LAND DISTURBANCE AND THE ENVIRONMENTAL POLLUTION FROM OIL - AND GAS - WELL DRILLING AND EXTRACTION, WITH SPECIAL REFERENCE TO NATURALLY OCCURRING RADIOACTIVE MATERIALS

D. A. Bradley
School of Physics, University of Exeter, United Kingdom

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Summary

Naturally occurring radioactive materials leached from oil and gas bearing formations can be transported to the Earth's surface as part of a mixture of aqueous fluids and hydrocarbons. Temperature changes in the ionic fluids will occur as the material rises to the surface, bringing about the possibility of complex salt scale formation. Removal and disposal of these unwanted deposits from tubulars and process equipment, with possible inclusions of radioactive salts, can pose a potential radiological problem. On the surface, the produced fluids are separated into their various phases and this too may bring about a partitioning of radioactivity. In oil production, the majority of the radioactivity will either be retained on clay particulates, this constituting a significant amount of the oily sludge waste, or can remain in the aqueous phase. The treatment of bulk volumes of affected waste materials containing hydrocarbons, heavy metals, and radioactivity can be a complex environmental management problem. Oil recovery, in which the raw sludge is passed through a filter press, is one way of obtaining a volume reduction, also removing a substantial quantity of the hydrocarbons from the remaining waste. Untreated sludge can otherwise be subjected to techniques such as land farming in which the sludge is mixed with soil and aerated to promote microbial digestion of the

hydrocarbon component. In the oil and gas industry, the levels of radioactivity in treated and untreated affected material has been observed to vary considerably, according to location, ranging from the inappreciable to rather significant levels. In this article a review is made of the various environmental issues surrounding the handling of naturally occurring radioactive materials (NORM), in the oil and gas industry.

1. Introduction: The Issues

In the exploration and production of hydrocarbons operationally important materials are used in great volumes, while the generation of waste in large amounts is also inevitable. In consideration of both classes of materials, it is perhaps not widely appreciated that some of these materials contain naturally occurring radioactivity of low specific activity (LSA). Management of the naturally occurring radioactive materials (NORM) presents a challenge in caring for the environment. The environmental issues can be made significantly more complex by the presence of hydrocarbons and heavy metals. Consider, for instance, that the pipe scales of Mississippi oilfields have been observed to contain native lead and lead sulfide with barite at levels of up to 10%. Pipe scale will almost certainly contain NORM, sometimes at appreciable levels. Reported values have ranged from as little as that typical of unexploited environments to in excess of perhaps two orders of magnitude greater than the prevailing environmental levels. Pipe scale can significantly inhibit flow, leading to cleaning of the tubulars or their disposal in situations in which remediation is not possible. Disposal of the scaled waste or scaled tubulars naturally leads to health and environmental concerns. Although this article does not propose to look at the occupational hazards that are associated with the handling of NORM, it should nevertheless be mentioned that risks to the individual can be kept extremely low. This will come about through the use of engineered controls and adherence to prescribed safe working procedures.

Environmental concerns relating to disposal of scaled waste apply both off and onshore. Cleaning of valves and wellheads can for instance be attempted offshore. Conversely, the cleaning of tubulars is more typically performed at onshore sites. In both situations, cleaning practices and disposal of waste are controlled by applicable legislation. Where national legislation does not exist, it is the practice of industry to be self-regulating. Standards of practice are a response to applicable national legislation or industry self-regulation and will almost certainly be traceable to international recommendations, an example of which are those of the IAEA Basic Safety Standards.

Authorized disposal is generally dependent upon the results of impact assessment. Scaled materials have been disposed to the marine environment under such circumstances. It is in fact the very nature of scale that the material has a low potential for disassociation and while this makes for great difficulties in cleaning operations it also ensures that the release of NORM from material disposed to the environment is greatly inhibited. With the broadening of the London Convention (a regular series of meetings convened by the International Maritime Organisation, IMO) the disposal of waste at low levels of radioactivity is now included under internationally agreed controls. Thus, the disposal to sea of low-level radioactive waste, designated Low Level Radioactive Waste (LLRW), is not expected to continue. In instances where disposal has been allowed monitoring of the marine environment has been an important part of such practice. To determine impact of NORM, samples of marine sediments and organisms (typically bivalves) have been analyzed for radioactivity. As far as onshore disposal of scaled waste is concerned, the waste disposal can be expensive and has thus far been generally limited to a small number of dedicated radioactive waste disposal facilities. Specialist operators whose prime activity is that of providing service to the nuclear fuel cycle industry more often than not, operate facilities of this type. In countries that do not support a nuclear fuels-cycle, industry storage of the NORM has been the more general practice.

