SAMPLING AND SAMPLE PREPARATION OF NORM RESIDUES

taking into account

Evaluation of Measurement Results by Resampling Methods

European ALARA Network for NORM
4th workshop

Nov. 29th - Dec. 1st 2011, Hasselt (Belgium)

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Definition of the Project

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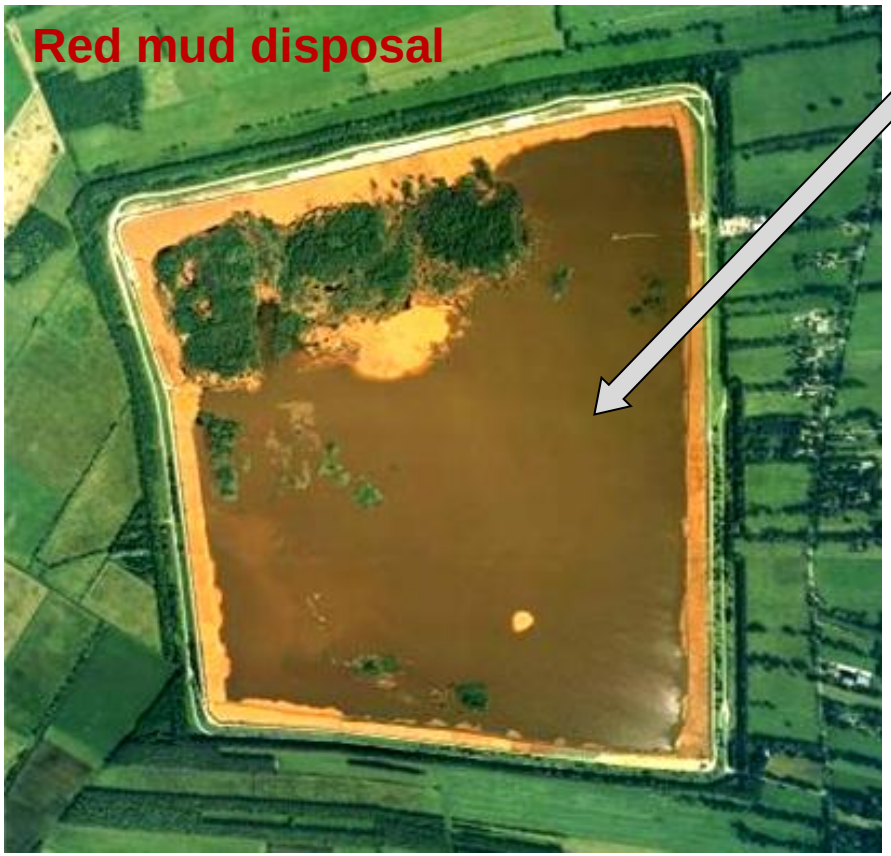
NORM residues



Radiological Assessment

< 1 mSv/a ?
(Reference value)

< 1 Bq/g ?
(Control limit)

