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Characterisation of NORM Contaminated Objects: Reliable & Efficient

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Abstract

The predominant contributors to the production of Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) and NORM-waste in the Nordic countries are the on- and offshore oil and gas producers. In oil and gas production processes, host rock formation water containing low concentrations of NORM is mixed with seawater containing high concentrations of sulphate. This leads to precipitation of NORM (Ra, Pb, Po)SO₄, which is deposited as either scale or sludge in the production equipment. NORM contaminated pipes, tubes, pumps and tanks, etc. are therefore subject to radiological characterization in order to ensure safe reuse or recycling as well as safe NORM waste handling. The procedures and measurement techniques may significantly affect the amount of material that is categorized as NORM contaminated equipment and NORM waste. At present, different procedures are used for categorization by the individual operators in the North Sea. A number of parameters considerably influence the measurement accuracy and may cause false categorization of the materials, for instance abnormal geometry of objects, various density and composition of the scale and sludge, inhomogeneous distribution of NORM, and measurement sensitivity and uncertainty. Incorrect characterization can however be prevented by the use of well certified methods and inappropriate handling of these materials can be avoided. The current report is partly a review of existing methods to perform initial characterization of NORM contaminated equipment and partly an experimental section dealing with the basic investigations required to evaluate factors affecting external dose rate measurements.

Key words

NORM, TENORM, Scaling, Oil & gas industry, radium, ²²⁶Ra, ²²⁸Ra, dose-rate

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Interim Report for the NKS-B CONCORE activity 2014

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Table of contents

	Page
1. Introduction	1
2. TENORM containing scale in the oil and gas industry.	4
3. Experimental characterization of scale-containing pipes.	6
3.1 Materials.	6
3.2. Scale homogeneity.	8
3.3. Emanation of radon from scale.	11
4. Results	11
4.1 Cleaned tubes.	11
4.2 Inhomogeneity of activity in contaminated tubes.	12
4.3 Emanation of radon from scale.	14
4.4 Geometrical parameters governing external dose-rate from investigated steel-pipes	15
5. Practical methods for characterization of NORM material	17
5.1 Characterization methods	17
5.2 Characterization using a dose rate meter (DRM)	18
5.3 Characterization using a contamination monitors (CM)	18
5.4 Characterization with the “puck” method	18
5.5 General comments	19
6. Conclusions	26
7. References	27
Appendix 1. Measurement instruments and characterization instructions	29
Appendix 2. Implementation in existing Nordic NORM management guidance	32
Appendix 3. Application of project results	33

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1. Introduction

The new BSSD (Euroatom Basic Safety Standards Directive), adopted by the member states in January 2014, further emphasize radiation protection issues in relation to NORM relative to the old standards. The reason for this is a growing consensus that natural radioactivity actually leads to significant doses to workers in a variety of industries. Table 1 presents a rough overview of average doses to workers in a variety of industries. The table shows that doses associated with NORM work are of equal magnitude - or even higher - than doses in industries, that traditionally are associated with risk of doses to employees.

Table 1. Worldwide average doses to workers in a variety of industries. (December 2014, IAEA Conference on Occupational Radiation Protection).

Medical	0.5 mSv/y
Industrial	1 mSv/y
Nuclear	1.5 mSv/y
Air crew	2 mSv/y
NORM	0.5-5.5 mSv/y

The wide range of doses to NORM workers - shown in Table 1 - is reflecting the fact that NORM industries are very diverse in terms of NORM quantities, activity concentration, isotopes, and primary exposure pathway. Within the NORM field exposure to radon in mines is by far the most significant dose contributor. But oil and gas industry also gains attention, as the amounts of NORM waste are large and the activity concentrations in oil and gas NORM can be significant. An important task in the implementation of the new BSS in national legislation is to balance the desire to minimize the quantities of radioactive waste and the need to protect workers, the public and the environment. The fact that NORM have such a high priority in the new BSS, emphasize the need to develop tools that can assist to maintain a safe working environment at NORM companies while keeping the amounts of NORM waste down.

Within the oil and gas industry the first step in the practical management of NORM is the characterization or screening of potential NORM contaminated equipment. The same applies to other industries where wet processes cause the precipitation of NORM in pipes, tanks and other process equipment. The main objective of this project is to develop a reliable and efficient method for the characterization of NORM contaminated equipment which may be applied in various enterprises such as oil/gas production, geothermal energy utilization, tree pulp processing, water treatment, fertilizer production, mining industry and others.

As an additional benefit this project can contribute to the Nordic EU countries' efforts to meet the requirements of the new BSS, which deals with the management of NORM.

Relevant references concerning the problems with NORM in the oil and gas industry may be found in IAEA (2003a, 2003 b) and van Weers et.al. (1997).

2. TENORM containing scale in the oil and gas industry.

The salty solutions frequently contained in reservoirs of oil and gas is known as "formation water". Group II cations, originally present in the reservoir bedrock, dissolve in the brine solutions through ion exchange processes thus making elements like calcium, strontium, barium and radium relatively abundant in the formation water. During drilling, a mixture of oil, gas, and formation water is pumped to the surface. The water is separated from the oil and gas into tanks or pits, where it is referred to as "produced water." Due to drops in pressure and temperature the solubility products of the group II mixed carbonates and sulphates may become exceeded which causes their precipitation as sulphate and carbonate scale on the inner walls of tubulars, valves, pumps and other drilling equipment. The deposition is particularly pronounced where the turbulent flow and centripetal forces are high but is also enhanced on surfaces where particles (acting as nucleation sites) deposit. Ordinary sea water may mix with the produced water thereby increasing the sulphate and carbonate concentrations thus further decreasing the solubility of the group II cations.

The origin of radioactivity in the scale is from the radioisotopes in the three decay chains (^{238}U , ^{235}U and ^{232}Th decay chains).

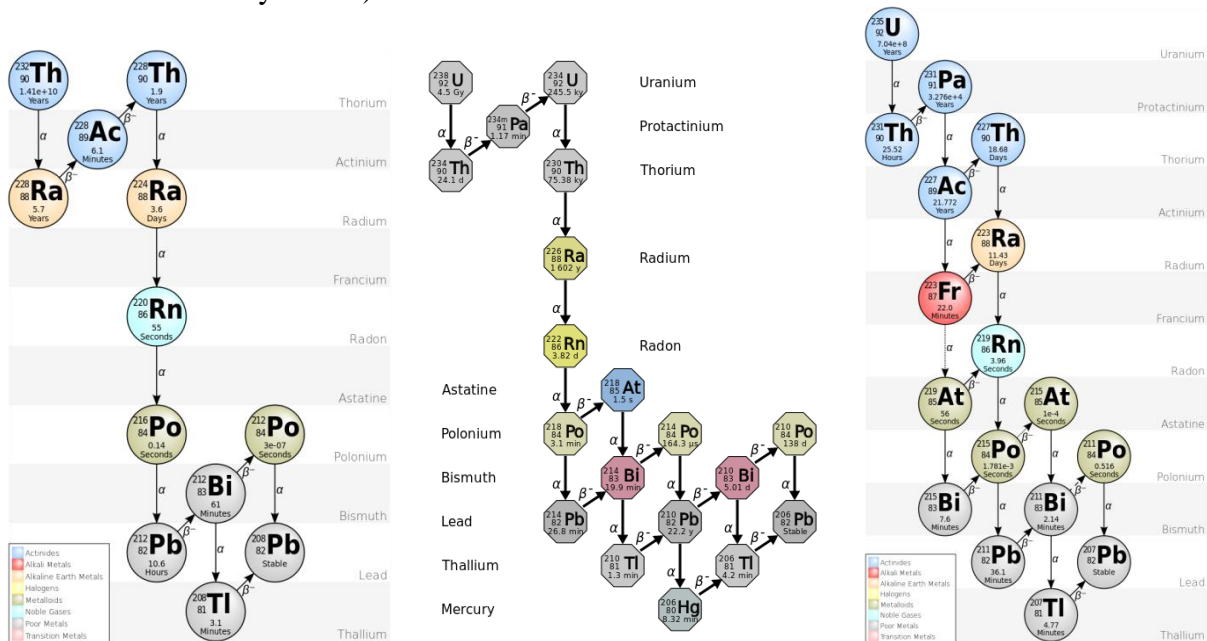


Figure 1. The three natural decay chains ^{232}Th , ^{238}U and ^{235}U (Source: Wikipedia).

