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industry**

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## CHARACTERISATION OF NORM IN THE OIL AND GAS PRODUCTION (E&P) INDUSTRY

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### ABSTRACT

Trace quantities of the primordial, radioactive elements uranium ( $^{238}\text{U}$ ) and thorium ( $^{232}\text{Th}$ ) have been present in the earth's crust since its formation. Consequently, these elements are also common in oil/gas bearing geological strata. Both  $^{238}\text{U}$  and  $^{232}\text{Th}$  are the 'parent' of a complex series of successive decays, in which their radioactive 'daughters' are formed. Some of these daughters may be co-produced with oil/gas well fluids and may concentrate in ordinary deposits (e.g. scale, sludge) in tubulars, flow lines and processing/treating equipment. Combining the geochemical properties of the relevant individual Naturally Occurring Radionuclides and an extensive world-wide literature survey on E&P NORM, its characteristics has been determined. From the circumstantial evidence presented, it is concluded that E&P NORM is fully characterised by analysing for  $^{226}\text{Ra}$ ,  $^{222}\text{Rn}$ ,  $^{210}\text{Pb}$  and (rarely)  $^{210}\text{Po}$  as well as  $^{228}\text{Ra}$  and  $^{228}\text{Th}$  (only present due to ingrowth).

### INTRODUCTION

Naturally Occurring Radionuclides (NOR's) from the  $^{232}\text{Th}$ - and  $^{238}\text{U}$ -series, which are omnipresent in the earth's crust, can be concentrated by technological activities, particularly those in which natural resources (e.g. coal, oil, natural gas, geothermal energy, various types of ore, mineral water) are extracted and/or processed [1, 2, 3]. This may subsequently lead to an effective dose<sup>‡</sup> delivered to the workforce, which is additional to the natural background dose. In this respect it should be noted that also technological achievements (e.g. jet aircrafts, space exploration) [1,2,3,4] may lead to an increase of radiation doses. For these phenomena a special class of radiation exposure, the so-called TENR (Technologically Enhanced Natural Radiation), has been introduced [1,4].

The acronym TENR is only used for '*exposures to truly natural sources of radiation (e.g. NOR's and cosmic radiation) which would not occur without (or which are increased by) some technological activity not expressly designed to produce radiation*' [1,4]. TENR is observed when NOR concentrations in process streams are altered as a consequence of physical and chemical processes associated with the applied technology, such that there exists potential for a) exposures to individuals or populations, b) environmental contamination, c) increased environmental mobility, d) incorporation of radioactivity in products and materials (e.g. concrete additive), e) recycling or re-use of materials contaminated with NOR's (e.g. tailings, sands, slags, equipment, scrap metals), and f) improper disposal.

The occurrence of TENR is of increasing concern in oil and gas extraction and processing (or for short: Exploration & Production, E&P) industry [5]. The substances, in which NOR's of the  $^{232}\text{Th}$ - and  $^{238}\text{U}$ -decay series show up in technologically enhanced concentrations, are generally denoted by the acronym NORM (Naturally Occurring Radioactive Material). Focusing on the main waste stream, also the acronym LSA (Low Specific Activity) scale/sludge is employed. All appearances and aspects of NORM in the E&P industry are extensively discussed in this paper.

The world average dose of ionising radiation as received by each individual amounts to about 3.1 milli-sievert per annum (mSv/a) in areas of normal background [4]. For regulatory and analytical purposes, this dose is normally subdivided into a *natural*, a *medical*, an *occupational* and a dose caused by *(un)controlled release of artificial radionuclides*:

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‡ Biological sensitivity of the individual organs and tissues with respect to the various types of radiation is accounted for in the effective dose. For the remainder of this paper the generic term dose will be used.

- the world-population averaged *natural* dose, which for almost 70% is stemming from NOR's of the  $^{238}\text{U}$ - and  $^{232}\text{Th}$ -decay series, amounts to about 2.4 mSv/a and is essentially and deservedly uncontrolled by regulatory authorities;
- the world-population averaged *medical* dose amounts to about 0.7 mSv/a, but typical diagnostic doses delivered to individuals will be in the 1 to 10 mSv/a range. While efforts are made to minimise medical diagnostic radiation exposure to patients, this exposure is uncontrolled in the tacit belief that the practitioner is best able to balance risk-benefit;
- the world-population averaged *occupational* dose amounts to about 2  $\mu\text{Sv/a}$ . As this dose is mainly received by so-called radiation workers, their average annual occupational doses range from 0.5 to 5 mSv/a; e.g. in the United Kingdom this amounts to 1.9 mSv/a [6]. The radiation exposure of workers is regulated in (inter)national legislation. Currently, the 1990 recommendations of the International Commission on Radiological Protection (ICRP), stating an occupational exposure limit [7] of 20 mSv/a, are being implemented into (inter)national legislation;
- the world-population averaged dose caused by the *(un)controlled release of artificial radionuclides* (e.g. atomic weapons testing, nuclear power production) amounts to 0.2  $\mu\text{Sv/a}$ , but will vary with time and with the actual geographical location of the individual. Exposure to artificial radionuclides is usually controlled by setting (inter)national disposal limits.

Like the other radiation doses, TENR doses are constituted from an external and an internal component. The ICRP recommends [7] to keep any radiation dose additional to the natural background dose (*vide supra*) to non-radiation workers and members of the public below 1 mSv/a.

- TENR doses to E&P industry workers are not yet been documented very well. In principle any occupational TENR dose due to the encounter NORM must be controlled through Standard Operational Procedures (SOP's) [5,8].
- Any potential TENR dose to the public at large is to be regulated by setting external dose-rate and NORM disposal limits. Currently environmental aspects related to E&P NORM disposal are being mapped out.

To acquire insight into the characteristics of E&P NORM an extended survey in the scattered literature has been carried out, the outcomes of which have been supplemented with analytical results [9] obtained by the Shell Research and Technology Centre, Amsterdam (SRTCA). This paper elucidates upon the origin of the NOR's, and the E&P industry product and waste in which these NOR's may appear in concentrations above legislative levels.

## NATURAL RADIOACTIVITY

NOR's can be subdivided into cosmogenic (e.g.  $^7\text{Be}$ ,  $^{14}\text{C}$ ) and primordial (e.g.  $^{40}\text{K}$ ,  $^{87}\text{Rb}$ ,  $^{138}\text{La}$ ,  $^{190}\text{Pt}$ ) radionuclides. While cosmogenic radionuclides are continuously formed in the outer atmosphere by cosmic irradiation, primordial (or terrestrial) radionuclides have been present ever since the planet *Earth* (life-time  $5.5 \times 10^9$  y) has been formed. The primordial radionuclides  $^{235}\text{U}$  (natural abundance = 0.72%,  $t_{1/2} = 7.1 \times 10^8$  y),  $^{238}\text{U}$  (99.27%,  $4.5 \times 10^9$  y) and  $^{232}\text{Th}$  (100%,  $1.4 \times 10^{10}$  y), are special, as these are heading a series of successive nuclear decays. Due to its low isotopic abundance,  $^{235}\text{U}$  is rarely encountered in practice and therefore this series will not further be discussed. Apart from the members of the  $^{238}\text{U}$  and  $^{232}\text{Th}$  series, NOR's do not generate a radiation hazard by any NNI activity.

Uranium and thorium are omnipresent in the *Earth's* crust at an average concentration of 4.2 and 12.5 ppm [10], corresponding to 0.05 becquerel  $^{238}\text{U}$  per gram ( $\text{Bq}[^{238}\text{U}]/\text{g}$ ) and 0.05  $\text{Bq}[^{232}\text{Th}]/\text{g}$  respectively. Apart from specific uranium and thorium ores, various natural substances (e.g. phosphate rock; mineral sands; aluminium, copper, lead, niobium, tantalum, tin, zinc ore) [1,2,3,4,5] show naturally enhanced concentrations of uranium and/or thorium.

The  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay schemes are given in Figure 1 and 2. When a geological formation containing  $^{238}\text{U}$  or  $^{232}\text{Th}$  has not been disturbed ('closed system') for more than a million years, the members of the

individual decay series will have the same activity concentration (Bq/g). In such a case the series are said to be in **secular equilibrium**. However, as the chemical concentration (mole/g) is proportional to the half-life time of the radionuclide, their number of atoms (concentration in [weight based] parts per million: ppm) varies considerably; e.g. 50 mBq[<sup>238</sup>U] and 50 mBq[<sup>226</sup>Ra] per gram correspond to about 4 ppm(U) and  $1.4 \times 10^{-6}$  ppm(Ra), respectively.

