# INVESTIGATIONS INTO THE CONCENTRATION RATIOS OF SELECTED RADIONUCLIDES IN AQUATIC ECOSYSTEMS AFFECTED BY MINE DRAINAGE EFFLUENTS WITH REFERENCE TO THE STUDY OF POTENTIAL PATHWAYS TO MAN

REPORT TO THE WATER RESEARCH COMMISSION

By

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The Steering Committee for this project consisted of the following persons:

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# EXECUTIVE SUMMARY

# INVESTIGATIONS INTO THE CONCENTRATION RATIOS OF SELECTED RADIONUCLIDES IN AQUATIC ECOSYSTEMS AFFECTED BY MINE DRAINAGE EFFLUENTS WITH REFERENCE TO THE STUDY OF POTENTIAL PATHWAYS TO MAN

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## EXECUTIVE SUMMARY

### Background and Motivation

South Africa is endowed with a vast wealth of mineral resources typified by the extensive gold deposits in the Witwatersrand region. In contrast, water resources are limited and are vulnerable to environmental impacts from the mining industry.

In the processing of South African gold-ores, uranium occurs as a by-product while several other radioactive isotopes from the uranium and thorium decay chains notably $^{226}$Ra are also present. The average uranium concentration in the ores (< 0,01%) is at least an order of magnitude lower than that mined in the rest of the world. The large volumes of ore processed for gold production, however, result in an appreciable production of uranium. Gold mining on the Witwatersrand has been in progress since 1884 and by 1987 a total of $3 \times 10^9$t of tailings were produced, covering a surface area of about 6500 ha. These tailings contain about 120 000t of unrecovered uranium whilst the radium residues of a further 30 000t of recovered uranium have been dumped with milled residue on slimes dams since 1952.

Because the volume of solid wastes in tailings is so immensely large, it is impossible to isolate these tailings from the environment over a prolonged period of time (de Jesus *et al.*, 1987). In addition, these mining activities and tailings on the Witwatersrand are in the close proximity to densely populated areas as well as agricultural land. These mines and tailings are spread over an 80 km long strip with approximately 3,8 million people living within 20 km of this area. It is therefore unavoidable that radium and uranium contaminated leachate may reach dams, lakes and streams which are accessible to either the public or agricultural land. Contamination of surface waters such as lakes and rivers may also occur by direct discharges of large volumes of radioactive process water or underground mine water into such water bodies. Farming practices can transfer contaminants to the terrestrial environment and thus to livestock, crops and vegetables. In turn, persons may be exposed to various radioactive contaminants. Uranium is known to have both radiotoxic and chemotoxic properties. $^{226}$Ra is a daughter product of uranium with a physical half-life of 1600 years and being a boneseeker, has a biological half-life of 48 years. It has a high degree of radiotoxicity.

A number of local studies have in the past been conducted to determine the occurrence of $^{226}$Ra in the environment as well as in agricultural produce (Malan, 1981; de Jesus *et al.*, 1987; Funke, 1990). However, none of these were aimed at determining the Concentration Ratios (CR) for transfer of these radionuclides to crops and livestock and the subsequent potential dose to humans.

The studies reported here inter-alia address these shortcomings for both uranium and radium. The need for investigation of such environmental impacts in South Africa has in recent years received fresh impetus by the requirements of the Council for Nuclear Safety, who is licensing affected mines under the Nuclear Energy Act and the Government Mining Engineer, who requires an Environment Management Review Plan under the Minerals Act.

## Objectives and Scope

The investigation had the following objectives:

- Selection of suitable localities where $^{226}$Ra and uranium might be present in significant concentrations. Such localities are those subject to mine drainage effluents which are in close proximity to gold and uranium mines as well as agricultural areas located in the immediate vicinity of such mines in the greater Witwatersrand area.
- Monitoring of these radionuclides from the point of release and, where possible, via aquatic to terrestrial ecosystems.
- Determination of the levels of these radionuclides in water, soil and agricultural products such as vegetables, milk and meat from farms potentially affected by contaminated effluents from mines and calculation of relevant concentration ratios.
- Experimental study to determine with different irrigation schemes the amount of $^{226}$Ra uptake through the leaves and roots of two vegetables. For the purpose of this experiment, two commercial strains of beetroot and cabbage were chosen.
- Calculation of the potential dose to man via different pathways using mathematical models.

The localities studied were limited to non-mining property except in isolated cases. The emphasis was on the aquatic pathway of release from mining property. The contribution of contaminated dust and radon gas emanation from mine dumps to the radiologic dose of the public was not investigated and merits a separate investigation.

## Results and Conclusions

A practical difficulty in the project was to identify areas with the combination of agricultural production, irrigation with mine-related water and that concentrations of radionuclides in the water be significant. Only a few meeting all the criteria were identified. One could conclude for the Witwatersrand area that no major agricultural production occurs with significantly contaminated water. Individual localities probably exist where concentrations could be high or could in future be high if mine effluent loads increase.

The high cost of radionuclide analysis particularly for $^{226}$Ra did not allow, in some cases, sufficient samples to be collected and analyzed to provide adequate statistical confidence in comparisons between sites and between countries.

The data available for concentration of radium and uranium in water from the localities sampled indicates levels generally well below guideline values . Luipaardsvlei, however, had uranium values somewhat above the guideline value of 44 $\mu g.\ell^{-1}$. Concentration values for uranium in soil were variable and above background reference levels in the USA.

The radium values for rooted emergent aquatic macrophytes indicate that such plants are scavenging radium from the reducing conditions in anoxic sediments. Concentrations of uranium and radium in aquatic plants and fish were generally lower than in other uranium mining countries, which correlates with the much lower ore concentrations locally.

The vegetable concentration ratio values (water) were generally greater than those relative to soil. This is, however, not an indication that more radium or uranium is taken up from the water, but just that the concentrations in the water are less than in the soil. In this study the highest concentration used for irrigation by a farmer was of the order of one hundred times lower than the radium concentrations in the soil. The crop uptake is more strongly dependent on the soil concentration than that in the water. The expected buildup of radium in soil due to irrigation is anticipated to be negligible. The concept of concentration ratios is useful but has a limitation particularly the lack of easily interpreted physical meaning. It should preferably be defined together with other field variables.

The concentration of uranium in farm grown vegetables had values generally greater than those from background areas in other countries. The radium values in the vegetables were similar to overseas values and in the case of lettuce, much lower. A statistically valid background site of date was not established in the study, so the high trend in uranium concentration can not be placed into perspective.

The dose assessment results for reasonably realistic, yet conservative assumptions for water and vegetable consumption show that doses to a maximum will be of the order of 1% of the annual limit of 250 μSv. Radium, as expected, has a higher contribution than uranium. If the drinking water radium concentration were a factor 5 higher, i.e. equal to the current EPA(USA) limit of approximately 0,2 $Bq.\ell^{-1}$ then the annual commited effective dose dose would be 39 μSv, still well below the dose limit. Only infrequently, and then very close to mine properties, have such concentrations been observed locally. Similarly a factor 5 or 20 increase in vegetable concentrations will still leave a good margin below the limit. Obviously, production of crops on mine dumps should be prohibited or strictly controlled as evidenced by the individual results from oats production.

The data collected in this study in the Witwatersrand area shows that no member of the public is expected to be exposed via aquatic pathways to any significant dose. If mine effluents are controlled to less than 0,2 $Bq.\ell^{-1}$ radium and less than 0,5 $Bq.\ell^{-1}$ uranium, the doses received from various aquatic pathways should be well within dose limits set. The concentration values quoted here are not intended to imply these should be concentration limits. Control limits are set by CNS on site specific assessments. The collective dose to the high density population of the Witwatersrand on a whole was not considered in this study but for the aquatic pathway it is expected to be low.

The contract objectives have largely been met. Localities where irrigation of agricultural crops with contaminated mine water were identified. There could, however, be affected sites that were not identified in the Witwatersrand area but the general trend indicates no major problem.

Various aquatic fauna and flora were sampled together with available agricultural crops. In many cases concentration ratios could be derived. The major limitation in the project however was the constraint imposed on the number of samples analyzed due to the high cost of radionuclide analyses. This limitation prevented in some cases statistically robust averages or trends to be determined. Relevant generalisation could however be made.

The pot experiment yielded useful data on radium uptake in roots and leaves of selected vegetables for flood and spray irrigation. Various aspects can be deduced from the pot experiments:

- relative to high concentrations of radium in the water the results show that the leaves of both beetroot and cabbage have about the same CR, while the roots of cabbage have a greater CR than those of beetroot.
- the difference between the effects of root only (or flood) irrigation and root plus leaf (or flood plus spray) irrigation is greatest for the beetroot where there is about x2 increase in soil CR and x4 increase in water CR.
- there is a transfer of radium from the leaves of the beetroot to its roots with spray irrigation.
- the actual concentration in the beetroot vegetable is greater than that in the cabbage.

The conclusion is that beetroot (and possibly other root crops) are more vulnerable to radium contamination than cabbage (or leaf crops) and that flood irrigation only is preferred to limit possible uptake. The latter is relevant to high water concentrations of radium but may not be the case for much lower concentrations found in practice.

The objective of modelling the dose to man was achieved and indicates the important role of radium in the water pathway.

On the zoological and botanical front, the concentration values for radium and uranium in certain species are the first reported in the literature. The relative concentrations in the aquatic food chain algae - fish - birds is of interest.

The further knowledge gained will be useful to mines that have to licence their operation in terms of the Council for Nuclear Safety's regulations under the Nuclear Energy Act. The general environmental impact studies (EMPR) required by the Minerals Act will also benefit from this study. With the growing population density in the Witwatersrand region the pressures for agricultural production and housing will encroach increasingly existing mine boundaries. Land users and planners will have to allow for the possibility of occupying contaminated land and water. This study alerts developers and mine owners to do preliminary studies on radionuclide levels in water, sediments and land, particularly where agricultural production occurs. This will assure the public that potential doses are well within limits.