Sampling

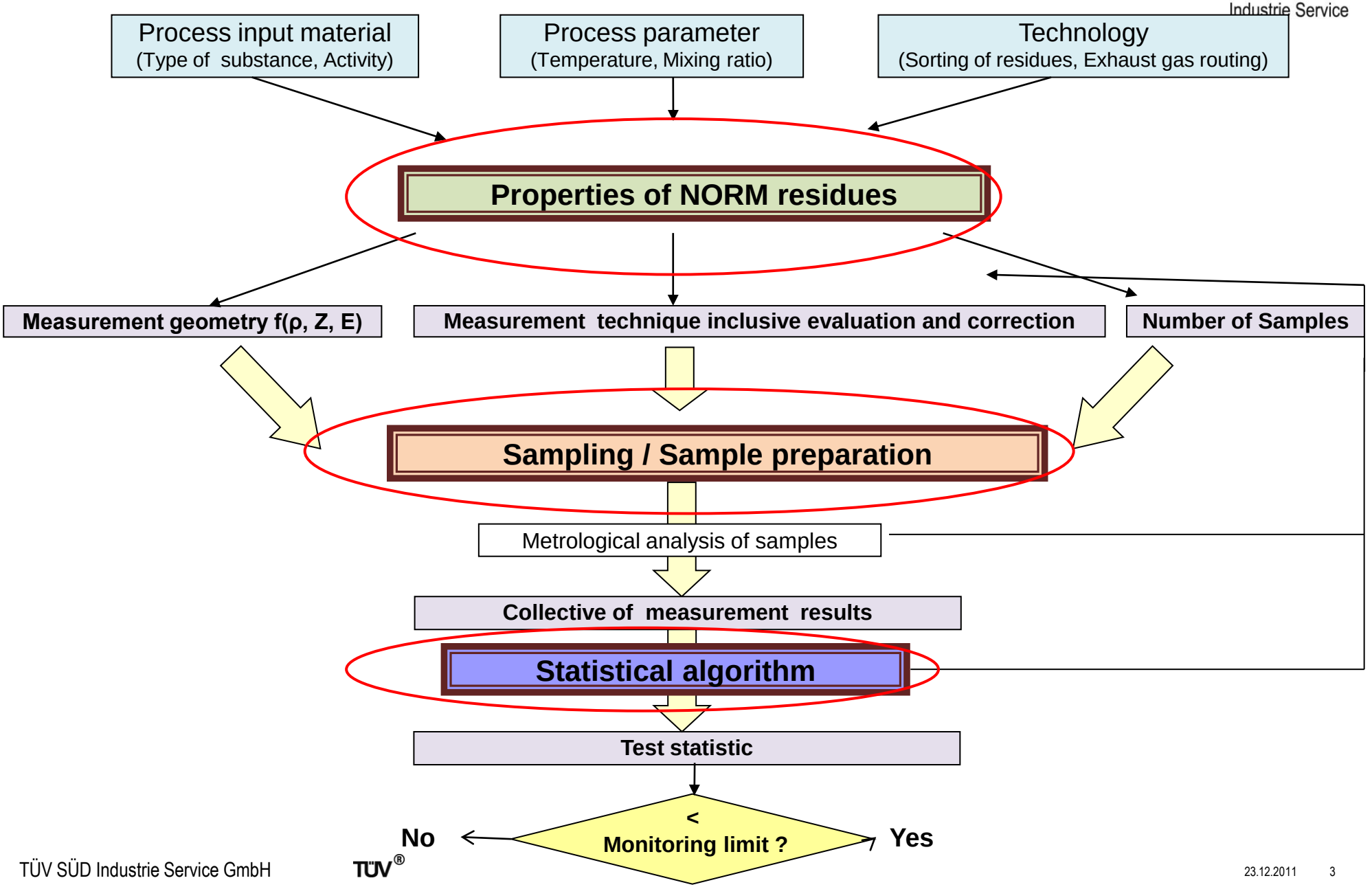
Sample preparation

Analysis of samples

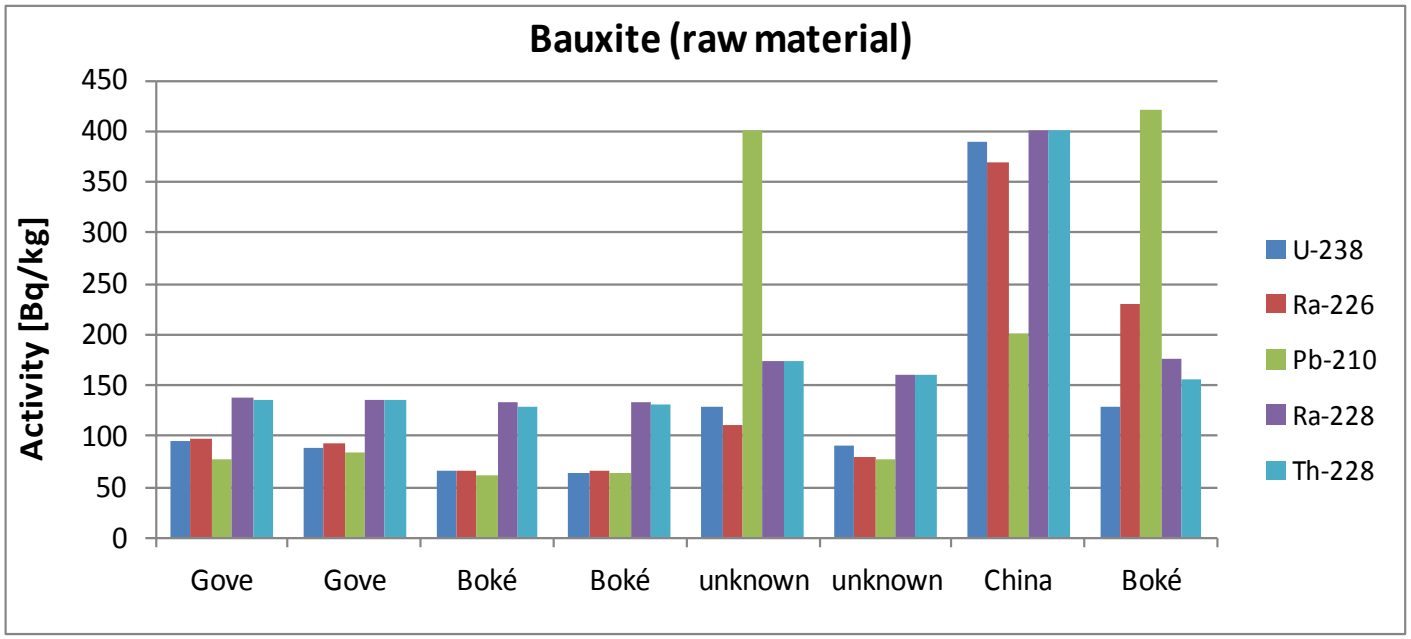
Statistical algorithm

Project by order of the Federal Office for Radiation Protection

Fundamental Relationships

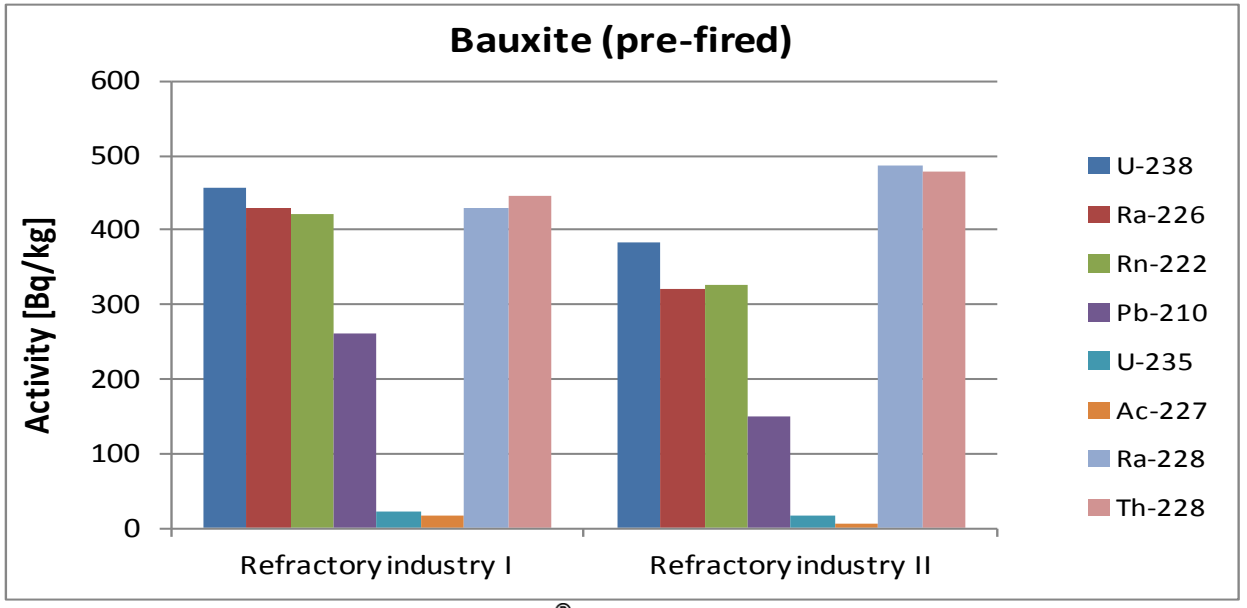


Properties: Nuclide Vector



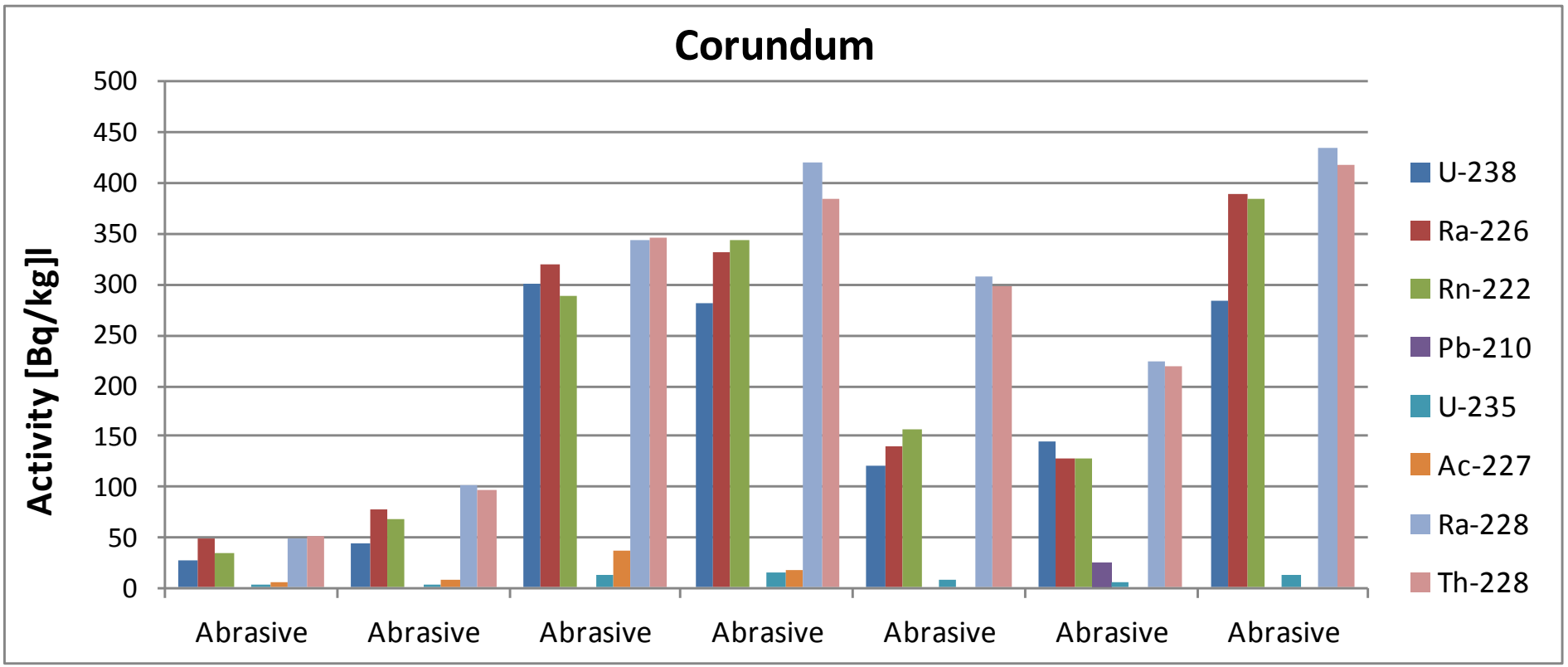
Reference:
Gellermann, R. (HGN), Schulz, H. (IAF), Weiß, D. (GRS), Schellenberger, A. (IAF), Müller, A. (HGN), Methodische Weiterentwicklung des Leitfadens zur radiologischen Untersuchung und Bewertung bergbaulicher Altlasten und Erweiterung des Anwendungsbereichs, Teil B; BMU – 2007-697, 2006

Leopold, K., Chemical types of bounding of natural radionuclides in TENORM, Inaugural thesis submitted in fulfilment of the requirements for the Degree of Doctor of Natural Sciences, Department of Biology and Geography of the University Duisburg-Essen, June 2007