If a geological formation is not 'closed' to radionuclide migration, e.g. if <sup>226</sup>Ra becomes mobilised and is deposited/accumulated somewhere outside the formation, no secular equilibrium will exist. At the site of deposition/accumulation the only way the <sup>226</sup>Ra concentration will change, is by in/outflow through transport phenomena and radioactive decay. No ingrowth of <sup>226</sup>Ra via radioactive decay of its ancestors will occur, as all NOR's above <sup>226</sup>Ra (c.f. Fig. 1) remain with <sup>238</sup>U. As at the site of deposition/accumulation ingrowth of <sup>226</sup>Ra by decay does not occur, <sup>226</sup>Ra is said to be **unsupported**.

A NOR should 'live' long enough to develop the characteristic geochemical properties of the element. In oil/gas production two typical time scales are important, i.e. a *geological time scale* (say far longer than 1,000 y) and a *production time scale* (order of magnitude; days to decades).

## GEOCHEMICAL PROPERTIES OF URANIUM, THORIUM AND RADIUM

The formation of oil and gas is a process taking place on a geological time scale. Due to their half-life times only <sup>238</sup>U, <sup>234</sup>U ('uranium'), <sup>230</sup>Th, <sup>232</sup>Th ('thorium'), and <sup>226</sup>Ra ('radium') may be encountered unsupported in this process. In order to get a grips on the natural occurrence of these elements with respect to oil/gas formation, their (geo)chemical characteristics are summarised:

- **Uranium** [11, 12, 13, 14] exists under the 4<sup>+</sup> and 6<sup>+</sup> state of valence
  - \* U<sup>4+</sup> (as UO<sub>2</sub>) is generally insoluble and exists only in reducing, acidic (pH < 4) sulphurous hydro-thermal waters. By oxidation, U<sup>4+</sup> passes easily to valence 6<sup>+</sup> as UO<sub>4</sub><sup>2-</sup> or U<sub>2</sub>O<sub>7</sub><sup>2-</sup>.
  - \* U<sup>6+</sup> does not exist in solution, but forms the complex uranyl ion UO<sub>2</sub><sup>2+</sup>, which is very soluble and relatively stable under oxidising and acidic (pH < 2) conditions. An increase of pH induces hydrolysis leading to the formation of various types of ionic complexes.

In general, reducing conditions increase with the depth, because of the relatively low 'reactive' oxygen content, the presence of metal sulphides and organic matter, the activity of anaerobic bacteria, etc. As a consequence, the following inorganic uranium-forms are typical for sedimentary rocks, where oil/gas reservoirs are found:

- ◊ *sandstone* UO<sub>2</sub> (uranite) & USiO<sub>4</sub> (coffinite) U contents up to 1.5 ppm ( $\approx$  20 mBq[<sup>238</sup>U]/g)
- ◊ *limestone* (UO<sub>2</sub>)(CO<sub>3</sub>) U contents up to 2.5 ppm ( $\approx$  30 mBq[<sup>238</sup>U]/g)

Owing to its solubility UO<sub>2</sub><sup>2+</sup> is chiefly transported in solution. However, under reducing conditions UO<sub>2</sub><sup>2+</sup> forms numerous complexes with organic compounds (e.g. humic acids), which facilitates uranium fixation by organic sediments (i.a. peat, lignite and coal) and mineral matter. Localisation of uranium in organic shales (up to 20 ppm or 250 mBq[<sup>238</sup>U]/g) is another typical example of this fixation. These organic substances are particularly important in absorption of uranium from water. Hence, the concentration for uranium in ground waters is low (0.01 to 5 ppm corresponding to 0.1 to 60 mBq[<sup>238</sup>U]/g).

Thermal diagenesis of organic matter, which produces hydrocarbons (*vide infra*), will enhance the uranium concentration, as uranium remains with the residual organic matter and does not follow the hydrocarbons produced [15].

- **Thorium** [12, 13, 14, 16] exists only at valence state 4<sup>+</sup>, and has a very stable degree of oxidation. All its compounds are insoluble, the solubility product for e.g. Th(OH)<sub>4</sub>, is close to  $10^{-42}$ . During weathering, thorium is easily hydrolysed and therefore has a limited mobility and a tendency to concentrate in residual minerals (bauxite, clay, etc.). Owing to its large ionic radius, Th<sup>4+</sup> is likely to be absorbed between the platelets on clay minerals. Its amount depends on the pH and on the abundance of other cations.

Owing to its insolubility, thorium is almost wholly transported in suspension. Thus, it concentrates in the silty fraction of shale as thorium minerals or as thorium-bearing accessory minerals. The following thorium-forms are typical for sedimentary rocks:

- ◊ *sandstone* ThO<sub>2</sub> (thorianite) & ThSiO<sub>4</sub> (thorite) Th contents up to 5 ppm ( $\approx$  20 mBq[<sup>232</sup>Th]/g)
- ◊ *limestone* Th contents up to 1.1 ppm ( $\approx$  5 mBq[<sup>232</sup>Th]/g)

Humic substances are also important in absorption of thorium from water. Hence, the thorium concentration in ground waters is very low ( $\pm$  0.007 ppb, corresponding to 0.3  $\mu$ Bq[<sup>232</sup>Th]/g).

- From a geological point of view the only important isotope of radium [12,13,17,18] is <sup>226</sup>Ra (1,600 y), which due to its tight relation with <sup>238</sup>U has an average concentration in the *Earth's* crust of about 10<sup>-12</sup> gram <sup>226</sup>Ra per gram crust (corresponding to 40 mBq[<sup>226</sup>Ra]/g).

Despite this low concentration in nature very often selective leaching of radium is observed, so that radium - regardless its tight relation to uranium - shows its own geochemical characteristics. The mobilisation of radium by leaching from minerals or rocks is governed by various factors like the physical mineral/rock condition, the nature of the radium occurrence in mineral/rocks, the chemical composition of the leaching waters. The primary migration or leaching process can be described in three stages:

- \* as a consequence of radioactive decay of its parents ( $\alpha$ -recoil) radium isotopes get into the water containing capillaries of the minerals/rocks;
- \* at the walls of the capillaries an adsorption equilibrium between radium at the wall and the radium in the capillary water is established; and
- \* caused by diffusion an equilibrium between the radium in the capillaries and the radium in so-called gravitation waters is established.

This migration is enhanced by reducing and acidic (pH < 7) conditions. Besides, the ionic composition of the leaching waters strongly increase radium mobility, in particular a high chlorine and low sulphate and carbonate concentration. Also cations may increase the transfer of radium into gravitational water, the influence decreases according the following series: H<sup>+</sup>>Ba<sup>2+</sup>>Pb<sup>2+</sup>>Sr<sup>2+</sup>>Ca<sup>2+</sup>>Mg<sup>2+</sup>>Na<sup>+</sup>>K<sup>+</sup>. The great radium mobility in a reducing zone is in strong contrast to its immobility in an oxidising zone.

All processes driven by the mobility of formation waters in a porous formation, e.g. radium transport, precipitation, adsorption, are indicated as secondary migration. During this migration radium may be transported as ions. Radium often attains significantly different concentrations due to its far better solubility as a free ion and its tendency to become stabilised in high ionic strength solutions, yielding <sup>226</sup>Ra concentrations to over 100 Bq/L for saline ground waters [19].

## OIL/GAS ORIGINATION vs. NATURAL RADIOACTIVITY

Hydrocarbons are formed [20] by thermal cracking of organic matter trapped in sedimentary rock (**source rock**; c.f. Fig. 3). With increasing burial, the temperature of the source rock increases and, after a given time at a given temperature ('source rock maturity'), the organic matter (kerogen) is gradually transformed into oil and/or gas. The most important oil/gas source rocks are *carbonates*, *limestones*, *shales* and *coal*. After expulsion from the source rock, oil and gas migrate either to a reservoir formation (**reservoir rock**) where it is trapped, or to the *Earth's* surface where it escapes as a seepage. Migration is not fully understood, but is thought to take place along faults and minor fractures within the rock sequence (**'carrier' rock**). As hydrocarbons are lighter than water, they generally migrate in an upwards or sideways direction from areas of higher to lower pressure.

An oil/gas reservoir is a porous *sedimentary* rock formation, capped with a layer of impermeable rock or salt (**cap rock**), through which liquids and gas cannot pass. The shape of a reservoir must allow gas or oil to accumulate, and the cap rock is essential to prevent them from migrating further upwards. Because of capillary forces, some of the water originally in the pores could not be displaced by the accumulating hydrocarbons (so-called connate or interstitial water). The volume of all pores and openings in a reservoir rock (porosity) ranges from 10 to 30% of total rock volume.