## ABSTRACT

Investigations were made into the occurrence and concentration values of $^{226}$Ra and uranium in some gold and uranium mine polluted aquatic environments in the Transvaal. An overview is given on aspects of the possible environmental effects of $^{226}$Ra and uranium as well as background data on factors affecting the biological uptake and transport of both radionuclides in some terrestrial and aquatic organisms. A brief discussion is given on the prevailing physical and chemical conditions of the irrigation water and agricultural soils at three localities where analyses were made for the presence and concentrations of both radionuclides in the water, in stream sediments, in selected aquatic plants, in a decapod macro-invertebrate, in fish and in water birds. In order to evaluate the potential effects of these radionuclides on man, studies were also conducted on the concentration ratios of $^{226}$Ra and uranium in these organisms and the abiotic environment in which they occur, including selected vegetable crops irrigated with mine polluted river water. The experimental uptake of radium by beetroot and cabbage under controlled environmental conditions was also investigated. These data were then used in a dose assessment model looking at various potential pathways of both radionuclides to man including via soil, drinking water, vegetables, a cereal and fish.

Field results obtained showed that in virtually all cases the presence and concentrations recorded for both radionuclides were (and possibly are also at other affected areas) at least an order of magnitude lower than concentrations found in countries such as Japan, Germany and the USA, and that in all cases concentration values found for both radionuclides in the present study fall below the maximum recommended guideline values laid down by the Council for Nuclear Safety (CNS) of South Africa.

The dose assessments for the scenarios chosen indicate that the annual effective dose for uranium and radium is a fraction of the maximum allowable limit for members of the public. However it is possible that certain site specific concentration values may require controls to limit exposure.

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## 1 INTRODUCTION

Gold mining on the Witwatersrand has been in progress since 1884 and by 1987 a total of $3 \times 10^9$t of tailings were produced, covering a surface area of about 6500 ha (de Jesus *et al.*, 1987). These tailings contain about 120 000t of unrecovered uranium whilst the radium residues of a further 30 000t of recovered uranium have been dumped with milled residue on slimes dams since 1952.

In the processing of South African gold-ores, uranium occurs as a by-product (Feather & Koen, 1975; Wittmann & Förstner, 1977; Malan, 1981) while several other radioactive isotopes from the uranium and thorium decay chains are also present. The average uranium concentration in the ores (< 0,01%) is at least an order of magnitude lower than that mined in the rest of the world (Funke, 1990). The large volumes of ore processed for gold production, however, result in an appreciable production of uranium. As a result, South Africa was the third largest producer of uranium in the world in 1987 with a quantity of approximately 5000t of $U_3O_8$. Although the uranium production of South Africa decreased to approximately 2000t in 1992, it is at this level still appreciable in volume (Ainslie, 1992).

If an equilibrium exists in the natural decay chain of $^{238}U$, every kilogram of $^{238}U$ is associated with $3{,}3 \times 10^{-4}$g of $^{226}Ra$. As there is no present economic use for radium, the $^{226}Ra$ occurring with the uranium is usually returned to the tailings dams. $^{226}Ra$ has a half-life of 1620 years and decays to $^{222}Rn$ (radon), which is a gas and which can then emanate from slimes dams. $^{222}Rn$ can have detrimental effects on man when its solid radioactive decay products ($^{218}Po$, $^{214}Pb$, $^{214}Bi$, $^{214}Po$, $^{210}Po$) attach themselves to airborne particles which can be inhaled. It is estimated that these radioactive particles are by far the largest contributor of total radiation exposure to man (90% and over) (Funke, 1990). However, because radon gas has a short half-life of 3,8 days, most of it decays into solid decay products before it reaches the surfaces of slimes dams (Funke, 1990). The total amount of $^{226}Ra$ associated with the recovered or unrecovered uranium is approximately 50 000g, or an equivalent of $1{,}85 \times 10^{15}$Bq.

Because the volume of solid wastes in tailings is so immensely large, it is impossible to isolate these tailings from the environment over a prolonged period of time (de Jesus *et al.*, 1987). In addition, these mining activities and tailings on the Witwatersrand are in the close proximity to densely populated areas as well as agricultural land. These mines and tailings are spread over an 80 km-long strip with approximately 3,8 million people living within 20 km of this area. It is therefore unavoidable that radium and uranium contaminated leachate may reach dams, lakes and streams which are accessible to either the public or agricultural land. Contamination of surface waters such as lakes and rivers may also occur by direct discharges of large volumes of radioactive process water or underground mine water into such water bodies. Affected streams usually bypass tailings dams which have a normal adsorption capacity for $^{226}Ra$ (Funke, 1990).

It is therefore inevitable that radioactive material can enter aquatic ecosystems such as lakes, rivers and wetlands in the Transvaal. These radionuclides can furthermore be present in irrigation water pumped from these water bodies when it is utilized by farmers and, in the process, are transferred to the terrestrial environment. Once in the terrestrial environment, these contaminants can then be taken up by livestock or accumulated by crops and vegetables via the soil.

$^{226}$Ra, being an alpha emitter and a bone seeker with a biological half-life of 48 years, is known to possess a high degree of radiotoxicity and exhibits a health hazard when it enters the human body either by ingestion or inhalation (Neuman *et al.*, 1955). Uranium is also known to have both radiotoxic and chemotoxic properties.

Taking into account the potential radiotoxicity of $^{226}$Ra and uranium, some local studies have in the past been conducted to determine the occurrence of $^{226}$Ra in the environment as well as in agricultural produce (Malan, 1981; de Jesus *et al.*, 1987; Funke, 1990). However, none of these were aimed at determining the Concentration Ratios (CR) for transfer of these radionuclides to crops and livestock and the subsequent potential dose to humans.

This project was initiated on the basis of this background, and with the following research aims in mind:

- **Selection of suitable localities where $^{226}$Ra and uranium might be present in significant concentrations.** Such localities are those subject to mine drainage effluents which are in close proximity to gold and uranium mines as well as agricultural areas located in the immediate vicinity of such mines in the greater Witwatersrand area.

- **Monitoring of these radionuclides from the point of release and, where possible, via aquatic to terrestrial ecosystems.**

- **Determination of the levels of these radionuclides in water, soil and agricultural products such as vegetables, milk and meat from farms potentially affected by contaminated effluents from mines and calculation of relevant concentration ratios.**

- **Experimental study to determine with different irrigation schemes the amount of $^{226}$Ra uptake through the leaves and roots of two vegetables.** For the purpose of this experiment, two commercial strains of beetroot and cabbage were chosen.

- **Calculation of the potential dose to man via different pathways using mathematical models.**

# 2 LITERATURE REVIEW

## 2.1 Uranium resources in South Africa

Minute amounts of uranium are present in every type of rock and soil in the earth's crust. The most extensive uranium deposits are mostly found in South Africa, Zaire, France, Czechoslovakia, Australia, Brazil, Canada and USA (Fisher, 1988). South Africa has the third largest uranium resource (294 000 t) in the Western World. An estimated 68% of the South African low cost reserves occurs in the Witwatersrand conglomerates and tailing dumps, 29% in Karoo sandstone and coal deposits and 3% in the Phalaborwa complex (Ainslie, 1993). Estimates on the average content of uranium in the Witwatersrand ores (usually uraninite) vary between 0,05 and 0,2 kg $U_3O_8.t^{-1}$ ore (Smit, 1989).

The auriferous conglomerates in the four contiguous Precambrian systems, namely the Dominion Reef, Witwatersrand, Ventersdorp and Transvaal systems contain the major concentrations of uranium minerals in South Africa. Of these, the Dominion Reef and Witwatersrand complex are the most important uranium resources. Although not geologically conformable, these four systems together cover tens of thousands of square kilometres in the Transvaal and the Orange Free State (von Backström, 1976).

## 2.2 Nuclear properties of uranium

The element Uranium, named after the planet Uranus which was observed only a few years earlier, was discovered in 1789 in pitchblende samples by the German metallurgist and chemist, Klaproth. Uranium-238 has a long half life of 4,5 x $10^9$ years. Because of its low specific (alpha only) activity, uranium itself is regarded as having a relatively low radiotoxicity. Its radionuclides, however, decay into a complex series of isotopes of the elements thorium, protactinium, radium, radon, polonium, bismuth, thallium and lead, some of which are significant sources of natural radiation in the environment, or in the human body, following its dietary intake (Long, 1987).

## 2.3 Uptake, metabolism and physiology of uranium in plants, animals and humans

### 2.3.1 Effects on micro-organisms and plants

According to Burkart (1991) no information exists on the chemotoxic or radiotoxic effects from natural and industrial sources of U, Th and their decay products on micro-organisms or plants.

### 2.3.2 Non - radiological effects of uranium in humans

The chemical form and solubility in body fluids of uranium determines the biokinetics and metabolism of this element in man (Scott, 1973). The compounds $UF_4$, $UO_2$, $UO_4(NH_4)_2U_2O_7$, $UO_3$ and uranyl nitrates are moderately transportable in the human body, while $U_3O_8$ and uranium hydrides and carbides are mostly insoluble and only slightly transportable (Morrow *et al.*, 1966). Biologically soluble uranium is absorbed into the blood from where it is filtered out by the kidneys and excreted via the urine.

A certain amount of uranium present in the blood stream is also taken up by the skeleton where it first deposits on the bone surface, but later redistributes in the bone volume (Bernard & Struxness, 1957). Uranium also tends to deposit in soft tissues in the body, especially in the kidneys. The metabolism and excretion of hexavalent uranium by the kidney has been described by Morrow *et al.* (1982). According to Hodge (1973) high doses of uranium exposure to man also has an effect on the blood vasculature throughout the body. Because of the effects of uranium on the blood chemistry, capillary permeability, blood pressure and edema may increase while the clotting ability of blood may decline. Minor damage in liver and muscle tissue may also be induced by uranium. The effects of uranium on the nervous system may be similar to those from poisoning by other metals (Yuile, 1973).

