AMERICAN UNIVERSITY OF BEIRUT

ASSESSMENT OF NATURAL RADIOACTIVITY IN DRINKING WATER IN SOUTH LEBANON

by
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A thesis
submitted in partial fulfillment of the requirements
for the degree of Master of Sciences in Environmental Sciences
to the Interfaculty Graduate Environmental Sciences Program
Environmental Technology
of the Faculty of Engineering and Architecture
at the American University of Beirut

Beirut, Lebanon
April 2019

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ACKNOWLEDGMENTS

First and foremost, I thank God Almighty for granting me the strength, knowledge and determination to accomplish this work and for sending into my path each and every person who assisted me during my research study.

My full gratitude goes to Professor George M. Ayoub for his support, compassion and patience; my thesis advisor whose guidance went beyond being only academic advices to become parental lessons that will stay with me throughout my life: A "Master" who believed in me, generously shared his wisdom and tenderly pushed me so toughly to raise me up during my moments of doubts.

Sincere appreciations are dedicated to Dr. Bilal Nsouli, the Director of the Lebanese Atomic Energy Commission of the National Council for Scientific Research (LAEC-CNRS), who with his scientifically engaged mind, opened wide the doors of the LAEC facilities for hosting me into this research project.

Special recognitions are owed to the examining members of my thesis: Dr. Maria Aoun for her extremely professional close-up technical assistance and her meticulous directives, Dr. Mahmoud Al Hindi and Dr. Rana Bilbeisi for their exceptionally high valuable feedback.

Particular thanks are extended to Dr. Omar El Samad, the Head of the Environmental Radiation Control Department at LAEC, and also to his team members, Mr. Hamzeh El-Jeaid and Mr. Radwan Sidaoui, who offered me lots of laboratory mentoring.

Tremendous acknowledgements are due to Mr. Ahmad Nizam, the former Director of the South Lebanon Water Establishment, for making possible the field work of the study, and to the dedicated field's technical crew members who enthusiastically escorted me for samples collection during weekends and at late night hours.

Last but not least, I would like to pay tribute to every single person who contributed in a way or another in the achievement of this thesis. Amongst them, I would like to specifically mention my "Third Eye" Sarah Sinno, Mrs. Zakiya Deeb, Dr. Ramez Zayyat, and my friend Zeina Hamze for their advices, support and/or assistance in the experimental part of my work.

AN ABSTRACT OF THE THESIS OF

Safaa Mohamad Said Mourad for Master of Science in Environmental Sciences
Major: Environmental Technology

Title: Assessment of Natural Radioactivity in Drinking Water in the South of Lebanon

Groundwater contains certain levels of radioactivity that arise from the decay of naturally occurring radioactive materials in the adjacent rocks. Radiation is known to have deleterious health effects. As such, assessment of the radioactive concentration levels is essential for determining the suitability of the water for human consumption.

The Southern region of Lebanon relies primarily on groundwater sources to fulfill its drinking water requirements. However, information on the radiological properties of the water is lacking. Consequently, this study was initiated to formulate a baseline assessment for natural radioactivity in the area's water sources. In this context, radioactivity concentrations were determined using liquid scintillation counter for gross alpha, gross beta and radon activity concentrations, and using alpha spectroscopy for those of uranium isotopes.

Measurement of gross alpha, gross beta, uranium (^{238}U, ^{234}U) and radon (^{222}Rn) activity concentrations in 42 drinking water supplies (wells, springs, and tap water), sampled over the wet and the dry seasons, showed that all activities concentrations were below the WHO guidance levels. The maximum recorded radioactivity concentrations were 42.9±3.7 BqL^{-1} for radon, 53.7±2.1 $mBqL^{-1}$ for ^{238}U, 55.9±2.3 $mBqL^{-1}$ for ^{234}U, 374.6±11.5 $mBqL^{-1}$ for gross alpha activities and 418±12 $mBqL^{-1}$ for gross beta activities; concentrations values, which indicate the suitability of the water for human consumption.

Seasonal variations were investigated with only gross alpha activity concentrations showing significant variation between the wet and the dry seasons. Whereas, radon was the only radionuclide to produce significant variation when comparing the radioactivity concentration levels between sources and tap water. On the other hand, no significant variation in radionuclides concentrations was observed between water from springs and that from wells.

The calculated effective dose in few locations was relatively high and the radiation dose from one of the sampled wells (Aitaroun) exceeded the WHO Individual Dose Criterion (IDC) level of 0.1 $mSvy^{-1}$ with an annual effective dose of 0.170 $mSvy^{-1}$, 0.127 $mSvy^{-1}$, and 0.103 $mSvy^{-1}$ for infants, adults, and children respectively. Yet the effective dose received from tap water form this well falls below the IDC level.

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ABBREVIATIONS

α	Alpha particle
β	Beta particle
γ	Gamma ray
υ	Neutrino
$(\bar{\upsilon_e})$	Antineutrino
ADC	Analogue to Digital Analyzer
Am	Americium
AUB	American University of Beirut
B	Becquerel
Be	Beryllium
Bi	Bismuth
BqL^{-1}	Becquerel per Liter
dps	Disintegration per second
C	Carbon
Ci	Curie
CNRS	National Council for Scientific Research
cpm	Count Per Minute
CR-39	Columbia Resin #39
Cs	Cesium
DIPN	Diisopropylnaphtalene
DVB	Divinylbenzene
EERC	Environmental Engineering Research Center

EIC	Electret Ion Chamber
EPERM	Electret Passive Environmental Radon Monitor
EU	European Union
H	Tritium
HCL	Hydrochloric Acid
He	Helium
IAEA	International Atomic Energy Agency
ICRP	International Commission on Radiological Protection
IDC	Individual Dose Criterion
K	Potassium
LAEC	Lebanese Atomic Energy Commission
LET	Linear Energy Transfer
LIBNOR	Lebanese Standards Institution
LNHB	Laboratoire National Henri Becquerel
MCA	Multi-Channel Analyzer
MDA	Minimum Detectable Activity
NCRP	National Council for Radiation Protection
NLT	Linear Non Threshold
NRC	National Research Council
P	Phosphorus
Pb	Lead
PIPS	Passive Implanted Planar Silicon
PMT	Photo Multiplier Tube
Po	Polonium

PSA	Pulse Shape Analysis
Pu	Plutonium
Q	Quality Factor
Ra	Radium
Rn	Radon
Ru	Rubidium
SLWE	South Lebanon Water Establishment
Sr	Strontium
SSB	Silicon Surface Barrier
SSNTD	Solid State Nuclear Track Detector
Sv	Sievert
$T_{1/2}$	Half-life
TDS	Total Dissolved Solids
Th	Thorium
TID	Total Indicative Dose
U	Uranium
UN	United Nations
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
WHO	World Health Organization
Y	Yttrium

To the soul of my mother who taught me the true meanings of patience and devotion,

To my father who led me through roads of integrity, ambition and determination,

To the martyrs who sacrificed their lives for South of Lebanon liberation.

CHAPTER I

INTRODUCTION

1.1 Introduction

Access to clean water is one of the United Nations sustainable development goals and it plays a major role in maintaining good health (Sachs 2012). Continuous assessment of drinking water characteristics is essential for validating the water quality and its suitability for human consumption. Radioactivity concentration is one of several water characteristics that may affect the quality of drinking water (WHO 2011) and thus should be effectively monitored.

Radioactivity transforms atoms to more stable ones by the emission of excess energy in their nucleus in the form of radiation. This radiation is energetic enough to cause ionization and eventually introduce biological transformations into the human body. As such, the presence of radionuclides in drinking water at certain concentrations makes this water unsafe for general public consumption.

There are two main sources of radiation that can contribute to radioactivity concentration in water: anthropogenic and naturally occurring (Pearson et al. 2016). Anthropogenic radionuclides, such as Cesium-137 (^{137}Cs), Plutonium-239 (^{239}Pu) and Strontium-90 (^{90}Sr) are produced artificially and are usually introduced into the environment through human activities that include, but are not limited to, nuclear weapons testing and usage, nuclear power plants, and nuclear accidents (Hu et al. 2010). On the other hand, natural radionuclides include cosmic rays entering the earth atmosphere and terrestrial rays that have an origin from the earth crust. Beryllium-7 (^{7}Be), Carbon-14 (^{14}C) and Tritium (^{3}H) are examples of natural

radionuclides produced by cosmic radiation. Whereas, terrestrial radiations include the long-lived radionuclides that undergo simple decay forming parent-daughter pair like potassium-40 (^{40}K) and rubidium-87 (^{87}Ru), and the components of the long-lived uranium (^{238}U), thorium (^{232}Th), and actinium (^{235}U) decay chains (L'Annuziata 2012). The long lived members of the ^{238}U, ^{235}U, ^{232}Th series as well as the singly occurring ^{40}K are primordial radionuclides that have been existing since the formation of earth. They are present in trace amounts in nature and in human environment. Thus, they are responsible for the major portion of human exposure to radiation form natural terrestrial sources (Khan et al. 2011). Stability of all three chains occurs after series of alpha and beta decays which end with stable lead (NRC 1999). Figure 1.1 shows the three main natural radioactive decay series, the half-life of each decay progeny in the 3 decay chains, and the decay modes which are mainly alpha and beta frequently accompanied with gamma rays (Crawford-Brown 2011).

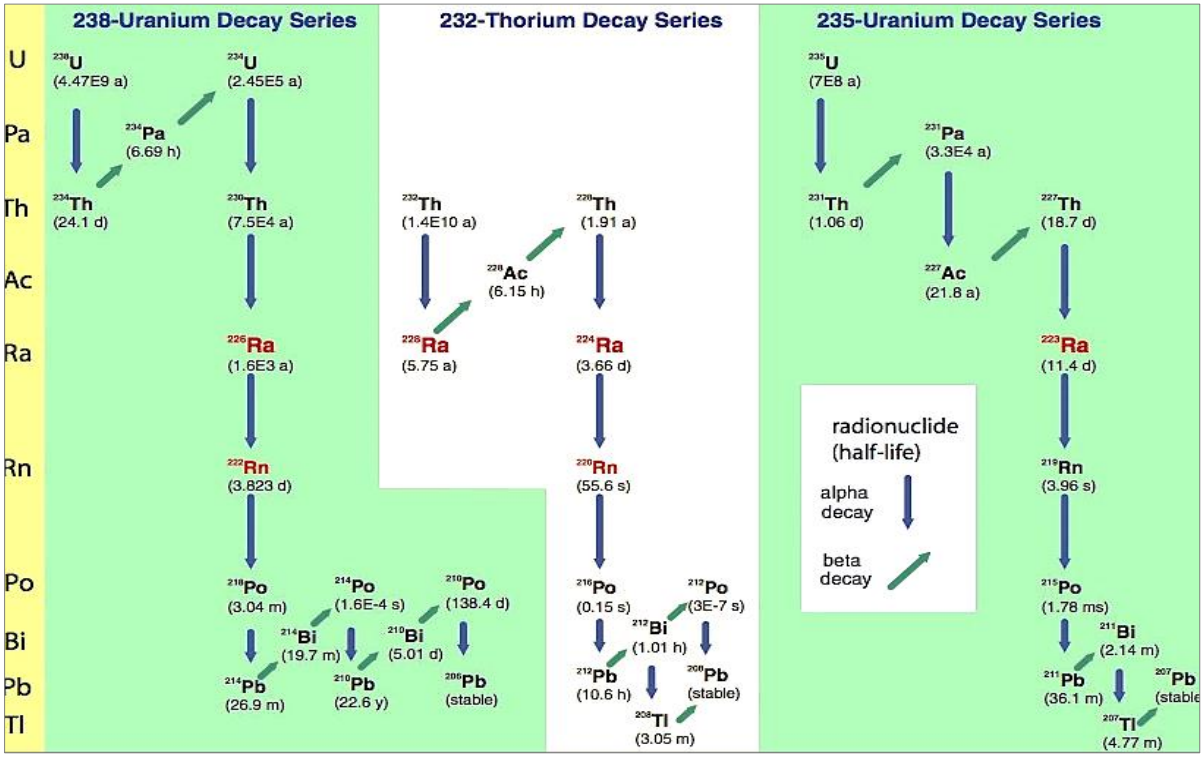


Figure 1.1 Decay series of ^{238}U, ^{232}Th and ^{235}U (Crawford-Brown 2011)

Natural radioactivity can get into drinking water through fallout of radionuclides produced by cosmic rays, or through discharge of terrestrial radionuclides into water bodies (L'Annuziata 2012). Several factors affect the radioactivity concentrations in drinking water. Depending on the specific radionuclide, these factors include the surrounding geological formations, soil characteristics, contact time, temperature, pH, and conductivity (Bajwa et al. 2017).

Naturally occurring radioactivity in water is of great importance when radiation exposure is concerned because it constitutes a significant percentage of background radiation exposure. The main radionuclides that are naturally occurring in water are uranium (^{238}U, ^{234}U), radium (^{226}Ra), radon (^{222}Rn), and polonium (^{210}Po). In fact, ^{226}Ra, a member of the uranium decay chain, and its progeny, radon, are responsible for the highest human radiation dose from naturally occurring radionuclides (IAEA 2014).

Several standards have been developed to control the health risks associated with prolonged radiation exposures. In the European Union (EU), the radioactivity levels for drinking water quality are regulated under two indicative standard limits: Tritium activity of 100 $mBqL^{-1}$ and a "total indicative dose" (TID) of 0.1 mSv/year representing the effective dose from radionuclides in drinking water except for ^{3}H, ^{40}K, and radon and its progenies (EC 2001). The Word Health Organization (WHO), on the other hand, set guidance levels for radiation dose from drinking water based on an "individual dose criterion (IDC) of 0.1 mSv/year. The IDC representing "a very low level of risk that is not expected to give rise to any detectable adverse health effect" (WHO 2004; WHO 2017). "Sievert" (Sv) is the unit of biological effect of ionizing radiation.

Based on the IDC, WHO developed a screening mechanism for radiological water quality assessment. The screening method consists of an initial measurement of gross alpha and gross beta activity concentrations produced by the simultaneous decay of radionuclides in the tested water. Further investigations, to check for other specific radionuclides are to be initiated if the activity concentration in the screening phase was found to exceed the set guidance levels of 0.5 BqL^{-1} for gross alpha activity concentrations and 1 BqL^{-1} for gross beta activity concentrations (one "Becquerel" (Bq) is the unit of radioactivity corresponding to the disintegration of one nucleus per second). Nevertheless, the set screening limits for gross alpha and gross beta activity concentrations are not to be considered as indications that the water is unsafe for consumption; they are rather meant to trigger proceeding to further radiological water quality assessments (WHO 2004; WHO 2017). For radiation exposure control purposes, guidance limits are set for the common natural and artificial radionuclides that might be present in drinking water. For instance, the limits set by the WHO for ^{238}U and ^{234}U are 10 BqL^{-1} and 1 BqL^{-1} respectively. WHO also provides a provisional guidance value of 30 μgL^{-1} for total uranium content based on its chemical toxicity rather than its radiotoxicity (WHO 2017). The 30 μgL^{-1} natural uranium, when the uranium activity ratio ($^{234}U/^{238}U$) is equal to 1 (in equilibrium), corresponds to 0.37 BqL^{-1} of ^{238}U. As for radon, WHO did not provide screening and guidance levels for it because 90% of the radiation associated to drinking water is attributed to exposure due to inhalation of the radon gas as it escapes from the water into the ambient air rather than from the ingestion of water (UNSCEAR 2000).

Whereas, the EU set a recommendation level of 100 BqL^{-1} for radon activities in water, and at higher than this level considerations for possible remedial actions are warranted (EC 2001).

In Lebanon, the characteristics of drinking water quality and contaminants limits were set by the Ministry of Environment (MOE) decision 52/1 (MOE 1996) and in decree number 1039 (MOI 1999), however radioactive concentrations were not included in this decision. Guidance for radioactivity concentrations has been addressed in the standards for drinking water issued by the Lebanese Standards Institution (LIBNOR) where the same activities limits set by WHO for gross alpha, gross beta, ^{238}U and ^{234}U activity concentrations were adopted (LIBNOR 2016). Nevertheless, the LIBNOR standards are non-binding and compliance is voluntary.

The radiological aspects of water quality in Lebanon are not included in routine quality control tests performed to check for the suitability of the drinking water for human consumption and the radioactivity in water is never included in regular monitoring of the drinking water supplies. As such, information on the radiological aspects of the Lebanese drinking water resources are scarce. In fact, environmental radioactivity in water in Lebanon was addressed through a handful of studies which performed preliminary radioactivity measurements to assess the associated radiation risks. Yet, none of these studies was focused on screening radioactivity in drinking water in the southern region of Lebanon.

For instance, a preliminary radon assessment study using E-PERM technology, performed in the main administrative districts of Lebanon, revealed that radon activities in water were below the 100 BqL^{-1} recommendation level set by the EU (Abdallah et al. 2007). E-PERM, the electret passive environmental radon

monitor system, is a passive method for radon measurement using electret ion chamber (EIC) (Kotrappa et al. 1988; Kotrappa et al. 1990). Radon concentrations in water were investigated in various locations in Lebanon using Columbia Resin #39 (CR-39) nuclear track detectors without recording any significant levels (Hashim et al. 2014). CR-39 is a kind of solid state nuclear track detectors (SSNTD) used for passive radon dosimetry (Ismail and Jaafar 2009; Nikezić 1994). Public exposure to radioactivity levels in Lebanon was assessed within the framework of the National Environmental Monitoring Program at the Lebanese Atomic Energy Commission (LAEC). Radioactivity in various matrices was assessed including water samples from monitored rivers and wells. Thorium and radium activity concentrations in rivers and gross alpha, gross beta, radium and uranium activity concentrations in wells samples were measured. The reported activities, in both medium, were lower than the WHO guidance levels, and the recorded average for annual radiation dose was determined to be lower than the total worldwide value (El Samad et al. 2012; El Samad et al. 2016).

In the Southern region of Lebanon in specific, to the best of our knowledge, the only two natural radioactivity studies that covered this region were performed to measure radon activities in air and did not target radioactivity in water. While noting some seasonal variations, Habib et al. 2018 (Habib et al. 2018) reported radon concentrations indoors (inside dwellings) and outdoors to be below the International Commission on Radiological Protection (ICRP) recommended levels of 300 Bq m^{-3} (ICRP 2009). On the other hand, Kobeissi et al., also investigated indoor and outdoor radon concentrations, reported few locations where the effective dose from radon

activity was found to be above the 1 $mSva^{-1}$ UNSCEAR recommended limit for general public members (Kobeissi et al. 2014; UNSCEAR 2000).

These results, in addition to the particularity of the Southern region of Lebanon which was subject to many military conflicts that raised suspicions of using depleted uranium, motivated proceeding to further radioactivity investigations in that region. It is worth mentioning, however, that public concerns about the military use of radioactive materials in South Lebanon might be exaggerated or unjustified especially that the LAEC performed a field study after the 2006 summer Israeli attack that confirmed that the uranium concentrations are within the normal environmental values and that enriched or depleted uranium are absent in the analyzed samples (El Samad et al. 2007).

1.2 Study Objectives

The exposure to radiation from trace radionuclides naturally occurring in drinking water carries some risk of harmful biological effect due to the induced ionization at the cellular level. In Lebanon, there are no regulations that control the allowable levels of natural radioactivity in drinking water, and information about radioactivity concentrations in water are very limited with a complete absence of such information in the Southern region of Lebanon.

Owing to the scarcity of data on environmental radioactivity in the region, in water specifically, the main objective of this research is to measure the natural radioactivity in drinking water in this region of Lebanon to assess its suitability for human consumption. For this purpose, gross alpha and gross beta, naturally occurring uranium isotopes and radon activity concentrations were measured, over

two seasons, in various types of drinking water supplies including springs, wells and tap water: Gross alpha and gross beta radioactivity concentrations for being the first evaluation technique in any radioactivity assessment, uranium for being on top of the most abundant naturally occurring decay chains and a possible contaminant of water sources from anthropogenic activities, and radon for being responsible for the highest radiation dose received from naturally occurring radioactivity and being attributed to lung cancers (ICRP 2010).

1.3 Significance of the study

The proposed research aims at assessing the natural radioactivity in drinking water in the Southern region of Lebanon. The measurements were taken over two seasons and from various drinking water supply sources, as such the study:

- Serves as a baseline assessment of natural radioactivity of drinking water and may be used as a reference for any future investigation of the radiological characteristics in this area and activities changes evaluation.
- Constitutes a screening analysis that determines the need for further testing to identify the presence of other radionuclides.
- Assesses the seasonal variations in activity concentrations of investigated radionuclides.
- Evaluates the effects of existing water treatment/distribution systems on radioactivity concentrations.
- Provides decision makers with information about the radiological quality of drinking water in Southern Lebanon and helps them in making decisions regarding future drinking water regulations.

- Serves as a preliminary monitoring indicator in seismic studies as it investigates radon level in groundwater which was proven to be a precursor for earthquake prediction (Baskaran 2016); especially that the study area lies mostly in a location largely affected by the main faults of Lebanon namely: Yammoune, Hasbaya, Rachaya, Serghaya and Roum – the fault that is expected to take the largest offset in "future geological time" (Khair 2001).

1.4 Limitations

This study is meant to be a preliminary assessment, with limited number of samples, due to several limitations that inhibit considering a larger number of samples for radioactivity analysis. These limitations include the requirement of meticulous sampling procedure, lengthy samples preparation process, expenditures associated with samples collection, preparation and analysis, counting equipment accessibility and time availability for the current project.

1.5 Thesis organization

The first chapter included the introduction in which the background of radioactivity in water as a contaminant, the objectives of the thesis, and the significance were described.

Chapter 2 defines the principle of radioactivity, its characteristics and biological effect, summarizes the occurrence of radioactivity in water especially for the radionuclides in question in this study, and describes the general applied

radioactivity measurement principles and particularly those to be followed in this study.

Chapter 3 describes the methodology that was followed during the research and the adopted radio-analytical processes.

Chapter 4, displays the results of the performed radio-measurements and the proposed interpretations in terms of seasonal variation, water treatment and geological formations.

Chapter 5 represents the deduced conclusions from the analyses of the results and depicts the proposed recommendations.

CHAPTER 2

LITERATURE REVIEW

2.1. Radioactivity

A dedicated account on radioactivity including its forms, exposure ways, biological effects and dose-response relation is supplemented in Appendix 1.

2.2. Natural Radioactivity in water

The environment contains several forms of anthropogenic and naturally occurring radioactivity which can get into the water bodies (ground waters and surface waters). Desorption from the soil and wash off by rain are main causes for the presence of naturally occurring radionuclides in water (^{40}K, ^{232}Th, ^{238}U, ^{234}U and their decay products - particularly ^{226}Ra, ^{228}Ra, ^{222}Rn and ^{210}Pb). Other reasons for the presence of naturally occurring radioactivity in water include the release of radio-elements due to processes that comprise naturally occurring radionuclides like processing of mineral sands, mining, and phosphate fertilizers production and use (Eisenbud and Gesell 1997).

The concentrations of naturally occurring radionuclides are not the same in all water bodies. It varies depending on the type of water body, the geological characteristics of the aquifer from which the water generates, composition of adjacent rocks, contact time, temperature, pH and conductivity (Bajwa et al. 2017). Also, the radioactivity depends on the degree of mineralization of the local geology. It tends to be higher in igneous granitic rocks that are rich in uranium, yet it can also be affected by the pH of the medium. Sedimentary rock and intermediate soils have

lower levels of radioactivity than granitic rocks (Table 2.1) (Eisenbud and Gesell 1997).

In addition, groundwater is expected to have radioactivity concentrations of naturally occurring radionuclides higher than in surface water due to their contact with specific geological formations especially when the physiochemical characteristics are favoring the solubility of natural radionuclides (Table 2.2).

2.2.1. Gross alpha and gross beta

Drinking water may contain radionuclides contaminants of natural or anthropogenic origin. Amongst these radioactive contaminants are alpha emitters radionuclides likes ^{238}U, ^{234}U, ^{232}Th, ^{226}Ra, ^{210}Po, and the beta emitters like ^{40}K, ^{210}Pb, ^{212}Pb, ^{228}Ra, … The measurement of gross alpha and gross beta activity concentration is not meant to be absolute determination of the entire radioactive contents in the sample. It is rather considered as a preliminary screening to determine the need for further investigation about specific radionuclides content in the tested water if the regulatory guidance levels were exceeded. The measurement of gross alpha and gross beta activity concentrations is not, however, expected to provide results as accurate as when performing radioanalysis for specific radionuclides after radiochemical separation (BSI 2018): A more expensive and time consuming procedure for assessment of drinking water quality.

