Measurement of surface contamination according to legal requirements

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Abstract. The measurement of radioactive contamination of metal surfaces is a common issue in NORM industries, e.g. because of the scale deposition in pipes and other equipment of oil/gas production facilities. Regulatory requirements for measurements may address radiological assessments aimed at occupational radiation protection or may be aimed at confirming compliance with surface contamination limits for the release of scrap from the dismantling of installations for recycling. Such requirements may apply to different parameters, e.g. to area specific alpha or beta activities or to summation formulas for the area specific activities of relevant long lived radionuclides, and differ from country to country. The measurement of surface contamination levels in large scrap volumes requires a method that is applicable in a straightforward way under field conditions with minimum expenditure of time. These objectives can be accomplished by means of beta contamination measurements combined with gamma spectrometric sample analyses for the calibration according to the activity ratios of the long lived radionuclides. A calibration of beta contamination measurements was performed by numerical calculations using a well-known Monte Carlo particle transport model. Apart from the detector sensitivity, the detector response is dominated by the area specific mass of the contaminated layer and by the activity ratios between the relevant long lived radionuclides. The calibration has been validated for surface contaminations of different type/origin, which comprise the radium isotopes ^{226}Ra, ^{228}Ra and their daughter nuclides. The comparison with measurements of scratch samples shows good agreement. Therefore, it may be concluded that the use of Monte Carlo particle transport model calculations represents a cost effective and very adaptive method for the calibration of surface contamination measurements.

1. Introduction

Surface contamination of pipes and other components by naturally occurring radionuclides of the decay chains of ^{238}U and ^{232}Th is a common issue in NORM industries. In the present paper, the measurement of radioactive surface contamination is considered by way of example for oil/gas production facilities, but the method applies similarly to other NORM industries.

The parent radionuclides ^{238}U and ^{232}Th have very long half lives and are ubiquitous in the earth's crust with specific activities that depend on the type of the reservoir rocks. Their decay produces chains of daughter nuclides of different physical characteristics with respect to their mobility, half lives, decay modes and energies of emitted radiation. Concerning the radioactive contamination of oil/gas production facilities it should be mentioned that the reservoir water contains Group II (Periodic Table) cations of calcium, strontium, barium and radium dissolved from the reservoir rocks. As a consequence, the produced water contains the long lived radium isotopes ^{226}Ra from the ^{238}U series and ^{228}Ra from the ^{232}Th series, whereas their parent nuclides are not mobilized with the formation water of the reservoir rocks. Due to the operation history, pipes, valves and other components of the production facilities are more or less contaminated with ^{226}Ra and ^{228}Ra and their daughter nuclides. It should be mentioned that in some oil/gas fields ^{210}Pb occurs in considerable amounts already in the produced water, which may cause a relatively high surface contamination with ^{210}Pb. An elevated surface contamination with ^{210}Pb may also be caused by the decay of ^{222}Rn. The radioactive decay chains of the radium isotopes ^{226}Ra and ^{228}Ra, which are the leading radionuclides of NORM in the oil/gas industry, are shown in Fig. 1.

