

THE ROLE OF
AQUATIC PLANTS AND SEDIMENTS
IN

RADIUM

CYCLING
IN A TROPICAL WETLAND.

A thesis submitted to the School of Biological Sciences,
Macquarie University
for the degree of Master of Science (Honours)

by

Alexander R Williams BSc (New England)

Australian Nuclear Science and Technology Organisation
Lucas Heights Research Laboratories
Private Bag No.1, Menai NSW 2234.

February 1988

MACQUARIE UNIVERSITY

HIGHER DEGREE THESIS (MASTER'S)

AUTHOR'S CONSENT

This is to certify that I, Alexander R. Williams
being a candidate for the M Sc (Hons)
am aware of the policy of the University relating to the retention and
use of higher degree theses as contained in the University's Master's
Degree Regulations generally and in particular Regulation 16(2).

In the light of this policy I agree to allow a copy of my thesis to be
deposited in the University Library for consultation, loan and photocopying
forthwith.

M. Pissone
.....
Signature of Witness

Alex R. Williams
.....
Signature of Candidate

Dated this 24th day of March 19 88

The Academic Senate on 19 July 1988 resolved that the candidate
had satisfied requirements for admission to the degree of MSc(Hons)
This thesis represents a major part of the prescribed program of study.

TABLE OF CONTENTS

	page
SUMMARY	
STATEMENT	
ACKNOWLEDGEMENTS	
Chapter 1 INTRODUCTION	1-1
1.1 The Problem	1-1
1.2 The Magela Floodplain	1-1
1.3 The Buffalo-Grazing Food-Chain	1-2
1.4 Radioactive Wastes from Uranium Mining	1-2
1.5 How Much Radium is Hazardous?	1-4
1.6 Literature on Radium Uptake by Aquatic Plants	1-4
1.7 Simulation Methods	1-5
1.8 Structure of the Thesis	1-5
Chapter 2 NATURAL RADIUM DISTRIBUTION ON MAGELA FLOODPLAIN	2-1
2.1 Introduction	2-1
2.2 Methods	2-1
2.3 Results	2-2
2.3.1 Soil	2-2
2.3.2 Plants	2-3
2.3.3 Buffalo Faeces	2-5
2.4 Comparison With Operational Monitoring Data	2-5
2.5 Discussion	2-6
2.6 The Accumulation Hypothesis	2-7
Chapter 3 BIOLOGY OF THE MAJOR PLANTS	3-1
3.1 Vegetation of the Magela Floodplain	3-1
3.2 Plant Growth Models	3-1
3.3 Mud Couch Grass - <i>Pseudoraphis spinescens</i>	3-2
3.4 The Water Lily - <i>Nymphaea violacea</i>	3-4
3.4.1 Water Lily Production	3-5
3.4.2 Natural Radium Distribution in <i>Nymphaea</i>	3-6
3.5 Conclusion	3-7
Chapter 4 RADIUM KINETICS IN WATER, PLANTS AND SEDIMENT	4-1
4.1 Introduction	4-1
4.2 Radium Uptake by Plants from Water	4-2
4.2.1 Methods	4-4

4.2.2 Results	4-5
4.2.2.1 Water	4-5
4.2.2.2 Green Tissue	4-5
4.2.2.3 Old Tissue	4-8
4.2.3 Conclusion	4-8
4.3 Radium Uptake by Plants from Sediment	4-9
4.3.1 Introduction	4-9
4.3.2 Experimental Sediment	4-9
4.3.3 Methods	4-9
4.3.4 Results	4-10
4.3.5 Discussion	4-10
4.4 Exchange of Radium Between Sediment and Water	4-11
4.4.1 Introduction	4-11
4.4.2 Transfer from Water Column to Bed Sediment	4-12
4.4.2.1 Methods	4-12
4.4.2.2 Results	4-12
4.4.3 Complete Mixing of Sediment and Water	4-13
4.4.3.1 Introduction	4-13
4.4.3.2 Methods	4-13
4.4.3.3 Results	4-14
4.5 Radium Uptake into Detritus	4-14
4.6 General Discussion	4-14
 Chapter 5 THE FLOODPLAIN SIMULATION MODEL	 5-1
5.1 An Idealization of the Magela Floodplain	5-1
5.1.1 Hydrological Sub-Model	5-1
5.1.2 Plant Growth Sub-Model	5-2
5.1.3 Detritus Sub-Model	5-3
5.1.4 Sediment Sub-Model	5-3
5.1.5 Effects of Increased Ionic Strength	5-3
5.1.6 Radium Uptake by Water Buffalo	5-4
5.1.7 Pollution Sub-Model	5-4
5.1.8 Radium Transfer to Man	5-5
5.2 Equilibrium Calculations	5-5
5.2.1 The Completely Mixed Model	5-5
5.2.2 The Completely Mixed Input-Output Model	5-6
5.3 The Kinetic Model	5-7
5.3.1 Computational Methods	5-9
 Chapter 6 PERFORMANCE OF THE MODEL	 6-1
6.1 Introduction	6-1
6.2 Validation Against Natural Radium Concentration	6-1
6.3 Validation Against Natural Radium Distribution Pattern	6-3
6.4 The Accumulation Hypothesis	6-3
6.5 The Linearity Hypothesis	6-3