2.2.2. Uranium

Uranium (Z= 92) is the heaviest naturally occurring radionuclide. It has three naturally occurring alpha emitter isotopes ^{238}U (99.28%), ^{235}U (0.71%) and

^{234}U (0.0058%) (Eisenbud and Gesell 1997). Naturally occurring uranium isotopes decay slowly with half -life of $4.468.10^9$ year for ^{238}U, $7.04.10^8$ years for ^{235}U and $2.455.10^5$ years for ^{234}U. It belongs to the actinides family and has 4 oxidation state: U3+, U4+, U5+ and U6+, but uranium ions in 4 and 6 oxidation states are the most commonly present ones and U4+ is the most soluble in water (IAEA 1988). Uranium ions can travel many kilometers from its rock source before they precipitate as uranium mineral like FeO(OH) (Finch and Murakami 1999).

Uranium can be found in all components of the environment. Its concentration in the earth's crust varies from 0.3 $\mu g \cdot g^{-1}$ to 12 $\mu g.g^{-1}$ (UNSCEAR 2008). The ^{238}U current worldwide average value is 33 $Bq.kg^{-1}$ in soil, but the reported concentrations vary largely to reach as high as 1,000 $Bq.Kg^{-1}$ (UNSCEAR 2008). In surface water, groundwater and oceans, uranium concentrations varies from less than 1 $\mu g \cdot L^{-1}$ to more than 10 $\mu g \cdot L^{-1}$ (WHO 2011). Uranium can be found in variant concentrations in most of rock types and soils. Its concentration can be as low as 0.003 ppm in meteorites rocks and as high as 120 ppm in phosphate rock (Table 2.1); the thing that can be translated into highly regional fluctuations in concentrations. In fact, magmatic rocks with high uranium content have more radioactivity than sedimentary zones (Eisenbud and Gesell 1997) . Furthermore, uranium tends to be of a higher concentrations near uraniferous aquifers and in areas with granitic intrusions (Wisser 2003).

Uranium is a heavy metal that has chemotoxic properties with deleterious effects on human health. Its chemotoxicity is even more predominant than its radiotoxicity. Actually, several studies have reported the nephrotoxic properties of

uranium. (Banning and Benfer 2017; Canu et al. 2011), while data related to the carcinogenic effect in human are still insufficient (WHO 2005).

Table 2.1 Average uranium concentrations in various rocks (Eisenbud and Gesell 1997)

	Uranium concentration	
Rock Type	PPM	$BkKg^{-1}$
Phosphate rock (Florida)	120	1500
Bituminous shale (Tennessee)	50-80	610-980
Normal granite	4	49
Acid igneous	3	37
Intermediate igneous	1.5	18
Limestone	1.3	16
Basic igneous	0.6	7.3
Ultra-basic igneous	0.03	0.37
Meteorites	0.003	0.037
Other sedimentary rocks	1.2	15

2.2.3. Radon

Radon is a naturally occurring noble radioactive gas that is odorless, colorless and tasteless. It originates from rocks charged with uranium. All ground water contains some radon concentrations ranging from 4 to 40000 Bq/L depending on the uranium content of the soil. Radon concentrations may present some seasonal variation depending on water table (UNSCEAR 2008). Actually, radon results from the decay process of ^{226}Ra, ^{224}Ra and ^{223}Ra that are themselves elements of the ^{238}U, ^{235}U and ^{232}Th decay series respectively. Soluble radon in water results from the

decay of dissolved ^{226}Ra and also from the emanation of the inert gas from adjacent soils and rocks into the water bodies (L'Annuziata 1998).

Radon decays by emission of the highly ionizing alpha particulates that can find its way into the human body through ingestion or through inhalation. Actually, radon is responsible for 54% of the naturally occurring background radiation. It is classified as a carcinogen that affects mainly the lungs (IARC 2012). Amongst the three radon isotopes, ^{222}Rn ($T_{1/2}$ = 3.82 days) has the major dose contribution due to its abundance and relatively longer half-life than the less important ^{220}Rn ($T_{1/2}$ = 55.6s) and the negligible ^{219}Rn ($T_{1/2}$ = 3.92 s) isotopes.

Radon concentration in water depends on many factors including uranium content of rocks, type of soil, porosity, salinity, temperature and depth of water. The solubility of radon, for example, decreases with an increase in temperature (Kulali et al. 2017). Radon can easily get released form water when it gets into contact with the air; the reason why surface water like in rivers and lakes generally tend to have lower radon concentrations. For instance, the radon activity concentrations can be as low as less than 1 BqL^{-1} in surface water and goes up to more than 1000 BqL^{-1} near uranium rich rocks (Table 2.2) (UNSCEAR 2008).

Table 2.2 Radon concentrations in various water

Type	**Formation**	**Radon concentration (BqL^{-1})**
Surface water	-	< 1
Groundwater	Sedimentary rock aquifers	1 to 50
Wells	-	10 to 300
Rocks	Crystalline rocks	100 to 1000
	Rock with high U concentration	> 1000

2.3.Measurement of radioactivity in water

2.3.1. Gross alpha and gross beta

Several methods have been developed for the measurement of gross alpha and gross beta activities in water such as those based on proportional counting and scintillation counting where a water sample is evaporated till dryness and measured on proportional or scintillation counters. These methods are dependent on the TDS and chemical composition of the sample which might affect the counting efficiency (Atsor et al. 2015; Bonotto et al. 2009; Jobbágy et al. 2010)

However, the excellent ability for liquid scintillation counters in discriminating alpha and beta energy and the simple preparation requirements, as well as the counter ability for performing low background reading are making it the procedure of choice for the measurement of gross alpha and gross beta activity concentrations in environmental samples, water in particular (Çakal et al. 2015; Stojković et al. 2017).

In our study, gross alpha and gross beta activities were measured using the liquid scintillation counting.

2.3.1.1.Liquid scintillation counting principle

Liquid scintillation counting technique relies on the principle of converting the kinetic energy of a nuclear emission into photons of light. In fact, samples containing radioactivity are dissolved or suspended inside a plastic or glass vial containing a scintillation cocktail. A series of interaction between the emitted nuclear energy of the sample and the components of the cocktail (solvent, scintillator like Fluor and sometimes an emulsifier) leads to the emission of a photon light with an

intensity proportional to the initial energy of the ionizing rays. The emitted photon light hits the cathode of two photomultiplier tubes (PMT) which amplify the incident light and emit a pulse. The PMTs convert the light into an electrical pulse with an energy proportional to the intensity of the amplified light, and eventually to the initial radioactivity in the sample (Kessler 2015). The LSC is equipped with a coincidence circuit that differentiates between real scintillation events and noise. Real events are the pulses emitted simultaneously by the two PMTs of the LSC and are detected by the coincidence unit. Only those pulses that are accepted by the coincidence circuit are processed further. The electrical pulses are registered as counts that are sorted according to their amplitude into separate channels and are then used collectively to generate a spectrum for the sample. The spectrum will be used to calculate the activity in the samples as counts per minutes.

Figure 2.1 below shows the details of the liquid scintillation counter and its components (Seoane and Llovet 2012).

Alpha and beta emitters can be counted simultaneously in liquid scintillation samples. The separation between alpha and beta nuclides energy is done based on the fact that alpha and beta radionuclide have different pulse decay time and produce different pulse shape at the PMT. It is done via an electronic circuit which distinguishes signal pulses according to their pulse height or voltage. This option is referred to by different instrument manufacturers as pulse shape analysis (PSA), pulse shape discrimination, and pulse decay analysis (L'Annuziata 2012). The simultaneous measurement of gross alpha and gross beta activity concentrations in water samples is possible due to adjustment of the setting of the PSA option in the liquid scintillation counter. The efficiency of the alpha-beta discrimination depends

on the energy of alpha and beta. Therefore the counter needs to be optimized for optimum discrimination using pure alpha and pure beta emitters with energies near to that expected to be present in the sample (BSI 2018).

Several scintillation cocktails have been developed for use with LSC. Making the right decision in selecting the most suitable cocktail, based on the matrices and the form of the samples to be counted, is essential for improving the efficiency of counting (Edler 2006). Some liquid scintillation counters are capable of detecting very low activities, like the trace amounts expected in most of environmental samples, due to an improved efficiency and enhanced ability for background discrimination. Employing heavy lead shielding, installation of improved detection system, and also fitting an optional low scintillating Bismuth Germanium Oxide detector guard are also some methods of improving the sensitivity of the LSC to make it more suitable for low activities counting (Kessler 2015; L'Annuziata 2012)

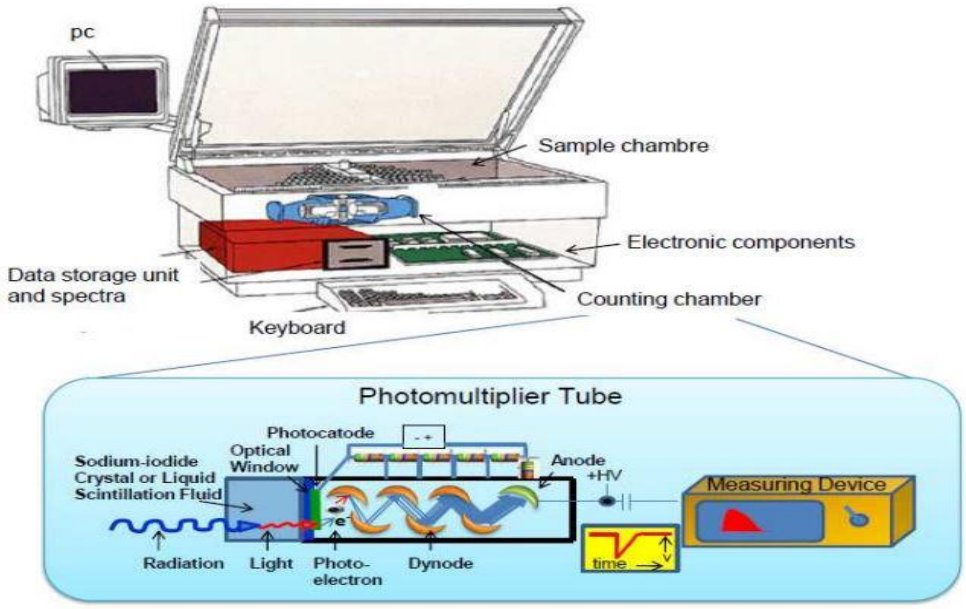


Figure 2.1 Liquid scintillation counter components (Seoane and Llovet 2012)

2.4.2. Measurement of Radon

Radon measurement in drinking water can be performed by the following three main principles: gamma ray spectroscopy, emanometry, and liquid scintillation counting.

In gamma spectroscopy, the radon concentration is obtained through the measurement of its gamma emitter decay products, ^{214}Bi and ^{214}Pb. This method is negatively influenced by the presence of ^{226}Ra, temperature, suspended materials, air bubbles, and the sample density and homogeneity, (BSI 2013a; Tsukuda 2008).

The emanometry principle for radon measurement is based on degassing the radon from water and reading the alpha particles with a detection system such as Lucas cell or Alpha Guard. This method is influenced by the water temperature and the possibility of concentration build-up of radon daughters inside the detector (BSI 2013b; Rangaswamy et al. 2016).

As for the use of liquid scintillation counting method for radon measurement, it relies on the principle of extracting radon gas from water in the sample into an immiscible scintillation cocktail. Water injected under the cocktail forms a two phase aqueous-organic separation. Radon trapped between the two phases is measured by LSC as described in section 2.3.1.1. This method is popular owing to the high solubility of radon in organic solvents such as the cocktail used in scintillation counting and also to the simple preparation steps. However, careful samples collection as well as setting proper alpha/beta discrimination and calibration are the main challenges for getting the desired level of efficiency in counting (L'Annuziata 1998).

In our study, radon activities were measured using the liquid scintillation counting.

2.4.3. Measurement of Uranium

Several methods have been reported in the literature for the analysis of uranium. Mass spectroscopy, liquid scintillation counting and alpha spectrometry are the most practiced techniques (Hao et al. 2011; Parrish et al. 2006; Yoon et al. 2012). However, since we will be using alpha spectrometry for uranium analysis in our study, we will be focusing more on this technology.

Mass spectroscopy measures the mass to charge ratio of ions and generates a mass spectrum which represents the masses of the sample components. It offers more sensitivity for measuring ^{238}U than the radiometric methods. However, it gives less information about its short-lived isotopes, for these the radiometric methods are a better choice for assessment. The simple sample preparation, short measurement time, and the small sample size are all advantages for mass spectroscopy (Hou and Roos 2008). However, the high capital and maintenance cost are limitations for its routine use for environmental analysis (Jia and Jia 2012).

Liquid scintillation counting on the other hand has been used in uranium analysis for its easy samples preparation requirements, its practicality and cost effectiveness, and because it can provide information about the radionuclide composition of the water sample using the alpha-beta discrimination technique. A draw back for the usage of LSC for uranium analysis is its inability to discriminate between ^{238}U and ^{234}U due to its poor resolution. This will create a potential for underestimation when accounting for the radiation dose received form the water especially in groundwater where ^{238}U concentrations are often lower than those of ^{234}U (L'Annuziata 2012).

Alpha spectroscopy with solid states detector is, however, the most commonly used technique for alpha emitters' measurement. It relies on isotopes differentiation based on its alpha energies. Table 2.3 shows the different uranium isotopes and their characteristics (LNHB).

Table 2.3 Uranium isotopes characteristics (LNHB)

Uranium Isotope	**Half-Life** (Years)	**Main emission energy** (KeV)
232	70.6 (± 1.1)	5263.48
		5320.24
233	159.1 (± 0.2) *10^3	4783.5
		4824.2
234	2.455 (± 0.006) *10^5	4722.4
		4774.6
235	704 (± 1) *10^6	4366.1
		4397.8
		4414.9
236	23.43 (± 1) *10^6	4445
		4494
238	4.468 (± 0.005) *10^9	4151
		4198

Alpha spectroscopy technique is based on transforming the alpha energy into electrical pulses plotted on a spectrum with a peak energy equal to that of the energy of the incident alpha particle. Considering that alpha penetrating range is very limited, a high resolution performance, low background, excellent stability and high permissible counting rates detector is required for proper detection of alpha particles

(El-Galy et al. 2011). In addition, the element to be measured needs to be chemically separated to eliminate chemical or radiological interference from other components in the sample, and the sample needs to be thin enough to avoid self-attenuation. As such, water sample needs to undergo certain preparation to be ready for the chemical separation and the formation of thin layer that can be read by the alpha spectrometer (L'Annuziata 1998). The artificial isotope ^{232}U, is usually added to the sample as a chemical recovery tracer (Lytle et al. 2014).

During sample preparation for uranium analysis, the water is acidified to avoid bacterial growth and to maintain the solubility of the trace elements in the sample and prevent it from getting adsorbed on the wall of the container. Following acidification, an artificial uranium radiotracer, ^{232}U, is added to the sample and is homogenized by stirring. This is followed by a concentration step especially when the uranium concentration in the sample are expected to be low. Concentration is performed mainly by evaporation or coprecipitation.

Chemical separation is performed to purify the sample and remove mineral and organic components, other than uranium, from the sample and to eliminate interference from other alpha emitters like thorium, radium neptunium and plutonium. Separation can be done by several methods like ion exchange and chemical extraction (BSI 2014)

After chemical separation, the uranium in the sample will be ready to be mounted for reading using alpha spectrometer. The two most practiced mounting methods are electrodeposition on a stainless steel disk and coprecipitation as fluoride (Fettweis and Schwenn 1998).

The alpha spectrometry components (Figure 2.2) consists of a counting chamber containing a detector where counting is made under almost vacuum conditions. The two most commonly used detectors are the Silicon Surface Barrier (SSB) and the Passive Implanted Planar Silicon (PIPS) low background semiconductor detectors. The sample is mounted inside the chamber on an adjustable height tray underneath the detector which has a specific active area for radiation detection. As the alpha particles interact with the active surface of the detector at the electronic level, very low amplitude electrical signals proportional to the initial incident radiation are produced. The low amplitude signals are amplified by a very low noise preamplifier, then are passed to a shaping amplifier circuit. Finally, the signals are sent to an analogue to digital convertor (ADC) and a multichannel analyzer (MCA) which build a spectrum based on the measurement of the pulse height. The spectrum is more like a "histogram of pulses classified as a function of their pulse height" (Fettweis and Schwenn 1998).

Alpha spectrometry offers accurate $^{234}U/^{238}U$ activity ratio determination. However, it requires meticulous chemical preparation and long counting time. Besides that, the alpha resolution may get affected if the mounted sample was not thin enough (Salonen et al. 2012).

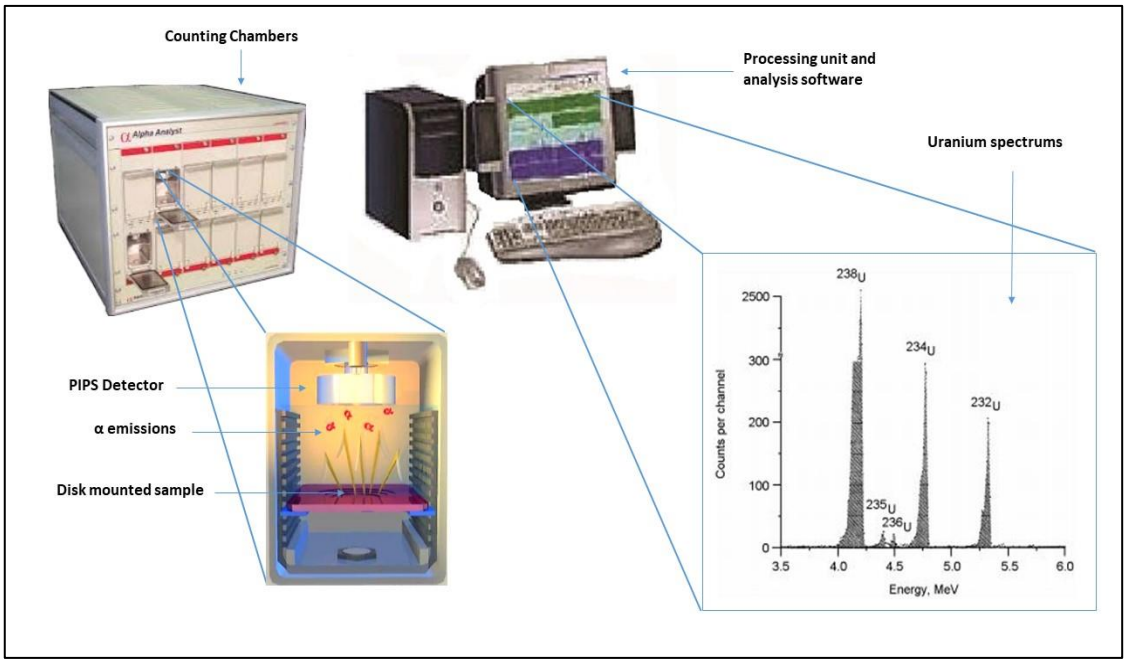


Figure 2.2 Alpha Spectrometry, adapted from (Canberra) and (Jia et al. 2002)

CHAPTER 3

MATERIALS AND METHODS

3.1. Study area

As previously mentioned, this research study aims at assessing the natural radioactivity in drinking water in the Southern region of Lebanon. The following sections will describe this region and its main characteristics as related to the intended research as well as the methods used for sampling, measurement and calculations:

3.1.1. Geographical information

This study is focused on assessing the natural radioactivity in the Southern region of Lebanon which is located between the geographical coordinates of 33.600°-33.055°N and 35.101°-37.796° E. The region extends from the western shores of South Lebanon at the Mediterranean Sea to the Mount Hermon (Jabal El-Sheikh) foothills in the East.

The study region consists of two governorates: *Al Nabatieh governorate* which is composed of four districts: Nabatieh, Hasbaya, Marjeyoun and Bint-Jbeil, and *South Lebanon governorate* which includes three districts: Saida, Jezzine and Sour. The region extends over an area of 2025 Km^2 which is about 20% of the total area of Lebanon (MOE 2001). It hosts 17.3% of the total population of Lebanon with 956,500 residents, 359,100 of them in Al-Nabatieh governorate and 597,400 in South Lebanon governorate (CAS 2009).

3.1.2. Climate and precipitation

Lebanon enjoys typical Mediterranean weather with cool rainy winters and warm dry summers marked with significant daily irregularities in temperature and precipitations between the costal zones and the more interior and elevated areas. In fact, the wet season approximately extends from mid-September till end of May while the dry season spreads from June till August. Actual precipitation data from weather stations in 4 districts of the Southern Lebanon (LARI 2018), reflect this wet and dry trends between January 2015 and September 2018 (Figure 3.1).

In the study region, the precipitation varies between less than 400 mm on the coastal zone, and less than 1000 mm in the more elevated inland areas, with an average of 700 mm of precipitation per year (Arrighi de Casanova 2009).

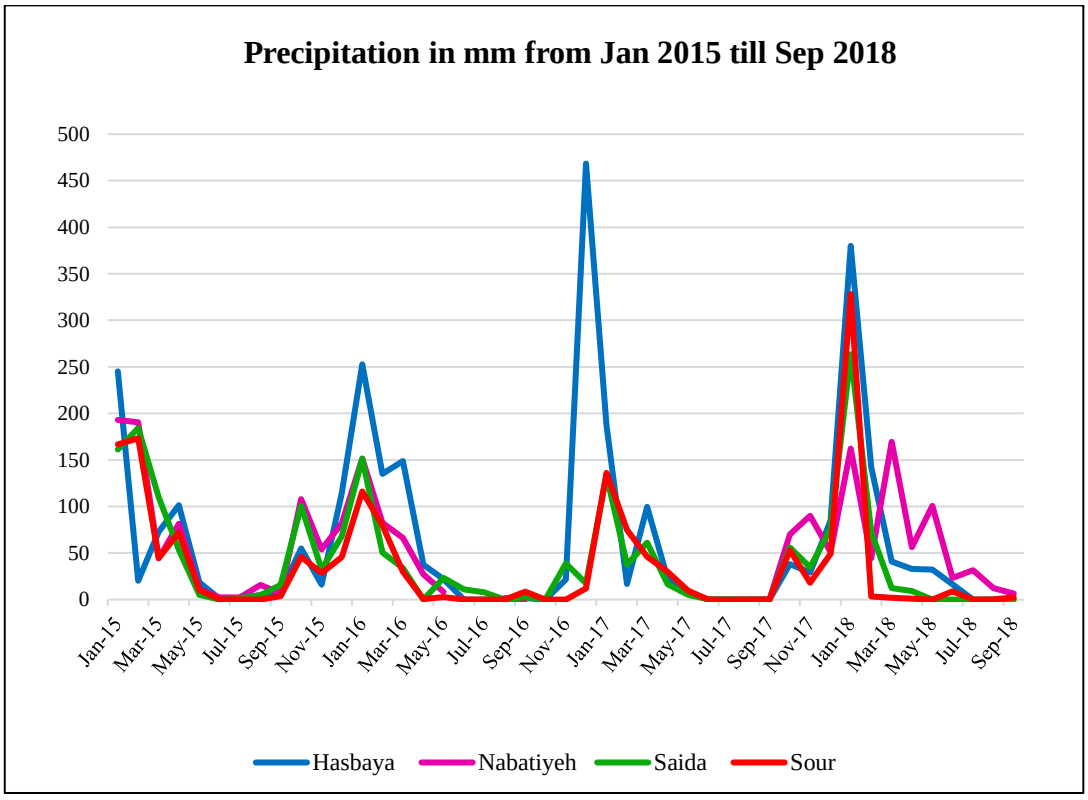


Figure 3.1 Precipitation trend in Southern Lebanon

3.1.3. Hydrogeology

Geological structure and lithology characteristics play major role in the formation of aquifers and the occurrence of natural radioactivity in rocks. It also determine the behavior of radionuclides in the water bodies in terms of leaching out of the adjacent rock formations into the water.