### 2.3.3 Somatic and hereditary effects of radiation

Besides its chemotoxic properties uranium is also radiotoxic. All substances known to emit ionizing radiation are considered as potentially inducing somatic or hereditary effects (International Commission on Radiological Protection (ICRP) ,1977). Hereditary or mutagenic effects affect the descendants of an exposed individual while somatic effects occur in the individual himself. Non-malignant damage to the skin, minor damage to tissue, carcinogenic effects and teratogenic effects are examples of somatic effects. Teratogenic effects are induced at high doses of ionizing radiation (Coggle, 1983). It may also result in developmental defects leading to impaired brain functions such as severe mental retardation. A case which clearly illustrates this phenomenon was the occurrence of severe mental retardation among the prenatally exposed survivors of the atomic explosions at Hiroshima and Nagasaki during the second world war (Otake *et al.*, 1987). This is, however, an example of severe radiation effects. Somatic as well as hereditary effects of radiation are usually considered as stochastic or deterministic (ICRP,1977). Deterministic (or non-stochastic) effects are those where the severity of the effect may vary with dose, and a threshold thus exists below which the effect cannot be determined. Stochastic effects, on the other hand, are those where the probability of an effect occurring is a function of the dose (without threshold).

Although uranium is known to be chemotoxic and radiotoxic, the unstable decay daughter isotopes of this element are more hazardous, both in the occupational and non-occupational environments as they are major contributors to the total radiation dose (Burkart, 1991). The ICRP (1991) suggested an annual limit for a radiation worker on intake of uranium via ingestion as high as $3 \times 10^6$ Bq or about 200 g of natural uranium. This statement by the ICRP (1990) is seen by many as conservative.

## 2.4 Tolerance levels and limiting concentrations of uranium

### 2.4.1 Normal daily intake and assessment of uranium levels in food groups

According to Wrenn *et al.* (1985), the average dietary intake of uranium from foods and water is about 1,75 $\mu g.day^{-1}$, while the population-weighted average for drinking water in the United States is 1,2 $\mu g.\ell^{-1}$ (Lappenbush & Cothern, 1985). ICRP (1975) assumes that a standard man has a daily intake of uranium via food and fluids of 1,9 $\mu g$.

A comprehensive investigation on the uranium content of human dietary items was conducted by Welford & Baird (1967). This study was primarily aimed at establishing typical levels of uranium intake by man and was conducted for typical diets in New York City, Chicago and San Francisco (Table 2.1). These results showed that although there were geographical variations in uranium content of individual foods, the total annual intake for the three cities were nearly similar. More than 70% of the annual intake of foodstuffs came from potatoes, meat, fresh vegetables and bakery products. Based on these figures, the calculated daily intake of uranium was 1,3 $\mu g.day^{-1}$ for New York, 1,4 $\mu g.day^{-1}$ for Chicago and 1,3 $\mu g.day^{-1}$ for San Francisco.

Hamilton (1972) determined the uranium content of a variety of foodstuffs representative of the United Kingdom diet including cereals, vegetables, meat, eggs, fish and milk. The average intake (g/person/day) is also represented while the annual intake ($\mu$g) and percentage annual intake was calculated. Results show that starchy roots (138 $\mu$g : 35%) represent the biggest portion of annual intake of uranium, while items such as eggs (3 $\mu$g : 0,9%), fish (3 $\mu$g : 0,8%) and milk (2 $\mu$g : 0,5%) provided the lowest portions of the annual intake.

### 2.4.2 Chemical toxicity threshold to uranium

Although little data are available on humans, extensive research on animals has been conducted on the threshold tolerance limits for exposure to uranium. The renal injury threshold is estimated at 10 $\mu$g U (as $UO_2F_2$).$kg^{-1}$ (body weight) for dogs and about 100 $\mu$g $U.kg^{-1}$ for rats (Morrow *et al.*, 1982). The threshold level for man is estimated to be between those of rats and dogs, being about 70 $\mu g.kg^{-1}$ (body weight). This estimated value corresponds to a renal injury threshold concentration value of 16,3 $\mu$g $U.g^{-1}$ kidney tissue in man. Morrow *et al.* (1982) however found that a steady-state concentration of 3 $\mu$g $UO_2F_2.g^{-1}$ (tissue) was sufficient to produce the typical symptoms of uranium poisoning in animal experiments. These results therefore raised questions on the suitability from a chemical point of view of currently recommended exposure limits.

The chemical toxicity of uranium varies according to its compound form. While insoluble uranium compounds are the least toxic, toxicity increases with chemical solubility. According to Morrow *et al.* (1982), the most toxic uranium compound is $UO_2F_2$, possibly due to the additive effects of exposure to hydroflouric acid (HF) which originates from the original uranium compound in solution.

### 2.4.3 Summary

Uranium is an ubiquitous element found primarily in high concentrations in primordial sources, phosphate fertilizers and the nuclear fuel cycle. The major transfer-pathway of uranium into crops seems to be by root uptake with concentration ratios for soil ranging between $10^{-3}$ and $10^{-4}$ (Long, 1987). The concentration of uranium in food materials is therefore commonly low but it is known that uranium levels are more significant in root and leafy vegetables than in meat, fish and milk (Long, 1987). Although substantial information is available on the occupational exposure and intake of uranium and uranium compounds, little information is available on the role of uranium in food chains and more studies are needed on this subject.

Table 2.1: Annual intake of uranium through various agricultural crops from New York, Chicago and San Francisco. (After Welford & Baird, 1967).

| Group | Uranium intake $\mu g.year^{-1}$ | | |
|---|---|---|---|
| | New York | Chicago | San Francisco |
| Bakery products | 66 | 58 | 58 |
| Grain products | 16 | 16 | 16 |
| Eggs | 3,4 | 3,4 | 3,4 |
| Fresh vegetables | 25 | 25 | 44 |
| Root vegetables | 12 | 9,4 | 12 |
| Milk | 16 | 31 | 62 |
| Poultry | 2,7 | 8,4 | 5,6 |
| Fresh fish | 3,4 | 6,8 | 4,5 |
| Flour | 23 | 14 | 8,4 |
| Macaroni | 1,2 | 1,4 | 1,9 |
| Rice | 5,8 | 18 | 4,3 |
| Meat | 79 | 104 | 46 |
| Shellfish | 9,5 | 29 | 31 |
| Dried beans | 4,5 | 11 | 11 |
| Fresh fruit | 76 | 76 | 42 |
| Potatoes | 111 | 101 | 101 |
| Canned fruit | 4,2 | 6,4 | 4,0 |
| Fruit juices | 1,1 | 1,1 | 3,4 |
| Canned vegetables | 4,0 | 2,0 | 2,0 |
| **Total** | 463,8 | 522,9 | 462,5 |

Figures based on average food consumption (USDA ,1955).

## 2.5 Historical introduction and overview of radium

### 2.5.1 Nuclear properties of radium

In 1898 Pierre and Marie Curie discovered the new element polonium and "a new element with very curious properties" (Curie *et al.*, 1898) which was found to be nine hundred times (later found to be over a million times) more radioactive than uranium. It was also found to be chemically similar to element 56 in group IIA, Barium. This element was named "radium" after the term "radioactivity" which was used to describe the spontaneous disintegration of matter.

Altogether twenty-five isotopes of radium have been identified. The most abundant of these naturally occurring isotopes are $^{226}$Ra ($^{238}$U daughter : α-emitter : half life of 1622 years) and $^{228}$Ra($^{232}$Th daughter (mesothorium) : ß-emitter : half life of 5,8 years) (Williams & Kirchmann, 1990).

### 2.5.2 Uses, health hazards and environmental effects of radium

Radium was initially utilized as a source of radioactivity for industrial and medical radiography. It also provided valuable information about radioactivity and the structure of matter. In the field of medicine, radium was especially used in the treatment of cancer.

Although the applications of radium provided a benefit to medicine and industry, the hidden dangers of this element soon became apparent. Following the subsequent illness of Marie Curie after handling quantities of this element, a New York dentist described in 1924 a disease diagnosed in a luminescent dial painter as "radium jaw". This was followed by a description of anaemia resulting from occupational poisoning (Martland *et al.*, 1925) and five deaths from "jaw necrosis" in a factory (Flinn, 1926). Due to its alkaline earth behaviour, $^{226}$Ra intake leads to its deposition in the human skeleton (Burkart, 1991). Following these deaths and many other radium poisoning related cases, the International Committee on X-Ray and Radium Protection, the forerunner of the present ICRP was established. At that time (1940), only 1,4 kg of radium had been extracted from the earth, but it was responsible for the deaths of more than one hundred people (Williams & Kirchmann, 1990).

A few years after the discovery of radium in uranium ores, it was commonly found in the environment. Tsivoglou *et al.* (1958) was the first to identify radium as an environmental pollutant from the uranium industry during investigations in the Colorado Plateau area in the USA. Since similar reports by Havlik (1970) in Czechoslovakia and Kirchmann *et al.* (1973) in Belgium, radium contamination of the aquatic and terrestrial environment originating from uranium, gold and phosphate mining and milling has been identified in many areas over the world, including South Africa.

The decay product of $^{226}$Ra, $^{222}$Rn as well as its short lived decay products are mostly found in mines and the indoor environment where $^{222}$Rn occurs as a gas. Currently the best quantified occupational disease is the link between $^{222}$Rn decay products in mines and the occurrence of lung cancer amongst mine workers (Thomas *et al.*, 1985 ;

McMichael, 1989).

## 2.6 Biological uptake and transport of radium by aquatic plants and animals

### 2.6.1 The mechanisms of radium uptake and transport

Due to the similar chemical properties of radium and calcium as well as the other alkaline earth elements such as Mg, Sr and Ba, it can be anticipated that radium may be taken up by cells via the same mechanism as the latter elements (Williams, 1984). Accordingly there could be a correlation between radium uptake across cell membranes and/or its distribution within tissues with that of calcium (Williams, 1984).

