RADIOLOGICAL DOSE ASSESSMENT OF NATURALLY OCCURRING RADIOACTIVE MATERIALS AT PETROLEUM GAS SEPARATION STATIONS RUMAILA, IRAQ

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Dedicated to my beloved family

To the spirit of my father, who died prematurely

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ABSTRACT

Accurate evaluation of radiological dose from naturally occurring radioactive materials (NORMs) generated by petroleum gas separation stations remains challenging. Protection of human health from unwanted radiation exposure, and minimization of environmental pollution from these NORMs are the main motivations of this work. These could be achieved by evaluating the activity concentrations (ACs) of various identified radioactive elements in the soil, sludge, and wastewater samples collected from seven petroleum gas separation stations located in Northern Al-Rumaila of Iraq. In this study, high purity germanium (HPGe) detector was used to measure the ACs of ^{232}Th, ^{226}Ra, and ^{40}K radioactive isotopes present in the soil, sludge, and wastewater samples. Radiometric analyses were performed on wastewater samples, while ^{238}U activities were recorded using fluorimetry technique. The gamma absorbed dose rates in air were measured using survey meters. Gross alpha/beta measurements were also conducted for the collected wastewater samples. Radiation hazard parameters were examined at these gas separation stations to estimate the potential radiological health risk in the sediments/sludge/wastewater samples. The findings showed that the mean value of ACs of ^{232}Th, ^{226}Ra and ^{40}K in soil samples measured were 106.05 ± 4.34 Bqkg^{-1}, 209.91 ± 8.43 Bqkg^{-1} and 302.20 ± 13.07 Bqkg^{-1}, respectively. The ACs for ^{226}Ra, ^{232}Th and ^{40}K in the sludge samples were considerably greater than IAEA guidelines. Furthermore, the ACs of ^{232}Th and ^{226}Ra were discerned to be higher, and ^{40}K was lower than the world average. Results of the radiometric analysis of water samples revealed a wide variation in the detected activities of ^{228}Ac, ^{226}Ra, ^{40}K and ^{238}U from below the detection limit until 11.17, 22.63, 19, and 0.92-6.46 BqL^{-1} respectively. Simulation was performed using RESRAD software to estimate the yearly dose where the yearly dose for various NORMs present in the samples were found to be maximum (64.67 μSv.y^{-1}) at DS4 station. Results of this study clearly demonstrated that radioactivity levels in the wastewater samples exceeded the limits set for by the World Health Organization for drinking water. If no remedial steps are implemented, then there is a potential for long term risks to the health of the community and those who rely on recycled water in this Iraqi region.