The geological formations in the study area extend form the Mesozoic up to the Cenozoic era, from the lower Jurassic to present period. However, there is prevalent domination of geological formations belonging to the cretaceous and lower tertiary period; mainly those of the Cenomanian (C4), and the Eocene (e) epochs. Other geological features occur as well in the region, but with a lesser extent, like the formations from the upper Jurassic period (J4-J7), the Aptian (C2), Turonian (C5), Senonian (C6) of the Cretaceous, Miocene (m) and Pliocene (p) of the Tertiary Paleocene, and the arable lands on the coastal zone belonging to the Quaternary epoch (qta) (Faour 2004). In addition, there are few intrusions of lower Cretaceous and Jurassic basaltic formations (Walley 1997).

Understanding the geology of the study area is essential for determining the underlying lithology. Figure 3.2, Figure 3.3 and Table 3.1 describe the geology and the lithological characteristics of the formations of each of the geological epochs as originally prepared by Dubertret (Dubertret 1955; Faour 2004).

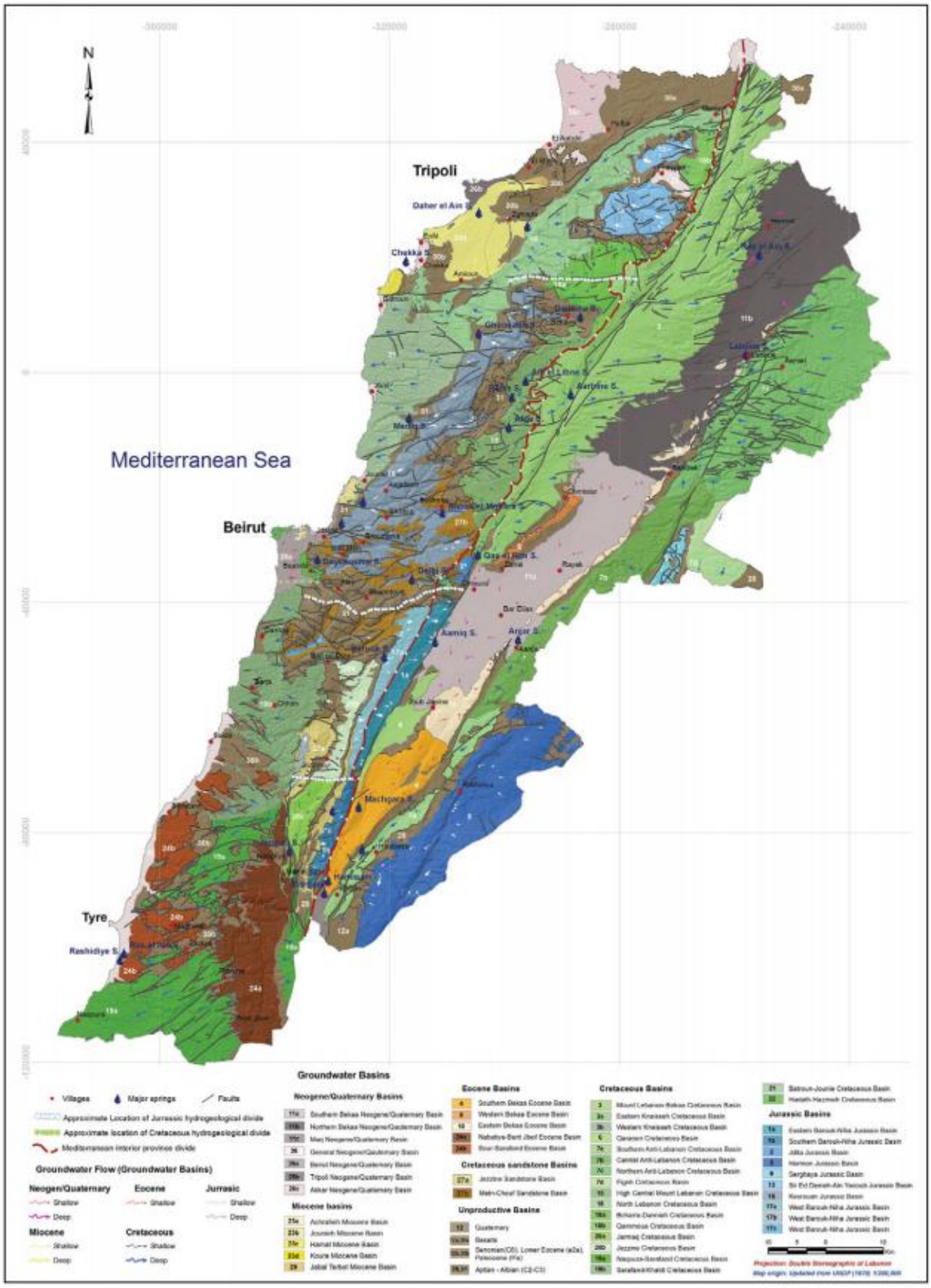


Figure 3.2 Hydrogeological map of Lebanon (MoEW/UNDP 2014)

Table 3.1 Stratigraphic column of Lebanon, by Dubertret (Faour 2004)

Era	Period	Epoch	Lithology
Cenozoic	***Quaternary***	Holocene, Pleistocene q	Marine deposits, river terraces, dunes, alluvial deposits
	Neogene	Pliocene p	Limestone, marl, volcanic materials (marine facies)
		Miocene m	Limestone, sandstone
	Paleogene	Eocene e	Limestone, chalky limestone
Mesozoic	***Cretaceous***	Senonian C6	Chalky marl
		Turonian C5	Chalky marl
		Cenomanian C4	Limestone, dolomitic limestone, marly limestone
		Albian C3	Marly limestone, marl
		Aptian C2	Dolomitic limestone, argillaceous sandstone, marl, limestone
		Neocomian-Barremian C1	Quartzy sandstone, beds or sets clayey grey
	Jurassic	Upper Jurassic J4-7	Dolomite and dolomite limestone

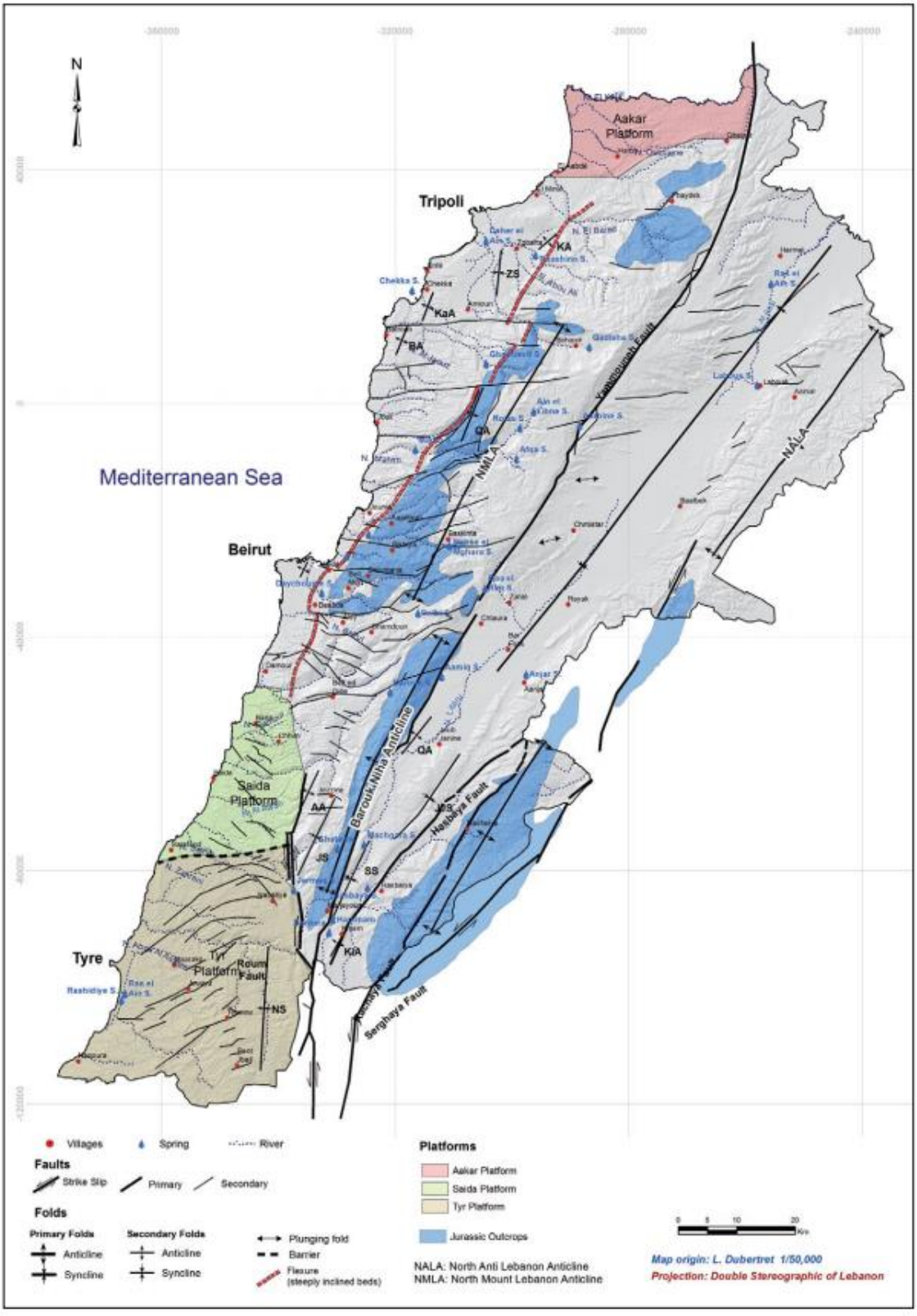


Figure 3.3 Faults of Lebanon (MoEW/UNDP 2014)

3.1.4. Water supplies

The study area relies mainly on groundwater of wells and boreholes, and on surface water of spring, rivers and dams to fulfill its need for drinking water. The public water supplies are mainly managed by the regional water authority, the South Lebanon Water Establishment (SLWE) in Southern Lebanon, and by the local municipalities in some villages. The public water supplies are usually subjected to water treatment processes to attain water quality standards for physiochemical as well as microbiological properties. Chlorination is the only water treatment that is applied to water drawn from wells and boreholes. Whereas, drinking water from rivers and dams usually undergoes more treatment steps to reach acceptable quality to be supplied for public use. These steps include primary treatment processes starting with filtration of insoluble solid subjects, removal of floating objects from the surface of water, gravity sedimentation in sedimentation tanks to remove solids that sink to the bottom, and chlorination before being pumped into the water transmission and distribution system. After treatment, water is sometimes directed to storage tanks or towers before delivery to the residents. Drinking water is supplied to consumers via distribution networks, or cisterns that collect the water from local treatment facilities or by hand-carried containers in some locations. The water supply chain is summarized in Figure 3.4 (MEW 2010).

In addition to the officially supplied water, there are also large number of licensed and unlicensed private wells that are used to satisfy the local needs for drinking water during periods when officially supplied public drinking water is not available (Farajalla et al. 2015). These unofficial and improperly managed wells are affecting the sustainability of groundwater aquifers especially during the shortage

periods which occur in the dry seasons (MOE/UNDP/ECODIT 2011). Beside water supplied from the aforementioned sources and due to the perceived uncertainties related to water quality delivered to the households taps, residents often resort to the purchase of bottled water as a source for drinking water (Semerjian 2011) and use tap water only for cooking, personal and domestic hygiene purposes (UN 2016).

In this research study, radioactivity is measured in natural water samples from wells, springs, and from tap water only; sealed bottled waters were not included.

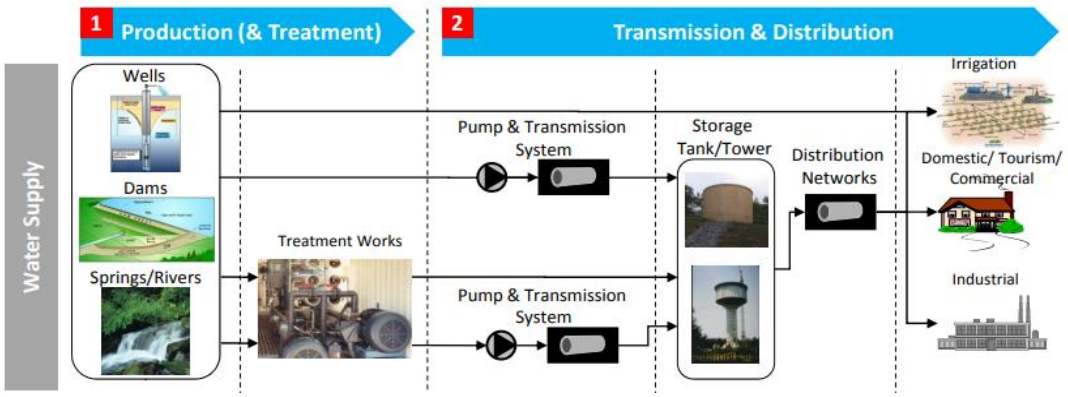


Figure 3.4 Water supply chain, adapted form (MEW 2010)

3.2. Sampling design and frequency

Samples from various locations in the study area were taken from drinking water sources and the respective taps. Location of sampling points and number of samples collected conformed to the general methods in environmental sampling and in natural radioactivity analysis studies where relatively small numbers of sampling points are taken. Sampling sites were chosen based on geographical location within a district and the convenience of access of the sampling points. Whenever possible, preference was given to take samples from the main sources supplying large number

of consumers. The number of collected samples was meant to provide an overview of radioactivity concentrations in the studied areas, rather than being a statistically representative sample of all existing water sources (Caridi et al. 2017; Jowzaeem 2013; Wang et al. 2017). In fact, lengthy preparation procedure, expenses, and counting equipment availability were limiting factors for conducting large number of samples in radioactivity analyses.

Six samples were collected from each of the seven districts of the region under investigation. Of the six samples, three were collected from groundwater and surface water sources (wells and springs) and the other three were collected from household taps related to the same water source (Figure 3.5) and (Table 3.2). Depending on the local geography, the distribution of drinking water through the network systems is sometimes done by gravity or through pumping into collection tanks or towers. Gravitational distribution is always the preferred option, so it happens sometimes that the water drawn from a certain source is directed towards a zone that is not within the administrative borders of the village or district from which it originates. Thus, tap water sampling was performed with close coordination with the SLWE to follow the distribution network and make sure that the selected tap for sampling at the consumer side, corresponds to the same sampled source.

Analyses conducted on tap water were intended to provide information on the behavior of radioactivity as it reaches the consumers' end, since radioactivity "can potentially enter the water supply at any point, or at several points, prior to consumption", or it can be "lost" during the treatment processes (WHO 2017). In fact, radionuclides such as uranium and thorium and radium have been found in sludge produced from potable water treatment (Fonollosa et al. 2015; Palomo et al.

2010a; Palomo et al. 2010b). Similar results were also reported by Al-Jaseem et al. who also reported some variation in indoor radon concentrations inside the treatment plant due to radon release into the air (Al-Jaseem et al. 2016). Radioactive contaminants (Gross alpha, gross beta, radium thorium, and uranium activities) were also proved to accumulate in the drinking water distribution systems as well (Lytle et al. 2014).

Water sampling from the same locations were conducted during the wet and dry seasons to investigate possible seasonal variations in radioactivity concentrations. Thus, drinking water samples were collected from 42 locations, and natural radioactivity investigations took place over two seasons through two sampling rounds – giving a total of 84 samples in both seasons.

Actual precipitation data were collected from weather stations installed in four out of the seven districts namely: Hasbaya, Nabatieh, Saida and Sour. Precipitation data from January 2015 to September 2018 show the general trend and durations of the wet and dry seasons: a wet season extending between September and May and a dry season extending from June till August (LARI 2018). Samples collection in this study was performed in the period extending between February 2017 and February 2018 (Figure 3.6).

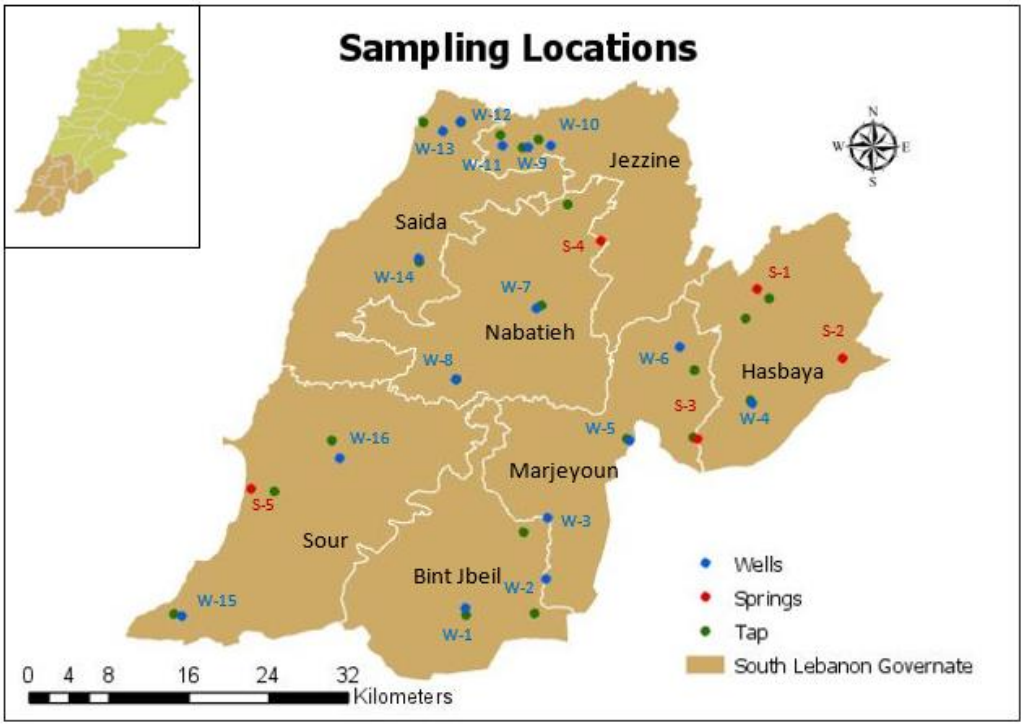


Figure 3.5 Sampling locations

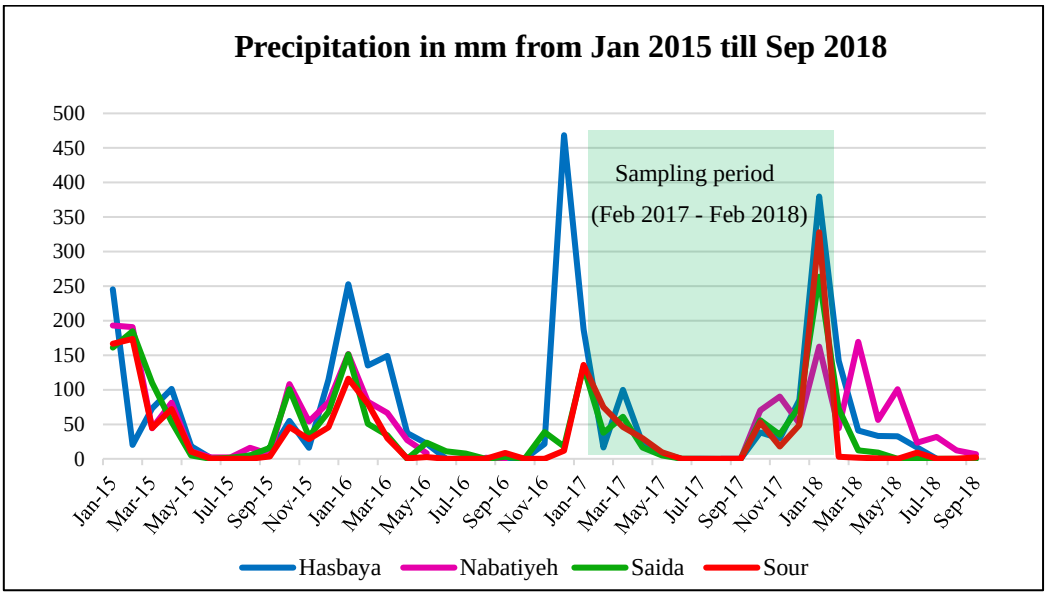


Figure 3.6 Precipitation pattern and sampling period

Table 3.2 Sampling locations

Governorate	District	Location Type	ID	Latitude	Longitude	Location
Al-Nabatieh	Bint Jeil	Well	W-1	33.121671°	35.410228°	Ain Ebl
		Well	W-2	33.147876°	35.482253°	Aitaroun - Al Nikaa
		Well	W-3	33.203112°	35.483715°	Wadi Slouki
		Tap	TW-1	33.105871°	35.410241°	Ain Ebl
		Tap	TW-2	33.116381°	35.472205°	Aitaroun
		Tap	TW-3	33.189461°	35.462301°	Shakra
	Hasbaya	Spring	S-1	33.408681°	35.672479°	Al Hasbani Spring
		Spring	S-2	33.345685°	35.749573°	Al Mghar - Shebaa
		Well	W-4	33.306193°	35.667943°	Halta
		Tap	TS-1	33.400189°	35.683524°	Al Hasbani Spring
		Tap	TS-2	33.382221°	35.662309°	Abou Kamha
		Tap	TW-4	33.308457°	35.666658°	Halta
	Marjeyoun	Spring	S-3	33.273491°	35.618981°	Al Wazzani Spring
		Well	W-5	33.272074°	35.558361°	Kfarkila Well
		Well	W-6	33.356518°	35.602999°	Khiam - Marj Al Khokh
		Tap	TS-3	33.274921°	35.614955°	Al Wazzani Spring
		Tap	TW-5	33.273643°	35.555284°	Kfarkila
		Tap	TW-6	33.335619°	35.616283°	Khiam
	Al-Nabatieh	Spring	S-4	33.452649°	35.532514°	Al Tasseh Spring
		Well	W-7	33.390536°	35.473815°	Fakhreddine Wells
		Well	W-8	33.326911°	35.401301°	Kfarsir
		Tap	TS-4	33.484931°	35.501921°	Kfarfila
		Tap	TW-7	33.393939°	35.478318°	Kfarjouz
		Tap	TW-8	33.327324°	35.401777°	Kfarsir
South Lebanon	Jizzine	Well	W-9	33.537030°	35.466211°	Ain Al Mir
		Well	W-10	33.538349°	35.486441°	Kfarfalous
		Well	W-11	33.538500°	35.442508°	Wadi Baanqoudine
		Tap	TW-9	33.536176°	35.461289°	Ain Al Mir
		Tap	TW-10	33.543371°	35.475594°	Kfarfalous
		Tap	TW-11	33.547605°	35.441254°	Wadi Baanqoudine
	Saida	Well	W-12	33.559331°	35.405771°	Haret Saida
		Well	W-13	33.550894°	35.389322°	Saida - Al Fawwar
		Well	W-14	33.436848°	35.367665°	Tefahta
		Tap	TW-12	33.559258°	35.405638°	Haret Saida
		Tap	TW-13	33.558952°	35.371873°	Saida
		Tap	TW-14	33.433312°	35.368191°	Tefahta
	Sour	Spring	S-5	33.228373°	35.216595°	Ras El Ain Spring
		Well	W-15	33.113798°	35.153511°	Naqoura
		Well	W-16	33.256769°	35.296524°	Wadi Jilou
		Tap	TS-5	33.226242°	35.237689°	Deir Quanoun
		Tap	TW-15	33.115664°	35.147111°	Naqoura
		Tap	TW-16	33.272177°	35.289476°	Teir Debba

3.3. Sampling procedures

Polyethylene bottles of 5 L, 2 L and 1 L capacities were used to collect water samples for uranium, gross alpha/gross beta, and radon activity measurements. On site, each bottle was rinsed three times with the source water. Radon escapes from water as it touches the air (Inácio et al. 2017). As such, samples were carefully collected to preserve their in-water radon. Sampling bottles used for radon, were fitted with a hermetic seal to avoid radon gas leakage during transport.

During the process of sampling from springs, the bottles were immersed below the water surface to avoid any contact with air during sampling. As for those collected from wells and taps, fresh water samples were taken after allowing the withdrawal pump (in case of wells) and the faucet water (in case of tap) to run for at least 5 minutes before taking the sample. In general, during sample collection, a plastic tube was properly fitted to the outlet faucet and extended into the collection bottles. The flow was adjusted to allow steady water stream and avoid turbulence and formation of bubbles or empty volumes in the tube or the bottles. The water was allowed to overflow the bottles enough time before gentle extraction of the tube from the collection bottle and tight sealing of the cap to eliminate voids inside the bottle. Containers were labeled, and collection time and coordinates were recorded. Measurement of the water temperature was not performed on the field, however, some wells were obvious to have a warmer temperature than the others.