In sludge, comprising the sedimentary remnant of crude oil, the tripartite problem of hydrocarbons, heavy metals, and naturally occurring radioactive materials exists. Sludge formation is promoted by the highly adsorbing properties of the clay and silt particulates that account for the bulk of the sludge. The highly adsorbing properties of the silt and clay particulates are effective inhibitors of the leaching of NORM and this is an important consideration in characterizing the waste-form and its environmental impact.

In some offshore oil and gas operations first stage separation of water and particulates from the oil or gas is carried out on the platform itself. Subsequently, the product (either crude oil or wet gas) is shipped or piped onshore for further treatment before shipment to the refinery. The large volumes of water, which have been separated from the product, are discharged offshore, while unwanted gas is flared or vented off. The latter is often carried out on unmanned platforms called “jackets,” and in such situations exposure to radon gas is not an issue. As with scaled waste and sludge, the potential impact of discharge of water containing NORM is of concern, particularly in regard to disturbance of the marine environment. It is typical for the discharge water separated from the crude oil or gas to be monitored for NORM.

The separators require regular cleaning, with often quite large volumes of silts accumulating within such vessels. In some instances it has been a practice to directly dispose of the accumulated silts to the marine environment, giving rise to both environmental and occupational concerns. In terms of concerns regarding occupational health (and all personnel offshore are occupationally engaged) it is typical for a platform to cease water intake operations during the time of disposal.

It is evident that discharges to the marine environment do occur. Given such a scenario the oil and gas industry does conduct benthos studies as part of an all-encompassing effort of to regulate its own operations.

NORM is also to be found in drilling muds (barites), which are used for lubrication and cooling in drilling operations, and the abrasive grits, which are used for onshore cleaning operations. Some abrasive grits contain higher levels of NORM than others. It is known, for instance, that tin slag will typically exhibit higher concentrations of NORM than copper slag, the latter being the abrasive grits of choice. Control can be maintained through buying specifications and purchase contracts, which make provision for sampling and testing of the abrasive grits.

In all of the above non-scale materials, including sludge, produced fluids, drilling muds, and abrasive grits, the levels of NORM are typically subtler than those found in affected scales. It should nevertheless be appreciated that the associated volumes of these materials tend to be significantly larger than that of scale.

A good deal of published material is to be found on natural radioactivity in scaled waste. In contrast, much less has been written on the technogenic elevation of naturally occurring radioactivity in the other affected materials, including the settled-out sludge residues of crude oil. While it is not typical for the technogenic enhancement of naturally occurring radioactivity in sludge to be as significant as that observed in scale, the sludge does give rise to the complex issue of a mixed waste, comprising hydrocarbons, heavy metals, and NORM. A number of processes have been developed to deal with the onshore management of sludge, the major fraction of the material accumulating in crude oil tank bottoms. Scheduled cleaning and maintenance of crude tanks is typically carried out once in perhaps every five to ten years. Enormous amounts of sludge can be retained in tank bottoms and during cleaning of the tanks the material must be removed and managed at an appropriate level. One of the options in handling the waste is that of sludge farming. The success of sludge farming is critically dependent on bio-remediation and in particular upon microbial digestion of the heavy content of hydrocarbons in the sludge.