Reference:
Own measurements

Properties: Nuclide Vector



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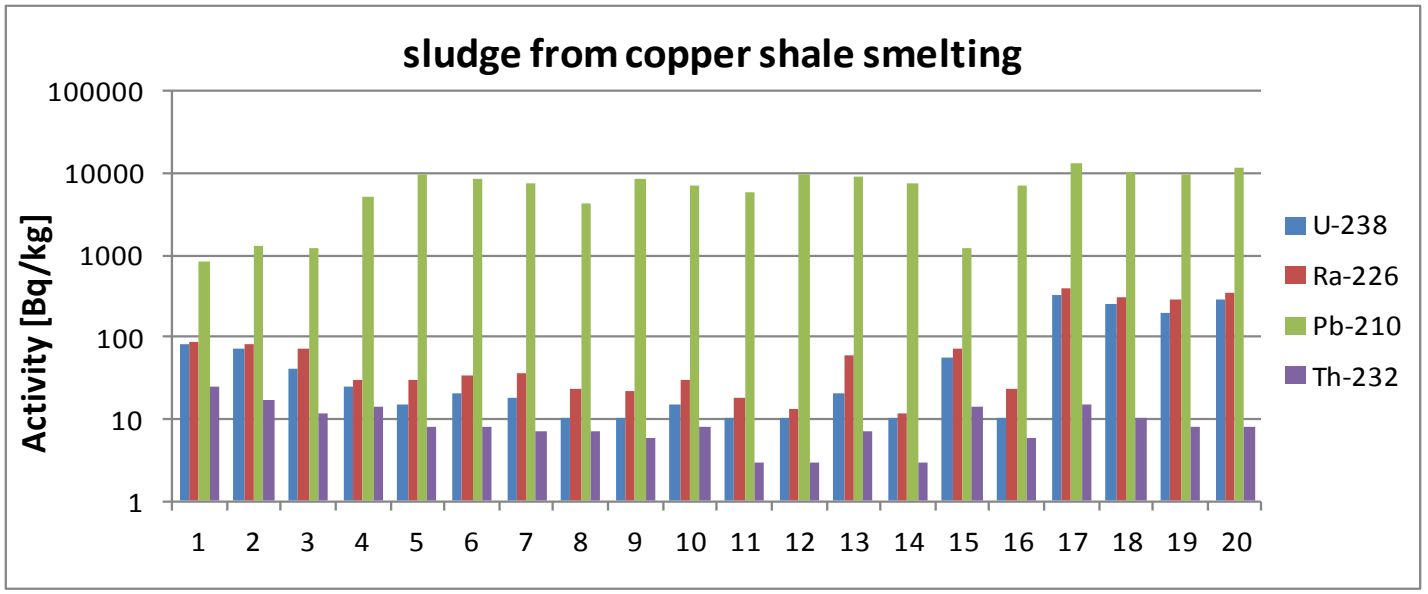

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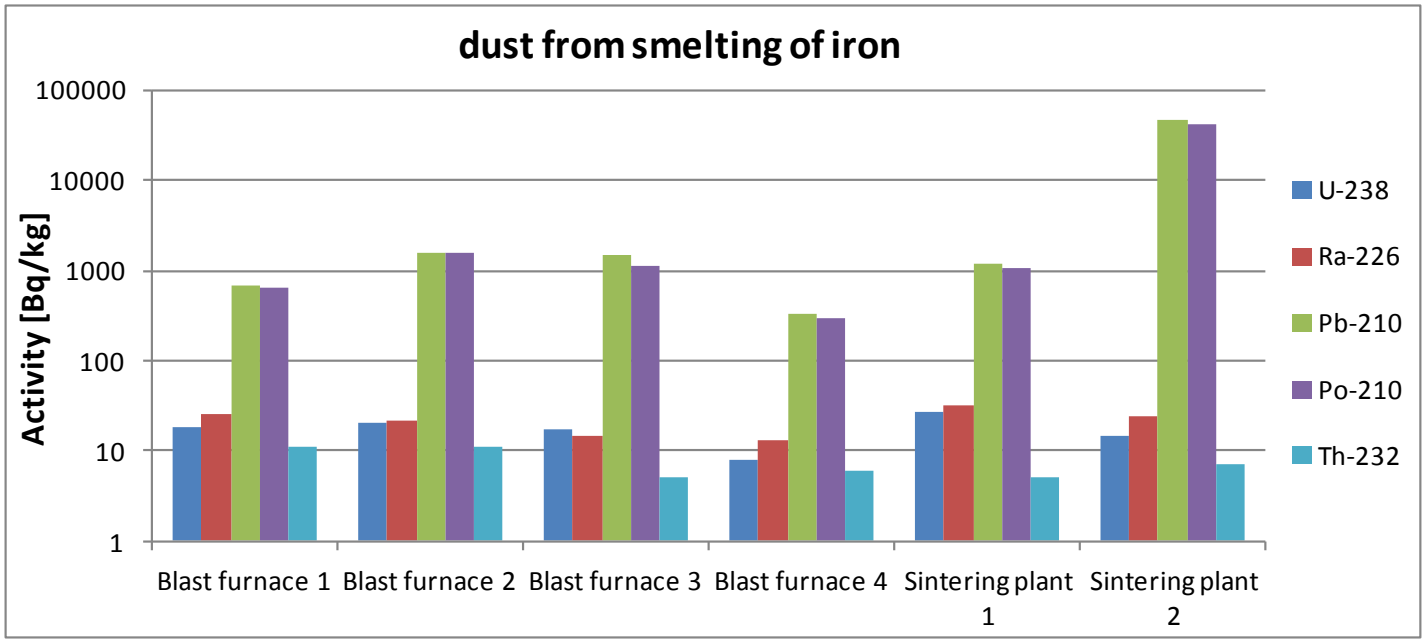
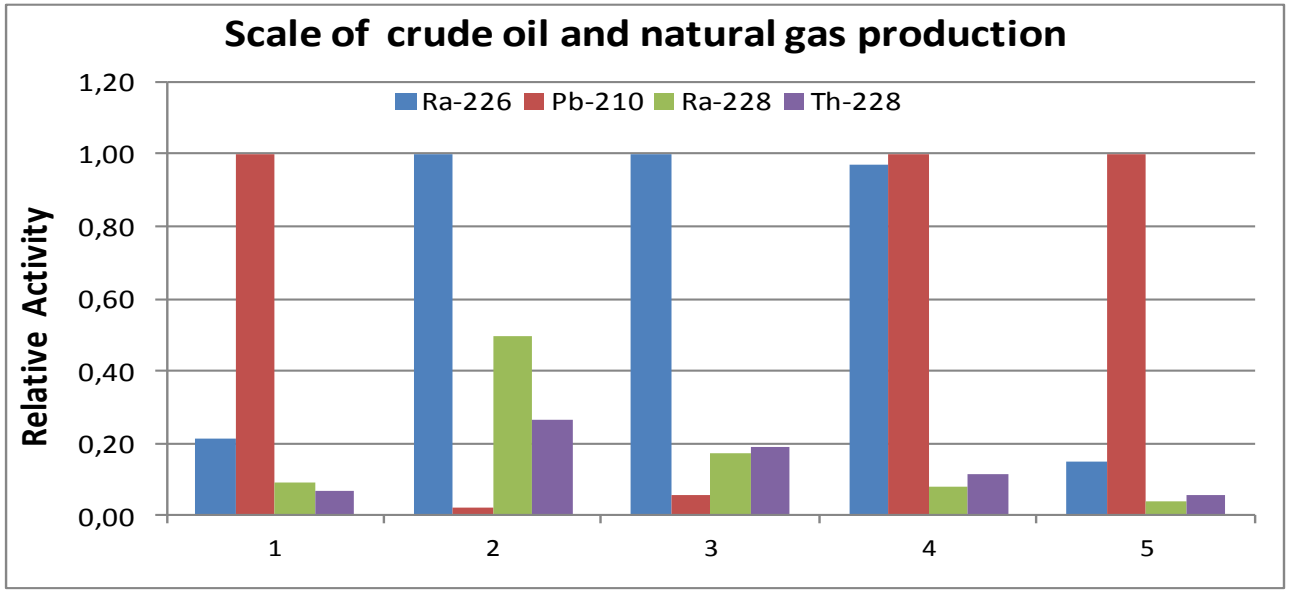
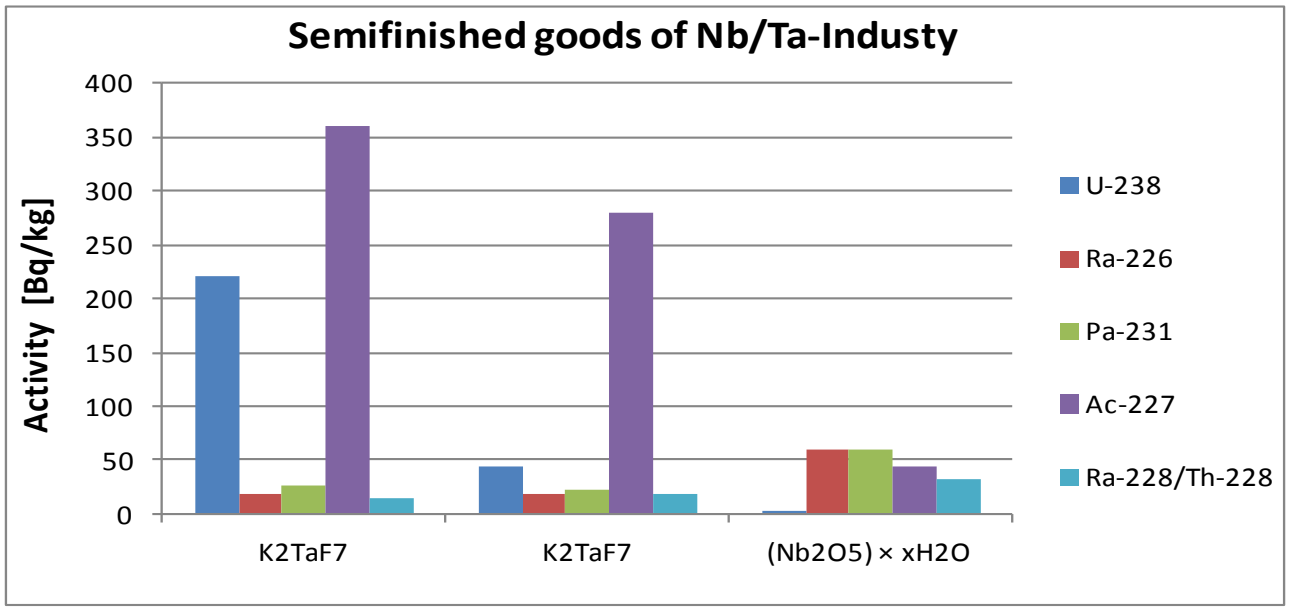


