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Influence of neutron moderating materials in the characterization of 200 L radioactive waste drums by neutron activation analysis

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Introduction

Motivation: Non-destructive characterization of toxic (non-radioactive) elements and water pollutants in radioactive waste.

- Repository “Konrad”
- Declaration of toxic elements
- Reducing risks to human health and environment
- Product quality control
- MEDINA facility

Source: Hamburger Abendblatt

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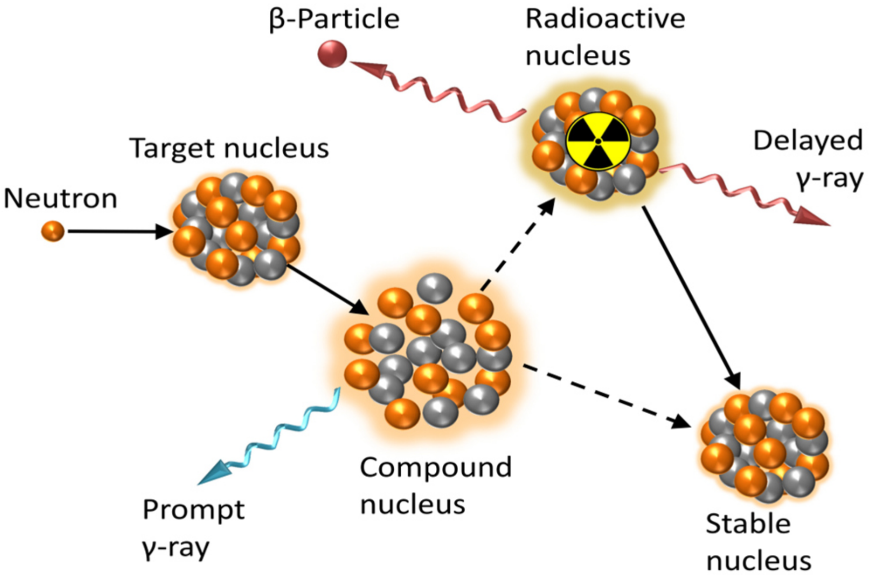
Introduction

Physical fundamentals

Prompt & Delayed-Gamma-Neutron activation analysis (P&DGNAA)



Basics

- *Non-destructive*
- *Multi-element analysis*
- *High sensitivity*

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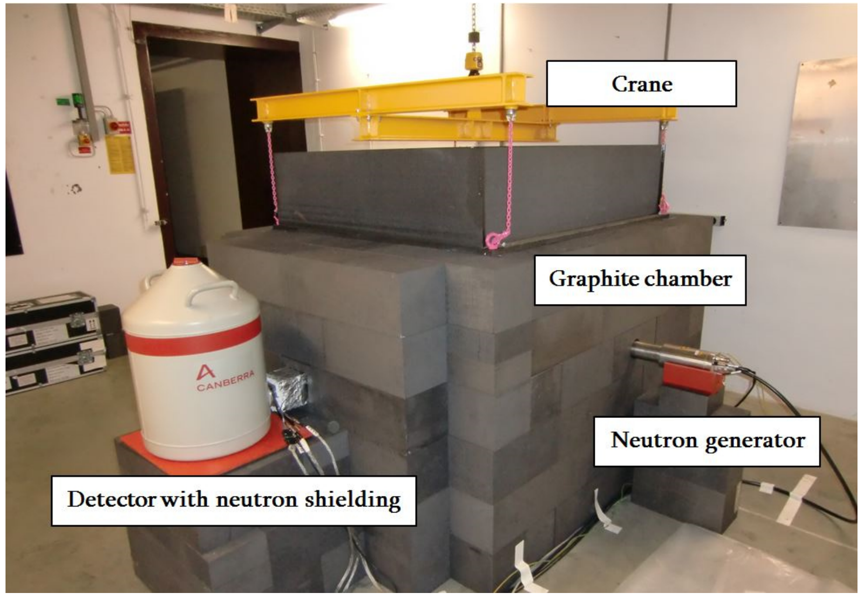
Multi-Element-Detection based on Instrumental Neutron Activation



