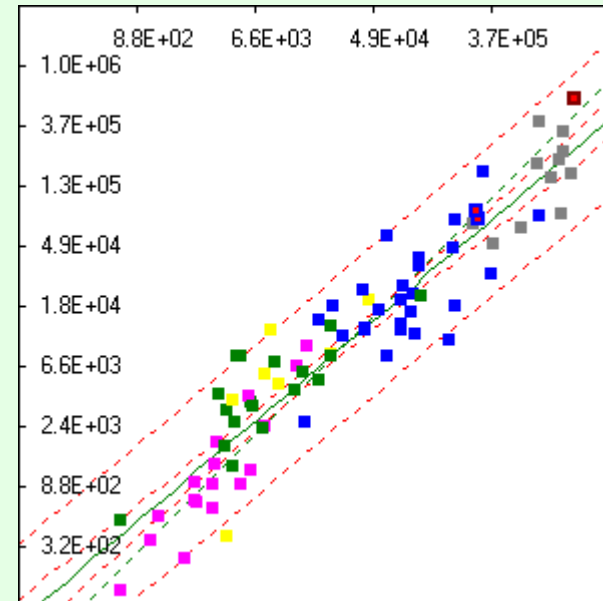
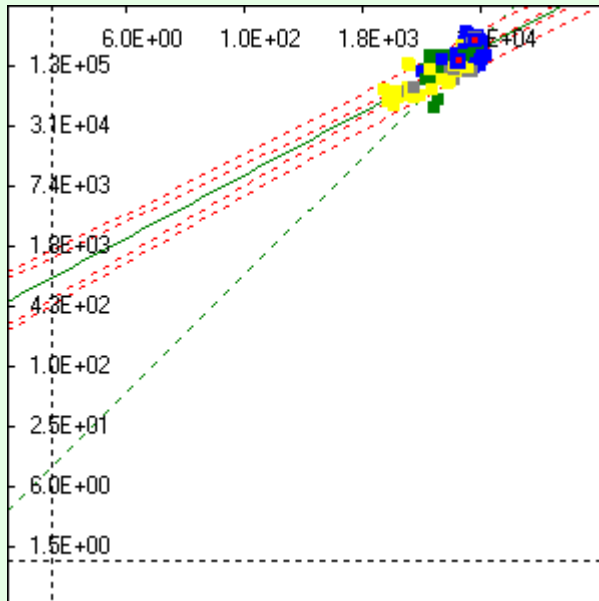
- Intermediate and Low Level Waste (I&LLW).
- Very Low Level Waste (VLLW).
- Clearance purposes, materials than can be release from regulatory control.

**THE CHARACTERIZATION DESIGN SHOULD BE
PLANNED IN ORDER TO OBTAIN A REPRESENTATIVE
SCALING FACTORS**

- **REPRESENTATIVENESS OF SCALING FACTORS**

Collect samples with a great range of dose rate, contact Beta, etc.

- ❑ Radiochemical analysis are expensive.
- ❑ Radiochemical analysis are time consuming.
- ❑ Collect samples above Detection Limit (dose rate, Beta, etc.).





- **MANNER OF SAMPLING**

Ways to improve the manner of sampling:

- ❑ The first way has been mention before, collecting samples with a great range of dose rate, contact Beta, etc..
- ❑ The second way, by Increasing the mass of the sample to be analyzed, how? COMPOSITE SAMPLE

Once the data range have been ensured, samples from specific I.V. with a similar dose rate (or contact beta, etc.), can be merged and therefore one only sample is created for radiochemical analysis, instead of sending them in a separate manner for radiochemical analysis.

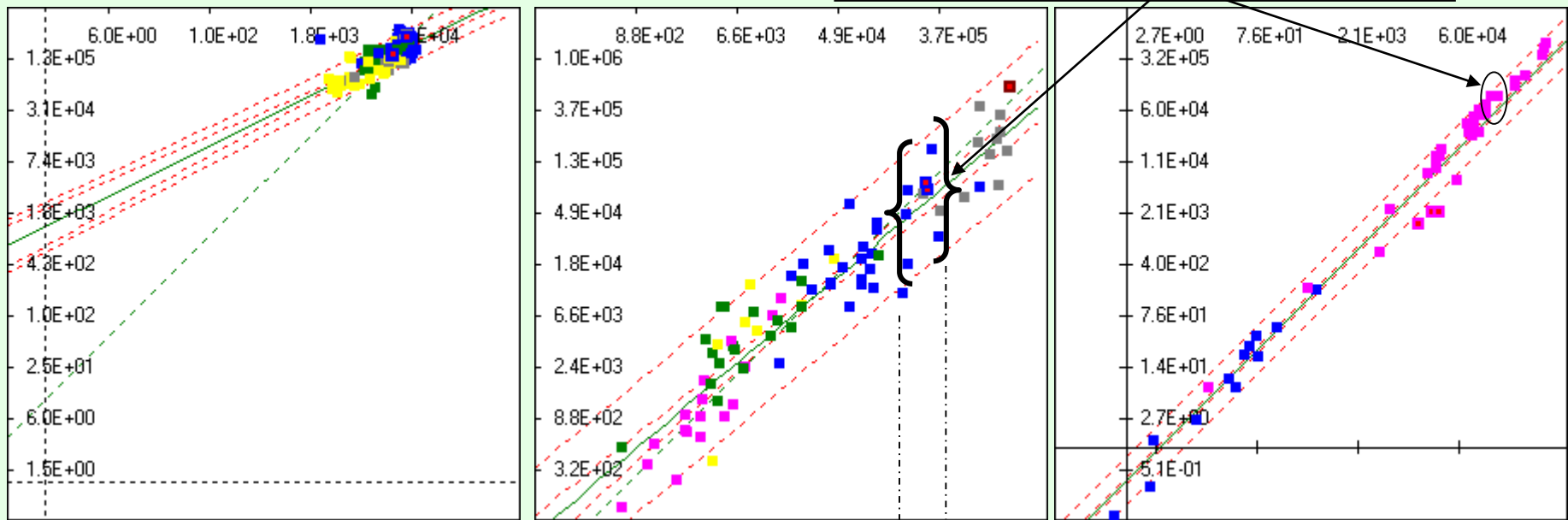
NPP Decommissioning:

Site&Facility Characterization: Representativeness



REPRESENTATIVENESS IMPROVEMENT OF SCALING FACTORS

Composite samples. Lower number of samples for radiochemical



Increasing accuracy

Decreasing uncertainty



NPP Decommissioning:

Site&Facility Characterization: SF issues



- **BUILDING ISOTOPIC VECTORS**

Main features to be taken into account:

- ❑ Consistent I.V. are obtained by finding correlation between isotopes.
- ❑ Scaling Factors search for correlation between easy to measure (K.N.) and difficult to measure ones.
- ❑ Many times, easy to measure isotopes well correlate between them. Also difficult to measure isotopes well correlate between them. Some advantages are obtained of these ratios.
- ❑ Radiochemical measurements are very expensive, try to use also operational data. Therefore special attention should be paid for decaying data.



NPP Decommissioning:

Site&Facility Characterization: SF issues



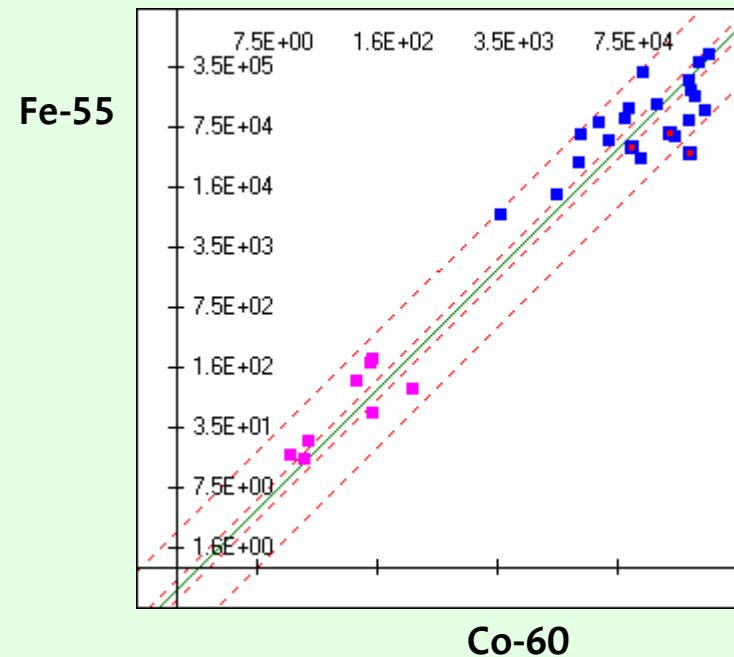
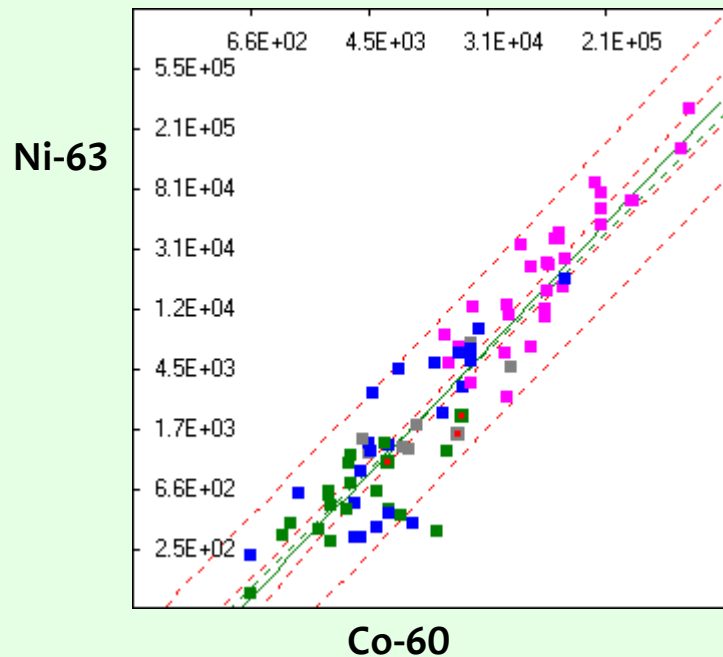
- **BUILDING ISOTOPIC VECTORS**

Built from Scaling Factors and easy to measure ratios:

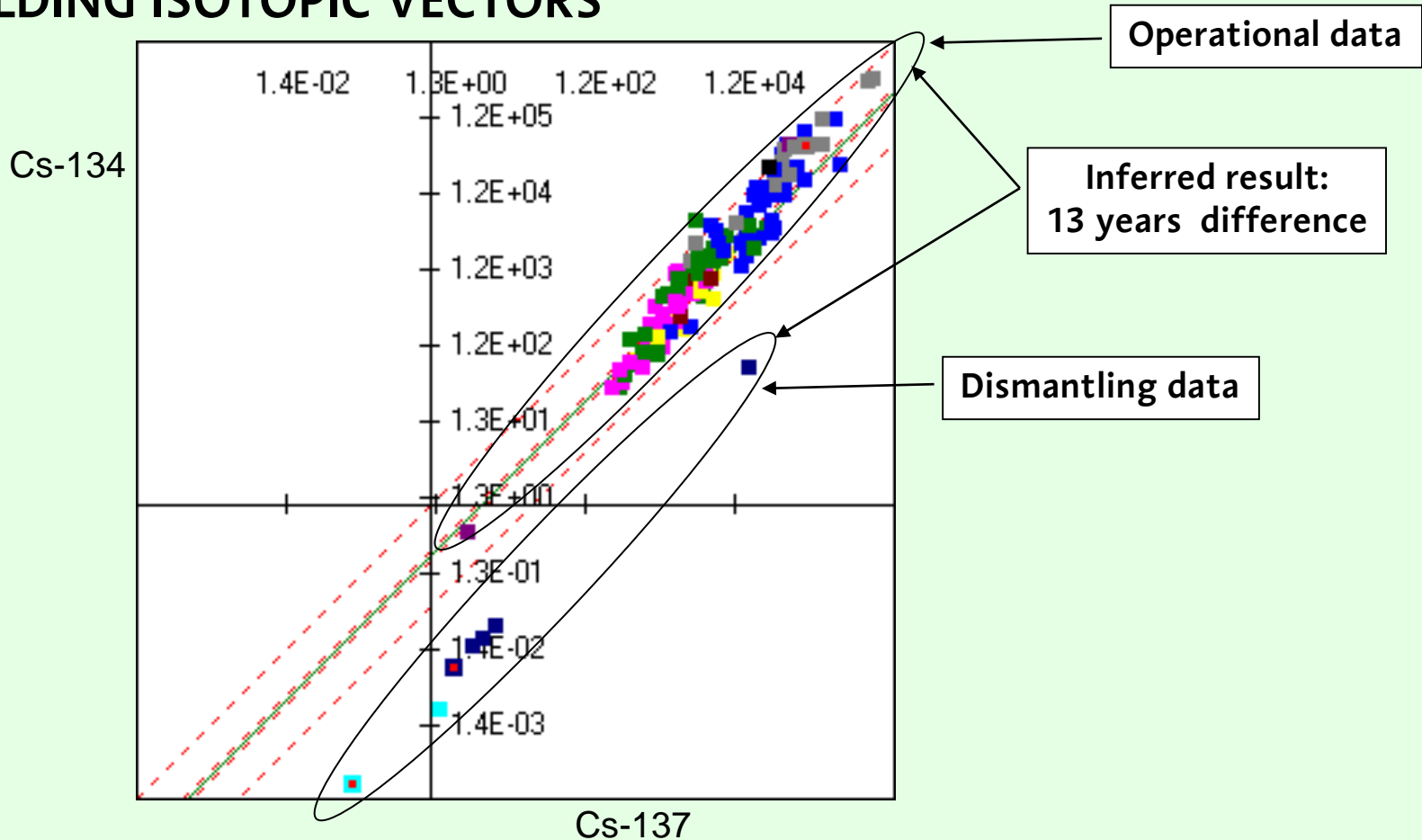
- ❑ Many S.F. or ratios do not show significant variations in different materials nature, due to similar chemical behavior (Ni63/Co-60, Mn-54/Co-60, Fe-55/Co-60).
- ❑ Many S.F. or ratios cannot show variations in different materials nature, due to the same chemical behavior (Ni59/Ni-63, Cs134/Cs137, Co57/Co-60). Useful information in order to guess the age of the studied material (the contamination date).
- ❑ Am-241 is mainly due to Pu-241 decaying, if the operational ratio is known, the current S.F. for the studied material could also give information of its age.

- BUILDING ISOTOPIC VECTORS**

- Many S.F. from different hypothesized I.V. show no significant differences.



BUILDING ISOTOPIC VECTORS





NPP Decommissioning:

Site&Facility Characterization: Similar SF



- **I.V. FINAL RESULTS**

- ❑ If several hypothesized I.V. have finally showed both the same S.F. and ratios, for every radio nuclides, they all can build a unique I.V.
- ❑ Some I.V. only differs from other in one S.F. or one ratio.
- ❑ Some I.V. can have different isotopes from others as a function of their relative abundances.
- ❑ It could be useful to first work with Gamma I. V. and finally use the total I.V.



NPP Decommissioning:

Site&Facility Characterization: Applicability

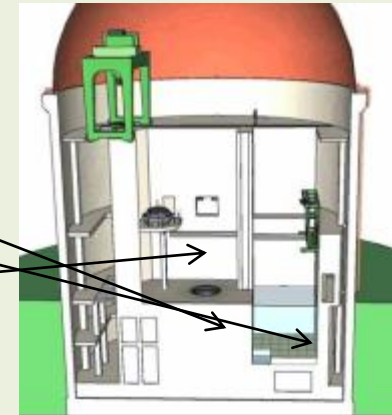


- **MAIN USAGES OF ISOTOPIC VECTORS**

- I&LLW PACKAGES
- VLL PACKAGES
- CONTAINERS WITH MATERIALS FOR CLEARANCE
- SURFACES AND BIG PIECES FOR CLEARANCE
- SOILS RELEASE FROM REGULATORY CONTROL
- RADIOLOGICAL PROTECTION
- ENGINEERING PROCESSES

DIFFERENT ISOTOPIC COMPOSITION FOR NPP NPP

- Biological shielding
- Spent Fuel Pool Materials.
- Refuel Cavity.
- Evaporator.
- Waste Package Storage N° 1.
- Outsides, Site Restoration.
- Rest of places/systems.



Main isotopes: Co-60 and Cs-137



NPP Decommissioning:

Materials: Radiological Classification



CONVENTIONAL.

Materials arising from zones not having radiological implications
(Conventional areas)

DECLASSIFIABLE.

Materials arising from controlled areas which, given their operating and radiological background, the plant radiometric studies and the characterizations performed during disassembly, are candidates for management as conventional materials. For this purpose, they are required to have levels of activity below those authorized by the regulatory authority (CSN).

RADIOACTIVE WASTES.

Wastes arising from radiologically controlled areas.



NPP Decommissioning:

Jose Cabrera Material Production



≈ 104.000 tons

Initial Situation



95.300 t

Concrete & Demolitions



Spent Fuel



175 t

Conventional Scraps



4.700 t

Recycling



Small Quantities

Toxic & Hazardous



Controlled LandFill

RadW



4.000 t

4% - Radioactive Waste Management

12 HI-STORM



All the materials have a final destination

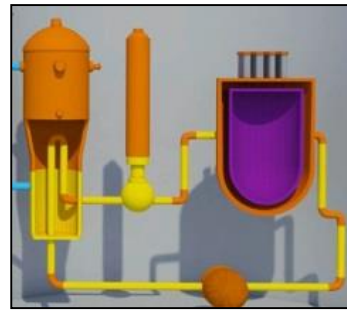


Final Status



NPP Decommissioning:

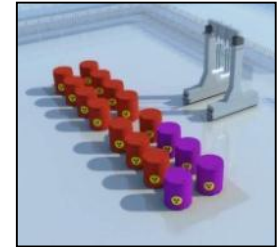
Jose Cabrera Radioactive Material Routes