Reference:
Trotti, F., Zampieri, C., Caldognetto, E., Ocone, R., Di Lullo, A., Margo, L., Jia, G., Torri, G., A Study Concerning NORM in Integrated Steelworks; Proceedings of the fifth international symposium on Naturally Occurring Radioactive Material (NORM V), Seville, Spain, 19–22 March 2007, International Atomic Energy Agency, Vienna, 2008

Properties: Nuclide Vector



Reference:
Message of Dr. E.-M. Steffan, ExxonMobil Production Deutschland GmbH, 06.03.2009



Reference:
Stepanov, S.V., Simakov, A.V., Petrov, S.V., Abramov, Ju.V., Isaev O.V., Methodological Problems of the Personnel Protection Supply in Case of Operations with Natural Radionuclide Materials, Proceedings of IRPA 10, 10th International Congress of the International Radiation Protection Association, "Harmonization of Radiation, Human Life and the Ecosystem", International Conference Center Hiroshima, Hiroshima, Japan, May 14-19, 2000

Properties: Temporal Variability

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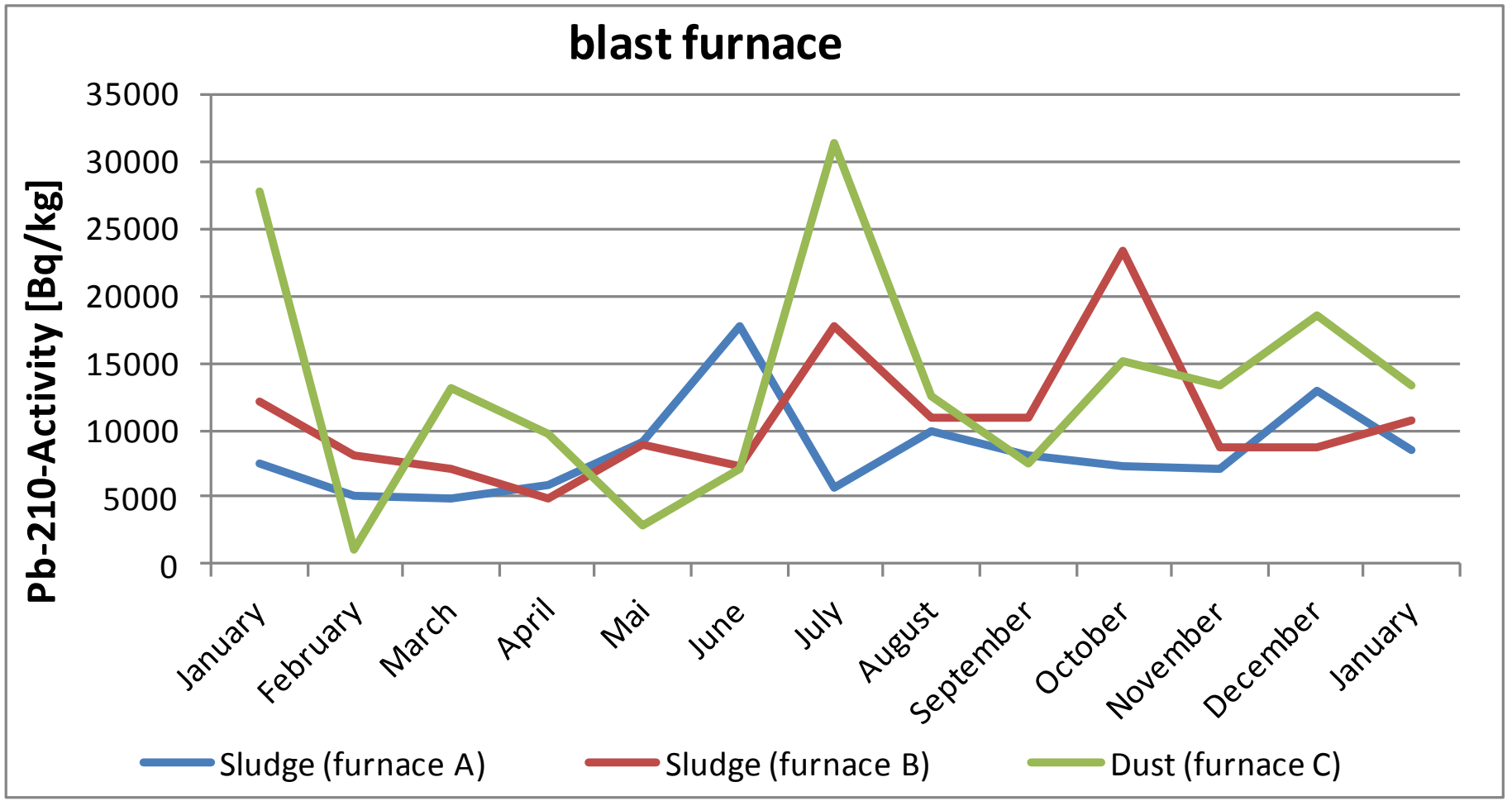

Reference:
Wieters, C.U. Experience and expectation of NORM in the German hot metal production, ALARA Workshop, European ALARA Network for Naturally Occurring Radioactive Materials - NORM in Dresden, Nov. 20th - 22nd 2007