Investigations conducted by Williams (1982) showed that among 40 samples of aquatic and semi-aquatic macrophytes which represented 19 species, there was no significant correlation between the distribution of $^{226}$Ra and the elements Ca, Mg, Na, Cu, Zn, Fe and S. A positive correlation existed, however, between $^{226}$Ra and Mn concentrations.

### 2.6.2 Aquatic plants

A certain number of qualitative and quantitative factors exist which either promote or constrain radium accumulation by aquatic plants, namely:

- mechanisms at cellular level,
- the rate and extent of radium accumulation by the whole plant,
- accumulation from sediment and translocation within plants and
- the role of plants in radium cycling within the aquatic environment (Williams, 1990).

It has, however, been stated by Coughtrey & Thorne (1983) that no attempt has yet been made to construct generalized dynamic models of accumulation and retention of radionuclides by aquatic plants. This is especially true for radium and where there is no information available, general principles are illustrated from relevant information on other trace metals such as radioactive Sr (Pally & Foulquier, 1983) and Ca.

### 2.6.3 Concentration ratios

All living organisms obtain their nutritional or elemental needs through the processes of ingestion, absorption and adsorption. Alternatively, these elements may be lost through exchange, excretion and decomposition (Conway *et al.*, 1974). The concentration of an element in aquatic and terrestrial organisms is usually expressed as a concentration ratio, i.e. the ratio between the amount of element per unit weight of organism compared to that of an equal weight of surrounding medium, being water, sediment or soil. However, a concentration ratio is only valid if a condition of equilibrium exists between the concentrations in the organism and in the water, sediment or soil. Therefore, under these conditions, the rate of elemental uptake is balanced by turnover or exchange of elements (Conway *et al.*, 1974). Another definition is provided by Hoffmann *et al.* (1977) which states that traditional food chain models require a plant/substrate transfer coefficient, referred to as a Concentration Ratio (CR). This ratio describes the amount of nuclide expected to enter a plant from its substrate, under equilibrium conditions.

If the definition of CR is accepted as a constant, rather than a function, the CR approach requires us to assume that the plant and substrate concentrations are linearly related. The line defining the relationship goes through the origin of the representative graph. This assumption implies that, under comparable conditions, the CR for an element is constant regardless of substrate concentration (Sheppard & Sheppard, 1985). However, it is a poor assumtion in some situations to accept that the plant and substrate concentrations are linearly related (Simon & Ibrahim, 1990). According to these authors the linearity assumption infers that the CR is the slope of the linear relationship of plant concentration as a function of soil concentration. However, the regression approach (i.e. the slope of plant concentration versus soil concentration in computing CR was found to be unsatisfactory by Gilbert & Simpson (1983) when there was any possibility that the data were lognormal rather than normal. From a statistical bias point of view the regression estimate of the CR is not a preferred estimation, even in the case of normally distributed data (Simon & Ibrahim, 1990). While examining uptake data for 12 biological systems, Williams (1982) came to the conclusion that the linearity assumption for the uptake of radium can only be validated for freshwater algae.

Williams (1982) observed that a non-linear uptake response by terrestrial plants to increased radium concentrations in the substrate may be one factor accounting for non-linear CR ratios. Sheppard & Sheppard (1985) also stated that the lack of correlation between plant and substrate concentrations could be explained if the element (radium or uranium) was mimicking an essential element. This would also, according to these authors, lead to a non-linear relationship between the CR and the substrate concentration at low concentrations. Sheppard & Sheppard (1985) also observed that uranium may behave as an essential element (mimicry) at low substrate concentrations. In the case of uranium, it was observed that if the CR value was used to predict plant concentrations from substrate uranium concentrations, it will underpredict plant concentrations if the substrate uranium concentration was less than 20 $\mu g.g^{-1}$.

It has been found (Epstein & Leggett, 1954 ; Fried & Shapiro, 1961) that competition for adsorption binding sites on root surfaces exists between alkaline earth elements. In soils with high concentrations of alkaline earth cations, radium uptake by plants may be supressed due to adsorption competition. A hypothesis on membrane protein oligomeric structure and transport function (Klingenberg, 1981), gave rise to the thought that radium and other alkaline earth elements are taken up by the same type of binding site, or "calcium gate" on the cell membrane (Williams, 1984). Rusanova (1964), Taskaev et al. (1977) and Marple (1980) also came to the same conclusion that alkaline earth competition may suppress radium adsorption, which is generally in line with the hypothesis of Klingenberg (1981). The presence of calcium in the soil can even supress the uptake of strontium and barium, and not only radium (Wallace & Romney, 1971).

Maas (1969) has shown with uptake by maize roots that the uptake rate of calcium or other alkaline earth ions increase asymptotically as a function of substrate concentration (or nutrient solution) towards a plateau value which may indicate saturation of the root surface binding sites and which therefore limits the uptake rate of elements.

Williams (1984) has shown that although the substrate radium concentration may never reach the saturation value required for binding sites on roots, these sites would always be saturated due to the concurrent contribution of calcium and other alkaline earth elements to these sites.

Available published data on the biological uptake of radium consist mostly of field survey results where the radium concentrations of the dietary items [Q] is compared to that of the supporting water medium or sediment [C]. From this, the bioconcentration factor (BF) or Concentration Ratio (CR) for radium can be calculated:

$CR = [Q] / [C]$ ---------- (1)

According to the ICRP (1979) and the United States Nuclear Regulatory Commission (USNRC) (1980), a possible linear relationship exists between [Q] and [C]. This implies that CR is independent of [C]. In the expression of results on the $^{226}Ra$ uptake by algae, aquatic vascular plants, insects, fish and mussels, the data are usually assembled in log[Q] vs. log[C] plots and the linearity of the graph tested using the standard error of regression.

Organisms like fish which are exposed to radium via water and food, and aquatic macrophytes which are exposed via water and sediments, are examples of organisms which take up this element from either or both of the two sources. If the concentration ratios are therefore independent of the concentrations in both exposure sources, then:

$[Q] = a_1[C_1] + a_2[C_2]$ ---------- (2)

A multiple regression of [Q] on $[C_1]$ and $[C_2]$, constrained through the origin of the graph, can facilitate the determination of the two concentration ratios, $a_1$ and $a_2$. This model has an important application to irrigation. Using this model, Myttenaere *et al.* (1969) was able to demonstrate that the transfer of Cs and Co to rice from irrigation water was 10-100 times greater than from soil. However, Williams (1981) states that there is insufficient data on radium available to describe its transfer during irrigation.

In the following example (algae), the goodness of fit of the regressions was estimated by the coefficient of determination $r^2$ which was expressed as a percentage. This provided an indication of the percentage of the variance that was accounted for by the regression. The statistical significance of the regression was taken from the square root of the coefficient of determination, which was the correlation coefficient, r. The significance was shown as a p-value, being the probability that the r value occurred by chance (Williams, 1984).

Data from the literature covers a concentration range of 7 orders of magnitude. A very good fit to the following regression was obtained:

$[Q] = 441[C]^{1,02}$ ---------- (3)

With: $r^2 = 94\%$ ($p << 0,001$)

The value of the regression coefficient ($b = 1,02$) is not significantly different from 1, it can therefore be concluded that uptake of Ra from water by algae is a linear process (Williams, 1984). The equivalent linear regression found was:

$[Q] = 432[C]$ ---------- (4)

giving a concentration ratio:

$CR = [Q] / [C] = 432$ --------- (5)

## 2.7 Biological uptake of radium by terrestrial plants

### 2.7.1 The extent of radium accumulation

The extent of contamination of terrestrial plants by radium sources can, according to Simon and Ibrahim (1990), be discussed under four major categories:

- In natural environments under normal conditions
- In regions with naturally high radioactivity
- In laboratory, greenhouse and field studies
- At uranium mill tailing sites.

Simon and Ibrahim (1990) compiled tables on the concentration of radium in terrestrial plants under these four categories. Portions of some of these tables are presented in some of the following sections.

#### 2.7.1.1 Radium in plants in natural environments

According to the National Council on Radiation Protection and Measurements (NCRPM) (1975), the world average activity concentration of $^{226}$Ra in soil is 0,03 Bq.g$^{-1}$. Iyengar (1990) found that world-wide $^{226}$Ra concentrations normally vary between 0,004 and 0,126 Bq.g$^{-1}$ while typical levels in most components of the human diet range between 0,004 and 0,2 Bq.g$^{-1}$ (United Nations, 1977). Table 2.2 provides some data on the concentration of radium in edible crops growing in normal uncontaminated soils at a number of locations in the world.

### 2.7.1.2 Radium contents of plants from regions with naturally high radioactivity

In certain areas of Iran where a high natural background of radium is found, Khademi *et al.* (1980) measured $^{226}$Ra concentrations in the soil, ranging from 0,022 to 37 Bq.g$^{-1}$. The $^{226}$Ra concentrations in the plants ranged between 0,007 and 0,240 Bq.g$^{-1}$ and therefore, although the sediment concentrations were high, concentration ratios for these plants were relatively low, varying between < 0,001 and 0,043.

Vasconellos *et al.* (1987) investigating the uptake of $^{226}$Ra by crops cultivated on the Pocos de Caldas plateau, an area in Brazil with a high natural radioactivity, showed that the maximum $^{226}$Ra content of the affected vegetables analyzed were an order of magnitude higher than in normal regions. The CR-values for these vegetables were low, ranging between 0,001 and 0,006.

