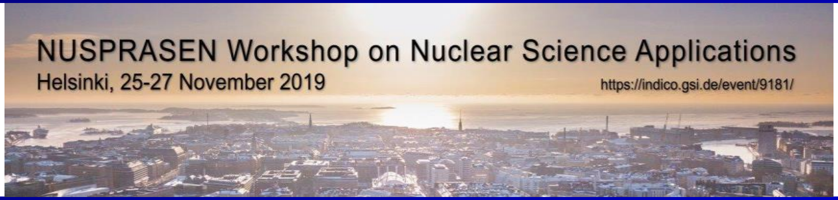

R&D, innovation and physicist knowledge needs for radioactive waste characterization.

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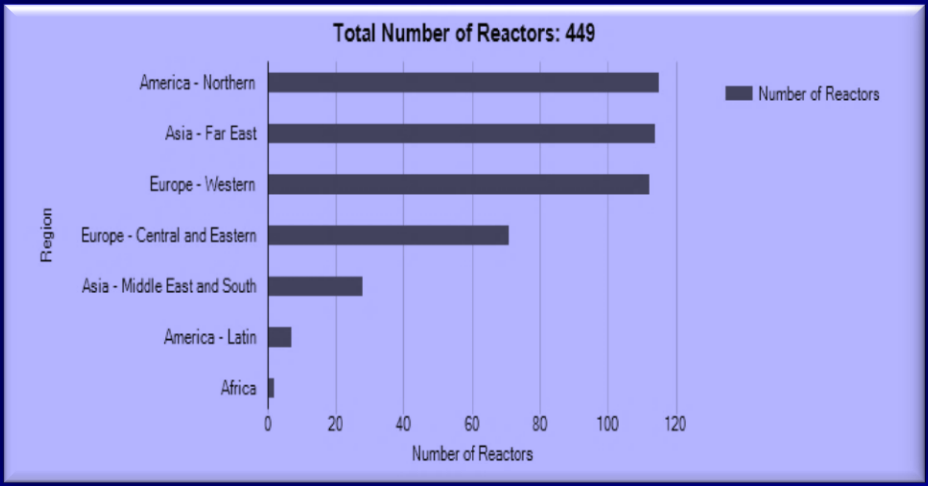
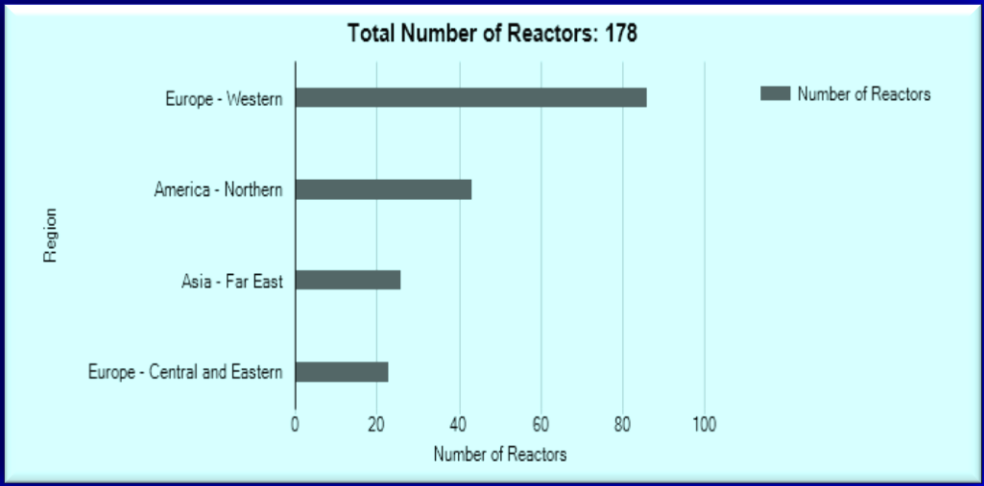
Nuclear Power Reactors in the World

OPERATIONAL REACTORS



PERMANENT SHUTDOWN REACTORS



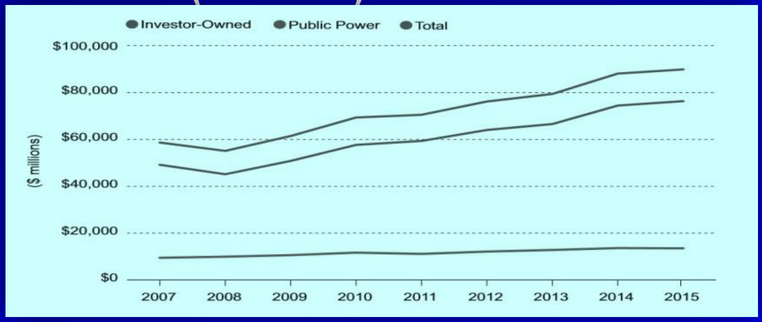
https://pris.iaea.org/PRIS/WorldStatistics/

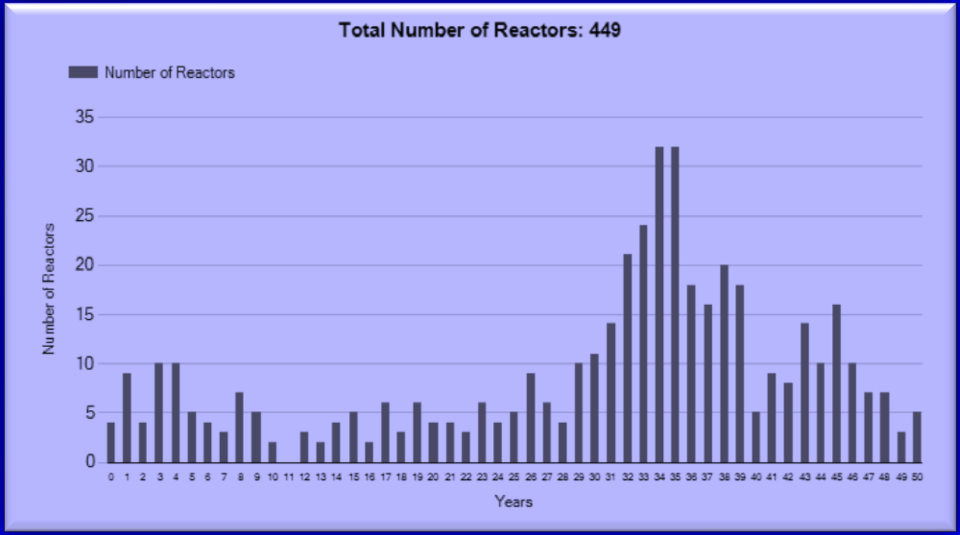
The largest number of nuclear power plants are either already closed or to be closed by 2025 is in Western Europe (>160).

