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Comparative assessment of the European and Latin American scenarios for NORM/TENORM exposure

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Abstract. The geological formation of the areas of high natural radioactivity are usually associated with mineral ores commercially important. As a consequence, extracting industries are installed in or near those areas. Several industrial products and byproducts can be obtained from thorium, uranium and potassium rich mineral ores, for example: niobium concentrate from pyrochlore; monazite, ilmenite, rutile and zirconite concentrates from monazite sands; phosphate fertilizers from apatite; tin and lead from cassiterite; gold and copper form a variety of thorium and uranium rich mineral matrices. Other industries like oil- and natural gas production and processing, production of thoriated tungsten lamps, welding, gas mantles and pigments produce non-negligible amounts of wastes containing technologically enhanced naturally occurring radioactive materials (TENORM). In addition, quite frequently unknown amounts of natural radionuclides end up embedded in a variety of consumer products. Thus, naturally occurring radioactive materials (NORM) as well as TENORM in consumer products and/or industrial wastes may be of importance as far as human exposures are concerned. This work will present a comparative assessment of the radiological significance of different NORM/TENORM exposure scenarios in Europe and Latin America.

1. INTRODUCTION

In the scientific community it has been known for about 25 years that workers in the natural gas- and oil industry can be exposed to technologically enhanced levels of natural radioactivity (TENR), although they are not a priori classified as occupationally exposed persons [1–3].

At the regulatory level the International Commission on Radiological Protection (ICRP) recognized in 1991 the need for regulatory control over TENR-sources. It recommended that the full system of radiation protection (adherent to the principles of justification, limitation and optimization) should apply for workers if the TENR-exposure scenario results in average annual doses exceeding 1 mSv [4]. Subsequently these recommendations have been adopted internationally as reflected e.g. in the recommendations by the International Atomic Energy Agency (IAEA) on radiation safety and the safety of radiation sources [5].

In Europe the EURATOM Radiation Protection Basic Safety Standards – EURATOM BSS – recommends in Item VII [6]:

- It is the responsibility of the Member State to identify all those work places where handling of material with increased TENR could occur.
- In such cases appropriate measures should be taken in terms of monitoring and/or reduction of the TENR-induced radiation exposure.
- The limit for mandatory action is set at 1 mSv/a.

- The activity concentration of naturally occurring radioactive material in waste of the non-nuclear industry should not exceed 500 Bq/g.
- If the limit of 500 Bq/g is exceeded a Member State can exempt this type of activity only if the individual annual dose of 10 μSv, respectively the collective dose of 1 man-Sv is not exceeded.
- Alternatively the results of an optimization process have to demonstrate that the exemption is indeed the optimum solution in terms of radiation protection.

Those industrial processes in Member States which could be of concern of Item VII of the EURATOM BSS were identified in Radiation Protection 88 [7]. Quite recently, the reference levels for workplaces processing materials with enhanced levels of naturally occurring radionuclides, which included the oil and gas extraction industry, were the subject of Radiation Protection 95 [8]. The Group of Experts established under the EURATOM Treaty identified the most likely pathways through which exposures to TENR occur, and proposed a four band system with marker points between each two of them for regulatory control above the limit at 1mSv/a. As far as regulatory control is concerned, the band structure is the following: band 1 (no need to consider regulatory control – equivalent to a band below an exemption level); band 2 (lower level of regulation); band 3 (higher level of regulation); and band 4 (process should not be permitted without a full individual assessment).

Taking into account the identified pathways for each case, screening and reference levels were derived to indicate the likely level of regulation for the radionuclides important for the relevant industry. Thus, for example, if the derived screening and reference level put the industrial process in band 1 there is no need for regulatory control. However, if the derived level would place the industrial process higher than in band 1, control measures would be applied, or the process should receive individual assessment prior to deciding which control measures to adopt [8].

In Brazil, there is not yet any established procedure as far as TENR is concerned. The TENR levels are still regulated through dose limits which are non-specific. There is, however, a broad regulatory legislation which can be applied, in principle, to any industry, including the extraction industries.