Specification

- *14.1 MeV D-T Neutron generator*
- *HPGe Detector (rel. Eff. 100%) with a thermal neutron shielding*
- *Rotary table*

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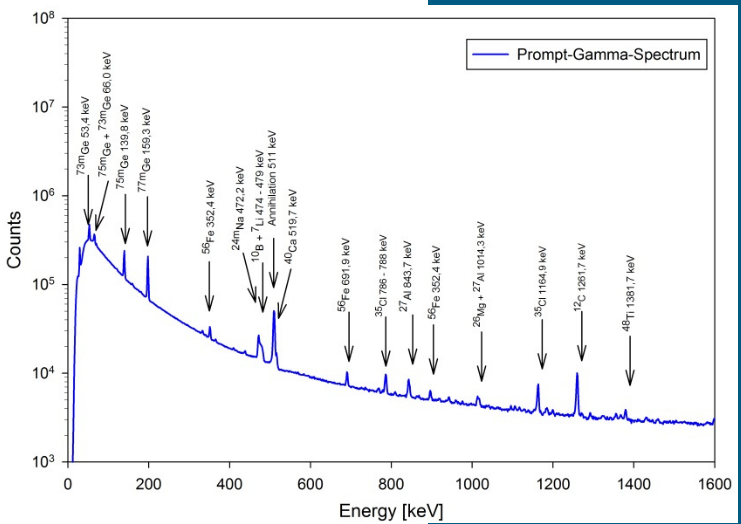


Prompt-Gamma-Spectrum
Counts
Energy [keV]
10^3
10^4
10^5
10^6
10^7
10^8
0
200
400
600
800
1000
1200
1400
1600
^{73m}Ge 53,4 keV
^{75m}Ge + ^{73m}Ge 66,0 keV
^{75m}Ge 139,8 keV
^{77m}Ge 159,3 keV
^{56}Fe 352,4 keV
^{24m}Na 472,2 keV
^{10}B + ^{7}Li 474 - 479 keV
Annihilation 511 keV
^{40}Ca 519,7 keV
^{56}Fe 691,9 keV
^{35}Cl 786 - 788 keV
^{27}Al 843,7 keV
^{56}Fe 352,4 keV
^{26}Mg + ^{27}Al 1014,3 keV
^{35}Cl 1164,9 keV
^{12}C 1261,7 keV
^{48}Ti 1381,7 keV

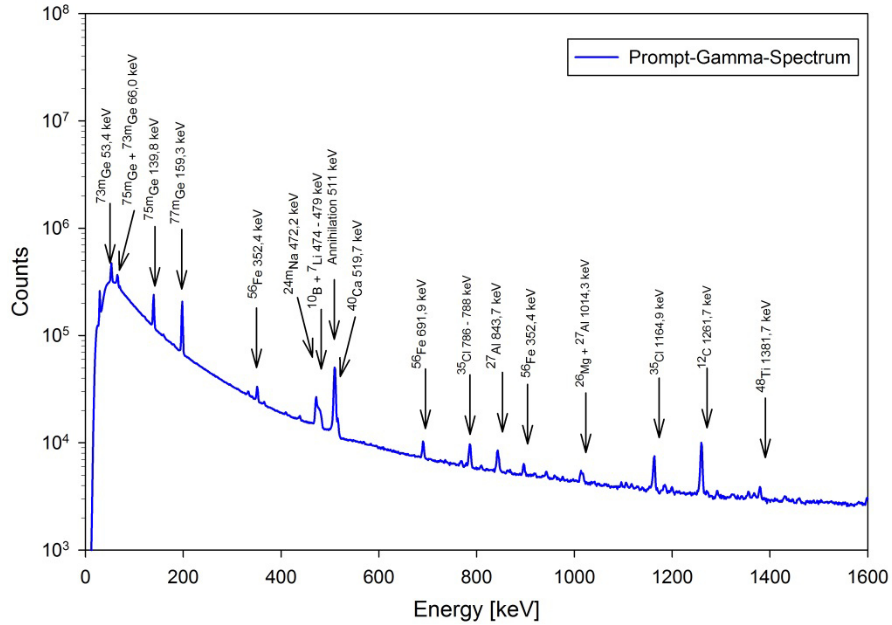
Method of quantification

Basic equation

Based on the results of the analyzed PGNAA spectra, the amount of elements can be quantified.