Estimated cost:

France (EDF, AREVA, CEA) 73 billion EUR

USA (US DOE):





Waste management system (in Hungary)

SCOPE OF DUTIES

Collection

Minimization

Treatment

Conditioning

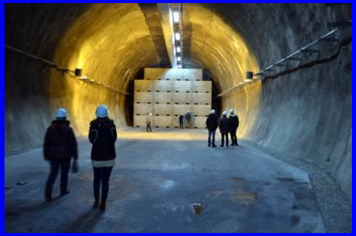
Characterization

Interim storage

Buffer storage

Overpacking

Disposal



Radioactive waste characterization

Package identification

- Five- or six-figure identification number
- Bar code

Packaging

- 200 litre drum
- 400 litre drum
- Four-cornered steel container
- Special container

Radiological parameters

- Dose rate
- Surface contamination
- Activity

Waste form properties

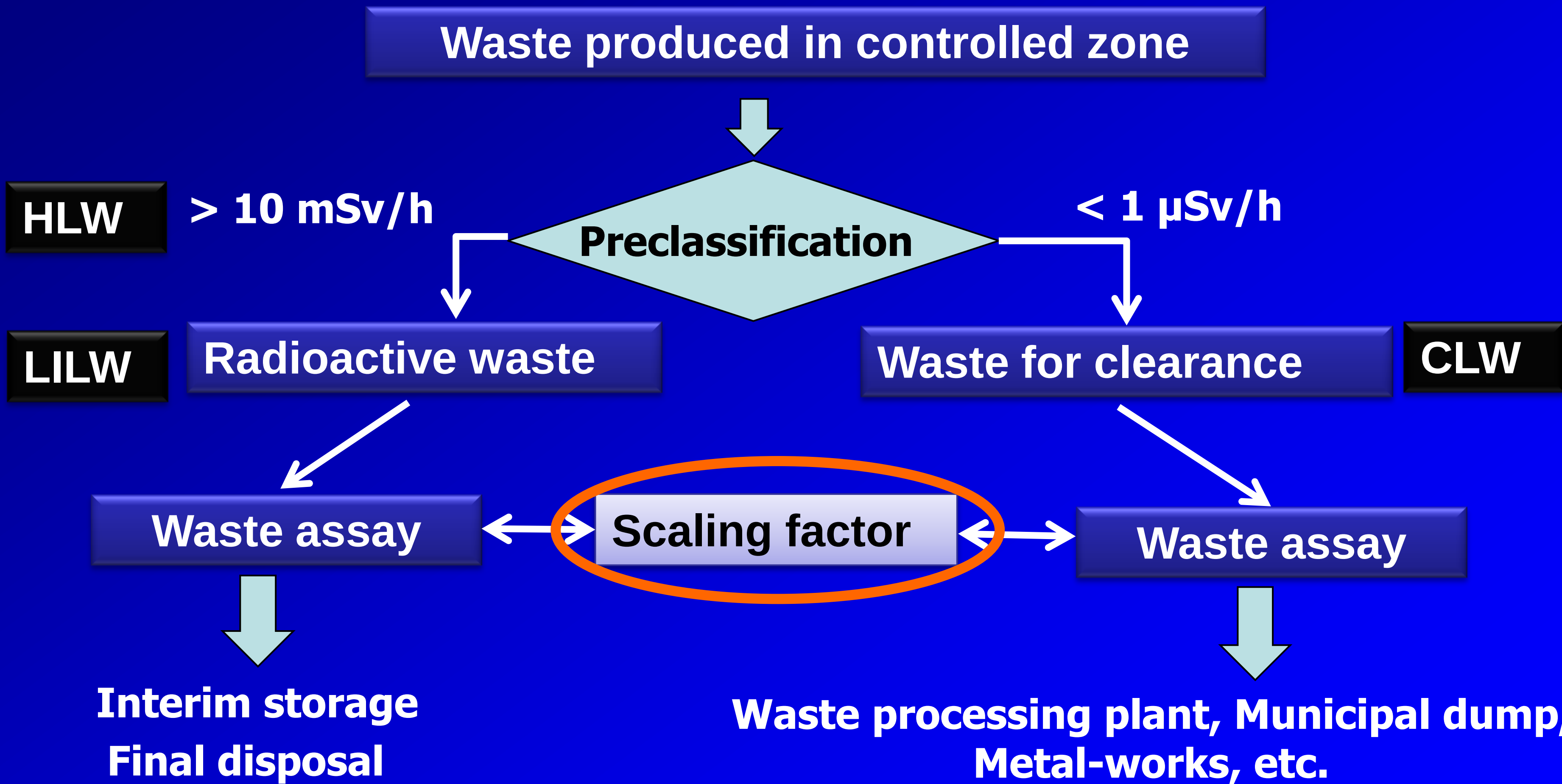
- Physical form
- Structural stability
- Impacts of heat and radiation
- Combustibility
- Forbidden components
- Gas generating
- Microbiological impact
- Free liquid
- Chelating and complexing agents
- Leaching
- Homogeneity

Process of Treatment

- Compacted
- Non-Compacted
- Solidified
- Special treatment

Classification of wastes (solid)

Classification of solid wastes:

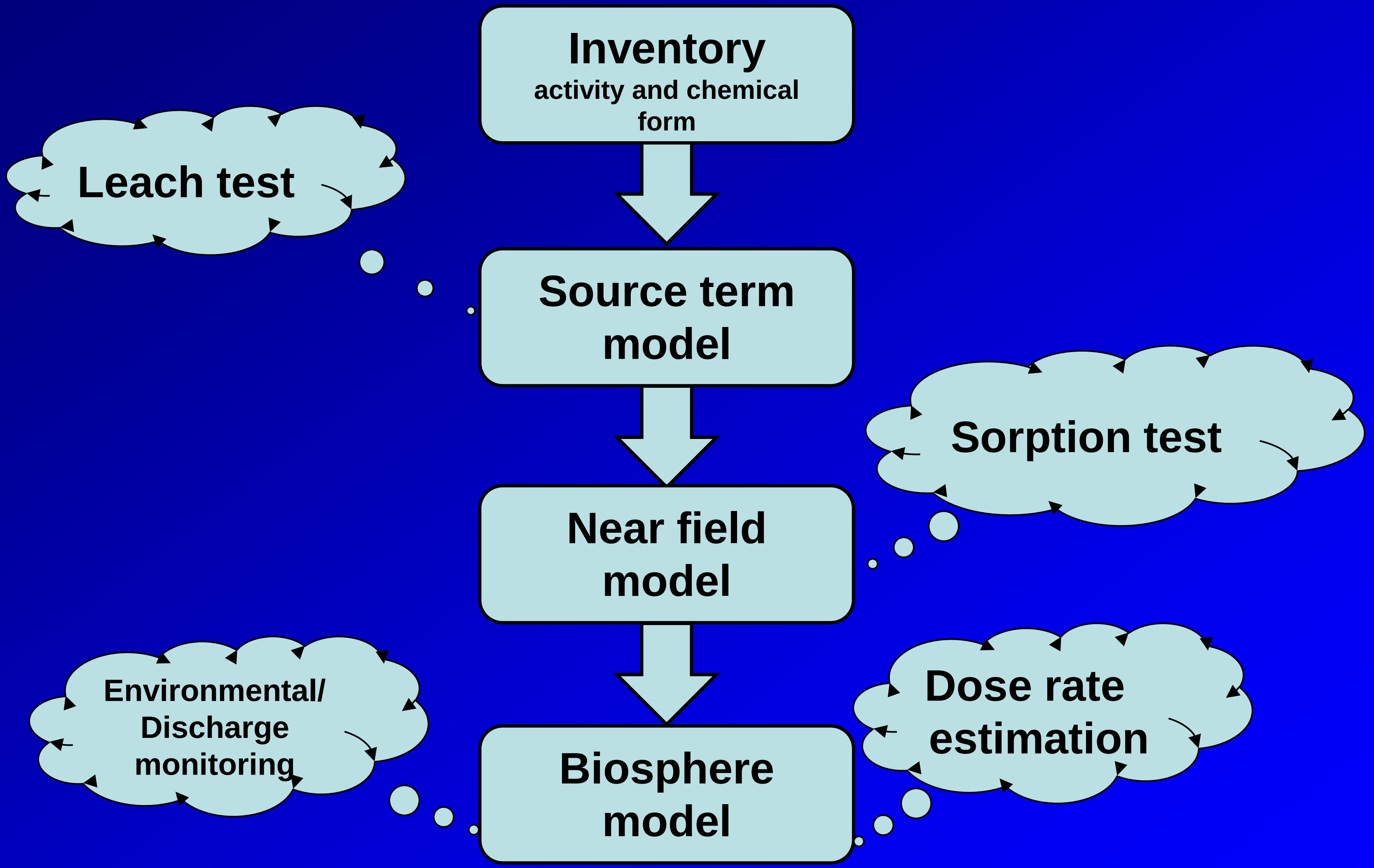


Scaling Factors

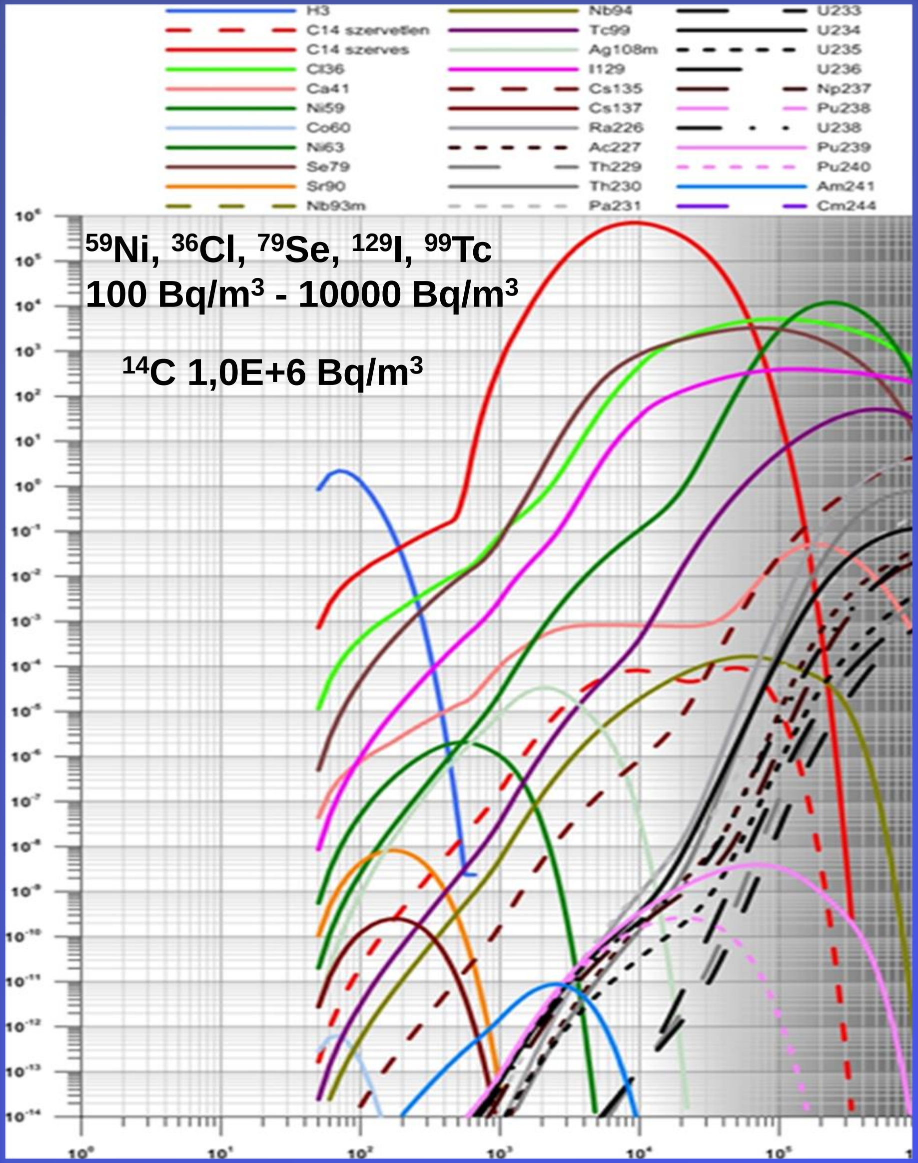
DETERMINATION OF NON-MEASURABLE NUCLIDES