PRIMARY CIRCUIT

40 t

4 HI-SAFE



ISFSI



CE-2a

+



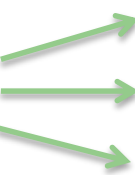
EL CABRIL

360 t



CE-2b

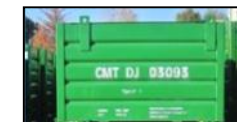
REST OF THE INSTALLATION



CMB



VLLW LARGE ITEMS



CMT

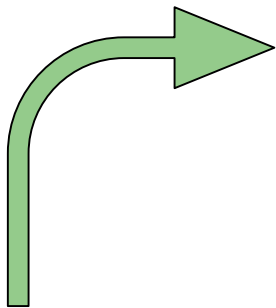


DRUM 220 L

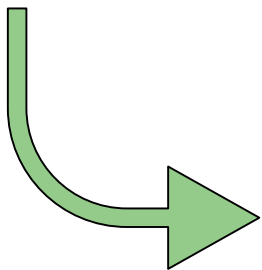


EL CABRIL

400 t

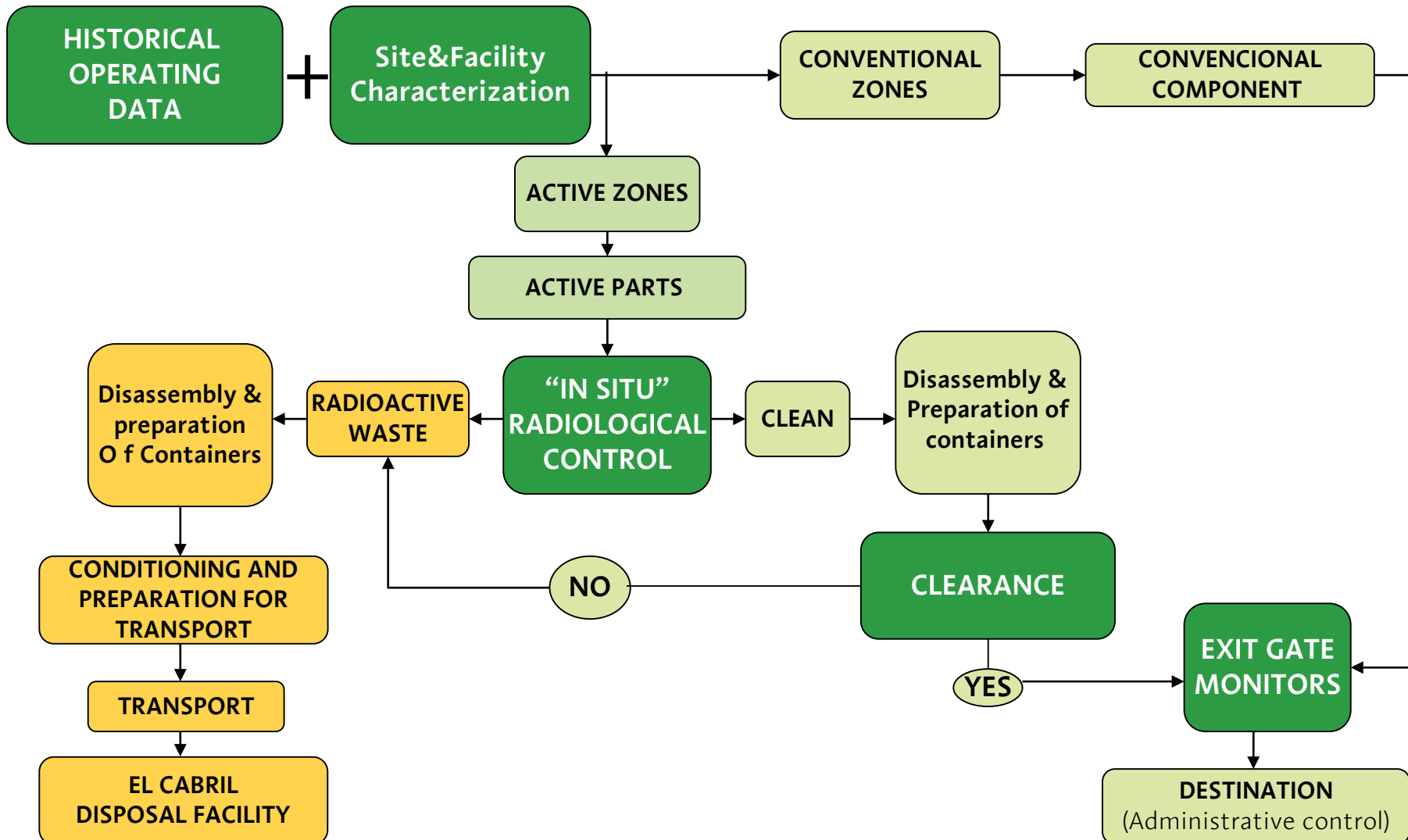


≈ 4.000 t



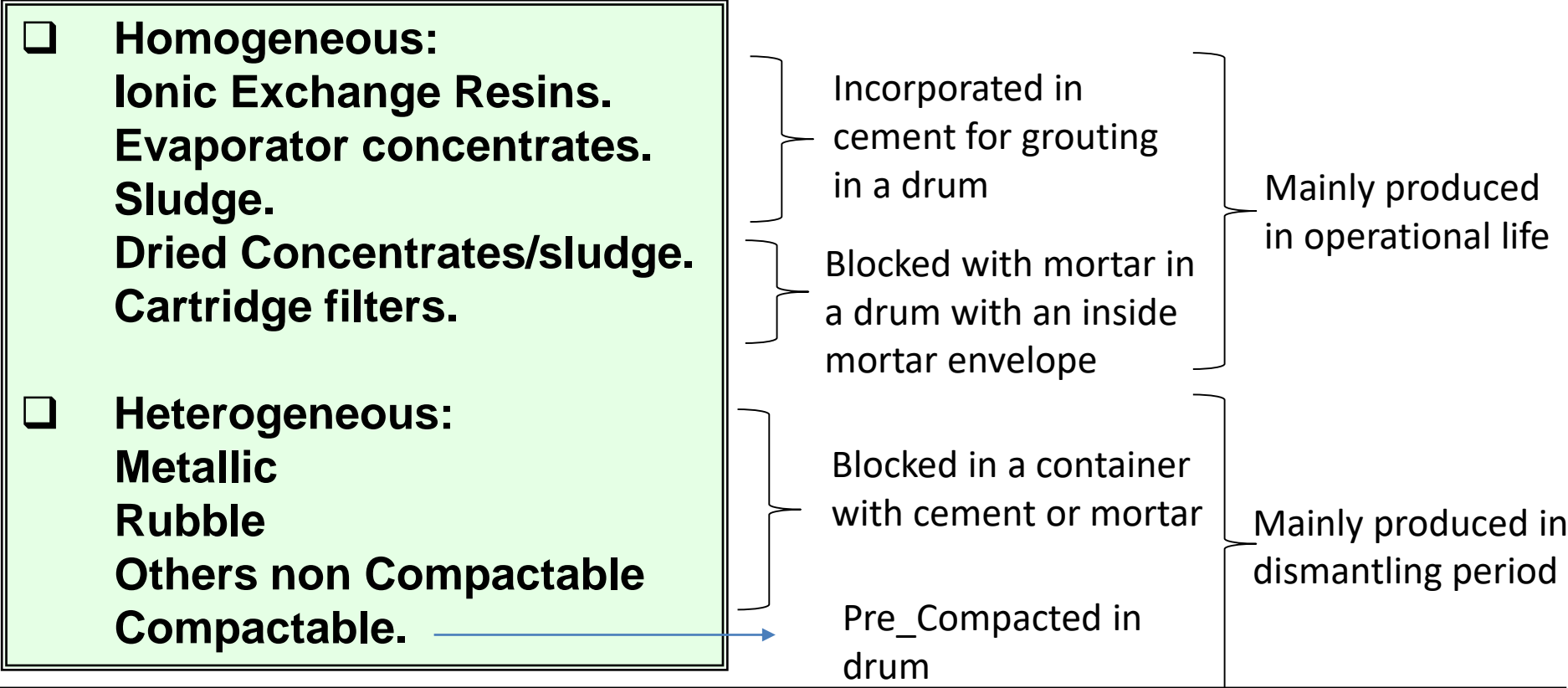
3.600 t

NPP Decommissioning: General Scheme for Materials



NPP Decommissioning:

Waste Streams Nature and Final Condition



Homogeneous and Heterogeneous waste are produced in both operational and dismantling stage of the NPP, but heterogeneous waste is the main stream produced during the dismantling period.

NPP Decommissioning:

Waste Acceptance Criteria



❑ Waste Package Level 1:

- Solidified Homogeneous waste (resins, sludge, evaporator concentrates):
 - Mechanical limits (compression, before and after immersion).
- Blocked waste (cartridge filters, dried sludge, ashes):
 - Thickness of the mortar/concrete sleeve
 - Mechanical limits (compression) of the sleeve.
- Heterogeneous waste:
 - Compactable waste: segregation process.
 - Non compactable waste: gap filling.

NPP Decommissioning:

Waste Acceptance Criteria



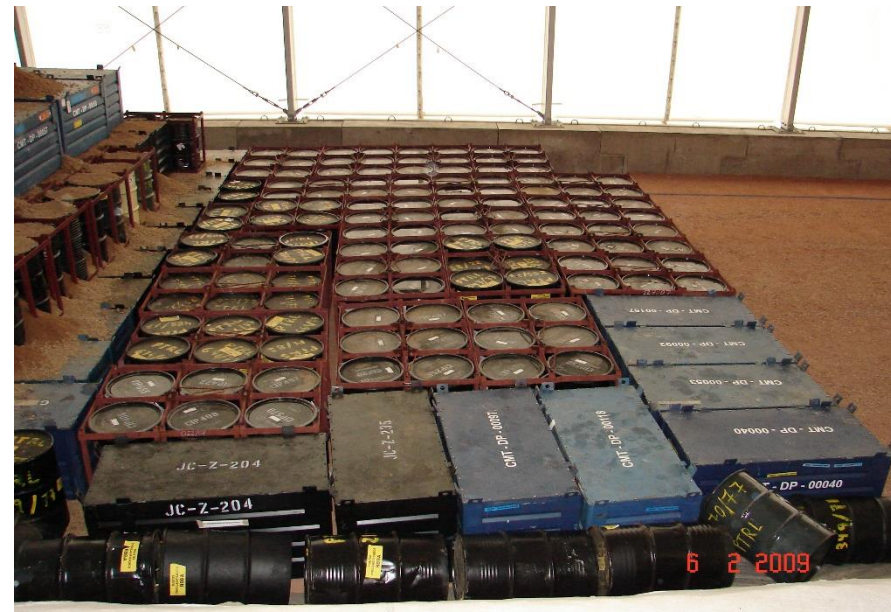
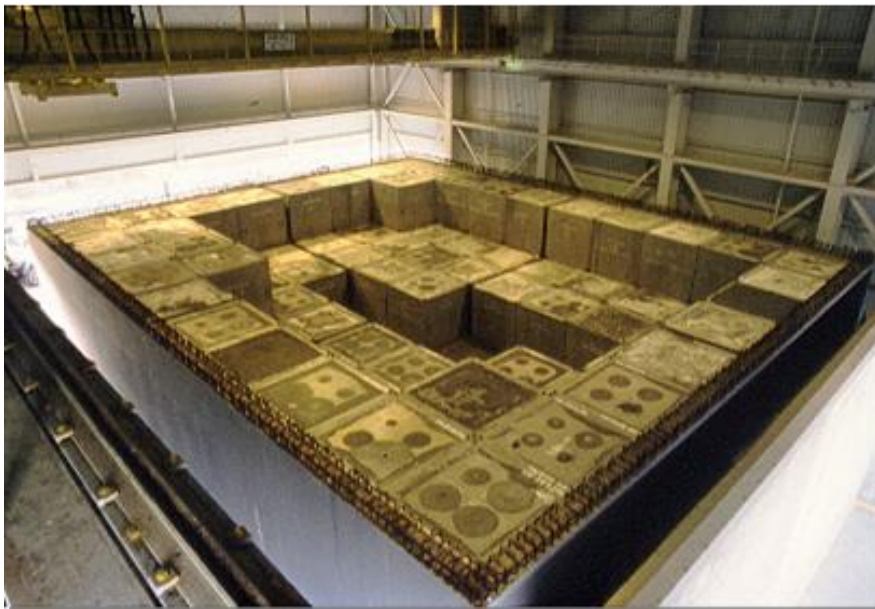
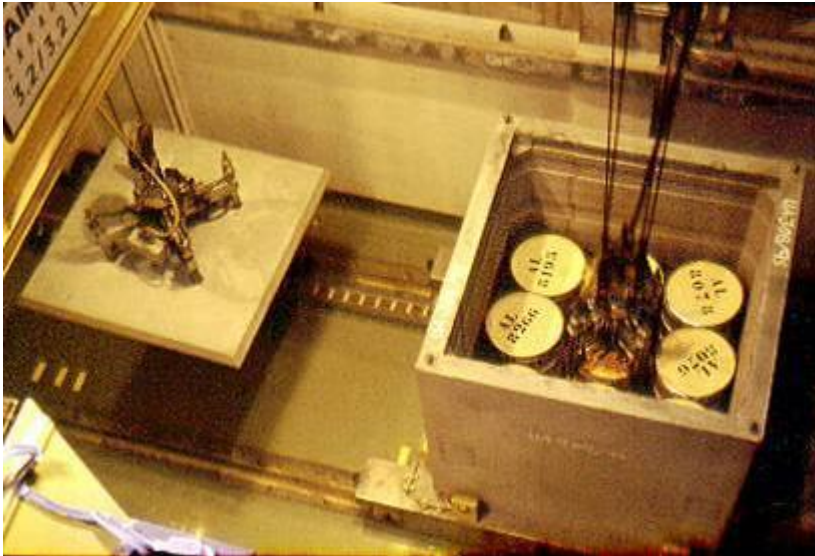
❑ Waste Package Level 2:

- Solidified Homogeneous waste (resins, sludge, evaporator concentrates):
 - Strongest mechanical limits (compression, traction. Before and after immersion, thermal cycles).
 - Leaching limits.
- Blocked waste (cartridge filters, dried sludge, ashes):
 - Thickness of the mortar/concrete sleeve
 - Strongest mechanical limits (compression), and thermal cycles of the sleeve.
 - Diffusion limits
- Heterogeneous waste:
 - Compactable waste: Try to avoid its production.
 - Non compactable waste: Try to avoid its production



NPP Decommissioning:

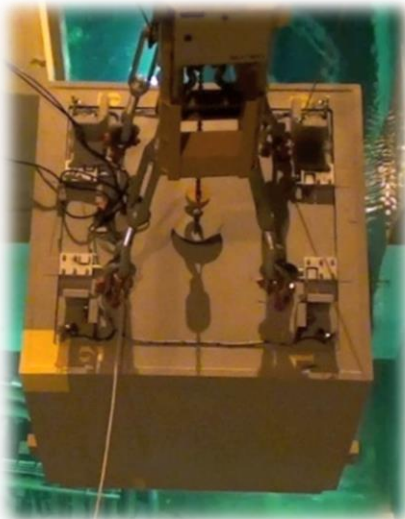
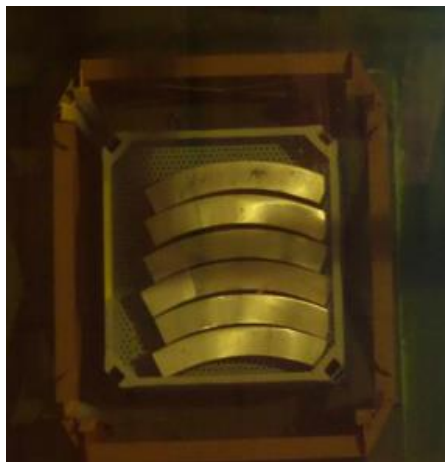
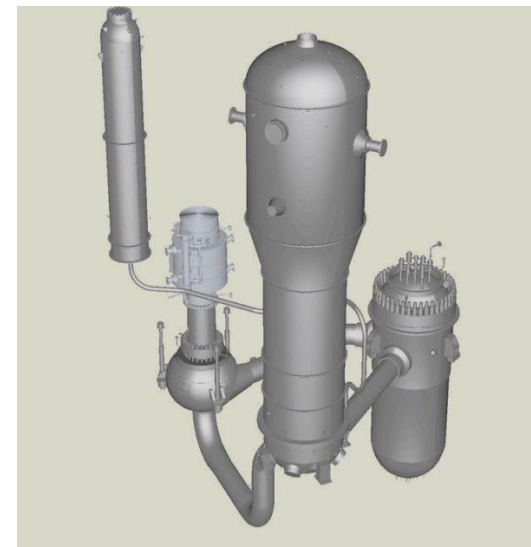
Waste Streams Nature and Final Condition





NPP Decommissioning:

Large components Cutting: Internals, Primary Circuit



NPP Decommissioning:

Items directly introduced in Disposal Units of El Cabril



NPP Decommissioning:

Items directly introduced in Disposal Units of El Cabril



NPP Decommissioning:

Packages Characterization: ISOCS



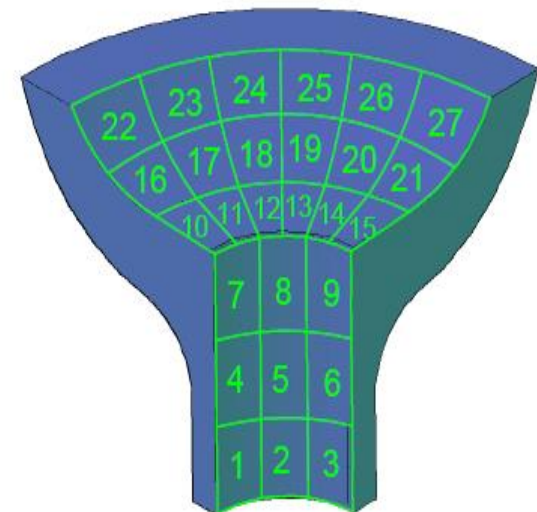
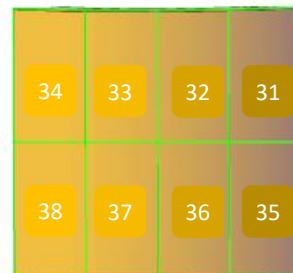
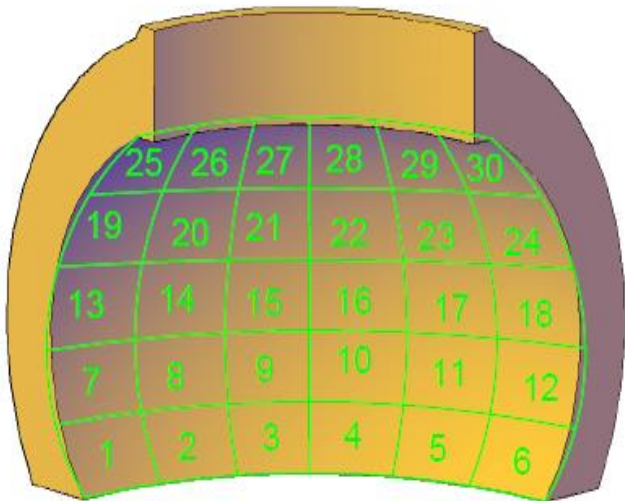
- ❑ Numerical calibration curve based on Monte Carlo scheme.
- ❑ Sensitivity analysis of different parameters that take influence on the measurement, like density heterogeneity, as long as heterogeneous activity distribution.

NPP Decommissioning:

Characterization of Large Items



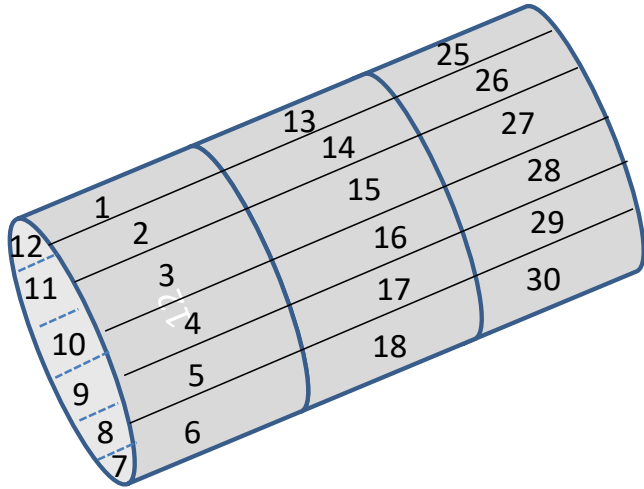
- ❑ Digitalize de Large Piece in Items of easy geometry.
- ❑ Determine the influence of each item on the rest of segments, in order to determine the intrinsic contribution of the Item itself discarding the influence of the rest of the Items.
- ❑ Simplify the model leaving null the contribution of the far Items which influence can be considered negligible in relation to the closest ones. This should be verified by dose rate measurements.
- ❑ Determine the activity of each Item, and the total activity by summing all.





NPP Decommissioning:

Characterization of Large Items



1	13	25
2	14	26
3	15	27
4	16	28
5	17	29
6	18	30
7	19	31
8	20	32
9	21	33
10	22	34
11	23	35
12	24	36

0	0	0
0	0	0
0	0	0
0	0	0
0	0	0
R5(18)	R17(18)	R29(18)
R6(18)	R18(18)	R30(18)
R7(18)	R19(19)	R31(18)
0	0	0
0	0	0
0	0	0
0	0	0
0	0	0
0	0	0





NPP Decommissioning:

Three main projects during the dismantling period



Three main activities, almost sequential, are involved during the dismantling period directly linked to both volume optimization of the VLLW and in situ characterisation:

Material Release: during the radiological disassembles activities, a large volume of material is generated from controlled zones that are candidate to pass a release process. The pieces are classified and sorted in containers to be finally measured for the verification of the fulfillment of the clearance limits.

Surface Release: after the removal of the materials from controlled zone, the next step is the process of systematic surfaces decontamination with the aim to release the building involved, and to be able to start the demolition of them.

Site Release; this is the final main process to be faced in order to leave the site as the licensed plan (green field, brown/gray field...).



NPP Decommissioning:

Material Clearance



Methodology and Levels have to be authorized (SPAIN)
Licensing document: Clearance Material Control Plan
Licensing tests: approved by Regulatory Authority
Clearance Levels (European Commission Recommendations)
GENERAL CLEARANCE LEVELS (N1): BSS 2013/59 (any solid material, does not require further regulatory control to ensure the destination)
SPECIFIC CLEARANCE LEVELS (N2): for a particular use or destination
RP-89: Recycling of metals
RP-113: Building and building rubble

BOX COUNTER (gamma spectroscopy system)

Activity of each radionuclide C_j in the material (Bq/g & Bq/cm²)

Scaling factors



NPP Decommissioning: Material Clearance: BOX COUNTER Device



NPP Decommissioning:

Material Clearance: Rule of decision



BOX COUNTER (gamma spectroscopy system)

Activity of each radionuclide C_j in the material (Bq/g & Bq/cm²)

To confirm a mixture of radionuclides is below the clearance level

$$SUF = \sum C_j / L_j \leq 1$$

C_j = specific activity in the material of radionuclide j (Bq/g)

L_j = the clearance level of radionuclide j (Bq/g)

Scaling factors

- Clearance Level
- N1: general levels (Bq/g)
 - N2: specific levels (Bq/g & Bq/cm²)
 - Metals
 - Rubble



NPP Decommissioning:

Material Clearance: QC and Reports



RELEASE REPORT

- CONTAINER data sheet
- Verification sheet of Material and measurement requirements
- Measurement and Final Calculations Reports
- Certificate of compliance

•Quality Control of the clearance process:

- Measurement Verification of the 5% of containers cleared /working day)

•Quality Additional control by laboratory analyses:

- Verification of clearance level (1% of containers cleared)

•Box-Counter CALIBRATION and VERIFICATION:

- Energy calibration: every 6 months
- Verification: every day

NPP Decommissioning:

Surfaces Clearance



CLEARANCE LEVELS: RP 113 (Demolition)