Properties: Homogeneity

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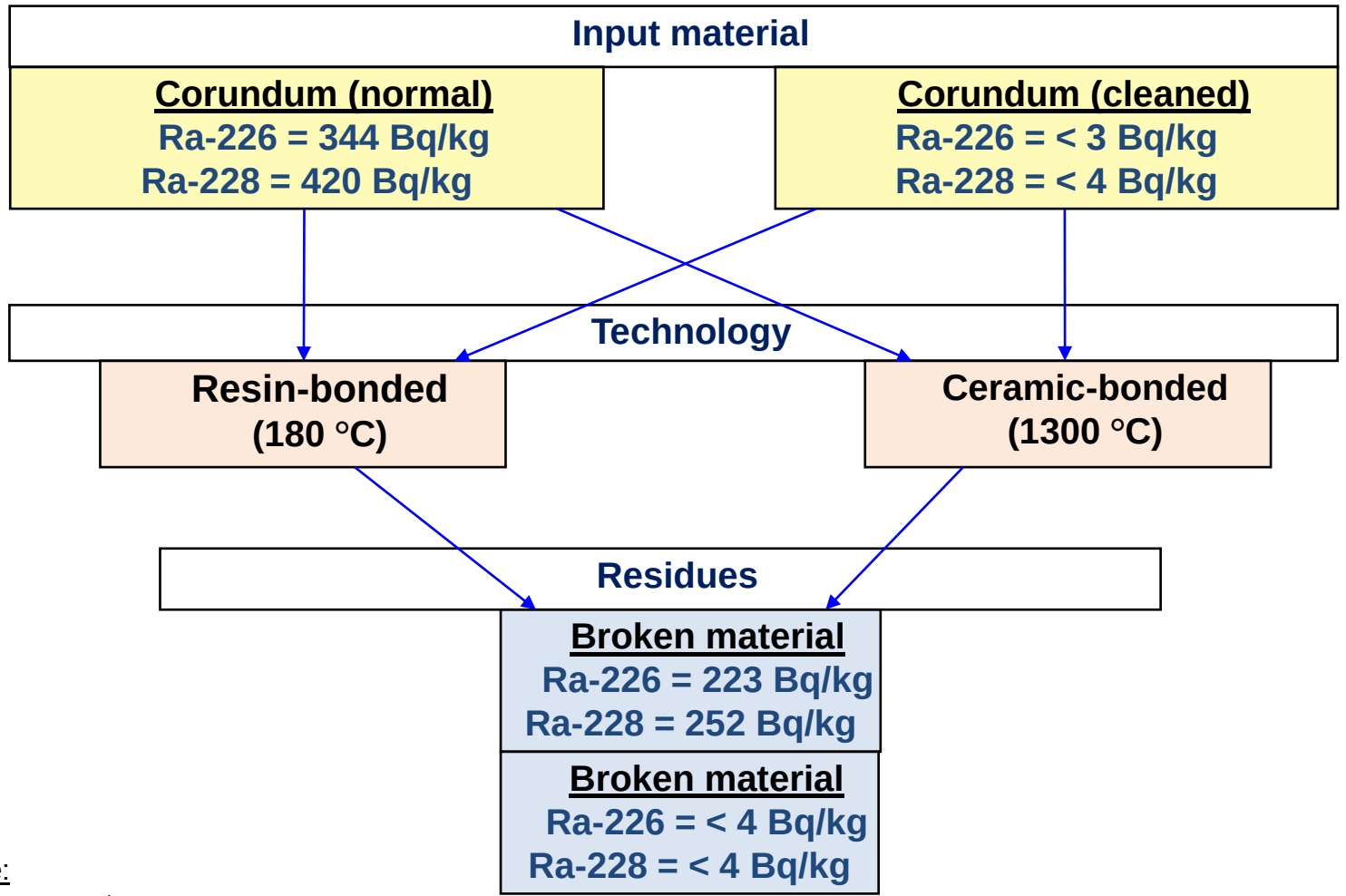
Example: Manufacture of corundum abrasives



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Own measurements

Properties: Homogeneity

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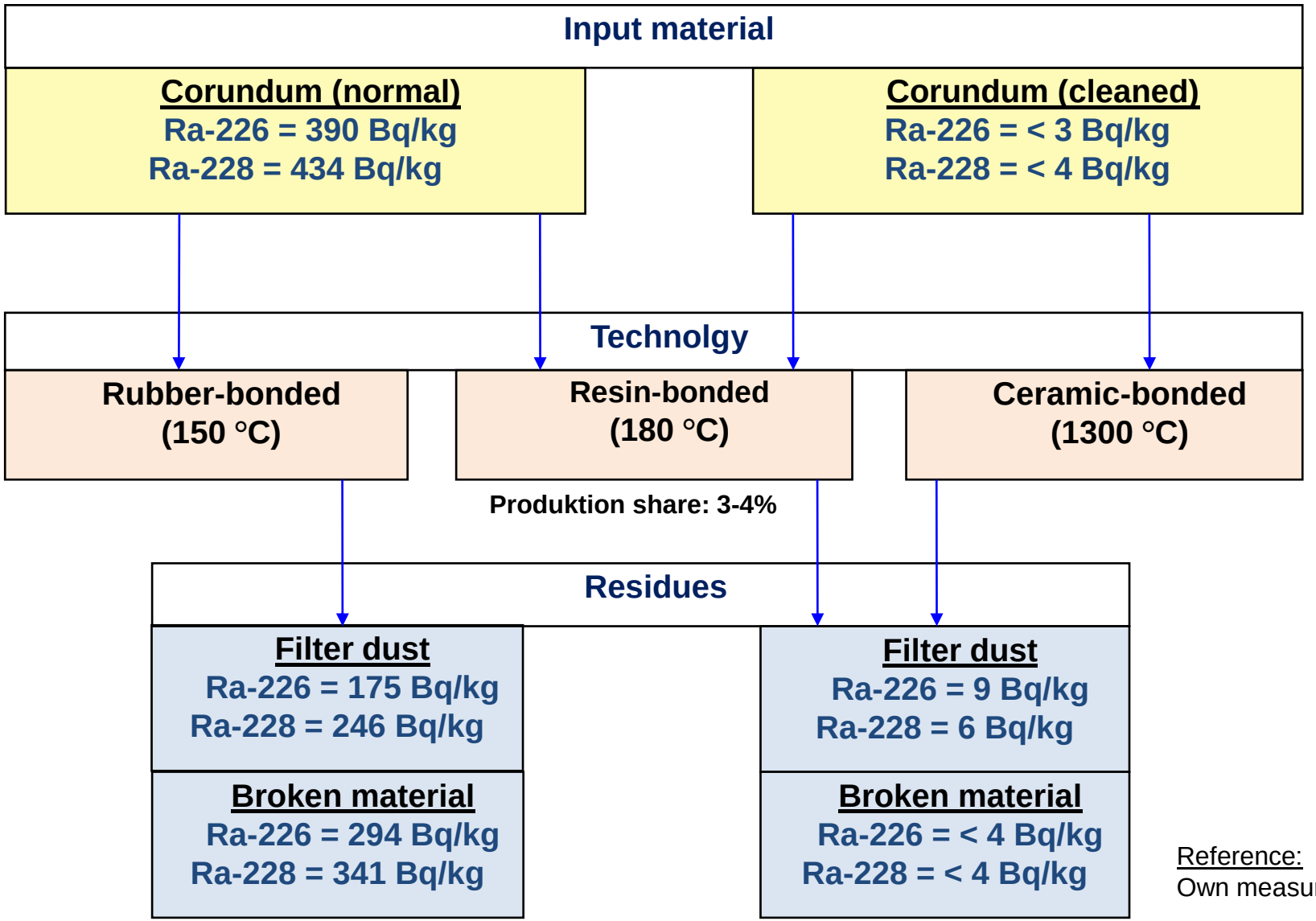
Example: Manufacture of corundum abrasives



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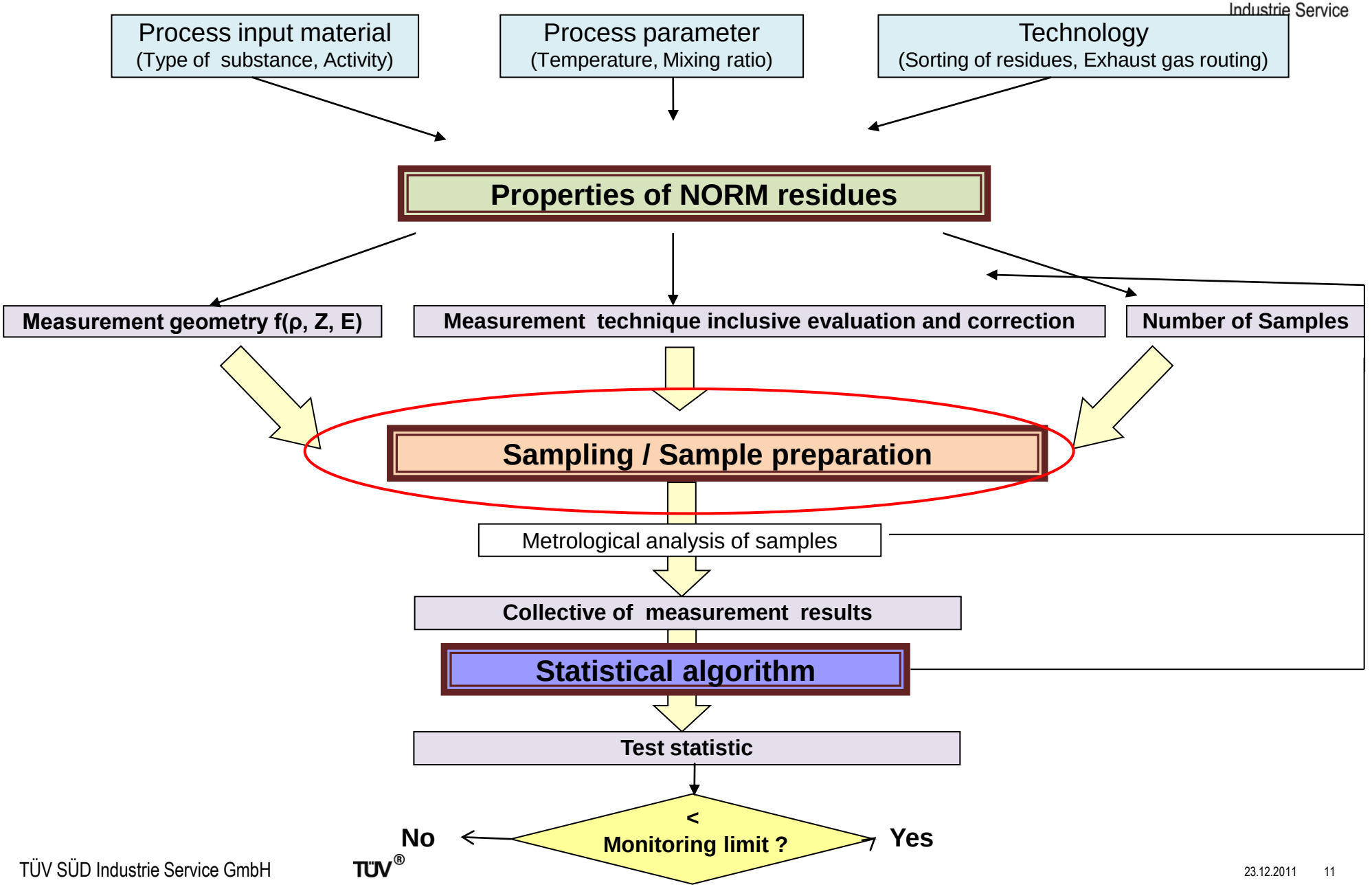
Fundamental Relationships