Discovered oil/gas reserves stem from various geological periods: 14% of the oil and 29% of the gas is of *Palaeozoic* origin; 54% of the oil and 44% of the gas is of *Mesozoic* origin and 32% of the oil and 27% of the gas is of *Cenozoic* (*Tertiary*) origin. In Western Europe, major source rocks originate from the Jurassic [North Sea crude oil/gas], from the *Permian* (*Rotliegend*) and *Early Triassic* [Southern North Sea, North Germany] and from the *Late Carboniferous* [Groningen gas]. The presence of natural radioactivity in these

geological strata has been recognised world-wide, and exploited as a means of identifying oil bearing formations, for more than 30 years. As a result of clay minerals, the main NOR found in common sedimentary rocks is  $^{40}\text{K}$  (0.0117%,  $1.4 \times 10^9$  y). Potassium levels in shale are resulting from the clay minerals illite and K-feldspar. Despite this, the presence of members of the decay series of  $^{238}\text{U}$  and  $^{232}\text{Th}$  can also be shown by the Natural Gamma Tool. Some typical NOR concentrations, relevant for reservoir strata [10,21], are listed in table 1.

Dependent on the original concentration of  $^{238}\text{U}$  and  $^{232}\text{Th}$  in type of 'rocks' mentioned above, members of the  $^{238}\text{U}$ - and  $^{232}\text{Th}$ -decay series may become contaminants of radiological importance. As the oil/gas migrates, NOR's may be taken up in the up-flowing fluid/gas stream. Also NOR's may be transferred into connate waters, potentially partly co-produced with oil/gas as production water.

- Source rock: uranium is predominantly associated with organic matter [15,22]. In samples with high total organic carbon (> 7%) uranium values range up to 1,100 ppm. For humic substances (as found in peat and bog, *i.e.* kerogen precursors) enrichment factors relative to ground water of up to 50,000 have been reported.

During thermal cracking of the kerogen (thermal diagenesis) uranium or thorium generally remain with the residual organic matter and, hence, are not carried with the hydrocarbons produced<sup>5</sup> [15]. In outcrop bitumens, uranium concentrations of 0.2 - 0.8%(m/m) (25 - 100 Bq[ $^{238}\text{U}$ ]/g) have been reported [22].

- 'Carrier' Rock: hydrocarbons, expelled from mature source rocks, may migrate along faults and minor fractures within the rock sequence, through one or more carrier beds having similar permeability and porosity as reservoir rocks, before being trapped by an impermeable or a very low permeability barrier forming an oil/gas reservoir.

Since practically all pores in the subsurface are water-saturated, movement of hydrocarbons within the network of capillaries and pores has to take place in the presence of the aqueous pore fluid. Such movement may be due to active water flow or may occur independently of the aqueous phase, either by displacement or by diffusion. There may be a single phase (oil and gas dissolved in water) or a multi-phase (separate water and hydrocarbon phases) fluid system.

Leaching of uranium and thorium in a reducing environment from the rocks passed either into passing fluids will not occur. Radium will also not leach into a hydrocarbon phase, but it may leach into the aqueous phase. When in addition gravitational displacement of this phase occurs (secondary migration), radium may ultimately be transported to the reservoir. The amount progressing into a reservoir is dependent on the amount of uranium present in 'carrier rock', the amount of radium leached, its transport time relative to the half-life time of  $^{226}\text{Ra}$  and the transport efficiency.  $^{226}\text{Ra}$  may appear up to a kilometre from its place of 'birth' [19].

- Reservoir Rock: reservoir material does not contain naturally enhanced radioactivity (*c.f.* table 1). Connate waters have been trapped in a potential reservoir before actual oil/gas accumulation occurred. As these waters have generally been at rest, *i.e.* no displacement during a geologically short period of one million years, NOR's present must originate from the reservoir material itself. In such a reducing environment only radium may leach from reservoir rock into connate water.

With a view on the geological time scale of the processes described thus far, enhanced  $^{226}\text{Ra}$  concentrations in connate water will most likely be caused by the natural radioactivity present in reservoir material. A model explaining that these concentrations are originating from normal (*c.f.* table 1) uranium and thorium levels in reservoir rock, has been proposed [22].

Some reservoir strata may contain thin shale or coal layers/spots containing naturally enhanced concentrations of uranium. Sometimes these layers/spot are mentioned as the reason for the enhanced radium concentrations in connate waters.

- Cap Rock: material is impermeable for hydrocarbons and gases and is often constituted of evaporites, *e.g.* salt layers, or shales. As these salt layers are generally formed by evaporation of ancient sea waters, the main NOR will be  $^{40}\text{K}$ , while leaching only occurs at the interface.

When oil/gas reservoirs are capped by black shales, containing relatively high amounts of natural

<sup>5</sup> If nevertheless under production conditions uranium or thorium transport occurs, it will be contained in the heavy oil fraction (bituminous residue), which is not produced in conventional oil recovery.

radioactivity (e.g. *Kimmeridgean* in the northern North Sea [23]), only at the interface of cap and reservoir rock (containing connate water) radium leaching might occur.

## OIL/GAS PRODUCTION

Under forced flow (or production) conditions potential exchange of NOR's with reservoir fluids has to be re-examined on a production time scale. Next to the NOR's of the previous section  $^{230}\text{Th}$ ;  $^{228}\text{Th}$  ('thorium'),  $^{228}\text{Ra}$ ,  $^{224}\text{Ra}$  ('radium'),  $^{222}\text{Rn}$  ('radon'),  $^{210}\text{Pb}$  ('lead') and  $^{210}\text{Po}$  ('polonium') have to be considered.

However, due to the half-life time of these NOR's their range of transport will be limited. Consequently, only NOR's in reservoir material and cap rock have to be considered. Transfer *c.q.* transport phenomena of these NOR's will shortly be discussed:

- Transport of uranium and thorium in production water is confined to the suspended particles fraction [24, 25, 26]. So any activity concentration in these particles will more or less reflect the natural uranium and thorium level of the reservoir rock (*c.f.* table 1).

The chemistry of an element is virtually independent of its isotope. Consequently, the chemical behaviour of thorium will not be different from the phenomena described for a geological time scale; *i.c.*  $^{230}\text{Th}$  (24 d) and  $^{228}\text{Th}$  (1.9 y) will not be leached in reservoir fluids.

- Radium leaches into connate water, which may ultimately be co-produced with oil/gas as production water. Dependent on the rate of diffusion of radium ions leached from reservoir rock into formation waters, also  $^{228}\text{Ra}$  (5.8 y) and  $^{224}\text{Ra}$  (3.7 d) may be encountered unsupported in production water. The rate of diffusion under forced flow conditions is relatively fast, because  $^{224}\text{Ra}$  (in relative absence of  $^{228}\text{Th}$ ) has been noticed in freshly produced water [27].
- Radon is a noble gas, which emanates from reservoir material, from cap rock or from connate water containing  $^{226}\text{Ra}$ . Dependent on the filling of the pores of the reservoir, it prefers the Natural Gas (NG) phase. In absence of NG it dissolves in the (light) hydrocarbon and aqueous phase (partitioning). However, at low pressures  $^{222}\text{Rn}$  (3.8 d) will always appear in the gas phase.
- Some connate waters are containing high concentrations of lead (> 100 mg/L) and zinc (> 300 mg/L) [28]. This lead is enriched in radiogenic stable lead isotopes, particularly  $^{206}\text{Pb}$  (*ex*  $^{238}\text{U}$ -series) at the expense of the only non-radiogenic stable lead isotope ( $^{204}\text{Pb}$ ). Besides these waters appear to contain relatively high concentrations of  $^{210}\text{Pb}$  (22 y) in absence of high  $^{226}\text{Ra}$  concentrations. This suggests that a selective lead transport *c.q.* leaching process is operational.

In relatively dry NG wells, sometimes lead deposits are observed, with lead stable isotope ratios [29] comparable to those described in the preceding paragraph. As water can not be the transport medium an independent transport of lead compounds via the gas phase is hypothesised. It is clear that both for the connate water and the dry NG wells stable lead serves as carrier for  $^{210}\text{Pb}$ .

Relatively light oil\*\* (hydrocarbon condensate or Natural Gas Liquids (NGL)), which is produced from some wells, sometimes also show enhanced concentrations of  $^{210}\text{Pb}$ . Though several reasons for this observation can be hypothesised, none of these has been investigated yet.