Since the uranium and thorium only at a limited extent are dissolved in the formation water the main isotopes of importance are the radium isotopes ^{226}Ra and ^{228}Ra . Due to the combined action of solubility, half-life and formation water residence time in the reservoir the composition of radium isotopes and their daughter products in the formation water may vary.

The on- and offshore oil and gas producers, especially in Denmark and Norway are the predominant contributors to the production of Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) and NORM-waste in the Nordic countries, although other industries such as metal foundries using zircon sand, fertilizer industry, building material producers also produce TENORM waste. TENORM contaminated pipes, tubes, pumps and tanks, etc. are therefore subject to radiological characterization in order to ensure safe reuse or recycling as well as safe NORM waste handling. As part of the normal operation, scales, deposits and sludge is removed from the process equipment at regular intervals to be able to either reuse them or recycle the metals. However, the concentration of NORM in some treated materials might still exceed the so-called exemption levels, that set the limit at which they must be treated as radioactive material. The annual production of this type of TENORM from the North Sea oil and gas producers is several tens of tons. As the relevant nuclides (Ra, Pb, Po) are characterised by very high dose coefficients upon intake, it is in the interest of operators, authorities as well as the society in general to ensure reliable and correct characterization and handling of material that exceeds the exemption levels. Moreover, decommissioning of oil and gas rigs is anticipated to contribute several hundreds of tons of NORM-waste in the near future. Storage and final disposal of such volumes is pending and it is thus in the interest of the same stakeholders to minimize the volumes of NORM-waste that is going to be placed in onshore repositories. Although NORM reinjection in boreholes and incineration of certain types of NORM material could be possible, the bulk of the produced NORM-waste must be expected to end up in national repositories.

The procedures and measurement techniques may significantly affect the amount of material that is categorized as NORM contaminated equipment and NORM waste. At present, different procedures are used for categorization by the individual operators in the North Sea. A number of parameters considerably influence the measurement accuracy and may cause false categorization of the materials, for instance abnormal geometry of objects, various density and composition of the scale and sludge, inhomogeneous distribution of NORM, and measurement sensitivity and uncertainty. Incorrect characterization can however be prevented by the use of well certified methods and inappropriate handling of these materials can be avoided.

A number of parameters considerably influence the measurement accuracy and may cause false categorization of the materials, for instance abnormal geometry of objects, various density and composition of the scales and sludges, inhomogeneous distribution of NORM, and measurement sensitivity and uncertainty. Mis-characterization and inappropriate handling of these materials might happen due to lack of well-certified methods. Parameters that may have a significant effect on the characterisation are:

- Density and composition of the scales and sludges (shielding and self-absorption)
- The absorbed water content of the material (shielding and self-absorption)
- The sample shape (complex object geometry)
- The measurement geometry (distance, time, shielding)
- The distribution of NORM (inhomogeneity)
- Instrument capabilities (technical suitability)
- Measurement sensitivity and uncertainty

Moreover, the authorities in all North Sea countries agree that all measurements should be performed on wet material in order to prevent the spreading of dust from dried out scale etc. and hence the risk of inhalation of aerosols. This procedure presents a problem for the characterization, since wet measurements introduce false negatives due to the shielding effect of water. Items that were originally characterized as negative by wet measurements, are frequently found to be positive by subsequent dry measurements, for example at recycling industries or by simple reuse by the original operator. In this way false negative readings lead to the spreading of radioactive material into the environment. It is therefore more than likely that relevant legislative provisions and conventions are compromised. False negatives find their way into reuse at rigs or in the metal recycling industry, putting workers at risk of internal radiation doses from high dose coefficient nuclides. On the other hand false positives are treated as radioactive waste, taking up space in NORM waste repositories.

Establishment of a reliable, efficient and commonly acceptable method for characterisation of NORM contaminated objects is not only an interest of NORM producers, but also become an urgent requirement of the authorities and the society to ensure relevant, safe and cost efficient management of NORM waste in the future.

This current project is anticipated to provide a common Nordic, scientifically based set of recommendations for practicable and reliable characterization of NORM contaminated objects. The project is suggested to focus on oil/gas industry NORM, but could be extended to establish recommendations for other industrial NORM from mining, metal and fertilizer industries. The initial phase of the project is envisaged to comprise: a) a primary compilation of existing procedures for characterization of NORM-contaminated equipment – carried out by the Nordic regulatory authorities, b) assembling of a detailed overview of the type distribution of NORM-contaminated items, c) selection and collection of types and numbers of specific items to be used in the project, and d) establishment of a detailed analytical plan to be presented to - and adjusted - with all project partners before implementation.

3. Experimental characterization of scale-containing pipes.

3.1 Materials

Although the total number of tubes used in the oil and gas industry is huge their size (length tens of metres) and the long deployment times (months to years) means that the number of available tubes with suitable dimensions at any given time may be limited. During the present project tubes were available first when a section of pipes needed to be replaced.

A set of steel tubes was obtained from a Danish NORM decontamination company. The original 2 tubes had at the company been cut into 4 pieces each, i.e. a total of 8 steel tube samples. Four of these were cleaned according to the company procedures while the other four were untreated but two of the tubes were cut along their length which had made the scale layer to become detached to the tube walls. A picture of the four cleaned tubes is seen in figure 2. In the initial information

given (as seen from the attached paper in figure 2) the tubes received were supposed to come from the same original tube (10m long) but later information revealed that they all were of different origin.



Figure 2. Steel pipes cleaned (descaled) from NORM by a decontamination company. The paper sheet shown on top of the tubes in the left picture shows the supposed cutting of the original tubes and the numbering of them, however later information revealed that the individual pipes were of different origin. Cleaned pipes were numbered No 2,4,6 and 8 while still contaminated pipes were No 1,3,5 and 7.

Two different dimensions of the tubes with respect to inner diameter and wall thickness were originally supposed to be delivered but only one dimension was obtained with a tube inner diameter of 75mm and a wall thickness of 7mm. The length of the tube samples were 60 and 80cm.

NORM containing pipe samples were packed in plastic bags and loose material was found to be present in some of the tubes. Tube No 7 was selected for further examination and was first investigated for outside contamination by swipe testing but were found clean. In the documents associated with the tubes a scale thickness of 1mm was indicated. From visual inspection (figure 3) this was found to be reasonable. Gently scraping of the scale with a knife showed that the outer part of the scale consisted of a relatively loose brown colored material. Analysis of the material by gamma spectrometry showed it to be of relatively low ^{226}Ra -activity, therefore assumed to be rust.



Figure 3. Visual appearance of the interior of a non-cleaned tube. The scale-thickness of the tube (tube No 7) was approximately 1mm.



Figure 4. Tube cut along its axis with loose scale material inside.

3.2 Scale homogeneity

Initial test on tube No 7 was to find out about scale homogeneity, or more correctly the spatial variations in surrounding gamma flux from the tube at a given distance tube to detector. Such variation may be due to different thickness of the scale material in the tube or due to varying concentrations (Bq/g) within the scale. Large variations in exposure along the tube axis, or when rotating around the tube would set higher demands on the procedure required for dose rate reading using dose meters. Due to the low excess dose-rate measured above background from the particular tubes and the limited access to high quality dose-rate readers during the experiment

period, gamma spectrometry rather than dose rate readings were used in this investigation. In any case gamma spectrometry would be required in the following calculations on external exposure.

The tube was placed in front of a HPGe-detector (figure 5) at 20cm distance and rotated 90 degrees between measurements to obtain a rough information on inhomogeneity, if any. Apart from the rotational homogeneity also the upper and lower parts of the tube was compared. The tube wall dimensions (7mm thickness) were such that they posed only limited attenuation for most intermediate and high energy gammas and the actual scale inhomogeneity would therefore be difficult to view during rotation of the tube. Variation in primary gamma emission as recorded by the HPGe-detector would however be larger than the exposure variation since scattered gammas would tend to even out the variation in dose rate caused by scale inhomogeneity.