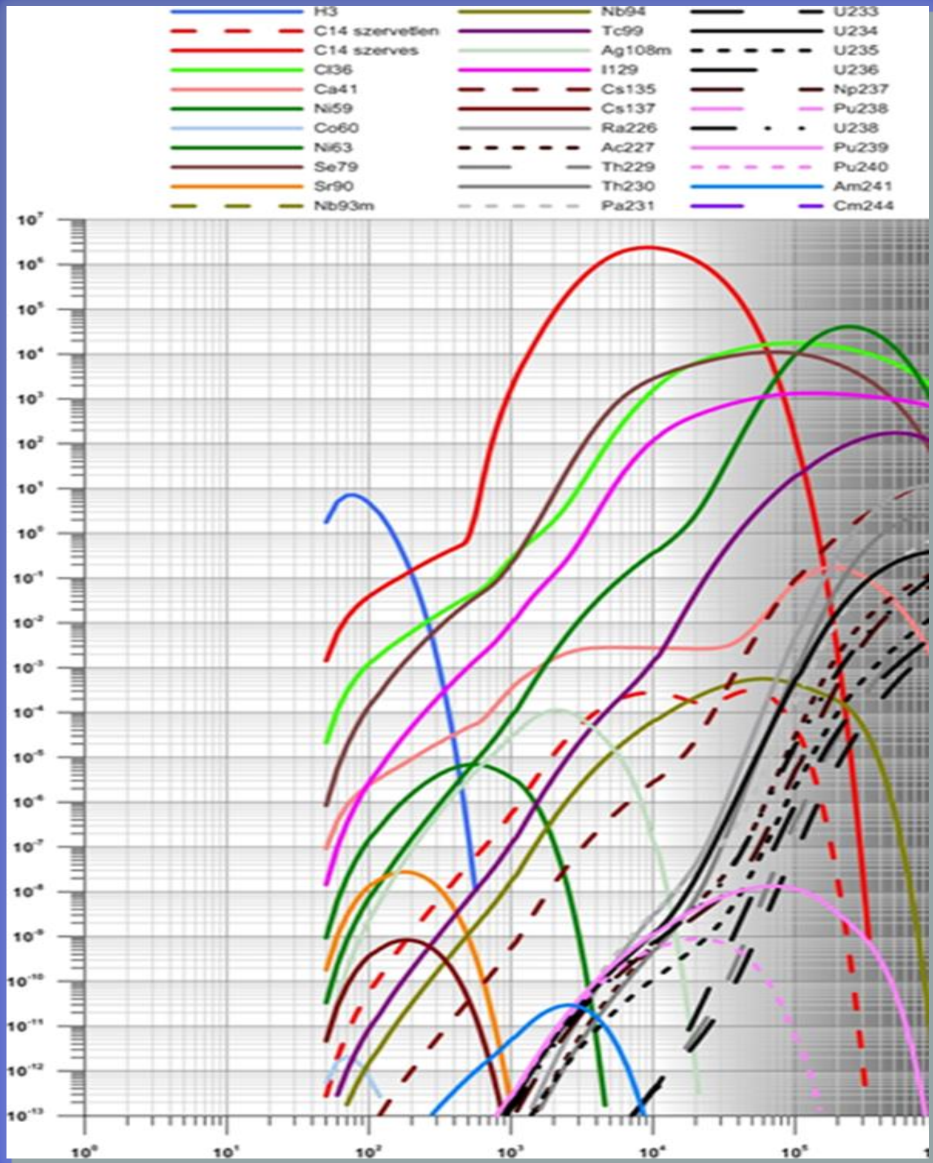
- The radioactive content of every waste type is directly or indirectly originates from the primary coolant.
- Every waste type contains easily measurable isotopes. There is a correlation between certain easily measurable and difficult-to-measure (DTM) isotopes
  - Linear relationship (SF model): $C_{DTM}=SF*C_{KN}$
  - Non-linear relationship (LOG model): $C_{DTM}=a*C_{KN}^{b}$
- The key nuclide (KN) has to be relatively long-lived to be traceable during the characterization process. The chemical behaviour of the KN has to be similar to that of the related DTM. The gamma-intensity of the KN has to be high enough for low-error measurements.
- The DTM isotopes' activity concentration are usually several order of magnitude lower than the „bulk" (mostly ^{60}Co + ^{137}Cs), due to
- Low formation probability (fission or activation); Low „escape" probability (from an intact fuel rod); Long half-life ► low specific activity; They are either pure alpha/beta emitters or have only very low energy and intensity X-ray or γ-ray lines.
- Result: The determination of their AC requires a radiochemical separation of various complexity and / or special measurement method.
  - Clearance Criterion: $\Sigma (A_i / A_{i\ ref.\ level}) \leq 1$

Safety Assessment/Analysis



Safety Analysis





Activity concentration/time [Bq/m3_v_time] **(Excavation Damaged Zone)** **Activity flux/time [Bq/year_v_time]**

Nuclides to determine

Nuclide	$T_{1/2}$ (year)	MDA [Bq/l]	Type of measurement technique
^{3}H	12,32	1,0	LSC
^{14}C	5730	1,0	LSC, AMS
^{36}Cl	301000	10,0	LSC
^{41}Ca	102000	10,0	AMS, LSC
^{54}Mn	0,85	NR	gamma spectrometry
^{55}Fe	2,73	1,0	X-ray spectrometry
^{60}Co	5,27	10,0	gamma spectrometry
^{59}Ni	75000	1,0	X-ray spectrometry
^{63}Ni	100	10,0	LSC
^{79}Se	327000	10,0	LSC/ICP-MS
^{90}Sr	29,1	1,0	LSC
^{93}Zr	1500000	1,0	ICP-MS
^{93m}Nb	16,1	10,0	X-ray spectrometry
^{94}Nb	20300	1,0	gamma spectrometry
^{99}Tc	211000	0,1	LSC
^{106}Ru	1,01	NR	gamma spectrometry (^{106}Rh)
^{107}Pd	6500000	1,0	LSC
^{108m}Ag	418	1,0	
^{110m}Ag	0,68	NR	gamma spectrometry
^{125}Sb	2,76	NR	
^{129}I	15700000	0,1	LSC, TW-gamma spectrometry
^{134}Cs	2,06	NR	gamma spectrometry
^{135}Cs	2300000	1,0	NAA ICP-MS
^{137}Cs	30,07	10,0	
^{144}Ce	0,78	NR	gamma spectrometry
^{154}Eu	8,8	NR	
^{234}U	244000	0,05	
^{235}U	704000000	0,05	
^{237}Np	2100000	0,05	alpha spectrometry
^{238}U	4470000000	0,05	
^{238}Pu	86,4	0,05	^{238}U, ^{235}U, ^{237}Np, ^{239}Pu
^{239}Pu	24400	0,05	ICP-MS
^{240}Pu	6540	0,05	
^{241}Am	432	0,05	^{241}Am, ^{235}U
^{243}Am	7380	0,05	gamma spectrometry
^{242}Cm	0,45	0,05	
^{244}Cm	17,9	0,05	