- As there are no stable isotopes of polonium, knowledge of its (bulk) chemistry is scarce. The observation of unsupported  $^{210}\text{Po}$  in NGL was therefore surprising [30]. Fortunately, the occurrence of unsupported  $^{210}\text{Po}$  is very rare.

## NOR CONCENTRATIONS IN E&P (BY-)PRODUCTS

Next to the analytical results obtained by SRTCA [9], a world-wide inventory of NOR concentrations in E&P (by-)products and waste streams was made. To facilitate comparison, reported concentrations have been converted to the SI-unit Bq, and are presented either as Bq/g, Bq/L or Bq/m<sup>3</sup>.

- In line with their geochemical properties and productional transport phenomena maximum reported concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Po}$  or  $^{232}\text{Th}$  in (produced) crude oil are low (table 2).

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\*\* technically: crude oil is a mixture of hydrocarbons existing in the liquid phase in reservoirs and remains liquid at atmospheric pressures, while NGL being liquid in the reservoir become gaseous at atmospheric pressures.

- $^{222}\text{Rn}$  will be transported to the surface with natural gas, or with crude oil, from which it will be removed during de-gasification (associated gas). Under production conditions  $^{222}\text{Rn}$  transport will mainly be determined by convection (Darcy's law). For a gas well a  $^{222}\text{Rn}$  convective transport length per half-life time of ca. 50 m is computed (production:  $5 \times 10^6 \text{ m}^3 \text{ NG (STP)}$ , reservoir: 20% porosity, 250 bar, 200 °C). Therefore, for a gas well as a rule of thumb:  $^{222}\text{Rn}$  originates from a sphere with radius 50 m around the centre of the perforation.

The reported concentration range of  $^{222}\text{Rn}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in Bq per cubic meter NG at standard temperature (21 °C) and pressure (1 bar) or STP is presented in table 3.

- Dependent on the pressure  $^{222}\text{Rn}$  will partition between all produced phases; crude oil, NG, natural gas liquids and water. There are no reports on  $^{222}\text{Rn}$  concentrations in crude oil and water, as it will be released at de-pressurisation. NGL is a valuable product, it is used for the production of base chemicals [31]. During fractionating of NGL  $^{222}\text{Rn}$  concentration factors of about 20 (gas volume base) and about 1,000 (after liquefaction) in the propane/propene fraction (table 4) have been observed [9,30,32].

Despite their low levels in NG (*c.f.* table 3) both  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  seem to occur unsupported in hydrocarbon condensate/NGL. However, the reason for this phenomenon is not well understood.

- In 1928 the first report on enhanced levels of radioactivity in production water, which are characterised by their high salinity ('brine'), was published [33]. Following this observation, oil fields have been extensively screened [24,34] in the 50's as a potential source for deposits of uranium. However, it turned out that the levels of radioactivity were due to enhanced levels of dissolved radium ions (table 5), while only minute quantities of uranium and thorium were demonstrated to be present [23,25,26,34,35,36,37,38,39]. Dis-equilibrium factors for  $^{238}\text{U}/^{226}\text{Ra}$  and  $^{232}\text{Th}/^{226}\text{Ra}$  up to  $1 \times 10^{-5}$  and  $4 \times 10^{-4}$  have been determined respectively [24,25,26,34,40]. Because of its high salinity, on-shore produced water is re-injected into injection or disposal wells, while off-shore it may be re-injected or disposed overboard.

A strong enhancement of radium concentrations relative to  $^{238}\text{U}$  or  $^{232}\text{Th}$  in the aqueous phase is also observed in many other (non E&P industry) cases; in a deep drilling project in Germany (over 100 Bq[ $^{226}\text{Ra}$ ]/L) [19], in geothermal waters (up to 60 Bq[ $^{226}\text{Ra}$ ]/L) [41], in coal mining (up to 390 Bq[ $^{226}\text{Ra}$ ]/L) [42], during ore processing (up to 10 Bq[ $^{226}\text{Ra}$ ]/L and 30 Bq[ $^{226}\text{Ra}$ ]/L) [43]. Therefore, the occurrence of unsupported  $^{226}\text{Ra}$  and/or  $^{228}\text{Ra}$  is a frequently observed, natural phenomenon and not explicitly/exclusively related to the occurrence of oil/gas reservoirs.

## NOR CONCENTRATIONS IN E&P WASTE STREAMS

The (sub)surface facilities for crude oil and NG production and subsequent treatment are very schematically given in figure 4. The process can shortly be characterised as follows:

- at the perforation in the reservoir, entering well fluids are transported via down hole production tubing ('tubulars') to the well head. The flow from several wells are commingled in the production manifold, after which it is led to subsequent separators for de-gasification and de-hydration. Inside a flotation cell the removed water is skimmed to remove any remnant oil. Produced oil is pumped through a metering package to the sales terminal.
- till the production manifold, a similar route is followed for gas production. The produced gas is separated from associated fluids, in a (hydrocarbon) condensate/water separator. The separated gas is heat treated and sent to absorbers to remove any water and other unwanted products (*e.g.*  $\text{CO}_2$ ,  $\text{H}_2\text{S}$ ), after which it is transported to the sales terminal.

Produced water may be disposed off or (re)injected into injection/disposal wells. Water flooding of a reservoir by reinjection of production water (off-shore: mostly sea water), below the oil/gas reservoir compensates for pressure decrease and extends the production life-time of a reservoir.

However, scales may cause problems in the operation of installations by plugging perforations, by clogging tubulars and valves, thereby restricting flow. Such plugging *c.q.* clogging results in production losses and necessitates costly cleaning exercises. The scales are formed at spots, where the solubility product of a particular pair of ions is exceeded; as brine (relatively high concentrations of calcium, barium and/or strontium, and low concentrations of sulphate and/or carbonate) is combined with injected



(sea)water, which may be relatively high in sulphate and/or carbonate, so-called scales will be preferentially deposited at spots, where severe gradients (temperature, pressure, concentrations, flow velocity *etc.*) are present [44]. Though the occurrence of LSA scales have frequently been reported for water flooded reservoirs, it should be noted that the (sea)water injection is not a prerequisite for the encounter of LSA scales [7,8,9].

The co-precipitation of radium with group IIa ions in (sulphate/carbonate) scales or sludges leads to enhanced external radiation levels. Spots in production *c.q.* treating facilities, where LSA scale/ sludge has been encountered, are schematically indicated in figure 4. The occurrence of LSA scale/ sludge is quite unpredictable and is not restricted to certain regions, but may be encountered all over the world [8,25,26,37,38,45,46,47,48,49]. Roughly three types of LSA waste are distinguished:

- Hard LSA scales may be deposited in tubulars close to the perforation, at well heads, at the production manifold and at certain spots (flanges, valves and pumps) in production flow lines [5,8,23,25,26,35,36,45,46,50,51,52,53,54]. Activity concentrations are presented in table 6.  
This type of scale is not limited to oil/gas production and also appears during coal mining (up to 200 Bq[<sup>226</sup>Ra]/g) [42] and ore processing (up to 400 Bq[<sup>226</sup>Ra]/g and 1,600 Bq[<sup>228</sup>Ra]/g) [43].
- Further downstream in a production installation (*e.g.* production piping, filters, vessels, at the water outlet of flotation cells, production water disposal/injection wells, overboard discharges) and associated equipment, soft to medium hard scales may be deposited [8]. Again the main NOR's in these scales are the progeny of unsupported radium isotopes (table 7).  
In some gas and oil wells medium hard to soft LSA scales, constituting of more than 50%(w/w) Pb may be encountered [5,8,25,29]. The material is deposited at spots with disturbed flow (*e.g.* close to flanges and valves, in bends, at connections in well tubing and near well heads) [52]. The first NOR of the <sup>238</sup>U-series encountered is (unsupported) <sup>210</sup>Pb.
- LSA sludges are deposited at various spots (*e.g.* pipeline bottoms, close to filters), but mainly in voluminous equipment ([NGL] storage tanks, separator and dehydration vessels, LPG delivery facilities) in the production train [5,8,37,52]. This material is deposited mainly because of hold up times in the large tanks and vessels. Generally, the activity concentration of this oily sludge is comparable to that of medium hard to soft LSA scales (table 8).  
The composition of an oily sludge is very variable; it may contain fine clay, sand, silt, iron oxide (rust: most likely acting as absorptive material for radium ions), abraded hard LSA scale. The main part of the activity level may also be caused by unsupported <sup>210</sup>Pb. During oil production <sup>226</sup>Ra/<sup>210</sup>Pb ratios [29] as low as 0.01, while during gas production <sup>226</sup>Ra/<sup>210</sup>Pb ratios [9,29] far less than 0.0001 are observed.  
Sludges, in which enhanced levels of radium or lead are present, are not limited to oil/gas production and also may appear in *i.a.* thermal spas (up to 1,200 Bq[<sup>226</sup>Ra]/g, 360 Bq[<sup>210</sup>Pb]/g and 70 Bq[<sup>228</sup>Ra]/g) [55].