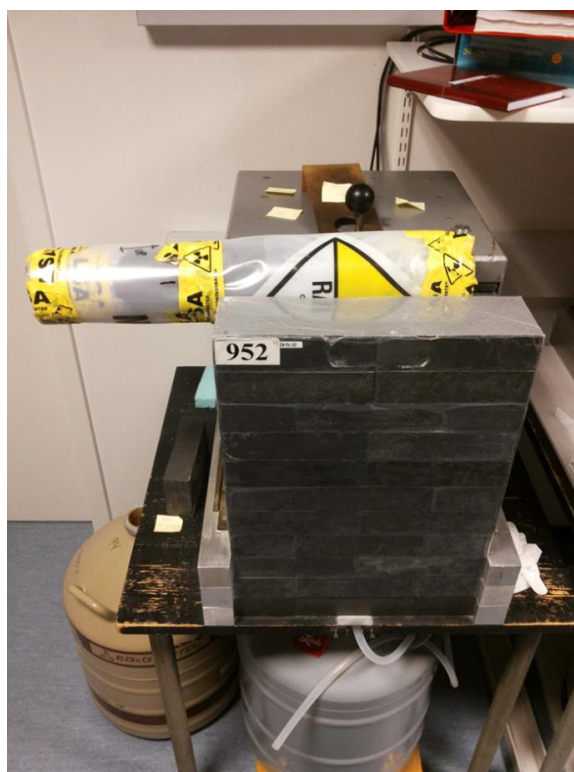


Figure 5. Experimental set-up to obtain a rough view of the exposure variation from tube No7. The central parts of the tube was analysed by rotating the tube 90° between measurements. The lower and upper end of the tube was also compared to each other.

To gain further information on spatial inhomogeneity of the activity in the tubes a 2"x2" NaI(Tl)-detector was placed in a simple lead cave with a 1 cm collimated opening (see figure 6). With a significantly better spatial resolution due to the collimation inhomogeneity, if any, would be more clearly seen using this set-up.

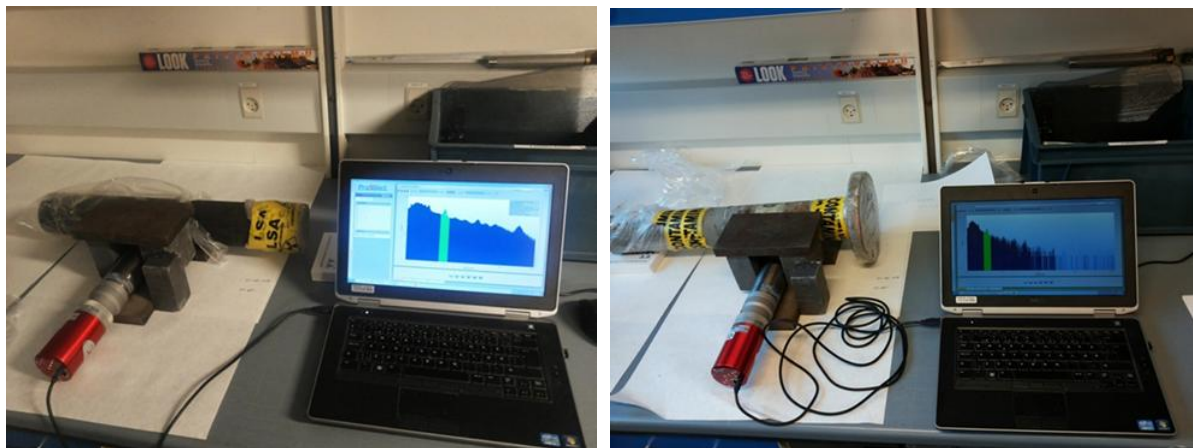


Figure 6. Inhomogeneity of received tubes investigated using a collimated 2"x2" NaI(Tl)-detector. The 609 keV line from ^{214}Bi was used for the survey.

Following gamma spectrometric measurements of the tube, scale was scraped from the interior of the tube. An approximately 10 cm^2 area was scraped using a simple knife (figure 7). The material was weighed and placed in a test tube and analysed by gamma spectrometry.

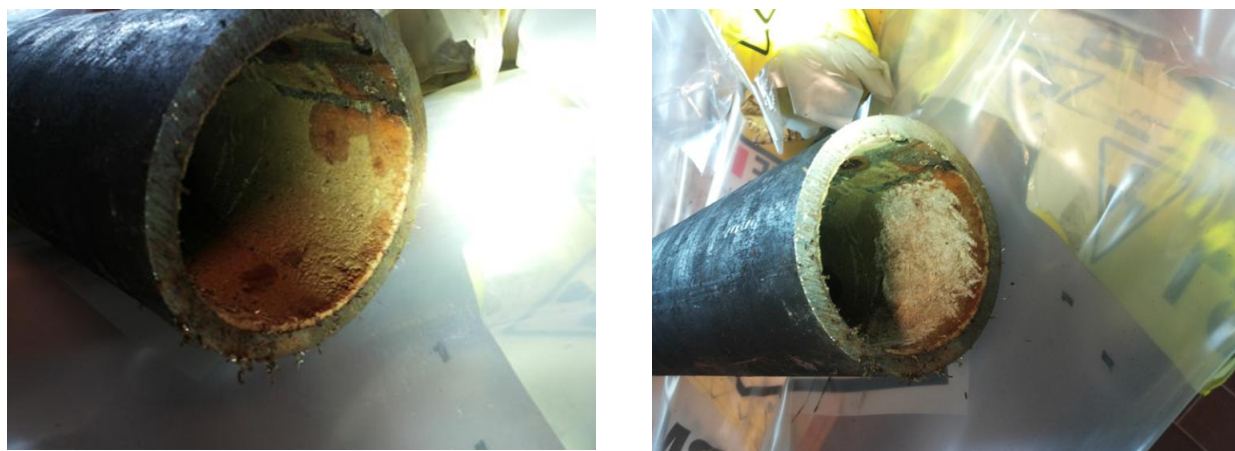


Figure 7. Tube before and after removing the upper rust layer covering the white $\text{Ba}(\text{Ca})\text{SO}_4$ scale material. The area of the scraped scale was approximately $2 \times 5\text{ cm}^2$. Detailed composition of the scale remains to be determined.

3.3 Emanation of radon from scale.

More than 80% of the external dose-rate from the tubulars described above originate from the short-lived ^{222}Rn daughters continuously generated in the scale material due to the presence of ^{226}Ra - ^{222}Rn . Any losses of radon from the scale material thus make a major impact in the exposure rate. Radon gas emanation, or the fractional escape of radon, from any material is a function of several factors and may be expected to be dependent on the scale composition, texture, moisture, air pressure and temperature. In the present project radon emanation from the scale was initially not planned but due to repeated observations on variations in external exposure rate reported by various companies it was decided to perform some preliminary investigations.

- a) The emanation may be expected to be dependent on if the scale material is left untouched in the tubes or if it is removed, crushed into smaller pieces thereby increasing the total surface to volume exposed to air. Experiments designed to obtain emanation from a specific material should thus aim for a minimum of alteration in order to mimic realistic conditions. In order to meet these conditions a tube with the original scale untouched was sealed with plastic and aluminum foil and left for 1 month before ejecting 2.5 ml of the internal gas volume using a syringe and a needle. The needle was gently removed and the syringe sealed using aluminized plastic and placed in a well-type HPGe in order to quantify radon daughters. A decrease in the ^{214}Pb and ^{214}Bi gamma-lines faster than expected (effective half-time of roughly 2h) from radon decay showed that the syringe was leaking. Repeated extraction of the internal radon gas from the sealed tube showed an even faster escape of radon from the syringe.
- b) A known amount of scale removed from one of the tubes was placed in a glass-flask equipped with a rubber cork and allowed to stand for 1 week before sampling using a syringe and needle as described above. In order to minimize pressure differences in the glass-flask only 0.5ml of air was withdrawn.

4. Results

4.1 Cleaned tubes

An important observation is that tubes cleaned by the descaling process appears to be clean as judged from both dose-rate readings and gamma spectrometry. Measurements by gamma spectrometry were all indistinguishable from the ambient background for the four tubes investigated.

4.2 Inhomogeneity of activity in contaminated tubes

Homogeneity of the gamma flux recorded from tube No 7 as shown in figure 5 is presented in table 2.

Table 2. Gamma flux (cps) variation for various gamma lines originating from the short-lived ^{222}Rn daughters ^{214}Pb and ^{214}Bi as well as from the short-lived ^{228}Ra daughter ^{228}Ac . All results are in counts per second divided with the fractional branching for each gamma line. The decreasing numbers with increasing energy reflects the decreasing efficiency of the detector.