ABSTRAK

Penilaian tepat terhadap dos radiologi dari bahan radioaktif semula jadi (NORMs) yang dihasilkan oleh stesen pemisahan gas petroleum kekal mencabar. Perlindungan kesihatan manusia daripada pendedahan sinaran yang tidak diingini, dan peminimuman pencemaran alam sekitar dari NORMs ini adalah tujuan utama kajian ini. Ini boleh dicapai dengan menilai kepekatan keaktifan (AC) pelbagai elemen radioaktif yang dikenal pasti dalam tanah, enap cemar, dan sampel sisa air yang dikumpul daripada tujuh stesen pemisahan gas petroleum yang terletak di Al-Rumaila Utara Iraq. Dalam kajian ini, pengesan germanium berketulenan tinggi (HPGe) telah digunakan untuk mengukur AC daripada isotop radioaktif ^{232}Th, ^{226}Ra, dan ^{40}K dalam sampel tanah, enap cemar, dan sisa air. Di samping itu, analisis radiometrik telah dijalankan ke atas sampel sisa air, manakala keaktifan ^{238}U telah dirakam menggunakan teknik fluorimetry. Kadar dos sinar gama diserap dalan udara diukur menggunakan meter tinjauan. Ukuran alpha / beta kasar juga telah dijalankan bagi sampel sisa air yang dikum. Beberapa parameter hazard sinaran juga dinilai di stesen pemisahan gas untuk menganggarkan potensi risiko kesihatan radiologi dalam sampel sedimen / enap cemar / sisa air. Dapatan kajian menunjukkan bahawa nilai min AC pagi ^{232}Th, ^{226}Ra dan ^{40}K dalam sampel tanah yang diukur masing-masing ialah 106.05 ± 4.34 Bqkg^{-1}, 209.91 ± 8.43 Bqkg^{-1} dan 302.20 ± 13.07 Bqkg^{-1}. AC pagi ^{226}Ra, ^{232}Th dan ^{40}K dalam sampel enap cemar adalah jauh lebih besar daripada garis panduan IAEA. Tambahan pula, AC daripada ^{232}Th dan ^{226}Ra ternyata lebih tinggi, dan ^{40}K adalah lebih rendah daripada purata dunia. Hasil analisis radiometrik sampel air mendedahkan variasi yang basar dalam keaktiran yang dikesan daripada ^{228}Ac, ^{226}Ra, ^{40}K dan ^{238}U dari bawah had pengesanan sehingga masing-masing ke 11.17, 22.63, 19, dan 0.92-6.46 BqL^{-1}. Simulasi telah dilakukan dengan menggunakan perisian RESRAD untuk menganggarkan dos tahunan di mana dos tahunan bagi pelbagai NORMs dalam sampel didapati maksimum (64.67 µSv.y^{-1}) di stesen DS4. Dapatan kajian ini menunjukkan dengan jelas bahawa tahap keradioaktifan dalam sampel sisa air melebihi had yang ditetapkan oleh Pertubuhan Kesihatan Sedunia untuk air minuman. Jika tiada langkah pemulihan dilaksanakan, maka terdapat potensi risiko jangka panjang kepada kesihatan masyarakat dan individu yang bergantung kepada air yang dikitar semula di rantau Iraq ini.

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LIST OF ABBREVIATIONS

AEDE	-	Annual effective dose equivalent
ASTM	-	The American Standard for Testing and Materials.
BEIR	-	The Biological Effect of Ionising Radiation.
Bq	-	Becquerel
Ci	-	Curie 1Ci=3.7 *10^{10} bq
D	-	absorbed dose rate in air
DCF	-	Dose conversion factor
DH	-	Dehydration Unit
DS1	-	Degassing Station One
DS2	-	Degassing Station Two
DS3	-	Degassing Station Three
DS4	-	Degassing Station four
DS5	-	Degassing Station five
EPA	-	Environmental protection agency
GPS	-	Global positioning system
Gy	-	Gray absorbed dose
HBRA	-	High background radiation area
Hex	-	External hazard index
H_{in}	-	The internal hazard index
HPGe	-	Hyper pure germanium detector
IAEA	-	International atomic energy agency
ICRP	-	The International Commission on Radiological Protection
MCA	-	Multi-channel analyzer
NCRP	-	National council on radiation protection
NIDS4	-	North Intermediate Degassing Station Four
NORM	-	Naturally Occurring Radioactive Material.

ROI	-	Region of interest
SIDS2	-	South Intermediate Degassing Station Two
TENORM	-	Technologically-Enhanced Naturally Occurring Radioactive Material
UNSCEAR	-	United nations scientific committee on the effects of atomic radiation
USNRC	-	United states nuclear regulatory commission
VOCs	-	Volatile Organic compounds
WNA	-	The World Nuclear Association

LIST OF SYMBOLS

^{210}Pb	-	Lead-210
^{214}BI	-	Bismuth-214
^{214}Pb	-	Lead-214
^{226}Ra	-	Radium-226
^{228}Ac	-	Actimium-228
^{232}Th	-	Thorium-232
^{235}U	-	Uranium -235
^{238}U	-	Uranium-238
^{40}K	-	Potassium-40
^{60}Co	-	Cobalt-60
^{85}Sr	-	Strontium-90
A_k	-	Specific activity of potassium
A_s	-	Specific activity
A_{Th}	-	Specific activity of thorium
A_U	-	Specific activity of uranium
C_K	-	Concentrations of potassium
C_{Th}	-	Concentrations of thorium
C_U	-	Concentrations of uranium
D	-	Absorbed dose
ɤ	-	Gamma rays
^{220}Rn	-	Thoron
^{222}Rn	-	Radon
α	-	Alpha particle
β	-	Beta particle