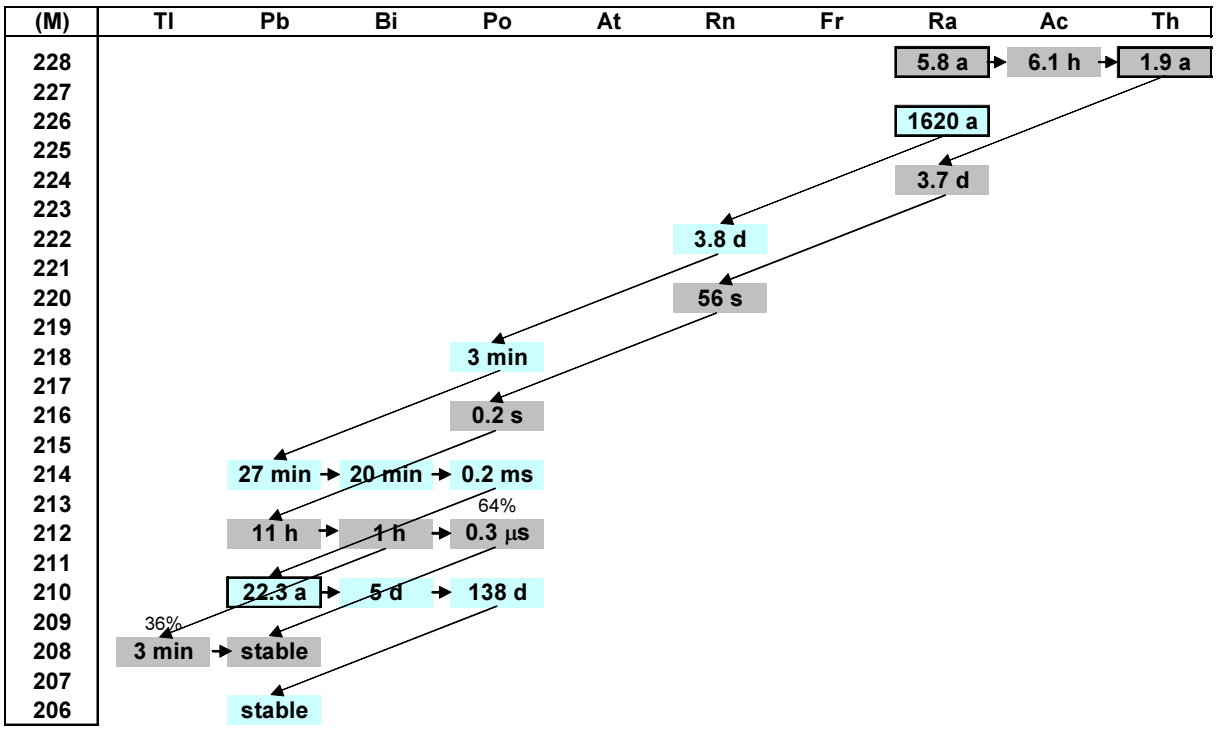

FIG. 1: Radioactive decay of Ra-226 and Ra-228 (half-life indicated for each radio-nuclide)

The radiological assessment of contaminated pipes or other components requires knowledge of the area specific activities of the long lived radionuclides (short lived nuclides are in radioactive equilibrium with their long lived parent nuclides). Taking into account that for the given circumstances ^{210}Po may be considered as short lived, the relevant long lived radionuclides, which in general will have different specific activities, are ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{228}Th. The alpha and beta emitting nuclides of the sub-chains, which comprise the long lived (leading) parent nuclide and its short lived daughter nuclides up to the next long lived nuclide are summarized in Table 1.

TABLE 1: ALPHA AND BETA EMITTERS IN THE SUB-CHAINS OF THE RELEVANT LONG LIVED RADIONUCLIDES

Sub-chain	Alpha emitters		Beta emitters	
	Radionuclides	No.	Radionuclides	No.
^{226}Ra+	^{226}Ra, ^{220}Rn, ^{218}Po, ^{214}Po	4	^{214}Pb, ^{214}Bi	2
^{210}Pb+	^{210}Po	1	^{210}Pb, ^{210}Bi	2
^{228}Ra+	—	0	^{228}Ra, ^{228}Ac	2
^{228}Th+	^{228}Th, ^{224}Ra, ^{220}Rn, ^{216}Po, ^{212}Bi (36%), ^{212}Po (64%)	5	^{212}Pb, ^{212}Bi (64%), ^{208}Tl (36%)	2

2. Regulatory requirements

Regulatory requirements for the release of pipes and other components as 'uncontaminated' for unrestricted reuse and/or for the clearance of scrap for smelting deal at least with two issues: the definition of the radiological parameter that has to be applied for the decision, and the definition of a limit for this parameter, below which the treatment of contaminated pieces according to a certain licensing regime is not required. For limiting surface contamination, different radiological parameters may be used, e.g. the area specific β activity S_β or the area specific α activity S_α. Sometimes, regulatory bodies combine limitations of the kind $S_\beta \leq L_\beta$ and/or $S_\alpha \leq L_\alpha$, where L_β and L_α are the

limiting values of beta and alpha activity, respectively. In this case, the more restrictive condition has to be applied.