	214Pb	214Pb	214Bi	214Bi	214Bi	214Bi		228Ac	228Ac
	295 keV	352 keV	609 keV	1120 keV	1238 keV	1765 keV		338 keV	968 keV
POSITION-1	22,8	20,8	16,8	13,1	13,6	9,8		1,8	0,9
POSITION-2	21,3	19,9	15,5	12,7	12,3	9,6		2,2	0,5
POSITION-3	21,8	20,9	16,5	11,8	12,9	9,6		1,5	0,8
POSITION-4	20,4	19,1	16,2	12,2	12,7	9,5		1,5	0,6
Average	21,6	20,2	16,2	12,5	12,9	9,6		1,8	0,7
1 stdev	1,0	0,9	0,5	0,6	0,6	0,1		0,3	0,2
% stdev	4,6	4,3	3,3	4,6	4,4	1,3		16,6	24,9

From table 2 it is obvious that the recorded inhomogeneity when analyzed in this way is insignificant. For the ^{226}Ra daughters the observed inhomogeneity is less than about 5% and corresponding dose-rate measurements should be even less. The concentration of ^{228}Ra is significantly less than for ^{226}Ra resulting in larger statistical fluctuations during the measurements. This is reflected in the larger apparent spatial variation for the ^{228}Ac lines. The $^{228}\text{Ra}/^{226}\text{Ra}$ ratio as estimated from the current measurements using gamma lines of approximately the same energy (i.e. ^{228}Ac 338 keV and ^{214}Pb 352 keV) results in about 7-8%.

It is worth noting that the $^{208}\text{Tl}/^{228}\text{Ac}$ ratio as judged from the 860 keV and 911 keV lines respectively indicate a $^{228}\text{Th}/^{228}\text{Ra}$ ratio of less than 10%. Since the ^{208}Tl high energy gamma line at 2614 keV contributes significantly to the Th-series gamma dose rate this will increase with time as a consequence of build-up of ^{228}Th from ^{228}Ra .

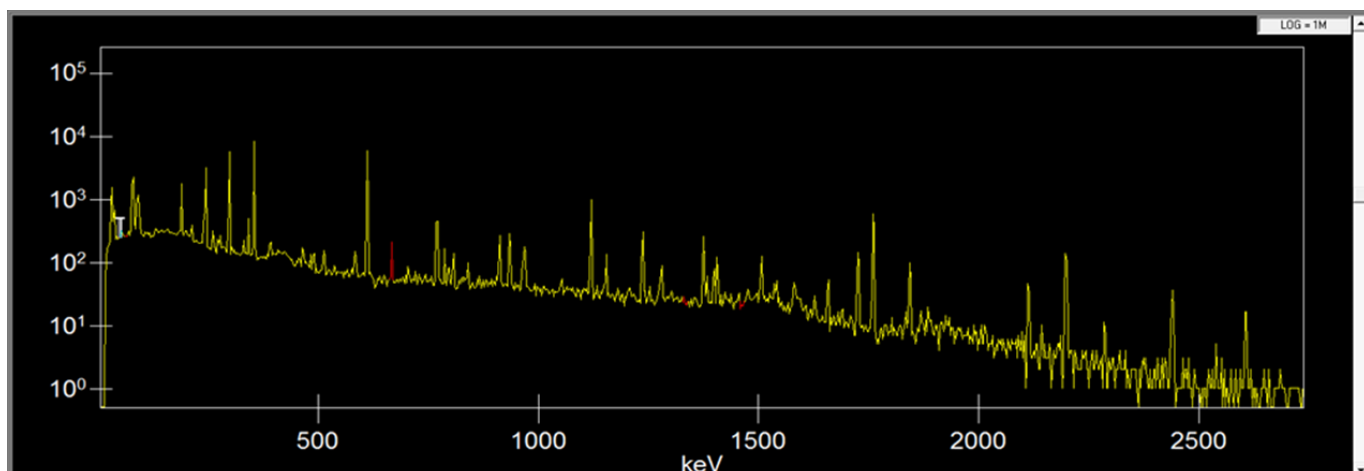


Figure 8. Gamma spectrum recorded from tube No7. Nearly all the gamma lines originate from the short-lived ^{226}Ra daughters ^{214}Pb and ^{214}Bi . Remaining gamma lines are dominated by the ^{228}Ra decay products, mainly ^{228}Ac and ^{208}Tl .

Of the 4 tubes obtained with remaining scale (untreated tubes) only two had sufficient levels of activity to be able to scan using the NaI-detector. For the other two tubes levels of scale were close to background levels (tube No 3 and 1A had count rates of 0.1-0.3 cps and below 0.1cps respectively) and were therefore not elaborated upon further. Results obtained by scanning the tubes using the NaI-detector showed that spatial inhomogeneity was of minor importance.

Table 3. Results of gamma-readings of the 609 keV ^{214}Bi line from tube No 7 and from an unlabeled tube obtained using a collimated 2"x2" NaI(Tl)-detector.

<u>Tube-7</u>	<u>NaI-cps 609 keV</u>		<u>Unlabelled tube with collar</u>	<u>NaI-cps 609 keV</u>
	25,34			2,35
	26,43			2,34
	23,42			1,98
	23,13			
	21,62			
	22,83			
	23,45			
	26,03			
	27,08			
	22,32			
Average cps (609 keV)	24,17		Average cps (609 keV)	2,22
1 Stdev	1,89		1 Stdev	0,21
%Stdev	7,84		%Stdev	9,48

Loose scale material in 3g portions from two different tubes were analysed in a well-calibrated geometry on a HPGe-detector. Material from the above analysed tube No 7 were previously determined to have a ^{226}Ra concentration of 91 kBq/kg. ^{228}Ra and ^{228}Th in the same sample was determined to be 9.0 and 3.6 kBq/kg respectively.

Table 4. Results from analysis of 3g portions of scale material from tube No2. Previous analysis of material from tube No 7 resulted in a ^{226}Ra concentration of 91 kBq/kg.

	Cps/g	Cps/g	Cps/g
Sample	^{226}Ra (352 keV)	^{228}Ra (911 keV)	^{228}Th (2614 keV)
Tube-7 (Det7-Rør-C) 91 kBq/kg ^{226}Ra	18,1047	0,4598	0,0436
Tube-2 NoA	18,4569	0,4277	0,0508
Tube-2 NoB	18,0529	0,4381	0,0513
Tube-2 NoC	18,0060	0,4314	0,0479

In conclusion it seems like the level of inhomogeneity is in the order of 10% or less which indicate that calculations of external dose rates may be done realistically assuming homogeneous activity distributions in the scale along the pipes.

4.3 Emanation of radon from scale

Results obtained by sampling of radon gas inside the sealed tube using a syringe must be considered relatively uncertain firstly due to difficulties in verifying if the sealed steel-pipe was air tight during the month of sealing. Temperature and pressure changes may induce gas exchange even though both ends were sealed. Secondly the sampling of radon gas using a syringe and its transfer to the well detector were associated with leakage of radon from the syringe. Both factors would underestimate the calculated release of radon from the scale. Given the amount of ^{226}Ra in the pipe as determined by the concentration (91 kBq/kg) and the total weight of scale in the tube as determined by the thickness (1mm) and the surface area and density (3.5 g/cm^3) the measured ^{222}Rn of 0.3 Bq/cm³ out of a total tube volume of 2450 cm³ results in an approximate radon emanation of 1.5%.

Analysis of radon extracted by the 0.5ml gas volume out of a 100ml volume glass-flask with 15g of scale resulted similarly in 10% radon emanation. A fraction which is considerably higher than the corresponding emanation from the tube but due to large uncertainties in both experiments this needs to be confirmed.

To conclude radon gas is emanating at a measurable rate from the scale present in the tubes. The magnitude is still to be determined. The consequence of radon emanation is the associated uncertainty in making exposure determinations using external hand instruments. The uncertainty may be insignificant but should be investigated further. A further consequence is the resulting potential variations in external dose rate when measuring the same pipes during wet and dry conditions. Radon gas emanation is usually found to be much larger in dry material relative to wet material. To minimise the problems in determining scale concentrations (Bq/kg) based on external gamma-ray measurements or hand held dose rate instruments the pipes internal part should at all times be kept slightly wet, or alternatively become wetted a few hours before conducting the measurements (radon daughter build-up time to 90% equilibrium is approximately 3h).

