SURFACE CONTAMINATION CRITERIA FOR EQUIPMENT CONTAMINATED WITH NORM

Meijne E., Weers A. van, Verhagen H.
NRG, P.O. Box 25, 1755 ZG Petten, The Netherlands
Email: meijne@nrg-nl.com

1 ABSTRACT

After implementation of Council Directive 96/29/Euratom in the new Dutch legislation (BS) the total activity concentration criterion (100 Bq/g) and total activity criterion for exemption from authorization will be replaced by nuclide specific exemption/clearance levels. In addition the Dutch Authorities intend to introduce exemption/clearance levels specifically for surface contaminated objects. These will replace the current practice to estimate the presence of material falling under authorization inside or on objects using a criterion of 3x the background count rate measured with a portable surface contamination monitor. The exemption levels for alpha emitters are likely to be set at a factor of ten more restrictive than for beta-emitters and will be applied to long-lived alpha emitters only. Since many NORM radionuclides are alpha emitters the lower clearance level will be applicable to NORM contaminated objects. Portable contamination monitors which in practice can measure NORM alpha contamination reliably do not exist. However, all long-lived alpha emitters relevant to NORM contain beta emitters in their decay chain. The ratio of beta-emitters to long-lived alpha emitters varies from 1:1 for Pb-210 containing deposits to 3:1 for old Ra-228 deposits. By assuming a ratio of 1:1 a conservative estimate of the long-lived alpha emitters in the contamination can be obtained for all types of NORM deposits from beta measurements.

To determine the activity per unit area the contamination monitors need to be calibrated. Ideally this should be done with calibration sources containing the same radionuclides as anticipated in the field. Almost all NORM deposits however emit beta particles with widely ranging $E_{\beta max}$. By selecting a moderately low-energy calibration source like Co-60 a conservative value for the surface contamination will be obtained.

2 INTRODUCTION

Council Directive 96/29/EURATOM [1] establishes basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation. The provision in the Directive will be implemented in Dutch legislation by the enforcement of the "Besluit Stralingsbescherming [BS]" [2]. In the Council Directive special attention is given to exposure to natural radiation sources. The provisions given in the Directive are in principle not only applicable to "practices" but also to ‘work activities’. Work activities are defined as activities, which involve the presence of natural radiation sources and lead to a significant increase in the exposure of workers or members of the public which cannot be disregarded from the radiation protection point of view. According to Title VII of the Directive Members States of EU have the obligation to investigate which work activities should be brought under some form of radiological control, using some or all of the stipulations of Titles III, IV, V, VI and VIII.

3 IMPLEMENTATION OF THE COUNCIL DIRECTIVE

A survey of relevant work activities in The Netherlands identified a large number of non-nuclear industries which may be of concern in the context of Title VII and to which provisions of BS may be applicable [3]. Of particular interest to these industries are the exemption/clearance levels for radioactive substances below which no reporting or prior authorization is required and which also determine if waste material has to be treated as radioactive waste. Instead of one value for the total activity concentration as was the case in the Council Directive 80/836, Table A of the Council Directive 96/29 specifies nuclide specific exemption values (Bq/g). These exemption values have been derived derived using exposure scenario's applicable to "practices" and thus involve small or moderate quantities of material. They are therefore not a priori suitable as exemption value for NORM material. Under contract with the Dutch authorities a number of studies have therefore been performed to define exemption (and clearance) levels for large quantities of residual materials from non-nuclear industries [4], [5], [6]. By taking into account the results of these studies the Dutch authorities established exemption levels that are applicable to "practices" as well as "work activities". They differ for a number of relevant NORM radionuclides from those specified in the Council Directive.
Registration is required when materials are possessed with concentrations and total amounts exceeding the exemption/clearance levels. Prior authorization is required if the exemption/clearance levels are exceeded by more than a factor of 10.