NR: Not relevant for long term safety case

Radioactivity measurement

Liquid scintillation counters

Alpha spectrometry

Gross alpha/beta measurement

Gamma spectrometry

Mass spectrometers

Environ MICADAS AMS

HELIX-SFT Noble gas mass spectrometer

QQQ-8800 ICP-MS

Q-MS

Delta Plus XP Stable Isotope Mass Spectrometer

Scaling Factors

DTM isotope	KN	Parameters		Coefficient of determination	Coefficient of correlation	Statistical test 95% confidence interval		Quality of fitting
Nuclide		m	b	r	R	F/F_{krit}	t/t_{krit}	
^{3}H	^{60}Co	0,138	2,84E+04	0,192	0,438	12,663	3,556	proper
^{14}C	^{137}Cs	-0,071	1,60E+04	0,034	-0,183	1,681	-1,296	bad*
		0*	1*					
^{36}Cl	^{137}Cs	0,398	3,31E-02	0,110	0,332	3,527	1,878	proper
^{41}Ca	^{137}Cs	0,494	1,49E-03	0,231	0,481	4,657	2,158	good
^{55}Fe	^{60}Co	0,554	2,70E+00	0,350	0,592	19,687	4,437	good
^{59}Ni	^{60}Co	0,479	6,30E+00	0,444	0,667	34,564	5,879	very good
^{63}Ni	^{60}Co	0,712	1,48E+01	0,828	0,910	242,328	15,567	excellent
^{79}Se	^{137}Cs	0,553	7,20E-02	0,288	0,537	0,910	0,954	good
^{90}Sr	^{137}Cs	0,406	8,13E-01	0,136	0,401	8,620	2,936	proper
^{94}Nb	^{60}Co	0,172	1,37E+01	0,103	0,321	0,518	0,720	proper
^{99}Tc	^{60}Co	0,246	1,62E-01	0,193	0,440	8,917	2,986	proper
^{108m}Ag	^{60}Co	0,888	1,89E-03	0,443	0,666	7,560	2,750	very good
^{129}I	^{137}Cs	0,701	2,33E-05	0,481	0,693	42,363	6,509	very good
^{234}U	^{238}Pu	0,266	2,02E-02	0,462	0,679	43,511	6,596	very good
^{235}U	^{238}Pu	0,311	2,70E-02	0,455	0,675	36,798	6,066	very good
^{237}Np	^{60}Co	0,556	1,96E-02	0,203	0,451	1,084	1,041	good
^{238}U	^{238}Pu	0,306	2,22E-01	0,433	0,604	41,373	6,432	very good
^{238}Pu	^{60}Co	0,415	1,27E-03	0,337	0,581	22,636	4,758	good
$^{239+240}$Pu	^{60}Co	0,545	2,41E-04	0,347	0,589	24,180	4,917	good
^{241}Am	^{60}Co	0,696	1,10E-04	0,499	0,706	43,810	6,619	very good
^{242}Cm	^{60}Co	0,441	1,85E-04	0,247	0,497	11,460	3,385	good
^{244}Cm	^{60}Co	0,605	7,68E-05	0,399	0,632	27,765	5,269	very good

