Development of an In-Situ Radiological Classification Technique for Material and Waste from CERN's Accelerators Facilities

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Content

- Introduction
- **Clearance** in Switzerland
- RADOS RTM 661/440 lnc, a **total gamma counter**
- **Determination** of Radionuclide Inventories
- **Visualization** of Radionuclide Inventories
  - Evolution of Activity over Time
  - Importance on Clearance Limit
  - Resulting Gamma Signal per Clearance Limit
- **Analysis** of Radionuclide Inventories
  - Minimum Gamma Signal per Clearance Limit
  - Enveloping Radionuclide Inventories
- Evaluation of **Geometry Effects** beyond calibration conditions
- Summary & Further Developments

Introduction

Material taken out of the accelerator facilities and experimental zones originates from maintenance, repair and upgrade actions as well as from decommissioning. Because of beam losses and subsequent particle interactions, radioactivity can be induced in certain accelerator components.

The particular conditions at CERN

- wide ranges of particle energies and irradiation conditions
- many different typed of materials

lead to a large number of different and time dependent radionuclide inventories.

Swiss Clearance Regulation

- Surface Contamination < 1 CS
  (sum over all radionuclides)
- Dose Rate < 0.1 μSv/h
  (10 cm distance)
- **Mass Specific Activity < 1 LE**
  (clearance limits = exemption limits)

$$R = \sum_i \frac{a_i}{LE(swiss)_i} < 1$$

Isotope	LE [Bq/g]
H-3	200
Be-7	400
Na-22	3
Na-24	20
Cl-36	10
Sc-44	30
Ti-44	2
V-49	600
Cr-51	300
Mn-54	10
Fe-55	30
Co-57	50
Co-58	10
Co-60	1
Ni-63	70
Cu-64	80
Zn-65	3
Ag-110	4
Cs-137	0.8
Eu-152	7

RADOS RTM 661/440 lnc

Technical Details

- Total gamma counting device with a 4π detector geometry (almost full solid angle)
- 6 large area plastic scintillation detectors
- Thick lead shielding (5 cm)
  - Low radiation background of 12-15 nSv/h
- Barcode reader
- Network connection
- Integrated scale
- Report printer
- QNX 6.5 (unix system)

RADOS RTM 661/440 lnc

Calibration and *lnc* Method

- Volume calibration performed with a metal plate phantom and Co-60 area sources (up to a density of 1 g/cm^3);
- For other radionuclides the build-in *lnc-method* (*leading nuclide correlation*) is used;

$$A_b = \frac{M}{lnc_b}$$

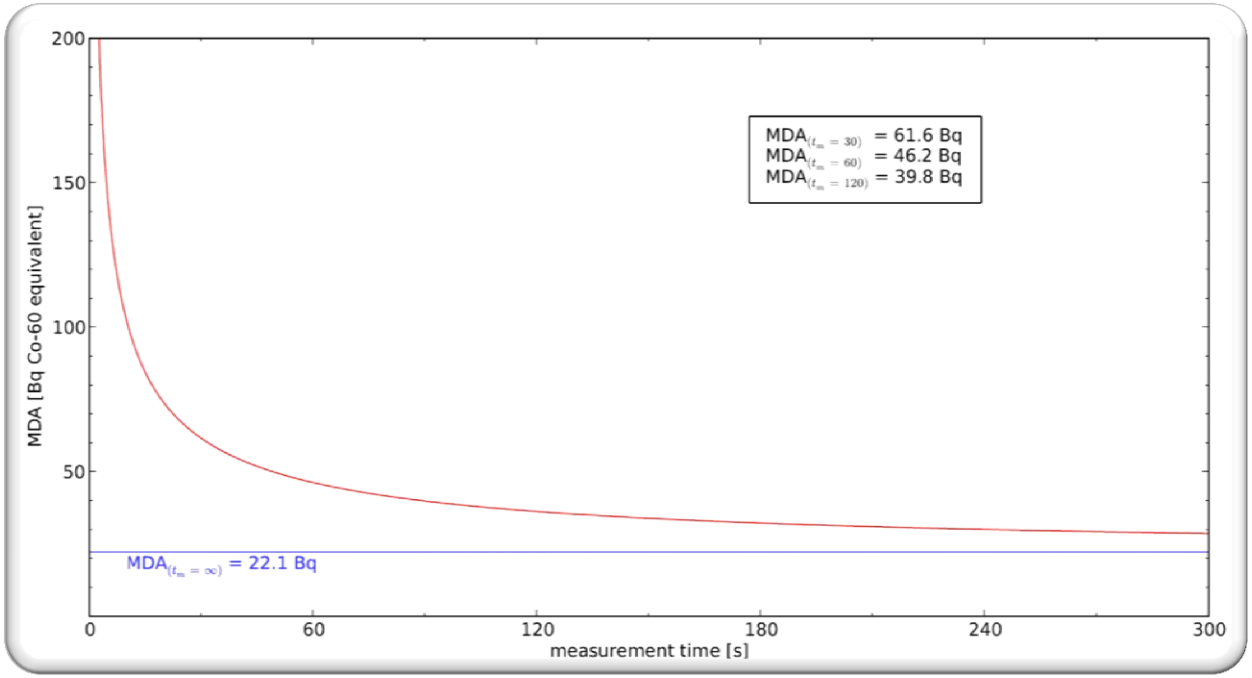
- A *lnc*-factor list is provided by the manufacturer for common radionuclides. The list is not exhaustive and certain radionuclides that can be produced by induced activation processes are missing.

➢ **A detector response curve over the photon energy span of interest was determined and allows us to calculate additional *lnc*-factors.** ('lnc fitter')

RADOS RTM 661/440 lnc

Minimum Detectable Activity, MDA

MDA of the RADOS RTM 661/440 lnc following the ISO11929



*curve calculated for
$t_0 = 180$ s
bg = 0.015 µSv/h
error quantiles:
$k_{1-\alpha} = k_{1-\beta} = 1.64$

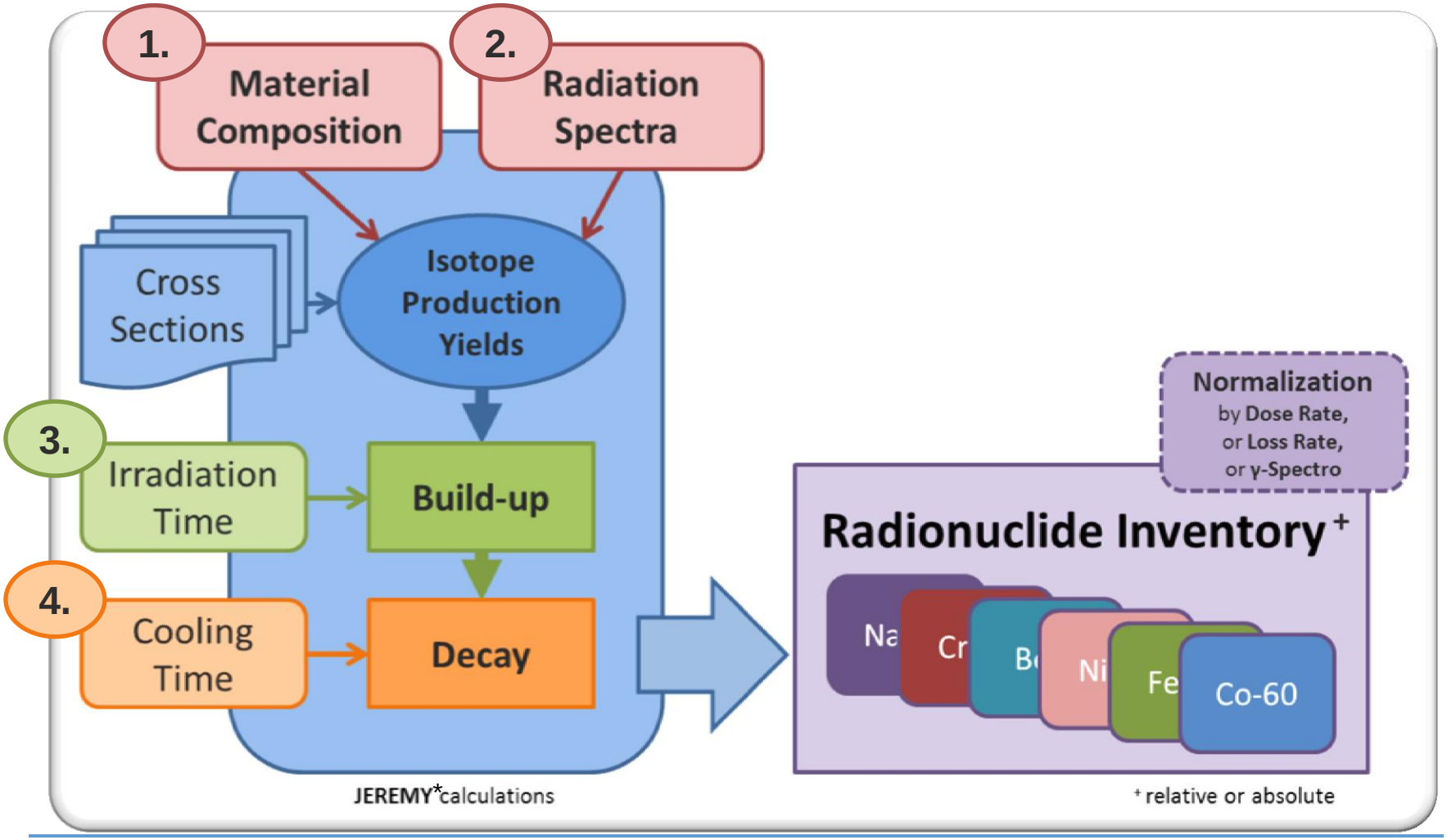
RADOS RTM 661/440 lnc

Limiting Factors

- Detection of gamma emitters ONLY;
- Total gamma counting, i.e. NO photon energy spectra;
- **Relative Radionuclide Inventory as input parameter required!**
- Calibration assumes homogeneous activity distribution;
- Sensitive on background variation (e.g. air activation);
- Sensitive on electrostatic effects (e.g. on plastic objects);
- Object size limited by the chamber volume
- Object weight is limited to 20 kg (manual loading)

Determination of Radionuclide Inventories

Scheme of determination process



*R.FROESCHL, M.MAGISTRIS, F.L.PEREIRA, "JEREMY, a code for the radiological characterization of accelerator components including detailed uncertainty estimation.", 2nd workshop on Accelerator Radiation Induced Activation(ARIA), Jerusalem, 2011