Crude oil or gas transmission lines have to be inspected at regular time intervals in order to remove flow hampering wall deposits or to trace potential malfunctioning at an early stage. For this purpose, spheres ('sphering') or pipeline inspection gauges are sent through these lines, thereby also generating debris or scrapings. Encountered activity concentration are presented in table 9. Again the first radionuclides of the <sup>238</sup>U and <sup>232</sup>Th decay series encountered are <sup>226</sup>Ra and <sup>228</sup>Ra, respectively, but occasionally <sup>210</sup>Pb concentrations higher than that of <sup>226</sup>Ra have been found; *i.e.* <sup>226</sup>Ra/<sup>210</sup>Pb ratios as low as 0.1 [9,29].

The occurrence of LSA scales/sludges is also associated with contamination [21] of the inside of equipment with NOR's:

- in *crude oil* production/treating facilities due to thin films of scales (initial state of formation) or to deposition of NOR's from the various streams
  - ◊ unsupported <sup>226</sup>Ra and/or <sup>228</sup>Ra, and occasionally unsupported <sup>210</sup>Pb;
- in (*associated*) NG production/treating due to thin, sometimes hardly visible, films of long-lived progeny of unsupported <sup>222</sup>Rn, *i.c.* <sup>210</sup>Pb
  - ◊ <sup>210</sup>Pb (after ingrowth in secular equilibrium with <sup>210</sup>Po), and occasionally unsupported <sup>210</sup>Pb;
- in NGL processing/treating facilities the additional enhancement of the <sup>222</sup>Rn concentration level in the propane/propene fraction may lead to thin invisible films of <sup>210</sup>Pb within propane/propene storage and

processing equipment [31,32,56 ,57 ,58 ]. Severe surface contamination inside pumps (up to 7,000 Bq[<sup>210</sup>Pb]/cm<sup>2</sup> has been observed [31,56,57,58,59 ,60 ]

◇ <sup>210</sup>Pb (after ingrowth in secular equilibrium with <sup>210</sup>Po), and unsupported <sup>210</sup>Pb and rarely unsupported <sup>210</sup>Po.

The occurrence of LSA scales/sludges inside (parts of) installations is generally associated (*vide supra*) with levels of external (γ)-radiation enhanced with respect to natural background γ-radiation:

- in crude oil production/treating facilities due to short-lived progeny of unsupported <sup>226</sup>Ra , ([via <sup>222</sup>Rn] <sup>214</sup>Pb and <sup>214</sup>Bi [c.f. Fig 1]), and/or unsupported <sup>228</sup>Ra, (<sup>228</sup>Ac, [via <sup>228</sup>Th/<sup>224</sup>Ra] <sup>212</sup>Pb and <sup>208</sup>Tl [c.f. Fig 2]). Its occurrence may be demonstrated by standard external radiation, intrinsically safe monitoring equipment. The external radiation level will persist until the LSA scale, sludge or contamination has been removed.
- in NG, associated gas and NGL production/processing plants and storage tanks due to fast ingrowth of short-lived progeny of unsupported <sup>222</sup>Rn, (notably <sup>214</sup>Pb and <sup>214</sup>Bi [c.f. Fig 1]). After stopping operations the external (γ)-radiation level decreases with the half-life of <sup>222</sup>Rn; after venting even with an apparent half-life of about 40 minutes.
- ⊗ the occurrence of unsupported <sup>210</sup>Pb and/or <sup>210</sup>Po is not associated with increased levels of external radiation. Therefore, this occurrence can only be proven through a sampling program and subsequent dedicated analysis, or through a survey of the interior of the installations with the aid of dedicated monitors/detectors; e.g. dedicated surface α/β-contamination monitoring equipment, which cannot be made intrinsically safe, but may be used within the context of a permit-to-work system.

Many reports state the presence of enhanced external radiation levels. In the US even a national survey of (increased) radiation levels associated with oil/gas production and gas processing has been carried out [61 ]. Radiation levels as encountered in the E&P industry are presented in table 10.

## RADIONUCLIDE CHARACTERISATION OF E&P NORM

From the extensive (literature) data presented above, it is observed that both <sup>238</sup>U and <sup>232</sup>Th are only present in minute concentrations, mainly in production water and associated scale/sludge. The encounter of <sup>238</sup>U and <sup>232</sup>Th is particularly associated with the occurrence of suspended (fine) cutting, sand and clay mineral particulates, and hence the <sup>238</sup>U and <sup>232</sup>Th concentrations will be non-enhanced with respect to their concentration in reservoir rock constituting minerals (c.f. Table 1). As these <sup>238</sup>U and <sup>232</sup>Th concentrations are far below the most stringent (proposed) levels for free disposal no radiological hazards are expected.

Furthermore, it is observed that the radium isotopes actually mark a clear separation line for the encounter of NORM in oil/gas production, treating or processing facilities. The most likely cause for this separation line is a distinct difference in geochemical properties (mobility under reservoir conditions) in combination with the 'effective'<sup>††</sup> half-life' of <sup>238</sup>U and <sup>232</sup>Th on one hand, and that of <sup>226</sup>Ra and <sup>228</sup>Ra and their progeny on the other hand. Therefore, it can be stated that no radionuclides above <sup>226</sup>Ra (*i.c.* <sup>238</sup>U, <sup>234</sup>Th, <sup>234m</sup>Pa, <sup>234</sup>U and <sup>230</sup>Th; c.f. Fig. 1) and <sup>228</sup>Ra (<sup>232</sup>Th; c.f. Fig. 2), respectively, will be present in this type of NORM. This is typical for E&P NORM in comparison with NORM encountered in ore processing industries [4,43], where these isotopes will be present in one of the waste (or, less likely, product) streams.

As unsupported <sup>224</sup>Ra may be encountered in fresh production water its activity concentration should be applied for the characterisation of E&P NORM. As the <sup>224</sup>Ra-subseries (c.f. Fig. 2) is rather short-lived (3.7 d) all its activity disappears on short term. Consequently, the <sup>224</sup>Ra activity is not required for characterisation. Due to the half-lives involved the equilibration of the <sup>228</sup>Ra-subseries is a slow process as <sup>228</sup>Th (1.9 y; c.f. Fig. 2) is part of this series. From a radiological protection point of view <sup>228</sup>Th is a

<sup>††</sup> The 'effective half-life' of a radioisotope under reservoir conditions is defined as a linear combination of the 'physical' half-life ( $t_{1/2}$ ) of the radionuclide and its 'mean residence time' in a phase during production from the reservoir or even during maturation, migration or accumulation processes in the subsurface.

crucial NOR. Therefore, its activity concentration is essential for E&P NORM characterisation. As the only way  $^{228}\text{Th}$  may appear at surface facilities is by ingrowth, its maximum concentration will only become equal to the  $^{226}\text{Ra}$  concentration after about 7 years after initial deposition. Consequently the assumption of secular equilibrium in the  $^{228}\text{Ra}$ -subseries is a conservative one.

For relatively dry NG wells,  $^{222}\text{Rn}$  forms a second distinct separation line for the potential encounter of NORM. The arguments presented above, indicate that  $^{222}\text{Rn}$  is stemming from reservoir rock near the perforation. In general, the determination of the  $^{222}\text{Rn}$  concentration in NG is included in the standard gas analysis during well-testing. Also during the production life time of a well the  $^{222}\text{Rn}$  concentration is regularly monitored. It has often been observed that with increasing depletion of a gas reservoir due to production also an increasing  $^{222}\text{Rn}$  concentration is found. In case high  $^{222}\text{Rn}$  concentrations are encountered at the well head, NG and NGL treating/storage facilities should be screened on contamination with  $^{222}\text{Rn}$  progeny, particularly  $^{210}\text{Pb}$  films.

Based on the literature survey presented above, it is observed that also  $^{210}\text{Pb}$  may mark an 'independent' separation line in the  $^{238}\text{U}$  decay series, as this radionuclide can be encountered unsupported. When carried with the water phase, this is likely caused by its specific (geo)chemical properties, but also indications exists that  $^{210}\text{Pb}$  might be transported through the gas phase [29,62].

Only a few cases of unsupported  $^{210}\text{Po}$  in hydrocarbon condensates have been reported; however, one should be aware that this radionuclide may be encountered.