$A_{DTM} = m * A_K{}^b$

A_{DTM} – activity of DTM

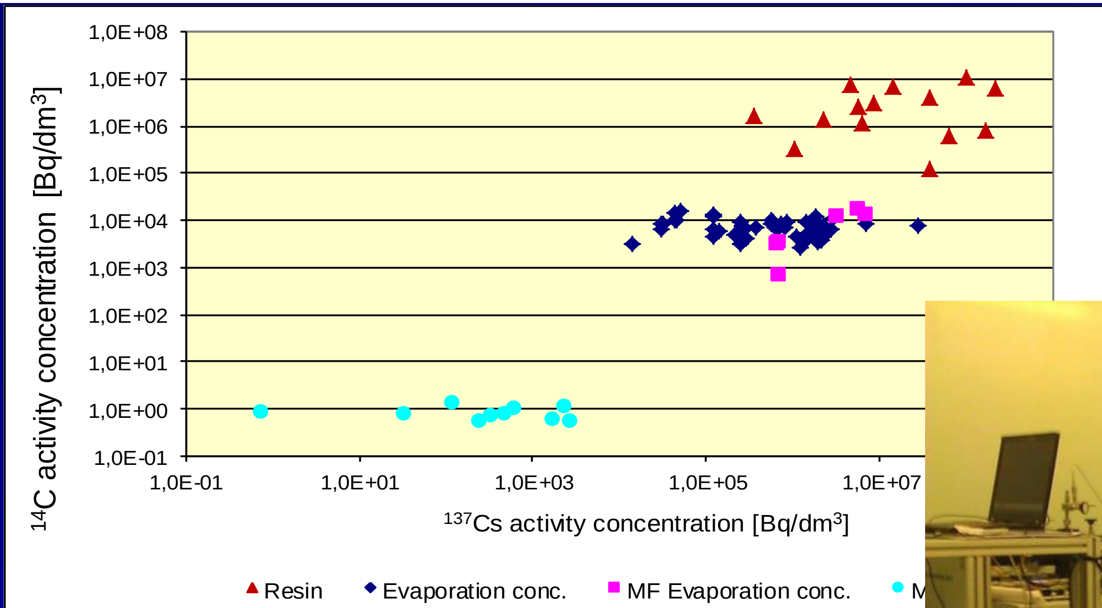
A_{KN} – activity of key nuclide

m,b – scaling factors

*suggested parameters

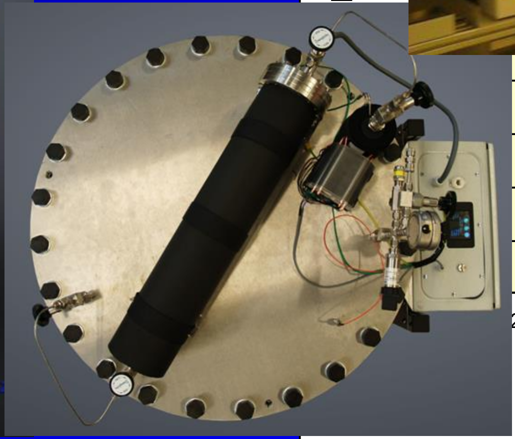
R<0,3	→	bad
If 0,3≤R<0,45	→	proper
If 0,45≤R<0,6	→	good
If 0,6≤R<0,75	→	very good
If 0,75≤R	→	excellent

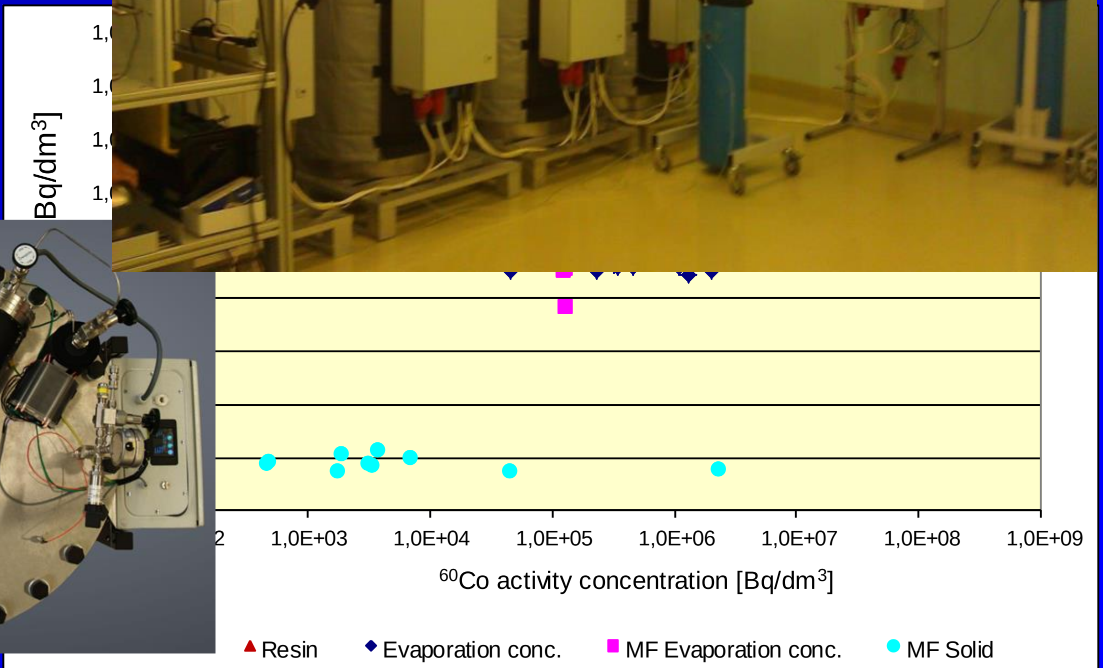
Scaling Factor (^{14}C)



How can we solve this problem?

- Individual sampling (difficult, not representative)
- Calculation (from what?)
- Gas generation?





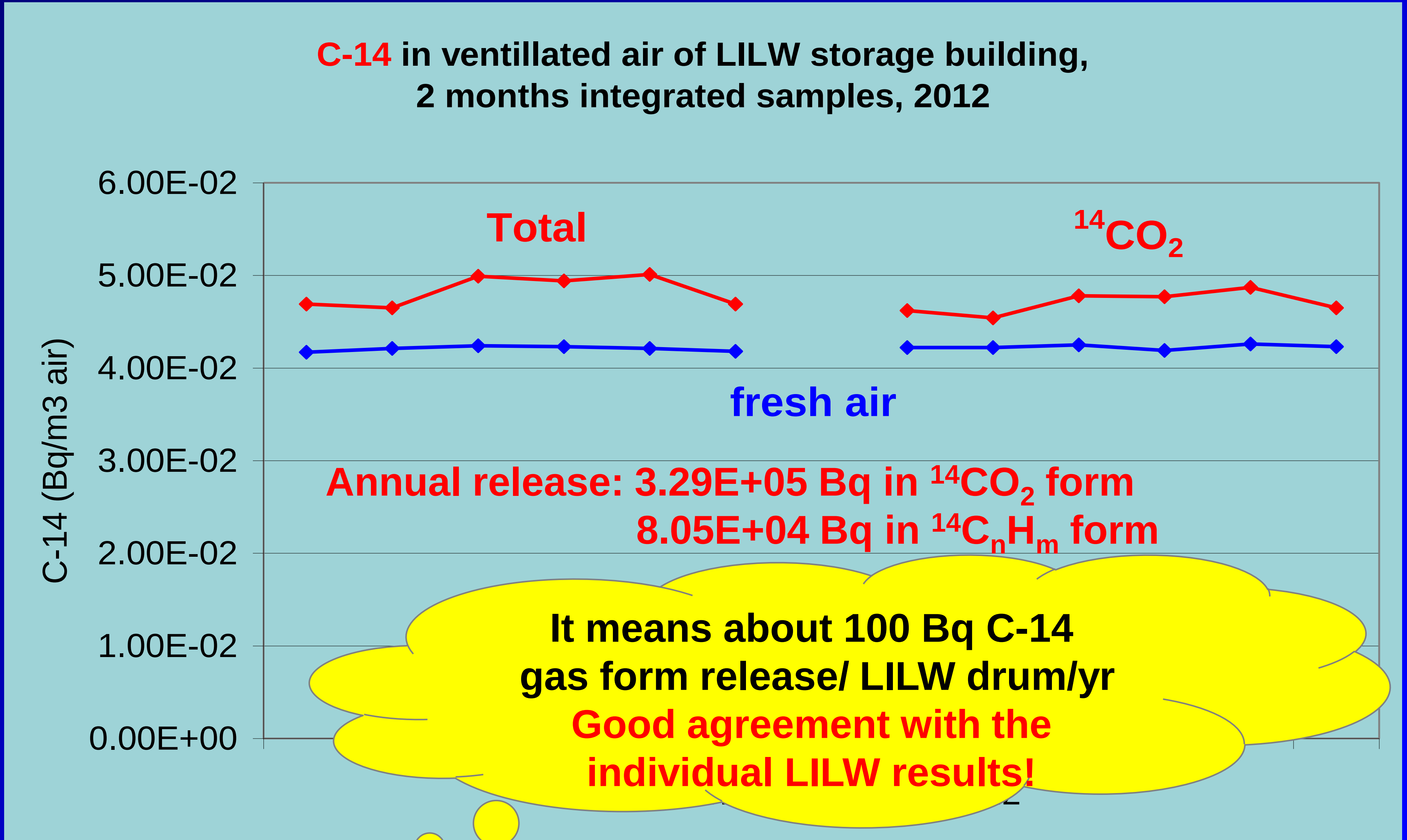
^{14}C release by the gas generation of L/ILW