The radiation protection ordinance of the Netherlands MR-NABIS [1] defines, for instance, the presence of NORM with an average mass per unit surface of less than 1 g/cm² on non-radioactive solids as 'surface contamination' and requires measurements of the area specific β activity on surfaces of non-radioactive materials. The regulations [1] require a treatment of contaminated parts according to the licensing regime if the area specific β activity S_β of accessible surfaces is above the limit of L_β = 4 Bq/cm^2.

Combined limitations are applied, for instance, in the Libyan 'NORM Management Manual' [2] that defines radioactive surface contamination as the presence of radioactivity on a surface in excess of L_β = 3.0 Bq/cm^2 or L_α = 0.3 Bq/cm^2, and these limits as action levels for the control of radiological work.

International regulations for the safe transport of radioactive material are set out in the IAEA Safety Standards [3] defining radioactive contamination as the presence of a radioactive substance on a surface in excess of 0.4 Bq/cm^2 for beta and gamma emitters and low toxic alpha emitters or of 0.04 Bq/cm^2 for all other alpha emitters (in the present case this concerns ^{226}Ra). If the area specific activity S_r of a relevant radionuclide is above its so-called exemption level (L_r), the object is regarded as a 'surface contaminated object'.

An alternative approach is recommended by the European Commission for the recycling of metals from the dismantling of nuclear installations [4]. Based on the assessment of exposure scenarios, area specific clearance levels L_r corresponding to a maximum individual dose of about 10 μSv/a were derived for long lived radionuclides r. For the nuclides relevant in scales of oil/gas production facilities, the following clearance levels are listed in Tab. 7-1 of [4]: $L_{Ra\text{-}226}$ = 0.3 Bq/cm^2; $L_{Pb\text{-}210}$ = 0.58 Bq/cm^2; $L_{Ra\text{-}228}$ = 0.39 Bq/cm^2 and $L_{Th\text{-}228}$ = 0.097 Bq/cm^2. For the clearance of scrap the so-called 'summation formula' $\sum_r \left(S_r \div L_r\right) \le 1$ has to be applied.

For a given type of radioactive contamination with known activity ratios between the relevant long lived radionuclides, the above mentioned requirements can be transformed into each other. Therefore, the compliance with any exemption or clearance levels defined by a regulatory body for surface contaminations may by proven, for instance, by measurements of the area specific β activity S_β.

Denoting by A_r (in Bq/g) the mass specific activity of the contamination for the radionuclide r and by μ (in g/cm^2) the area specific mass, the area specific activity S_r (in Bq/cm^2) is determined by

$$S_r = A_r \times \mu \tag{1}$$

In the following, we select ^{226}Ra as the 'reference nuclide' and define the activity ratios

$$R_r = A_r / A_{Ra-226} \tag{2}$$

Using the numbers of alpha and beta emitters listed in Table 1, a limit L_α can be transformed, for instance, into a derived limit for S_β:

$$L_\beta(L_\alpha) = L_\alpha \times \frac{2 \times (1 + R_{Pb-210} + R_{Ra-228} + R_{Th-228})}{4 + R_{Pb-210} + 5 \times R_{Th-228}} \tag{3}$$

The derived limit for the area specific β activity S_β, which corresponds to the summation formula of [4], is given by the following equation:

$$L_\beta(L_r) = \frac{2 \times (1 + R_{Pb-210} + R_{Ra-228} + R_{Th-228})}{\sum_r R_r / L_r} \quad (4)$$

Example: For scale from contaminated pipes of a former flare line of an oil/gas production facility in Libya, by means of gamma spectrometric measurements of scratch samples the following activity ratios were established: $R_{Pb-210} = 0.7$; $R_{Ra-228} = 0.5$ and $R_{Th-228} = 0.7$. Due to Eq. (3), the limit of $L_\alpha = 0.3$ Bq/cm^2 defined in [2] is equivalent to a derived limit for the area specific β-activity of $L_\beta(L_\alpha) = 0.21$ Bq/cm^2. A derived limit of $L_\beta(L_r) = 0.44$ Bq/cm^2 resulting from Eq.(4) would have to be applied due to [4]. The exemption level $L_{Ra-226} = 0.04$ Bq/cm^2, which is valid according to [3], yields a derived limit of $L_\beta(L_{Ra-226}) = 0.23$ Bq/cm^2. The limit of $L_\beta = 4$ Bq/cm^2 defined in [1] is about one order of magnitude above the previously mentioned derived limits. From the radiological point of view it presents, however, a reasonable limit, which corresponds according to [4] to a maximum individual dose of approximately 0.1 mSv/a.