3.4. Water properties

Leaching of radioactive materials form the adjacent aquifer rocks into the water bodies is dependent on the physiochemical properties of the water. The acidity,

conductivity, temperature and TDS are prone to change during the wet season (Rodríguez-Rodríguez et al. 2018). Thus, seasonal variations in the radionuclides contents of the water might be noted. As such, in addition to radionuclides analysis, pH, TDS and conductivity were measured, at the laboratory, during both seasons as they are major factors affecting the solubility of radioactive materials in water. In fact, pH, TDS and conductivity were measured for each of the collected samples. TDS measurement was intended to evaluate the dissolved particles in water, as its presence may be indicative for the presence of suspended radionuclides (NRC 1977) and affects the counting efficiency of the low penetrating alpha particles.
Tests for the three parameters were conducted in accordance with APHA standard methods (APHA 2012). The Cyberscan pH 11 and the Cyberscan Con 11 meters supplied by Eutech instruments were used for pH, conductivity and TDS measurement, respectively.

3.5. Analytical Methods for Radioactivity

Samples' preparations for gross alpha and gross beta activity concentrations measurement and determination of the physiochemical properties of the samples were performed at the Environmental Engineering Research Center (EERC) at the American University of Beirut (AUB).

Whereas radioactivity measurements (gross alpha and gross beta, uranium and radon concentrations) were performed in the LAEC's facilities. The testing methodologies followed the adopted procedures in LAEC for each of the intended radioactivity test. The accuracy of the adopted procedures is assured by several successful participations in international proficiency tests for environmental

radioactivity measurement. These tests are usually organized for interlaboratory radioanalytical methods comparison. In fact, the LAEC facilities have been used to study environmental radioactivity in various matrices as well, not only in drinking water (Aoun et al. 2015; El Samad et al. 2016; El Samad et al. 2017; El Samad et al. 2013; El Samad et al. 2007).

3.5.1. Gross Alpha and gross Beta

Gross alpha and gross beta activities were determined at the LAEC facilities using the low background Tricarb 3180 TR/SL, supplied by Perkin Elmer. This LSC is equipped with pulse shape analyzer (PSA) and optimized for reading gross alpha and gross beta activities. Pure alpha emitter, ^{214}Am, and pure beta emitter, ^{90}Sr/^{90}Y standards were used for the optimization of the PSA and determination of the detector efficiency (Todorović et al. 2012).

Liquid scintillation method for gross alpha, and gross beta activity measurement has been applied in many research studies (Çakal et al. 2015; Hamzah et al. 2011; Happel et al. 2004). This technique provides information about radioactivity concentration from various radionuclides excluding the volatile radionuclides like ^{3}H, ^{14}C and radon as evaporation eliminates the dissolved radon in the samples (Sanchez-Cabeza and Pujol 1995).

Preparation of the water samples for simultaneous gross alpha and gross beta activities counting using the liquid scintillation technique involved the reduction of the test sample by evaporation, acidity adjustment, transfer to a scintillation vial and then mixing with liquid scintillation cocktail (Zapata-Garcia et al. 2009). Samples preparation was performed in the EERC at AUB and the liquid scintillation

counting was performed at LAEC facilities. Preparation for gross alpha and gross beta activity measurement was performed by withdrawing 200 ml of water from the 2 L sampling bottle, adjusting its pH to 2.5 using hydrochloric acid (5 M HCl) and then slowly evaporating the sample (at 80°C) till the volume is reduced to 20 ml. This was followed by transferring 8 ml of the solution obtained, having a pH of 1.5, to a scintillation vial, adding 12 ml of OptiPhase HiSafe 3 scintillation cocktail, gently shaking the vial to homogenize the content then performing the counting on the liquid scintillating counter. HiSafe 3 is a diisopropylnaphtalene (DIPN) based cocktail with emulsifier and it provides excellent α/β discrimination (Edler 2006).

A blank was prepared for each set of samples to be used as indicative for the background. The blank was prepared by adding and mixing 8 ml of 1.5 M HCl with 12 ml of cocktail. The minimum detectable activity (MDA) of the system is 1 $mBqL^{-1}$ for alpha and 5 $mBqL^{-1}$ for beta activities for a 300 minutes count time per sample.

The gross alpha and gross beta activities in water sample were calculated from the measured activity by the LSC using the following relationship:

$$A_{(\alpha)} = \frac{(cpm_{\alpha} - cpm_{\beta kg}) * V_2 * 1000}{60 * V_1 * V_3} \quad (3.1)$$

$$A_{(\beta)} = \frac{(cpm_{\beta} - cpm_{Bkg}) * V_2 * 1000}{60 * V_1 * V_3} \quad (3.2)$$

Where: A_{α} is gross alpha activity concentration in BqL^{-1}

A_{β} is the gross beta activity concentration in BqL^{-1}

cpm_{α} is gross alpha activity in the concentrated volume in counts per min.

cpm_{β} is gross beta activity in the concentrated volume in counts per minute

cpm_{Bkg} is the activity of the blank vial in counts per minute

V_1 is the initial volume in liters

V_2 is the volume after evaporation in liters

V_3 is the transferred volume in liters

3.5.2. Radon measurement

Radon measurement was also performed using the LSC at LAEC's facilities. Samples' preparation and counting was performed within 24 to 36 hours after samples collection to avoid decay of radon gas due to its relatively short half-life. OptiPhase HiSafe3 was used for samples preparation in this study. It has been reported as a cocktail option for radon measurement by LSC (Aleissa et al. 2012), however, a water immiscible cocktail could have provided better phase separation results (BSI 2015).

Samples were prepared by placing 12 ml of OptiPhase HiSafe3 cocktail in a Teflon coated polyethylene scintillation vial that prevents radon leakage outside the vial. This was followed by pipetting 8 ml from the collection bottle at around 4 cm below the water surface to avoid radon escaping, and injecting it slowly under the surface of the scintillation cocktail in the vial to avoid turbulence which might lead to radon loss. The scintillation vial was closed with a cap lined with aluminum that also helps preventing radon escaping from the scintillation vial during counting. A blank was prepared by adding only the 12 ml of cocktail to the scintillation vial. The samples were stored in darkness to eliminate luminescence and were read after 3

hours to allow equilibrium between radon and its short lived progeny. Each vial was counted for 300 minutes.

The radon activity concentration was determined using the following formula (Aleissa et al. 2012):

$$C = \frac{(R - R^0)\, Exp^{(\lambda \Delta t)}}{V\, \varepsilon} \quad (3.3)$$

Where:

C is the radon concentration in BqL^{-1}

R is the total count rate in the water sample measured in counts per second

R^0 is the count rate of the blank vial in counts per second

λ is the decay constant of ^{222}Rn in day^{-1}

Δt is the time interval between the collection of the sample and the counting time

V is the sample volume in liters

ε is the efficiency of the detector

3.5.3. Uranium

Uranium measurement was performed at the Environmental Radiation Control Division laboratories of the LAEC facilities using the routinely adopted method for uranium analysis: Samples preparation was done by coprecipitation, chemical separation by ion exchange, and sampling mounting by electrodeposition, then the thin layer was measured by alpha spectroscopy. Figure 3.7 summarizes the steps of uranium samples preparation for alpha spectroscopy measurement.

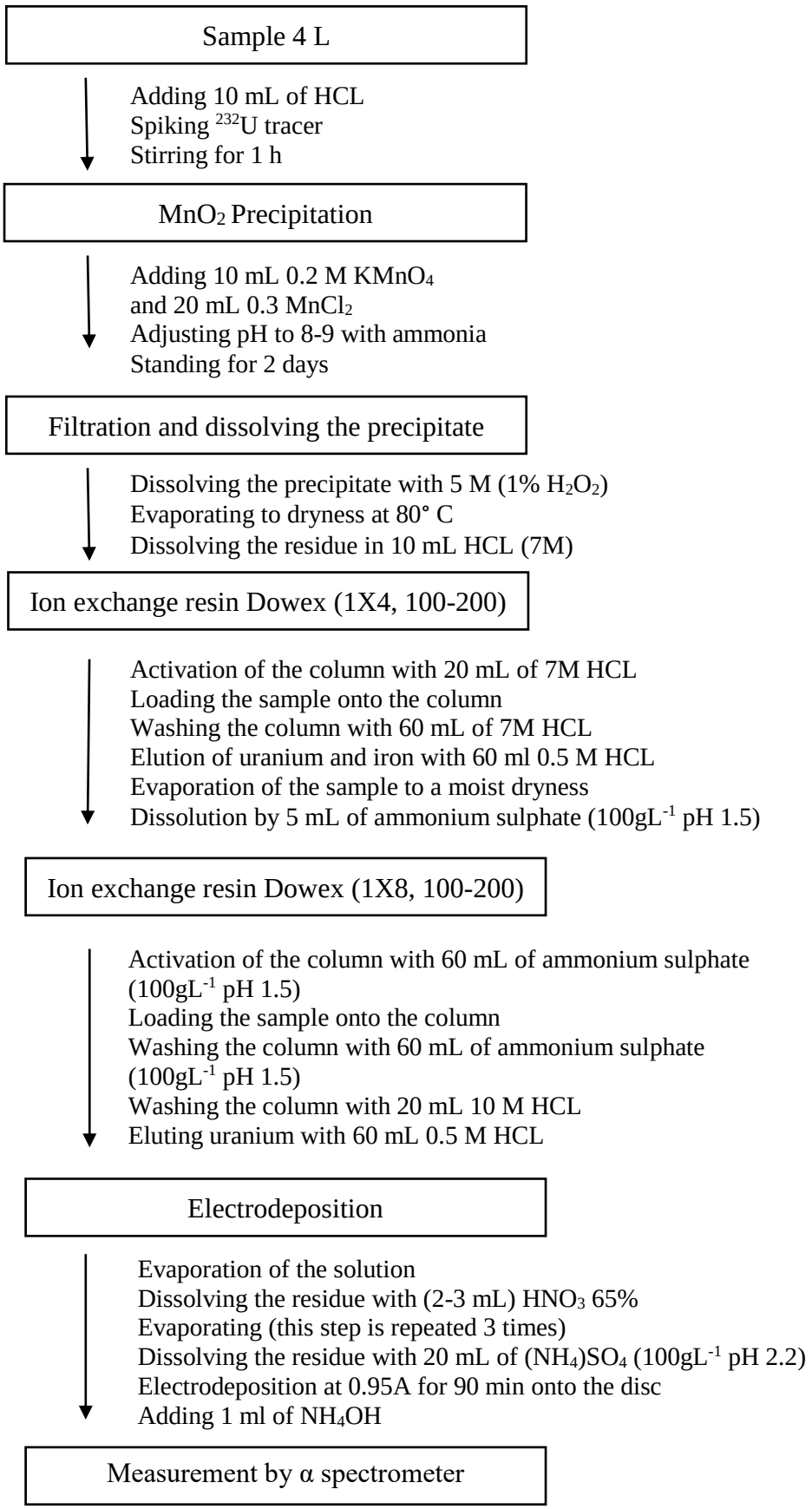


Figure 3.7 Steps of uranium samples preparation for spectroscopy measurement

3.5.3.1.Sample preparation

A 4 L water sample was first acidified in a laboratory grade glass beaker by adding 10 ml of concentrated hydrochloric acid (HCL). Then the sample was spiked with 0.3 ml of the artificial ^{232}U radiotracer to determine the chemical recovery at the end of the procedure. The current activity of the ^{232}U radiotracer used, $T_{1/2}$ = 70.6 years, (LNHB), was calculated from its original activity using the equation 3.4 (Lawson 1999):

$$A_t = A_0 e^{-\lambda \Delta t} = A_0 e^{-[\frac{ln2}{T_{1/2}}(t-t_0)]} \quad (3.4)$$

Where:

A_t is the activity remaining after the elapse time (Δt, $t-t_0$)

A_0 is the initial activity

λ is the decay constant, expressing the decay rate as a factor of the half-life

Δt, $t-t_0$ is the elapsed time

$T_{1/2}$ is the physical half-life of the radionuclide

After spiking with the ^{232}U, the solution was oxidized with 10 ml of 0.2 M potassium permanganate (0.2 M $KMnO_4$). Homogenization of the produced pinkish solution was allowed for 1 hour by spinning on a magnetic stirrer. These steps are made to allow coprecipitation of radionuclides with MnO_2 (Todorović et al. 2012).

One hour after stirring, 20 ml of 0.3 M manganese chloride (0.3 M $MnCl_2$) were added in excess to the mixture and the pH was adjusted to become to between 8 and 9 with Ammonium Hydroxide (NH_4OH). Upon this modification, the mixture turns into brown and a brown precipitation starts to get formed. The mixture was left for two days to allow for total precipitation.

After 2 days, the precipitate was filtered by Buchner filtration and then dissolved with a solution of 5M HCL and hydrogen peroxide (H_2O_2) at 1% concentration. The resulting solution was gently evaporated till dryness (at 80°C).

3.5.3.2.Chemical separation

The dried sample was dissolved with 10 ml of 7M HCL and transported to the first anion exchange column: a 30 cm × 1 cm ion exchange column loaded with 10 cm of ion exchange resin. The used resin is a strong base anion resin, chloride form, containing 4% divinylbenzene (DVB) with particle size of 100 mesh to 200 mesh, DOWEX (1X4, 100-200) and activated with 20 ml of 7M HCL. After allowing the sample to pass through the column, the latter was washed with 60 ml of 7M HCL to retain the uranium and iron complex on the column and get other radionuclides like thorium and radium removed from the sample. After that the column was eluted with 60 ml of 0.5M HCL.

The eluted sample containing uranium and iron complex was evaporated to moist dryness and then dissolved with 5 ml of ammonium sulphate ($(NH_4)_2SO_4$) ($100gL^{-1}$, pH 1.5). The sample was then transported to the second anion exchange column: a 30 cm × 1 cm ion exchange column loaded with 10 cm of a strong base anion resin, chloride form, containing 8% divinylbenzene (DVB) with particle size of 100 mesh to 200 mesh, DOWEX (1X8, 100-200) and activated with 60 ml of $(NH_4)_2SO_4$ ($100gL^{-1}$, pH 1.5). The goal of this second separation is to elute the uranium alone. After loading the sample, the column was washed with two solutions: 60 ml of $(NH_4)_2SO_4$ ($100gL^{-1}$, pH 1.5), then with 20 ml of 10M HCL. At last, the uranium alone was eluted from the second column with 60 ml of 0.5M HCL.

3.5.3.3.Electrodeposition

After eluting the solution that contains the separated uranium, the obtained sample was subject to further processing to produce a thin layer that can be effectively measured by alpha spectrometry. In our study, this was done by electrodeposition. In electrodeposition, the metal cation, uranium, dissolved in the electrolyte is reduced by applying different voltages between two electrodes. As a result, the actinides in the sample are deposited in hydroxide form (BSI 2014).

The obtained solution which contains the separated uranium was evaporated till dryness and then dissolved with 2 to 3 ml of nitric acid (HNO_3) 65% and then evaporated. This step was repeated 3 times. Then the residue was dissolved with 20 ml of $(NH_4)_2SO_4$ ($100gL^{-1}$, pH 2.2). The solution was then put in the electrodeposition cell which contains an inox disk as a cathode and a platinum rod of 1 mm diameter as the anode. The disk was mounted, for 90 min, on the cathode of the apparatus supplying 0.95A to allow the deposition of uranium on the inox disk as a thin layer. One minute before cutting the circuit, 1 ml of ammonium hydroxide (NH_4OH) was added to the electrodeposition cell. The disk was then washed with distilled water, labeled and measured with alpha spectroscopy.

3.5.3.4.Measurement with alpha spectroscopy

The inox disk, with the thin deposited layer of uranium, was measured with the alpha Canberra spectrometer (Alpha Analyst) equipped with PIPS detector. The alpha spectrometer at LAEC facility is mounted with options that enhance its ability for detecting expected low activities in environmental samples. These options include detectors with small leakage current that reduces the sources of electronic

noise, an active exposure surface area that allows optimum detection efficiency for alpha particles in low activity environmental samples, and also a high energy resolution system allowing the distinction between alpha particles with small energy difference. The LAEC alpha spectrometer is a benchtop type composed of eight independent vacuum chambers allowing simultaneous counting of eight samples. Each chamber has twelve levels adjustable height tray that is meant to change the distance between the sample and the detector. Changing the distance can affect the detection efficiency and also the strength of the generated pulse. The software of the alpha spectrometer analyses the pulses resulting from the interaction of the alpha particles with the PIPS detector located inside the chamber, and generates a spectrum of pulses corresponding to each radionuclide detected in the sample. Figure 3.8 shows an example of the spectrum produced by alpha spectroscopy and it shows the separate peaks of uranium isotopes (Abbasisiar et al. 2004).

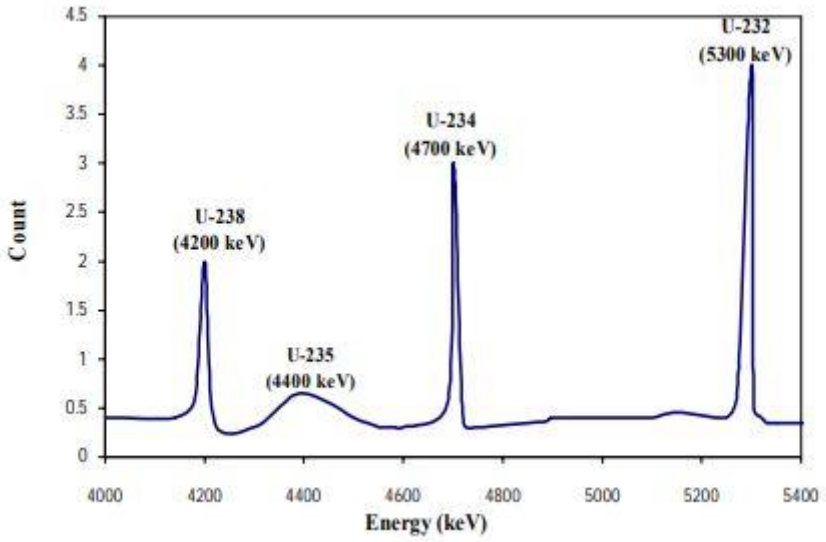


Figure 3.8 Spectrum of uranium isotopes (Abbasisiar et al. 2004)

3.5.3.5. Activity calculation

Activity calculation for uranium isotopes was performed using the following equations:

$$\text{Radiochemical recovery \%} = \frac{N_{Tracer}}{\varepsilon \times t \times A_{tracer}} \times 100 \quad (3.5)$$

$$A_{U\text{-}238} = \frac{N_{U-238} \times A_{tracer}}{V \times N_{Tracer}} \quad (3.6)$$

$$A_{U\text{-}234} = \frac{N_{U-234} \times A_{tracer}}{V \times N_{Tracer}} \quad (3.7)$$

Where: N_{tracer} is the net peak area of the tracer

ε is the detector efficiency

t is the counting time in seconds

A_{tracer} is the activity of the tracer at the counting time

$A_{U\text{-}238}$ is the activity of ^{238}U at counting time in BqL^{-1}

$N_{U\text{-}238}$ is the net peak area of ^{238}U

$A_{U\text{-}234}$ is the activity of ^{234}U at counting time in BqL^{-1}

$N_{U\text{-}234}$ is the net peak area of ^{234}U

3.6. Dose Calculation

Drinking water is always susceptible for containing certain undesired levels of naturally occurring or artificially made radionuclides. As such, assessing the concentrations of radioactive materials is essential for monitoring the drinking water quality. Radioactivity assessment shall be intended for estimating the associated risk

of stochastic effects, cancer risk in particular, with an objective of reducing the probability of radiation induced cancer to an acceptable risk level. Radiation doses were estimated to assess the contribution of the water intake from the sampled locations to public exposure from natural radioactivity and evaluate its potential health hazards. Doses were calculated for the three age groups, infant (1-2 years), child (7-12 years) and adult (> 17 years) based on the yearly water consumptions estimated by UNSCEAR and WHO 150L, 350L and 730L (UNSCEAR 2000; WHO 2017).

The equation used for doses calculation is the following (El-Mageed et al. 2013):

$$DR_w = A_w \times IR_w \times IDF \quad (3.8)$$

Where:

DR_w is the effective dose in $mSvy^{-1}$

A_w is the radon activity in water in BqL^{-1}

IR_w is annual intake of water in L

IDF is the effective dose equivalent conversion factor ($mSv\ Bq^{-1}$)

The effective dose per unit of intake by ingestion of measured natural radionuclides for each of the three age groups are presented in Table 3.3 (UNSCEAR 2000).

Table 3.3 Effective dose per unit intake by ingestion

Radionuclide	Effective dose per unit intake ($nSvBq^{-1}$)		
	Infant	Child	Adult
^{238}U	120	68	45
^{234}U	130	74	49
^{222}Rn	23	5.9	3.5

Another approach for dose calculation from radon ingestion is reported whereby a more conservative estimate of dose coefficient is applied. In this method, annual intakes by infant, child, and adult of 100, 75 and 50 liters respectively, or a weighted estimate of annual water intake of 60 Ly^{-1} is considered (Ravikumar and Somashekar 2013; UNSCEAR 2000). In our study, and to account for worst case scenario, we assumed that all the annual water consumption will be taken from the sampled locations and we considered annual water consumption of the 150 L, 350L for infant and child, and 730L for adult (i.e. 2 liters per day). This was done to stay consistent with WHO guidance levels that were set based on a radioactivity concentration in a daily water consumption of 2 liters that will result in an effective dose not exceeding 0.1 mSv per year (WHO 2017).

3.7. Statistical data analysis and mapping of results

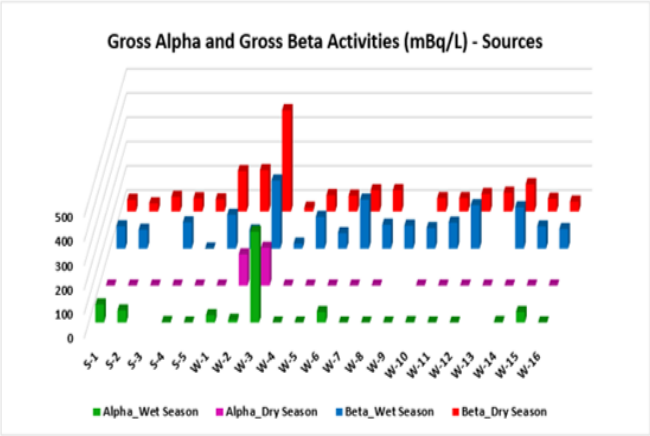
Statistical analyses were performed to study the seasonal variation of radionuclides concentration between the wet and the dry seasons, the variations in activities concentrations between sources and tap water, and also between springs water and wells water. None of the measured radioactivities data (gross alpha, gross beta, ^{238}U, ^{234}U and ^{222}Rn) could fit into a normal distribution. As such, nonparametric tests were used to perform the statistical analyses: Paired, two-tailed Wilcoxon test was used to study the wet-dry and sources-tap variations, whereas, unpaired, two-tailed, Mann-Whitney test was used for spring-well variation. Graph generation and data analyses were performed using the statistical software Prism Graphpad 8. A level of confidence of 95% with a p-value ≤ 0.05 was chosen to define the significance of results. Maps and geoanalysis were generated using the ArcGIS Pro software.

CHAPTER 4

RESULTS AND DISCUSSION

4.1. Results of measured radioactivity

The results of all performed tests are presented in Figure 4.1, Figure 4.2, Figure 4.3, and in Appendix 2 as well. In general, the gross beta activity concentrations are higher than those of the gross alpha activity concentrations. The variation in ^{238}U, ^{234}U and ^{222}Rn from one location to another could be indicative that the water in these locations originates from different basins and that it crosses different geological formations, or it can be linked to differences in the physiochemical properties of the water. It is, however, to be noted that no measurements were conducted to determine the associated beta emitting radionuclides which could include but are not limited to ^{40}K, ^{210}Pb and ^{228}Ra. Further investigations are needed for determining the activity concentrations of individual beta emitting radionuclides for the interest of building knowledge about the individual radionuclides occurrence in the area under investigation not only for regulatory compliance.