Recommendations for Sampling

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Homogeneity and Variability:

It is expected with a general inhomogeneity and variability of the accumulated NORM residues. The sampling must be guided accordingly.

Kind of Sampling:

- Random sampling of individual samples
- Sampling volume of 1 liter at first
- Sampling in one step without separation
- No mixing or gross samples, multistage sampling, cluster sampling, hot-spot-sampling, no repeated, time-separated sampling

Number of Samples:

- Minimum of ten individual samples to be taken per charge
- The total number of samples must be made dependent on the distance of the mean value of the monitoring limit and the coefficient of variation of the statistical analysis
- great piles to be sampled with a stratified sampling

Recommendations for Sample Preparation

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Sample Preparation:

- Based on the material properties (ρ, Z, E) the measurement geometry is defined. This determines the sample preparation.
- At peaks with gamma-lines energies < 100 keV, additional processing steps are possibly necessary (changes in the measurement geometry).
- If the grain size of the sample for the 1-liter geometry is too high (from about 2 cm grain diameter): milling and homogenization.
- If the measured material must not be milled, it is proposed, the drying to perform after the measurement and to relate the obtained measurement results of the wet sample on the dry mass.

Recommendations for Analysis of Samples

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Radionuclides:

- Consideration of the following relevant radionuclides:

Uranium series	Thorium series	Actinium series
U-238 Th-230 Ra-226 Pb-210 Po-210	Th-232 Ra-228 Th-228	Pa-231 Ac-227

activity U-234 ≅ activity U-238
activity U-235 ≅ 0,05 * activity U-238

- At a basic investigation, all relevant key radionuclides must be measured; later reference nuclides can be calculated with the aid of activity ratios
- From a metrological determined equilibrium Ra-228/Th-228 is not to conclude, that the whole series, including Th-232, is in a radioactive equilibrium.
- The need for the recording of Po-210 must be decided based on a temporal analysis.
- Ac-227 and Pa-231 were in some of the investigated cases, enriched compared to U-235.

Recommendations for Analysis of Samples

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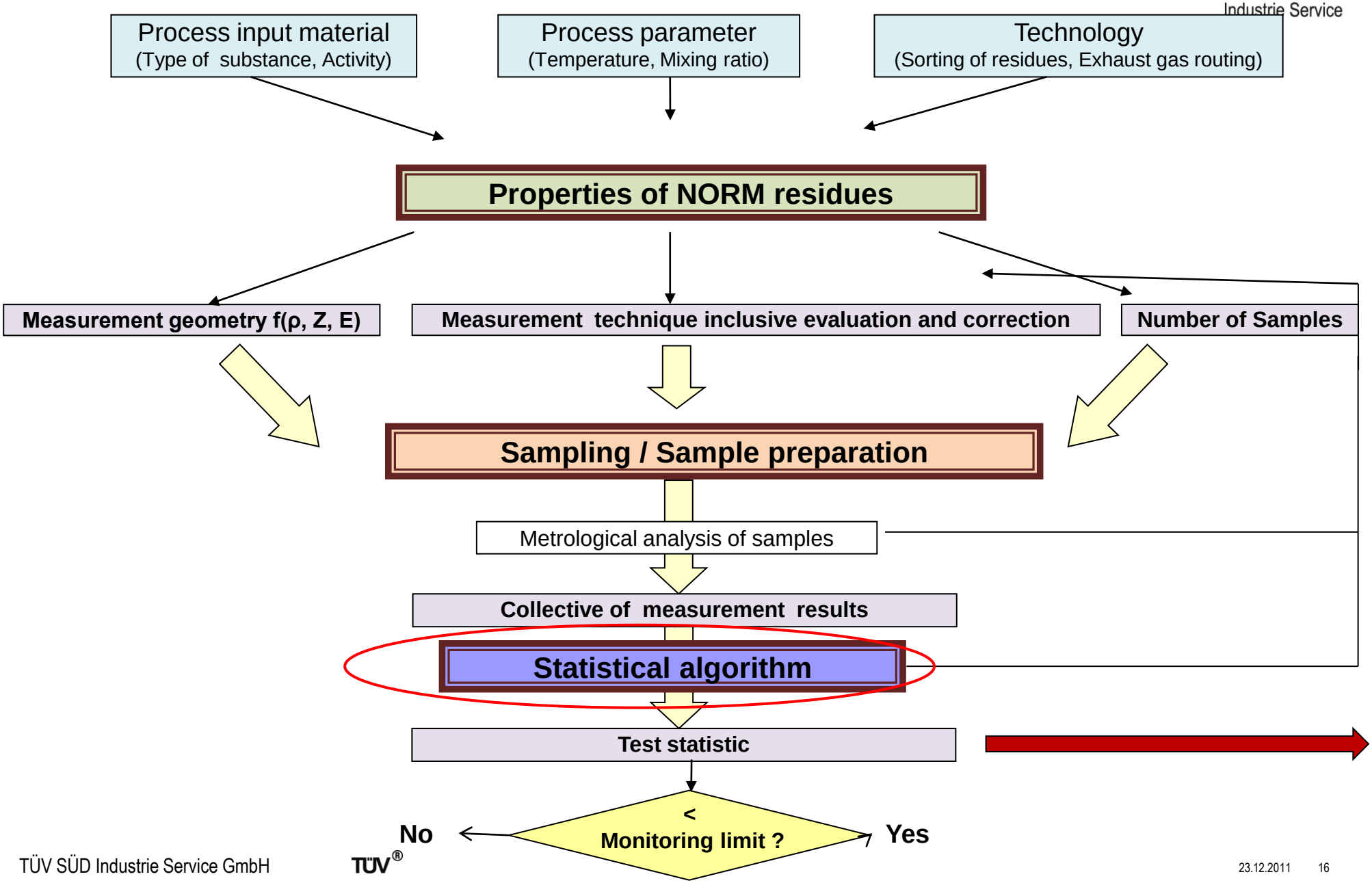
Measurement Process :

- The times for setting the corresponding equilibria have to be waited.
- Due to the uncertainty of the modeling of the control limits and the systematic uncertainties associated with each sampling, it is not appropriate to demand measurement uncertainties of less than 10% at the measurement of samples in the laboratory.
- A classification of residues according to their radionuclide ratios and their dominant radionuclides U-238_{max} and Th-238_{max} is orienting possible (compared with representative nuclide vectors).
- In the specific case - at least exemplary for some residue charges - all key radionuclides are to be measured (including Th-230 and Th-232).