In land farming, the crude sludge is transferred from crude tanks to plots of land located within the terminal facility. The plots typically take the form of a series of engineered cells, designed to strongly inhibit release of the material to the environment. The cells generally comprise concrete bunds, the retained plots typically being lined by membranes of heavy duty PVC and clay, and a surrounding isolated waste water management system which acts to prevent spilled water from leaking to the drainage systems. In the plots the transferred sludge is mixed with natural soils, turned and watered on a routine basis, the whole process leading to the promotion of microbial digestion of the hydrocarbon content. One outcome of the process of promoting microbial digestion of the hydrocarbons is an inadvertent dilution of the technologically enhanced concentration of the heavy metals and radioactivity.

As an important aside, it should be mentioned that following cleaning of crude tanks an opportunity becomes available for inspection of the tank bottoms. Oil terminal tank bottom failures have occurred in the past, leading to extensive environmental impacts to the underlying ground and water system. Signs of deterioration in tank bottoms are generally looked for using ultrasonic inspection techniques, while for tank-walls other non-destructive testing (NDT) modalities are also used. In addition to the conduct of regular NDT, engineered controls are also in place around each tank in an effort to contain seeping fluids or the massive losses of cataclysmic failure as has occurred in the crumpling of tank walls. Among the control measures are bunding and lining of the area surrounding each tank.

In contrast with land farming, there are a number of volume reduction techniques, the majority of which will inadvertently lead to concentration of radioactivity. Volume reduction techniques include oil recovery by means of filter press, centrifugation, and soil washing. Volume reduction through incineration is less favored due to the possibility of release to the atmosphere of noxious gases and gaseous and particulate radioactive materials. Electrostatic precipitators and gas scrubbing techniques are known to be efficient in removing hazardous materials from an incinerator stack. This said, use could also be made of models, which can predict the downwind distribution of any released materials. In particular sophisticated versions of the Gaussian plume diffusion model exist which can be made to account for micro-climatological features induced by surrounding terrestrial conditions and by buildings.

In this introductory section an attempt has been made to review the issues, which surround NORM, in as far as it is associated with the oil and gas industry, particularly in regard to controlling its impact upon the marine and onshore environments. NORM is delivered into the biosphere by the actions of oil and gas production and accumulated as a waste. The material concentrations are typically technologically enhanced above the more dilute and distributed levels which are to be found in the geosphere and as such the hazards associated with this additional environmental loading require an appropriate level of management. Treatment of waste, resulting from extraction of oil and gas from hydrocarbon-bearing formations, inadvertently gives rise to concentration or dilution of the radioactivity. Volume reduction systems will remove a large proportion of the hydrocarbons as well as a large fraction of the heavy metals contained within the oily waste. Reduction in the volume will also be accompanied by an approximately proportionate increase in the activity concentration of the alpha active material in the remaining waste.

In the following sections we will take a more detailed look at some of the issues raised in this introductory section.

2. NORM in the Oil and Gas Industry

The elements uranium and thorium occur naturally within Earth's crust. Both of these elements are unstable and both decay through a series of intermediate radionuclides referred to as progeny. In the case of uranium and thorium both series of decays eventually terminate in isotopes of stable lead. It should perhaps be noted here that the lifetime of a radioactive isotope is most usually measured in terms of the time taken for its activity to decrease to half of its original level, this period of time generally being referred to as the "half-life." The half-life of the primordial radionuclides ^{238}U and ^{232}Th are 4.5 billion years and 14 billion years respectively. Decay of the chain of radionuclides is accompanied by a number of alpha decays and beta decays, sometimes in association with gamma emissions. The manifest concentrations of the parent elements and progeny in the earths' crust are a result of both this physical decay and of complex transport phenomena, which involve geochemical and sometimes biological processes.

While the uranium and thorium generally remains *in situ*, the radium products, which result from this decay process, are more readily leached out by the formation water. Fluids brought to the surface comprise a mixture of formation water, leached products, oil, gas and particulates of soil and clay, in association with attached radium particulates and their decay products. Partitioning of the various phases can be a result of natural processes such as sublimation, the latter resulting from changes in temperature of the fluids, and as a result of anthropomorphic intervention brought about by the actions of top-side vessels and other processing equipment. The radioactivity, which appears in several phases, is concentrated by such physical processes. In the case of the thermodynamic process of sublimation, scale is formed, radium salts being included in the complex scale. Conversely in the formation of sludge radium particulates in the produced fluids are adsorbed onto the surfaces of the silt and clay material. As an aside it can be mentioned that similar associations with NORM occur in the waste products of minerals mining operations and in the various associated downstream activities, including minerals benefication, separation and refining, milling, and sizing. It must also be noted that the technogenic elevation of radioactive progeny of uranium and thorium is quite inadvertent.