Starting from this radionuclide characterisation of E&P NORM, screening and surveys for its encounter should be carried out in order to protect the workforce from uncontrolled exposure to TENR, both during normal operations and during maintenance stops [5]. Besides, in this way uncontrolled dispersal of radioactive materials into the environment is prevented.

Any enhanced external radiation levels can be used as indication for the occurrence of NORM, but as both unsupported  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  do not give rise to enhanced external radiation levels, the absence of external radiation is not conclusive. Therefore, both production streams -- *i.c.* NG ( $^{222}\text{Rn}$ ) and NGL ( $^{222}\text{Rn}$ ,  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$ ) -- and waste streams -- *i.c.* production water, scale, sludge and scrapings (all:  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ , (rarely  $^{210}\text{Po}$ ),  $^{228}\text{Ra}$  and  $^{228}\text{Th}$ ) -- should be sampled during operations *c.q.* maintenance stops, and analysed by a standardised procedure [63] for the relevant NOR's. The analytical results will be used for an assessment of potential radiological safety and environmental hazards.

## CONCLUSIONS

In summary, it is concluded that for a radionuclide characterisation of E&P NORM

- **no screening for NOR's** in the top of the  $^{238}\text{U}$  decay series ( $^{238}\text{U}$ ,  $^{234}\text{Th}$ ,  $^{234\text{m}}\text{Pa}$ ,  $^{234}\text{U}$ ,  $^{230}\text{Th}$ ) and for  $^{232}\text{Th}$  is required as the first radionuclides encountered will either be  $^{226}\text{Ra}$  (*c.f.* Fig. 1), and/or  $^{228}\text{Ra}$  (*c.f.* Fig. 2); and that

from a Health, Safety and Environmental (HSE) point of view E&P NORM is fully characterised by analysing samples (if relevant) for

- $^{226}\text{Ra}$  (secular equilibrium with  $^{222}\text{Rn}$ ,  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ ) and/or
- $^{222}\text{Rn}$  (secular equilibrium with  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ )
- $^{210}\text{Pb}$  (secular equilibrium with  $^{210}\text{Bi}$  and  $^{210}\text{Po}$ ) and/or
- $^{210}\text{Po}$  and/or
- $^{228}\text{Ra}$  (secular equilibrium with  $^{228}\text{Ac}$ ) and/or
- $^{228}\text{Th}$  (ingrowth via  $^{228}\text{Ac}$  secular equilibrium with  $^{220}\text{Rn}$ ,  $^{216}\text{Po}$ ,  $^{212}\text{Pb}$ ,  $^{212}\text{Bi}/^{208}\text{Tl}$  and  $^{212}\text{Bi}/^{212}\text{Po}$ )

Table 1 - Mean and range of potassium, thorium and uranium concentration in sedimentary rocks<sup>10,21</sup>.

Sedimentary Rock Class ↓	potassium (K)			thorium (Th)			uranium (U)		
	(%)		Bq <sup>40</sup> K/g	(ppm)		Bq <sup>232</sup> Th/g	(ppm)		Bq <sup>238</sup> U/g
	mean	range	mean	mean	range	mean	mean	range	mean
<b>DETRITAL</b>									
Sandstone & Conglomerate	2.1	0.01 - 9.7	0.7	12.4	0.2 - 362	0.05	4.8	0.1 - 80	0.06
Orthoquartzites	1.2	0.1 - 8.5	0.4	9.7	0.7 - 227	0.04	4.1	0.1 - 62	0.05
Arkoses	2		0.06	1.5 5		0.006 0.02	0.5 1.5	0.5 - 3	0.005 0.02
Shale grey/green black	3.5 3	0.9 - 8.5	1.1 0.9	16.3 13	5.3 - 39	0.07 0.05	5.9 3	0.9 - 80 3 - 4 8 - 20	0.07 0.04
Clay	0.6	0.1 - 2.6	0.2	8.6	1.9 - 55	0.03	4.0	1.1 - 16	0.05
<b>CHEMICAL</b>									
Carbonates	0.6	0.02 - 8.4	0.2	14.9	0.03 - 132	0.06	3.6	0.03 - 27	0.04
Limestone	0.3 0.5	0.01 - 3.5	0.09 0.2	1.8 3	0 - 11	0.007 0.01	2.0 13	0.03 - 18	0.02 0.16
Evaporites							< 0.1		< 0.001

Table 2 - Activity concentration of <sup>238</sup>U, <sup>226</sup>Ra, <sup>210</sup>Po and <sup>232</sup>Th in crude oil

Radionuclide	Reported Range (Bq/g)	References
<sup>238</sup> U	0.0000001 - 0.01	9,15,24,25,27,64 ,65 ,66 ,67 ,68 ,69 ,70
<sup>226</sup> Ra	0.0001 - 0.04	24,27
<sup>210</sup> Po	0 - 0.01	9,71
<sup>232</sup> Th	0.00003 - 0.002	9,25,72 ,73

Table 3 - Activity concentration of <sup>222</sup>Rn, <sup>210</sup>Pb and <sup>210</sup>Po in natural gas

Radionuclide	Reported Range (Bq/m <sup>3</sup> )	References
<sup>222</sup> Rn	5 - 200,000	4,9,25,30, 32,34,35,62,71,74 ,75 ,76
<sup>210</sup> Pb	0.005 - 0.02	74
<sup>210</sup> Po	0.002 - 0.08	9,30,74

Table 4 - Activity concentration of <sup>210</sup>Pb and <sup>210</sup>Po in NGL/hydrocarbon condensate

Radionuclide	Reported Range (Bq/L)	References
<sup>222</sup> Rn (NGL)	0.01 - 1,500	9,30,31,32,56
<sup>222</sup> Rn (C <sub>3</sub> .liq)	0.01 - 4,200	9,30,31,32,56
<sup>210</sup> Pb	0.3 - 230	9,31
<sup>210</sup> Po	0.3 - 100	8,9,30,56

Table 5 - Activity concentration of <sup>238</sup>U, <sup>226</sup>Ra, <sup>210</sup>Pb and <sup>232</sup>Th, <sup>228</sup>Ra, <sup>224</sup>Ra in production water

Radionuclide	Reported Range (Bq/L)	References
<sup>238</sup> U	0.0003 - 0.1	22,25,26,27,34,38,70
<sup>226</sup> Ra	0.002 - 1,200	9,17,22,24,25,26,27,33,34,35,36,38, 77 ,78 ,79 ,80 ,81
<sup>210</sup> Pb	0.05 - 190	22,25
<sup>232</sup> Th	0.0003 - 0.001	22,25,26,27,38
<sup>228</sup> Ra	0.3 - 180	9,24,25,27,33,35,36,80
<sup>224</sup> Ra	0.5 - 40	24,25

**Table 6 - Activity concentration of <sup>238</sup>U, <sup>226</sup>Ra, <sup>210</sup>Pb, <sup>232</sup>Th and <sup>228</sup>Ra in hard scales**

Radionuclide	Reported Range (Bq/g)	References
<sup>238</sup> U	0.001 - 0.5	26,35,47,82
<sup>226</sup> Ra	0.1 - 15,000	8,9,23,26,27,34,36,37,39,46,47, 50,51,52,70,82,83
<sup>210</sup> Pb	0.02 - 75	9,25,39
<sup>210</sup> Po	0.02 - 1.5	39
<sup>232</sup> Th	0.001 - 0.002	26,39
<sup>228</sup> Ra	0.05 - 2,800	8,9,34,36,39,47,73,83

**Table 7 - Activity concentration of <sup>238</sup>U, <sup>226</sup>Ra, <sup>210</sup>Pb, <sup>232</sup>Th and <sup>228</sup>Ra in soft/medium hard scales**

Radionuclide	Reported Range (Bq/g)	References
<sup>238</sup> U	0.001 - 0.05	26,38
<sup>226</sup> Ra	0.8 - 400	8,18,29,36,38
<sup>210</sup> Pb	0.05 - 2,000	25,29,31
<sup>232</sup> Th	0.001 - 0.07	26,38
<sup>228</sup> Ra	0.05 - 300	18,25,29

**Table 8 - Activity concentration of <sup>238</sup>U, <sup>226</sup>Ra, <sup>210</sup>Pb, <sup>210</sup>Po, <sup>232</sup>Th and <sup>228</sup>Ra in sludges**