4 EXEMPTION/CLEARANCE OF NORM CONTAMINATED OBJECTS

In addition to solid materials like for example sand, slag and sludges the non-nuclear industry also generates disused or re-usable components which are themselves not radioactive but which have radioactive material (in the case of NORM often encountered as scale) attached to their surfaces. In past practice it had shown to be impractical and often impossible to reliably quantify the activity concentration and total activity in the surface contamination for decisions on exemption. Therefore it was investigated if separate exemption levels for surface contamination could be derived for NORM contaminated objects. Council Directive 96/29 does not specify exemption levels for these so-called surface contaminated objects (SCO). Article V of the Council Directive however gives National authorities the possibility to establish clearance levels for materials containing radioactive substances if they follow the basic criteria used in Annex I and take into account any other technical guidance provided by the Community. Exemption values for SCO will probably be specified in a Ministerial Guideline to which is referred in Article 25.7 of BS ("other methods can be appointed to identify damage in those cases that exemption criteria for activity concentrations per unit of mass do not give a proper indication of the damage that handling of these radioactive materials will cause. The characteristics of NORM containing surface contamination and portable measuring equipment which are relevant to the establishment of surface contamination in relation to criteria for NORM material are further discussed below.

5 NORM SCALE CHARACTERISTICS

The radiological aspects of NORM are mainly caused by the long-lived

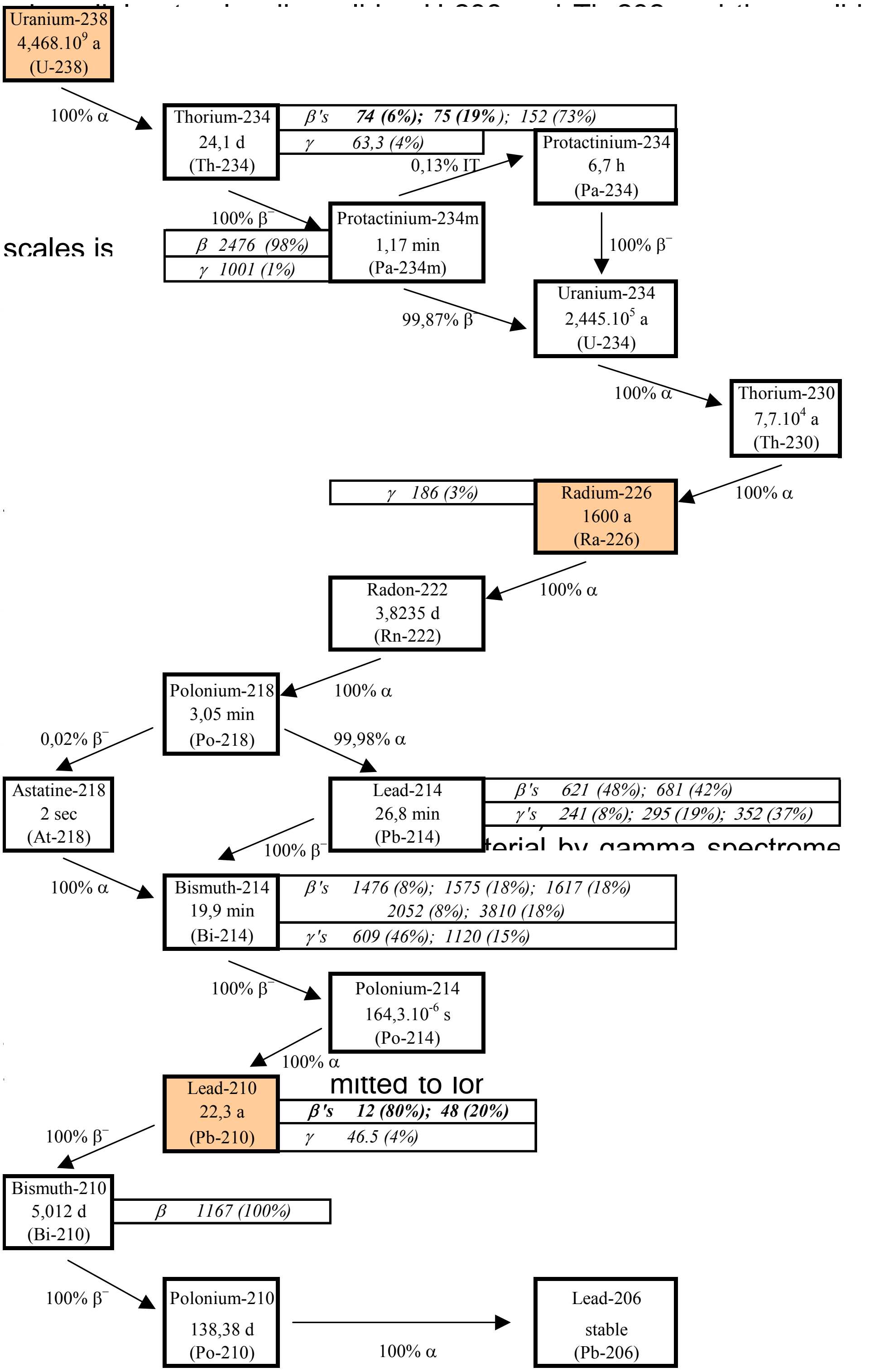

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Fig 1 Decay chain of U-238 including $E_{\beta\,max}$ and $E\gamma$ (keV) of the most important beta particles and gamma photons emitted and emission probabilities (%). The long-lived radionuclides are marked gray.

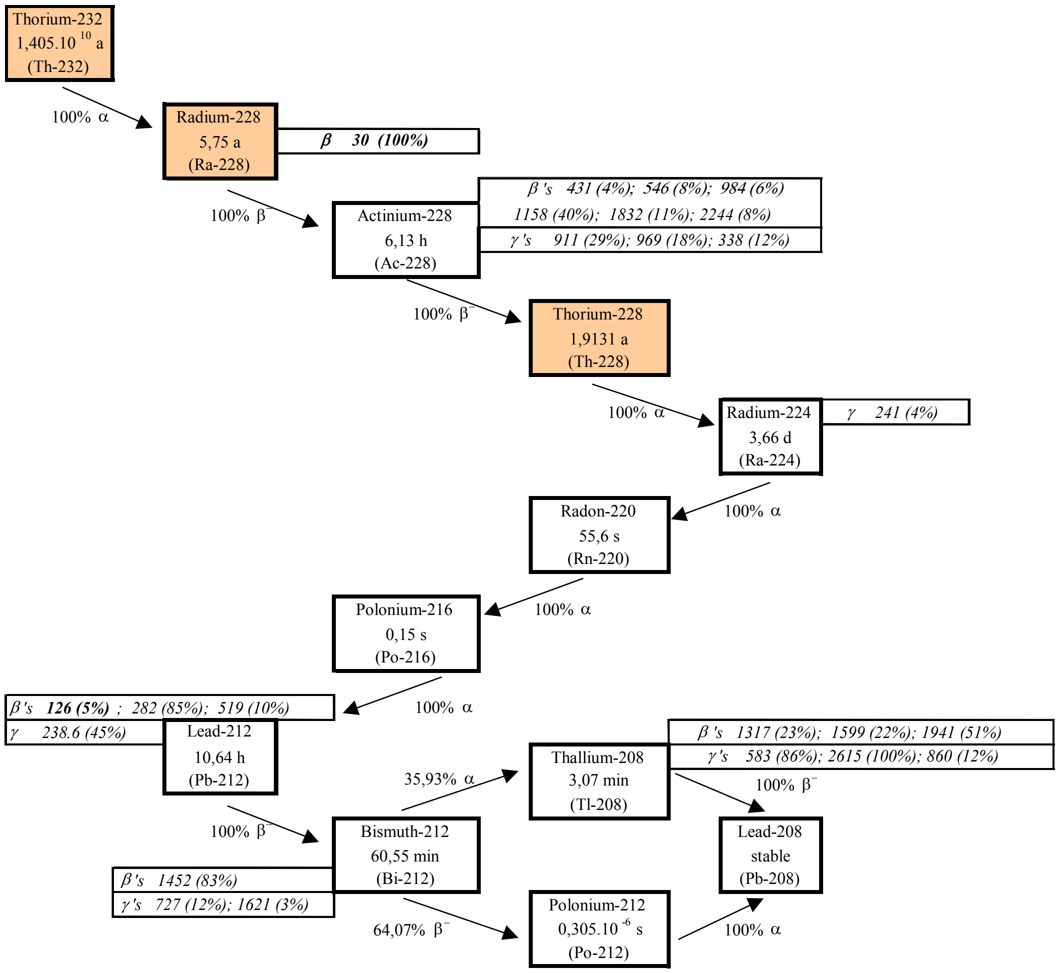

Fig 2 Decay chain of Th-232 including $E_{\beta max}$ and $E\gamma$ (keV) of the most important beta particles and gamma photons emitted and emission probabilities (%).
The long-lived radionuclides are marked gray.