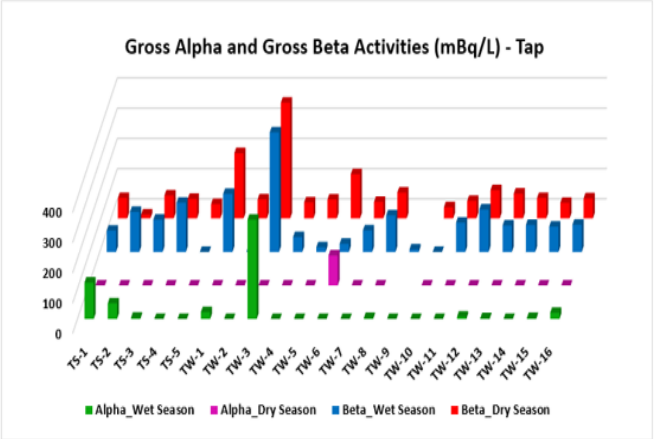
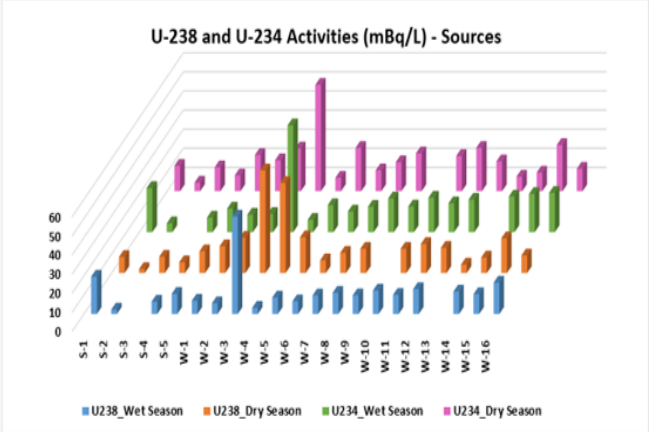


Figure 4.1 Gross α and β Activities - Sources & Tap water in Wet & Dry seasons



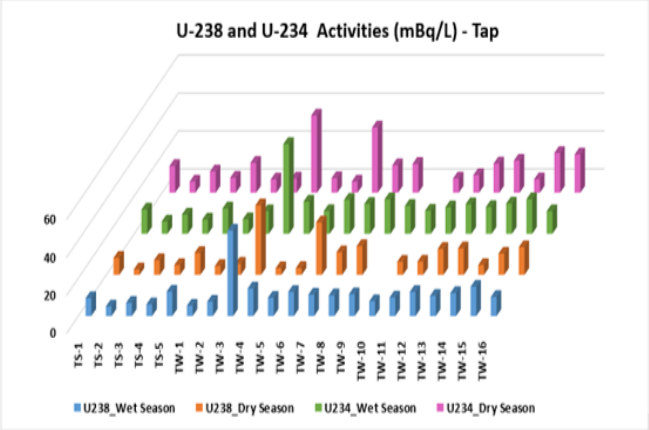
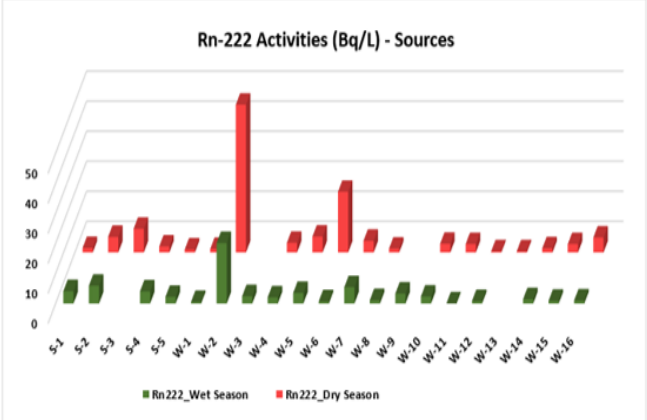


Figure 4.2 ^{238}U and ^{234}U Activities - Sources & Tap water in Wet & Dry seasons



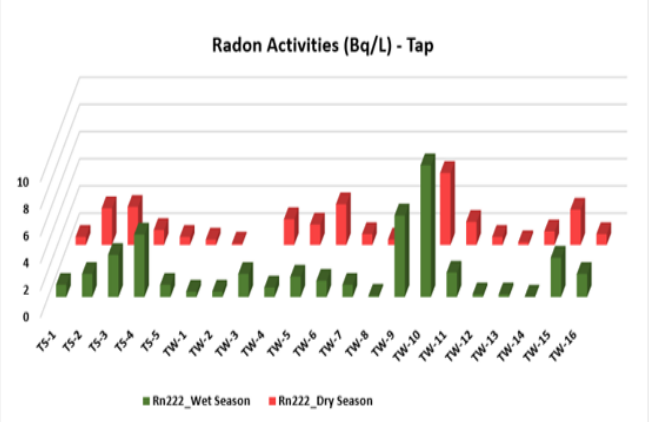


Figure 4.3 ^{222}Rn Activities - Sources & Tap water in Wet & Dry seasons

4.1.1. Gross alpha and gross beta activities

Gross alpha and gross beta activities in the wet and dry seasons are displayed on the geological maps in Figure 4.4. As noted from Figure 4.1 and Figure 4.5, all gross alpha and gross beta activities were found to be below the WHO screening limits of 0.5 BqL^{-1} and gross beta limits of 1 BqL^{-1}.

Gross alpha activities in the measured locations ranged between less than the minimum detectable activity (MDA) of 1 $mBqL^{-1}$ and 374.6±11.6 $mBqL^{-1}$ in the wet season, and between less than MDA and 155.8±13.2$mBqL^{-1}$ in the dry season. In fact, the gross alpha activities of 71.25 % of the sampled locations were below the detection limit of the measuring equipment. The higher levels for gross alpha radioactivity (374.6±11.6 $mBqL^{-1}$ and 155.8±13.2 $mBqL^{-1}$) were recorded in well number 3 (W3, Wadi Slouki, in Bint-Jbeil district) and its corresponding tap water (TW3). This well is located within the Nabatieh/Bint Jbeil Eocene basin located in an area dominated mainly by geological formation of the Eocene period composed mainly of marly, chalky limestone and breccia limestone (MoEW/UNDP 2014). Other locations that exhibited relatively high values of gross alpha activities included W2 (Aitaroun well, 128.6±9.43 $mBqL^{-1}$), TS1 (Tap water of Al Hasbani river, 120.2±9.4 $mBqL^{-1}$), and TW6 (Tap water in Al-Khiam, 99.1±9.2 $mBqL^{-1}$). Well 2 belongs to the same basin as W3, while W6 lies within the Southern Bekaa Eocene basin. However, the geological formation of W2 and W6 are similar to those of W3. On the other hand, S1 falls within a geological area of the cretaceous period (C4) composed of dolomitic and marly limestone and is fed by the Southern Anti-Lebanon Cretaceous basin (MoEW/UNDP 2014). The major naturally occurring

alpha emitting radionuclides are ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, and ^{210}Po (Turhan et al. 2013).

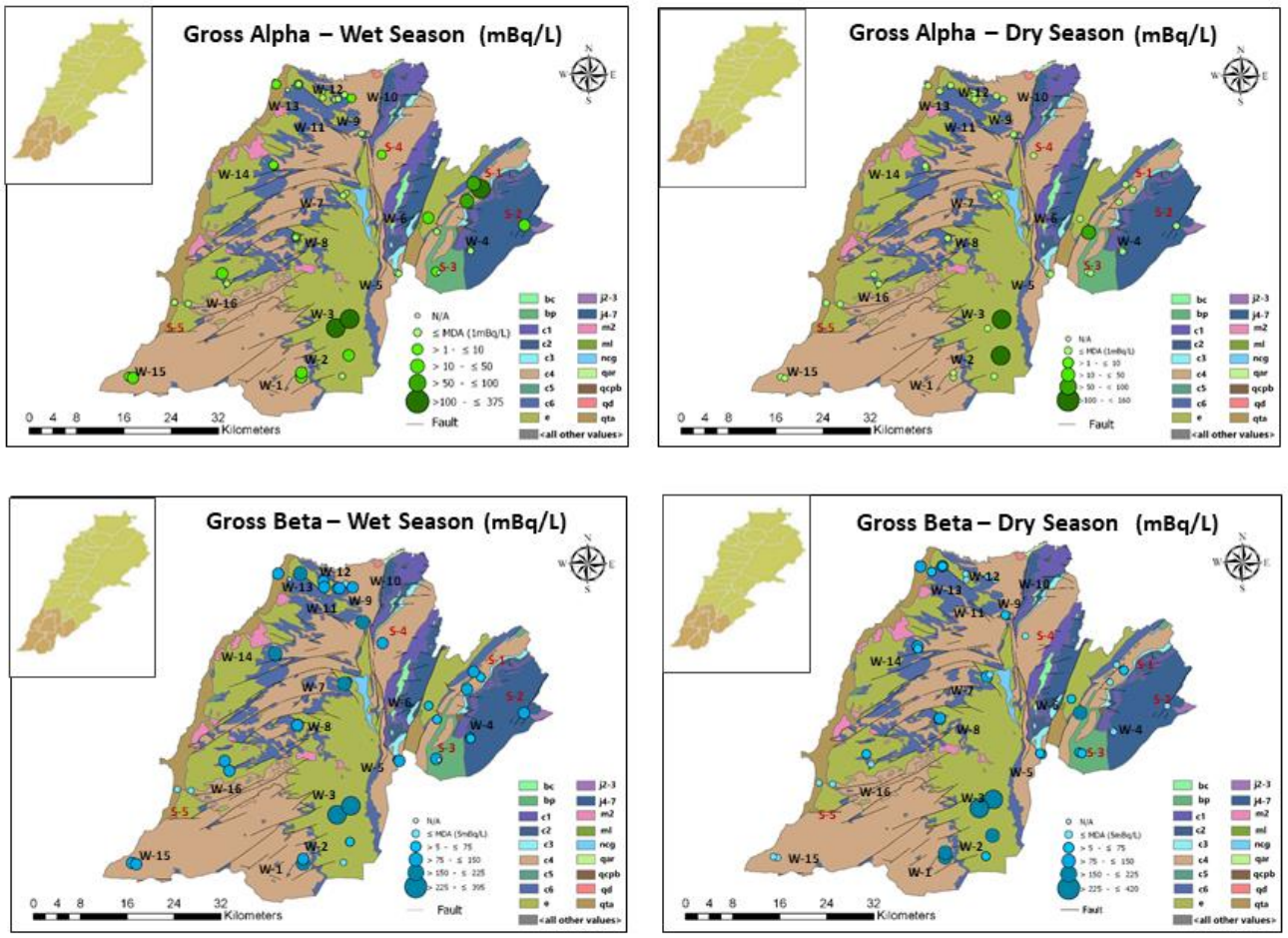


Figure 4.4 Gross alpha and gross beta activities displayed on the geological map

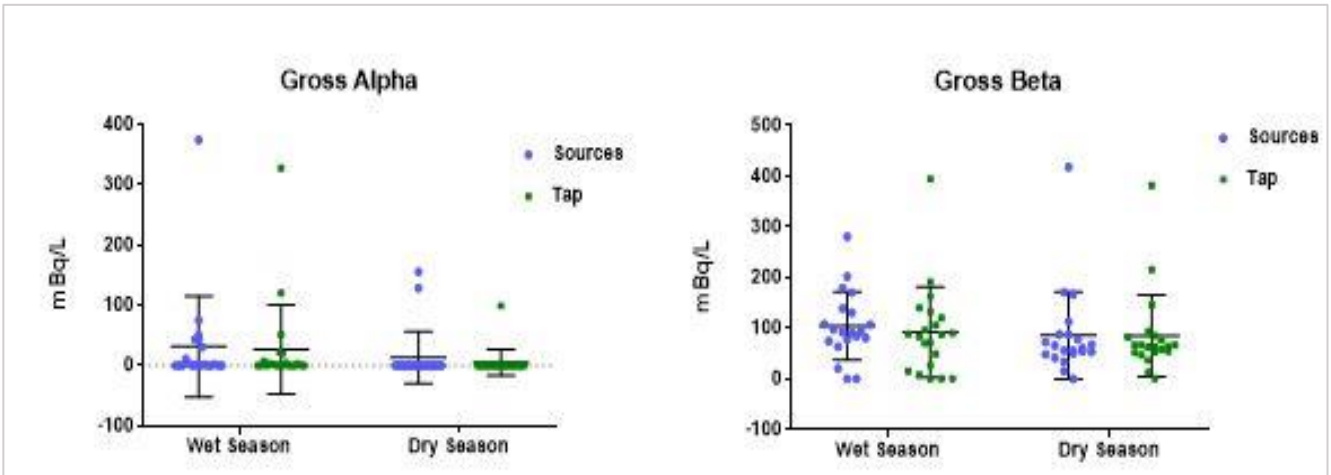


Figure 4.5 Seasonal variations for gross alpha and beta activities in sources and tap water

Gross beta activities, were in general higher than those of gross alpha; Damla et al reported similar findings (Damla et al. 2009). Only 5% of the samples were below the 5 $mBqL^{-1}$ MDA of gross beta particles. Gross beta activities varied between less than its MDA (< 5 $mBqL^{-1}$) and 395±8.3 mBL^{-1} in the wet season, and between 12.8±0.6 $mBqL^{-1}$ and 418±12.1 $mBqL^{-1}$ in the dry season. The highest activities (418±12.1 $mBqL^{-1}$ and 395±8.3 $mBqL^{-1}$) were observed in W3 (Wadi Slouki) during the dry season and in tap water from well 3 (TW3) during the wet season. Relatively high gross beta activities were also observed in TW1 (Ain Ebl, 215.6±7.3 $mBqL^{-1}$), W7 (Fakhreddine, 202.1±10.3 $mBqL^{-1}$), W12 (Haret Saida, 179.2±10.3 $mBqL^{-1}$), and W14 (Tefehta, 170±7.3 $mBqL^{-1}$). These levels of activities may be due to the geological formation or due to the physiochemical properties of the area. Actually, W7 shares the same lithological properties described previously for W3 and lies within the Nabatieh/Bint-Jbeil Eocene basin. Whereas, W1, W12, and W14 are of the cretaceous period; where W12 and W14 are of the Senonian age (C6) characterized by its marly chalks with phosphate and its chalky marly limestone lithology, and W1 originating from the highly karstified and well bedded dolomitic limestone of the Cenomanian (C4). Naturally occurring gross beta emissions are mainly due to ^{210}Pb and ^{228}Ra as well as ^{40}K radionuclides (Görür et al. 2011).

The majority of gross alpha and gross beta concentrations are of low values. In fact 90% of gross alpha radioactivity concentrations in the measured samples fall below the 10% value of 0.5 BqL^{-1}, the WHO guidance level for gross alpha activity concentrations (i.e. < 50 $mBqL^{-1}$), and 72.5% of gross beta radioactivity concentrations are less than the 10% value of 1 BqL^{-1}, the WHO guidance level for gross beta concentrations (i.e. < 100 $mBqL^{-1}$).

Wilcoxon test performed to study the variation in gross alpha and gross beta activities during the wet and the dry seasons resulted in p value of 0.0024 ($p < 0.05$) for gross alpha activities. A result that describes a significant variation in gross alpha activity concentrations between the two seasons. Whereas, such significance was absent in the case of gross beta activities ($p > 0.05$). The similar test conducted to assess concentrations variations between sources of the water supplies and those of tap water did not show any significance in both gross alpha and gross beta activities ($p > 0.05$). This elucidates that the primary applied treatment systems and the distribution networks did not affect the gross alpha and gross beta radioactivity concentrations in water. In addition, insignificant variation was observed while testing gross alpha and gross beta activity concentrations in wells and in springs (Figure 4.6). Mann-Whitney test resulted in a $p > 0.05$, interpreted as insignificant influence of water depth on its radioactivity content. However, this insignificance is expressed with uncertainty due to the small and uneven n value of springs and wells in this study.

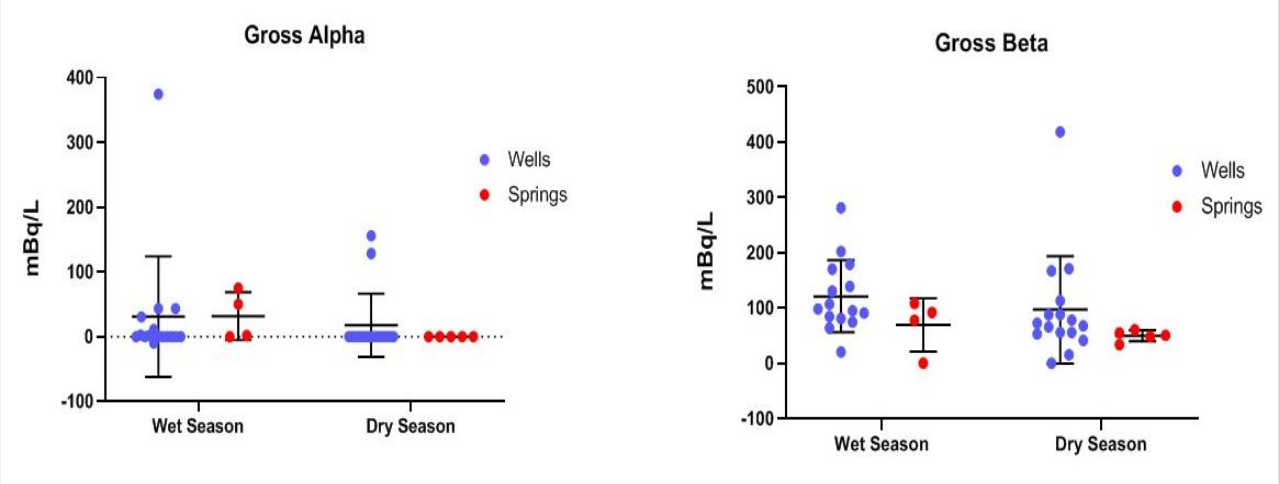


Figure 4.6 Seasonal variations for gross alpha and gross beta activities in wells and in springs

The results of the measured activities were compared to other studies performed worldwide (Table 4.1). Gross alpha and gross beta activity concentrations were comparable to those in Turkey, Iran, Syria, India and Jordan (Abbasi and Mirekhtiary 2017; Al-Shboul et al. 2017; Degerlier and Karahan 2010; Sarvajayakesavalu et al. 2018; Shweikani and Raja 2015). However, large differences were observed in other locations like Nigeria (Seydou and Abdullahi 2016) as depicted from Table 4.1. This variation could be linked to differences in the geological and lithological formation in each region.

Table 4.1 Comparison of gross alpha and gross beta activities in different world locations

Country	Type of water	Gross alpha $mBqL^{-1}$	Gross beta $mBqL^{-1}$	Measurement Technique	Reference
Turkey	Sea, Lake, River, Tap	0.3 - 22.9	24 - 290.7	Gas proportional counter	(Degerlier and Karahan 2010)
Iran	Wells & Springs	12 - 115	23 – 332	Gas proportional counter	(Abbasi and Mirekhtiary 2017)
Syria	Tap water	28 - 340	30 – 550	Liquid scintillation counter	(Shweikani and Raja 2015)
Jordan	Wells & Tap	82 - 484	216 – 984	Liquid scintillation counter	(Al-Shboul et al. 2017)
India	Bore well	0.69 – 15.26	14.6 – 108.5	Liquid scintillation counter	(Sarvajayakesavalu et al. 2018)
Nigeria	Boreholes	1030	18690	Gas proportional counter	(Seydou and Abdullahi 2016)
Lebanon	Wells, Springs, Tap	< MDA - 375	< MDA - 418	Liquid scintillation counter	This study

4.1.2. Uranium

Uranium activity concentrations extended between 2.3 $mBqL^{-1}$ and 51 $mBqL^{-1}$ and between 4.6 $mBqL^{-1}$ and 55.9 $mBqL^{-1}$ during the wet season for ^{238}U and ^{234}U respectively. These concentrations remained almost the same during the dry season where they ranged between 2.1 $mBqL^{-1}$ and 52.7 $mBqL^{-1}$, and 4.5 $mBqL^{-1}$ and 55.7 $mBqL^{-1}$ for ^{238}U and ^{234}U respectively. This was confirmed by the nonparametric tests (Wilcoxon and Mann Whitney) which revealed insignificant variations ($p > 0.05$) in the wet-dry seasons, source-tap water, and also in spring-well water ^{238}U and ^{234}U activity concentrations. Actually, the removal of uranium form water is mostly done by precipitation induced by water additives in treatment plants (Baeza et al. 2017). Such technologies are not practiced in water treatment in the study area, the factor that explains the insignificant variation in uranium concentration between sources and tap water.

Figure 4.7 displays the ^{238}U and ^{234}U activities plotted on the geological map, Figure 4.8 shows the seasonal variations of their concentrations and Figure 4.9 depicts the seasonal variations in wells and in springs.