Determination of Radionuclide Inventories

1. Material Compositions

- Determination of a set of *Common Materials*
- Worst Case assumption with *Pure Iron* (no elementary Co)

(a) *Common Steel*

Element	Weight [%]
Fe	67.6
Cr	18.0
Ni	10.0
Ti	1.0
Mn	1.0
Si	1.0
Mo	1.0
Co	0.1
C	0.1
P	0.1
S	0.1

(b) *Common Copper*

Element	Weight [%]
Cu	86.2
Ni	10.0
Fe	1.0
Be	1.0
Mn	1.0
Al	0.1
Co	0.1
Cr	0.1
P	0.1
S	0.1
Pb	0.1
Si	0.1
Zr	0.1

(c) *Common Aluminium*

Element	Weight [%]
Al	93.5
Si	1.0
Cr	1.0
Cu	1.0
Fe	1.0
Mg	1.0
Mn	1.0
Ti	0.1
Ni	0.1
V	0.1
Zn	0.1
Zr	0.1

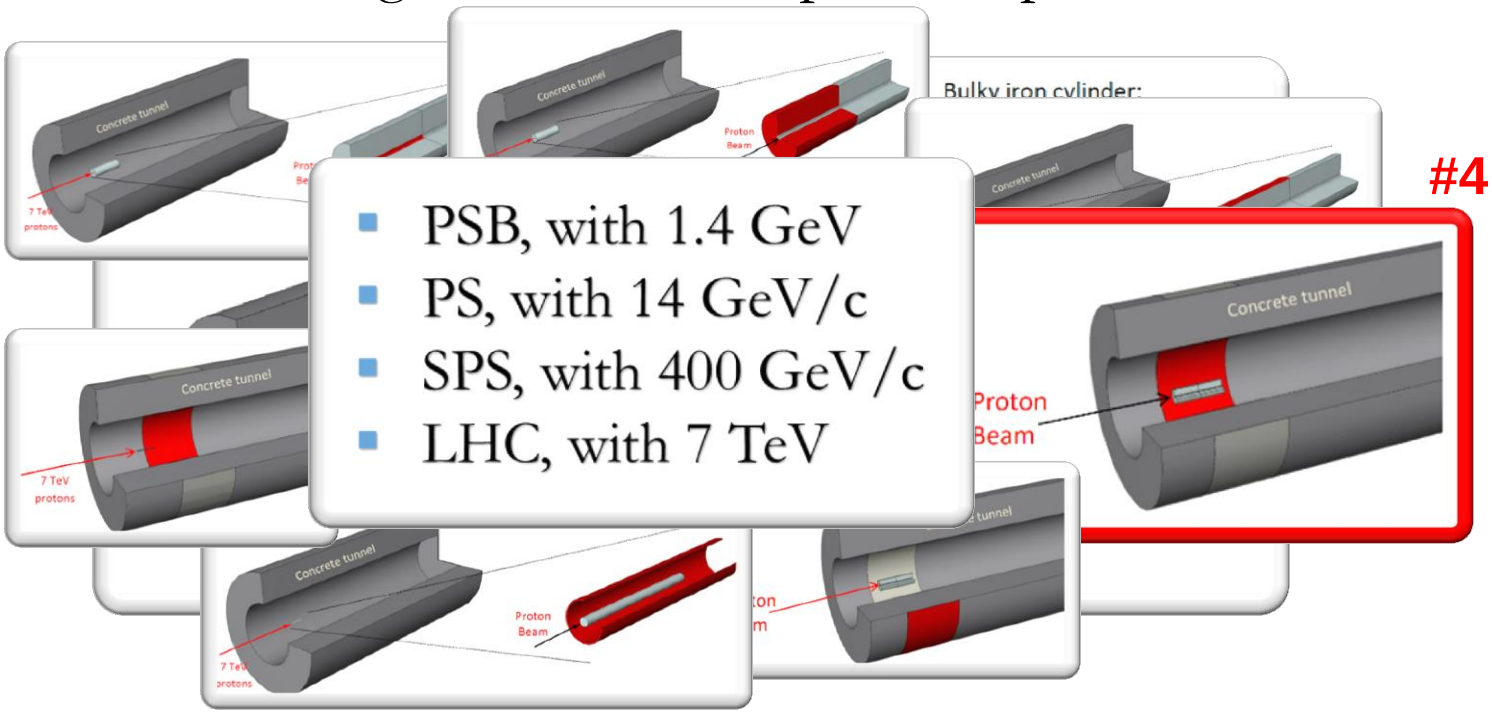
+ *Pure Iron* 100 % Fe

Determination of Radionuclide Inventories

2. Particle Spectra

Classification of representative particle spectra (ActiWiz* project) by

- ActiWiz position & Accelerator energy
- 7 positions x 4 energies = 28 sets of particle spectra



- PSB, with 1.4 GeV
- PS, with 14 GeV/c
- SPS, with 400 GeV/c
- LHC, with 7 TeV

*H. VINCKE and C. THEIS, "ActiWiz - optimizing your nuclide inventory at proton accelerators with a computer code," Progress in Nuclear Science and Technology, vol. 4, pp. 228-232, 2014.

Determination of Radionuclide Inventories

3. Irradiation Times

- **60 days** - a very short irradiation period that is the typical time span between two technical stops
- **1 year** - a short irradiation period, e.g. a typical interval between accelerator runs
- **3 years** - a medium-length irradiation period that corresponds to a typical short technical life time of an object placed in the accelerator
- **10 years** - a long irradiation period, e.g. a typical medium technical life time of an object placed in the accelerator
- **30 years** - a very long irradiation period that represents the typical nominal life time of the CERN accelerators

Determination of Radionuclide Inventories

4. Cooling Times

- **1 to 7 day**(s) - i.e. for maintenance interventions
- **1 to 5 week**(s) - i.e. administrative time before immediate shipping or machining
- **1 to 12 month**(s) - i.e. non time-critical shipping or tooling
- **1 to 30 years**(s) - i.e. typical waiting time for historical waste

Visualization of Radionuclide Inventories

Motivation

4 materials x 28 spectra x 5 irradiation times x 41 cooling times = 22'960 Radionuclide Inventories

were calculated by interfacing the JEREMY code with a self-developed PYTHON script.

I. Visualization: A practicable and clear way to make use of this large amount of data was developed.

II. Analysis: A method to evaluate and compare different radionuclide inventories was developed.

Visualization of Radionuclide Inventories

Activity Fractions Plot 1/2

Visualization of activity fractions over cooling time:

$$a_b^{\mathrm{rel}}(t_{\mathrm{cool}}) = \frac{a_b(t_{\mathrm{cool}})}{\sum_i a_i(t_{\mathrm{cool}})}$$

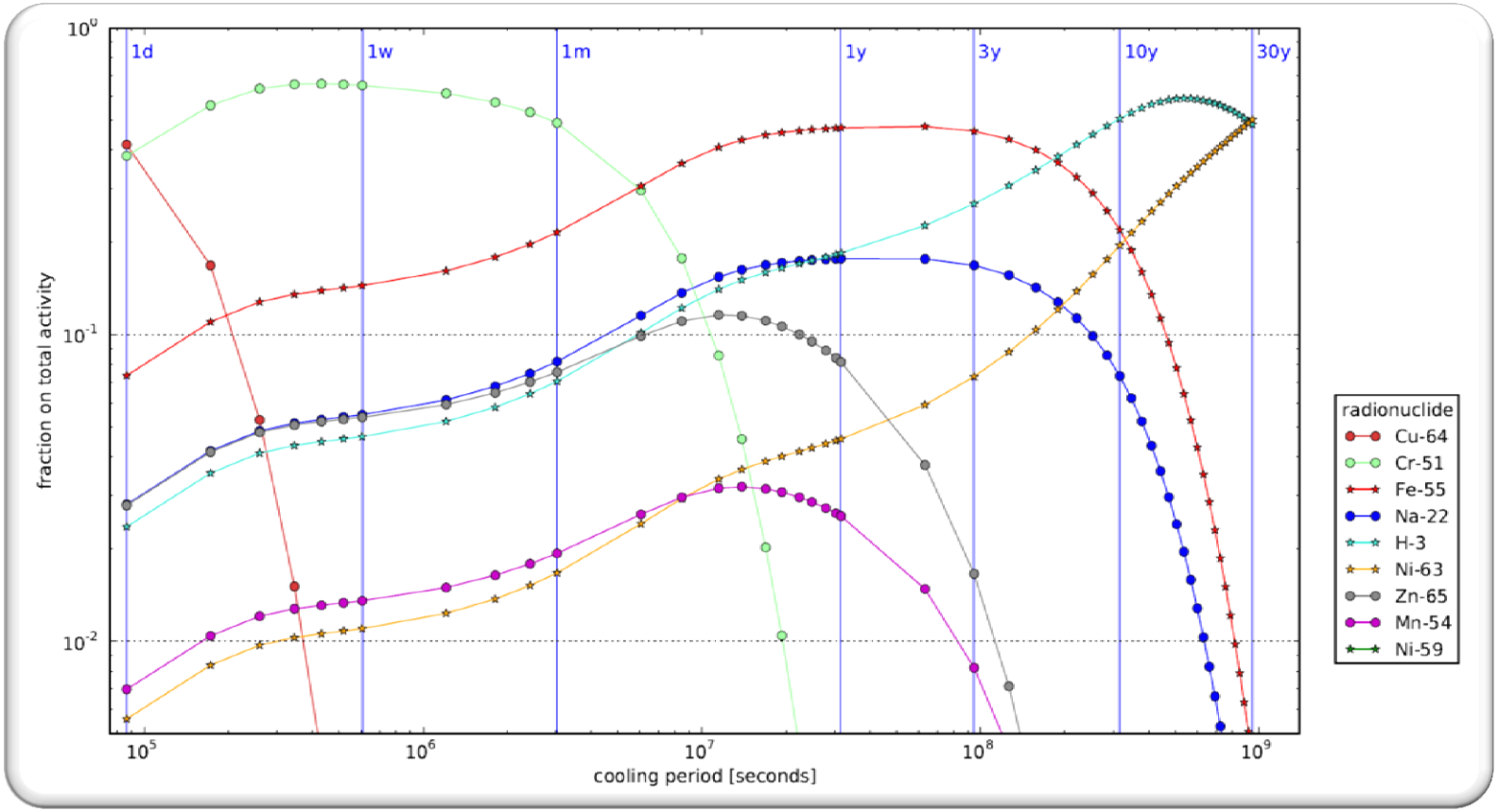
- End of Irradiation period = initial point for the plot;
- Evolution over the cooling time range;
- Circle markers = *Key radionuclide** ('gamma emitter')
- Star markers = *Difficult-to-measure radionuclide**