6 CURRENT PRACTICE

At present the Dutch legislation only specifies criteria for specific activity concentration. It is however impractical and often impossible to take (representative) scale samples from inner side of contaminated objects. The current practice is to measure possibly contaminated objects with portable surface contamination monitors measuring beta particles. If the measured count rate exceeds 3x the background value this is considered an indication of the presence of "radioactive" material. As a result the equipment has to be treated as radioactive waste and the installation from which it is derived is considered a NORM contaminated installation, unless it can be proven by means of sampling and analysis that the activity concentration in the scale is lower than 100 Bq/g.

Table 1 Ratio of beta : alpha particle emitters in different types of NORM derived surface contamination

Radionuclide	Half-life radionuclide	Type of emitter	1 Ra-226 deposits (till Pb-206)	2 Ra-226 deposits (till Po-214)	3 Pb-210 deposits (till Pb-206)	4 U-238 deposits (till Pb-206)	5 Ra-228 deposits (till Pb-208)	6 Th-232 deposits (till Pb-208)	7 Pb-scale[1)]	8 SO_4-scale[2)]
U-238		**alfa**				**1**				
Th-234	*24.1 d*	*beta*				*1*				
Pa-234m	*1.17 m*	*beta*				*1*				
U-234	$2.4\ 10^5$ a	**alfa**				**1**				
Th-230	$7.7\ 10^4$ a	**alfa**				**1**				
Ra-226	1600 a	**alfa**	**0.5**	**1**		**1**			**0.05**	**0.25**
Rn-222	3,8 d	alfa (T½ < 10 days)	0.5	1		1			0.05	0.25
Po-218	3.05 m	alfa (T½ < 10 days)	0.5	1		1			0.05	0.25
Pb-214	*26.8 m*	*beta*	*0.5*	*1*		*1*			*0.05*	*0.25*
Bi-214	*19.9 m*	*beta*	*0.5*	*1*		*1*			*0.05*	*0.25*
Po-214	$164\ 10^{-6}$ s	alfa (T½ < 10 days)	0.5	1		1			0.05	0.25
Pb-210[3)]	22.3 a	beta	0.5		1	1			0.85	0.25
Bi-210	*5.0 d*	*beta*	*0.5*		*1*	*1*			*0.85*	*0.25*
Po-210	138.3 d	**alfa**	**0.5**		**1**	**1**			**0.85**	**0.25**
Pb-206										
Th-232	**$1.4\ 10^{10}$ a**	**alfa**						**1**		
Ra-228[3)]	5.75 a	beta					1	1	0.05	0.25
Ac-228	*6.13 h*	*beta*					*1*	*1*	*0.05*	*0.25*
Th-228	**1.9 a**	**alfa**					**1**	**1**	**0.05**	**0.25**
Ra-224	3.66 d	alfa (T½ < 10 days)					1	1	0.05	0.25
Rn-220	55.6 s	alfa (T½ < 10 days)					1	1	0.05	0.25
Po-216	0.15 s	alfa (T½ < 10 days)					1	1	0.05	0.25
Pb-212	*10.6 h*	*beta*					*1*	*1*	*0.05*	*0.25*
Bi-212	*60.6 m*	*beta / alfa (T½ < 10 days)*					*1*	*1*	*0.05*	*0.25*
Tl-208/Po-212	3 m / $0.3\ 10^{-6}$ s	alfa (T½ < 10 days) / beta					1	1	0.05	0.25
Pb-208										
Detectable beta : long-lived alpha particle emitters			**1.5 : 1**	**2 : 1**	**1 : 1**	**5 : 5**	**3 : 1**	**3 : 2**	**1.1 : 0.95**	**1.5 : 0.75**
Detectable beta : (all) alpha particle emitters			1.5 : 2.5	2 : 4	1 : 1	5 : 7	3 : 5	3 : 6	1.1 : 1.3	1.5 : 2.5

1) Pb-scale Pb-210 : Ra-226 : Ra-228 : Th-228 = 0.85 : 0.05 : 0.05 : 0.05. Type of scale that occur frequently in the oil&gas industry.