6.5.1 A Multi-Factorial Experiment	6-4
6.5.2 Results of the Factorial Experiment	6-5
6.5.3 Influence of Individual Variables	6-8
6.5.3.1 Species	6-9
6.5.3.2 Biomass	6-9
6.5.3.3 Length of the Wet Season	6-10
6.5.3.4 Bioturbation	6-10
6.5.3.5 Erosion	6-10
6.5.3.6 Ionic Strength	6-10
6.5.3.7 Sediment K_d	6-10
6.5.3.8 Extra Rainfall	6-10
6.5.3.9 Pollution Type	6-11
6.6 The Role of Plants and Sediment in the Fate of Radium	6-11
6.7 Sensitivity Analysis	6-11
6.8 Conclusion	6-12
 Chapter 7 RADIATION DOSE ASSESSMENT	 7-1
7.1 Environmental Protection Philosophy in the Nuclear Industry	7-1
7.2 Dose Assessment Models for the Alligator Rivers Region	7-3
7.3 A Derived Limit for the Buffalo Grazing Food Chain	7-3
7.4 Application of the Derived Limit	7-5
 Chapter 8 SIMULATION AND THE MONITORING PROGRAM	 8-1
8.1 Introduction	8-1
8.2 The Natural Background	8-1
8.2.1 Pre-Operational Radium Concentration in Water	8-1
8.2.2 Ecosystem Stability	8-2
8.2.3 Operational Radium Concentration in Water	8-3
8.2.4 Soil Monitoring Results	8-3
8.2.5 Food Monitoring Results	8-3
8.3 Sensitivity of Analytical Methods	8-4
8.4 Simulation of Waste Water Disposal	8-4
8.5 Conclusion	8-5
 Chapter 9 DISCUSSION AND CONCLUSIONS	 9-1
9.1 The Accumulation Hypothesis	9-1
9.2 The Linearity Hypothesis	9-1
9.3 The Role of Plants and Sediment in the Fate of Radium	9-1
9.4 Evaluation of Methods	9-2
9.4.1 The Minimum Spanning Tree Sampling Method	9-2
9.4.2 The Nymphaea Growth Model	9-2
9.4.3 Uptake-and-Loss Experiments in a Closed System	9-3
9.4.4 Radium-226 Analysis by Liquid Scintillation Counting	9-4
9.4.5 Air-Photo Study of Ecosystem Stability	9-4
9.4.6 General Error Model for Radium-226 Analysis	9-4

9.3.7 Computational Experiments

9-4

LITERATURE CITED

10-1

APPENDICES

- A.1 Radium, An Historical Introduction
- A.2 Pre-Mining Radium Distribution on the Magela Floodplain
- A.3 Growth and Productivity of the Water Lily *Nymphaea violacea*
- A.4 Radium Uptake by Aquatic Plants (review paper)
- A.5 Radium Analysis and Sample Preparation Methods
- A.6 Computer Program Used to Implement the Model
- A.7 Stability of vegetation on the Magela floodplain
- A.8 Error Analysis: Evaluation of Interlaboratory Comparisons
Against a Precision Model

SUMMARY

The Magela floodplain, in the Northern Territory of Australia, drains two large uranium deposits: Ranger, where mining began in 1980, and Jabiluka, which is undeveloped. The floodplain is a seasonal freshwater lagoon in which dense stands of aquatic plants grow during the annual wet season and through much of the dry season. Radium (Ra) is a waste product from uranium mining that may enter the floodplain through leaching or seepage from the mine site or through accidental or controlled release of waste waters. No Ra pollution of Magela Creek has yet been detected but preliminary studies suggested that small amounts could be a significant health hazard to local Aborigines who drink creek water and eat a variety of aquatic foods including water buffalo that graze on the floodplain. If effluent Ra does enter the floodplain it may be flushed out to sea by subsequent floodwater or, more likely, be taken up onto vegetation or bed sediment. The vegetation may, in turn, remobilize Ra from the sediment into the water column. The purpose of the present work was to investigate the relative importance of these processes and to predict the potential for Ra transfer through the buffalo-human food-chain.