The increasing public awareness of environmental issues is putting pressure on the Brazilian legal system to solve complex environmental problems resulting from the application of inadequate laws which ignore long term implications to the environment [9]. The lack of clear cut national guidelines concerning the disposal of accumulated TENR wastes has already engendered ridiculous judicial battles in the courts of justice between environmentalists and the monazite industry in Brazil.

2. TENR-EXPOSURE SCENARIOS IN THE OIL- AND GAS INDUSTRY

2.1. Source terms

In Europe the recycling industry is being increasingly supplied with metal originating from the dismantling of installations in the oil- and gas industry. Such material is frequently contaminated by natural radioactive nuclides from the U- and Th-decay series, such as ^{238}U, ^{235}U, ^{232}Th, ^{234}Th, ^{226}Ra, ^{228}Ra and ^{210}Pb. The contamination of the equipment results mainly from :

- Solid radium (^{226}Ra) contained in scale deposited on the inner surface of pipes, vessels and tanks.

- Gaseous radon (^{222}Rn) concentrated in ethane and propane fractions due to the fact that the boiling point of Rn lies in between that of propane and ethane. Elevated Rn-activity concentration have been measured at several such sites of processing plants [10].
- Short lived Rn-decay products (Rn-d) deposited on internal surfaces of various equipment components. The radioactive decay results in a radioactive contamination by the long lived radioactive lead-isotope ^{210}Pb (T ½ = 22.3 a) and the growth of the alpha-emitting polonium isotope ^{210}Po (T ½ = 138d).

The association of TENR and petroleum genesis have already been proposed, to the extent of that should be a correlation between the age of oil and those of ^{238}U and ^{232}Th found in the organic materials from which petroleum is originated and TENR produced in the oil and gas industries [11]. Experimental work to confirm such association is still to be done.

Until recently there was a legal monopoly to extract oil in the whole territory of Brazil exercised by a government owned company. Such monopoly made this company the largest Brazilian industrial and commercial enterprise. As one of the unwanted consequences of such concentration of commercial and, to a considerable degree, political power, the Brazilian environmental regulatory agencies were unable to enforce non-specific regulations concerning TENR to such a powerful company. However, many thousands of cubic meters of TENR wastes and pipes with radioactive scales have been accumulated by the Brazilian oil extraction industry throughout the years. The current meeting may help State and Federal regulatory agencies to draft more specific regulations concerning TENR applicable to the Brazilian oil extraction industry. Such regulations are becoming even more urgent in view of the fact that end of the monopoly of the oil industry enables other companies to extract oil and gas in the Brazilian territory.

There are also large oil and gas extraction industries in other Latin American countries, such as Argentina, Bolivia, Ecuador, Mexico, and Venezuela. However, unlike the European Union Member States, Brazil and these other Latin American countries have not yet made any effort to discuss, propose and finally adopt a common regulatory structure concerning TENR, which may be applicable to the extraction industries, including the oil and gas industry.

By and large, the size of the TENR source terms in the Latin American oil- and gas producing countries are either unknown, or at least poorly identified. Joint efforts ought to be made to improve the current dissatisfactory situation.

2.2. Exposure pathways

Radiation exposure of workers in the oil- and gas industry can occur under the following conditions:

- Gas samples taken near well-heads show an average Rn-concentration of 0.5 Bq/L [12].
- In Germany almost 30 million tons of brines are raised to the surface annually as an unwanted by-product [12]. Increased exposure to external gamma radiation can occur in the vicinity of vessels and tanks filled with brines.
- Maintenance operations on scale-contaminated process equipment can generate undesirably high dust levels in the breathing area of workers [13]. Such operations occur e.g. during sand-blasting of the inside of tanks, opening of valves for repair purposes, brushing of surfaces or grinding of metal. Scales contain the following radionuclides (maximum value in Bq/g): ^{226}Ra (1000), ^{210}Pb (72), ^{228}Ra(360), ^{228}Th (360). The range of typical values in brines and scale is shown in Table I.

Table. I. Range of typical activity concentration values for ^{226}Ra in brines and scale from the gas- and oil industry [14].