2) SO_4-scale Pb-210 : Ra-226 : Ra-228 : Th-228 = 1 : 1 : 1 : 1. A second type of scale that is often encountered in contaminated objects derived from the oil&gas industry.

3) Pb-210 and Ra-228 emit beta particles with an $E_{\beta max}$ lower than 150 keV, which are not or with negligible sensitivity detected by commonly used portable surface contamination monitors.

Tabel 2 Overview of a number of different portable surface contamination monitors and their sensitivity in $Bq/cm^2/cps$ for beta particles emitted by various calibration sources

Number	Manufacturer	Type of instrument	Type of detector	Measuring principle	Count rate background (cps)	Sensitive area (cm^2)	$^{90}Sr/^{90}Y$ $E_{\beta max}$ 2260 keV Sensitivity ($Bq/cm^2/cps$)	^{36}Cl $E_{\beta max}$ 710 keV sensitivity ($Bq/cm^2/cps$)	^{60}Co $E_{\beta max}$ 318 keV Sensitivity ($Bq/cm^2/cps$)
1	Automess	6150 AD	6150 AD17	GM	0.1	7	0.7	0.66	1.24
2	Mini Instruments	7.10	ZP-1490	GM	0.1	9	0.5	0.4	0.7
3	Mini Instruments	5.10	DN-212	GM	0.1	21	0.2	0.17	0.37
4	Mini Instruments	900	EP-15f	GM	0.1	21	0.17	0.15	0.29
5	Rados/Herfurth	Microcont	HXE-60R	Xe-proportioneel	4	60	0.15	0.2	0.35
6	Rados/Herfurth	RDS-110	GMP-115	GM	0.5	16	0.13	0.15	0.37
7	Nuclear Enterprise	PCM5	DP2R	ZnS + plast.scint.	0(α) 5(β)	50	0.11		0.47
8	Nuclear Enterprise	Electra	DP2R	ZnS + plast.scint.	0(α) 5(β)	50	0.11		0.47
9	Nuclear Enterprise	Electra	DP6A	ZnS + plast.scint.	0(α) 3(β)	100	0.061	0.067	0.26
10	Nuclear Enterprise	Electra	DP3R	ZnS + plast.scint.	0(α) 4(β)	100	0.058	0.077	0.19
11	Rados/Herfurth	Microcont	RPD 1D4 5x15 S	Plastic scint.	25	65	0.04	0.04	0.08
12	FAG	Contamat	111M	Xe-proportioneel	12	155	0.03	0.03	0.06
13	Automess	6150 AD	6150 AD-k	Xe-proportioneel	0(α) 7(βγ)	170	0.021	0.023	0.042
14	FAG	Contamat	111E	Methaan prop.	4	165	0.02	0.02	0.02
15	Berthold	LB-122		Methaan prop.	0(α) 7(βγ)	205	0.017	0.017	0.022
16	Rados/Herfurth	Microcont	HXE-260/25	Xe-proportioneel	20	260	0.014	0.015	0.050
17	Berthold	LB-122		Xe-proportioneel	15	216	0.020	0.022	0.046
18	Rados/Herfurth	Microcont	HXE-260/10	Xe-proportioneel	20	260	0.012	0.013	0.028

A large number of different portable surface contamination monitors is used in the field. An overview (not comprehensive) is given in Table 2. They vary with respect to measuring principle, sensitivity and background count rates [8]. The beta sensitivity of a detector is determined by the size of the sensitive window of the detector, while the background count rate is more dependent on the volume of the detector.

The use of a surface contamination criterion based on a number of times the background value has a number of disadvantages. Due to the differences in size of the sensitive window and sensitivity of various detectors, a measured count rate of 3x the background corresponds for each detector with a different value of surface contamination present. As shown in Table 3 a count rate of 3x the background value measured with a AD17 probe corresponds with a surface contamination of 0.25 Bq/cm^2, while the same count rate measured with a DP2R probe corresponds with a surface contamination of 4.7 Bq/cm^2. In addition the actual surface contamination present when counts rates of 3x the background are measured will vary dependent on the background value. At sea, where the background is lower than on land, values of 3x the background count rate correspond with surface contamination values that are approximately 3x lower compared to those on land. Another disadvantage of the current situation is that the surface area over which the contamination can be averaged is not officially specified. In practice the counts measured by a particular detector are averaged over the size of the sensitive window. A spot contamination of 20 Bq with beta particles with an $E_{\beta max}$ of 318 keV on 1 cm^2 measured with a PCM5-DP2R (size sensitive window: 50 cm^2) will give a count rate of 1.2x the background value, while the same spot measured with an Automess 6150AD3-6150AD17 (size sensitive window: 7 cm^2) will give a count rate of 24.0x the background value (Table 4).