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CHAPTER 1

INTRODUCTION

1.1 Background of the Study

In the oil and gas industries worldwide, the naturally occurring radioactive materials (NORMs) consisting of radionuclide such as ^{232}Th, ^{228}Ra and ^{238}U are accumulated and concentrated in various types of containers for their effective disposal. Consequently, they can be characterized based on their chemical and physical processing in the natural oil and gas refineries [1]. Furthermore, the brine water (wastewater) that is created in this process normally isolated from oil is set for disposal into the environment without accessing their radioactive dose level. This wastewater is channelled into the inoculation well or evaporated in the open atmosphere [2]. It is also injected into the source to increase the oil yield via a method called "water flooding". In some cases, produced water amount is greater than the amount of oil produced. Thus, it forms the major amount of waste radioactive material produced from the oil and gas industries [3] . It is important to monitor the radioactivity level of these waste for environmental and health safety.

It is worth mentioning that most of the companies operating in the Middle East region usually dispose their wastes water into unlined pits and lagoons. Subsequently, the projection water is drained to underground leaving radioactive precipitate within the soil it needs proper. This required remedial or treatment action in accordance with

radiation protection principles. Over the years, several remediation projects are undertaken to treat the contaminated soil for reducing the hazard to workers, environment, and public [1, 3]. One of the largest oil field in Iraq (Rumaila) produce huge amount of water and sludge that are regularly released in the surroundings for evaporation. Recently, it is realized that such uninhibited dumping of such wastes leads to severe environmental pollution unless regulated. Besides, oil field workers and civilians living in this region are often exposed to the high radiation dose. To minimize the radiation exposure related health risk, constant monitoring of the dose level is necessary. Thus, it is important to identify the radionuclide present in the soil for subsequent determination of the effective remediation protocol. It is the main recurring theme of the present study

Earlier studies revealed that Radium (^{226}Ra and ^{228}Ra) is the primary radionuclide present in the contaminated soil around this oil field [2]. Accordingly, strategies must be taken to remove the radium isotopes that are present in this kind of polluted soil. The long half-life of Radium isotope (1620 yr for ^{226}Ra and 5.8 yr for ^{228}Ra) and associated decay products (^{224}Ra) make them highly threatening to human and other living organisms. Various types of radioactive waste that are generated from oil and gas industries are generally removed from the production units and stored in special container or discharged in the environment as low radioactive dose wastes. Nevertheless, strict control of radioactive waste from being released in the environment that cause pollution and health hazard is compulsory via accurate monitor. In-depth research on the mechanism of radioactive waste removal from oil field and their impact on health and ecology need special attention in terms of remedial measure [4].

Smith *et al*. (1996) reported that during oil and gas extraction process substantial amount of NORMs at very high concentrations are accumulated as stream of waste by-products. Furthermore, it was found that in these wastes the isotopes of ^{238}U and ^{232}Th appeared as the most prolific NORMs. As by-products, they occur on the subsurface of the oil and gases that are produced in the industries. The main radioactive nuclides or NORMs that are greatly alarming as waste products are ^{226}Ra,

^{228}Ra, ^{238}U and ^{232}Th decay series. Besides, the radio-nuclides those are formed as decay products of ^{226}Ra, and ^{228}Ra are also environmentally concerning [4].

Manufacturing and dispensation tools may include remaining amounts of NORM-contaminated water, scale, or sludge. Consequently, it may cause removal and exposure setback when the dispensation tools or equipment's are taken off-line for repair, maintenance, or substitution [5]. Equipment's or container used for natural gas processing and production are also gets polluted by forming a thin layer of poisonous radioactive ^{210}Pb in the inner surfaces. In short, NORMs are omnipresent in the oil and natural gas processing systems unless carefully removed.