3. Measurement of radioactive surface contamination

3.1. General comments

If a scale sample can be scratched from the surface of a contaminated piece, direct measurement of the surface contamination is possible by measuring the area specific mass μ and the specific activities A_r of the relevant long lived radionuclides. The area specific activities S_r are then determined by Eq. (1), and the area specific β activity S_β is calculated by

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However, disadvantages of this approach are the exhausting and time-consuming extraction of scratch samples and the relatively high expenses for the gamma spectrometric measurements of the specific activities A_r. In many cases it is impossible to obtain a sufficient sample mass. The contaminated surface layer is frequently so thin that a scratch sample cannot be taken at all. Therefore, an alternative method for measuring the surface contamination will be necessary, especially in the case of low levels of contamination, which have to be checked against exemption or clearance levels.

The measurement of surface contamination in large scrap volumes or in complex production facilities requires a method, which is applicable in a straightforward way under field conditions with an a minimum expenditure of time. These objectives can be accomplished by means of beta contamination measurements. They have to be combined with gamma spectrometric analyses of a few scratch samples, as the calibration has to take into account the existing activity ratios of the relevant long lived radionuclides. This is necessary because of nuclide specific energy spectra of the emitted β particles, which influence the detector response. The calibration has also to take into account the area specific mass μ of the contaminated surface layer, as it influences the self-absorption of emitted β particles within the scale layer.

3.2. Application of a beta sensitive surface contamination probe

By means of a beta sensitive surface contamination probe that has been calibrated for the given conditions (i.e. the activity ratios between the relevant long lived radionuclides and the area specific mass), sufficiently accurate or, at least, conservative measurements of S_β are possible. The surface contamination probes are in general sensitive for beta particles as well as for gamma rays. Therefore, in addition to the counts C_β (in cps = counts per second) resulting from the required area specific β activity S_β of the measured area, counts $C_{\gamma,area}$ due to γ emissions originating from the contamination of this area, and counts $C_{\gamma,surr}$ originating from γ emissions from the surrounding contaminated surface and from other pieces also contribute to the total count rate C_{total} of the measurement device:

$$C_{total} = C_\beta + C_\gamma \quad \text{with} \quad C_\gamma = C_{\gamma;area} + C_{\gamma;surr}\,. \tag{6}$$

To determine the β count rate C_β, in addition to the measurement of the total count rate C_{total}, the γ count rate C_γ has to be measured at the same position by shielding the detector against β particles by a 3 mm thick aluminum plate. After this, C_β is calculated according to Eq. (6).

3.3. Calibration of a beta sensitive surface contamination probe

The determination of the area specific β activity S_β from a measured β count rate C_β requires the calibration of the measurement device. The detector response depends on the specific activities of the relevant long lived radionuclides and on the area specific mass μ of the contaminated layer. The calibration may be performed experimentally or by means of Monte Carlo particle transport calculations for the detector used, yielding for each relevant radionuclide r a detector response function $D_r(\mu)$ in units of cps per Bq/cm^2. Given an area specific activity S_r of the radionuclide r (r = ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th), the detector response D_r defines the count rate $C_{\beta;r}$ (in cps) for β particles emitted by radionuclide r and its short lived daughters:

$$C_{\beta;r} = n_r \times D_r \times S_r \tag{7}$$

where n_r denotes the number of β particles emitted per decay of radionuclide r by this nuclide and/or its short lived daughters (for the four sub-chains ^{226}Ra+, ^{210}Pb+, ^{228}Ra+ and ^{228}Th+ the value of n_r is 2, see Table 1). The ratio between the area specific β activity S_β and the β count rate C_β:

$$C_\beta = \sum_r C_{\beta;r} \tag{8}$$

resulting from combining Eqs. (1), (5) and (7) is:

$$\frac{S_\beta}{C_\beta} = \frac{2 \times \mu \times \sum_r A_r}{2 \times \mu \times \sum_r D_r \times A_r}\,. \tag{9}$$