The impact of thoron (^{220}Rn) emanation from the same scale may be considered less relevant in this case due to the factor of thirty lower ^{228}Th and the short half-life of thoron (55s) lowering the probability of emanation before decaying.

4.4 Geometrical parameters governing external dose-rate from investigated steel-pipes

Important parameters governing the dose rate from a steel tube at a given source strength (Bq/cm^2 of the scale material) are:

Tube length
 Tube wall thickness
 Distance from tube
 Scale thickness and density.

The last parameter (scale thickness and density) is only of importance for very significant scale, i.e. when constituting a significant fraction of the equivalent thickness of the steel wall.

Figures below show the dependence on dose rate of these parameters at a fixed source strength of ^{226}Ra + daughters (assumed to be in secular equilibrium). Calculations were done using Microshield software (Grooves) and include dose-buildup due to scattered gammas in the material and air. Tube diameter were in all calculations from the tubes investigated above, 75mm.

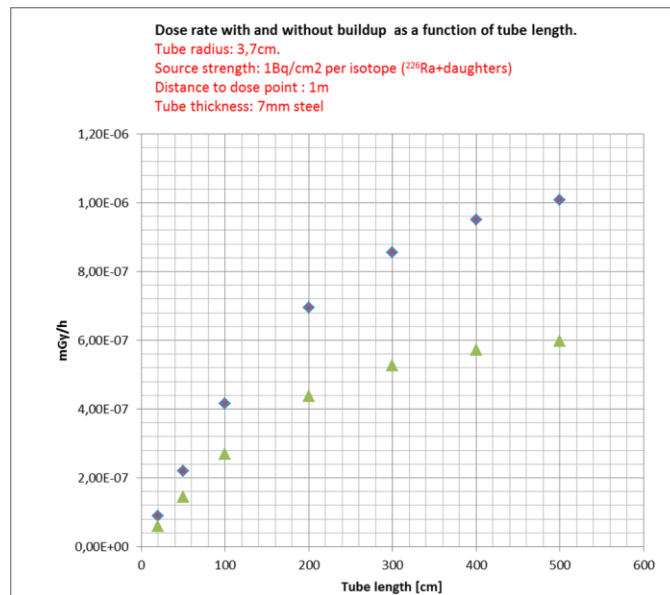


Figure 9. Dose rate as a function of tube length at 100cm distance from axis of tube.

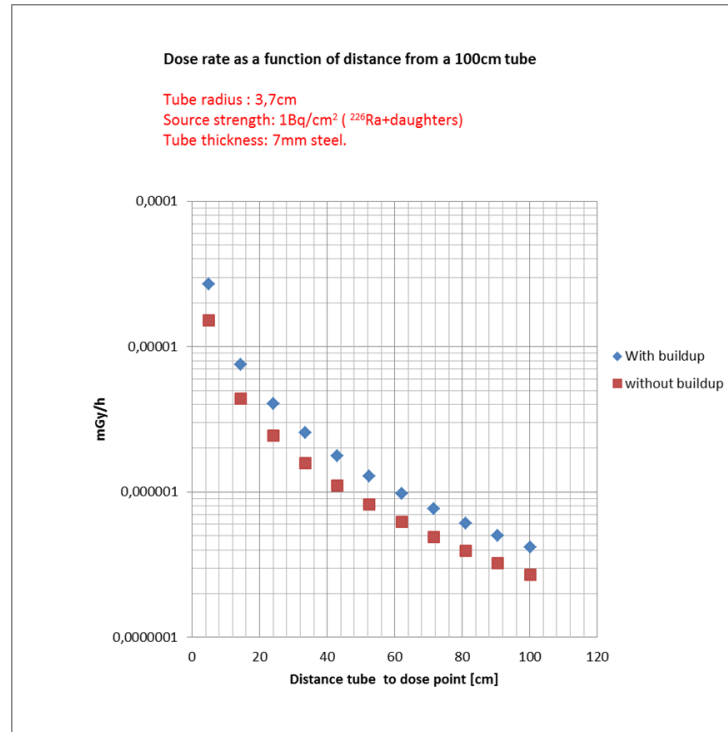


Figure 10. Dose rate as a function of distance from tube surface of a 100cm long tube.

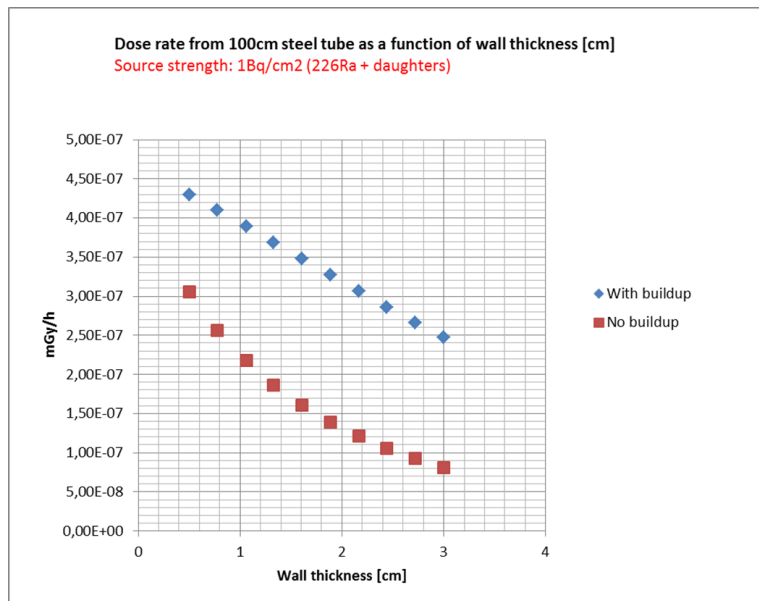


Figure 11. Dose rate as a function of steel wall thickness of a 100cm long tube.

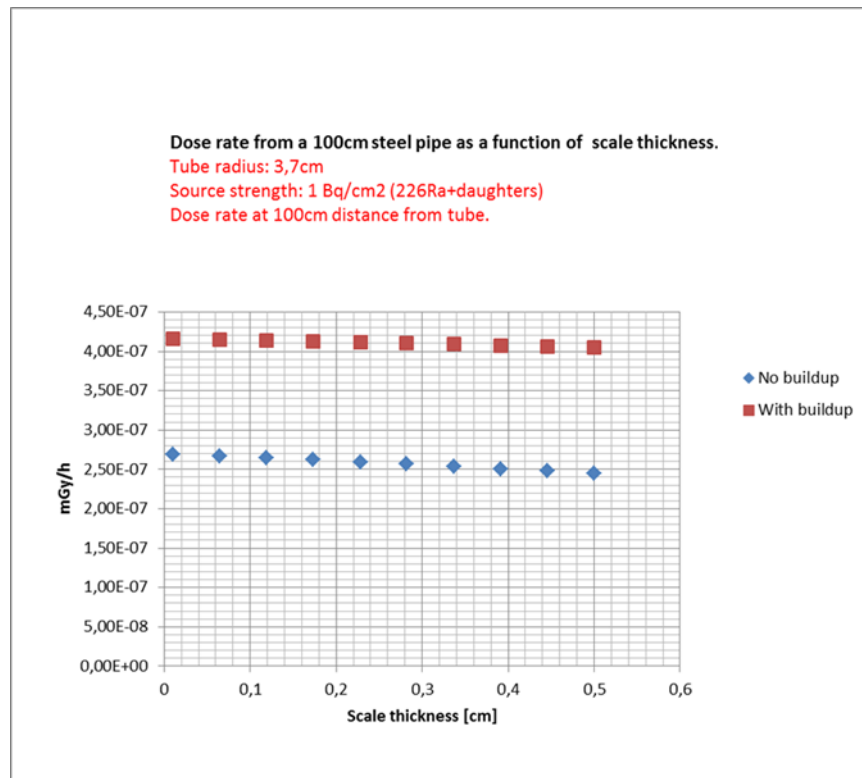


Figure 12. Dose rate as a function of scale material thickness of a 100cm long tube.

5 Practical methods for characterization of NORM material

Information about characterization of NORM was collected from thirteen guidelines. Of these thirteen, four guidelines are from national authorities¹ or presentations thereof, seven are from operators or their advisors and two are written by international organizations.