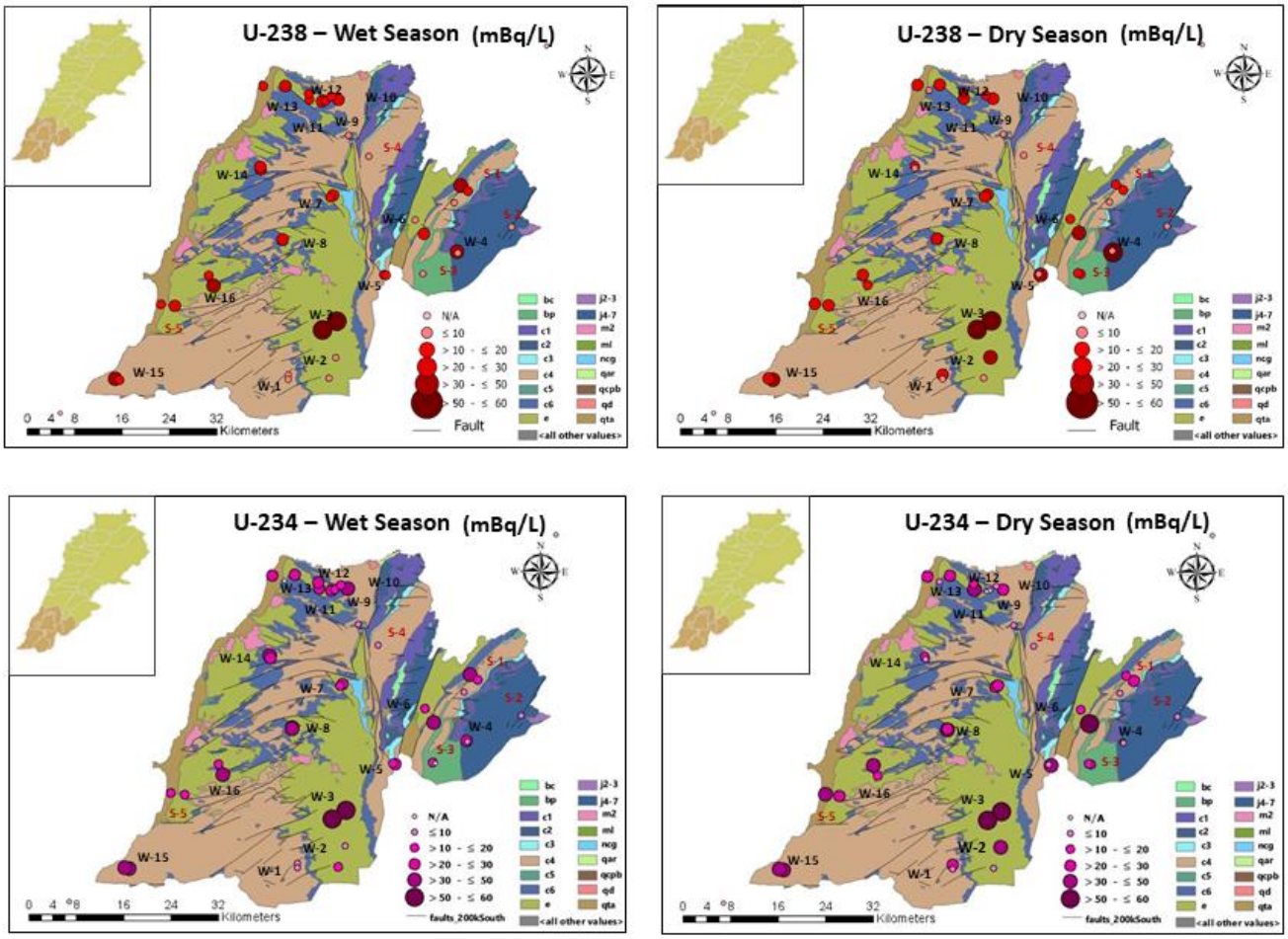


Figure 4.7 ^{238}U and ^{234}U activities displayed on the geological map

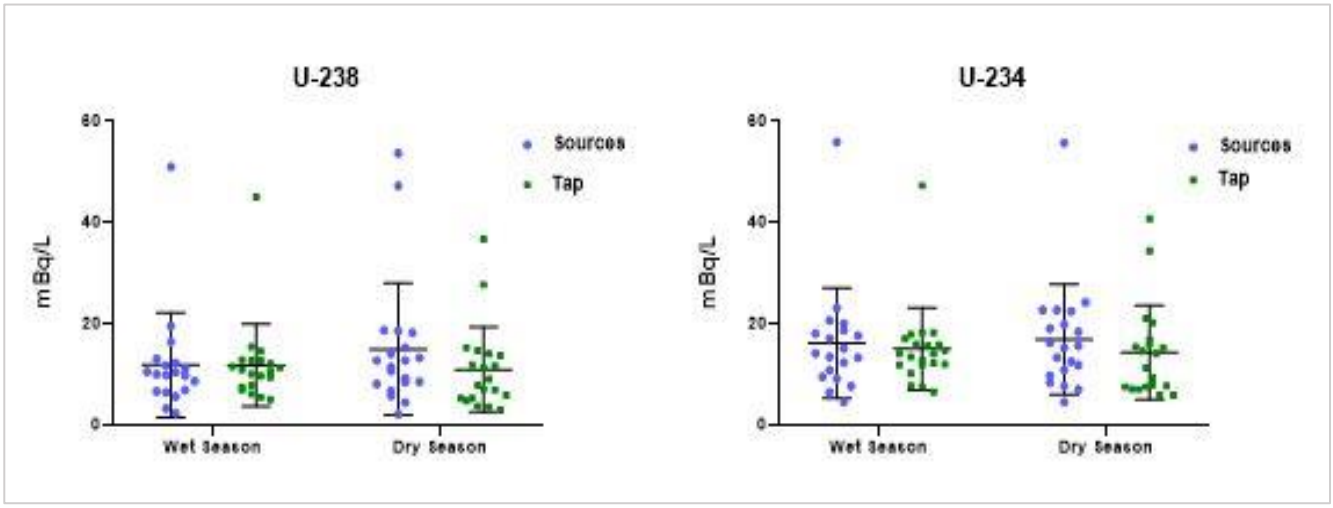


Figure 4.8 Seasonal variations of ^{238}U and ^{234}U activities in sources and tap water

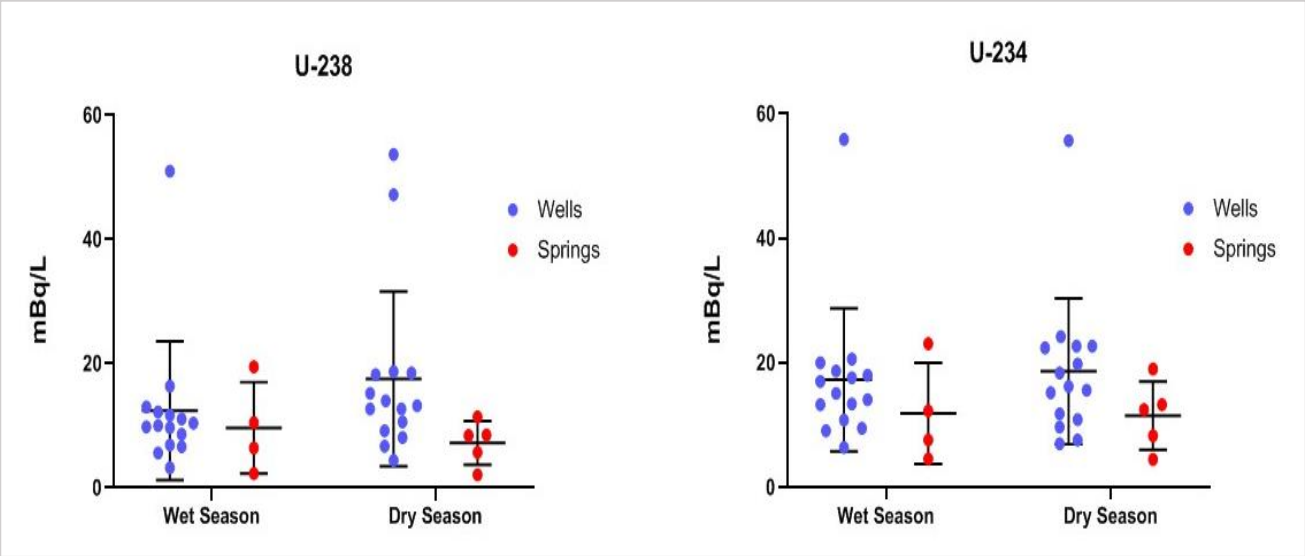


Figure 4.9 Seasonal variations of ^{238}U and ^{234}U activities in springs and wells

The measured results of all samples were below the WHO guidance level of 10 BqL^{-1} and 1 BqL^{-1} for ^{238}U and ^{234}U respectively. Like in the case of gross alpha and gross beta, the majority of ^{238}U and ^{234}U activities were of very low concentrations where all measured activities were less than 1% of the WHO guidance level for ^{238}U (<100 $mBqL^{-1}$) and less than 6% of that value for ^{234}U (< 60 $mBqL^{-1}$). Samples with relatively high ^{238}U concentrations were noted in W3 and TW3 (Wadi Slouki), W4 (Halta) and TW6 (Al-Khiam) with concentrations reaching 53.7±2.1 $mBqL^{-1}$, 47.2±2.1 $mBqL^{-1}$ and 27.7±0.9 $mBqL^{-1}$ respectively. Whereas, ^{234}U high activities were observed in W3 (Wadi Slouki) and TW6 (Al-Khiam) as well as in W15 (Al Naqoura) and TS1 (Ain Ebl) with measured ^{234}U concentrations reaching 55.9±2.3 $mBqL^{-1}$, 34.4±1.1 $mBqL^{-1}$, 24.2±2.5 $mBqL^{-1}$ and 23.1±1.2 $mBqL^{-1}$ respectively. Well number 15 belongs to the cretaceous basin and has the same geological formation as that of W12 and W14. Whereas, W4 lies within the Jurassic basin that belongs to the upper Jurassic formation rich in dolomite and dolomitic

limestone. Most likely, W3 is associated with deep seated hydrothermal activity that is allowing uranium to be deposited on the karstified limestone formation (Bell 1963). The high uranium activities in this well can be due to migration of deposited uranium into the adjacent highly recharged aquifer. On the other hand, leaching of phosphate fertilizers from soil to groundwater aquifers has been reported in the literature (Smidt 2011). This can explain the relatively high uranium concentrations mainly in the agricultural areas of Al-Khiam plane.

The calculated value of $^{234}U/^{238}U$ activity ratio fluctuates between 1.04 and 2 in the wet season and between 2.2 and 0.14 in the dry season. These ratio values lie within the average values of the $^{234}U/^{238}U$ activity ratio which is reported to range between 0.51 and 9.02 in natural groundwater (Boryło and Skwarzec 2014). The variation ratio of $^{234}U/^{238}U$ in the two seasons constantly varied around unity which suggests that the $^{234}U/^{238}U$ was not influenced with time or with change in the water table during the aquifers filling periods (Lowson and McIntyre 2013). All samples demonstrated ^{234}U activity concentrations higher than that of ^{238}U. This might be an indication that control of dissolved uranium isotopes in waters were affected by the dissolution and precipitation especially that the latter can reduce the ^{238}U in the water but has less influence on $^{234}U/^{238}U$ ratios (Galhardi and Bonotto 2017). This was not the case in W4 where $^{234}U/^{238}U$ activity ratio varied between 2 in the wet season and 0.14 in the dry season. This might be due to factors that induced leaching and transport of ^{234}U from adjacent rocks to the groundwater aquifer during the wet season (Galhardi and Bonotto 2017).

Comparison with other studies performed worldwide revealed that there is a wide spectrum of uranium activities concentrations in the areas under study

(Table 4.2). These differences may be due to the geological formation and to the physiochemical properties of the underlying lithologic composition that influence uranium solubility, or may be due to anthropogenic activities.

Table 4.2 Uranium activities in water resources in different world locations

Country	Type of water	^{238}U ($mBqL^{-1}$)	^{234}U ($mBqL^{-1}$)	Measurement Technique	Reference
Greece	Groundwater	43.9 – 117	24.6 – 179	Alpha spectrometry	(Noli et al. 2016)
Jordan	Tap water	15.4 – 33.3	16.6 – 119.1	Alpha spectrometry	(Al-Amir et al. 2012)
Saudi Arabia	Wells	140 – 390	140 - 320	Alpha spectrometry	(Alkhomashi et al. 2016)
Slovania	Wells & Tap	0.17 – 372	0.22 - 362	Alpha spectrometry	(Benedik et al. 2015)
Italy	Well, springs, tap, bottles	0.206 – 103	0.249 - 135	Alpha spectrometry	(Jia et al. 2009)
Lebanon	Wells, springs, tap	2.1 – 53.7	4.5 – 559	Alpha spectrometry	Current study

4.1.3. Radon

Figure 4.10 displays the radon concentration on the geological map and Figure 4.11 present sources-tap and springs-wells seasonal variations of the radon activities. Radon stand out as the highest values when compared to the other measured radiation sources. The measured values were found to be in the order of several BqL^{-1}, while gross alpha, gross beta and uranium activities were all less than 1 BqL^{-1}. All sampled locations demonstrated radon concentrations below the EU set recommendation level of 100 BqL^{-1}. Actually, radon activities range between the detection limit (< 0.03 BqL^{-1}) and a maximum of 49.2 BqL^{-1}, recorded at the well at

location W2 (Aitaoun in Bint Jbeil district). The high radon concentration as compared to other radionuclides can be elucidated by tracing back the origin of dissolved radon in water. In fact, the dissolved radon is not only due to the recoil and leaching of inert radon gas from the rocks and soil grains into the adjacent water body but also could originate from the decay of dissolved ^{226}Ra in water, which is one of the decay daughters of ^{238}U series. That, added to the occurrence of higher disintegration events due to the relatively short half-life of radon and its progenies, explain the higher concentrations of dissolved radon in water as compared to dissolved ^{226}Ra and eventually its parent, uranium (Chales J. Passo 1993).

The minimum activities of measured radon approached the minimum detectable activity of the equipment in both the dry and wet seasons, and the majority of samples are of low levels of radon concentrations. For instance, about 96% of the sampled locations are below 20% of the 100 $mBqL^{-1}$ EU recommended radon activity concentrations in water, i.e. less than 20 $mBqL^{-1}$.

Nonparametric statistical analysis demonstrated the absence of significant variations between the wet and dry seasons and also between sources and wells ($p > 0.05$). The dearth of seasonal variation might be attributed to the daily precipitation pattern which showed a limited number of rainy days as compared to other wet seasons. In contrast, Wilcoxon test performed to assess the variation in radon concentrations in sources versus tap water demonstrated significant variations ($p = 0.0019$). This may be due to the escape of radon gas during treatment and transport, and also can be due to radon decay during storage (Shweikani and Raja 2015).

Comparison of the results of this study with other studies in the world performed for radon concentrations in water is presented in Table 4.3 below. The

overall values of measured radon concentrations in water in this study are somehow comparable to the one performed by Abdallah et al in having most of the locations at a very low concentrations (Abdallah et al. 2007). However, areas with elevated concentrations are not the same. For instance samples taken from Ain Ebl, which was the highest in Abdallah et al (49.6 BqL^{-1}), is of negligible values in this study (0.5 $mBqL^{-1}$ and 1.2 $mBqL^{-1}$ during wet and dry seasons respectively). In contrast, relatively elevated radon concentrations were spotted in Al Khiam (20.2 $mBqL^{-1}$) but the reported level in Al Abdallah et al were higher than ours (31.6 $mBqL^{-1}$). Aitaroun, the highest radon concentration in this research (49.2 $mBqL^{-1}$), was not covered previously. The reported radon samples are close to those reported in in Syria, 7.5 $mBqL^{-1}$ to 28.4 $mBqL^{-1}$ (Shweikani and Raja 2015), but the concentrations reported in Al Jawa in Saudi Arabia, 1.45 $mBqL^{-1}$ to 9.15 $mBqL^{-1}$ were lower than those in this study (Althoyaib and El-Taher 2014). On the other hand, radon concentrations were higher in Turkey, 1.85 $mBqL^{-1}$ to 99.27 $mBqL^{-1}$ (Erdogan et al. 2017) and much more elevated at Al Khartoum in Sudan 1.58 $mBqL^{-1}$ to 345.1 $mBqL^{-1}$ (Idriss et al. 2011).

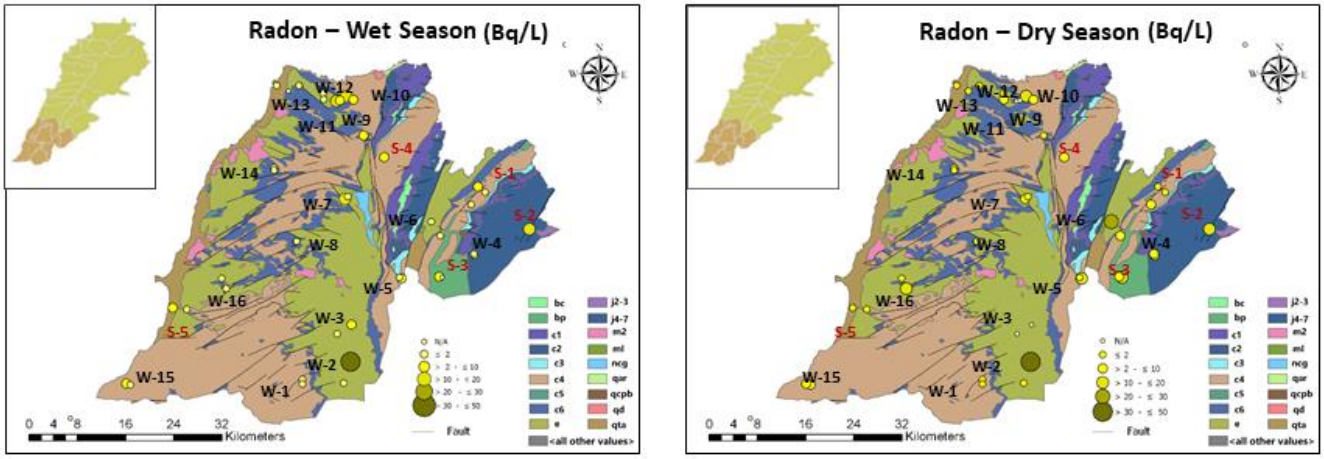


Figure 4.10 Radon activities displayed on the geological map

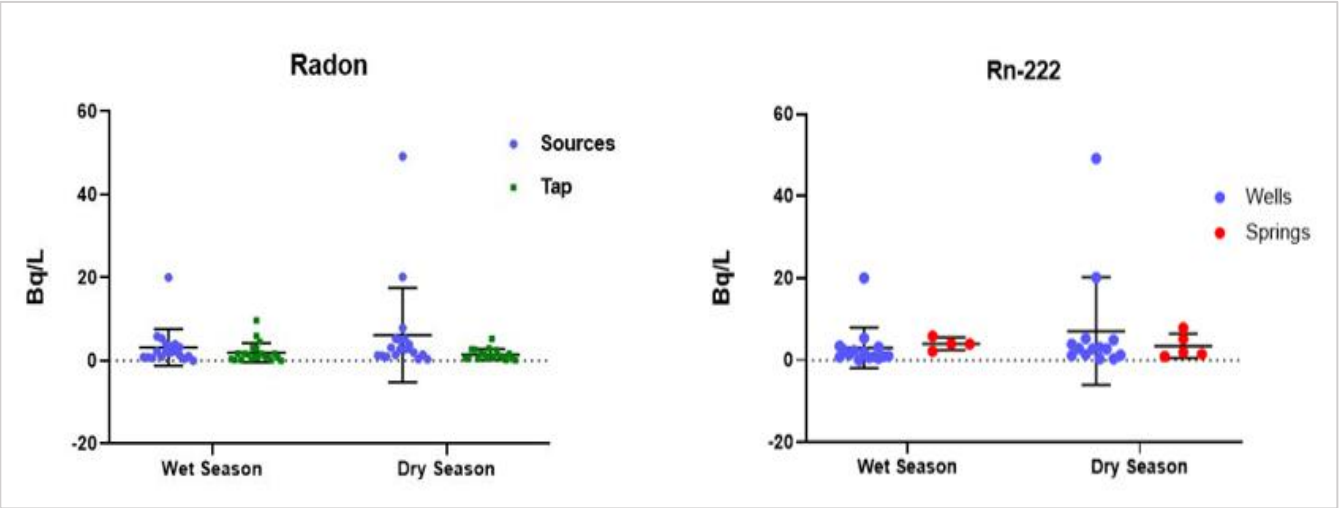


Figure 4.11 Radon activities in wet and dry seasons

Table 4.3 Radon activities in water resources in different world locations

Country	Type of water	^{222}Rn (BqL^{-1})	Measurement Technique	Reference
Syria	Wells Tap	7.5 - 28.4 2.8 – 15.3	Emanometry	(Shweikani and Raja 2015)
Turkey	Wells & Springs	1.85 – 99.27	Emanometry	(Erdogan et al. 2017)
Al Jawa, SA	Wells	1.45 – 9.15	Emanometry	(Althoyaib and El-Taher 2014)
Khartoum	Wells	1.58 - 345.1	Gamma spectrometry	(Idriss et al. 2011)
Lebanon	Well, springs	0.91 – 49.6	Electret ion chamber	(Abdallah et al. 2007)
Lebanon	Wells, springs, tap	2.1 – 53.7	Liquid scintillation counting	This study

4.2. Correlation study

Table 4.4 demonstrates the correlation between radionuclides and some of the physicochemical properties of water. Several factors play a role in the distribution of radioactivity in water including the geological and the lithological characteristics of each area. The relationship between each radioactivity component and the measured physiochemical properties revealed that the ^{238}U and ^{234}U were the only pair where a significant correlation exists (r = 0.879). This is true because ^{234}U is a decedent of the ^{238}U radionuclides series. The strong correlation was also observed between the conductivity and TDS (r = 0.967) which is expected because these two parameters are related to each other by definition. On the other hand, weak correlation was noted between ^{238}U and ^{234}U and beta activities, the same was observed between conductivity, TDS and ^{238}U and ^{234}U. Negative weak correlation was also noted between radon and pH, conductivity and TDS.

Table 4.4 Spearman correlation between radioactivities and a number of physicochemical properties of water

Spearman R	Alpha	Beta	U-238	U-234	Radon	pH	Conductivity	TDS
Alpha	1							
Beta	0.506	1						
U-238	0.142	0.220	1					
U-234	0.197	0.284	0.879	1				
Radon	0.043	-0.133	-0.077	-0.087	1			
pH	-0.157	0.105	0.071	0.021	-0.361	1		
Conductivity	0.008	0.199	0.341	0.343	-0.208	-0.221	1	
TDS	0.100	0.242	0.331	0.354	-0.242	-0.223	0.967	1

4.3. Spatial distribution of radio-activities

For better understanding of the distribution of radionuclides in the Southern region of Lebanon, interpolation of the expected activities of the radionuclides in the locations not sampled was done using the Inverse Distance Weighted (IDW) of the GISpro geostatististical analyst tools (Figure 12, Figure 13 and Figure 14). It shows that radionuclides are more prone to be present in the east side of the southern region. This could be linked to the local geology of the region as most of the relatively high concentrations were present around the areas that mainly belong to the eocene period. On the other hand, there is a domination of cretaceous geological formation with similar lithological composition in most of the studied area which are characterized mainly by its limestone, marly, dolomitic components. This might explain the wide spread of low activities since these lithological compositions are generally of low uranium content (Ielsch et al. 2017). Another factor that could affect the special distribution of radioactivity is the structural formation of the studied area located in the vicinity of the major faults of Lebanon. For instance, the consistently relatively high concentration in wells located in Bint-Jbeil district, W3 and W2, might be affected by the Roum fault which could be allowing radon to escape from the adjacent rock formations into the water aquifer. While Hasbaya and Marjeyoun districts are located near the southern edge of the Yammoune major fault and also near the Rashaya and Hasbaya secondary faults. This can also explain the hot spots observed in these areas. Similar results of higher radon concentrations in water bodies near faults lines were previously reported (Abadi et al. 2016; Khan et al. 2009). In fact, the ultimate source of radon in water is the uranium inside the earth core; and as radon emanation through earth crust is related to the porosity, faults in

tectonic plates constitute an easy pathway for radon emanation and eventual dissolving into water.

The ingnificant seasonal variation in spatial distribution can be linked to other factors that might affect the mobility of radioactivity. For instance, radionuclides concentration in water, uranium in particular, are affected by several factors including the chemical properties of water, occurrence of O_2 and complexing agents such as carbonate, partial pressure of CO_2, pH and conductivity (Rishi et al. 2017). Further studies can confirm or negate these hypotheses.