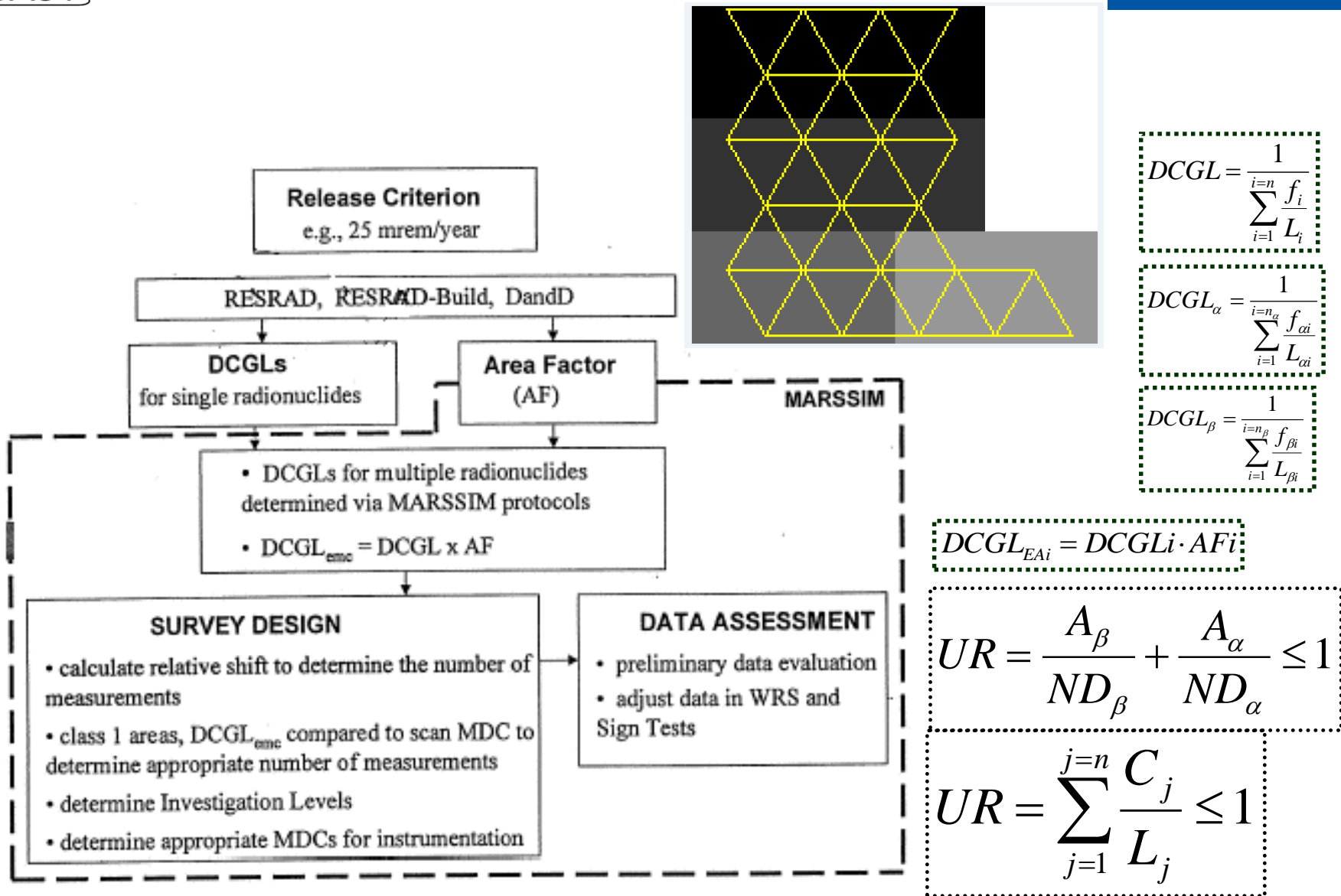
Dose: 10 μ Sv / year

METHODOLOGY: MARSSIM

- To confirm that buildings and structures are not contaminated, so that:
 - they can be demolished under conventional procedures
 - the materials arising demolition can be dispatched without any restriction.
- Cut the concrete of Reactor Cavity, Spent fuel Pool and Biologic Shield in approximately 2mx2mx2m concrete slabs for sending to el Cabril as L&ILW and VLLW.
- Systematic removal of superficial contamination from surfaces of radiological buildings (Auxiliary and Reactor).
- Removal of embedded radiological pipes from surfaces for allowing the final measurements with the required DQO.

NPP Decommissioning:

Surfaces Clearance: MARSSIM Approach



MARSSIM APPROACH

- Derived Concentration Guideline Levels, DCGLs, determined outside MARSSIM methodology.

$$DCGL_{gross} = \frac{1}{\frac{f_1}{DCGL_1} + \frac{f_2}{DCGL_2} + \dots + \frac{f_n}{DCGL_n}}$$

- MARSSIM does not describe the DCGL methodology but provides information of how to determine them (RESRAD, RESRAD BUILD, etc.).
- For a Survey Unit (SU) MARSSIM needs the DCGL, the best estimation of its activity and data dispersion, in order to obtain a representative value of the mean activity of the SU.



NPP Decommissioning: Surfaces

Clearance: MARSSIM Approach



MARSSIM START POINT

- MARSSIM describes a statistical methodology to obtain a representative sampling of SU, with a 95% confidence, or 5% of both type I and II errors.
- Mean value (Bq/cm^2) taken from the N data measured, sampling points, from the SU.
- The ratio of the mean value to the DCGL is a fraction of the dose criteria considered.
- Only for type 1 class, an additional aspect to the dose criteria has to be taken into account, due to the extra contribution above mean value of the elevated concentration areas.
- DCGL for Elevated Measurement Comparison: DCGL_{EMC} .

NPP Decommissioning:

Surfaces Clearance: MARSSIM Approach



- ❑ Relative Shift, basic parameter to establish the final N data to final survey.

$$\text{Relative Shift} = \frac{DCGL - SU\text{Activity}}{S\text{Deviation}}$$

- ❑ MARSSIM advises to assign the most actual values for both the SU activity and its Standard Deviation.
- ❑ The N data to be taken, that is to say the sampling process, is fully considered as representative one.
- ❑ Correctness of sampling is fulfilled due to the location of the N data grid (for class 1 and 2) or to their random location (class 3).



NPP Decommissioning:

Surfaces Clearance: MARSSIM Approach



Survey Unit Classification		Statistical Test	Elevated Measurement Comparison	Sampling and/or Direct Measurement	Scanning
Impacted	Class 1	Yes	Yes	Systematic	100% Coverage
	Class 2	Yes	Yes	Systematic	10-100% * Coverage (systematic and judgmental)
	Class 3	Yes	Yes	Random	Judgmental
Non-Impacted		No	No	No	None



NPP Decommissioning:

Surfaces Clearance: MARSSIM Approach



- ❑ MARSSIM scenario A, null hypothesis (H_0), SU is contaminated above DCGL.
- ❑ MARSSIM advices: bad estimation of SU activity increases the probability of type II errors (reject the SU), but the type I error is not influenced and kept anyway (Regulator).
- ❑ Intentional dilution is probably detected by MARSSIM methodology due to the sampling process applied.

NPP Decommissioning:

Surfaces Clearance: MARSSIM Approach



- ❑ Required in MARSSIM methodology.
- ❑ Once the Grid with N measurements has been defined, an Area Factor has to be evaluated using the area of the pattern (triangular or cubic).
- ❑ If the scan MDA is greater than $DCGL_{EMC}$, a new Area Factor should be taken due to the lack of detecting elevated activities below the $DCGL_{EMC}$.

$$\text{New Area Factor } (AF_{NEW}) = \frac{\text{Scan MDC}}{DCGL}$$

- ❑ After performing the measurements, if there are some areas with elevated concentration, the actual area extension has to be determined for each one and the actual Area Factor for each one is also calculated.
- ❑ Finally, the contribution above the mean value (calculated using N measurements) of each elevated activity area has to be accounted.

$$\frac{\delta}{DCGL} + \sum \frac{(\text{average concentration for elevated area} - \delta)}{(\text{area factor for elevated area})(DCGL)} < 1$$

Where δ is the mean (average) concentration in the survey unit as determined from the measurements



NPP Decommissioning:

Jose Cabrera Surfaces Works

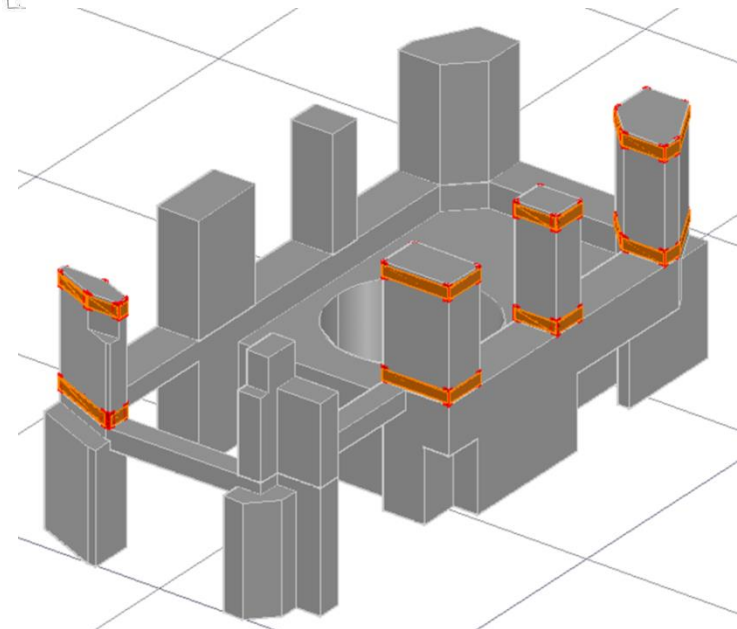
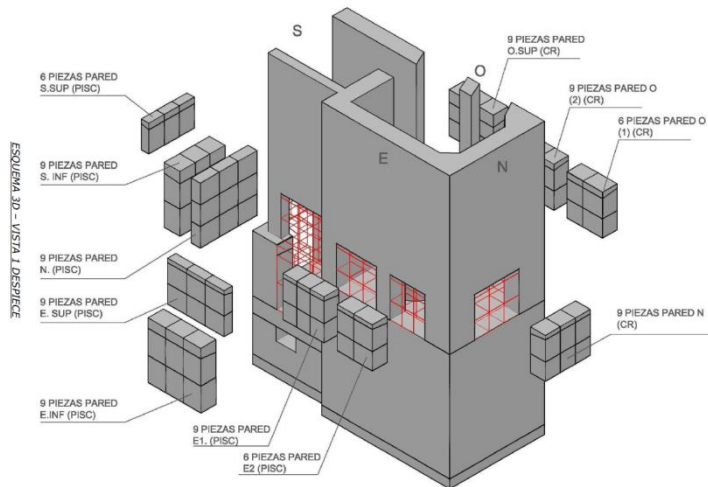
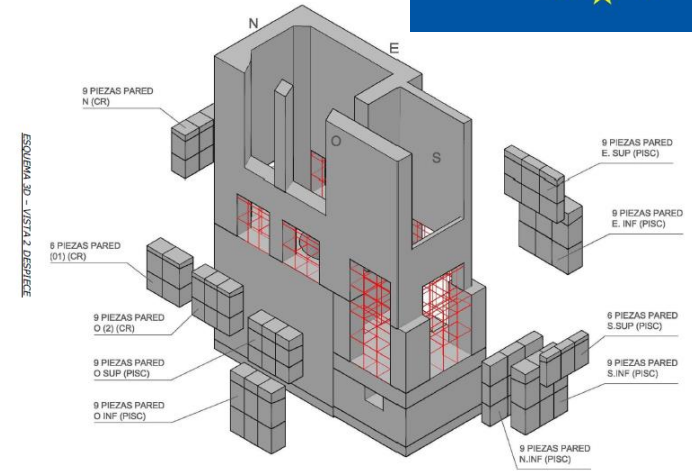
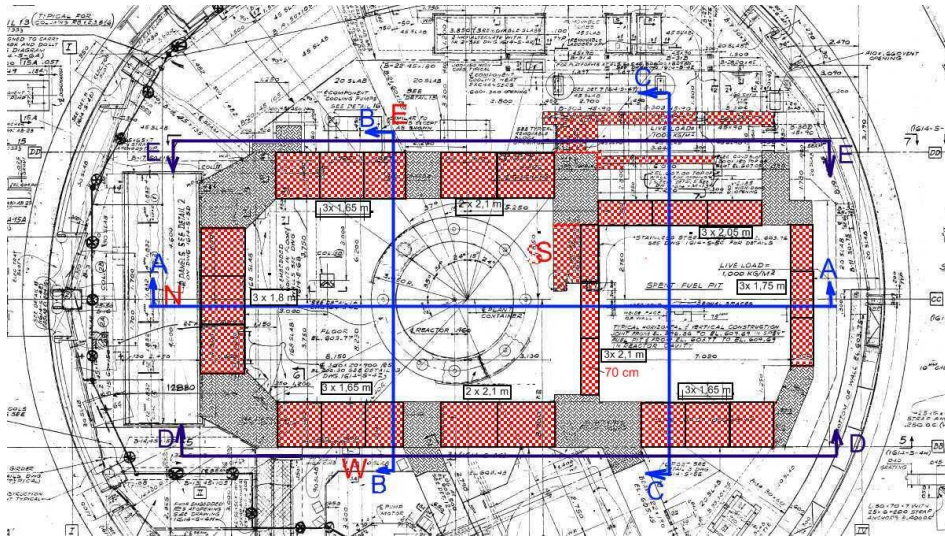


- ❑ Cut the concrete of Reactor Cavity, Spent fuel Pool and Biologic Shield in approximately 2mx2mx2m concrete slabs for sending to el Cabril as L&ILW and mainly as VLLW.
- ❑ Simultaneously, starting the process of systematic removal of superficial contamination from surfaces of radiological buildings (Auxiliary and Reactor).
- ❑ Removal of embedded radiological pipes from surfaces for allowing the final measurements with the required DQO.
- ❑ Characterization of Survey Units for the Final Status Survey, FSS



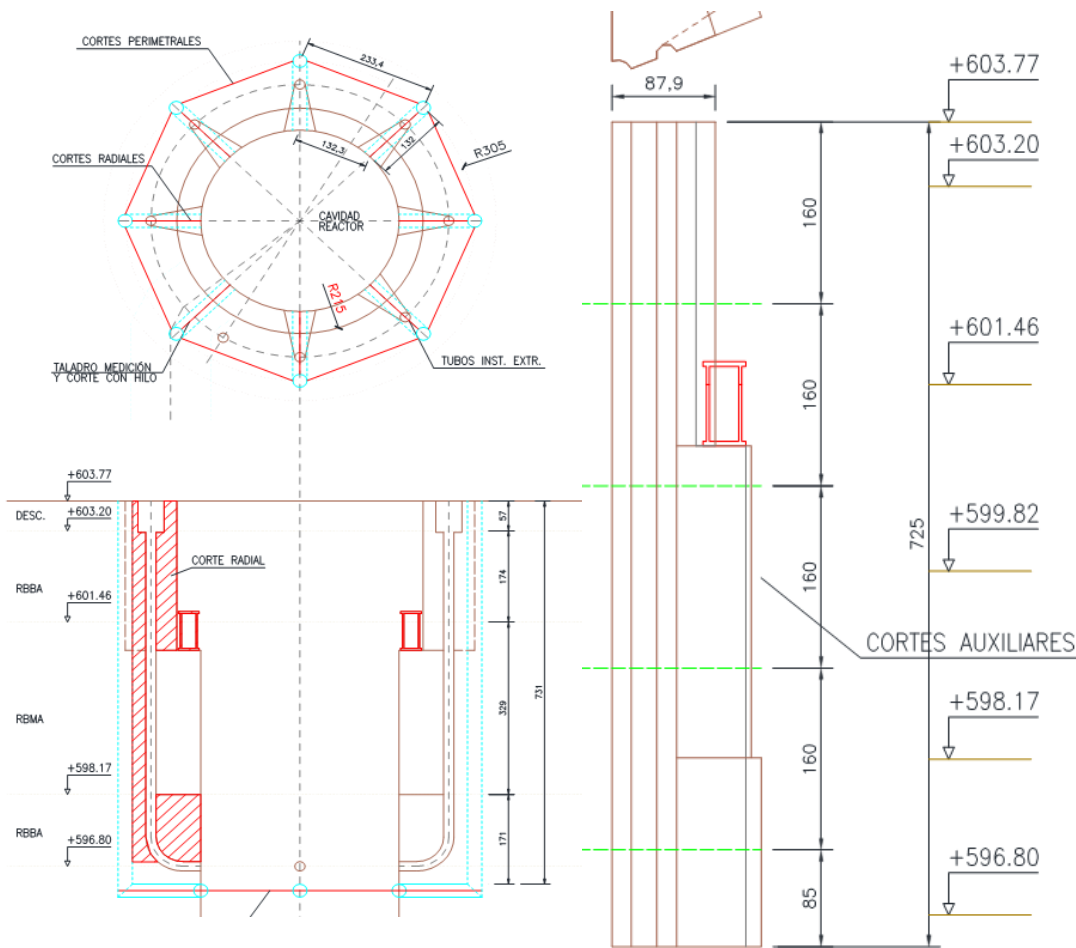
NPP Decommissioning:

Jose Cabrera Surfaces Works



NPP Decommissioning:

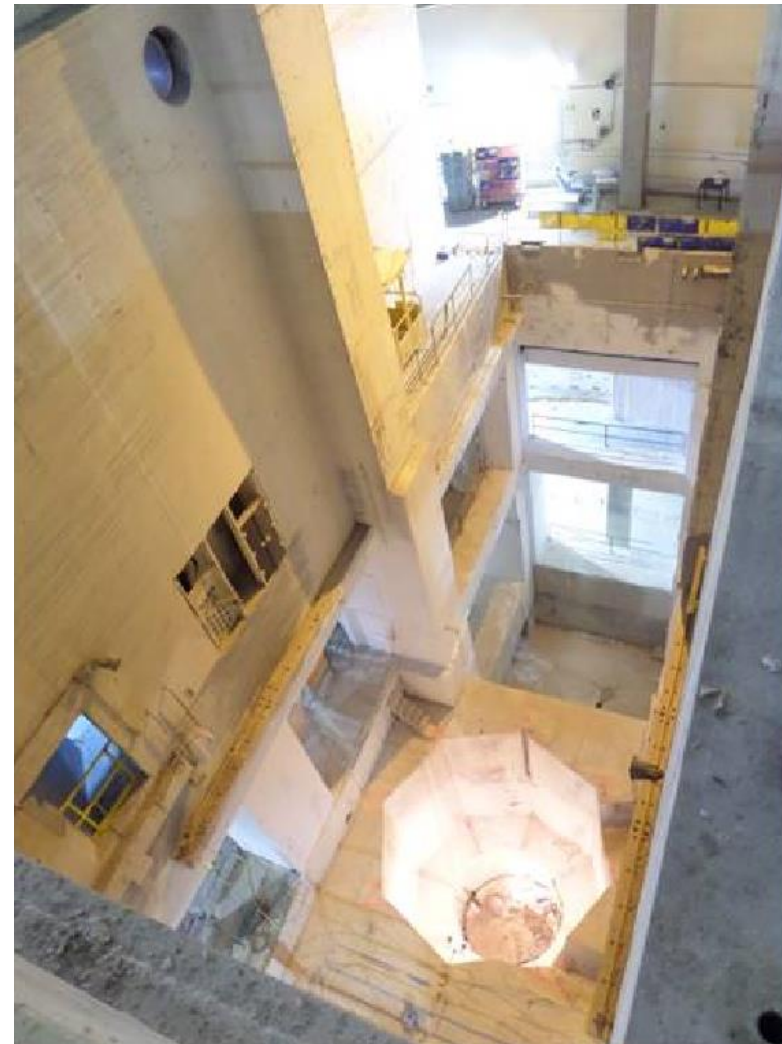
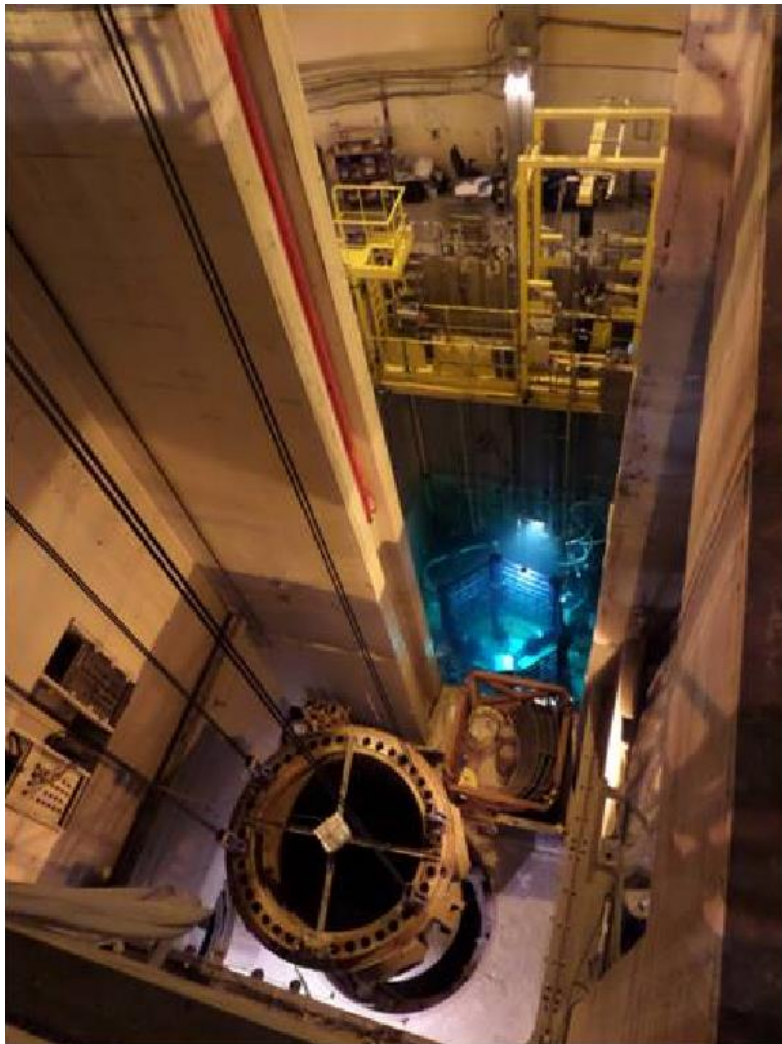
Jose Cabrera Biological Shield Cutting