$$m = \frac{P(E_\gamma) \cdot M}{N_A \cdot \varepsilon(E_\gamma) \cdot \Phi_{th} \cdot \left[\sigma(E_\gamma) + \frac{0{,}44 \cdot \sigma(E_\gamma) + I(E\gamma)}{R}\right] \cdot f_t}$$

m	Element mass
P	Photopeak-Area
M	Molar Mass
N_A	Avogadro Constant
$\varepsilon(E_\gamma)$	Photopeak-efficiency
Φ_{th}	Thermal neutron flux
R	Ratio of thermal and epithermal neutrons
$\sigma(E_\gamma)$	Partial gamma-ray production cross section for thermal neutrons
$I_\gamma(E_\gamma)$	Partial gamma-ray production cross section for epithermal neutrons
f_t	Time parameter



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Method of quantification (mixed waste)

Process overview of the quantification method

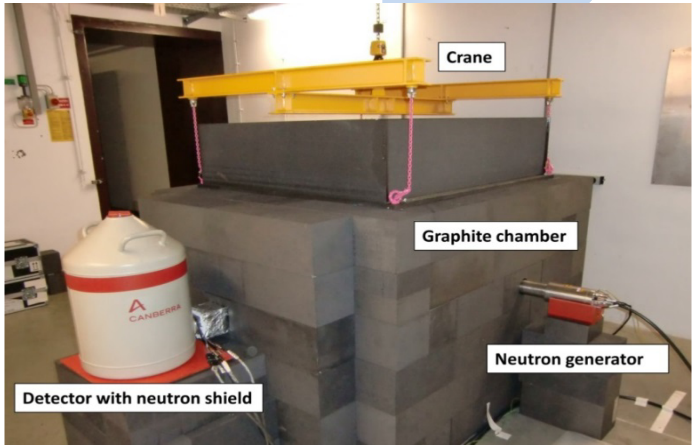
A priori
Drum Dimension
Mass
Density

Assumption
Concrete matrix
Homogeneous distribution

Efficiency calculation
Numerical Simulation

Thermal neutron flux
Patented & standardized method

Quanti-fication
Results



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Method of quantification (mixed waste)

- For the photopeak-efficiency calculation we assume that the waste matrix is concrete.
- Quantification needs the thermal neutron flux in the matrix, i.e. in the steel drum.
- The gamma-ray-lines of iron of the steel drum are used for the determination of the average thermal neutron flux [1,2].
- Determination of the element concentration in the matrix.

1. • A priori information
2. • Assumption
3. • Numerical calculation of the photopeak-efficiency
4. • Determination of the thermal neutron flux at the drum
5. • Quantification and determination of the elements in the matrix

[1] *Patent* EP 2596341 A1: *Neutron activation analysis using a standardized sample container for determining the neutron flux.* Forschungszentrum Jülich (2011)
[2] International Patent Application WO 2012/010162 A1; Australian Patent AU2011282018

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Samples

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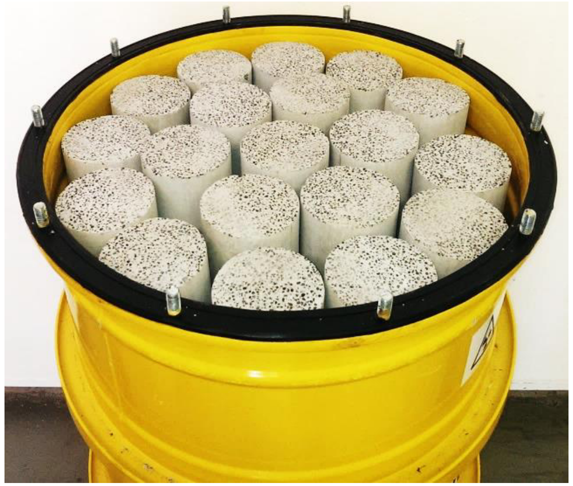
Study: 10 **concentrically** and **axial** mixed waste configurations.