Khodashenas *et al*. (2012) investigated the occurrences of NORMs in the oil production unit, evaporation pond, and drilling site in the Khuzestan province (Iran). Radionuclides such as ^{232}Th and ^{40}K were detected in the soil samples and ^{226}Ra was found in both soil and water samples. The ACs of ^{232}Th was ranged between 8.7 and 403 Bq.kg^{-1}, while the minimum concentration for ^{40}K was much larger (82 Bq.kg^{-1}) where the maximum concentration was found to be 815 Bq.kg^{-1}. Soil samples revealed very low concentrations of ^{226}Ra, typically between 10.6 and 42.1 Bq.kg^{-1} with some exceptions (282, 602, and even 1480 Bq.kg $^{-1}$). The range of ^{226}Ra contents in the water sample was ranged from very low (0.1 Bq.L^{-1}) very high (30.3 Bq.L^{-1}). Overall, results showed that on an average the NORM concentrations in these areas were lower than the usual concentration levels in typical oil and gas fields. However, necessary measures were undertaken for minimizing the environmental impact of such NORMs [5].

It is needless to mention that the traces of NORMs in the waste products of oil and gas processing industries have been continuously recognized since the mid-thirties of the last century [6]. Moreover, there was no concern regarding the NORMs assisted health hazards until the mid-1980s. Later, the oil and gas refinery industry and regulators realized the widespread occurrences of highly pollutant NORMs and their adverse environmental impacts. Thereafter, it was found that the activity levels of these

radio-nuclides could be quite high and a real threat to human health and environment. Despite the wide acknowledgement of these radio-nuclides radiation exposure related health concern to both workers and the general public the precise quantification of NORM-contaminated waste streams, the extent of health hazard and subsequent regulatory measure on NORM remains debatable. In this spirit, the present research takes an attempt to bridge the gap related to some NORM related issues as described hereunder.

1.2 Problem Statement

Several petroleum industries are spread all over the landscape in Iraq. Previous studies revealed that the soil and water in each of these sites contains significant amounts potentially active NORMs [7]. However, accurate evaluation of radiological dose of such NORMs generated by the petroleum and gas separation stations is not yet performed. The awareness regarding the protection of the human health from these NORMs [8], assisted unwanted dose exposure must be developed through careful evaluation and quantifications of the emanating radiation dose. Inhibition of the environmental pollution from these NORMs remains a challenge. Thus, systematic radio-metric measurements are necessary to determine the activity concentrations (ACs) of various radioactive elements in the soil/sludge and wastewater samples spread around petroleum gas separation stations located at Northern Al-Rumaila of Iraq. This study is expected to generate new information regarding the baseline radiometric data on NORMs present in the petroleum and gas separations stations. This radioactivity is purely natural which are not generated by other nuclear or war related activities in the country. To achieve such goal, the baseline data from diverse soil samples must be acquired for evaluating the health hazards of the onsite workers in the cited gas separation stations and public living in those sites. Precise radiometric information on the ACs of potentially hazardous NORMs such as ^{232}Th, ^{228}Ac, ^{226}Ra, ^{40}K, and ^{238}U in the sediments/soil/sludge and wastewater samples obtained from the petroleum gas separation stations located around Northern Al-Rumaila (Iraq) are lacking.

It is significant to determine the ACs level in the sediments/sludge samples to check their permissible limit as set by WHO and IAEA. Thus, quantification of the mean ACs for these NORMs is vital to prevent further environmental pollution and health risks. It is important to verify whether the ACs of the identified NORMs are lower than the world average. A correlation between the ACs of different NORMs is not yet established. This information can be obtained by calculating the relevant hazard parameters responsible for the potential radiological health risk due to sediments/sludge and wastewater samples. When these calculated dose rates exceed the permissible limit then strategies should be taken for immediate remedy. Radiometric analysis of these samples is required to determine the exposures impact on the onsite workers. The radiological health information of the mean outdoor annual effective dose, radium equivalent activity, external and internal hazard indices needs careful estimation. The evaluation of radioactivity levels in the wastewater samples is essential to determine the drinking water safety limit set by the World Health Organization (WHO). Present radiological dose assessment on all these degassing stations will reveal the degree of NORMs related radioactive contamination. Based on the collected information and data analysis it will be possible to solve the problem involving environmental sustainability, safety of the workers, and public.