* Classification in 'Key / Difficult-to-measure radionuclide' according to ISO21238

Visualization of Radionuclide Inventories

Activity Fractions Plot 2/2

- Example shows *Common Aluminium* after 30 years of irradiation at ActiWiz position #4 in the PSB



Visualization of Radionuclide Inventories

Stacked Bar Chart of Clearance Limit Fractions 1/2

Visualization of the fractions of the clearance limit

$$LE_b^{\mathrm{rel}}(t_{\mathrm{cool}}) = \frac{\frac{a_b(t_{\mathrm{cool}})}{LE_b}}{LE(t_{\mathrm{cool}})} \qquad \text{with} \qquad LE(t_{\mathrm{cool}}) = \sum_i \frac{a_i(t_{\mathrm{cool}})}{LE_i}$$

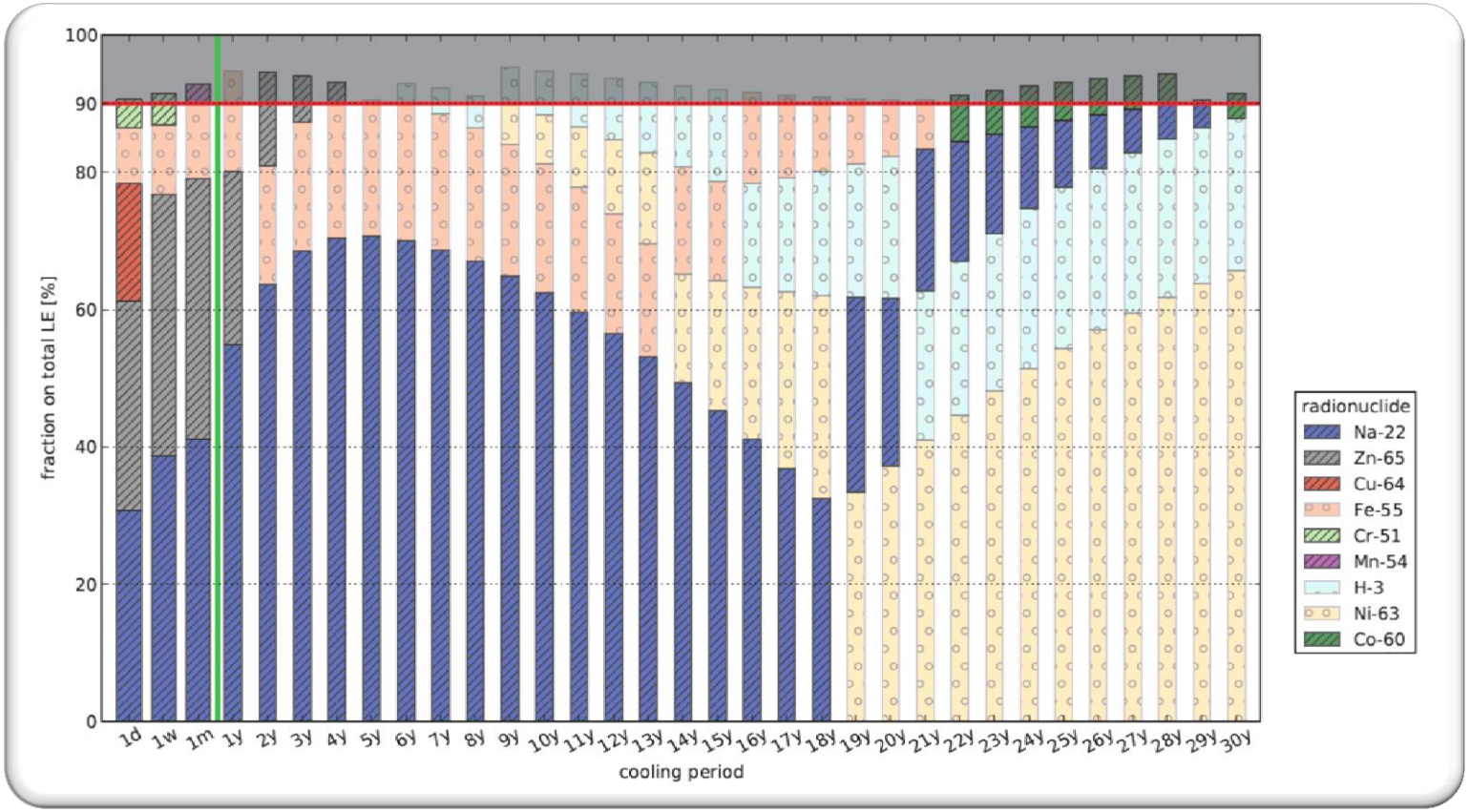
- Activities weighed with the corresponding clearance limits;
- Normalized to the activity threshold for clearance of LE = 1
- Fractions are ordered by size and piled up on each other up to 90 % of the total limit (according to the Swiss clearance guide-line ENSI-B04*).
- Intensive colors and stripe hatch = *Key radionuclide*
- Pastel colors and circle hatch = *Difficult-to-measure radionuclide*

*Swiss Federal Nuclear Safety Inspectorate ENSI, "ENSI-B04, Freimessung von Materialien und Bereichen aus kontrollierten Zonen," 2009.

Visualization of Radionuclide Inventories

Stacked Bar Chart of Clearance Limit Fractions 2/2

- Example shows *Common Aluminium* after 30 years of irradiation at ActiWiz position #4 in the PSB



Visualization of Radionuclide Inventories

Resulting Gamma Signal per Clearance Limit 1/3

Visualisation of the resulting gamma signal per clearance limit

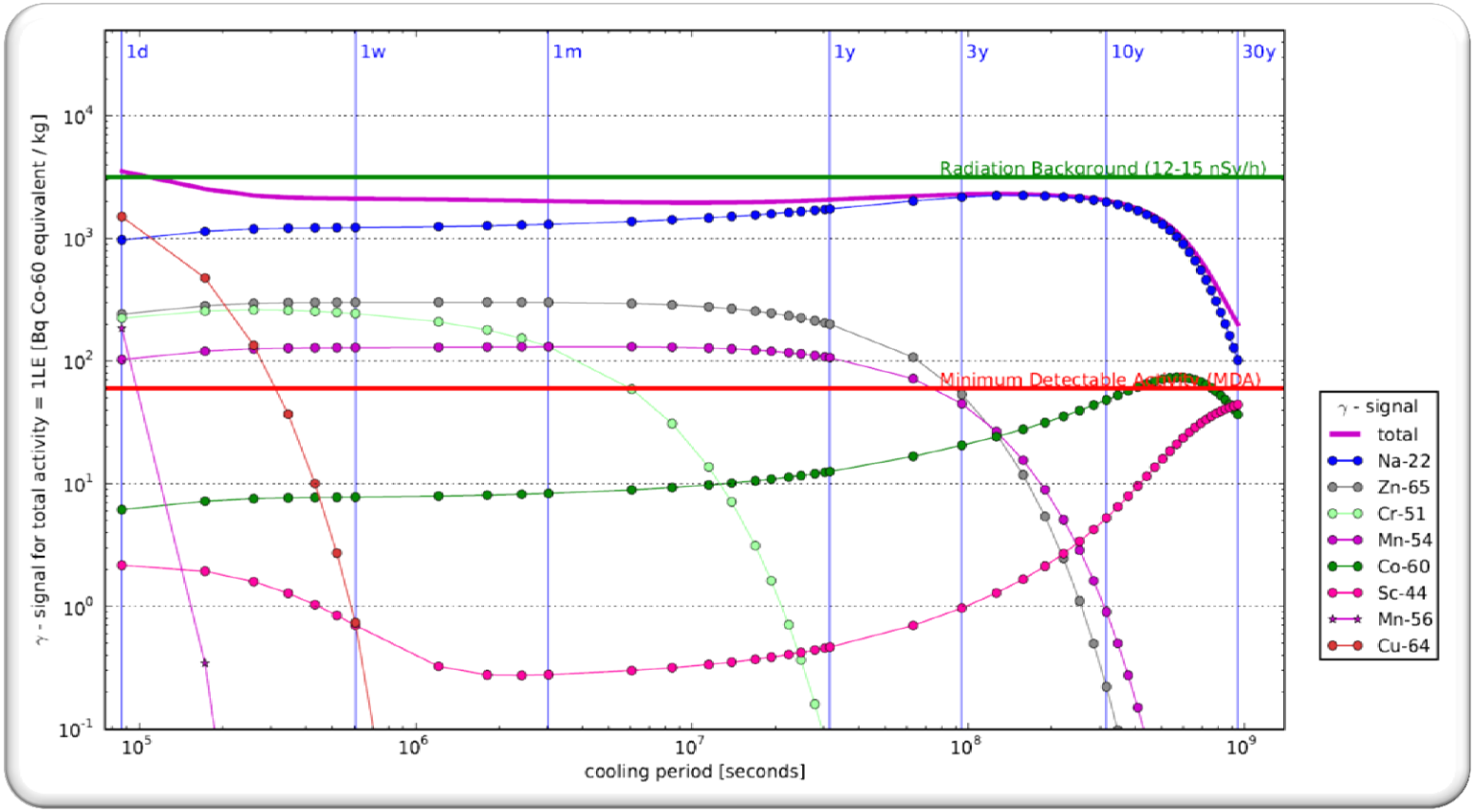
$$S_b(t_{\mathrm{cool}}) = \frac{a_b(t_{\mathrm{cool}}) \sum_E \epsilon_{b\,E}\, lnc_{\mathrm{mono}}(E)}{LE(t_{\mathrm{cool}})}$$

- Gamma signal expressed in *Bq Co-60 equivalent per kg*;
- Device efficiency taken into account by applying the determined detector response curve ('lnc fitter');
- Device MDA of 60 *Bq Co-60 equivalent*
  (Chamber volume calibration, homogeneous mass and activity distribution up to 50 kg, material density of 1 g/cm^3);
- Radiation background of 3'170 *Bq Co-60 equivalent*
  (corresponds to 12-15 nSv/h in the lead shielded measurement chamber).