An association between NORM and the production of oil and gas has been known for in excess of 90 years. On the other hand, exposure to NORM has developed as a relatively recent issue. It appears that the first such radiological surveys of NORM were performed in the early 1970s, the initial interest being in exposure due to radon and its progeny in gas processing plants. More recently, considerable interest has focused on the potential occupational risks that have grown out of the cleaning of scaled tubulars and the environmental issues of disposal of scaled waste. As previously mentioned, occupational problems can be effectively dealt with by the institution of safe working procedures and the use of relatively simple engineered controls, in particular those which inhibit ingestion and inhalation of NORM. Treatment and disposal of the waste are issues requiring very much greater levels of efforts in their confrontation. Much less attention has been given to treatment and disposal of sludge even though very large volumes of material can be involved.

The problems of production of scale can be confronted by use of chemical scale inhibitors. Scale inhibition can nevertheless lead to the appearance of affected material at points further along the process line, possibly at more diluted concentrations, but often, as in sludge, exacerbated by the presence of hydrocarbons and heavy metals. Even if scale inhibitors are used, NORM will still tend to appear in the sludge and discharge water. Since the additional material is technogenic this is sometimes referred to as Technologically Enhanced NORM or TENORM. In a number of situations, significant quantities of technogenic NORM have been found, often displaying quite obvious elevations over prevailing background. Much larger volumes of technogenic NORM display very subtle elevations. In both situations the associated issues of radiological health of the public and environmental protection need to be considered.

The issues to be dealt with here relate to the way in which storage and disposal of NORM are handled by the oil and as industry. The waste streams of the extractive industries are constituted of enormous amounts of materials, this being in line with the enormous amounts of material extracted from the earth. The United Nations Scientific Committee on Ionizing Radiation (UNSCEAR) 1993 report on Sources and Effects of Ionizing Radiations included an estimate of annual worldwide production of crude petroleum and natural gas of approximately 3×10^{12} kg and 10^{12} m^3 respectively. The annual generation of sludge of even modestly sized oil/gas fields can perhaps run to a few tens of thousands of tons of material, while the volumes of produced water and gas are clearly significantly more than that of the sludge. Kolb and Wojik observed that in 1983 about 30 million tons of brines were brought to the surface in association with oil and gas.

3. Mechanisms of Enhancement of NORM

In the oil and gas industry it has been known for some years that as a result of its operations radioactive material can sometimes appear as inclusions in scale formations in tubulars and process vessels as well as in the sludge that settles out in crude storage tanks. Scaled tubulars have been known to include ^{226}Ra in concentrations as high as 100 kBq kg^{-1}, and not much less for ^{228}Ra. This so-called TENORM derives from the formations from which oil and gas are extracted and is typically associated with the water phase.

The radionuclides ^{238}U and ^{232}Th and their radioactive progeny, many of which give rise to relatively toxic alpha emissions, exist at concentrations which are often higher in black shales than many other types of terrestrial media. Selective leaching of isotopes of radium, which exist as progeny of ^{238}U and ^{232}Th, results from the presence of formation water. Radium is a group IIA element, as are calcium, barium, and strontium. As a consequence, radium often follows the chemistry of these other group IIA elements. In sulfate and carbonate rich waters, cationic and anionic concentrations provides a favorable basis for precipitation and, given the pressure and temperature changes which produced fluids undergo, leads to the formation of simple and complex salts and scale formation. Examples of simple salts are $CaSO_4$, $BaSO_4$, $CaCO_3$, sometimes forming the basis of complex salts, as in for instance through co-precipitation of radium. Scale formation leads to enhancement of natural radioactivity and working with scaled tubulars and process equipment can impact upon health and the environment. Radium salts have also been observed to be transported throughout the system, with the further possibility that radioactivity levels in sludge may inadvertently become increased by the use of scale inhibitors. Sludge also contains significant quantities of clay silts and adsorption onto these materials may lead to a further mode of enhancement of radioactivity. Enhancement can also result from migration of dissolved radon in produced water and sludge, an issue that has been raised in attempting to provide a rationale for the existence of high levels of for instance ^{210}Po. In this latter case, the absence of high levels of radium has lead to the ^{210}Po being assumed to exist at unsupported levels. In storage tanks, radon progeny can build up in the product and also on the tank walls, the latter arising through plate-out of activity. One example of the build-up of radon progeny in product is that of ^{210}Pb; this particular radionuclide has been observed in sludge at levels of a few tens of Bq g^{-1} and more. Whilst sludge may constitute a low specific activity material, the accumulation of waste can lead to relatively large accumulation of activity and disposal is an entirely pertinent question.