Using the activity ratios R_r defined in Eq. (2), the area specific β activity S_β is determined by the measured β count rate C_β via

$$S_\beta = C_\beta / D_\beta(\mu) \quad \text{and} \tag{10}$$

$$D_\beta(\mu) = \frac{\sum_r D_r(\mu) \times R_r}{\sum_r R_r}\,. \tag{11}$$

$D_\beta(\mu)$ denotes the effective detector response for β emissions. It depends on the given ratios of the specific activities of the long lived radionuclides and on the area specific mass μ.

The nuclide specific detector response functions $D_r(\mu)$ were calculated for the β sensitive surface contamination probe 'NORM1' (RADOS GmbH), which is a plastic scintillation detector with a window size of 17.1 cm × 4.4 cm ≅ 75 cm^2. Due to its geometrical dimensions (L ×W × H = 28.2 cm × 6.2 cm × 3.8 cm), it is suited for measuring surface contamination also in pipes with inner diameters of ≥ 3 inches (75 mm) (taking into account the thickness of 3 mm of the aluminum plate that has to be used for the shielding of β particles for the measurement of C_γ).

The detector response functions $D_r(\mu)$ were calculated with version 4C of the Monte-Carlo N-Particle transport code MCNP (developed by the Los Alamos National Laboratory and distributed by bodies such as the OECD-NEA [5]). The results are shown in Fig. 2.

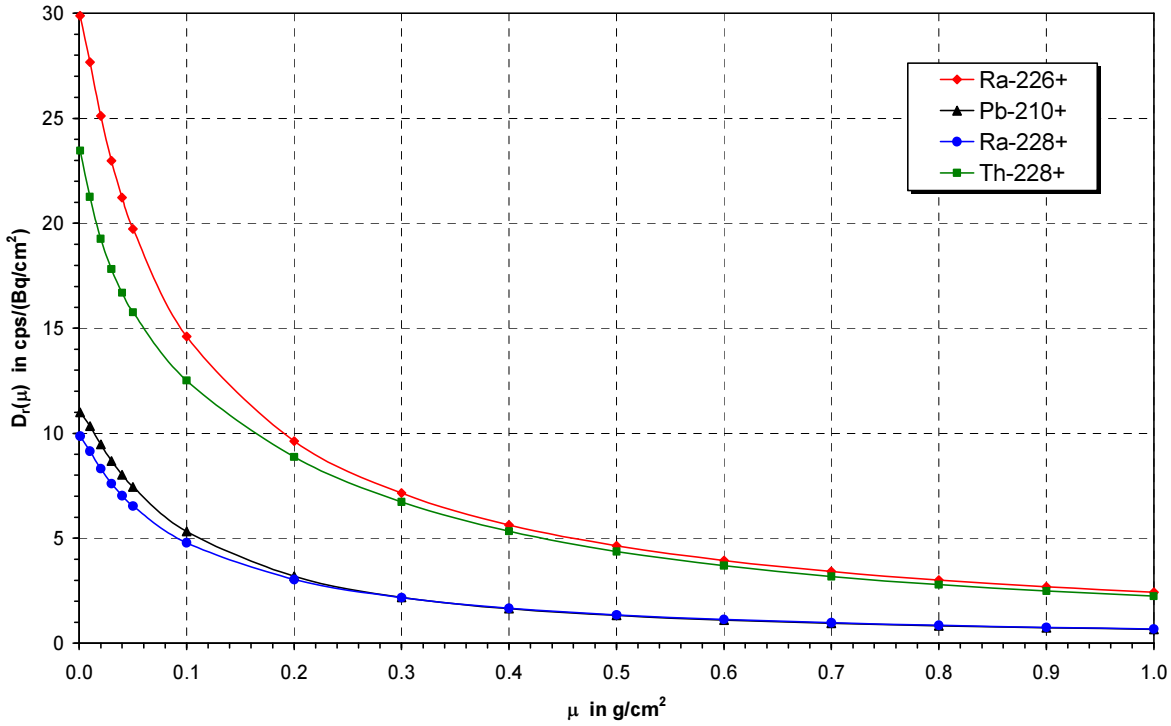

FIG. 2: Detector response functions $D_r(\mu)$ for sub-chains of relevant radionuclides