5.1 Characterization methods

Although most countries have regulations and guidelines indicating exemption levels for NORM materials, it has only been possible to find thirteen actual metrological instructions on how to characterize or screen material containing NORM. Of these only four are prepared by regulators.

¹ Three countries

These thirteen guidelines have been examined. Annex 1, 2 and 3 present metrological instructions for characterization of NORM material from national authorities, oil and gas operators (including consultancies) as well as international organizations respectively.

Characterization of the NORM is generally performed using one of three characterization - or screening - methods:

- Measuring with dose rate meters (DRM).
- Measuring with contamination monitors (CM).
- Measuring with the "puck" method.

Measuring using DRM or CM is the most common. The "puck" method is developed and used in Norway. The following describes the advantages and limitations of these methods as well as more general comments.

5.2 Characterization using a dose rate meter (DRM)

Measuring with DRM can be performed relatively quickly. Nine of the thirteen guidelines outline a characterization process using DRM. However, the DRM method has its limitations and two guidelines recommend CM as a more reliable measuring method. DRM may in some cases be used to test for contamination on the inside of the tube by measuring on the outside of them. Some isotopes cannot be detected in this way.

It is recommended in some of the guidelines to use a sensitive dose rate meter. One regulator indicates that the equipment must be capable of detecting dose rates down to 100 nSv/h.

5.3 Characterization using a contamination monitors (CM)

Ten of the thirteen guidelines describe characterization using CM.

The method takes a bit longer than DRM and cannot be used to estimate the contamination inside the equipment by measuring on the outside. On the other hand DRM is easy to operate and the results can via a conversion table often be directly converted from the CPS to Bq/cm² (Several operators have conversion tables for conversion from CPS to Bq/cm²).

One guideline outlines a method including the sampling of sand or scale to be measured by CM in an internal laboratory. The method has similarities with the "puck" method, see below.

5.4 Characterization with the "puck" method

The "puck" method is not an actual characterization or screening method, but an in situ method for calibrated analyzes of NORM, where the result can be directly compared with the exemption

levels. The sample is placed in a special container - a "Puck" - and the measurement is performed with a CM in beta mode. The method has a practical detection limit of 2-3 Bq/g. Apparently, the "Puck" method works well when it comes to dry material (scale) and less well if the material is wet (sludge).

5.5 General comments

It is specified in six out of the thirteen guidelines, that it is important to keep the probe close to the surface/equipment when measuring. Five of these six guidelines recommend a fixed spacer.

This is deemed to be an important point.

One guideline specifies that the measurements are very sensitive to water/grease on the surface of the equipment and one guideline points out that the final measurement should be done at dry equipment.

54 % of the guidelines note that measures should be taken to prevent the spread of dust from possible NORM material and 46 % of the guidelines recommend keeping the material wet.

For health reasons, there should be no relaxation of the provision to keep the equipment wet during handling, unless other appropriate measures are taken to prevent exposure of employees to NORM dust. It is therefore important to examine whether the water affects the measurement results, and if so to what extent.

According to one guideline the isotopic composition of NORM vary from sector to sector in the North Sea. NORM from the north is dominated by ^{226}Ra and ^{229}Ra , while NORM from the southern part mainly contains ^{210}Po and ^{210}Pb , and this fact must of course be reflected in the choice of measuring equipment. This is possibly part of the explanation for the differences in the individual guidelines in the choice of measuring equipment.

Eleven of the thirteen guidelines indicate a trigger level for indication of NORM contamination. There are some differences between the trigger levels in these guidelines. In eight guidelines trigger levels are indicated as the background level or 2-3 times the background level. Three guidelines indicate trigger levels in Bq/cm^2 ranging from 0.37 to 2.7.

None of the thirteen guidelines address directly the challenges of measuring items with different geometry. It is however still of importance to investigate the impact of different geometry of the inspected objects and define to what degree this can cause measurement challenges, especially considering NORM with isotopes that do not possess enough energy to be detected through a metal wall.

Table 5. National Guidelines for characterization of NORM, nationally funded development projects and presentations thereof

Country	Document	Method for in situ characterization	Remarks
Denmark	Sundhedsstyrelsen, 2005. Vejledning om håndtering af NORM fra olie- og gasindustrien (Danish Health and Medicine Authority, 2005. Guidance on the management of NORM from the oil and gas industry.)	<p>The guide specifies two methods:</p> <ol style="list-style-type: none"> 1. Measuring of dose rate. Use a dose rate meter (DRM) with high sensitivity, which can measure dose rates at 100 nSv/h. 2. Measurements using a contamination monitor (CM) to measure alpha and beta radiation are often most suitable. The monitor is placed in a holder which ensures fixed distance to the sample. <p>At radiation above the normal background, samples must be taken for further analysis.</p> <p>Ensure that the sample is representative, for example, by mixing a number of samples.</p>	Preferably use working methods that keep the radioactive material wet to prevent airborne contamination.
Norway	Per Varskog, Zpire Ltd. EAN-NORM 6 th Workshop, Madrid, Spain, 02.12.2013. NORM monitoring.	<p>The presentation specifies two screening methods and the “Puck” method :</p> <ol style="list-style-type: none"> 1. Screening with a dose rate meter (DRM) is primarily to define areas with enhanced radiation. Skilled operators may use the DRM outside their “purpose”, e.g. to use it to assess contamination. 2. Screening with a contamination monitor (CM) with a beta sensitive probe is used to measure surfaces to assess NORM contamination. Criterion for positive result should be 2-3 times background. The background is measured as close to the measure point as possibly (Be aware that representative background may be difficult to perform). In Norway “2 x background” measure on land is often equivalent to 1 Bq/g for loose materials. 3. The “Puck” method is an inter-field method for calibrated measurement of NORM. A NORM sample is placed in a special container - a "Puck" - and the measurement is performed with a CM in beta mode. The method has a practical detection limit of 2-3 Bq/g. 	The “Puck” method works good at dry material (scale) and bad at wet material (sludge).

Norway	Statens strålevern, 1997. Hefte 12. Avleiring av naturlig radioaktive stoffer I olje- og gassproduksjon. (NRPA, 1997. Booklet 12. Accumulation of natural radioactive substances in the oil and gas production.)	<p>It is recommended to use DRM that are sensitive enough to measure dose rates at 100 nSv/h by characterization of contaminated equipment, as these instruments are robust and easy to use. There must be measured as close to the surface as possible and in fixed distance.</p> <p>Trigger level for dose rate is twice the background level. Background level is established as the dose rate of one meter above the ground near the work area.</p> <p>At very thin NORM deposits with high activity concentration is DRM measurements are not useful. Instead characterized pipes and other equipment using CM.</p> <p>Equipment from gas production can be covered inside of thin coatings of NORM with ^{210}Pb. In these cases, measured directly on the contaminated surface with an instrument that is sensitive to beta radiation.</p>	For the cleaning of equipment; use methods, which keep the material wet in order to avoid release of dust.
Canada	Fact Sheet. Naturally Occurring Radioactive Material (NORM). Canadian Nuclear Safety Commission. November 2014.	Radiation readings above typical background may indicate the presence of NORM. Subsequent determination of the type of the material is important.	Best practices include amongst other things minimizing operations that generate dust containing NORM.

Table 6. Internal guideline for characterization of NORM for operators in the oil and gas industry

Operator No. ²	Document	Method for in situ characterization	Remarks
1	NORM - Naturally Occurring Radioactive Material, 2010.	Method for use on oil rigs for control of radiation and activity of NORM materials. The equipment is estimated to be NORM contaminated if the CM measured more than 24 CPS or dose rate determined by a DRM is more than 2.5 $\mu\text{Sv/h}$. (The guide is accompanied by a conversion table, where it appears that 24 cps is equivalent to 2.7 Bq/cm^2).	
2	It is perfectly NORMal. A booklet on NORM. Natural Occurring Radioactive Material.	Use a CM for characterization of possible NORM material. Results can be read in the CPS, which is converted to Bq/cm^2 using a conversion table. One must be careful with the measurement. The sensitive part of the probe must be kept 10 to 15 mm from the surface of the material. Avoid contact between the probe and the material. It takes some time to get stable results. Classification limit is 2.7 Bq/cm^2 .	
3	Managing NORM. Naturally Occurring Radioactive Material. January 2011 and cover letter of 6 January 2011.	Before opening any equipment you must measure dose rate with a DRM. The area is classified as radioactive at dose rates above 2.5 $\mu\text{Sv/h}$. In open equipment and possible contaminated surfaces, you have to measure with a CM. The apparatus can measure alpha, beta and gamma radiation and the measurement read in the CPS. Background radiation is recorded and subtracted from the measurements. The result can be converted to Bq/cm^2 by means of a table. The trigger level is 2.7 Bq/cm^2 . Materials that exceed these values are considered as NORM contaminated, and sent ashore for decontamination.	NORM must be constantly kept wet to prevent dust formation. Using personal protective equipment in dusty work.