### 2.7.1.3 Radium uptake in laboratory, greenhouse and field plot studies.

Table 2.3 provides a summary of radium uptake by plants from laboratory, greenhouse and experimental plots. Investigations done by D'Souza & Mistry (1970) showed that $^{226}$Ra uptake in beans grown in a spiked radium solution was considerably higher than in plants grown in soil. The CR-values (85 : shoots ; 1787 : roots) for this plant reflect the enhanced uptake of $^{226}$Ra in a solution. The lack of exchange sites and complexing ligands in the solution known to be present in soils, could explain the high $^{226}$Ra uptake in solution. Experimental exposure of a variety of crops to Ra-contaminated soil by Popova *et al.* (1964) yielded extremely low CR values (0,0012 - 0,011) even though the $^{226}$Ra content of the soil was high (39,2 Bq.g$^{-1}$). As the chemical form of radium was unknown in this experiment, it could possibly explain the low CR-values for these plants. This particular chemical form or species was for some reason not accessible to the exposed plants. Investigations done by Kirchman *et al.* (1980) revealed that the root of the sugar beet has a lower $^{226}$Ra content than the above ground foliage (dry weight basis). Kirchman *et al.* (1966) also observed that the tubers of potatoes contained ten times less radium than the leaves. However, contradictory observations showed that roots contain higher amounts of $^{226}$Ra than the shoots (Popova *et al.*, 1964 ; Verkhovskaya *et al.*, 1966 ; Gunn & Mistry, 1970 ; D'Souza & Mistry, 1970 ; Rayno *et al.*, 1980 ).

### 2.7.1.4 Uptake of radium from uranium mill tailings

Uranium milling and extraction causes residues (tailings) which usually contain elevated $^{226}$Ra levels which are several orders of magnitude higher than that in normal soil. Therefore, uranium mill tailings could provide a significant source of $^{226}$Ra contamination for terrestrial and some aquatic plants. Simon & Ibrahim (1990) report that the $^{226}$Ra content in plants growing at uranium mill sites are highly variable. Substrate concentrations, sampling locations, the degree of leaf surface contamination, the method used to clean vegetation prior to $^{226}$Ra analysis and the milling process (acidic or alkaline) are some of the factors which may contribute to the variability in Ra-concentrations amongst the different studies found in the literature. Table 2.4 provides a few radium concentrations found in edible plants grown near or on uranium

Table 2.2: Concentrations and concentration ratios (CR) of $^{226}Ra$ in plants and edible crops under normal environmental conditions. (After Simon & Ibrahim, 1990)

| Country | Food item | n | Weight basis | Plant conc. ($Bq.kg^{-1}$) AM(68% CI) Probability | Plant conc. ($Bq.kg^{-1}$) GM(68% CI) Probability | Soil ($Bq.kg^{-1}$) | CR |
|---|---|---|---|---|---|---|---|
| Fed. Rep. Germany | Cabbage | 2 | F | 0,063(0,03-0,11) | | | |
| Fed. Rep. Germany | Carrot | 3 | F | 0,11(0,02-0,22) | 0,096(0,037-0,18) | | |
| USSR | Corn | | | | | | 0,07 |
| USA | Corn | 1 | F | 0,056 | | | |
| Italy | Fruit (mix) | | F | 0,014 | | 26,5±4,2 | |
| USA | Lettuce | 2 | F | (0,17-0,20) | | | |
| USA | Root vegetables | 2 | D | 0,089(0,070-0,11) | 0,089(0,070-0,11) | | |
| Italy | Vegetables (mix) | | F | 0,036(0,027-0,044) | | 26,5 ± 4,2 | |
| USA | Vegetables (mix) | 6 | F | 0,048(0,019-0,078) | 0,041(0,023-0,078) | | |
| India | Wheat | 5 | D | 0,448(0,33-0,56) | 0,433(0,33-0,59) | | |
| USSR | Wheat grain | | | | | | 1,0 |

**Abbreviations:** A: ash; D: dry; F: fresh; a: plants without surface contamination; b: plants with surface contamination; AM: Arithmetic Mean; GM: Geometric Mean; CI: Confidence Interval; n: number of samples

Table 2.3: Concentrations and concentration ratios (CR) of $^{226}$Ra in plants and edible crops found under experimental conditions. (After Simon & Ibrahim, 1990)

| Experimental conditions | Plant Group | Weight basis | n | Conc. ($Bq.kg^{-1}$) Plant | Conc. ($Bq.kg^{-1}$) Substrate | CR |
|---|---|---|---|---|---|---|
| Nutrient solutions containing carrier free $^{226}$Ra | Bean (kidney) | D<br>D | | | | 85 (shoots)<br>1787(roots) |
| Contaminated pots containing bean (*Vicia faba*) | Leaves (young) | A | | 459 | 37000 | 0,012 |
| | Leaves (old) | A | | 2730 | 37000 | 0,074 |
| | Stem | A | | 1650 | 37000 | 0,045 |
| | Fruit | A | | 992 | 37000 | 0,027 |
| | Roots | A | | 13200 | 37000 | 0,358 |
| Contaminated soil, unknown chemical form of radium | Bean | D | | 56 | 39200 | 0,0015 |
| | Cabbage | D | | 56 | 39200 | 0,0015 |
| | Barley | D | | 44 | 39200 | 0,0012 |
| | Squash | D | | 407 | 39200 | 0,011 |
| | Lettuce | D | | 210 | 39200 | 0,0057 |
| | Spinach | D | | 67 | 39200 | 0,0018 |
| | Sunflower | D | | 63 | 39200 | 0,0017 |
| Field plots, phosphate fertilizer added | Wheat | A | 5 | 18 | 30 | 0,61 |
| | (straw) | D | | 1,5 | 30 | 0,05 |
| | Wheat | A | 6 | 8,9 | 30 | 0,30 |
| | (grain) | D | | 0,1 | 30 | 0,005 |
| | Sugar beet | A | 5 | 9,3 | 34 | 0,27 |
| | (foliage) | D | | 1,5 | 34 | 0,04 |
| | Sugar beet | A | 5 | 23 | 34 | 0,68 |
| | (root) | D | | 0,63 | 34 | 0,02 |

Abbreviations: D: dry; A: ash; n: number of samples

**Table 2.4: Concentrations and concentration ratios (CR) of $^{226}$Ra in plants and crops grown on uranium mill tailings systems and under control conditions.
(After Simon & Ibrahim, 1990)**

| Location and site | Plants | Weight basis | Conc. (Bq.kg$^{-1}$) Y(a) Plant | CR (Y/X) |
|---|---|---|---|---|
| USA (New Mexico) Residential areas near mill site | Tomato | W | 0,041 | |
| | Pea | W | 0,052 | |
| | Pea pods | W | 0,13 | |
| | Green bean | W | 0,22 | |
| | Carrot | W | 0,22 | |
| | Beet | W | 0,381 | |
| | Grass | D | 18 | |
| | Grass | D | 7 | |
| | Grass | D | 840 | |
| Fed. Rep. Germany (Black Forest, near U, Co and Au mines) | Vegetable (mix) | F | 0,04-6,3 | 0,028 |
| | Potato | F | 0,04-1,5 | 0,011 |
| | Fruit (mix) | F | 0,1-7 | 0,071 |
| | Grass (hay) | F | 0,3-20 | 0,15 |

**Abbreviations:** (a): Y arithmetic mean (AM) of plant concentrations; (b): X arithmetic mean (AM) of soil concentrations; D: dry; F: fresh; W: wet; n: number of samples; Y/X: mean of ratios mill tailings. Surveys conducted by Holtzman *et al* (1979) showed $^{226}$Ra concentrations in crops grown near uranium mill tailings which range between 0,000 041 and 0,840 Bq.g$^{-1}$ were similar or lower than those collected in regions with high natural radioactivity such as Brazil (Eisenbud, 1964). Concentration ratios obtained for crops collected at a mill site in Germany (Schüttelkopf & Kiefer, 1982) also showed a relatively close correlation to the CR values for crops collected in areas of high natural radioactivity as well as those exposed under experimental conditions.

### 2.7.2 Factors affecting the availability of radium from soil to plants

A large array of physical, chemical, biological and environmental factors play a role in the availability of trace elements from the soil for plant uptake. According to Fried & Shapiro (1961), several processes exist which must take place for plant uptake to occur, namely:

- Release of ions from the solid phase of the soil to soil solution
- Movement (involving diffusion and mass flow) of these ions to locations where plant roots exist
- Exchange of ions onto the root surfaces and transport across the root membranes
- Translocation into plant tissues

The availability of nutrients to plants from soil is governed by the solubility of the element associated with the solid phase (Cataldo & Wildung, 1978) and by the thermodynamic activity of the uncomplexed ion (Jenne & Luoma, 1977). These factors which may influence root uptake are dependent on the existence of a soluble species adjacent to the root membrane. However, the degree of availability of nutrients in the soil solution is dependent on the soil-plant relationship (Bray, 1954). According to Menzel (1965) the root system of the plant may alter the pH of the soil, influencing nutrient uptake. Mobility of nutrients is defined as the overall process whereby ions reach absorbing root surfaces and therefore facilitating the sorption of these ions into the plants (Bray, 1954). Therefore, mobility is the process involving the movement of ions by diffusion into the film of water between the root surface and the soil. Radionuclides such as radium which are soluble in the soil solution or which are isotopes of elements that have metabolic functions in plants are most readily absorbed by the roots of plants (Menzel, 1965).

The retention and availability of radium for plant uptake is therefore dependent on sorption and desorption processes in the soil. Under natural conditions a number of factors may have an influence on radium availability and uptake and which includes the effect of calcium and other cations, organic matter in the soil, the degree of leachability from soil and the chemical species (Simon & Ibrahim, 1990).

#### 2.7.2.1 Calcium and other cations

As mentioned earlier, the sorption and desorption (exchange) of radium in soil are influenced by other cations in the soil-solution system. Cations such as calcium are held as exchangeable ions on the surfaces of negatively charged, colloidal, clay and organic matter in the soil. Although it was observed that radium was adsorbed on soil in the presence of $Ca^{2+}$, the degree of radium adsorption decreased considerably as the calcium concentration increased in the soil (Nathwani & Phillips, 1979). Due to differences in ionic size, the ability of $Ca^{2+}$ to replace $Ra^{2+}$ in soil may be very low (Simon & Ibrahim, 1990).