Visualization of Radionuclide Inventories

Resulting Gamma Signal per Clearance Limit 2/3

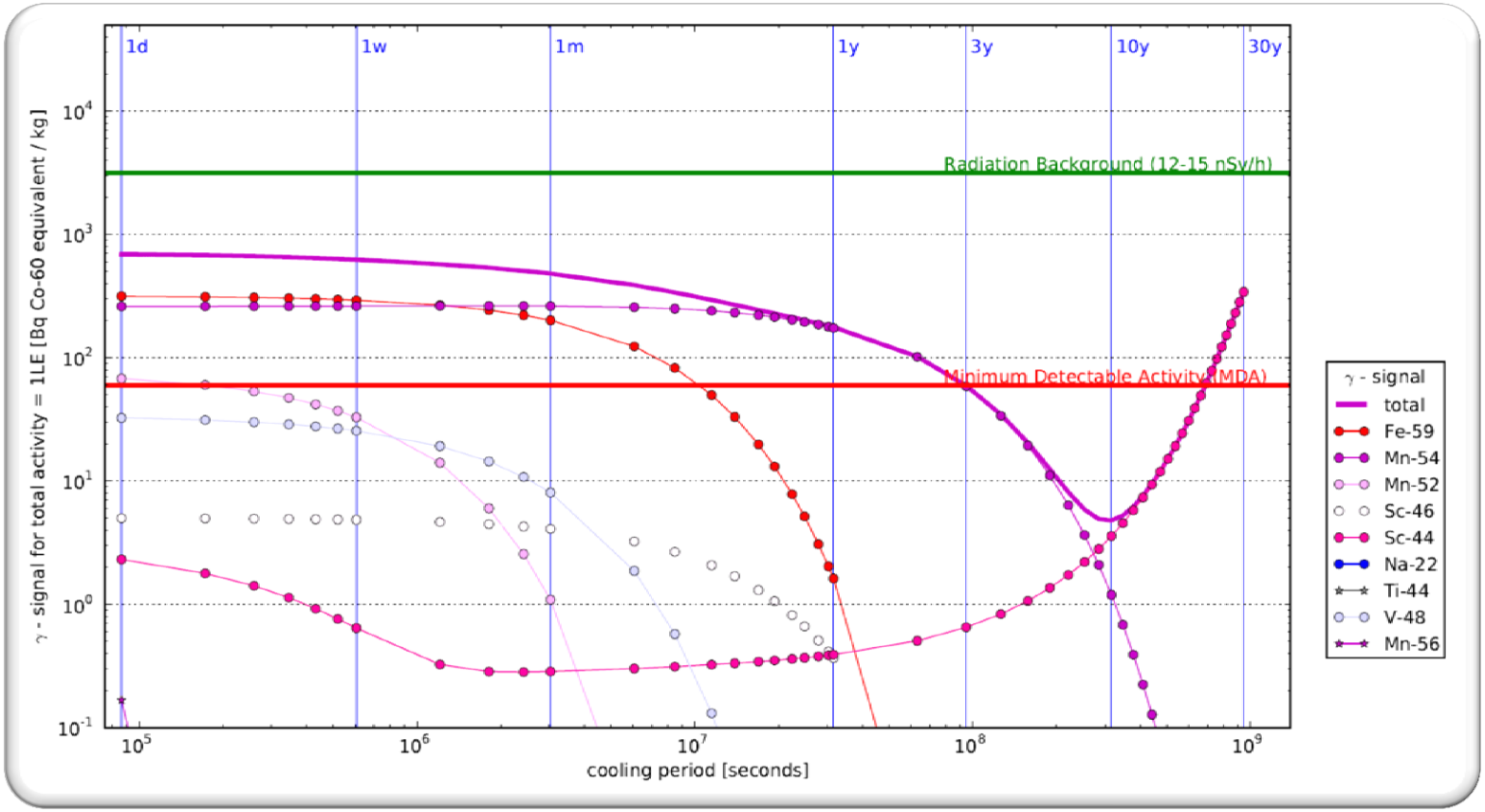
- Example shows *Common Aluminium* after 30 years of irradiation at ActiWiz position #4 in the PSB



Visualization of Radionuclide Inventories

Resulting Gamma Signal per Clearance Limit 3/3

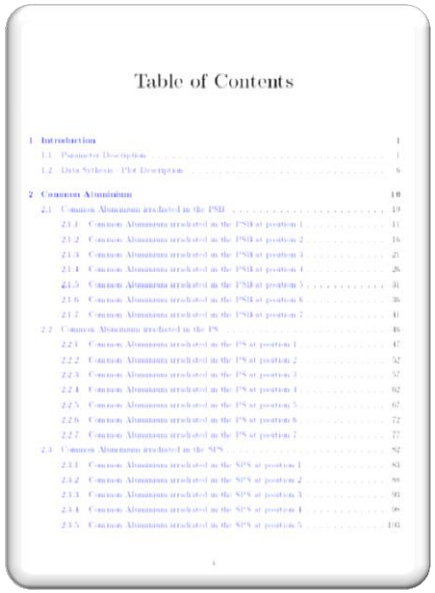
- Example shows *Pure Iron* after 30 years of irradiation at ActiWiz position #4 in the PSB

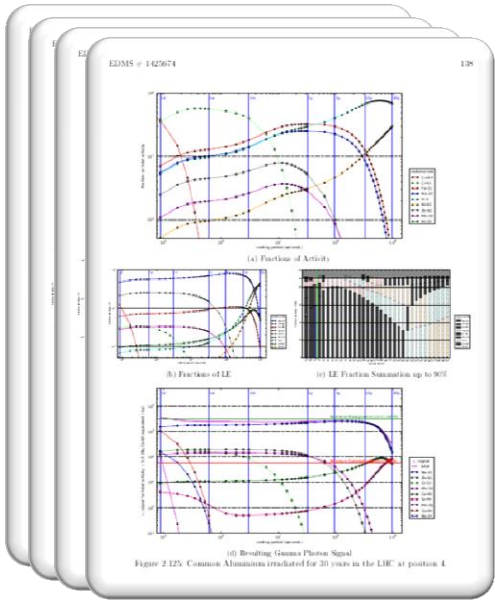


Visualization of Radionuclide Inventories

Synthesis Catalogue Available

- Synthesis of all visualization plots in a catalogue ordered by material, accelerator and irradiation scenario;
- 600 page document is published and in use for radiological characterization of waste.





Analysis of Radionuclide Inventories

Minimum Gamma Signal of *Common Materials* 1/3

- Table allows to compare the minimum resulting gamma signal per clearance limit as a function of the irradiation time and the object position to the device MDA. The presented minimum is taken over all considered cooling times as well as beam energies;
- The values are given in *Bq Co-60 equivalent per kg*;
- For the three *Common Materials* the resulting gamma signal is well above the device MDA of 60 *Bq Co-60 equivalent*.

Analysis of Radionuclide Inventories

Minimum Gamma Signal of *Common Materials* 2/3

- Minimum gamma signals for *Common Aluminium*

Position	Minimum gamma-ray signal for LE =1 [Bq ^{60}Co *equivalent* kg^{-1}]					Minimum ↔
	60 days	1 year	3 years	10 years	30 years	
# 1	651 (30 y)	639 (30 y)	560 (30 y)	440 (30 y)	387 (30 y)	387
# 2	952 (30 y)	934 (30 y)	826 (30 y)	644 (30 y)	526 (30 y)	526
# 3	984 (30 y)	965 (30 y)	849 (30 y)	644 (30 y)	481 (30 y)	481
# 4	587 (30 y)	571 (30 y)	484 (30 y)	333 (30 y)	200 (30 y)	200
# 5	1034 (30 y)	1015 (30 y)	897 (30 y)	690 (30 y)	534 (30 y)	534
# 6	808 (30 y)	794 (30 y)	701 (30 y)	555 (30 y)	481 (30 y)	481
# 7	954 (30 y)	937 (30 y)	829 (30 y)	649 (30 y)	535 (30 y)	535
Minimum ↕	587	571	484	333	200	**200**‡

Analysis of Radionuclide Inventories

Minimum Gamma Signal of *Common Materials* 3/3

- The radionuclide inventory resulting in the lowest signal per clearance limit can be taken as enveloping radionuclide inventory for the considered parameter range.

Radionuclide	Activity fraction		
	Common Steel [%]	Common Copper [%]	Common Aluminium [%]
^{55}Fe	**74**§	–	0.41
^{63}Ni	0.26	**67**§	**50**§
^{3}H	21	29	**48**§
^{60}Co	**2**§‡	**2.4**§‡	**0.04**§‡
^{54}Mn	**0.67**‡	–	–
^{44}Sc	**0.27**‡	**0.25**‡	**0.045**‡
^{22}Na	**0.25**‡	–	**0.11**‡

§ Fraction included in the inventory because of its importance on the LE (ENSI B-04)
‡ Fraction included in the inventory because of its significant gamma-ray emission

Analysis of Radionuclide Inventories

Minimum Gamma Signal of *Pure Iron* 1/3

- For the reference material *Pure Iron*, there can be radionuclide inventories within the considered parameter range with resulting gamma signals per clearance limits which are lower than the device MDA;
- The Adapted parameter range was adapted so that within this range clearance measurements with the RADOS RTM 661 are feasible;
- Table allows to compare the minimum resulting gamma signal per clearance limit as a function of the irradiation time and the object position to the device MDA. The presented minimum is taken over all considered cooling times as well as beam energies;
- The values are given in *Bq Co-60 equivalent per kg*.

Analysis of Radionuclide Inventories

Minimum Gamma Signal of *Pure Iron* 2/3

- Minimum gamma signals for *Pure Iron*
- Red signal values are below the device MDA

Position	Minimum gamma-ray signal for LE = 1 [Bq ^{60}Co *equivalent* kg^{-1}]					Minimum ↔
	60 days	1 year	3 years	10 years	30 years	
# 1	320 (11 y)	320 (10 y)	326 (9 y)	368 (8 y)	498 (7 y)	320
# 2	73 (12 y)	74 (12 y)	76 (11 y)	100 (10 y)	172 (8 y)	73
# 3	18 (9 y†)	18 (9 y†)	18 (8 y†)	25 (7 y†)	45 (8 y†)	18
# 4	1 (5 y†)	1 (5 y†)	1 (4 y†)	2 (3 y†)	4 (3 y†)	1
# 5	45 (12 y†)	45 (11 y†)	47 (11 y†)	63 (11 y)	112 (9 y)	45
# 6	167 (11 y)	169 (11 y)	174 (10 y)	218 (8 y)	345 (7 y)	167
# 7	40 (10 y†)	40 (10 y†)	42 (9 y†)	55 (9 y†)	97 (8 y)	40
Minimum ↕	1	1	1	2	4	**1**

† Shortest cooling time for which the resulting gamma-ray signal per exemption limit of the corresponding radionuclide inventory is below the device MDA.