The study began with a survey of the natural Ra distribution in soil, plants and buffalo faeces on the floodplain in 1975-76 before mining began in 1980. This showed a natural accumulation of Ra at the beginning of the floodplain and from this it was predicted that effluent Ra may behave in a similar way and be held up in the floodplain rather than being flushed out to sea. Laboratory experiments were then carried out on the major plant species from the floodplain, the semi-aquatic grass *Pseudoraphis spinescens*, and the water lily *Nymphaea violacea* (the latter studied by my colleague J.R.Twining), to measure the rates of Ra exchange between the water column, the plants and the sediment.

A computer model was constructed from these data to simulate Ra transfer through the floodplain. The model consisted of water flow from a monsoonal rainfall pattern over a simplified catchment and through a rectangular floodplain divided into 12 square "plainettes" (3x3 km each). The water carried natural Ra from the catchment and as it flowed through each plainette it exchanged Ra with plants, detritus, suspended sediment, bed sediment and subsoil at rates determined from the experimental and field data. Hypotheses about the fate of Ra were tested by manipulating the state variables and introducing simulated effluent into the input water.

These experiments with the model identified direct Ra uptake from the water column onto plants and bed sediment as the major process in Ra transport through the system.

Approximately similar amounts of Ra were taken up by these two components and a small proportion remained in the water column. As a result, nearly all the effluent Ra was taken up in the first 10 kilometres (the floodplain is 36 kilometres long) as predicted from the field survey. There was negligible translocation of Ra within the plants (determined from the laboratory experiments) and Ra uptake onto suspended sediment was not significant because its mass was small compared with the masses of bed sediment and plants.

When effluent Ra was introduced into the model and traced through the buffalo-human food-chain, a fourfold increase in Ra concentration in water over the entire wet season led to an average 2-fold increase in the human dietary intake of Ra. Increases in bioturbation and effluent salinity strongly suppressed Ra transfer through the food-chain. Variations in plant species or biomass or variations in the adsorption capacity of the sediment had only a small effect. Variations in rainfall and suspended sediment load had negligible effect on the fate of Ra. These effluent simulation experiments predicted that the Ranger mine could dispose of a million cubic metres of Ra-contaminated waste water over a 10-day period in the early part of the wet season without significant impact on the local Aborigines or the environment.

STATEMENT

This thesis contains no material that has been submitted to any other university or institution for a higher degree. The information herein is the result of my own research except where acknowledgement of others is given.

A handwritten signature in black ink, reading "Alex R Williams", followed by a small flourish.

Alexander R Williams

Australian Nuclear Science and Technology Organisation
Lucas Heights Research Laboratories
Private Bag No.1, Menai NSW 2234.

ACKNOWLEDGEMENTS

I wish to thank my colleague John Twining for carrying out technical aspects of the first preliminary experiment, for assisting with field work in 1981, 1983 and 1984, for sharing his equipment with me and for generally facilitating our working relationship with his generous and cheerful manner. The Office of the Supervising Scientist provided travel funds, accommodation and some equipment for field work. Chemical analyses of the field survey samples were carried out by B. G. O'Brien, K. F. Mears, the former Analytical Chemistry Section of the AAEC and the Australian Mineral Development Laboratory, Adelaide. Des Davy supervised my work at Lucas Heights, Associate Professor E.W.R. Barlow supervised my work at Macquarie University and they both provided useful comments on early drafts. Dr. Ross Jeffree, Max Giles, Dr. Arthur Johnston (OSS) and Associate Professor Mark Westoby (Macquarie University) also provided comments on several parts of the work. Dr. Jerrard Barry gave advice on numerical methods and he and Stephen Wong gave assistance in debugging the computer program. Celia Chrimes typed some of the figure captions.

"Behold, BEHEMOTH ...
under the lotus plants he lies,
in the covert of the reeds
and in the marsh".
Job 40:15,21