Specification: Drum with Concrete

m_{Drum}	52.3 kg
$m_{Concrete}$	195.5 kg
$\rho_{Concrete}$	1.62 g cm^{-3}
$\rho_{Filling}$	1.02 g cm^{-3}

Specification: e.g. mixed configuration

m_{Drum}	52.3 kg
$m_{Concrete}$	133.1 kg
m_{PE}	38.2 kg
$\rho_{Filling}$	0.87 g cm^{-3}

Specification: Drum with PE

m_{Drum}	52.3 kg
m_{PE}	120.8 kg
ρ_{PE}	0.94 g cm^{-3}
$\rho_{Filling}$	0.61 g cm^{-3}

Measurements

Measurement parameters

- $n_{Emission} = 8 \cdot 10^7 \mathrm{n\ s}^{-1}$
- $t_{Pulse} = 60\ \mu s$
- $T_{Repetition} = 1\ ms$
- $t_{Gate} = 940\ \mu s\ (Acquire)$
- $t_{Livetime} = 3600\ s$
- $\Lambda_{Cell} \approx 3\ ms$ (Thermal neutron Die – Away Time [3])

→ Almost constant thermal neutron flux

Measuring the prompt-gamma-ray count rate of

- H-1 $(E_\gamma = 2223\ keV)$
- B-10 $(E_\gamma = 477\ keV)$
- Si-28 $(E_\gamma = 3539\ keV)$
- K-39 $(E_\gamma = 770\ keV)$
- Ca-40 $(E_\gamma = 1943\ keV)$
- Fe-56 $(E_\gamma = 691\ keV)$

→ Determining the element concentration.

[3] F. Mildenberger, E. Mauerhofer (2015) *Thermal neutron die-away times in large samples irradiated with a pulsed 14 MeV neutron source. Nucl. Chem, J Radioanal. doi: 10.1007/s10967-015-4178-2*

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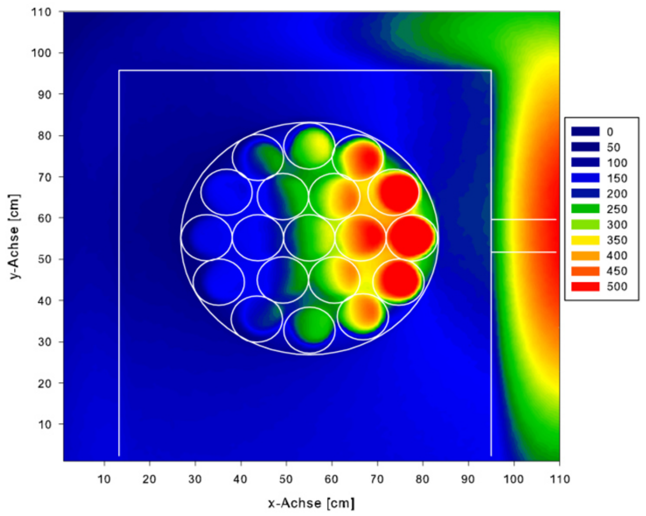