Analysis of Radionuclide Inventories

Minimum Gamma Signal of *Pure Iron* 3/3

- Minimum gamma signals for *Pure Iron* after adapting the parameter range. The maximum cooling time is limited to 2 years of cooling.

Position	Minimum gamma-ray signal for LE = 1 [Bq ^{60}Co *equivalent* kg^{-1}]					Minimum ↔
	60 days	1 year	3 years	10 years	30 years	
# 1	2505 (24 m)	2396 (24 m)	1885 (24 m)	1439 (24 m)	1364 (24 m)	1364
# 2	2465 (24 m)	2354 (24 m)	1823 (24 m)	1361 (24 m)	1285 (24 m)	1285
# 3	1533 (24 m)	1433 (24 m)	1010 (24 m)	698 (24 m)	652 (24 m)	652
# 4	264 (24 m)	240 (24 m)	152 (24 m)	97 (24 m)	90 (24 m)	90
# 5	2635 (24 m)	2525 (24 m)	1989 (24 m)	1504 (24 m)	1419 (24 m)	1419
# 6	2533 (24 m)	2423 (24 m)	1899 (24 m)	1439 (24 m)	1361 (24 m)	1361
# 7	1748 (24 m)	1642 (24 m)	1181 (24 m)	830 (24 m)	780 (24 m)	780
Minimum ↕	264	240	152	97	90	**90** = **1.5 x MDA**

Evaluation of Geometry Effects

Self-absorption beyond Calibration Conditions 1/2

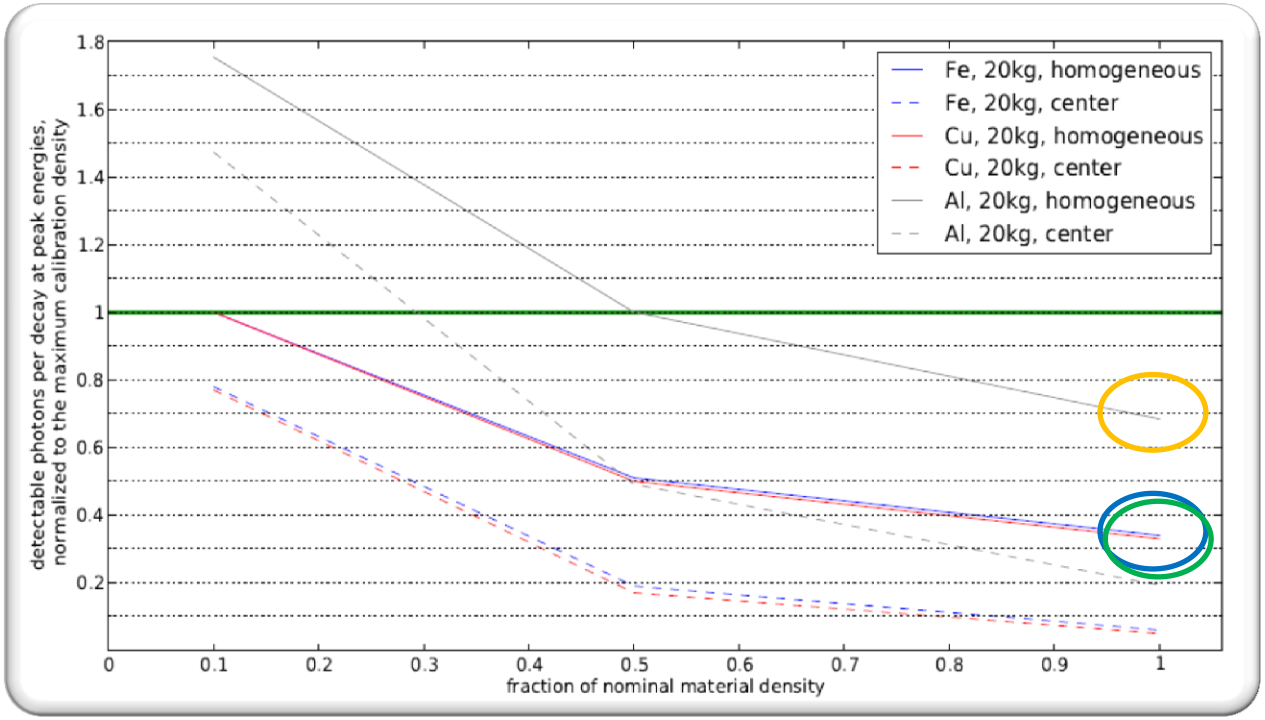
- The used calibration covers materials with densities up to 1 g/cm^3;
- Attenuation effects due to higher densities can be taken into account by applying safety factors;
- Safety factors for the material families Aluminium, Copper and Iron were determined via FLUKA* Monte Carlo simulations. The photon emission was simulated for spheres of the different materials and with varying apparent material densities of 0.1 ϱ_{mat}, 0.5 ϱ_{mat} and 1.0 ϱ_{mat};
- Homogeneous distribution of key radionuclide in sphere volume; (Co-60 in Copper and Iron, Na-22 in Aluminium)
- Values are normalized to maximum density of the present calibration, i.e. to 0.1ϱ_{mat} for Copper and Iron as well as to 0.5 ϱ_{mat} for Aluminium.

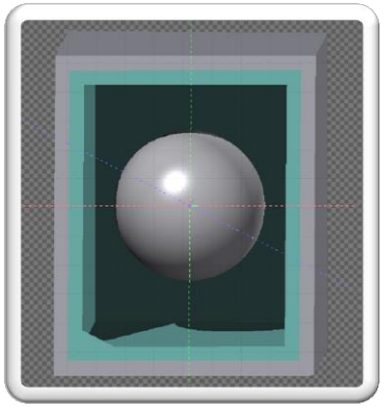
*1. A. FERRARI, P.R. SALA, A. FASSO, J. RANFT, "FLUKA: a multi-particle transport code," CERN 2005-10 (2005), INFN/TC_05/11, SLAC-R-773, 2005;
2. G. BATTISTONI, S. MURARO, P.R. SALA, F. CERUTTI, A. FERRARI, S. ROESLER, A. FASSO and J. RANFT, "The FLUKA code: Description and benchmarking," Proceedings of the Hadronic Shower Simulation Workshop 2006, Fermilab 6-8 September 2006, M. Albrow, R. Raja eds., AIP Conference Proceeding 896, 31-49, (2007)., 2007.

Evaluation of Geometry Effects

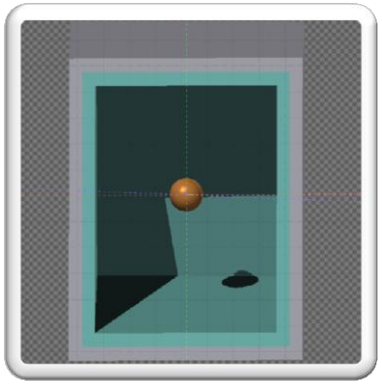
Self-absorption beyond Calibration Conditions 2/2

- Maximum Simulated Safety Factors
  0.69 for Al, 0.34 for Fe and 0.33 for Cu





Al sphere, 20 kg, $\rho = 0.1\ \rho_{Al}$



Cu sphere, 10 kg, $\rho = \rho_{Cu}$

Summary

- The developed characterization method is applicable and can be used to characterize material and waste from CERN accelerator facilities;
- The RADOS RTM 661/440 lnc device MDA is sufficiently low to robustly detect the resulting gamma signal per clearance limit of the considered *Common Materials* over the considered parameter range;
- The clearance of further materials has to be studied more in detail and the field of validity accordingly adapted;
- Geometry effects can be integrated by applying safety factors, which were determined for the most restrictive scenarios.

Further Developments

The method and its programmed scripts are highly modular and can be adapted to further changes or needs, as e.g.

- The new Swiss clearance limits that are announced for the near future
- Additional measurement devices by integrating the corresponding detector response curves
- More material compositions or particle spectra

Thank you for your attention!

Comments, remarks and ideas are welcome!

Contact details:

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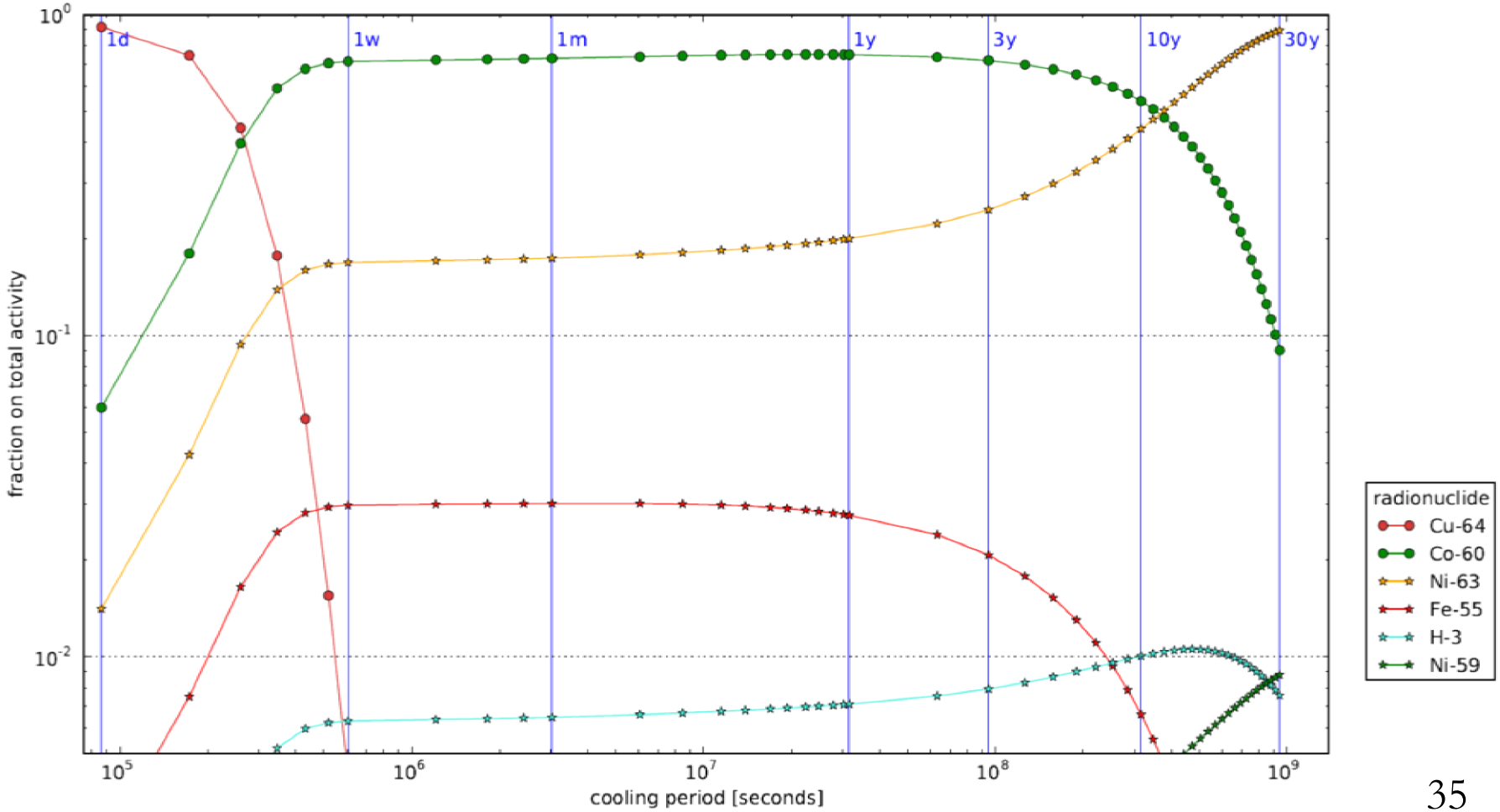
Back-up Information Slides