### 2.7.2.2 Organic matter and clay content

Research conducted showed that organic matter and clays tend to be the major constituents contributing to the adsorption of $^{226}Ra$ onto soil (Simon & Ibrahim, 1990). Stevenson (1985) also states that in most mineral soils, practically all of the humic (organic) materials occur in association with clay minerals. Organic matter adsorbs about ten times more radium than clay, which is known to be more adsorptive than other soil minerals. This phenomenon is due to the high "cation exchange capacity" (CEC) of organic matter (Nathwani & Phillips, 1978). Therefore, in soil with low organic matter and low clay content, a few sites will be available for radium adsorption.

At a neutral or normal soil pH, both the clay surfaces and humic (organic) substances are negatively charged. Electrostatic interaction is thus not in favour of the observed associations and therefore their stabilities may be based on ligand exchange involving polydentate groups of the humic material and/or of the formation of ternary surface complexes (Schindler, 1990).

Investigations done by Kirchmann *et al.* (1966) revealed that an inverse relationship existed between the log of the $^{226}Ra$ content in plants and the amount of sorption material in the soil. In a three-year greenhouse study to compare Ra-uptake from soil with different organic matter contents, Grzybowska (1974) indicated that high radium-uptake occurred with low organic matter content as organic matter tends to bind radium. However, Whicker & Schultz (1982) state that decaying plant material may increase the mobility of certain radionuclides by producing complexing agents or cations which tend to displace those adsorbed on the soil.

### 2.7.2.3 Radium leachability from soil

Qualitative and quantitative information about radium desorption and mobility from soil and the potential for plant uptake can be obtained from leaching tests (Simon & Ibrahim, 1990). The presence of radium detected in lysimetric water by Rusanova (1962) indicated that radium was leached into the soil solution and was therefore available to plants. The same author also determined that various soil types differed in the degree of radium leachability. Leaching tests and measurements of *in situ* leaching of tailings dams in South Africa (De Jesus, 1987) were consistently showing that the water moved between 1 000 and 5 000 times faster than the $^{226}Ra$. The $^{226}Ra$ movement was between 0,3 and 4 cm per annum for the laboratory leaching tests.

### 2.7.2.4 Chemical form and other factors

The chemical forms of radium, the presence of iron, the soil pH and the soil texture may influence Ra-mobility in soil (Simon & Ibrahim, 1990). Variations in the quality of water extractable radium may, according to Kalin & Sharma (1982) have an influence on the chemical form of $^{226}$Ra. These authors also indicated that an inverse relationship existed between the tailings concentration and the water extractable $^{226}$Ra and that its uptake was influenced by the water solubility of the element. The quantity of $^{226}$Ra that can exist in solution, may also be influenced by the chemical species of this element. As a result of the milling process of uranium mines, some mill tailings may contain large quantities of sulphate ions. The presence of trace quantities of barium in soil may therefore induce the co-precipitation of radium resulting in the reduced leachability of radium (Nathwani & Pillips, 1978 ; Ibrahim *et al.*, 1984).

During the comparison of the leachability of radium from alkaline and acidic mill tailings, Dreesen *et al.* (1981) found that the leaching of the fractions from the tailings were 6% and 0,3% respectively. The chemical form of radium as well as the effect of pH may have played a major role in differences observed in the leachability of these two types of tailings. Research by Taskaev *et al.* (1976) has shown that the soil type influences the extraction of $^{226}$Ra. The formation of soluble organo-iron compounds also influences the mobility of $^{226}$Ra in an $FeCl_3$ medium. Nathwani & Phillips (1978) found that soil texture influenced the leachability of radium from soil. Coarser soils showed greater desorption properties to radium than finer soils.

### 2.7.3 Distribution of radium in plant tissue

Tissue barriers, specific co-ordinating substances, metabolic incorporation and other causes not fully understood (Biddulph, 1960) cause heterogeneous concentrations and distribution patterns of elements in the different parts or organs of plants. An acropetal gradient, i.e. a decreasing concentration gradient from the oldest to youngest growth and towards the apex of the plant, can occur. In the case of radium a concentration gradient was also found from roots to stems and from stems to shoots in many different plant types under different growing conditions.

Leafy vegetables and beans grown in pots containing radium-contaminated soil showed an acropetal gradient (Popova *et al.*, 1964). Although the roots generally contained the highest radium of all the plant organs, these authors indicated that the radium content in the older leaves sometimes exceeded that of the roots. Exposure of barley, peas and maize (Gunn & Mistry, 1970) and kidney beans (D'Souza & Mistry, 1970) to radium containing nutrient solutions showed that radium was preferentially accumulated by the roots and prevented from entering the shoots.

The existence of an acropetal concentration gradient suggests a low metabolic activity of radium resulting in no secondary distribution to other parts or tissues of the plant. Radium has been observed to flow unidirectionally (from roots towards shoots) and was also regulated by growth rate of the plant and leaf age. Once radium enters the transpiration stream, it remains in the leaves without further distribution. Furthermore, it does not enter the metabolic cycle which is known to redistribute some elements to areas of younger growth (Vavilov *et al.*, 1964).

Elements absorbed directly through the leaves of plants may have a different distribution pattern to those which are absorbed through the roots and stem base. While alkaline earths such as Ca, Sr and Ba are freely translocated through the xylem of the plant, their transportation encounters discrimination in the phloem. Immobilization and accumulation of Sr and Ca takes place when it is absorbed through the leaves of the plant (Biddulph, 1960).

No data from the literature are available on radium being absorbed through the leaves of plants. However, it is expected that radium would be immobile and may pose a similar behaviour to those of other alkaline earth elements (Simon & Ibrahim, 1990).

### 2.7.4 Speciation of radium in plants

The mobility of radium within the plant and the degree to which it is recycled from plants to soil is dependent on the chemical form or species of radium. Although no identification or characterization of the exact chemical species of radium in plants has been conducted, the results of some chemical speciation studies were however available. According to Verkhovskaya *et al.* (1969), a difference may exist between the chemical forms or species of radium deposited in different organs or plants. Comparisons between water-soluble, exchangeable and non-exchangeable fractions of radium have indicated that the relatively more mobile species localize in the plant tops rather than in the roots. Popova & Kyrchanova (1974) have indicated that mobile species of $^{226}$Ra were mostly present in plants while Taskaev *et al.* (1977) showed that from 2 to 34% of $^{226}$Ra in the green parts of plants was water soluble. Most of the mobile $^{226}$Ra situated in the above-ground shoots occurs as a free ionic form, incorporated in soluble mineral salts and as organic complexes. Radium, however, also associates with carbonates, oxalates and other less soluble compounds (Simon & Ibrahim, 1990).

## 2.8 The behaviour, effects and radiation dosimetry of radium in man

During the assessment of radiation effects resulting from the intake of radionuclides, the following information is often required:

- The distribution and retention of the radionuclides in human tissues
- The position of sensitive cells in the human body in order to develop both stochastic (probabilistic) and non-stochastic (threshold) effects
- The relationship between radiation dose and risk (Stather, 1990)

There is limited information available on the effects of many radionuclides on man. Therefore, animal studies provide insight into the assessment of possible consequences of human exposure to radionuclides and assist in establishing limits on radionuclide intake (Stather, 1990). Substantial human data on both the adverse effects of radium as well as its distribution and retention in the human body are, however, readily or potentially available (Stather, 1990).

### 2.8.1 Behaviour and effects on the human body

After radium has entered the human body either through ingestion or inhalation, a fraction of it is translocated to the blood. The physico-chemical form of radium taken into the body and the affinity of the transportable fraction of radium for the biological transport systems in the lungs and gut determine the amount of radium absorbed by the blood following entry into the body. As mentioned earlier, the skeleton acts as the main site of deposition after radium has entered the blood. According to the ICRP (1975), 87% of the body radium in man is distributed in the skeleton. Although a percentage of the radium present in the body is also absorbed by some soft tissues, a substantial fraction is however rapidly excreted through the gastrointestinal and urinary tracts (Stather, 1990). The ICRP (1991) suggested an annual limit on intake for $^{226}$Ra of 9 x $10^4$ Bq or 2.5 x $10^{-6}$ g. This is about 500 000 times less than the limit for uranium (See section 3.3 : **Selection of radionuclides**).

#### 2.8.1.1 Ingestion

During an investigation conducted by the ICRP Task Group on Alkaline Earth Metabolism in Adult Man (ICRP, 1973), fractional absorption values of radium from the adult GI (gastrointestinal) tract ($f_l$) were obtained which ranged between 0,15 - 0,21. From prisoners who ingested elevated levels of $^{226}$Ra in drinking water (Stenhey & Lucas, 1956), the ICRP Task Group calculated an $f_l$ value of 0,21. The ICRP also derived an $f_l$ value of 0,15 for three adult control individuals which were presumed to be under equilibrium conditions.

No data on absorption of radium in children or young persons are however available (Stather, 1990). Because calcium is required for skeletal growth in young persons and, due to the chemical similarity of Ca to that of radium, it can be expected that a higher level of uptake of radium would occur throughout childhood and into adolescence (Stather, 1990).

#### 2.8.1.2 Clearance from blood

After radium has entered the blood, there is a quantitative similarity but a qualitative difference between the behaviour of Ra and Ca. After intake, a rapid clearance of all the alkaline earth elements takes place mainly from the blood to the skeleton. However, a substantial difference exists between the clearance rates of these elements, both in man and animals (Stather, 1990).