Result – Neutron flux

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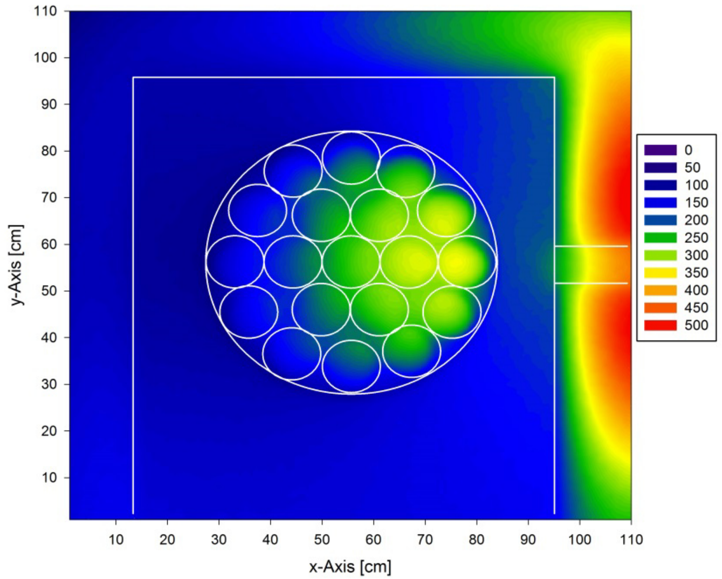
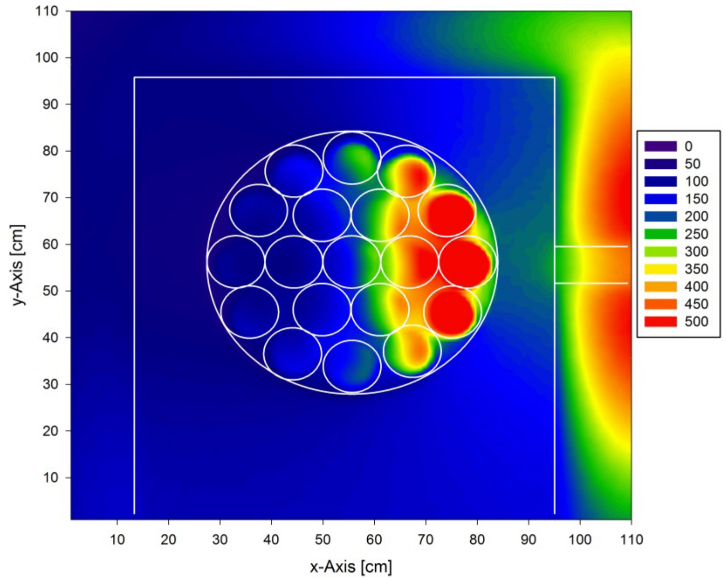
Thermal neutron flux distribution (MCNP5) Range: 10-100 meV



Drum filled with concrete.