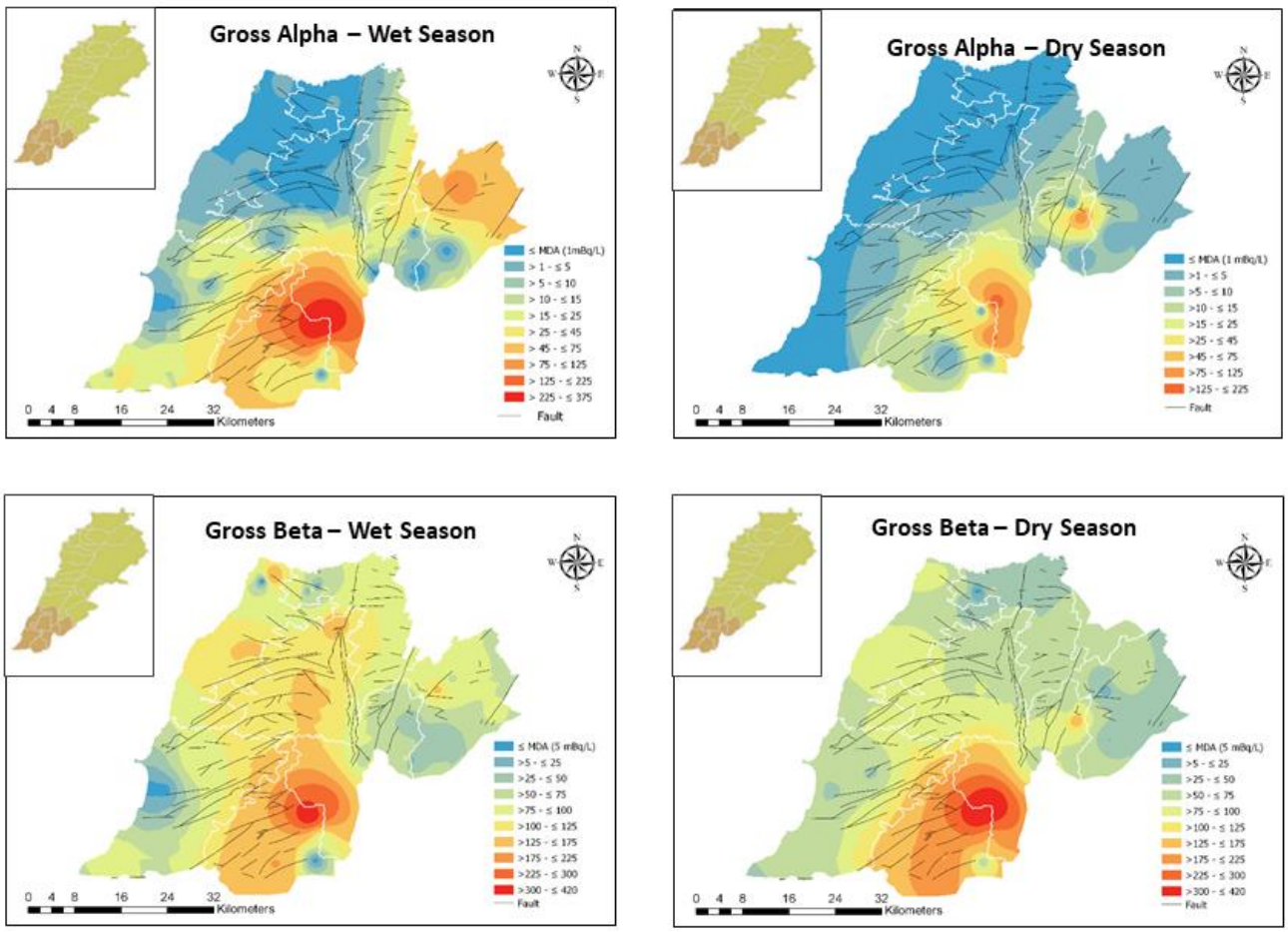


Figure 4.12 Gross alpha and gross beta activities distribution in wet and dry seasons

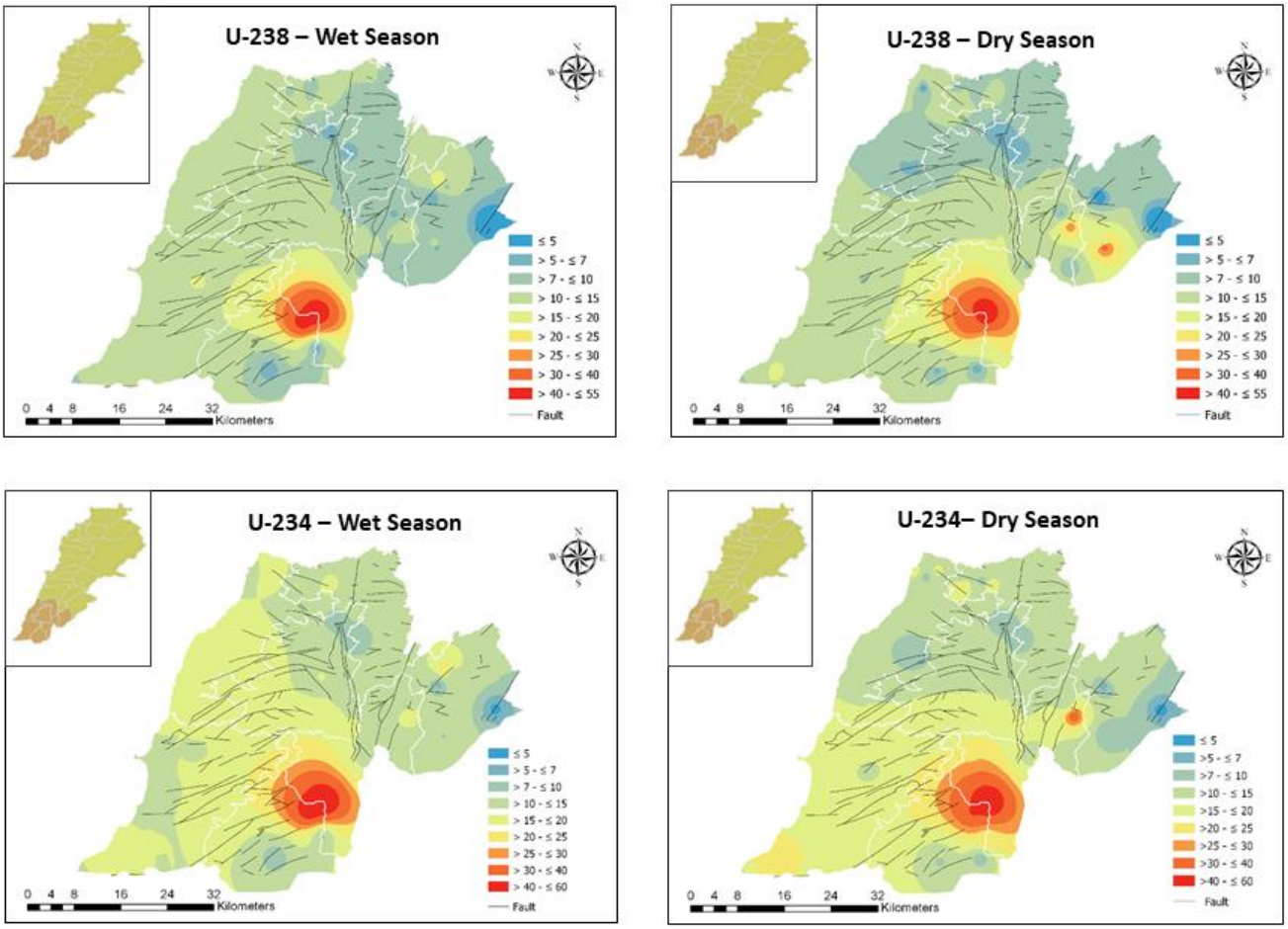


Figure 4.13 ^{238}U and ^{234}U distribution in wet and dry seasons

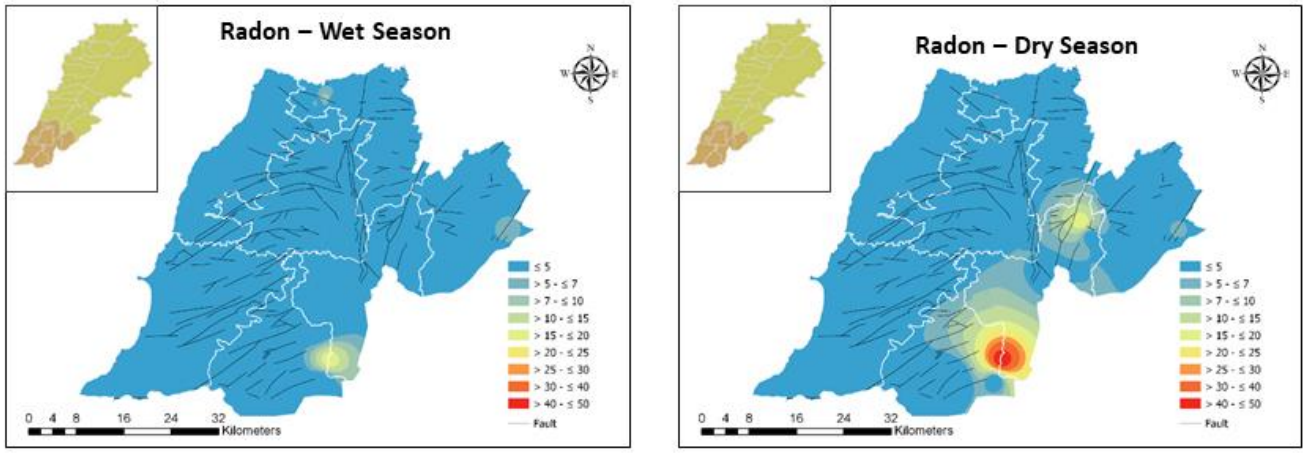


Figure 4.14 Radon distribution in wet and dry seasons

On the other hand, Wilcoxon tests were perfomed to assess the behavior of pH, TDS and conductivity in the two seasons and significant variations were recorded between the wet and dry seasons. P values of 0.033, 0.01 and <0.0001 were recorded for pH, TDS and conductivity respectively (Figure 4.15). However, in contrast to what was expected, these significant vatiations in water properties did not affect too much the radioactivity concentrations of the water.

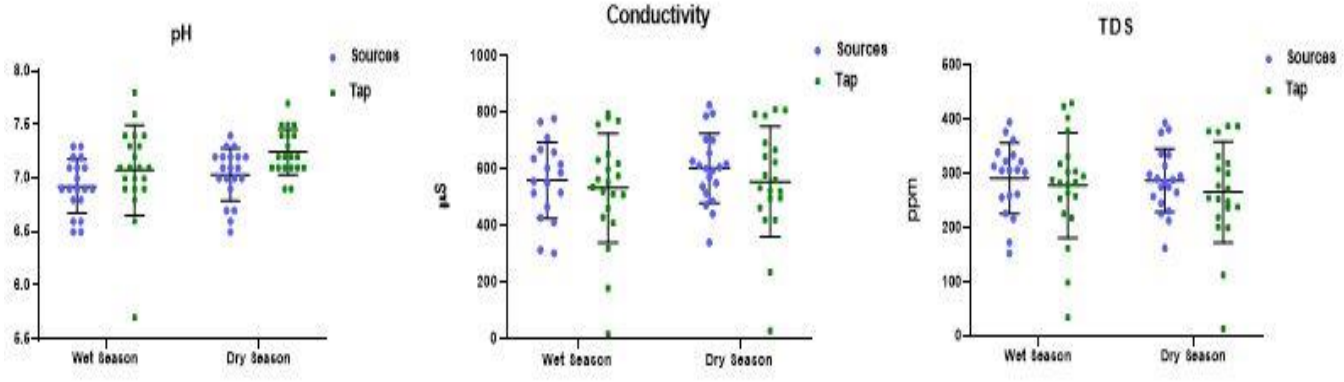


Figure 4.15 Seasonal variations of pH, conductivity and TDS

4.4. Dosimetry Study

The annual effective doses, calculated based on the activities measured during the wet and the dry seasons, and taking into consideration only ^{238}U, ^{234}U and ^{222}Rn, are represented in Figure 4.16 and Figure 4.17. Doses were estimated for three age groups: infant, child and adult assuming that all the annual consumed water are taken from the sampled locations (130, 350 and 730 liters per year for each of the age groups). The effective dose from each of the radionuclides is presented in Table S3. The total doses range between 0.54 μSvy^{-1} and 170.5 μSvy^{-1} for infants, 0.54 μSvy^{-1} and 102.6 μSvy^{-1} for children, and 0.7 μSvy^{-1} and 127.1 μSvy^{-1} for adults. However, the means of the calculated doses were as low as 11.2 μSvy^{-1}, 7.1 μSvy^{-1}, 8.8 μSvy^{-1}

for infant, child and adult respectively the thing that reflects the domination of low doses received from the sampled locations. Received doses, calculated based on activities measured during the wet and the dry seasons, from each radionuclide (^{238}U, ^{234}U and ^{222}Rn) alone and for each of the age groups, are displayed in Appendix 4.

It is noted that radon contribution in the total dose from the ingestion of water is the highest. This is of a particular importance especially that the infants, the most radiosensitive age group, receive the highest radiation dose as compared to children and adults. Most of the calculated doses were far below the WHO recommended level for radiation doses from water ingestion (100 μSvy^{-1}) (WHO 2017).

Few locations exhibit ingestion doses that are relatively high like 70.2 μSvy^{-1}, 42.1 μSvy^{-1} and 52.2 μSvy^{-1} in Al Khiam well (W6), and 33.8 μSvy^{-1}, 20.5 μSvy^{-1} and 25.4 μSvy^{-1} in Kfarfalous tap water (TW10), for each of the age group respectively. One of the wells, Aitaroun (W2), exceeded the recommended dose limit during the dry season, with doses of 170.5 μSvy^{-1}, 102.6 μSvy^{-1}, and 127.1 μSvy^{-1} for infants, children and adults respectively. However, the radiation doses in the corresponding tap water were of low value varying between 9.5 μSvy^{-1}, 5.8 μSvy^{-1} and 7.2 μSvy^{-1} for the three age groups respectively. This indicates that the water from this location can still be used as a safe drinking water supply despite the relatively higher radon content at the source.

These results are of a specific interest mainly because the reported doses do not take into considerations the dose contribution of possible beta emitters that might be present in the sampled water. Further investigations in these locations are recommended to build a complete radiological profile for these locations.

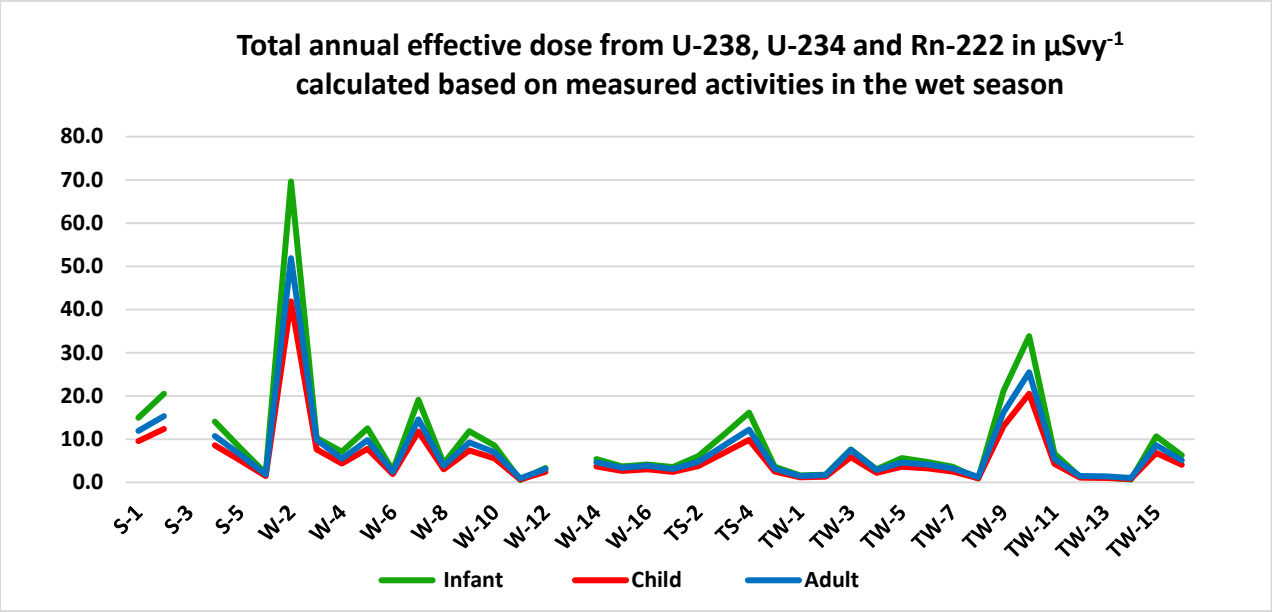


Figure 4.16 Total annual effective dose from ^{238}U, ^{234}U and ^{222}Rn calculated based on activities measured during the wet season

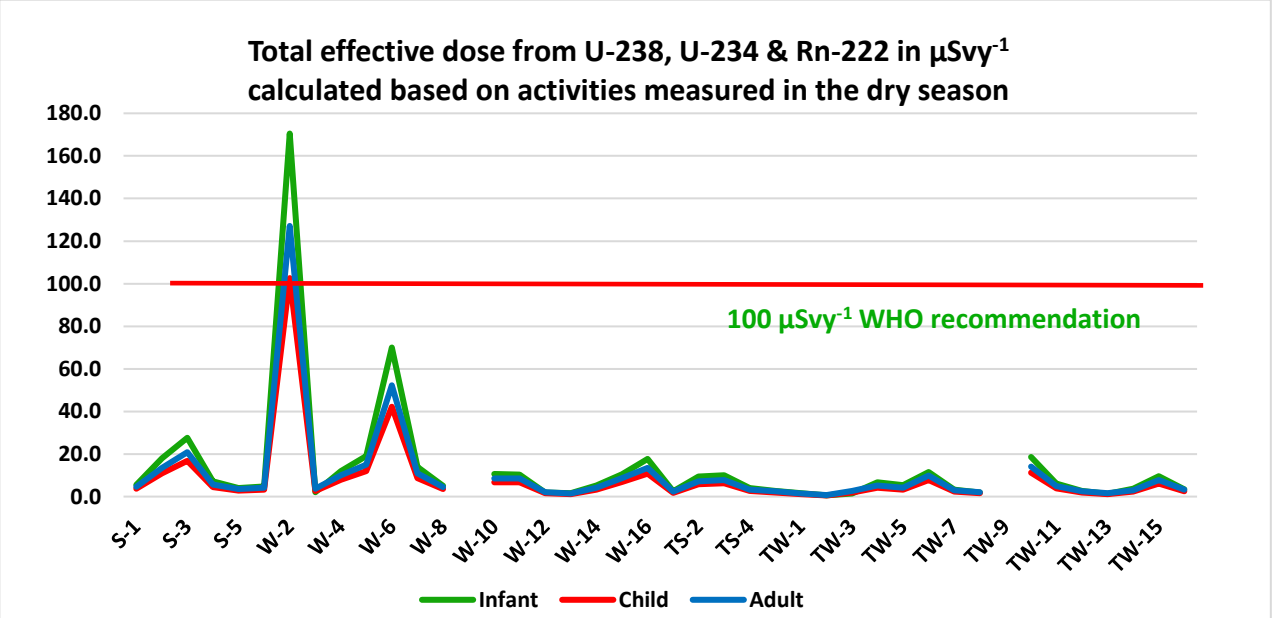


Figure 4.17 Total annual effective dose from ^{238}U, ^{234}U and ^{222}Rn calculated based on activities measured in the dry season

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1. Conclusion

The preliminary radioactivity assessment in the Southern region of Lebanon revealed gross alpha and gross beta, ^{238}U, ^{234}U and ^{22}Rn activity concentrations to be lower than the screening levels set by the World Health Organization. Gross beta activity concentrations were higher than gross alpha activity concentrations. Effective doses in few locations exhibited relatively higher values, and one of them exceeded the WHO IDC value. Seasonal variations were assessed and no significant difference in activity concentrations was noted between the wet and dry seasons except for gross alpha activity concentrations. Comparison of radioactivity concentrations in water sampled directly from the sources and in the corresponding tap water also demonstrated insignificant variation, except for the case of radon. In addition, no significant variation in radioactivity concentrations was noted between springs and wells water.

Spatial distribution mapping revealed that the most elevated concentration levels, for gross alpha and gross beta, uranium and radon are located at the South-West side of the studied area. This was almost consistent in both wet and dry seasons. Mild correlations were detected between the measured radioactivity and the physiochemical components.

The current work constitutes a preliminary study that can be used as a baseline reference for any future natural radioactivity investigations in the Southern region of Lebanon. Follow up investigations, especially in locations exhibiting

higher radiation doses are recommended. Furthermore, to build a complete radiological profile of this region, further assessment of specific naturally occurring radionuclides and studying their prevalence in association with the underlying geological composition and distribution of hydrological aquifers is well advised.

5.2. Recommendations

Based on the results of this study, the following recommendations are proposed:

- Developing national regulations that takes into consideration the assessment of the radiological quality of public sources of drinking water before being served for general public consumption.
- Performing a follow-up assessment of natural radioactivity in the studied area to detect any variations especially in locations where relatively elevated radioactivity concentrations were spotted and including more sampling points.
- Investigating the presence of other naturally occurring radionuclides in drinking water to include those that were not covered in this study with concentration on beta emitters ^{40}K, ^{210}Pb and ^{228}Ra specifically – only for the sake of investigating their presence, not for regulatory compliances.
- Including more investigation points to improve the statistical significance of the samples and performing similar natural radioactivity assessments in other regions of Lebanon to build a complete radiological profile for the country.
- Establishment of decentralized radio analytical laboratories and offering the needed trainings for its operation.

APPENDIX 1

INTRODUCTION TO RADIOACTIVITY

Radioactivity

Radioactivity is the process of spontaneous transformation (disintegration) of an unstable nuclide (radionuclide) into a more stable one through the emission of ionizing radiation i.e. energetic enough to make the atom lose electrons and become ionized. This process is also called radioactive decay. The atoms of each radionuclide disintegrate at a unique decay rate specific for each radionuclide. The decay rate of each isotope is expressed by its *physical half-life ($T_{1/2}$)*, which is the time required for an amount of radioisotope to decrease to one-half of its original activity. The relationship between the activity of a radionuclide at any point of time and the initially present activity is expressed by equation A1.1 (Lawson 1999):

$$A_t = A_0 e^{-\lambda \Delta t} = A_0 e^{-[\frac{ln2}{T_{1/2}}(t-t_0)]} \quad \text{(A1.1)}$$

When radioactive materials get into the body, two other expressions related to radioactivity are commonly used. *Biological half-life* which is the time needed for the body to biologically eliminate half of the introduced activity, and *Effective half-life* which integrates both physical and biological half-lives. The effective half-life is the time needed for the removal of half of the activity from the body by the collective processes of the radioactive decay and the biological elimination. It is calculated using equation A1.2 below (Lombardi 2006):

$$T_e = (T_p \times T_b) \ / \ (T_p + T_b) \quad \text{(A1.2)}$$

where: T_e is the effective half-life

T_p is the physical half-life

T_b is the biological half-life

The radioactivity, referred to as activity, is measured by the rate at which atoms undergo radioactive disintegration per second (dps), or Becquerel (B), where 1 B is equal to 1 dps. An older unit for measuring the amount of radioactivity in a source is the Curie (Ci), which is equal to $3.7 .10^{10}$ Bq and represents the activity of a one gram of ^{226}Ra (Mettler Jr and Guiberteau 2012).

There are several modes for radioactive decay that lead to the emission of different types of radiation: *electromagnetic* like Gamma (γ) rays and X-Rays, and *particulate* like alpha particles (α), beta particles (β), neutrons and protons. Most of natural radionuclides decay mainly through one or a combination of alpha, beta and gamma radiation. Therefore, only alpha, beta and gamma decay processes will be discussed.

Alpha particles

Alpha particle (α) is a relatively massive particle that contains 2 protons and 2 neutrons and carries two positive charges which makes it identical to a helium nucleus (^4_2He) without its electrons. They are particles of relatively large mass and low penetrating power. Actually, they can be easily stopped by a thin sheet of paper and cannot penetrate the dead layer of the skin (Gilmore 2011). Alpha particles have high ionizing power and high linear energy transfer (LET), meaning that they deposit all of their energy over a short distance of matter, few centimeters in air and few

micrometers in solids. This property makes alpha particles the most dangerous type of radiation when it gets to the inside of the body (Antoni and Bourgois 2017). Actually, the damage introduced by alpha particles is 20 times more powerful than that can be caused by beta particles or gamma radiation: it has a *quality factor* of 20 (NCRP 1993). The quality factor (Q) is a dimensionless factor intended to "relate the biological effectiveness of the absorbed dose in tissue" (Veinot and Hertel 2005), meaning that it "reflects the effectiveness of a particular type of radiation resulting in the same biological effect as another type of radiation" (Seeram and Travis 1997).

Alpha particles are emitted from the decay process of heavy elements with large atomic number (Z > 83) like ^{238}U, ^{234}U, ^{232}Th, ^{210}Po and ^{222}Rn. Alpha decay occurs as per equation A1.3 below:

$$ {}^{A}_{Z}X \quad \rightarrow \quad {}^{A-4}_{Z-2}Y \quad + \quad {}^{4}_{2}He \qquad \text{(A1.3)} $$

Beta decay

Beta particles are much less massive than the alpha particles. Beta particles are emitted in a continuous spectrum of energy with a maximum energy of decay, E_{max}, characteristic for each radionuclide. The charge of the emitted beta particle can be either negative (electron) or positive (positron) (Gilmore 2011):

- Beta negative ($^{-}\beta$) occurs when ratio of neutron:protons ratio in an atom is too high. In such a case, a neutron is transformed into a proton, whereby an electron and an electron-type antineutrino ($\bar{\upsilon_e}$) are emitted; thus the atomic number increases by 1. Beta negative decay occurs as per equation A1.4 below:

$$ {}^{A}_{Z}X \quad \rightarrow \quad {}_{Z+1}{}^{A}Y \quad + \quad {}_{-1}{}^{0}\beta \quad + \quad {}^{0}_{0}\upsilon_{\mathrm{e}} \qquad \text{(A1.4)} $$

- Beta positive ($^{+}\beta$) is the result of a low neutron:proton ratio and it occurs as a proton is converted into a neutron, whereby a positron and a neutrino (υ_e) are emitted. The atomic number decreases by 1 in ($^{+}\beta$) decay, equation A1.5:

$$ {}^{A}_{Z}X \quad \rightarrow \quad {}^{A}_{Z-1}Y \quad + \quad {}^{0}_{+1}\beta \quad + \quad {}^{0}_{0}\bar{\upsilon}_e \qquad \text{(A1.5)} $$

Examples of radionuclide which decay with beta emission include Tritium (^{3}H), Carbon-14 (^{14}C), Strontium-90 (^{90}Sr), Yttrium-90 (^{90}Y), Thorium-234, (^{234}Th), Radium-228 (^{228}Ra) and Lead-211 (^{211}Pb). Beta particles are more penetrating than alpha particles. The penetrating range is directly proportional to its energy. In general a low atomic number material, like plastic or thin layer of aluminum, is needed to shield beta particles (Mattsson and Hoeschen 2013).