Comparing the clearance rates of $^{47}$Ca, $^{85}$Sr, $^{133}$Ba and $^{223}$Ra from the blood of a healthy 60-year-old man after intravenous administration, Harrison *et al.* (1967) facilitated the analysis by administering four injections in succession over ten weeks. A period of two to three weeks was allowed between each injection. Both $^{133}$Ba and $^{223}$Ra were more rapidly cleared from the plasma than either $^{47}$Ca or $^{85}$Sr. Any radioactivity cleared from the plasma was either deposited in the tissues or excreted.

Considerable differences in the routes of excretion exist. Calcium is mainly excreted with the urine while radium tends to be present more in the faeces (Stather, 1990). Due to the high intestinal clearance of radium, much more radium was excreted than calcium (Stather, 1990). Measuring the excretion of $^{226}$Ra in man (11 female : 1 male) 103 and 104 days after radium intake, Rundo & Holtzman (1976) determined that faecal elimination was the main route of elimination, accounting for about 98% of the total radium activity lost from the body. At 104 days, the overall rate of elimination was estimated at 1,5% of the body content per year, suggesting a biological half-life of about 46 years (Rundo & Holtzman, 1976).

### 2.8.1.3 Structure of and deposition in the human skeleton

Because the behaviour of radium is chemically similar to that of calcium in the human body, radium is often referred to as a bone volume seeking radionuclide (ICRP, 1979). It is however necessary to know the role played by calcium in the development of the skeleton to fully understand the behaviour of radium in the human body.

Although the exact mechanism responsible for new bone formation is unknown, an increase in calcium in the area of formation takes place and apatite crystals form in the newly formed matrix. If radium or any other alkaline earths are present in the circulating blood and plasma, substitutions may be possible in the crystalline structure and therefore these elements may be incorporated into the bone crystals replacing calcium. Consequently, any bone being laid down during exposure and where high levels of radium has recently entered the blood, will contain much higher levels of activity than bone laid down when the levels of Ra in blood were low.

Any enhanced deposition occurs on all growing bone surfaces, but especially in the Haversian systems. In these Haversian systems where active bone accretion takes place, these areas become labelled during radium intake. However, those which are in a resting or resorbing phase retain only very small amounts as part of a more generalized "diffuse" deposit.

A constant exchange process is taking place between the Ca-ions in the extracellular fluid and the Ca present in bone mineral. This exchange process is caused by diffusion of ions from the blood stream to the extracellular fluids, therefore bathing the micro-crystals of bone mineral. The small size of the crystals result in a large surface ion exchange area.

### 2.8.1.4 Summary

A short review has been presented on the information available on the presence and behaviour of radionuclides in the environment and the radiation effects and behaviour of radium in the human body. Data from the literature have shown numerous cases illustrating the effects of $^{226}$Ra in the aquatic and terrestrial environments. Literature has also indicated that the substantial amounts of data available on the consequences provide a valuable source of information on the effects of incorporated radionuclides in man. It is expected that radium has a higher radiotoxicity than uranium and thus has a more important potential environmental impact. The literature survey shows that there is less data available on uranium effects on the environment than radium. To fully understand the mechanisms involved in the transport and uptake of $^{226}$Ra and uranium as well as the application of dose assessment modelling, as much information and data as possible need to be gathered. This also applies to the South African situation. Based on previous investigations (Malan, 1981; de Jesus *et al.*, 1987) the present investigation was aimed at contributing to this pool of information, especially in the South African context.

The absence of literature on the uptake of $^{226}$Ra from spray irrigation via plant leaves, together with a single reference to the effect of a higher CR for plants grown in spiked $^{226}$Ra solutions (section 2.7.1.3) make it imperative for the present study to further investigate this gap.

### 2.8.2 Dose assessment and modelling

#### 2.8.2.1 The concept of modelling

The different public exposure pathways of dust inhalation, radon and radon daughter inhalation and aquatic and food pathways need to be quantified for regulatory authorities and to assure adequate protection of the public and environment. Because of the complexity of environmental interactions and the varied modes of exposure, dose assessment is undertaken using mathematical computer models (Till, 1983 ; Halbert, 1991).

A typical model developed for the assessment of radionuclide transfer in a terrestrial environment is composed of an ensemble of submodels and mathematical equations which represent a large number of different processes and mechanisms. Pathways for the transport of radionuclides from the source to humans is illustrated in Figure 2.1.

According to such models, the following pathways can be identified and mathematical models designed for each:

- Direct exposure
- Inhalation
- Atmosphere-to-vegetation transfer
- Atmosphere-to-soil transfer
- Soil-to-vegetation transfer
- Aquatic transfer
- Irrigation-to-vegetation transfer
- Animal products

Essentially, models consider release mode from the source (gaseous or liquid), transport, dilution and dispertion, reconcentration, bioaccumulation and transfer up the food chain and subsequent exposure of humans either directly or by inhalation and consumption of water and foodstuffs. The habits of the population and data on food types and masses or volumes consumed are important for realistic dose assessment.

The models usually have the option to evaluate the dose from a number of different nuclides to:

a) the maximum exposed individual,
b) the critical group, and
c) the collective dose to the wider population

Doses to adults, teenagers or children can be evaluated, both for the whole body or individual body organs. The internal dosimetry submodels are based on established transfer factors between conceptualized body compartments.

#### 2.8.2.2 Model application

Various models have been developed world wide such as by the IAEA (1984) and the United States Nuclear Regulatory Commission (1982). Models that were available locally were LADTAP II (Simpson & McGill, 1980) from the Oak Ridge National Laboratory (U.S. Department of Energy), a mainframe version and GENII, the Harford Environmental Radiation Dosimetry Software System (1988) of Pacific Northwest Laboratory. The GENII code operates on a PC and was chosen for its ease of use for this project. It incorporates the internal dosimetry models recommended by the International Commission on Radiological Protection (ICRP).

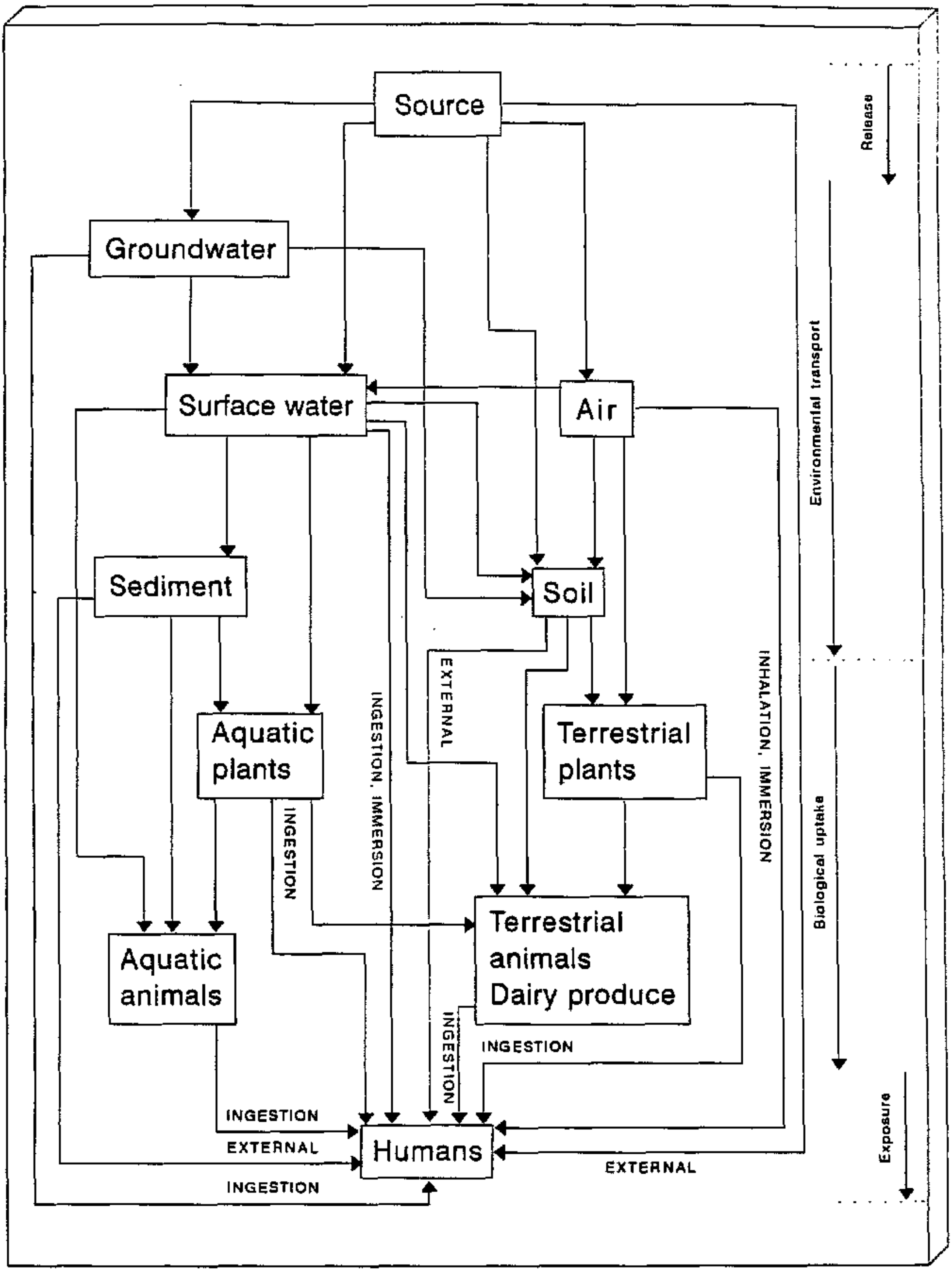

Figure 2.1 Pathways for the transport of radionuclides from the source to humans After National Technical Planning Group (1981).

# 3 MATERIALS AND METHODS

## 3.1 Factors affecting the selection of sampling localities

The criteria for suitable sampling localities required that significant concentrations of radium and uranium should occur in surface water or sediments associated with mining activities on the Witwatersrand and that agricultural production should occur using such contaminated water. In general, the localities should exclude mining land.