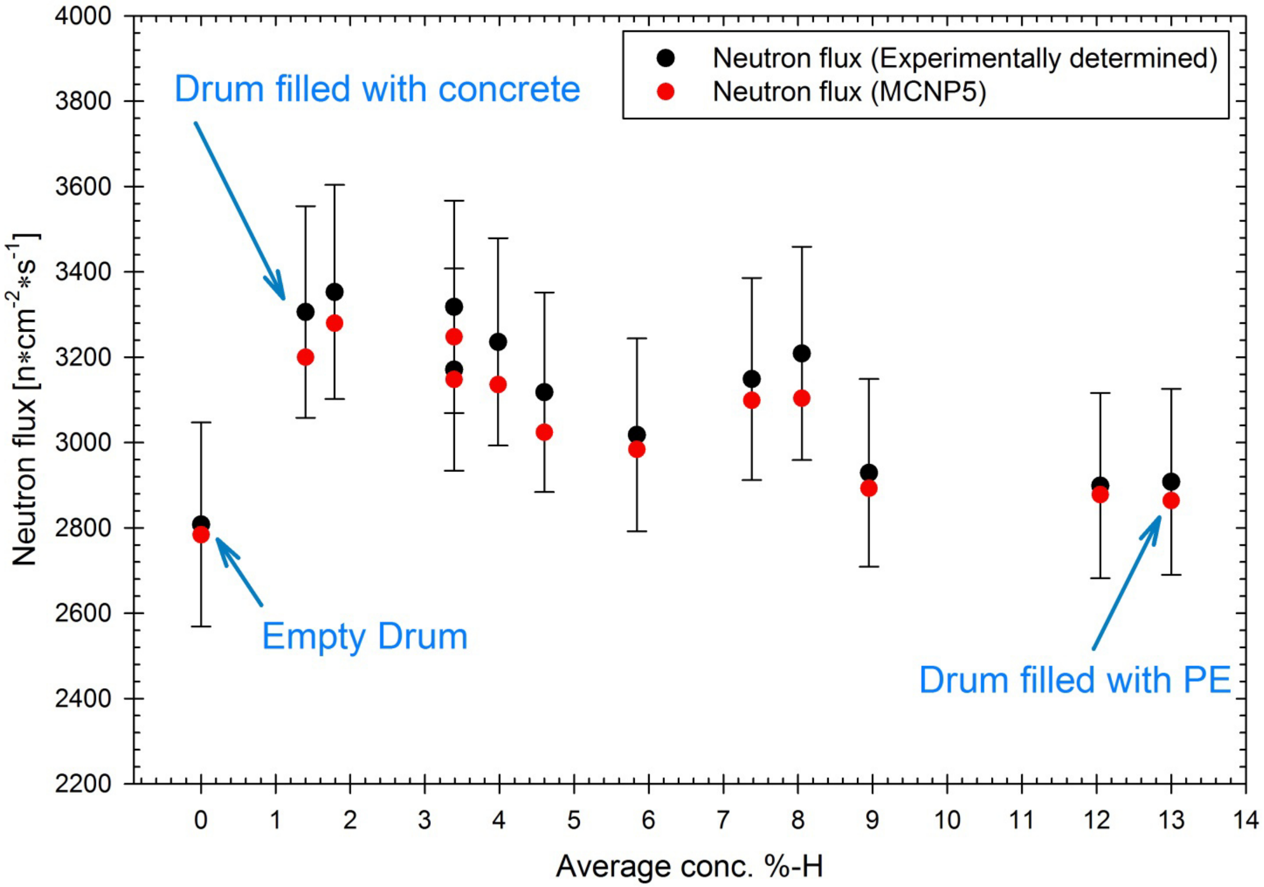


Drum filled with PE.

Sample	Φ_{th} ***steel drum (Experiment) [n cm^{-2} s^{-1}]***	Φ_{th} ***steel drum (MCNP5) [n cm^{-2} s^{-1}]***
Empty Drum	2808 ± 239	2798 ± 1
Drum filled with Concrete	3306 ± 248	3260 ± 1
Drum filled with PE	2908 ± 218	2888 ± 1

Result – Neutron flux

- Comparison between the experimental thermal neutron flux determination and the simulated (MCNP5) thermal neutron flux at the steeldrum.
- Results agree good in their uncertainties.



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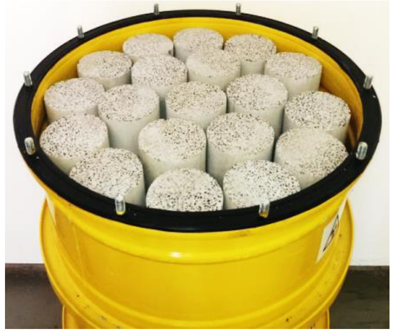
Result – Quantification

Drum with Concrete

Element	m_{Ref} [g]	m_{Det} [g]	Deviation
H	2,793 ± 22	2,813 ± 210	+ 0.7 %
B	18.5 ± 0.1	21.5 ± 1.8	+ 16 %
Si	34,713 ± 595	36,621 ± 6,480	+0.5 %
K	2,793 ± 44	2,777 ± 208	- 0.5 %
Ca	46,483 ± 615	47,370 ± 6,366	+ 1.9 %



Drum with Concrete & PE

Element	m_{Ref} [g]	m_{Det} [g]	Deviation
H	6,828 ± 546	5,240 ± 393	- 23.2 %
B	12.65 ± 0,09	14.1 ± 1.1	+ 11.3 %
Si	23,163 ± 405	29,158 ± 2,187	+ 25.9 %
K	1,864 ± 30	2,074 ± 156	+ 11.3 %
Ca	31,017 ± 419	30,254 ± 2,269	- 2.5 %

Drum with PE

Element	m_{Ref} [g]	m_{Det} [g]	Deviation
H	15,709 ± 150	15,571 ± 1,138	- 0.9 %

Result – Quantification

Accuracy of the model

For the accuracy of all concentrically and axially measurements a χ^2-test was made.

