

Determination of Baseline Natural Radioactivity Levels in the Suez-Canal Water Stream

By

Sayed Ali Mohamed Hassan El-Mongy

**B.Sc. Chemistry (Ain Shams University)
M.Sc. Radiochemistry (Ain Shams University)**

THESIS

**For the Degree of Doctor
of Philosophy
in**

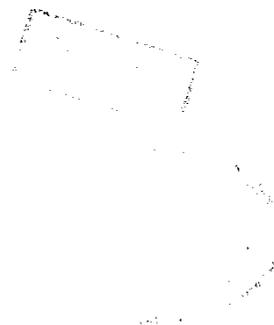
Chemistry

**Faculty of Science
Ain Shames University
CAIRO - EGYPT
1995**

541.38
S.A



51540

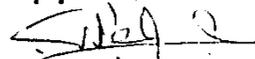


Determination of Baseline Natural Radioactivity Levels in the Suez-Canal Water Stream

Thesis Supervisors

Prof.Dr. M.S.Abdel Mottaleb
Prof.Dr. Abd El Gawad S. Emara
Prof. Dr. Aly Islam M.Aly

Approved



Abdel Gawad Emara

A. I. M. Aly



Prof.Dr.Fahmy A.F.M.

A. F. M. Fahmy

Head of Chemistry Dept.

Faculty of Science

Ain-Shams University



ACKNOWLEDGEMENT

The author wishes to express his very deep gratitude to Prof. Dr. M.S. Abdel Mottaleb Professor of Inorganic Chemistry, faculty of Science, Ain-Shams University, for sponsoring this work.

The author is greatly indebted to Prof. Dr. Abdel Gawad S. Emara head of the Nuclear Materials Department, Atomic Energy Authority, who has initiated this work. He is also acknowledged for his deep interest, scientific discussions and for his big effort in the revision and final presentation of the thesis.

The author wishes to express his thanks to Prof. Dr. Aly Islam Metwaly Aly, Professor of Chemical Engineering and head of Siting and Environmental Dept ,Atomic Energy Authority, for his sincere help and supervision.

The author wishes to express his thanks to Prof. Dr. K. L. Kratz, Professor of Nuclear Chemistry, Mainz University, Germany, for his sincere help, discussion and supervision.

The author would like to cordially thank ass. Prof. Dr. M. S. El-Tahawy, the acting head of the Central Laboratory for Environmental Radiation Measurements and Training (CLERMIT), for his sincere help, discussion of the results and final revision of the work.

The author would also like to deeply thank the family of the CLERMIT, especially Prof. Dr. M.F. Ahmed (Ex. head) and ass. Prof Dr. N.M. Ibrahiem for their sincere help and discussion during the gamma spectroscopy measurements.

Thanks are due to all of my colleagues in the CLERMIT for their sincere help and cooperation.

Aim_of_the_Work

This work aims at measuring the natural or background radioactivity levels in the Suez canal water stream. These data serve as a reference or baseline activity level of the canal water before any radiocontaminants are intentionally or accidentally released to the canal. The purpose of this environmental monitoring is also to describe the impact of a given nuclear facility (e.g. nuclear powered ships), at a specific time and location, and to obtain data that can be used as a basis for emergency action. In addition a radiological map can be depicted for radioactive distribution along the canal stream. Water, sediment, beach soils and different species of fishes have been collected along the canal, from Port-Said in the north to the Suez in the South of the Canal. The physical, chemical and laser fluorimetric analyses of the water samples have been carried out. The mechanical and X-ray diffraction analyses of the sediments have also been performed. The radionuclide concentrations (K-40, U-238, Th-232 and Cs-137) have been determined using γ spectrometric analysis. The above mentioned investigations have been carried out in Egypt. Neutron activation and delayed neutrons analyses, have been carried out for the different sediment samples at the Mainz Triga reactor, Gutenberg university, Germany. The obtained data show that the natural radioactivity levels of K-40, U-238 and Th-232 in the Suez-canal stream are comparable with the average international value levels (UNSCEAR DATA). Man-made Cs-137, which is a result of global fall-out, has been observed in some samples in very low concentrations (less than 3 Bq/Kg).