Radionuclide Inventory Visualization

Activity Time Evolution

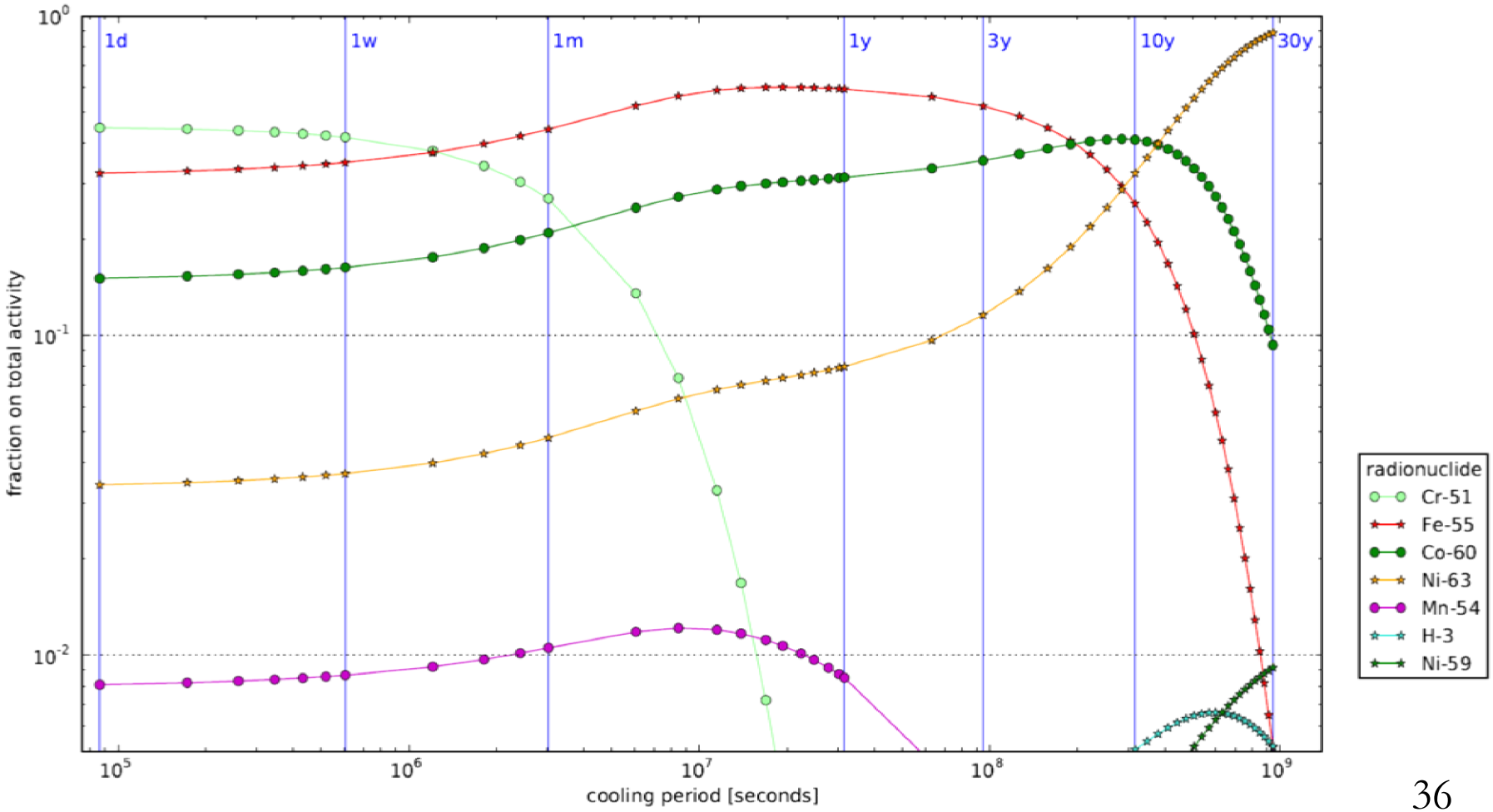
'Common Copper' after 30 years of irradiation at position #4 in the PSB



Radionuclide Inventory Visualization

Activity Time Evolution

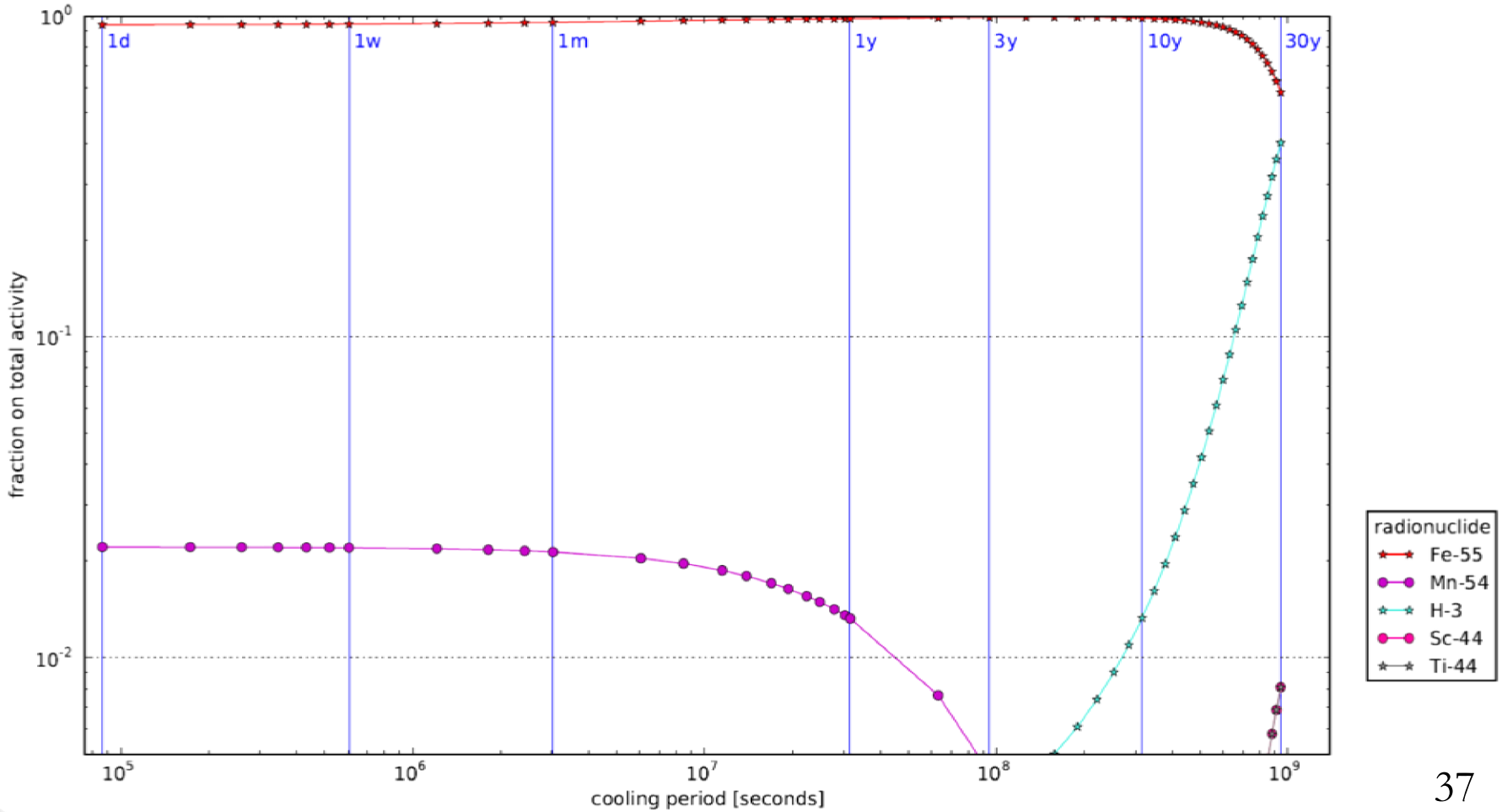
'Common Steel' after 30 years of irradiation at position #4 in the PSB