Radionuclide	Reported Range (Bq/g)	References
<sup>238</sup> U	0.005 - 0.01	9,26,47
<sup>226</sup> Ra	0.05 - 800	8,9,18,26,47,51,52,83
<sup>210</sup> Pb	0.1 - 1,300	9,24,25,30,37,56,57,58
<sup>210</sup> Po	0.004 - 160	9,30,37,51,55,56,57
<sup>232</sup> Th	0.002 - 0.01	9,26
<sup>228</sup> Ra	0.5 - 50	9,47,83

**Table 10 - Activity concentration of <sup>226</sup>Ra, <sup>210</sup>Pb and <sup>228</sup>Ra in scrapings**

Radionuclide	Reported Range (Bq/g)	References
<sup>226</sup> Ra	0.01 - 75	9,52,70
<sup>210</sup> Pb	0.05 - 50	9
<sup>210</sup> Po	0.1 - 4	9
<sup>228</sup> Ra	0.01 - 10	9

**Table 10 - Observed External Radiation Levels at the Outside of Processing Facilities**

Facility	Radiation Level <sup>#</sup> ( $\mu$ Sv/h)	References
<b>Crude oil processing/treating</b> <ul style="list-style-type: none"> <li>· down hole tubing, safety valves (internal)</li> <li>· well heads, production manifolds</li> <li>· production lines</li> <li>· separator scale (internal)</li> <li>· separator scale (external)</li> <li>· water outlets</li> </ul>	up to 300 0.1 - 2.5 0.3 - 4 up to 200 up to 15 0.2 - 0.5	8,25,38,46,47, 48,51,61,82,84 , 85
<b>Associated/Natural Gas processing/treating</b> <ul style="list-style-type: none"> <li>· downhole tubing</li> <li>· piping, filters, storage tanks, reflux lines</li> <li>· sludge pits, brine disposal/injection wells, brine storage tanks</li> </ul>	0.1 - 2.2 up to 80 up to 50	25,30,32,38, 46,61
<b>NGL processing</b> <ul style="list-style-type: none"> <li>· filters</li> <li>· NGL pump</li> <li>· C<sub>3</sub> storage tanks</li> <li>· NGL/C<sub>3</sub> shipping pumps, C<sub>3</sub> reflux pumps, elbows, flanges</li> </ul>	up to 90 up to 200 up to 60 0.1 - 2.8	31,32,37, 56,57,60

<sup>#</sup> Generally external radiation levels are quoted in Gy/h, but for reasons of comparison the UNSCEAR conversion factor, 1 Gy/h  $\mu$  0.7 Sv/h [4] is used.

**FIGURE 1**

<sup>238</sup>U decay-series.

Relevant process streams, in which various NOR's may show up are indicated.

**FIGURE 2**

<sup>232</sup>Th decay-series.

**FIGURE 3**

General Scheme for the Formation of Oil and Gas Accumulations

Schematic presentation of primary and secondary migration in the initial and advanced stage of basin evolution [20].

I: Initial phase of primary and secondary migration. II: Advanced stage of primary and secondary migration and formation of accumulation.

**FIGURE 4**

Schematic Lay Out of an Offshore Facility for Crude Oil and Natural Gas Production.

Spots, where enhanced radiation levels, or LSA scales/sludges have been encountered [5], are indicated by the radiation trefoil.



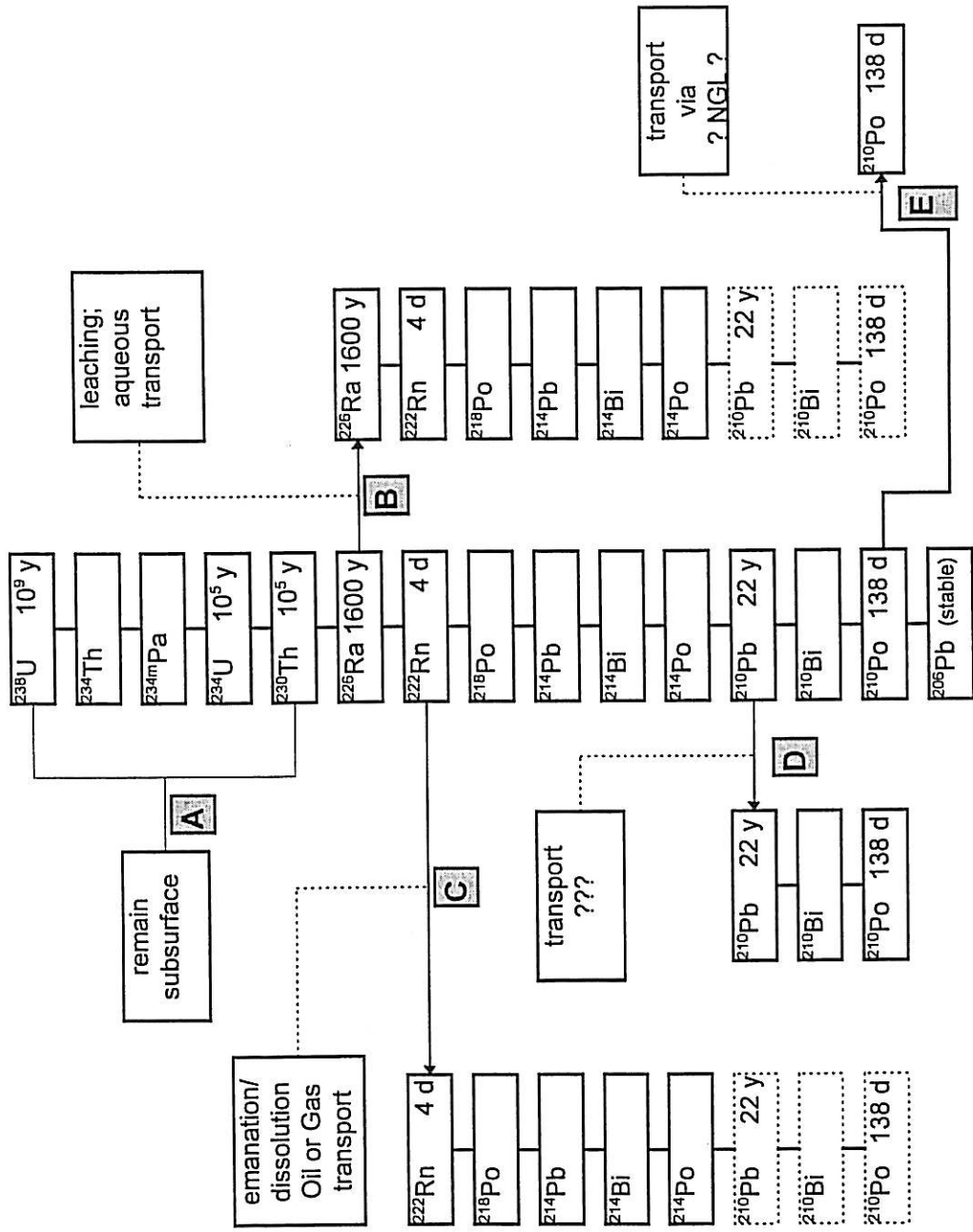


Figure 1: The  $^{238}\text{U}$  decay series and its sub-series:

A: geochemically immobile  $^{238}\text{U}$ ; B:  $^{226}\text{Ra}$  sub-series; C:  $^{222}\text{Rn}$  sub-series; D:  $^{210}\text{Pb}$  sub-series; E:  $^{210}\text{Po}$  sub-series. Nuclides in the sub-series, which do not readily reach secular equilibrium, are shown in dotted boxes