² Operators are rendered anonymous in this document.

4	Radioactive substances procedure. 1 August 2009.	<p>A sample (approximate 2-3 dl) of the sand or scale is carefully transported to the internal lab, without any kind of spill.</p> <p>Use a CM equipped with an alpha and beta sensitive probe to determine the level of radioactivity in the sand/scale. The probe shall have a distance of not more than 1 cm from the sample when measuring. Use a period of 10 minutes to count, both on each sample and the background.</p> <p>The CPS value for the samples are determined and compared with the CPS value for the background radiation.</p> <p>The trigger level is two times background radiation.</p>	Keep all solids wet to prevent dust generation.
5	Guidelines. Work with radioactive deposits and contaminated objects. 1997.	<p>Dose rate measurements: For the classification of piping and components, it is Important that the measure is done so close to the surface of the object as possible in a given, fixed distance. If radioactive substances are deposited inside the tube, the specific activity is not determined by measurements on site. The specific activity may be estimated from the dose rate and an internal experience material.</p> <p>For measurement in gas / condensation system: use a monitor with a probe calibrated against ^{210}Pb, for measurement of beta radiation. If the measured radiation level is higher than the background level, the activity should be measured with high resolution gamma spectroscopy.</p>	It is important to avoid creating dust from the work. This is done most effectively by keeping the equipment humid.
6	Guidelines for naturally occurring radioactive materials, 2002.	Monitoring instrument: Surface contamination monitor. Limits trigger level for NORM: 0.37 Bq/cm^2 .	There is no surface contamination limit in current Australian regulations. Nevertheless, the skin contamination limit of 0.37 Bq/cm^2 has been used.

7	<p>Procedure. Onshore And Offshore Local Rules for Work With Ionising Radiation Involving (LSA) And (NORM), 2013,</p>	<p>NORM from different parts of the North Sea includes different isotopes; in the north is frequently detected NORM with ^{226}Ra and ^{228}Ra and in the southern sector ^{210}Po and ^{210}Pb are frequently observed. Within these large areas there may be minor sections with a different isotope composition in NORM. The measuring equipment shall reflect the specific isotopes in the NORM.</p> <p>In the case of NORM with ^{226}Ra and ^{228}Ra a gamma sensitive detector can be used to detect NORM at the inside of the equipment by measuring the outside of the equipment (this may be difficult if the metal is thick). Are the isotopes ^{210}Po and ^{210}Pb dominant, can contamination inside the equipment not be detected without opening the equipment, since the isotopes do not possess enough energy to be detected through a metal wall. There must be measured directly on contaminated surfaces, using a Mini900EP15, PCM5 or Electra with DP2.</p> <p>There is no direct trigger level in the document for determine if pipes and other equipment are NORM contaminated. There is, however, set a trigger level - or success criterion - for decontamination of contaminated equipment. The final monitoring must not exceed two times the background level, where the background level must not exceed 5 CPS.</p>	<p>All final monitoring after decontamination must be carried out when the equipment is dry and not wet.</p>
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Table 7. International organizations' guidelines for characterization of NORM

Organization	Document	In situ characterization method	Remarks
International Association of Oil & Gas Producers	Guidelines for the management of Naturally Occurring Radioactive Material (NORM) in the oil & gas industry, Report No. 412, September 2008.	<p>The guide specifies two methods:</p> <ol style="list-style-type: none"> 2. Measuring of dose rate (gamma). Use sodium iodide detector as a quick and qualitative method. For a more accurate indication use Compensated GM tube. 3. Measurement using CM (alpha and beta). Use uncompensated GM tubes. Probe must be as close to the surface as possible without touching it. <p>Radiation above the normal background, indicate NORM contamination.</p> <p>NORM action limit: Equipment, clothing etc. shall be considered "NORM-contaminated" if contamination measures double radiation background level.</p>	<p>Surface coating with water or oil/grease would attenuate any NORM contamination present on the surface and give lower than anticipated indication on the detector.</p> <p>Many surfaces may be difficult to directly monitor due to their surface condition or geometry.</p> <p>When working with equipment that can be NORM contaminated, equipment must be kept wet during the work, to avoid NORM dust to be airborne.</p>
IAEA	Radiation Protection and the management of Radioactive Waste in the oil and Gas Industry. Training course Series 40. Vienna, 2010.	<p>It is recommended to select the measuring equipment as specified below:</p> <ul style="list-style-type: none"> • For well tubing strings use Gamma log. • For closed topside production, transport/off flooding systems, decontamination facility and storage facility use calibrated dose rate meter. <p>Measurement results are compared with the rules set down by the regulatory body.</p>	<p>Apply, when practicable, only those work methods that keep NORM contamination wet or that confine it to prevent airborne contamination.</p>

6. Conclusions

The new BSSD (Euroatom Basic Safety Standards Directive), adopted by the member states in January 2014, further emphasize radiation protection issues in NORM relative to the old standards. The reason for this is a growing consensus that natural radioactivity actually leads to significant doses to workers in a variety of industries. The actual identification of contaminated material is however a major problem since this is based on the 1Bq/g limit while instrumentation used to sort out contaminated equipment either is based on dose rate or simple gross count-rate. This results in interpretations of contamination levels which are either over or underestimated. In order to improve the measurement strategy the current project has focused on the characterization of contaminated equipment to be able to understand the basic factors that may govern uncertainty in the measurements.

Characterization of pipes received during the course of the project shows that the decontamination operation performed by the responsible company is very satisfactory and leaves non-detectable residues of activity on the analysed pipes. Analysis of the scale containing pipes shows that the radioactivity distribution is spatially homogeneous with variations in the order of 10% or less on the decimetre scale. Radioactivity content varied from about 100 to 10 Bq/g of ^{226}Ra in scale between the tubes received. The $^{228}\text{Ra}/^{226}\text{Ra}$ ratio was approximately 0.1 and the $^{228}\text{Th}/^{228}\text{Ra}$ ratio about 0.1. The external dose-rate from scale contaminated tubes was calculated as a function of tube length, material thickness, distance and scale density assuming only ^{226}Ra +daughters contamination. The equilibrium between radon (^{222}Rn) and radium is a key factor in both calculating and measuring external dose rate from the tubes. The dynamic radon emanation which depends on several physical factors limits the possibility of obtaining coherent results when performing several measurements. This complicates judgement on how to handle contaminated equipment. The rapid measurements performed off-shore when inspecting and/or changing equipment, often few minutes after closing operation, may thus be misleading due to disequilibrium between several short-lived radioisotopes and their more long lived parent isotopes. This does not only include radon but also ^{224}Ra which occurs at abundant concentrations in formation water. On top of this variability caused by disequilibrium conditions the overall uncertainty is increased as a consequence of the measurement procedures using variable procedures and staff.

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Appendix 1

Measurement instruments and characterization instructions

There are no radiation measurement instruments that are specifically designed for being used as a characterization tool for NORM in the oil and gas industry. No hand-held instrument is able to measure the different radiation types and energies one may come across in connection with NORM.

There are two measurement principles to characterize NORM in the field:

1. Measurements with a dose rate meter (mSv/h or μ Sv/h).
2. Measurements with a contamination monitor (cps or cpm).

Contamination monitors and dose rate meters cannot be used to determine the material's concentration of radio nuclides or determine if the exemption levels are exceeded. These measurements serve to identify NORM, to assess the radiological conditions at the workplace and to select samples for further analysis.

In the following, the term “tube” is used refer to the tubes, pumps, tanks, valves etc. that is being measured on, since tubes are the most common kind of equipment.

Dose Rate meters

There are several types of dose rate meters on the market. For NORM, a type with a sensitive detector, which is able to measure low dose rates (approx. 0.1 μ Sv/h) should be selected. Such an instrument can in some cases be used for measuring on the outside of the tube suspected of containing NORM.