Radionuclide Inventory Visualization

Activity Time Evolution

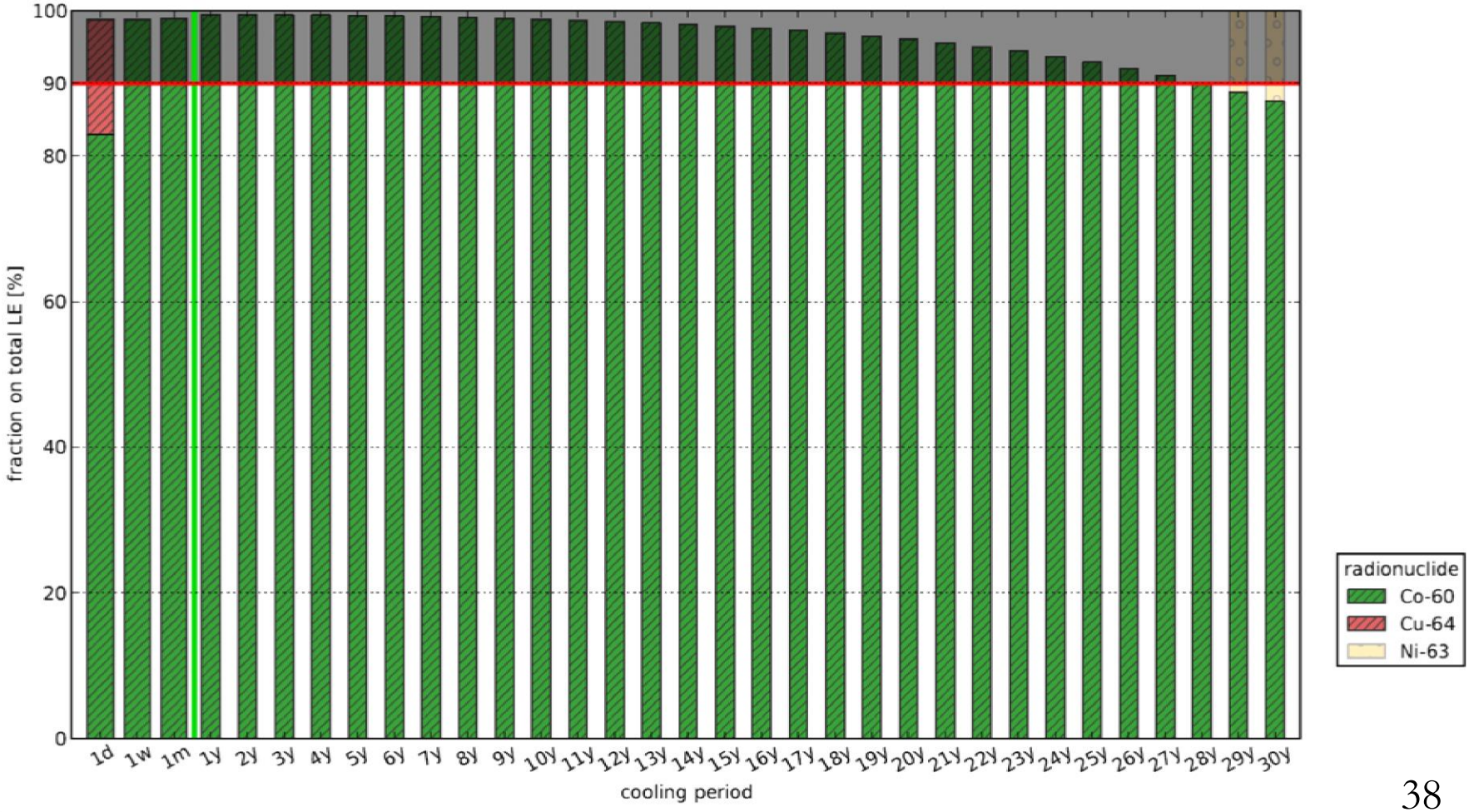
'Pure Iron' after 30 years of irradiation at position #4 in the PSB



Radionuclide Inventory Visualization

Stacked bar chart of importance on clearance limit

'Common Copper' after 30 years of irradiation at position #4 in the PSB



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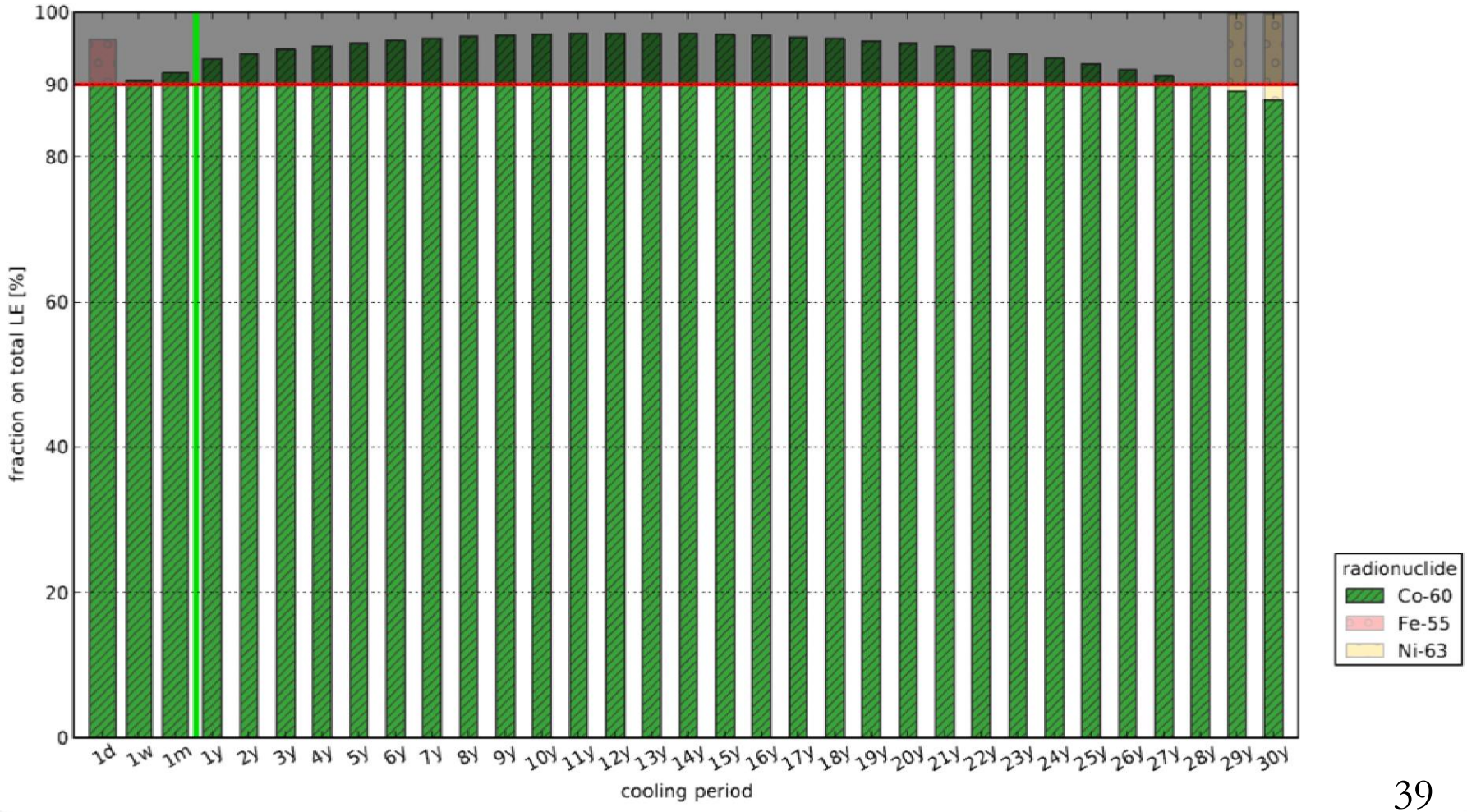
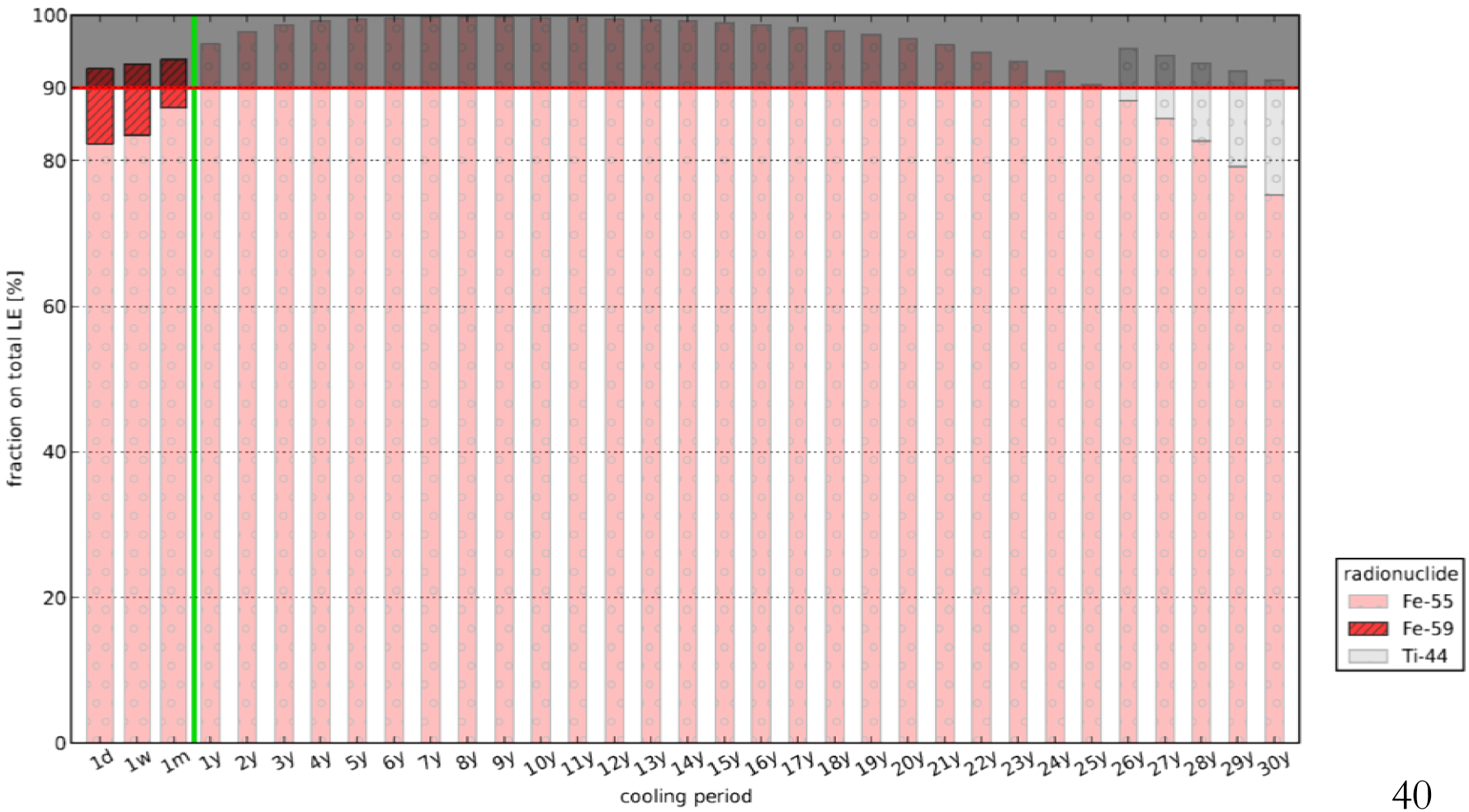