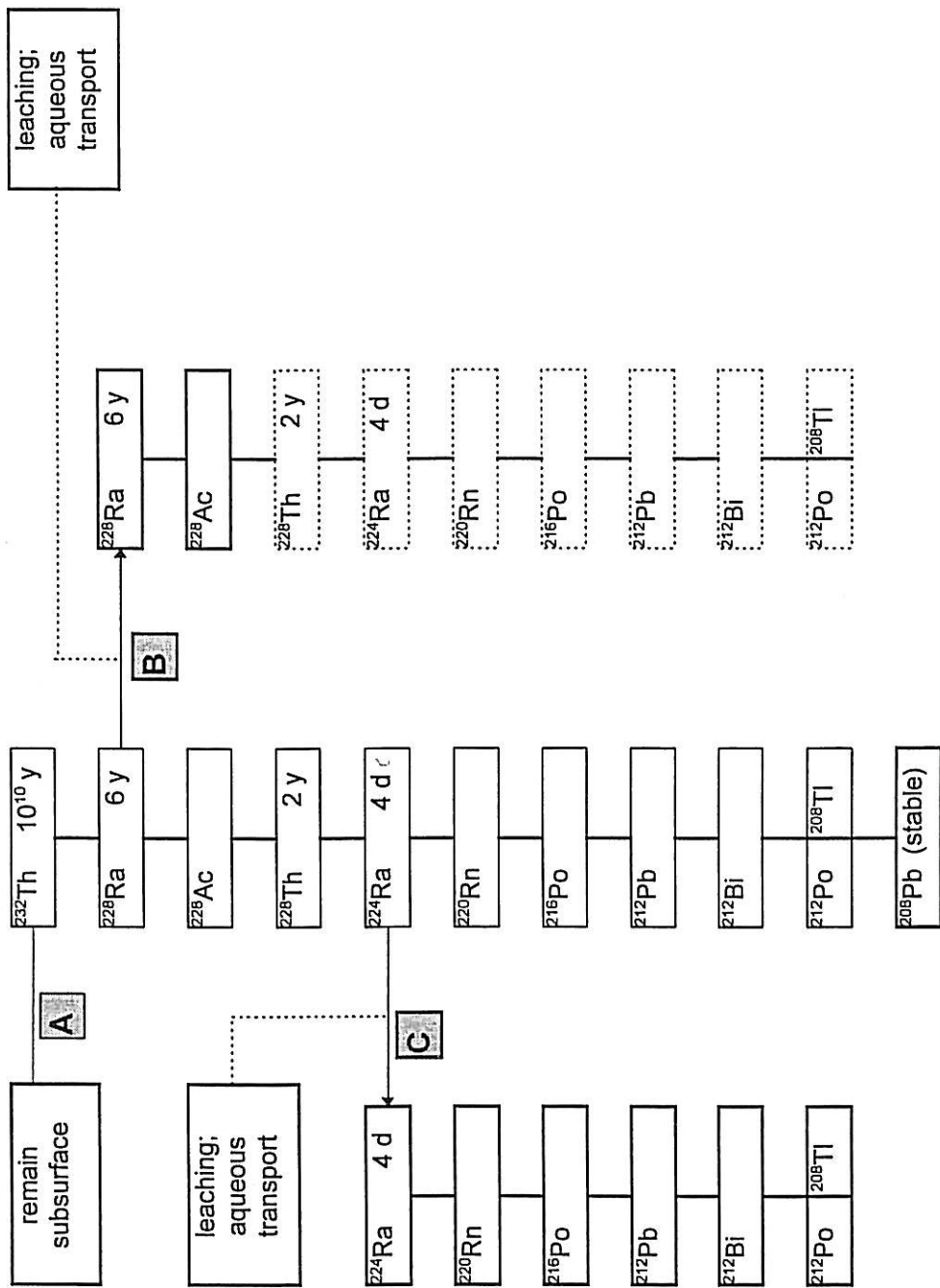
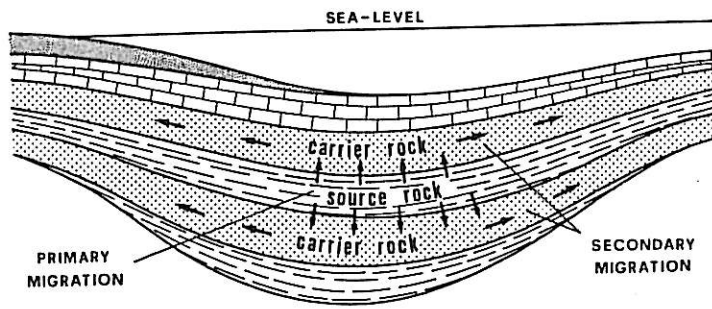


Figure 2:  $^{232}\text{Th}$  decay series and its sub-series: A: geochemically immobile  $^{232}\text{Th}$ ; B:  $^{228}\text{Ra}$  sub-series; C:  $^{224}\text{Ra}$  sub-series. Nuclides in the sub-series, which do not readily reach secular equilibrium, are noted in dotted boxes



I

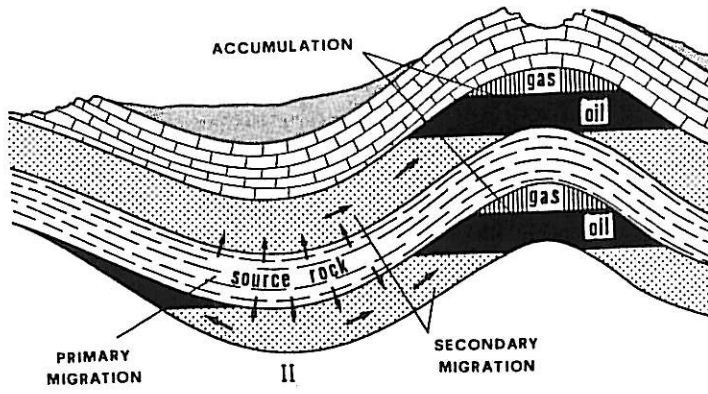


FIGURE 3

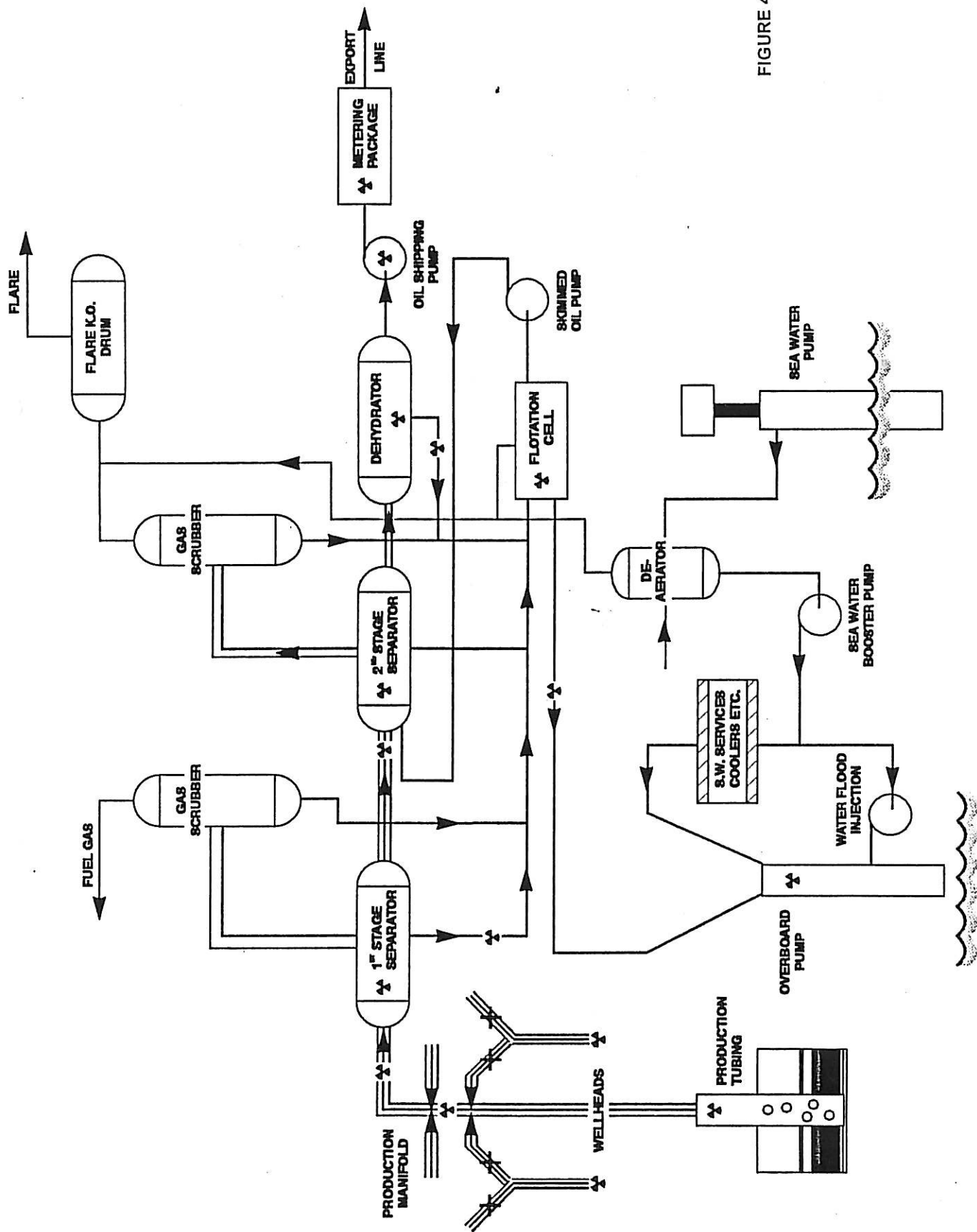


FIGURE 4

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