CONTENTS

TITLE :	Page
* Aim of the Work	
* CHAPTER I	INTRODUCTION and LITERATURE REVIEW
I) Origin of the elements	1
II) Classification of the radionuclides	2
ii-a) Naturally occurring radioactive nuclides	
ii-b) Artificially produced radioactive nuclides	
III) The marine environment	8
IV) Suez-Canal waterway	10
V) Radioactivity pathways	11
VI) Interaction mechanisms of radionuclides in the environment	11
VII) Levels of natural radionuclides in the environment	23
VII-a) Level of radionuclides in rocks	24
VII-b) Level of radionuclides in soils and sediments	26
VII-c) Level of radionuclides in water ways	30
VII-d) Level of radioactivity in marine organisms	31
* CHAPTER II	Theory and Applications of Gamma Spectroscopy ,Delayed Neutrons Instrumental Neutron Activation Analysis, X-ray fluorescence and laser fluorimetry
I) Gamma spectrometric analysis	33
II) Sources of system's background	34
III) Secular equilibrium and naturally occurring decay series	35

IV) The environmental applications of spectrometry	36
V) Sources of neutrons :	37
V-i) Radioactive neutron sources	
V-ii) Neutron generator	
V-iii) Nuclear reactors	38
VI) Classification of neutrons	39
VII) Delayed neutron activation analysis (DNAA).	41
VII-a) Delayed neutrons physics	
VII-b) Delayed neutron emission probability	42
VII-c) Delayed neutron activation analysis applications	46
VIII) Instrumental neutron activation analysis(INAA).	48
VIII-a) Introduction	
VIII-b) Activity produced after irradiation	49
VIII-c) Absolute and comparative techniques	52
VIII-d) Advantages and limits of INAA	55
VIII-e) Applications of INAA.	
VIII) x-ray spectrometric analysis	57
VX) Laser fluorimetric analysis	58

CHAPTER III

EXPERIMENTAL WORK

A - Sampling and Sample Preparation

I) Sampling and samples selection	61
II) Sampling of bottom sediments	62
III) Sampling of shore sediments(soil)	63
IV) Sampling of waters	66
V) Sampling of fishes	68
VI) Mapping, sampling and samples preparation of Suez-Canal.	69

B- Set-up and Methods of Analysis	
I) Gamma spectrometric analysis	73
I-a) The system specification	
I-b) Calibration of the system	76
I-c) Radionuclides of interest	79
II) β - Delayed neutron technique	80
II-a) Pneumatic system	
II-b) Neutron counting system	81
II-c) Interference sources of neutron counter	82
II-d) Method of analysis by DNAA	84
III) Instrumental neutron activation analysis(INAA)	85
III-a) Method of the analysis	
III-b) γ -lines and activation products of U and Th	86
III-c) Measurement of the induced activities	
IV) X-ray spectrometry	87
V) Method of analysis of laser fluorimeter	89
VI) Data Treatment	90
VI-i) DNAA results	
VI-ii) INAA results	
VI-iii) Decay time correction	
VI-iv) Calculation of errors	91
* CHAPTER IV	RESULTS and DISCUSSION
I) Results of water analysis	92
II) Results of sediment analysis	95
II-a) Mechanical and x-ray analysis	
II-b) Background radioactivity levels	96
i) Gamma ray measurements	
ii) Neutron activation measurements	108

III) Level of natural radioactivity in fish samples	114
* CHAPTER V.	
SOME DOSE CALCULATIONS TO THE GENERAL PUBLIC	117
I) External dose rate due to swimming in Suez Canal water	
II) External dose due to recreational activities on beaches	
III) Internal dose rate due to consumption of fish	
* SUGGESTED EMERGENCY MONITORING PROCEDURES	125
* REFERENCES	128
* ANNEX OF TABLES	140
* SUMMARY AND CONCLUSIONS	156
* ARABIC SUMMARY	

Chapter I

Introduction
and
Literature Review

INTRODUCTION

Until the beginning of this century man's only exposure to ionizing radiation originated from natural sources which in most habitats near sea level, results in a total body dose of about 100 mrad/year (1). In a few places known for their abnormally high radioactivity the whole-body dose can be ten or more times higher than the average sea-level figure.