1.3 Research Objectives

Based on the problem statement or research gap the following objectives are set.

i. To identify the potential NORMs in the soil/sediment/sludge and wastewater samples spread around the petroleum gas separation stations located at Northern Al-Rumaila of Iraq for accurate radio-metric dose assessment.
ii. To determine the activity concentrations of various NORMs useful for NORM management, disposal activities, radiometric information about

the potential health risk, and environmental protection following IAEA and WHO guidelines.

iii. To compare the measured ACs of these NORMs in various samples with relevant safety requirement and radiation protection guide of ICRP.

iv. To determine the radiological health information in terms of the mean outdoor annual effective dose, radium equivalent activity, sensitivity, external and internal hazard indices

1.4 Scope of Study

The scope of this study is comprised of the following:

i. Collection of the soil/sediment/sludge and waste water samples from the region around the petroleum gas separation stations located at Northern Al-Rumaila of Iraq. Identification of the NORMs that caused potential health risks and environmental pollution. Obtaining the baseline data for collected 36 soil samples to evaluate the health risks of the personnel working in such gas separation stations and civilians living in the proximity.

ii. Use of hyper pure germanium (HPGe) detector to measure the ACs of detected radioactive isotopes (NORMs). Use of fluorometric technique for determining the 238U activities in these water samples and Measurement through gross alpha/beta for evaluating the dose in the collected wastewater samples.

iii. Accurate radio-metric dose assessment for effective NORM management and disposal activities using decontamination apparatus, encapsulation via down-hole, under-ground disposal, land spreading through dilution, apparatus smelting, and unlimited superficial interment of polluted equipment's. Selection of decontaminated apparatus for effective ACs evaluation is because of lack of information

in the literature. Comparison of the measured radiation doses of these NORMs with relevant safety requirement and radiation protection guide ICRP.

iv. Assessment of the radiological health information in terms the mean outdoor annual effective dose, radium equivalent activity, sensitivity, external and internal hazard indices and Human Exposure Assessment by using RESRAD software simulation. Estimation of the maximum individual dose equivalent associated to the selected activities and their subsequent use as the basis for comparison between disposal options.

1.5 Significance of Study

Present study will elucidate the radiometric information about the potential health risk and environmental pollution due to the occurrence of NORMs in the petroleum gas separation stations in Iraq. This will allow the country's administration to develop a strategy for the remediation of unwanted radiation dose exposure with proper nuclear safety. This study will certainly develop an awareness regarding the natural radioactivity related health risks for workers and common public living in those regions. Precise radiometric information on the ACs of potentially hazardous NORMs including ^{232}Th, ^{228}Ac, ^{226}Ra, ^{40}K and ^{238}U in the soil/sediment/sludge and wastewater samples in this area will be necessary to protect people from health hazards [9].

It is important to detect the NORMs and check the AC level in the samples to determine the allowable dose limit as set by WHO and IAEA. So far, no data on radioactivity level of these elements exist. Accurate dose assessment is always needed as stated by UNSCEAR (2000). Though it is well known that the natural radioactivity levels of Ra, Th and K progenies contribute more than half of the total exposure received by workers in gas separation station and public due to natural radiation but proper evaluation has not been made yet in the context of Iraq. Thus, systematic sampling, experimentation, and data analysis is expected to provide very significant

baseline data useful for extended research. In this way, precautionary measure can be undertaken to prevent the feasibility of sudden radiological accidents. For nuclear emergencies and subsequent remediation, monitoring of radioactivity dose level in air has been diversely used as an early warning system [8]. Collected data from these Iraqi gas separation stations will be greatly beneficial for the evaluation of elevated background radiation originating from potentially active NORMs including ^{232}Th, ^{226}Ra, and ^{40}K in the region.