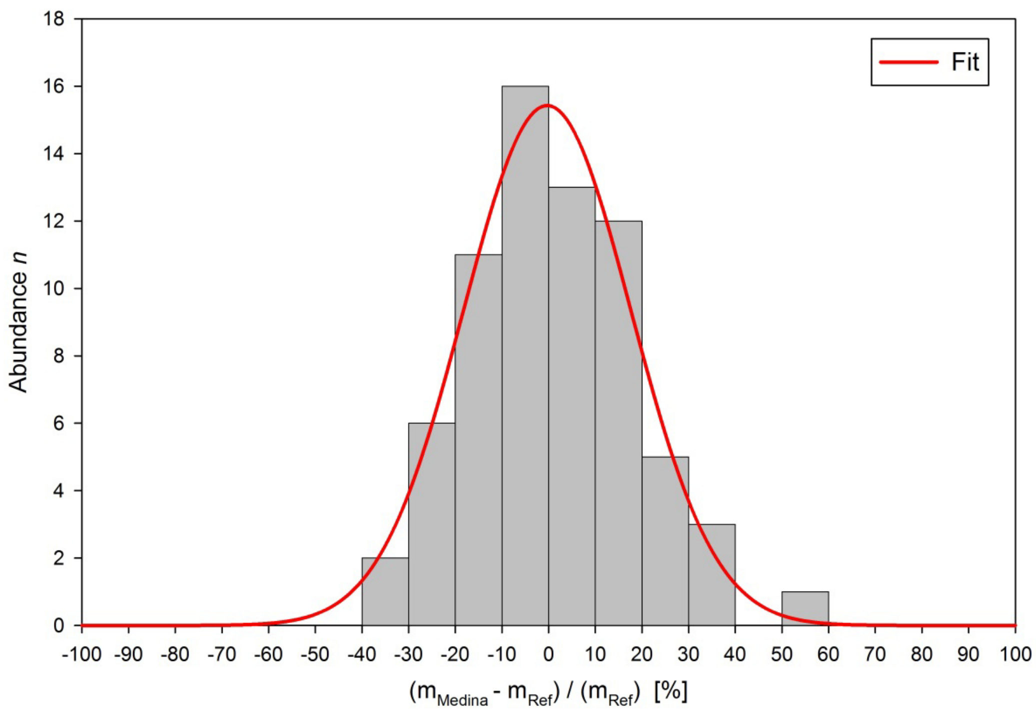
$$\chi^2 = \sum_{j=1}^{m} \frac{(O_j - E_j)^2}{{\sigma_{O,j}}^2 + {\sigma_{E,j}}^2}$$

Isotop	χ^2
H-1	1.60
B-10	3.05
Si-28	0.68
K-39	0.93
Ca-40	1.10
Ti-48	1.26
Average	**1.44**

Deviation of the model

The conservative deviation is between

-40 % and +40 %.



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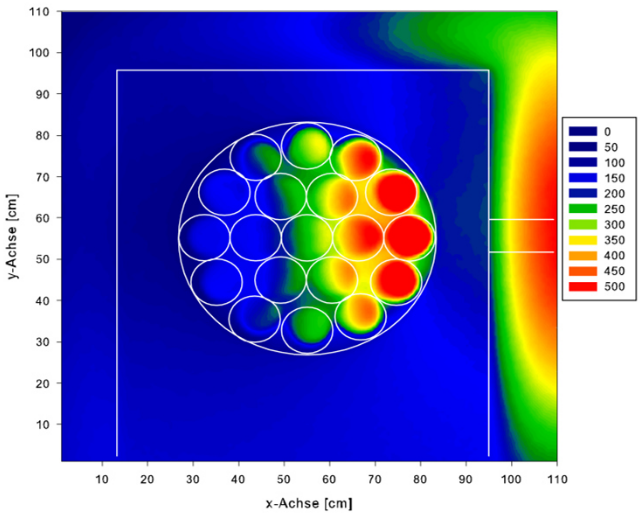

110
100
90
80
70
60
50
40
30
20
10
y-Achse [cm]
10 20 30 40 50 60 70 80 90 100 110
x-Achse [cm]
0
50
100
150
200
250
300
350
400
450
500

Conclusion/Outlook

Conclusion

- "No" *a priori* information needed.
- Assumption is that the waste matrix is homogeneous concrete.
- Results with a conservative uncertainty of ± 40 %.
- Experimental and simulated results are in a good agreement.
- The quantification model provides reasonable results for mixed waste configurations with high moderating materials.

Outlook

- Measurements of <u>local concentrated</u> toxic elements like Cadmium in mixed waste samples (concrete and PE).
- Improvement of the quantification method.

Any questions?

Backup

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References

[1] *Patent* EP 2596341 A1*: Neutron activation analysis using a standardized sample container for determining the neutron flux*. Forschungszentrum Jülich (2011)

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