	^{226}Ra
brines	7.6 – 286 [Bq/l]
scale on equipment	202 – 1000 [Bq/kg]

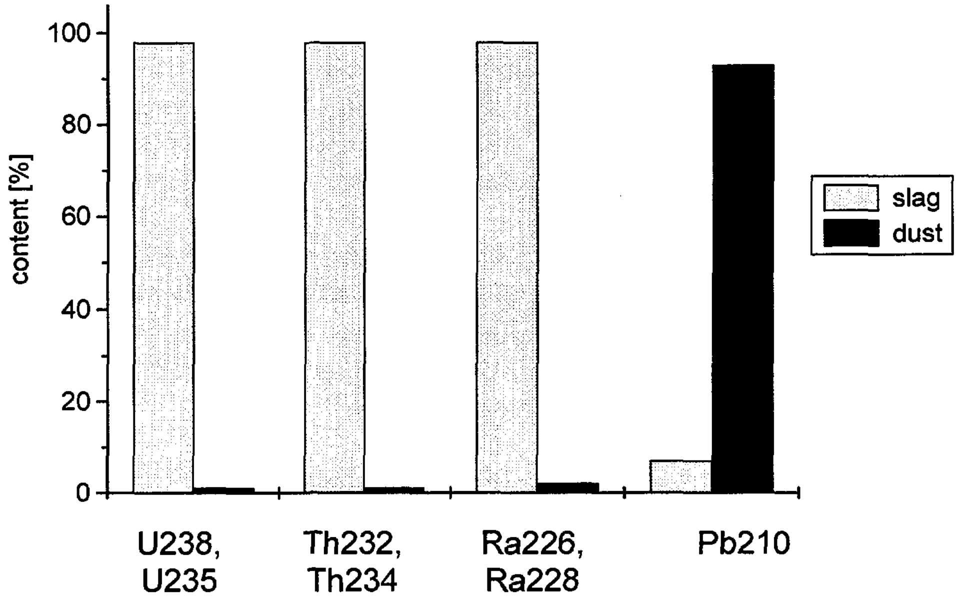

Figure 1. Nuclide distribution in slag and filter-dust after melting of TENR-contaminated metal during commercial recycling processes.

Furthermore the disposal of large amounts of contaminated wastes resulting from the various stages at processing plants can pose an environmental problem due to its content of long lived radionuclides (e.g. T ½ ^{226}Ra= 1600 a).

Finally the recycling of TENR-contaminated metal can pose a contamination problem for the operator of such a recycling plant. During the process of melting down ^{226}Ra and its decay products concentrate mainly in the resulting slag, whilst ^{210}Pb (gaseous at operating temperatures ~ 1400 C°) can be found predominantly in the dust filtration system of the exit-air [15](see Figure 1).

The Group of Experts of EURATOM considered the most likely pathways through which exposures to humans might occur as the following: inhalation of radon; ingestion of dirt and dust; skin contamination [8]. Taking into account such exposure pathways, and the effective dose limits at point marks between the four bands mentioned in section 1, this Group of Experts estimated screening concentration levels for ^{226}Ra plus progeny for oil and gas removal, scales in pipes, and sludge to be between 30 and 300 Bq/g. Thus, for concentrations below 30 Bq/g there would be no need to consider regulatory control, and above 300 Bq/g individual assessment would be necessary prior to deciding which control measures to take.

Advantage may be taken from exposure pathways observed in the studies of natural radioactive areas. In particular, the highest concentrations of ^{226}Ra, ^{228}Ra, and ^{228Th} in food and water samples from selected Brazilian areas of high natural radioactivity have been published [16]. Environmental behavior rather than laboratory conditions may be useful to establish TENR pathways, starting from the source-term until the exposure of man.

2.3. Resulting Doses

On the one hand, according to the recent compilation of data in UNSCEAR, the absorbed dose rate in air, including cosmic and terrestrial radiation, range from 0.02 to 30 μ Gy/h in areas of high natural radiation background [17]. On the other hand, in the thorium rich areas of Brazil external exposure radiation levels were reported to range from 0.3 to to 18 μ Sv/h [16], which is somewhat consistent with the exposure which would be obtained from worldwide UNSCEAR data. Moreover, the concentration of radionuclides from the uranium and thorium series in foods and drinking water throughout the world has also been compiled by UNSCEAR [17]. The age weighted annual intake effective dose from ingestion of uranium and thorium series radionuclides was estimated to be 160 μSv. These doses resulting from exposure to naturally occurring radioactive materials should be taken into due account when estimating the relative importance of TENR due to any particular anthropomorphic activity.