7 SURFACE CONTAMINATION CRITERIA FOR NORM

Introduction of a surface contamination criterion makes it possible to correct for differences in sensitivity between detectors and background values and to specify the surface area over which the surface contamination can be averaged. Guidelines for the evaluation of surface contamination are given in the international standard ISO 7503-1 [9].

Surface contamination can be evaluated by direct and indirect methods of measurements. As already mentioned in chapter 3 the characteristic of most NORM scale deposits make them less suitable for indirect evaluation of surface contamination by smear test as the majority of the deposits will consist of fixed surface contamination.

Most surface contamination monitors are designed for the assessment of beta particles. They generally have low intrinsic detection efficiencies for photons. Thin window instruments may be sensitive to alpha radiation in addition to beta radiation. A total mass of about 7 mg.cm^{-2} of a low atomic number material will preclude the response to alpha particles of less than about 6.5 MeV [10], which includes the alpha particles emitted by most NORM radionuclides. The source efficiency for alpha-emitters for practical contamination sources, like scales, therefore can easily be very near to zero. Direct measurement of alpha emitters in NORM scales is therefore not feasible.

To determine the activity per unit area the instruments should be calibrated using reference sources of known emission rate per unit area. ISO 7503-1 specifies how the instrument efficiency must be determined and which sources are appropriate for reference sources. As shown in Table 2 the sensitivity of the various detectors (Bq/cm^2/cps) depends on the energy of the beta particles. Ideally, calibration should be conducted with sources of the same radionuclides as anticipated in the field. In cases where this is not possible or impractical radionuclides should be selected with maximum beta energies close to those expected. In figure 1 and 2 the $E_{\beta max}$ of the relevant NORM radionuclides are given. From this figure it is clear that a large number of different beta particles all with a different $E_{\beta max}$ are emitted by any type of NORM derived surface contamination, except pure lead deposits. If the detector sensitivity is determined with Co-60 as calibration source a conservative value for the surface contamination will be obtained for thin layers of scale where source efficiency losses by absorption are considered negligible. If Sr-90/Y-90 is used as a reference source to determine the detector sensitivity the surface contamination will be underestimated. The value for surface contamination in Bq/cm^2 is obtained by multiplying the net counts measured by the sensitivity of the detector in Bq/cm^2/cps. For NORM deposits this value can be used as a measure for the long-lived alpha radionuclides present.

8 CONCLUSIONS AND RECOMMENDATIONS

For surface contamination measurements a large number of different portable surface contamination monitors can be used. Introduction of a surface contamination criterion as opposed to the current practice based on a criterion of 3x the background value makes it possible to correct for differences in sensitivity and background values of the various detectors. This will resolve some of the problems surface contamination measurements have given in the past.

Portable contamination monitors currently in use measure beta particles. In addition to beta-emitters NORM material also contains long-lived alpha emitters.The number of measurable beta particles ($E_{\beta max}$ > 150 keV) emitted by NORM deposits is always higher than the number of long-lived alpha emitters present in these deposits. By assuming a conservative ratio of 1:1 beta particle measurements can be used to estimated the contamination with long-lived alpha emitters.

The calibrations performed with reference sources usually are carried out under ideal circumstances e.g.: optimal geometry, short distance between source and detector, optimal source efficiency and long counting times. In the field measuring conditions will always be less optimal. Guidelines will have to be given to determine how to deal with these variables.