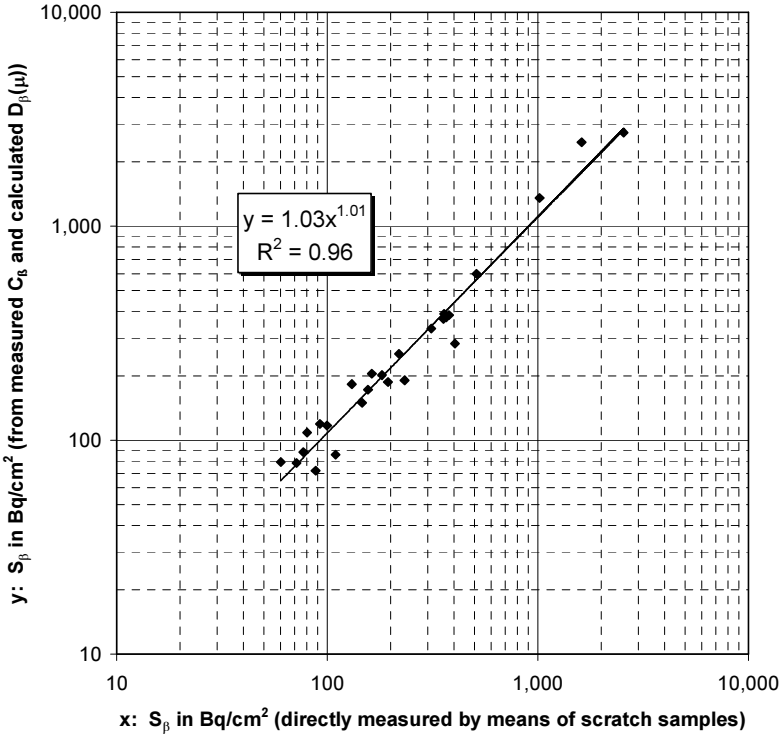

FIG. 3: Comparison of area specific β activities S_β determined directly by measurements of scratch samples and estimated alternatively from measured β count rates C_β with calculated detector response functions $D_\beta(\mu)$ via Eq. (10); 26 scale samples of six different types (activity ratios) and area specific masses μ from 0.05 to 1.5 g/cm^2

The detector response functions for the sub-chains ^{210}Pb+ and ^{228}Ra+ are relatively small compared with those for ^{226}Ra+ and ^{228}Th+. This is due to the low energy of each one of the β particles emitted by these sub-chains: decay of ^{210}Pb with $E_{\beta;max}$ = 63 keV and of ^{228}Ra with $E_{\beta;max}$ = 39 keV.

The calculated detector response functions are validated by means of experimental calibration data for different types of NORM layers with known specific activities of ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{228}Th and varying layer thicknesses. With respect to field conditions, area specific β activities S_β directly measured by gamma spectrometry on the basis of scratch samples taken from layers of different scale types have been compared with surface contamination measurements performed with the 'NORM1' probe using the calibration given in Eqs (10) and (11). The comparison shown in Fig. 3 confirms a good agreement. It may be concluded that the use of Monte Carlo particle transport model calculations represents a cost effective and very adaptive method for the calibration of surface contamination measurements.