4. The Hazards of Alpha Active Materials

At a fundamental level, the diversity of matter is apparent both in terms of atomic and nuclear composition. Atomic composition and structure ultimately dictates the chemical form of matter while nuclear composition and structure dictates stability. Non-stable forms move towards stability through reconfigurations of the structure of the atom, mediated by the emission of alpha particles, beta particles, and gamma rays. Particulate emission results in modification of the neutron to proton ratio. Neutron to proton ratios for all elements heavier than lead give rise to nuclear instability, with alpha emission being a common mode of decay.

The hazards associated with exposure to low-levels of alpha active material external to the body are generally quite small. This fact is directly related to the large capacity for alpha particles to deposit their energy in even thin layers of media; the alpha particles are said to be high Linear Energy Transfer (LET) radiations. In comparison, beta particles and gamma rays are of lower LET and are therefore more penetrating. As such, they generally present a greater external hazard. The rapid absorption which results from large loses of energy of alpha particles, even in media of density as low as air, means that they are almost entirely absorbed in clothing and in the outer layers of skin. In the same way, when taken up in the body, alpha active material will heavily irradiate the surrounding healthy tissues. As a result, the greater hazards of exposure to alpha active materials apply to their ingestion and inhalation. Once in the lungs, the radiosensitive epithelial tissues, which line the lungs, will be heavily irradiated. Compared to the uptake of other radioactive media of equivalent activity, the uptake of alpha active material leads to a particularly enhanced probability of the occurrence of detrimental health effects.

The degree of harm resulting from uptake of alpha active material is obviously enhanced at higher levels of activity, particularly at higher concentrations, and long retention times. Haaker has reviewed the issue of NORM absorbed through the gastrointestinal (GI) tract and retention of NORM in the lungs. In respect of inhalation a new respiratory tract model and revised biokinetic model have now become available. Present inhalation dose coefficients are based on an activity mean aerodynamic diameter (AMAD) of particles of size 5μm rather than previous use of AMAD's of 1μm. This has lead to a reduction of previous estimates of internal exposure to NORM radionuclides.

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Biographical Sketch

In 1975 **David Bradley** graduated with a Bachelor of Science (Physics) degree from the University of Essex. He has a Masters degree (Radiation Physics), awarded by the University of London in 1976, and obtained his PhD in 1985 while lecturing in Physics at the University of Science of Malaysia (USM). His career, which began with work as a radiotherapy physicist at Charing-Cross Hospital (London) in 1976, has in more recent years turned to teaching and research also, in addition, providing radiation and environmental protection consultancy. Currently he is a senior lecturer and member of the biomedical physics research group at the School of Physics, University of Exeter. His research interests concern the general area of interactions of radiation with matter, these interests being more often than not in respect of applications and consequences of radiations in industry and medicine. Journal publications number in excess of 100 papers. In addition, he has been the editor of a number of books and special issues of journals that focus on ionising radiations. DAB is a Fellow of three professional Institutes: the UK based Institutes of Physics (IOP) and of Physics and Engineering in Medicine (IPEM), and the Malaysian Institute of Physics (IFM). Presently he is Editor-in-Chief of the Elsevier journal *Radiation Physics and Chemistry* and a Council Member of the International Radiation Physics Society (IRPS).