Interim storage building with 3000 LILW drums

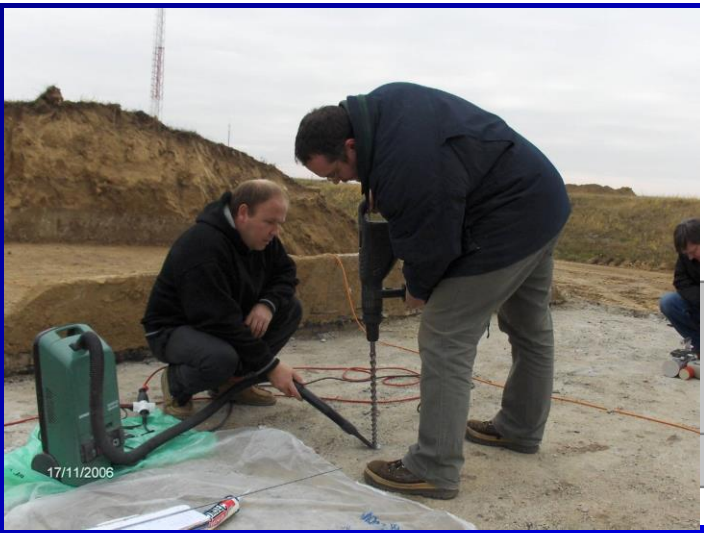
controlled ventilation and C-14/H-3 sampling

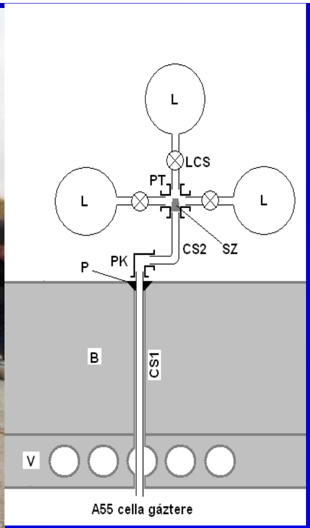
^{14}C release by the gas generation of L/ILW

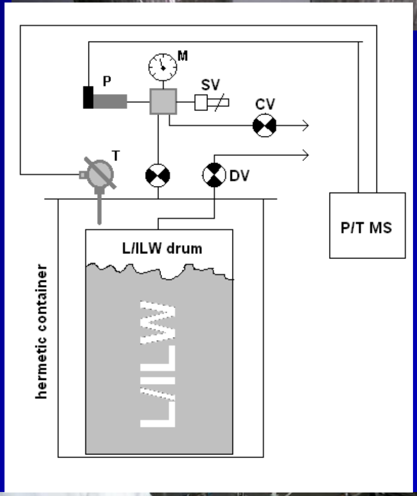
Interim storage building with 3000 LILW drums controlled ventilation and C-14/H-3 sampling



^3H inventory estimation with $^3H/^3He$ method







$$N(^3He)=\Delta N(^3H)=N(t_1)-N(t_2)=N(t_1)\cdot(1-e^{-\lambda\Delta t})$$

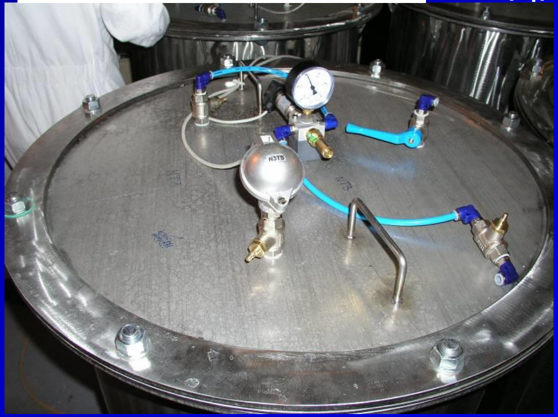
$N(^3He)$ is the number of tritiogenic ^3He atoms