Fundamental Relationships



Statistical algorithm

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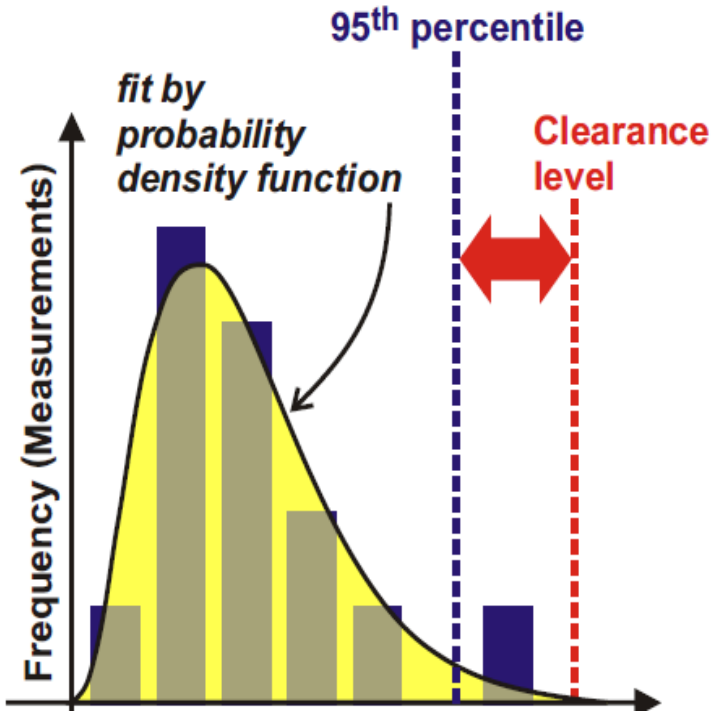
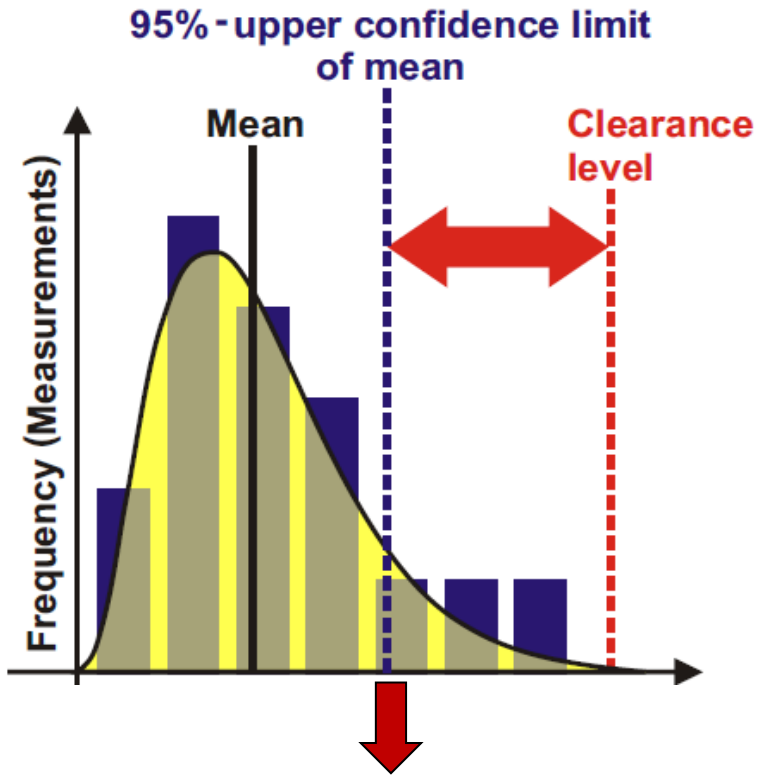
Test statistic





in Germany for NORM: 95 % UCL of mean

Statistical algorithm

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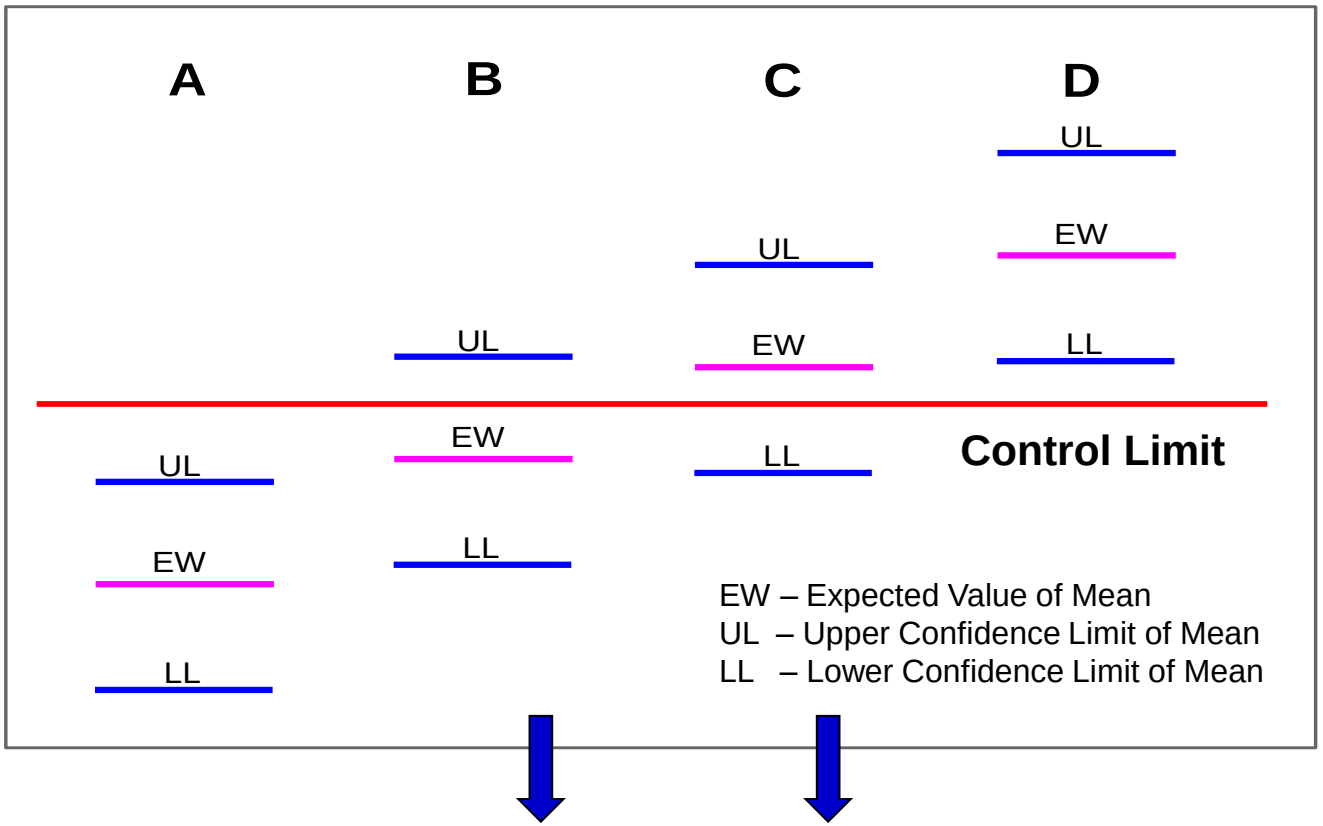
Test statistic

Possible positions of the confidence intervals of the expected value of the mass-related activity to the control limit



<u>Case B/C: Possibility of falling below the control limit:</u>
Increase the number of random samples; Increase the measuring accuracy

Statistical algorithm

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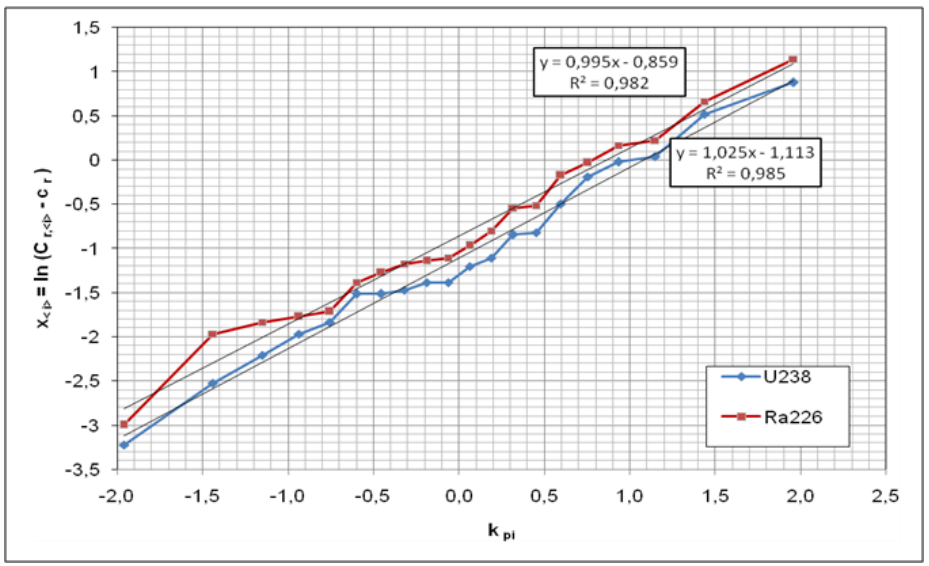
In Germany: Recommandation of the Commission on Radiological Protection (SSK): „Classical“ statistical algorithm to determine the upper confidence limit (UCL)