Instruments for measuring dose rates usually contain a scintillation monitor or a GM-detector. These detectors can only determine gamma radiation. NORM from the oil and gas industry emits alpha and/or beta radiation, but not all NORM nuclides emit high-energy gamma radiation, which is suitable for the dose rate measurements.

Since alpha radiation and most of the beta radiation will not be detected by a dose rate meter, dose rate measurements can give an imprecise estimate of NORM quantities, if the NORM contamination is dominated by nuclides with low-energy gamma energies or no gamma radiation at all.

Some radio nuclides as Po-210 and Pb-210, which emit low-energy gamma radiation or no gamma radiation, will not be measured on the outside of the tubes. These nuclides are dominant in the NORM from for example gas tubes.

Since the dose rate decreases with the square of the distance from the source (here NORM), the measurement must be carried out in a low fixed distance.

6 Characterization instructions – dose rate meter

Use a dose rate meter with a fixed spacer.

Make sure the dose rate meter work on a test source before and after the actual measurements. Note results.

Measure background dose rate near the workplace, but still outside the area with the elevated dose rate. Note background dose rate.

Press the fixed spacer firmly against the tube. Wait for a stable measurement result and note it.

If the dose rate from the tube exceeds 2 times the background, the pipe must be regarded as potentially contaminated for further handling. NORM must then be sampled for analysis at a laboratory.

The measurement can be carried out on wet pipes. A thin film of water will not be able to screen the gamma radiation.

Be sure that there are no other radioactive sources around, when dose rate and background is measured (logging sources, test sources, flow or density meters and similar).

Contamination Monitors

It is important to choose a contamination monitor, which is optimized for the relevant type of radiation to measure. Furthermore, it should be noted that probes for contamination monitors have thin windows of glass or film, and thereby are more fragile than dose rate monitors. A damaged probe will result in a misleading measurement. It is recommended to equip the probe with a fixed spacer, so the measurement can be done quite close to the equipment without destroying the probe.

Contamination monitors that can measure alpha and beta radiation are often the most suitable for thin layers of NORM. Since the vast majority of radio nuclides in NORM emit alpha and beta radiation, an alpha-contamination monitor or combined alpha-beta monitor is suitable.

Note that the alpha particles have a very limited range even in air. Even when measured directly on top of the NORM, self-absorption can make it impossible to "translate" the instrument readings (often in cps or cpm) directly to a nuclide concentration in the NORM material. This requires that the instrument is precisely calibrated to the type of radiation and the material that one wishes to measure.

There are instruments that can translate count rates (cps) to absolute units as Bq/cm^2 , but this requires that the measured material is very homogeneous, and that the instrument is calibrated to the relevant nuclides. NORM mostly consists of a mixture of radio nuclides that rarely appear in the same conditions. It's usually not possible to make assumptions about nuclide composition that gives a meaningful determination of Bq/cm^2 .

For the screening of NORM for Ra-226 in Bq/g it is however possible to construct an experimental setup with a beta sensitive contamination monitor. The monitor is attached to a fixed distance to the sample. A standard used for calibration is needed. This standard must have similar physical composition as the sample.

The standard and samples are packed in a standard container that roughly corresponds to the monitor

sector's window. It is possible to measure the standard activity and compare it to the samples' activity in the number of counts over a time interval, for example 1 minute. The background level will be determined by measuring without a sample.

7 Characterization instructions – contamination monitor

Use a contamination monitor - calibrated to the relevant nuclides - with a fixed spacer.

Make sure the contamination monitor works on a test source before and after the actual measurements.

Note results.

Measure background radiation. The background must be measured within short distance from the work area. Note results.

Press the fixed spacer firmly against the pipe to be measured. Wait for a stable measurement result and note it.

If the dose rate from the equipment exceeds 2 times the background, the equipment must be regarded as potentially contaminated for further handling. NORM must be sampled for analysis at a laboratory.

Sampling of NORM for analysis

The NORM samples must be representative. If the material is inhomogeneous, several samples must be taken.

A quantitative analysis of the NORM activity must contain at least the identification and concentration of the relevant nuclides. Apply documented reproducible measurement methods, as gamma spectrometry

Packaging and quantity of samples must be settled with the laboratory.

Appendix 2

Implementation in existing Nordic NORM management guidance

In connection with the revision of the Danish guidance on the handling of NORM from the oil and gas industry, results from the project will be incorporated after a mini-hearing on the results and methods among Danish oil and gas operators.

A thematic day for oil and gas operators will be held at the National Institute of Radiation Protection (Denmark). The results of the project will be presented in the form of new guidelines for characterization of NORM. Wherever possible, the thematic day will include both theory and practical exercises.

At the annual courses in Basic Radiation Protection at the National Institute of Radiation Protection (Denmark), training in the handling of NORM is an optional alternative to be chosen by individuals, who wish to be radiation protection officers in the NORM area or just want to work with NORM. The results of CONCORE will be communicated through this training.

Appendix 3

Application of project results

Although all laboratory tests used in this project are executed on pipes from the oil and gas industry, the project results could be used in a wide range of other industries, where there are deposits of NORM material in pipes, tanks, etc. In wet processes undergoing changes in pressure, temperature, pH, redox conditions, etc. there may be precipitations. Even very low levels of radionuclides in the material flow can be concentrated in these precipitations and thus form NORM. The following sections present a number of Nordic industries where NORM formation can occur. There are attached brief comments on each-industrial type.

Shale gas:

Extraction of shale gas may cause formation of NORM material both in the drilling phase and during the normal operation. The shale layers, from which the gas is recovered, can have a relatively high content of uranium and uranium daughters. The drilling waste may therefore be categorized and treated as NORM material. As the gas has a low content of radon, there may be deposits of radon daughters in the tubes and other equipment, as seen in the pipe systems used for gas from the North Sea.

Water treatment:

Groundwater is usually treated in several ways before it can be used as drinking water. In the context of the processes used, there can be several possibilities for the formation of NORM. It is often necessary to add oxygen to the water by aerating it, and this can result in precipitation of calcium carbonate and iron oxides. If the water contains radionuclides, these may become concentrated in the formed deposits. In certain areas, high levels of radon in the groundwater make active carbon filtration necessary, and it can lead to carbon filters with levels of radon and radon daughters high enough to necessitate treatment as NORM.

Geothermal industry:

Extraction of geothermal heat can cause scale formation in heat exchangers, pipes and other equipment. As the volume of water passing such a facility may be considerable, even very low levels of radionuclides in the water may lead to the formation of NORM deposits.

Paper industry:

In the context of the processes used in the manufacture of paper, there may be formation of deposits or scale containing NORM in pipes, tanks, etc.

Mining:

Ore often contains a mixture of elements and some of which may be undesirable and difficult to handle. For example, iron ore can contain uranium isotopes and their daughters. By processing, the ore is subjected to a number of chemical treatments that can cause precipitation and concentration of uranium and uranium daughters in some parts of the production system.

Title	Characterisation of NORM Contaminated Objects: Reliable & Efficient
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Abstract max. 2000 characters	<p>The predominant contributors to the production of Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) and NORM-waste in the Nordic countries are the on- and offshore oil and gas producers. In oil and gas production processes, host rock formation water containing low concentrations of NORM is mixed with seawater containing high concentrations of sulphate. This leads to precipitation of NORM (Ra, Pb, Po)SO₄, which is deposited as either scale or sludge in the production equipment. NORM contaminated pipes, tubes, pumps and tanks, etc. are therefore subject to radiological characterization in order to ensure safe reuse or recycling as well as safe NORM waste handling. The procedures and measurement techniques may significantly affect the amount of material that is categorized as NORM contaminated equipment and NORM waste. At present, different procedures are used for categorization by the individual operators in the North Sea. A number of parameters considerably influence the measurement accuracy and may cause false categorization of the materials, for instance abnormal geometry of objects, various density and composition of the scale and sludge, inhomogeneous distribution of NORM, and measurement sensitivity and uncertainty. Incorrect</p>

characterization can however be prevented by the use of well certified methods and inappropriate handling of these materials can be avoided. The current report is partly a review of existing methods to perform initial characterization of NORM contaminated equipment and partly an experimental section dealing with the basic investigations required to evaluate factors affecting external dose rate measurements.

Key words

NORM, TENORM, Scaling, Oil & gas industry, radium, ^{226}Ra , ^{228}Ra , dose-rate