The National Academy of Sciences — National Research Council (NAS-NRC) of the United States published recently a report with the results of an investigation to assess, among other issues, whether there is relevant scientific information that has not been used in the developments of guidelines to NORM [18]. Among the conclusions and recommendations of the NAS-NRC report it is worth mentioning the following:

"*1. The committee concludes that background radiation levels of NORM are highly relevant to regulation of TENORM because the radionuclides being regulated as TENORM are identical with those in nature. Arguments concerning small differences in the target regulatory level at small fractions of the natural background tend to pale into insignificance in comparison with natural background levels and their local and regional variations.*

2. Considering only external photon exposure, the committee notes that EPA (Environmental Protection Agency)'s proposed 0.15 mSv (15 mrem) standard is equivalent to an incremental increase in the concentration of radium-226 in soil of about the usual natural background level of 0.0 4 Bq/g (1 pCi/g). In view of the ubiquitousness of ^{226}Ra in soil and the substantial local variation of natural background, it is likely to be difficult to implement a 0.15 mSv (15 mrem) soil-cleanup standard for radium, particularly when the contamination is only marginally above the local background. That is especially the case if potential exposures to indoor radon are included in complying wth the standard.

3. As a practical matter, the implications of existing levels and the variability of natural radionuclide concentrations and doses received by humans should receive careful consideration in the regulation of TENORM."

3. CONCLUSIONS

1. The impact on society (occupationally exposed persons and members of the public) can only be estimated (Fig.2). Due to the lack of an adequate international database this estimate is associated with large uncertainties. Using the currently available data [19] it can be shown that the global impact from a one-year operation of the oil industry is equivalent to about 30 years of operation by the gas industry.

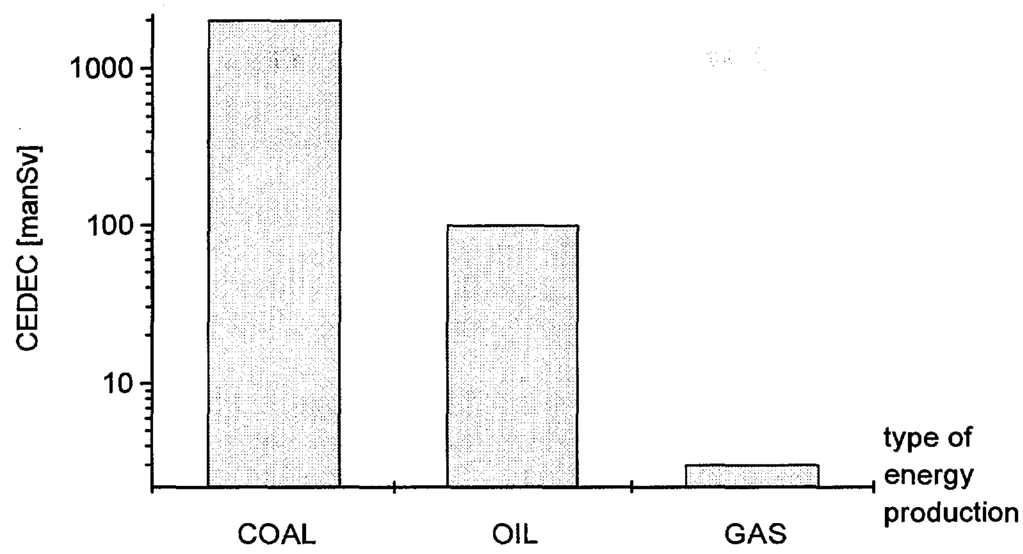

Figure 2. Collective effective dose equivalent commitment (CEDEC) for one year of world practice.

2. However, it is considerably less than the radiological impact from the coal industry (CEDEC: approx. 92 000 manSv).
3. A common basis for the international regulation of TENORM, taking into proper account natural background levels and their local and regional variation, should be found to establish a regulatory structure acceptable throughout the world, as it is the case with the IAEA BSS, and to a lesser extent the proposed EURATOM BSS.

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