Gamma decay

Gamma rays are emitted from the nucleus of an exited atom as a way to get rid of the excess energy. It is mostly associate with the alpha or beta disintegrations as they leave the product nuclide in an exited state after the decay (L'Annuziata 1998). They are electromagnetic rays with the same characteristics of light but with much shorter wavelength and higher energy (Amsler 2015). Thus, they have a high penetrating power and need high density material to be shielded (Mattsson and Hoeschen 2013).

Table A1.1 summarizes some properties of each of the three main radiation types and its characteristics.

Table A1.1 Properties of common radiation types

Name	Symbol	Range	Penetrating Power	Ionizing power	Quality factor (Q)	Shielding
Alpha particle	α	Short	Very low	Very high	20	None
Beta particle	β	Moderate	Intermediate	Intermediate	1	Low atomic number material (plastic, aluminum)
Gamma radiation	γ	Long	Very high	Very low	1	High density material (Lead)

Biological effects of radiation

Generalities

Ionizing radiations are capable of ejecting electrons form the atoms and can introduce molecular changes at the cellular levels. As such, ionizing radiation can produce biological damage. Biological effects are determined by the type of emitted radiation, the part of body receiving the radiation, the rate at which the dose is received, the exposure mode and the age at which the dose was received.

Hazardous exposure to ionizing radiation can be divided into two groups: external and internal (Ritz et al. 2000). External exposure occurs as the radiation arrives to the body from outside the skin and the hazard is eliminated when the radiation source is shielded or moved away. External exposure follows three modes: (1) *Distant source*: occurs from an outside source of radiation capable of penetrating the body, like gamma rays or the X-Rays emitted during the decay process of ^{238}U, ^{235}U, ^{210}Po and ^{226}Ra or the high energy beta emitted from radionuclides like ^{32}P; (2) *Contact:* due to handling of or getting into contact with a radioactive material or a substance containing radioactive materials. In such a case, the radioactive materials

can get deposited directly on the skin; (3) *Immersion in a cloud*: this occurs in the presence of gaseous or volatile radioactive materials (Delacroix et al. 1998).

On the other hand, the internal exposure happens when the source of radiation gets into the body through inhalation, ingestion, injection or open wounds. Examples of internal irradiation is the decay of natural radionuclides after getting to the inside of the body through the ingestion of water or through the food chain. Internal irradiation can be due to the internal radioactive material like ^{40}K, ^{14}C, or the decay products of the ^{238}U, ^{235}U and ^{232}Th radioactive series (Delacroix et al. 1998). In internal exposure, the radiation remains inside the body till it decays or until it gets excreted i.e. urination or exhalation (Trapp and Kron 2008).

Biological effects of radiation are divided into two categories: stochastic and non-stochastic effects (ICRP 1991). *Stochastic* effects are probabilistic effects dependent on the amount of radiation dose. Stochastic effects have no threshold for occurrence and the probability of occurrence increases as the dose increases, and with each radiation dose. The first appearance of radiation effect can be seen early after the moment of irradiation or at a later stage like in the case of cancer (Sherer et al. 2018) which is the most serious stochastic effect. *Non-stochastic* effects are deterministic, meaning that an effect will definitely appear when a dose threshold is reached. Deterministic effect are of concern when high radiation doses are encountered. This can happen when high radiation doses are received over a short period of time (acute effect), or from a cumulative low doses of radiation received over a long period of time (latent effect). In non-stochastic effects, the severity of the response increases with an increase in the radiation dose (Trapp and Kron 2008). Another important aspect of radiation exposure is that the radiation damage can

affect the living organism who have been exposed to radiation (somatic effect), or it can be projected to unborn future generation (genetic effect). Table A1.2 summarizes the biological effects of radiation.

Radiation exposure from naturally occurring radioactive materials is generally of the low level exposure dose. It constitutes 80% i.e 2.4 mSv/year, of the global average annual dose per person from all sources of environmental radiation, which is 3 mSv per year (UNSCEAR 2008). The Sievert (Sv) is the unit of measurement for the effective dose from radiation. The effective dose measures the effect of radiation taking into account the type of radiation and the irradiated body part (Sherer et al. 2018).

Table A1.2 Biological effects of radiation

<table>
<tr><th>Characteristics of effects</th><th>Occurrence time</th><th>Object</th><th>Effects/damage</th></tr>
<tr><td rowspan="2">Stochastic Effects (Probabilistic)</td><td rowspan="2">Latent Effects</td><td>Genetic Effect</td><td>Heredity effect</td></tr>
<tr><td rowspan="3">Somatic Effects</td><td>Cancer</td></tr>
<tr><td rowspan="2">Non-stochastic Effects (Deterministic)</td><td>Cataract
Embryologic effect</td></tr>
<tr><td>Acute Effects</td><td>Skin burns
nausea, hair loss
Acute radiation sickness
Chronic radiation sickness
Digestive system
Central nervous system damage
Sterility</td></tr>
</table>

Dose-Response Models

Knowledge about the biological effects of radiation are derived from studies on human and animal exposure. The response to high dose radiation leading to deleterious effects, including cancer, is well understood (Brenner et al. 2003). Data on high dose radiation effects were collected from early mine and radiation workers, survivors of nuclear bombs and accidents, and from patients exposed to therapeutic and diagnostic procedures (Ozasa et al. 2011; Ritz et al. 2000). However, the health effects related to low doses of radiation are still not definite (Tran and Seeram 2017). For instance, the average environmental natural radioactivity is 2.4 mSv/year and it is as high as 24 mSv/year in some locations in the world, yet no noticeable increase in health risks was observed in long-term epidemiologic studies (Hauri et al. 2013; Nair et al. 2009; Tao et al. 2000). Nevertheless, this was not the case in other studies which concluded that there might be some association between background radiation and increased cancer risks in childhood like Leukemia and Central Nervous System tumors (Kendall et al. 2013; Spycher et al. 2015).

Several models (Figure A1.1) have been developed aiming at estimating the excess cancer risk from low-dose exposures. Estimation was done based on extrapolation since intentional irradiation for research purposes is neither appropriate nor ethical (Seeram and Travis 1997). All dose-response models are based on predicting radiation effects at doses below 100 mSv, which is the lowest dose where excess cancer has been observed (Tang and Loganovsky 2018). Below the 100 mSv dose, the epidemiological studies could not conclusively identify an increased risk of cancer (Brenner et al. 2003). Dose-response models are briefed as follows:

- *Linear-Quadratic* model suggests that excess cancer risk per unit dose (risk per Sievert) is less important at lower doses than at higher doses.
- *Supralinear* model predicts that excess cancer risk per unit dose (risk per Sievert) at low doses is more serious than those at higher dose (Zanzonico et al. 2016).
- *Hormesis* model implies that individuals exposed to low doses of radiation have a lower risk for developing cancer i.e. prediction of beneficial and protective effects of radiation at low doses (Calabrese and Blain 2011; Calabrese et al. 2015; Vaiserman 2010)
- *Linear No-Threshold* (NLT) model infers that the cancer risk per unit dose is the same from low to high doses. It suggests that there is no threshold below which the risk of cancer is excluded i.e. no dose, no matter how small it is, is a safe dose. For radiation safety purposes, this model has been adapted for risk estimation by the authoritarian advisory institutions: ICRP, NCRP, UNSCEAR, (ICRP 2007; NCRP 1993; NCRP 2001; UNSCEAR 2000). However, recent radiobiological experiments are revealing results that are not in line with the LNT hypothesis (Averbeck et al. 2018; Sanders 2009).

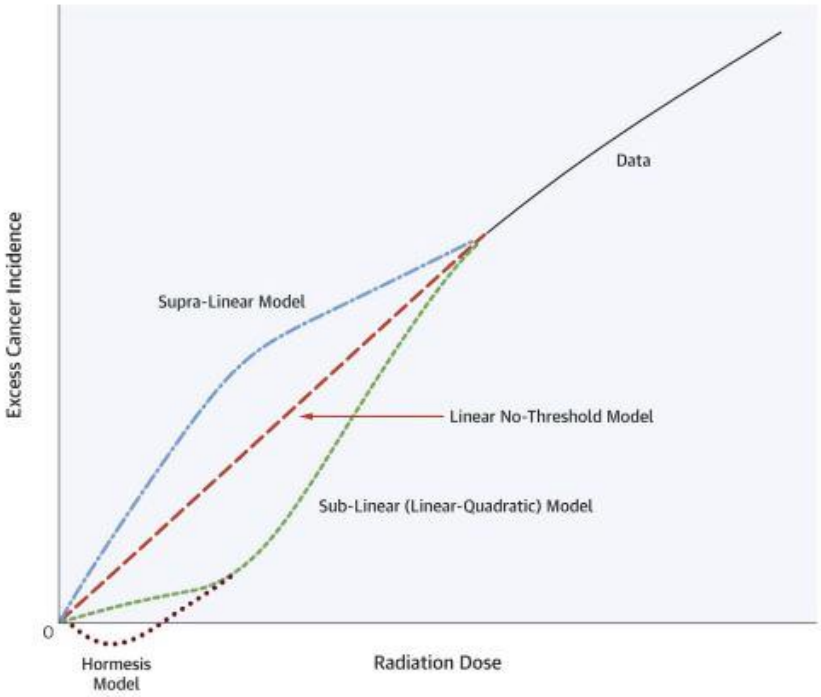


Figure A1.1 Dose-response curve for carcinogenesis (Zanzonico et al. 2016)

APPENDIX 2

MEASURMENT RESULTS FOR ALL LOCATIONS DURING THE WET AND THE DRY SEASONS

Unique ID	Alpha Wet mBq/L	Alpha Dry mBq/L	Beta Wet mBq/L	Beta Dry mBq/L	^{238}U Wet mBq/L	^{238}U Dry mBq/L	^{234}U Wet mBq/L	^{234}U Dry mBq/L	Radon Wet Bq/L	Radon Dry Bq/L
S-1	75	< MDA	91.7	48.6	19.5	8.4	23.1	13.3	4.1	1.5
S-2	50	< MDA	77.7	33.9	2.3	2.1	4.6	4.5	5.9	5.2
S-3	-	< MDA	-	60.6	-	8.5	-	12.5	-	7.9
S-4	2.1	< MDA	108.3	54.8	6.4	5.7	7.6	8.3	4	2
S-5	< MDA	< MDA	< MDA	50.5	10.5	11.4	12.3	19	2.2	1
W-1	30.4	< MDA	139.2	167	6.9	14	9.1	16.2	0.5	1.2
W-2	11.1	128.6	74.4	170.8	5.6	18.5	9.5	22.4	20.1	49.2
W-3	374.6	155.8	280.8	418	51	53.7	55.9	55.7	2.4	-
W-4	< MDA	< MDA	20.6	15.5	3.2	47.2	6.4	7	2	3.2
W-5	< MDA	< MDA	130.6	67.8	8.6	18.7	14.1	22.7	3.5	5.3
W-6	43.6	< MDA	63.9	65.8	6.6	6.7	10.8	10.9	0.7	20.2
W-7	< MDA	< MDA	202.1	87.8	9.7	10.6	13.3	15.2	5.4	3.9
W-8	< MDA	< MDA	98.3	88.3	11.1	12.7	17.6	19.8	1.1	1.3
W-9	< MDA	-	94.8	-	9.8	-	13.4	-	3.3	-
W-10	1.9	< MDA	84.4	55.8	12.2	12.7	18	18.4	2.3	2.9
W-11	< MDA	< MDA	106.9	55.8	10	15.2	15.1	22.7	0	2.8
W-12	< MDA	< MDA	179.2	72.5	13	13.2	17	15.6	0.8	0.4
W-13	-	< MDA	-	78.3	-	4.4	-	7.6	-	0.4
W-14	2.5	< MDA	170	112.8	11.7	8.1	18.7	9.7	1.4	1.4
W-15	43.6	< MDA	91.2	53	10.4	18.2	20	24.2	0.9	2.8
W-16	0.8	< MDA	80.8	41.3	16.4	9.2	20.6	11.8	1	5
TS-1	120.2	< MDA	70.2	67.3	9.4	9	12.8	14.1	0.9	0.6
TS-2	51.5	< MDA	132.5	12.8	5	2.9	6.5	5.9	1.7	2.7
TS-3	4.1	< MDA	106.3	75.7	6.9	7.8	10.2	11.2	3.1	2.8
TS-4	< MDA	< MDA	162.5	64.4	6.1	5.2	7.5	7.9	4.6	1.1
TS-5	< MDA	< MDA	< MDA	47.2	12.8	11.6	13.4	15.8	0.9	0.6
TW-1	22.5	< MDA	191.7	215.6	5.4	4.7	7.7	7	0.4	0.4
TW-2	< MDA	< MDA	< MDA	63	7.5	5.9	11.9	7.5	0.4	0.1
TW-3	327.9	< MDA	395	382.6	45	36.7	47.3	40.7	1.7	-
TW-4	< MDA	< MDA	48.9	53	14.5	3.5	17	7.7	0.7	1.9

TW-5	< MDA	< MDA	15.3	62.5	9.6	3.6	12	5.9	1.5	1.5
TW-6	< MDA	99.1	26.7	145.8	12.7	27.7	17.9	34.4	1.2	3
TW-7	< MDA	< MDA	70.8	54.8	11.3	11.9	15.8	14.5	0.9	0.8
TW-8	3.3	< MDA	120.6	85.3	10.9	15.2	18.2	15.2	0.1	0.4
TW-9	< MDA	-	8.3	-	11.5	-	14.7	-	6	-
TW-10	< MDA	< MDA	< MDA	36.7	7.8	6.9	12.2	7.5	9.7	5.3
TW-11	< MDA	< MDA	96.7	57.8	9.7	7	14	9.3	1.8	1.7
TW-12	7.7	< MDA	140.2	93	12.7	13.7	15.6	15.4	0.2	0.6
TW-13	2.5	< MDA	87.4	81.9	10.6	14	14.1	16.9	0.2	0.2
TW-14	< MDA	< MDA	89.2	67.3	12.1	5.2	15.9	7.1	0.03	1
TW-15	3.3	< MDA	83.6	51.3	15.4	11.2	18.2	21	2.9	2.6
TW-16	20.3	< MDA	90	66.3	10	14.6	11.9	20.2	1.7	0.8

APPENDIX 3

ANNUAL EFFECTIVE DOSE OF ^{238}U, ^{234}U AND ^{222}Rn FROM WATER INGESTION OF WATER FOR INFANT, CHILD, AND ADULT CALCULATED BASED ON ACTIVITIES MEASURED IN THE WET AND THE DRY SEASONS

ID	^{238}U, ^{234}U and ^{222}Ra Annual effective doses calculated based on activity concentrations measured in the Wet season								
	U-238 Infant µSv	U-238 Child µSv	U-238 Adult µSv	U-234 Infant µSv	U-234 Child µSv	U-234 Adult µSv	Rn-222 Infant µSv	Rn-222 Child µSv	Rn-222 Adult µSv
S-1	0.35	0.46	0.64	0.45	0.60	0.83	14.15	8.47	10.48
S-2	0.04	0.05	0.08	0.09	0.12	0.16	20.36	12.18	15.07
S-3	-	-	-	-	-	-	-	-	-
S-4	0.12	0.15	0.21	0.15	0.20	0.27	13.80	8.26	10.22
S-5	0.19	0.25	0.34	0.24	0.32	0.44	7.59	4.54	5.62
W-1	0.12	0.16	0.23	0.18	0.24	0.33	1.73	1.03	1.28
W-2	0.10	0.13	0.18	0.19	0.25	0.34	69.35	41.51	51.36
W-3	0.92	1.21	1.68	1.09	1.45	2.00	8.28	4.96	6.13
W-4	0.06	0.08	0.11	0.12	0.17	0.23	6.90	4.13	5.11
W-5	0.15	0.20	0.28	0.27	0.37	0.50	12.08	7.23	8.94
W-6	0.12	0.16	0.22	0.21	0.28	0.39	2.42	1.45	1.79
W-7	0.17	0.23	0.32	0.26	0.34	0.48	18.63	11.15	13.80
W-8	0.20	0.26	0.36	0.34	0.46	0.63	3.80	2.27	2.81
W-9	0.18	0.23	0.32	0.26	0.35	0.48	11.39	6.81	8.43
W-10	0.22	0.29	0.40	0.35	0.47	0.64	7.94	4.75	5.88
W-11	0.18	0.24	0.33	0.29	0.39	0.54	0.07	0.04	0.05
W-12	0.23	0.31	0.43	0.33	0.44	0.61	2.76	1.65	2.04
W-13	-	-	-	-	-	-	-	-	-
W-14	0.21	0.28	0.38	0.36	0.48	0.67	4.83	2.89	3.58
W-15	0.19	0.25	0.34	0.39	0.52	0.72	3.11	1.86	2.30
W-16	0.30	0.39	0.54	0.40	0.53	0.74	3.45	2.07	2.56
TS-1	0.17	0.22	0.31	0.25	0.33	0.46	3.11	1.86	2.30
TS-2	0.09	0.12	0.16	0.13	0.17	0.23	5.87	3.51	4.34
TS-3	0.12	0.16	0.23	0.20	0.26	0.36	10.70	6.40	7.92
TS-4	0.11	0.15	0.20	0.15	0.19	0.27	15.87	9.50	11.75
TS-5	0.23	0.30	0.42	0.26	0.35	0.48	3.11	1.86	2.30
TW-1	0.10	0.13	0.18	0.15	0.20	0.28	1.38	0.83	1.02

TW-2	0.14	0.18	0.25	0.23	0.31	0.43	1.38	0.83	1.02
TW-3	0.81	1.07	1.48	0.92	1.23	1.69	5.87	3.51	4.34
TW-4	0.26	0.35	0.48	0.33	0.44	0.61	2.42	1.45	1.79
TW-5	0.17	0.23	0.32	0.23	0.31	0.43	5.18	3.10	3.83
TW-6	0.23	0.30	0.42	0.35	0.46	0.64	4.14	2.48	3.07
TW-7	0.20	0.27	0.37	0.31	0.41	0.57	3.11	1.86	2.30
TW-8	0.20	0.26	0.36	0.35	0.47	0.65	0.35	0.21	0.26
TW-9	0.21	0.27	0.38	0.29	0.38	0.53	20.70	12.39	15.33
TW-10	0.14	0.19	0.26	0.24	0.32	0.44	33.47	20.03	24.78
TW-11	0.17	0.23	0.32	0.27	0.36	0.50	6.21	3.72	4.60
TW-12	0.23	0.30	0.42	0.30	0.40	0.56	0.69	0.41	0.51
TW-13	0.19	0.25	0.35	0.27	0.37	0.50	0.69	0.41	0.51
TW-14	0.22	0.29	0.40	0.31	0.41	0.57	0.10	0.06	0.08
TW-15	0.28	0.37	0.51	0.35	0.47	0.65	10.01	5.99	7.41
TW-16	0.18	0.24	0.33	0.23	0.31	0.43	5.87	3.51	4.34
Avr	0.21	0.28	0.39	0.30	0.40	0.56	8.82	5.28	6.53
Min	0.04	0.05	0.08	0.09	0.12	0.16	0.07	0.04	0.05
Max	0.92	1.21	1.68	1.09	1.45	2.00	69.35	41.51	51.36

ID	**^{238}U, ^{234}U and ^{222}Ra Annual effective doses calculated based on activity concentrations measured in the Dry season**								
	U-238 Infant µSv	**U-238 Child µSv**	**U-238 Adult µSv**	**U-234 Infant µSv**	**U-234 Child µSv**	**U-234 Adult µSv**	**Rn-222 Infant µSv**	**Rn-222 Child µSv**	**Rn-222 Adult µSv**
S-1	0.15	0.20	0.28	0.26	0.34	0.48	5.18	3.10	3.83
S-2	0.04	0.05	0.07	0.09	0.12	0.16	17.94	10.74	13.29
S-3	0.15	0.20	0.28	0.24	0.32	0.45	27.26	16.31	20.18
S-4	0.10	0.14	0.19	0.16	0.21	0.30	6.90	4.13	5.11
S-5	0.21	0.27	0.37	0.37	0.49	0.68	3.45	2.07	2.56
W-1	0.25	0.33	0.46	0.32	0.42	0.58	4.14	2.48	3.07
W-2	0.33	0.44	0.61	0.44	0.58	0.80	169.74	101.60	125.71
W-3	0.97	1.28	1.76	1.09	1.44	1.99	-	-	-
W-4	0.85	1.12	1.55	0.14	0.18	0.25	11.04	6.61	8.18
W-5	0.34	0.45	0.61	0.44	0.59	0.81	18.29	10.94	13.54
W-6	0.12	0.16	0.22	0.21	0.28	0.39	69.69	41.71	51.61
W-7	0.19	0.25	0.35	0.30	0.39	0.54	13.46	8.05	9.96
W-8	0.23	0.30	0.42	0.39	0.51	0.71	4.49	2.68	3.32

W-9	-	-	-	-	-	-	-	-	-
W-10	0.23	0.30	0.42	0.36	0.48	0.66	10.01	5.99	7.41
W-11	0.27	0.36	0.50	0.44	0.59	0.81	9.66	5.78	7.15
W-12	0.24	0.31	0.43	0.30	0.40	0.56	1.38	0.83	1.02
W-13	0.08	0.10	0.14	0.15	0.20	0.27	1.38	0.83	1.02
W-14	0.15	0.19	0.27	0.19	0.25	0.35	4.83	2.89	3.58
W-15	0.33	0.43	0.60	0.47	0.63	0.87	9.66	5.78	7.15
W-16	0.17	0.22	0.30	0.23	0.31	0.42	17.25	10.33	12.78
TS-1	0.16	0.21	0.30	0.27	0.37	0.50	2.07	1.24	1.53
TS-2	0.05	0.07	0.10	0.12	0.15	0.21	9.32	5.58	6.90
TS-3	0.14	0.19	0.26	0.22	0.29	0.40	9.66	5.78	7.15
TS-4	0.09	0.12	0.17	0.15	0.20	0.28	3.80	2.27	2.81
TS-5	0.21	0.28	0.38	0.31	0.41	0.57	2.07	1.24	1.53
TW-1	0.08	0.11	0.15	0.14	0.18	0.25	1.38	0.83	1.02
TW-2	0.11	0.14	0.19	0.15	0.19	0.27	0.35	0.21	0.26
TW-3	0.66	0.87	1.21	0.79	1.05	1.46	-	-	-
TW-4	0.06	0.08	0.11	0.15	0.20	0.28	6.56	3.92	4.85
TW-5	0.06	0.09	0.12	0.12	0.15	0.21	5.18	3.10	3.83
TW-6	0.50	0.66	0.91	0.67	0.89	1.23	10.35	6.20	7.67
TW-7	0.21	0.28	0.39	0.28	0.38	0.52	2.76	1.65	2.04
TW-8	0.27	0.36	0.50	0.30	0.39	0.54	1.38	0.83	1.02
TW-9	-	-	-	-	-	-	-	-	-
TW-10	0.12	0.16	0.23	0.15	0.19	0.27	18.29	10.94	13.54
TW-11	0.13	0.17	0.23	0.18	0.24	0.33	5.87	3.51	4.34
TW-12	0.25	0.33	0.45	0.30	0.40	0.55	2.07	1.24	1.53
TW-13	0.25	0.33	0.46	0.33	0.44	0.60	0.69	0.41	0.51
TW-14	0.09	0.12	0.17	0.14	0.18	0.25	3.45	2.07	2.56
TW-15	0.20	0.27	0.37	0.41	0.54	0.75	8.97	5.37	6.64
TW-16	0.26	0.35	0.48	0.39	0.52	0.72	2.76	1.65	2.04
Avr	0.23	0.31	0.42	0.30	0.40	0.56	13.23	7.92	9.80
Min	0.04	0.05	0.07	0.09	0.12	0.16	0.35	0.21	0.26
Max	0.97	1.28	1.76	1.09	1.44	1.99	169.74	101.60	125.71

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