Proben-Nr. <i>	$C_{U238,i}$	$C_{Ra226,i}$	k_{Pi}	Proben-Nr. <i>	$C_{U238,i}$	$C_{Ra226,i}$	k_{Pi}
1	0,12	0,14	-1,960	11	0,38	0,47	0,063
2	0,16	0,23	-1,440	12	0,41	0,54	0,189
3	0,19	0,25	-1,150	13	0,51	0,67	0,319
4	0,22	0,26	-0,935	14	0,52	0,69	0,454
5	0,24	0,27	-0,755	15	0,69	0,93	0,598
6	0,30	0,34	-0,598	16	0,91	1,06	0,755
7	0,30	0,37	-0,454	17	1,06	1,27	0,935
8	0,31	0,40	-0,319	18	1,12	1,34	1,150
9	0,33	0,41	-0,189	19	1,76	2,03	1,440
10	0,33	0,42	-0,063	20	2,49	3,23	1,960



$$s_{r,n} = \frac{1}{k_{(n-1)/n}} \ln\left(\sqrt{\frac{C_{r<n>} \cdot C_{r,<n-1>}}{C_{r<n/2>} \cdot C_{r<n/2+1>}}}\right)$$

$$R^2(c) = \frac{\left(\sum_{i=1}^{n} x_{<i>}(c) \cdot k_{Pi}\right)}{(n-1) \cdot \sigma_n^2(c) \cdot \sum_{i=1}^{n} {k_{Pi}}^2}$$

$$S(c) = \frac{n}{(n-1)\cdot(n-2)} \cdot \sum_{i=1}^{n}\left(\frac{x_i(c) - \mu_n(c)}{\sigma_n(c)}\right)^3$$

$$UL_{U\text{-}238} = c_{U\text{-}238} + E_{U\text{-}238}^{(n;P)} = (0{,}08 + 1{,}03)\ \text{Bq/g} = 1{,}11\ \text{Bq/g}$$

$$UL_{Ra\text{-}226} = c_{Ra\text{-}226} + E_{Ra\text{-}226}^{(n;P)} = (0{,}09 + 1{,}26)\ \text{Bq/g} = 1{,}35\ \text{Bq/g}$$

Statistical algorithm

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biggest drawback of this method:

applies only for normal and log-normal distributed populations

Search for a method with the following properties:

❶ Consistent procedure for all applications (even independent of the coefficient of variation)

❷ Fundamental suitability to test the 95%-VG

❸ Parameter-free method (no proof of the assumed probability density function is necessary)

❹ Efficient method, which means fast convergence of the calculated estimates to the actual parameters.

❺ Robust method that generates plausible results.

❻ Into account the measurement uncertainty in the process.

➢The „bootstrap“-method is the base.
➢Implementation of the type B measurement inaccuracy (modification 1)
➢Application of the BCa method (modification 2)

Bootstrap: Definition

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$$\hat{F} \rightarrow \vec{x}^{\times};$$

Bootstrap-Vector

Sampling with replacement

with: $\vec{x}^{\times j} \equiv \begin{pmatrix} x_1^{\times j} \\ x_2^{\times j} \\ \cdots \\ x_n^{\times j} \end{pmatrix}$; in which:

$$x_i \equiv \frac{A_{U-238,\max,i} + A_{Th-232,\max,i}}{CL} \quad 1 \le i \le n$$

$$1 \le j \le B;\ B > 1000$$

CL = Control Limit

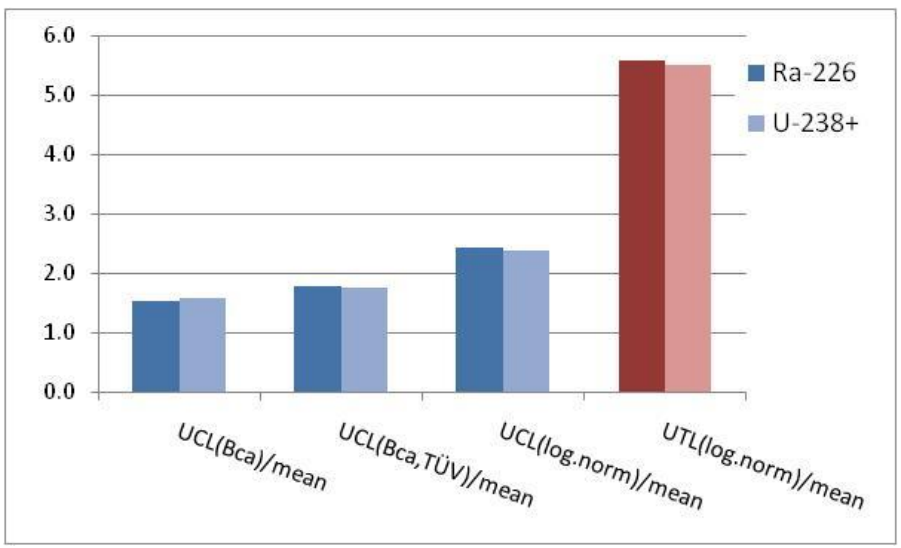
95%-UCL$_{boot}$: $\hat{\theta}^* + z^{(1-\alpha)} s\hat{e};$

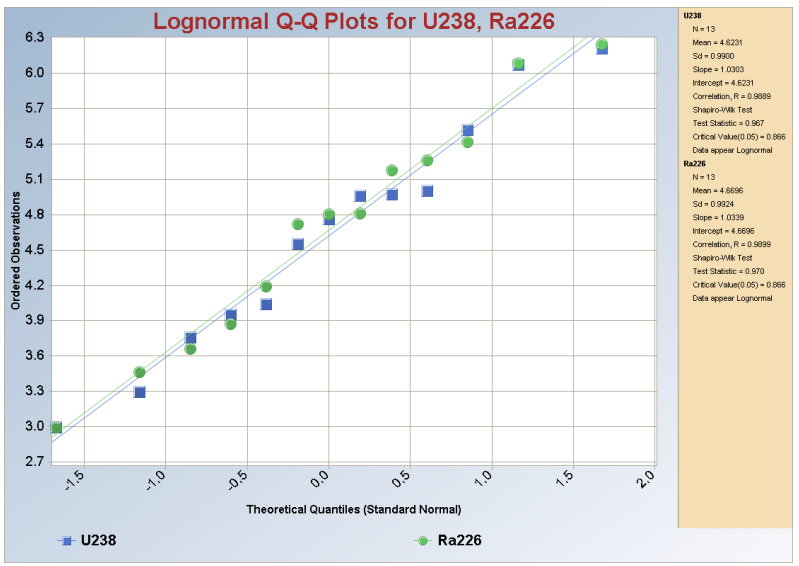
$$s\hat{e} = \sqrt{\frac{\sum_{j=1}^{B} [\hat{\theta}^{\times}(j) - \hat{\theta}^{\times}]^2}{B-1}} \quad mit: \ \hat{\theta}^{\times} = \sum_{j=1}^{B} \hat{\theta}^{\times}(j) / B;$$

Statistical algorithm

Examples of calculating the 95 % **U**pper **C**onfidence **L**imit (UCL) (Values in [Bq/g]):

		Classical method (95%-UCL)	**Bootstrap-method (95%-UCL$_{BCa}$); B=1000**
Constructed [SSK, 2004]	n=20, Ra-226	**1,35**	**1,35 / 1,41 / 1,39**
Real data [Reichelt, 2005]	n=10, Ra-226	**0,42**	**0,34 / 0,33 / 0,34**





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Conclusions

- It seems purposeful, numerical methods also in the statistical treatment of data to investigate at their applicability.

- Real development work is still necessary.