NPP Decommissioning:

Jose Cabrera Cavities





NPP Decommissioning:

Surfaces Clearance: General Scheme



BASIC PROCEDURE

1. Classify the SU (type 1, 2 or 3), historical site assessment.
2. Characterization of the SU.
3. Determination of the Mean Value and Standard Deviation of characterization data.
4. Determine the DCGL.
5. Relative Shift calculation.
6. MARSSIM scenario A to be applied, and the test of data to be used (Sign test or Wilcoxon Rank Sum test).
7. Establishes the N final survey data to be measured and the location of them inside the SU.
8. Determine the Area Factor for the grid obtained (Class 1 or 2).
9. Define the means for the measurement (non spectrometric or spectrometric devices).
10. Perform the survey.
11. Data analysis, Sign test or Wilcoxon test. Elevated activity areas analysis, If any.
12. Final decision, release or reject the SU.
13. SU Release Report.

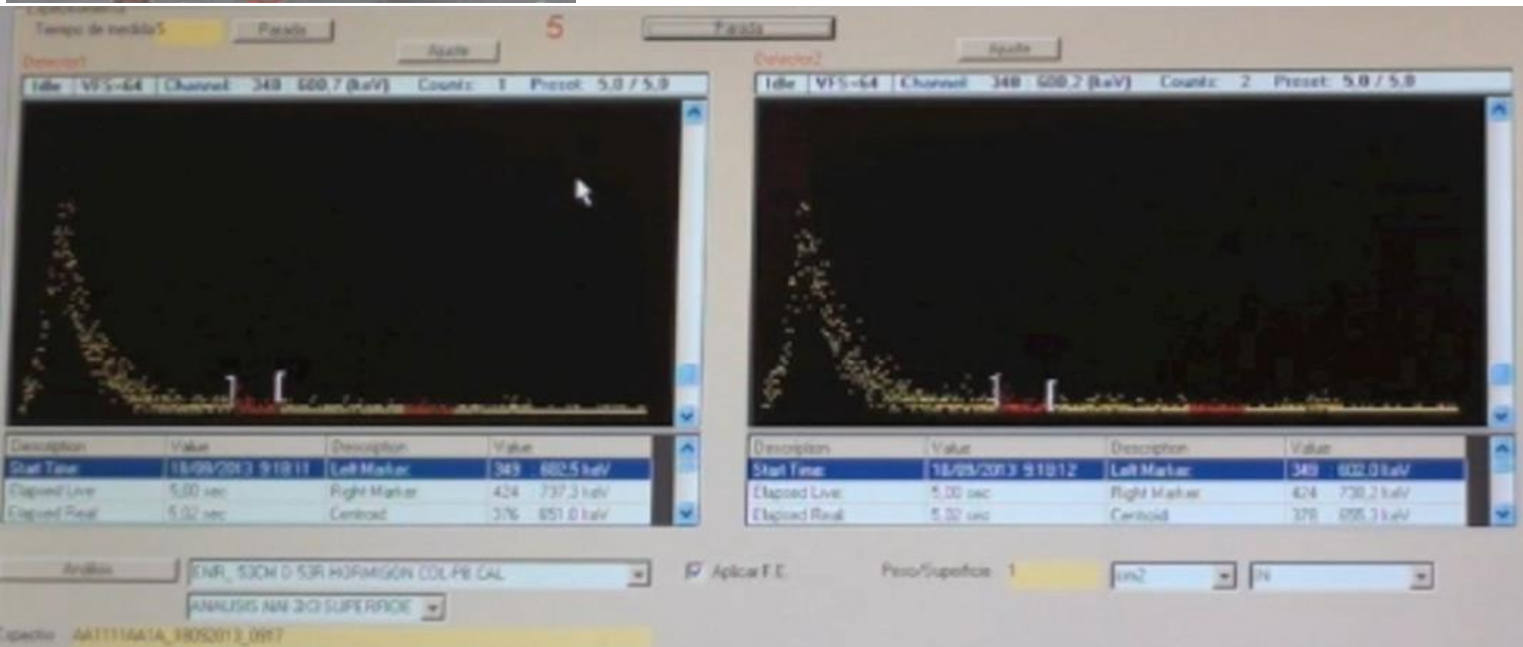
NPP Decommissioning:

Surfaces Clearance: Instrumentation



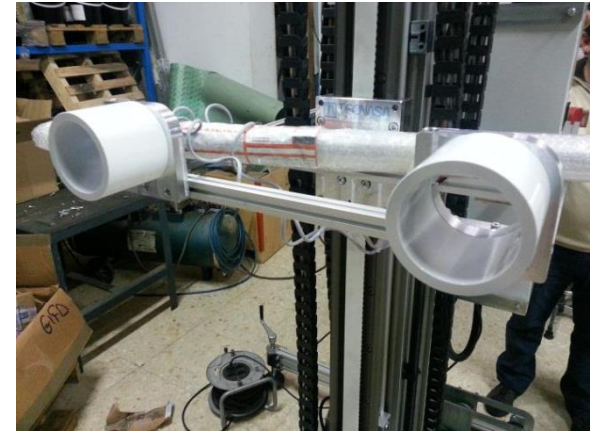
Id	X1	X2	Y2	Y4	Y5	Y6
81	680.3	5.187	10.3	4.4	2.7	4.2
13	680.0	10.7	3.4	5.1	3.0	5.6
23	679.2	14.2	12.9	6.3	3.4	2.2
23	678.3	15.0	13.1	5.7	4.4	7.8
23	683.5	14.3	10.8	5.2	3.9	3.0
82	684.3	15.1	11.3	6.3	4.0	4.2
23	683.2	14.3	8.5	3.8	6.0	3.0
21	1096.7	277.6	295.5	118.7	96.0	6.0
23	2540.3	648.2	702.5	277.5	222.2	7.5
23	2886.7	696.7	780.7	302.8	263.9	19.4

Id	X1	X2	Y2	Y4	Y5	Y6
38	686.4	16.1	12.0	5.7	3.1	5.2
21	689.7	10.0	8.6	5.7	3.0	8.1
21	691.0	16.3	13.5	5.8	4.3	7.7
21	694.0	12.2	11.7	5.4	3.9	4.4
21	690.4	16.1	12.9	2.9	4.3	6.7
83	672.4	14.3	12.5	4.5	4.1	7.1
21	695.5	13.9	12.0	3.8	1.4	3.4
15	686.4	14.7	11.0	4.2	2.6	5.9
21	684.9	14.3	9.6	7.2	2.9	4.3
23	634.0	11.1	9.2	5.8	1.9	6.3



NPP Decommissioning:

Surfaces Clearance: Instrumentation





NPP Decommissioning:

Surfaces Clearance: Instrumentation, DRONES



NPP Decommissioning: Site Release Process



PROYECTO	INGENIERÍA PRINCIPAL DE P.D.C. DE C.N.J.C.	
INSTAURADA POR	CENTRAL NUCLEAR JOSÉ CABRERA	
TÍTULO DE INGENIERÍA	FIGURA 1 DISEÑO DE LA DISTRIBUCIÓN DE LAS UNIDADES DE LIBERACIÓN	
EMPRESA	enresa	
	CÓDIGO DE DOCUMENTO	060-PC-JC-0430
FECHA	05/11/2011	05/11/2011



NPP Decommissioning:



Site Release: Release Levels

Radionucleido	NL (Bq/g)	Escenario más restrictivo
Am-241	8,83E+00	Obras/Mantenimiento
C-14	1,26E+02	Agrícola-residencial
Cm-244	1,91E+01	Obras/Mantenimiento
Co-60	1,39E-01	Obras/Mantenimiento
Cs-134	2,51E-01	Obras/Mantenimiento
Cs-137	5,97E-01	Obras/Mantenimiento
Fe-55	4,13E+04	Agrícola-residencial
H-3	3,54E+03	Agrícola-residencial
Nb-94	2,10E-01	Obras/Mantenimiento
Ni-59	5,86E+03	Agrícola-residencial
Ni-63	2,47E+03	Agrícola-residencial
Pu-238	9,75E+00	Obras/Mantenimiento
Pu-239	8,90E+00	Obras/Mantenimiento
Pu-241	3,68E+02	Obras/Mantenimiento
Sr-90	1,92E+00	Agrícola-residencial
Tc-99	6,19E+00	Agrícola-residencial

REFERENCE RESIDUAL LEVELS

- Useful for defining the Lower than Detection Limit Values.
- For deciding whether or not there is contamination.
- Their influence on the Radiological Criteria is lower than 10%.



NPP Decommissioning:

Site Release: Basic Approach



- Remediation is going to be systematically performed when residual activity is above RL's, with the main objective, insofar as possible, of leaving soil with activity below the RRL's.
- Before the backfilling process, MARSSIM methodology is to be applied to the exposed soil in order to decide whether or not the RU is release.
- When no remediation is required, MARSSIM methodology is directly applied to the RU to assess the release process.
- In addition to the MARSSIM approach, when applied, a number of pits has to be collected in the location of the N measurements to provide that no subsurface residual values are involved.



NPP Decommissioning:

Site Release: Basic Approach



- Dynamics analysis of the Release Unit covering 100% of its surface.
- Systematic pits in those RU's in which are not expected to have, by operational information and the initial characterization, residual values in depths greater than the ones achieved by this technics.
- Additional boreholes in those RU's in which are expected to have, by operational information and the initial characterization, residual values in depths greater than the ones achieved by pits. Or in those RU's which harbor buried structures and pipes with radiological functions.