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Tabel 3. Sensitivity for surface contamination with beta-emitters with an energy comparable to Co-60 of three types of detectors with a flat window under conditions of high background (land) or low background (sea)

Manufacturer	Type of instrument	Type of detector	Count rate background on land (high) or sea (low) (cps)	Sensitive area detector (cm^2)	^{60}Co sensitivity ($Bq/cm^2/cps$) $E_{\beta max}$ = 300 keV	Equivalent surface contamination of background countrate Bq/cm^2	Surface contamination in Bq/cm^2 at a count rate equivalent to 3x the background value
High background conditions (land)							
Automess	6150 AD	6150 AD17	0.1	7	1.24	0.124	0.248
Nuclear Enterprise	PCM5	DP2R	5	50	0.47	2.35	4.7
Rados/Herfurth	Microcont	HXE-260/25	20	260	0.050	1.00	2.0
Low background conditions (sea)							
Automess	6150 AD	6150 AD17	0.03	7	1.24	0.04	0.08
Nuclear Enterprise	PCM5	DP2R	1.7	50	0.47	0.78	1.6
Rados/Herfurth	Microcont	HXE-260/25	6.7	260	0.050	0.34	0.7

Tabel 4. Comparison of the respons of three types of detectores for a local spot contamination of 20 Bq on 1 cm^2 with a beta-emitters with an energy compared to Co-60 under condition of high background (land) and low background (sea)

Manufacturer	Type of instrument	Type of detector	Count rate background on land (high) or sea (low) (cps)	Sensitive area detector (cm^2)	^{60}Co sensitivity ($Bq/cm^2/tps$) $E_{\beta max}$ = 300 keV	Local contamination on 1 cm^2 (Bq)	Mean contamination below detector window Bq/cm^2	Count rate (cps)	Reading as multiple of background
High background conditions (land)									
Automess	6150 AD	6150 AD17	0.1	7	1.24	20	2.9	2.4	24.0
Nuclear Enterprise	PCM5	DP2R	5	50	0.47	20	0.40	5.9	1.2
Rados/Herfurth	Microcont	HXE-260/25	20	260	0.050	20	0.08	21.6	1.1
Low background conditions (sea)									
Automess	6150 AD	6150 AD17	0.03	7	1.24	20	2.9	2.3	70.1
Nuclear Enterprise	PCM5	DP2R	1.7	50	0.47	20	0.40	2.5	1.5
Rados/Herfurth	Microcont	HXE-260/25	6.7	260	0.050	20	0.08	8.3	1.4

9 REFERENCES

1. Council Directive 96/29/EURATOM of 13 may 1996 laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation, Office journal L159, 29 june 1996.

2. Ontwerp-Besluit Stralingsbescherming, Implementatie van de Euratomrichtlijnen 96/29/Euratom en 97/43/Euratom, versie 20 april 2001.

3. Timmermans, C., Weers, A.van, Inventarisatie van werkzaamheden met blootstelling aan natuurlijke stralingsbronnen. Onderzoek verricht in opdracht van het ministerie van Sociale Zaken en Werkgelegenheid, Publicatie No. 121, mei 1999.

4. Timmermans,C., Scenarios and reference levels for the disposal and reuse of large quantaties of residues from the non-nuclear industry, 22727-NUC 97-9002, 1997.

5. Timmermans, C., Conditionele vrijgavegrenzen voor toepassing als bouwmateriaal van reststoffen van de niet-nucleaire industrie, 22982-NUC 98-5308, 1999.

6. Steenhof, V.J.M., Opslagmogelijkheden voor zeer laag radioactief afval in Nederland, DHV, 1999.

7. Sources and effects of ionising radiation, United Nations Scientific Committee on the effects of atomic radiation, UNSCEAR 1993 Report to the General Assembly, with Scientific Annexes, United Nations, New York, 1993.

8. Meijne, E., Van Weers, A., Verhagen, H and Kok, M., Radioactieve oppervlaktebesmetting. Een inventarisatie van normen voor meting van oppervlaktebesmetting en van vrijgave-criteria en een evaluatie van hun toepassing in de praktijk, 21516/00.37027/C, 2000.

9. ISO 7503-1:1988 (E). Evaluation of surface contamination - Part 1: Beta emitters (maximum beta energy greater than 0,15 MeV) and alpha emitters.

10. Calibration of survey instruments used in radiation protection for the assessmetn of ionizing radiation fields and radioactive surface contamination, NCRP report no. 112, 1991.