Our knowledge of the natural radioactive background serves as a reference starting point if we wish to evaluate the contribution of the additional sources of ionizing radiations that result from man's exploitation of the atom. The phenomenon of natural radioactivity was first discovered by Bequerel just before the end of the 19 century. The phenomenon of natural radioactivity has been used ,for example, to reveal the structure of the atomic nucleus, to estimate the age of the earth, and to measure the rates of sediment formation on ocean bottoms. With the advent of man utilization of nuclear energy, studies of the natural levels of radioactivity have become necessary to understand fully the environmental influences of the radioactivity produced by the atomic energy industry. Only by having knowledge of the amounts of natural radioactivity and manner in which it varies , the intelligent interpretations of monitoring data can be ensured, whether in the vicinity of a nuclear facility, on land, in the oceans, in the atmosphere, or in the tissues of man. Under the terms of global law, impact studies are required before undertaking any major construction work. An environmental radioecology reference survey is also necessary for nuclear facilities to provide a qualitative and quantitative assessment of artificial radionuclides at the site. The qualitative and

quantitative knowledge of natural radioactivity in the ecosystems is important in itself since it is related to the most abundant radionuclide K-40 and other radionuclides (Ra-226 and Pb-210) liable to cause radiation protection problems under extreme conditions. This information is also valuable in understanding the geochemical cycle of certain stable elements (isotopes or chemically similar elements) and provides background data for determining the relative impact of artificial radioactivity from authorized waste discharge from nuclear plants, nuclear powered ships, fall-out from weapon testing and nuclear accidents. An assessment of the interaction between man and the natural - radiation environment requires a good understanding of the distribution and abundances of the naturally occurring radioelements uranium and thorium and K-40 in rock, water, soil, air, biota and food.

LITERATURE REVIEW

1) ORIGIN OF THE ELEMENTS :

One theory of the origin of the elements was originated by G.Gammow and R.A.Alpher and is known as the big-bang theory (2). They originally postulated that in the beginning of our universe all matter in it existed as neutrons in some super giant nucleus (the ylem) which exploded. During the explosion neutrons were converted through β -decay ($t_{1/2}$ 11 min.) to protons, and the protons combined with neutron and other protons to start the process of forming heavier elements. In this fashion all the elements were formed in a matter of less than an hour by a process of neutron capture.

Out of this expanding cloud the stars and planets were formed through condensation process. Through hydrogen and helium burning, neutrons are formed. A few of the reactions can be cited;



and many others certainly occur. As the heavier elements form in the stars, the neutron production increases considerably, since such reactions become more prevalent that heavier elements are the ones involved in the charged particle and γ -induced reactions. The mode of production of the elements changes from that of helium capture to that of neutron capture, so that the elements from iron to bismuth can be formed by a slow process of neutron capture (n, γ reactions), interrupted by β -decay whenever this is faster than the next capture step. Such a process is known as the slow,

or s-process. The reaction probability for the capture of neutrons increases with the atomic number of the element. Despite the abundances of the elements decrease with increasing atomic number, the probability of the formation of heavier elements increases with the atomic number. The s-process can not explain the formation of the elements heavier than bismuth as the transbismuth elements have a number of short-lived species which would prevent the possibility of the formation of thorium and uranium in the amounts observed in nature. The abundance of the heaviest elements are believed to occur through another process. The supernova (explosion of star) stage is a very short-lived one during which neutron production is extremely intense and provides a method whereby the earlier barrier of the short-lived isotopes between polonium and francium is no longer effective and the heaviest elements are rapidly synthesized. This mode of element formation is known as the rapid or r-process. The intensity of the neutron flux as well as the very short time preclude the possibility of β -decay as a competitor to neutron capture in the r-process. This can result in a different isotopic distribution of the element for the r-process compared to that formed in the s-process.

III CLASSIFICATION OF THE RADIONUCLIDES:

The radionuclides present in nature can be classified into two groups (2,4,5):

- A) Naturally occurring radioactive nuclides
- B) Artificially occurring radioactive nuclides

A) NATURALLY OCCURRING RADIOACTIVE NUCLIDES :

The naturally occurring radioactive nuclides can be