1.6 Novelty of Work

i. For the first time a radiometric evaluation of NORMs/TENORMs was made experimentally for the petroleum gas separation stations in the context of Northern Al-Rumaila of Iraq.
ii. Large number of natural soil, sediment/sludge and wastewater samples was collected from the studied region for the identification of the NORMs/TENORMs.
iii. Using RESRAD software, analytical simulation was performed to estimate the yearly dose for various NORMs/TENORMs present in the sample and the experimental results are validated.
iv. The potential health risks and environmental pollution caused by the presence of radionuclides such as 232Th, 228Ac, 226Ra, 40K and 238U 232Th, in those samples were evaluated.
v. This is the first report for Iraq on the detection of NORMs/TENORMs ACs level in the samples and comparison with the allowable dose limit recommended by WHO, IAEA and UNSCEAR.
vi. Present findings on precise radiometric information on the ACs of potentially hazardous NORMs/TENORMs will allow the national authority/administration to take necessary measures and develop strategies for protecting people from health hazards in terms of environmental pollution and drinking water contamination.

vii. This new baseline data on NORMs/TENORMs at gas separation stations in Iraq will help for remediation of unwanted radiation dose and better nuclear safety management.

viii. This study will develop an awareness regarding the NORMs/TENORMs related health risks for workers and common public living in those regions.

ix. The existence of excess radioactivity levels in the wastewater samples beyond the limits set for drinking water by the WHO suggested the immediate implementation of remedial steps to avoid long term health risks to the community those rely on recycled water in this region.

1.7 Thesis Outlines

This thesis is organized into five chapters as follows:

Chapter 1 introduces the problem by providing a brief background and the rationale of the research. The research gap is articulated as problem statement, which allowed the present researchers to set appropriate research objectives. For achieving the research goals a brief outline is rendered as the scope of study. Furthermore, the significance of the study is highlighted.

Chapter 2 presents the detailed overview of the relevant literature on the entitled thesis. It includes the basic concept of radioactivity, NORMs, radionuclide, and radiation dose, environmental radiation originated from NORM produced in the gas separation stations, radioactivity in sludge, radioactivity in soils, radioactivity in water, review of terrestrial gamma radiation dose in the gas and petroleum industries of various nations, review of terrestrial gamma radiation dose in Iraq and health effects caused by such natural radiation.

Chapter 3 describes the detailed methodology of the research that is used to accomplish the proposed objectives. This research used three techniques for measuring the specific activity of NORMs present in the samples (wastewater, sediments, soil, and sludge) collected from seven petroleum gas separation stations located at Northern Al-Rumaila of Iraq. As mentioned in Chapter 3, the baseline data from 71 soil, 70 sludges, and 36 waste water samples are obtained to evaluate the health risks of the personnel working onsite in such gas separation stations and civilians living in the proximity. These techniques include the gamma ray spectrometry system with High Purity Germanium (HPGe) detector, gross alpha and beta, as well as fluorescence measurement.

Chapter 4 underlines the experimental results, the detail analysis, thorough discussion, and comparisons with other related findings. Data analysis on total 177 samples of soil, sludge, and water were conducted. These samples are collected from the petroleum gas separation stations (Northern Roumaila, Iraq) to perform the experiments. A quantitative analysis is carried out to determine the ACs of the radio-nuclides such as ^{214}Ra, ^{232}Th and ^{40}K that were present in the NORM. The conversion factors for external gamma dose rate from the natural radio-nuclides were determined. The contributions of each radionuclide (^{232}Th, ^{226}Ra and ^{40}K) to gamma dose rate were calculated. The ACs of ^{226}Ra, ^{232}Th and ^{40}K were used to estimate the contribution from each gas separation station and soil type to terrestrial radioactivity.

Chapter 5 concludes the thesis. The radiological dose assessment from the NORMs that are generated by the petroleum and gas separation stations of Northern Al-Rumaila (Iraq) was the focus of this study. This research started with two primary recurring themes. The first one was to protect the human health from unwanted dose exposure and subsequent minimization of the environmental pollution from these NORMs. The second one was to develop the awareness among the communities subjected to occupational health hazards. This research opened up many new avenues those are worth looking. Based on these openings a future outlook is provided to way forward for further investigations.

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