NPP Decommissioning:

Site Release: Basic Approach



- Dynamics analysis of class 2 RU covering up to 50% of its surface.
- Dynamics analysis of class 3 RU covering up to the 10% of its surface.
- Pits in those class 2 RU's in which are expected to have some fraction of the release levels as residual activity.
- Pits in those class 3 RU's in which are expected to have some small fraction of the release levels as residual activity.



NPP Decommissioning:

Site Release: Basic Approach



Class 1 RU's :

- A first approach with a grid of 15 m of length side to identify/quantify the places to be remediated.
- Increase the density of measurement decreasing the size of the grid in the location to be remediated, just to best define the boundary that change from the clean area to the contaminated one.
- **The size of the grid should be greater than the size of the means used to remediate.**

Class 2 RU's :

- A first approach with a grid of 20 m of length side to identify/quantify the places to be remediated.

Class 3 RU's :

- A first approach with a grid of 30 m of length side to identify/quantify the places to be remediated.

All these information will be used to better define the N measurements to be taken in the **Final Status Survey**.



NPP Decommissioning:

Site Release: Basic Approach



GEOSTATISTIC

- ❑ **Its main goal is to quantify as best as possible the amount of terrain to be remediated.**
- ❑ **One additional objective is to estimate the residual activity of the terrain that is going to be left**, that have to be lower than the limits with a fixed confidence interval. But in any case, this terrain is going to be measure in a detail manner later on in the **Final Assessment** phase.
- ❑ **Geostatistics** is a valuable tool when **data are structured**.
Processes that follow physical laws of contaminants transport in which it is expectable to show correlation among them in different places.
- ❑ When **data are not structured**, there is **no difference between geostatistics and classical statistics** (e.g. MARSSIM).
In a trench that had packages, there is no expected correlation among the activity of different places inside the trench with distance.
Or after a systematic scarified of surfaces in buildings, the residual activity is not expected to show correlation.



NPP Decommissioning:

Site Release: Basic Approach



Structured Data

- From a detailed estimation by using geostatistics in order to infer the activity of each container. Additional non systematic in situ measurements as a verification tool.
- With no previous analysis. The classification is done during the remediation process by means of systematic in situ measurements.
- Both, the more current cases, a pre-classification by using geostatistics and in situ measurements to definitely classify the remediated material..

No structured data

- It is not possible to have a detailed previous estimation unless there is a large number of pits/boreholes, non operative process.
- Directly remediate/classify the materials.

NPP Decommissioning:

Site Release: Boreholes





NPP Decommissioning:

Site Release: Pit/Boreholes





NPP Decommissioning:

Site Release: Characterization



- We operate with two BOX COUNTER and two ISOCS.
- In situ measurements before remediation in addition to geostatistics tools would be useful for estimating the activity in blocks size comparable to the means and containers to be used.
- In situ measurements during the extraction process are required. Ratemeters, total Beta/Alfa devices, INa Gamma devices.
- VLLW containers or BIG BAG to be send to the washing process.
- Clearable containers to the BOX COUNTER measurements.
- In situ measurement to the left terrain in order to check the suitability of the remediated process, otherwise keep remediating.
- Washed soils will be measured by BOX/ISOCS devices.
- The final waste of the washing process, dried finer part of VLLW will be measure by ISOCS.
- In situ measurement in the washing process in order to check and track its efficiency.



NPP Decommissioning:

Site Release: Characterization



Remediated RU's

- Before backfilling.
- Dynamic scanning with 100% coverage.
- N detailed static measurements.
- N pits.

Non remediated RU's

- Dynamic scanning with a coverage in accordance with their class.
- N detailed static measurements.
- N pits.



NPP Decommissioning:

Site Release: Backfilling



- With clean material from outside.
 - Sand.
 - Rubble crushed.
- With released material.
 - Soils unconditionally released.
 - Rubble conditionally release and properly diluted with clean rubble as RP113 requires

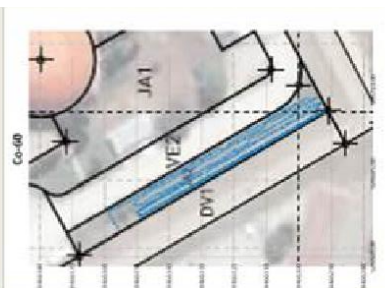
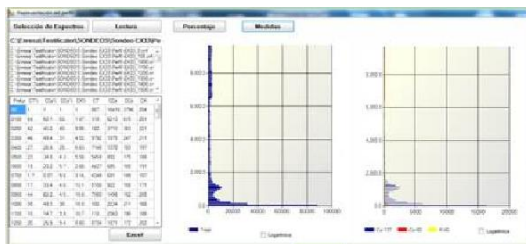
NPP Decommissioning:

Site Release: Instrumentation



NPP Decommissioning:

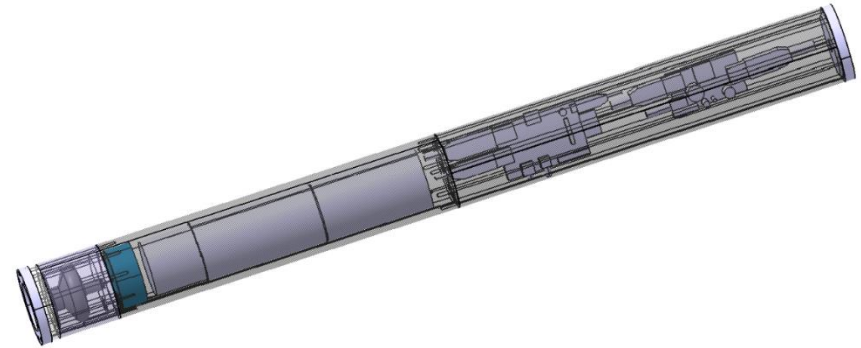
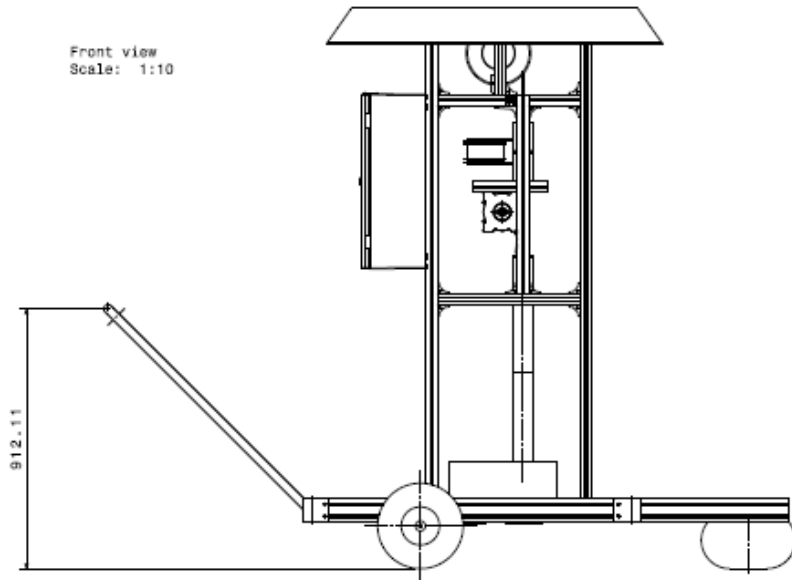
Site Release: Instrumentation





NPP Decommissioning:

Site Release: Instrumentation





NPP Decommissioning:

Site Release: Instrumentation



Conexión TMedida 120 s Pozo ppSUF Fecha 23/10/2015 12:15 Intervalo 50 mm 120 Salir

Detector conectado Descripción verificacion SUF

PosicionDeseadaZ = 100 mm Posicionar Final 3000 mm

Medida Automática CNID Parámetros Genie2K Nivel freático 11000 12000 Eficiencias Sobre nivel freático test130x50.CAL En nivel freático test130x50.CAL Bajo nivel freático test130x50.CAL

Estado Maquina ValidMovimiento Posicion.Z = 99 mm AlarmActive Speed.Z = 0 mm/seg gCargaBateria = 91

Parametros Maquina JogSpeed.Z = 100 mm/seg AccDec.Z = 100 mm/seg2 Inc.Z = 0 mm VZ = 50 mm/seg

Gráfico de actividad vs. Posición (mm)

Isotopo	Actividad	Incididumbre	LID	UD
K-40	5.11E-01	7.88E-02	2.23E-01	1.07E-01
CO-60	3.39E-02	6.59E-03	2.72E-02	1.31E-02
CS-134			3.67E-02	1.8E-02
CS-137	1.89E-01	1.47E-02	3.48E-02	1.70E-02
BI-214			8.23E-02	4.0E-02
PB-214	5.89E-01	3.11E-02	5.14E-02	2.51E-02

Borrar informacion

Error en la escritura. Iniciando software. Error en la lectura programada temporizada. Service Conectado. Error en la lectura programada temporizada. Error en la lectura programada temporizada.

Date Time Fecha motor 20/02/2007 3:13:36

E/S digitales gIN1_Pulsador_bajar gIN2_Pulsador_subir gIN3_Circuito_seguridad gIN4_FinalCarrera

Comandos Maquina Enable StartIncremento StartMove StopMove JogPos.Z JogNeg.Z Reset

NPP Decommissioning:

Site Release: Instrumentation



NPP Decommissioning: Site Release: Instrumentation



testi_30x50ajustado.geo

Edit dimensions - Well or Marinelli Beaker

Description: testi_30x50ajustado

Comment: arlite

Units: mm cm m in ft

No.	Description	d.1	d.2	d.3	Material	Density	Rel. Conc.
1	Liner - side	8	100	500	pvc	1.39	
2	Liner - end	20			lead	11.35	
3	Source - side	300			dirt1	1.6	1.00
4	Source - end	0			(none)	0	0.00
5	Source-Detector	200	0				

OK
Cancel
Apply
Help
View Drawing...

Geometry

- ISOCS Well or Ma
 - Liner - side
 - Liner - end
 - Source - side
 - Source - end
 - Detector

Template Drawing

WELL OR MARINELLI BEAKER

Fit to Window Close Help



NPP Decommissioning:

Materials/Surfaces/Site Release



METHODOLOGY APPROVAL FROM THE REGULATORY BODY

To Demonstrate that Enresa has properly developed the means and resources to implement the surface clearance, in relation to the following:

- Design the sampling for the SU & RU.
- Proper devices to use, spectrometric and non spectrometric.
- Perform the final survey of SU & RU.
- Final decision, to accept or to reject the SU & RU.
- Release Report of SU & RU.
- Controlling and tracking the SU & RU.



NPP Decommissioning:

Characterization and infer into a suitable Waste Form



THANK YOU VERY MUCH FOR YOUR ATTENTION !