Radionuclide Inventory Visualization

Stacked bar chart of importance on clearance limit

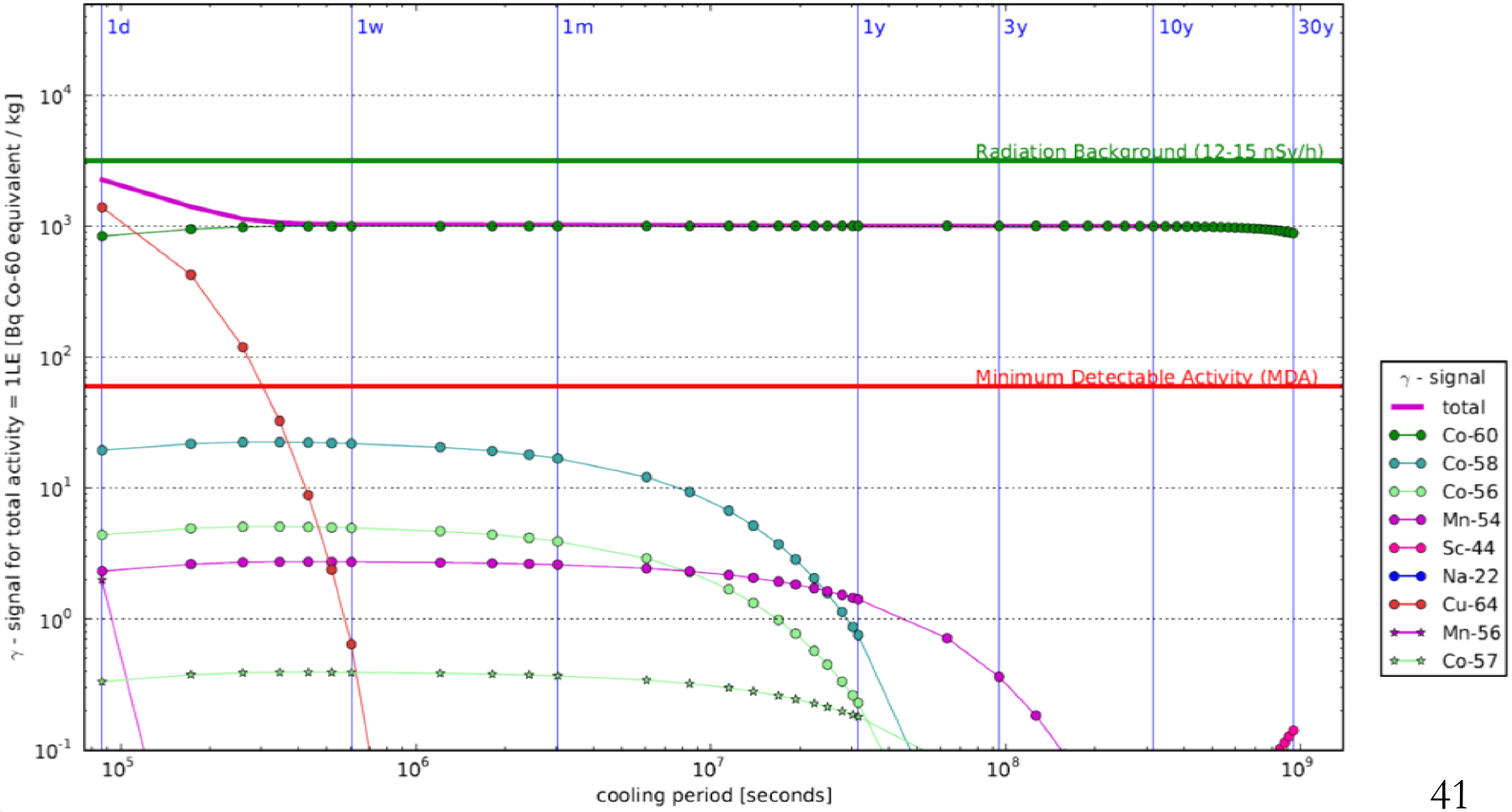
'Pure Iron' after 30 years of irradiation at position #4 in the PSB



Radionuclide Inventory Visualization

Resulting Gamma Signal per Clearance Limit

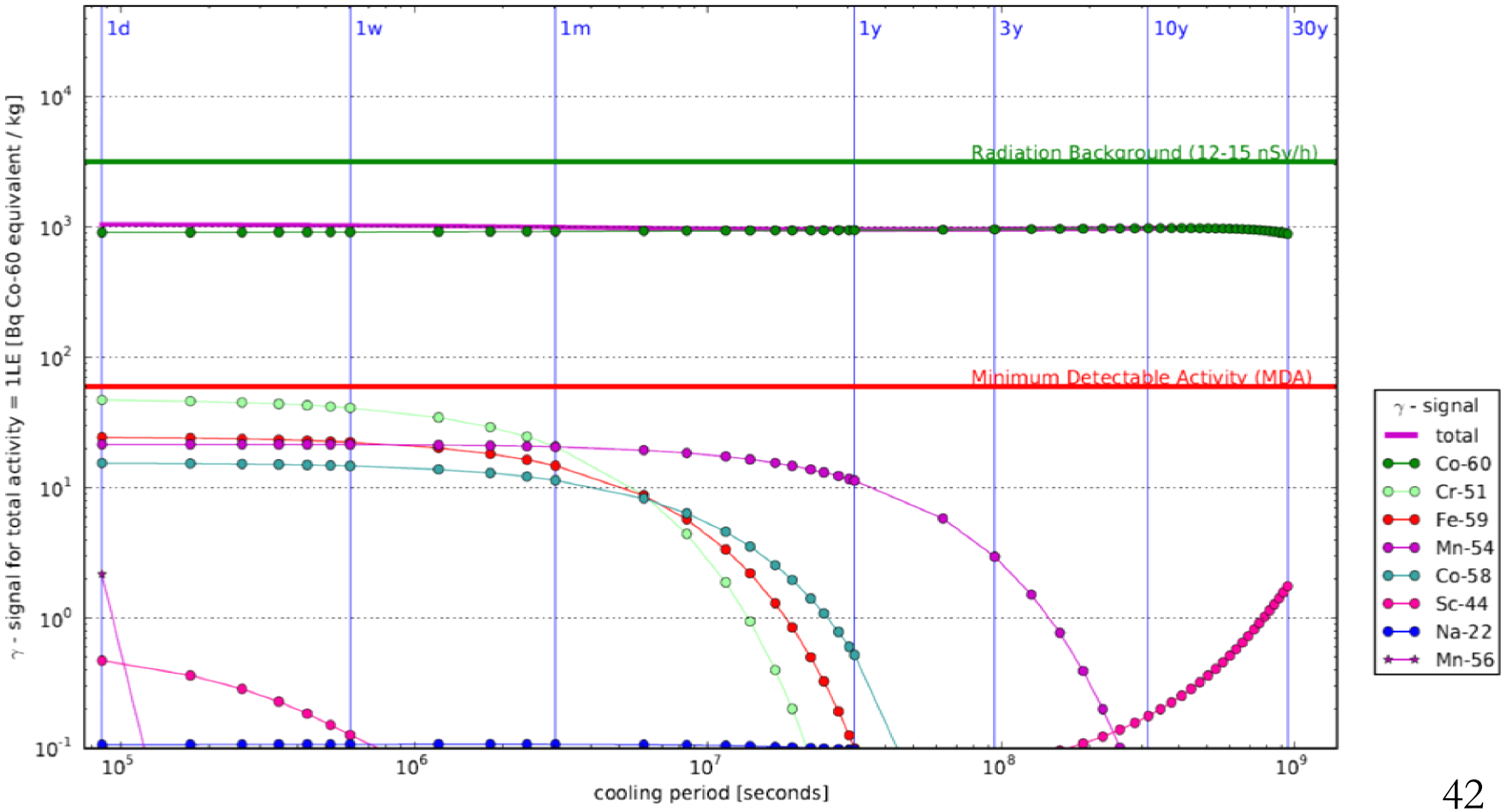
'Common Copper' after 30 years of irradiation at position #4 in the PSB



Radionuclide Inventory Visualization

Resulting Gamma Signal per Clearance Limit

'Common Steel' after 30 years of irradiation at position #4 in the PSB



Radionuclide Inventory Analysis

Minimum gamma signal per clearance limit – 'Common Materials'

'Common Steel'

Position	Minimum gamma-ray signal for LE =1 [Bq ^{60}Co *equivalent* kg^{-1}]					Minimum ↔
	60 days	1 year	3 years	10 years	30 years	
# 1	597 (10 y)	596 (9 y)	603 (8 y)	646 (7 y)	758 (6 y)	596
# 2	792 (8 y)	791 (8 y)	796 (7 y)	823 (6 y)	855 (6 y)	791
# 3	901 (8 y)	901 (7 y)	904 (7 y)	917 (6 y)	911 (30 y)	901
# 4	939 (3 y)	940 (3 y)	941 (2 y)	949 (30 y)	891 (30 y)	891
# 5	785 (9 y)	786 (8 y)	790 (8 y)	814 (7 y)	844 (6 y)	785
# 6	593 (9 y)	594 (9 y)	600 (8 y)	643 (7 y)	726 (6 y)	593
# 7	925 (7 y)	925 (7 y)	927 (6 y)	938 (5 y)	921 (30 y)	921
Minimum ↕	593	594	600	643	726	**593**‡

Radionuclide Inventory Analysis

Minimum gamma signal per clearance limit – 'Common Materials'

'Common Copper'

Position	Minimum gamma-ray signal for LE = 1 [Bq ^{60}Co *equivalent* kg^{-1}]					Minimum ↔
	60 days	1 year	3 years	10 years	30 years	
# 1	889 (30 y)	888 (30 y)	882 (30 y)	864 (30 y)	837 (30 y)	837
# 2	901 (30 y)	899 (30 y)	886 (30 y)	846 (30 y)	740 (30 y)	740
# 3	947 (30 y)	945 (30 y)	936 (30 y)	908 (30 y)	822 (30 y)	822
# 4	973 (30 y)	972 (30 y)	966 (30 y)	947 (30 y)	884 (30 y)	884
# 5	932 (30 y)	931 (30 y)	921 (30 y)	890 (30 y)	800 (30 y)	800
# 6	911 (30 y)	910 (30 y)	901 (30 y)	873 (30 y)	810 (30 y)	810
# 7	954 (30 y)	953 (30 y)	944 (30 y)	918 (30 y)	836 (30 y)	836
Minimum ↕	889	888	882	846	740	**740**‡