3.4. Proof of compliance with surface contamination limits

A direct measurement of the area specific mass μ, which has to be known for calculating the detector response function, cannot be realized in general, for instance in the case of a very thin scale contamination that may remain after pipe cleaning. To overcome this difficulty, an alternative approach for the determination of μ can be applied if the mass specific β activity A_β of this scale type is known from former measurements ($A_\beta = \Sigma\, n_r \cdot A_r = S_\beta/\mu$). It can be assumed that A_β is a relatively constant parameter. On this basis, the area specific mass μ can be estimated for each measurement point from the measured β count rate C_β by solving the following equation, which results from Eqs (1) and (10), with respect to μ:

$$\frac{C_\beta}{\mu \times D_\beta(\mu)} = A_\beta \cong \text{const}. \qquad (12)$$

As the detector response function $D_\beta(\mu)$ tends to its asymptote (~1/μ) with increasing area specific mass μ because of self-absorption, this approach cannot be applied for very thick scale layers. However, for area specific masses of contamination that are relevant for proof of compliance with regulatory limits, this approach can be used for the calculation of derived limits LC_β for the measured β count rate corresponding to a contamination limit L_β. As shown by way of example in Fig. 4 for the scale type that is characterized by the activity ratios R_r considered in the example of Section 2, the calculation of derived limits LC_β is not affected by uncertainties concerning the specific β activity A_β in the relevant region of β count rates.

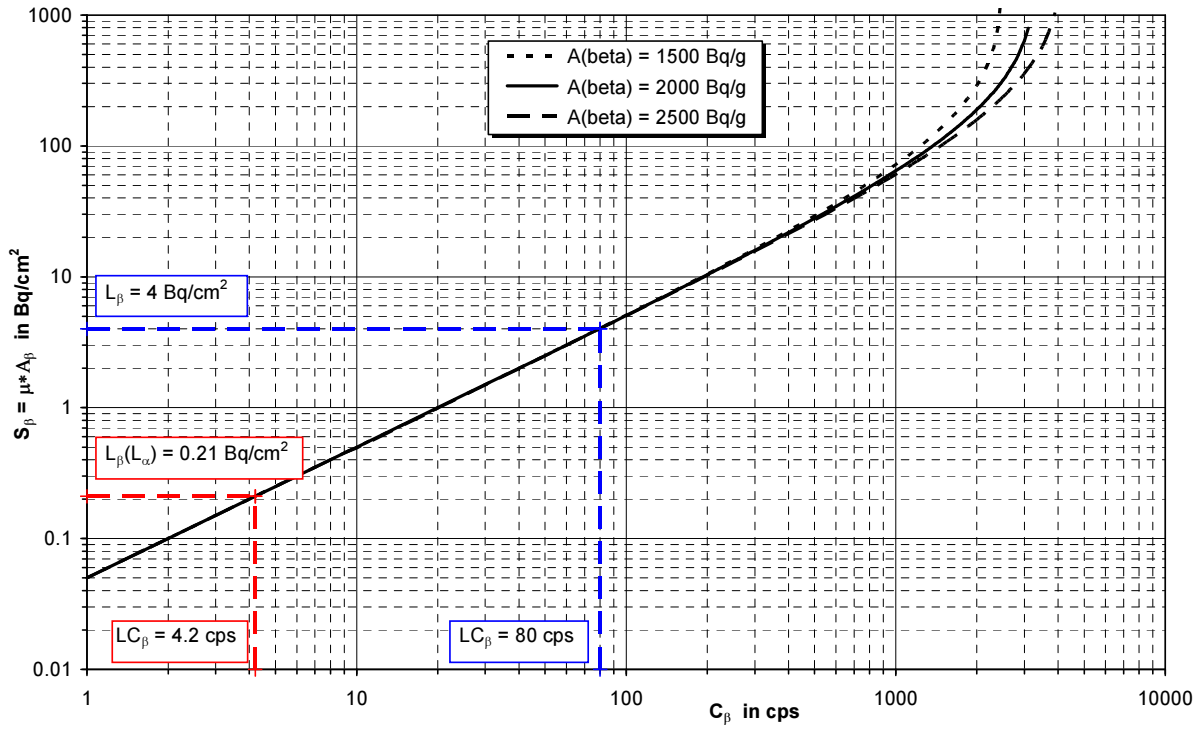

FIG. 4: Derived limits LC_β for β-count rates C_β for the scale type considered in the example of section with a specific β-activity of $A_\beta \cong (2000 \pm 500)$ Bq/g, which correspond to the limits L_α = 0.3 Bq/cm^2 ($L_\beta(L_\alpha)$ = 0.21 Bq/cm^2) and L_β = 4 Bq/cm^2, respectively

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