$\Delta N(^3H)$ is number of tritium atoms that have already decayed

$N(t_1)$ and $N(t_2)$ are the numbers of tritium at time t_1 and t_2

Δt is the difference between t_1 and t_2

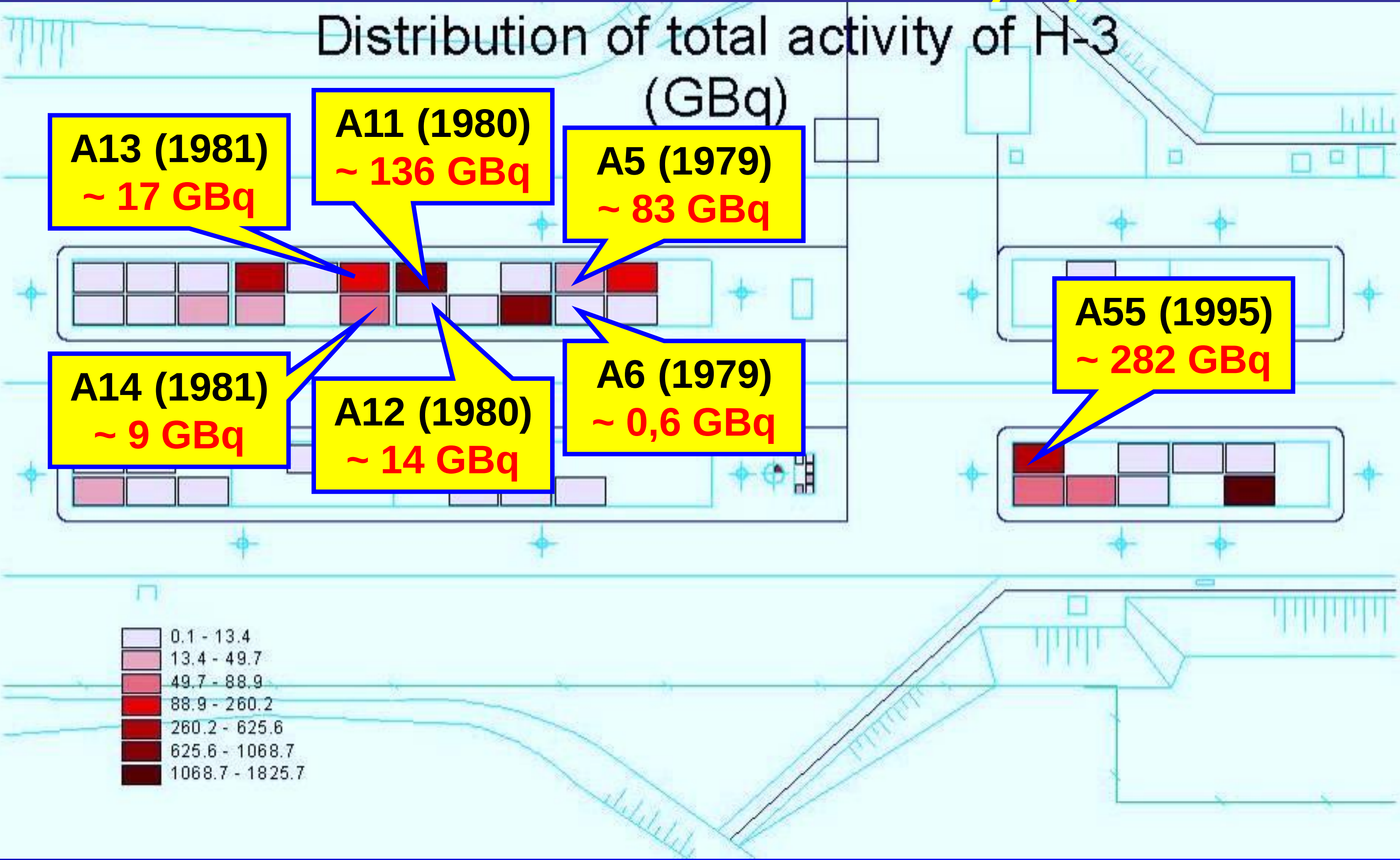
λ is the decay constant of tritium.



HELIX (Thermo Instruments)

^{3}H inventory estimation with $^{3}H/^{3}He$ method

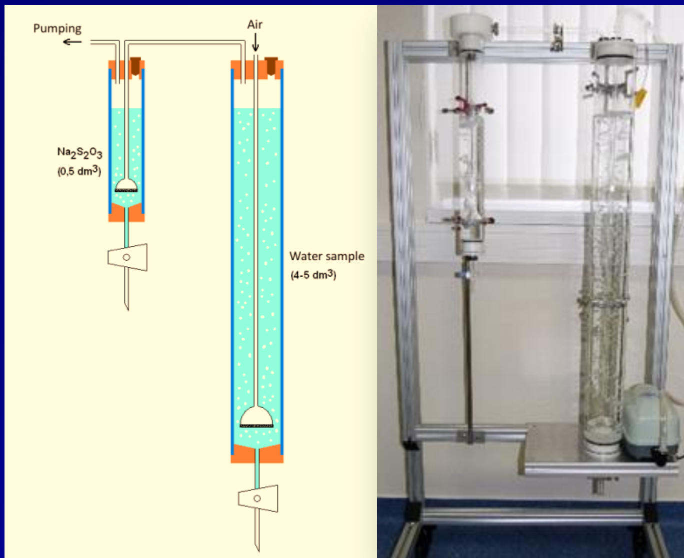
Estimation of total restored ^{3}H activity by ^{3}He results



^{3}H → ^{3}He, number of produced ^{3}He atoms is equal with number of decayed 3H atoms, if the vault is closed (enough) for gases…

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

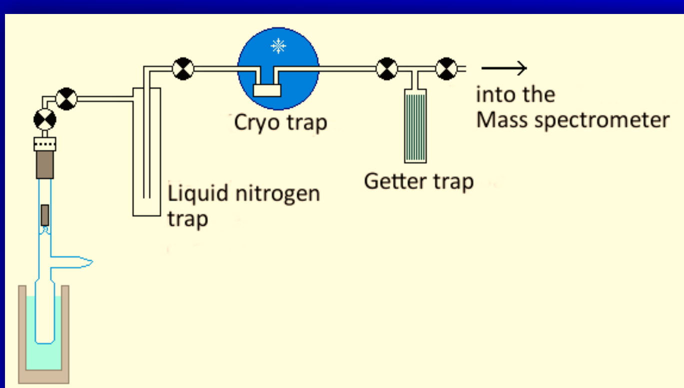
^{129}I activity measurement with ^{129}I/^{129}Xe method



Carrier: Potassium-Iodide,-Iodate + H_2SO_4
Purge with airflow
Trapped in Sodium thiosulfate

Degassing, set aside

Xe freezing out



VG5400 (Fison Instruments)

$$C = N_{doug} \cdot \frac{e^{\lambda t_{SD}}}{\left(1 - e^{-\lambda t_{DM}}\right)} \cdot \lambda \cdot \frac{1}{V_{samp}}$$

t_{SD}: $t_{sampling}$ - $t_{degassing}$

t_{DM}: $t_{measurement}$ - $t_{degassing}$

Summary

- **We can measure most of the DTM isotopes with the proper detection limit in liquid radioactive waste**
- **New methods are welcome (faster, cheaper, no radiochemical pre-treatment are necessary to apply them).**
- **We have significant uncertainty (in some cases) when applying Scaling-Factor. In most cases it is impossible to check its uncertainty.**
- **R&D needed to <u>develop fast, onsite measurement</u> techniques (decommissioning, safety improvement programs, re-characterization of old disposal facilities, legacy waste)**
- **The success of minimization, treatment and conditioning depends largely upon this knowledge.**
- **The result of the „under" characterization that the cost of the disposal will increase! (more robust waste packages, engineering barriers)**

Thank you for your attention!

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