During the selection of suitable sampling locations, the following factors which may have had an influence on $^{238}$U and $^{226}$Ra activity and agricultural use at particular sites were taken into account:

### 3.1.1 Geology of the study area

Although the association between gold and uranium has been known since 1923 (Travener, 1957), the extraction of uranium from gold ore only started in the late forties. The main uraniferous component of Witwatersrand ores is the primary uranium mineral uraninite (usually black or dark brown, massive and non-crystalline) which usually occurs in a complex matrix of conglomerates (Liebenberg, 1955 ; 1972). The reefs of the West Rand mostly contain brannerite ($(U,Ca,Ce)(Ti,Fe)_2O_6$), which is a uranium-bearing titanate (Feather & Koen, 1975).

The average uranium (as $U_3O_8$) concentrations in the Witwatersrand and the Far West Rand vary between 50 $g.t^{-1}$ to about 200 $g.t^{-1}$ (2 Bq $^{238}U.g^{-1}$) ore (Funke, 1990). They can even be as high as 640 $g.t^{-1}$ in the Klerksdorp area and 600 $g.t^{-1}$ in the OFS goldfields. However, these figures are in contrast with those of Canada, USA, and Australia where the uranium ore content are two orders of a magnitude higher (Funke, 1990).

In contrast with uranium, the concentrations of $^{226}$Ra are much lower and average about 50 $\mu g.t^{-1}$ (2 Bq $^{226}Ra.g^{-1}$) discarded sludge for a 200 $g.t^{-1}$ ore. Analyses of slimes dams have shown that $^{226}$Ra levels vary between 0,6 and 3,1 $Bq.g^{-1}$ for the Klerksdorp and OFS mining areas where richer uranium ores are associated with the presence of gold. This is in contrast with the East Rand where activity levels between 0,2 and 0,6 $Bq.g^{-1}$ were measured on slimes dams (Chamber of Mines of South Africa, 1979). These results are confirmed by de Jesus *et al.* (1987) who obtained the same tendency by measuring the $^{226}$Ra activity on various slimes dams in these mining areas. On the East Rand (0,49 ± 0,16 $Bq.g^{-1}$) and Central Rand (0,41 ± 0,15 $Bq.g^{-1}$) a relatively low $^{226}$Ra activity was measured while the Klerksdorp mining area produced the highest average $^{226}$Ra activity of 1,84 ± 1,21 $Bq.g^{-1}$ (Table 3.1). On the West Rand (1,18 ± 0,80 $Bq.g^{-1}$) and OFS-goldfields (1,23 ± 0,99 $Bq.g^{-1}$) the $^{226}$Ra-activity was more or less the same (Table 3.1), but lower than that of Klerksdorp (de Jesus *et al.*, 1987). The same tendency has also been observed during an airborne gamma-survey conducted by Grundling & Schoeman (1989) from Nigel in the east to Stilfontein in the far west. These results also showed a relatively low gamma-activity in the Eastern and Central Rand, while the West Rand and Klerksdorp-Stilfontein areas showed relative gamma-activity varying from > 1000 to about 3600 counts per second.

Table 3.1: Activity of $^{226}$Ra on various slimes dams on the East, West and Central Rand as well as the Klerksdorp and OFS goldfields (After de Jesus *et al.*, 1987).

| Mine | $^{226}$Ra Concentration (Bq.g$^{-1}$) | Mine | $^{226}$Ra Concentration (Bq.g$^{-1}$) |
|---|---|---|---|
| EAST-RAND | | WEST RAND | |
| Brakpan | 0,64 ± 0,05 | West Rand Consolidated | 1,22 ± 0,08 |
| Geduld-East | 0,58 ± 0,05 | Randfontein Estates | 2,69 ± 0,10 |
| Grootvlei | 0,58 ± 0,06 | East Champ d'Or | 0,53 ± 0,04 |
| New State Areas | 0,56 ± 0,06 | Blyvooruitzicht (1) | 0,64 ± 0,05 |
| Daggafontein | 0,38 ± 0,06 | Blyvooruitzicht (2) | 1,32 ± 0,07 |
| Sallies | 0,21 ± 0,06 | West Driefontein | 0,71 ± 0,05 |
| **Mean** | **0,49 ± 0,16** | **Mean** | **1,18 ± 0,8** |
| CENTRAL RAND | | ORKNEY/KLERKSDORP | |
| Vill. Main Reef (2) | 0,47 ± 0,03 | Buffelsfontein | 3,40 ± 0,26 |
| Vill. Main Reef (1) | 0,43 ± 0,03 | Vaal Reefs | 2,69 ± 0,14 |
| Vill. Main Reef (3) | 0,34 ± 0,03 | Western Reefs | 1,69 ± 0,08 |
| City Deep (3) | 0,16 ± 0,01 | Hartebeespoort | 0,54 ± 0,05 |
| City Deep (1) | 0,22 ± 0,02 | Stilfontein | 0,86 ± 0,05 |
| City Deep (2) | 0,69 ± 0,04 | **Mean** | **1,84 ± 1,21** |
| ERPM (3) | 0,37 ± 0,02 | ORANGE FREE STATE | |
| ERPM (4) | 0,47 ± 0,03 | Merriespruit | 2,67 ± 0,15 |
| ERPM (1) | 0,48 ± 0,03 | Harmony | 1,09 ± 0,12 |
| ERPM (5) | 0,31 ± 0,03 | Welkom | 0,48 ± 0,04 |
| ERPM (2) | 0,59 ± 0,03 | Western Holdings | 0,69 ± 0,05 |
| **Mean** | **0,41 ± 0,15** | **Mean** | **1,23 ± 0,99** |

### 3.1.2 Agricultural conditions

Although the East, Central and West Rand as well as the Klerksdorp and OFS goldfields fall within a summer rainfall area with an average precipitation of 400-720 mm per year, there are, however, subtle differences between the rainfall and soil composition of these areas which may have an influence on the suitability of sampling locations. The East Rand (Nigel, Springs, Brakpan and Boksburg), Central Rand (Germiston and Johannesburg) and West Rand (Roodepoort, Krugersdorp and Randfontein) have a higher average precipitation than the Klerksdorp mining area and the OFS goldfields (Weather Bureau, 1992). The soil conditions in the East, Central and West Rand are also more favourable for agricultural and irrigation purposes as the soil contains a higher average organic content and has a longer retention time for moisture. In these areas, a number of dams and lakes are found whilst extensive wetland areas also supply water to farmers for irrigation. Therefore, the Witwatersrand is generally more suitable for vegetable production as well as fodder for dairy cattle. However, the far West Rand, Klerksdorp and OFS are known for soils which consist of weathered calcareous dolomite which is not effective in storing water. The Klerksdorp and OFS mining areas are therefore not quite suitable for the production of irrigation-intensive crops and are generally known as maize-producing areas which are not irrigation-intensive. Due to the bigger impact that mining might have on agriculture, the Witwatersrand area was selected for this study.

### 3.1.3 Aquatic pathways for radionuclide contamination of agricultural land.

Two possible pathways exist whereby suspended or dissolved $^{226}$Ra and uranium can reach surface waters near gold and uranium mines:

#### 3.1.3.1 Seepage from slimes dams

An extensive survey conducted by de Jesus *et al.* (1987) to determine $^{226}$Ra-levels on slimes dams on the Witwatersrand has shown that levels vary from 0,2 Bq.g$^{-1}$ on the East Rand to 3,1 Bq.g$^{-1}$ on the West Rand. The average activity for all these slimes dams was about 0,5 Bq.g$^{-1}$ (Table 3.1) compared to 0,04 - 0,08 Bq $^{226}$Ra.g$^{-1}$ for normal soil. Many of the slimes dams which have been sampled have fallen into disuse in the past few decades. The state of conservation of these slimes dams therefore varies considerably. Some of these slimes dams have been subjected to plant rehabilitation programmes while others have been covered by gravel or burnt-out coal to prevent any further wind or water erosion (Cook, 1973 ; Cartwright, 1983 ; Blight & Cladwell, 1984 ; Jones *et al.*, 1989). However, there are some slimes dams which show signs of erosion and silting. It is especially the finer fractions which are clearly visible around the slimes dams. However, experimental data obtained by de Jesus *et al.* (1987) has shown that the mobility of $^{226}$Ra is about 1300-4500 times slower than that of water and that the $^{226}$Ra-activity averaged <0,07 Bq.$\ell^{-1}$ in seepage water sampled from the bases of some slimes dams. This seepage water could reach agricultural land, but the probability of large volumes of seepage occurring, is very low.

#### 3.1.3.2 Contamination of surface waters

Contamination of surface waters (rivers, lakes and wetlands) usually occurs where mining process water is released in waterways (de Jesus *et al.*, 1987). Surveys done in the vicinity of various mines as well as several sampling locations downstream from the points of release of effluents have shown that, except for a few isolated cases, the $^{226}$Ra-levels have decreased to such an extent in the water and sediments, that an activity of 0,04 Bq.$\ell^{-1}$ was measured within a few kilometres from the point of release. Samples taken from lakes in the vicinity of gold/uranium mines and which are accessible to the public, have shown that the $^{226}$Ra-levels are nearly similar to that of natural waters (de Jesus *et al.*, 1987) (Table 3.2) but sediment values are elevated in some localities. The greatest potential impact on irrigated lands is thus within a few kilometres downstream of mine discharges, and sampling localities had to be selected where agricultural activities was closest downstream from mine discharge points.

These results show general trends and identify certain areas. While not pinpointing problem areas, they have been very valuable in aiding selection of suitable sampling locations.

## 3.2 Sampling localities

Bearing in mind the criteria for sample location and the factors elaborated on above, various in-field inspections were carried out as highlighted below.

### 3.2.1 East Rand

This investigation originated on the East Rand and the Marievale Farm in particular was selected. This farm is located near the Marievale gold mine in the vicinity of Springs and Nigel (Figure 3.1). This farm is situated about 2 km downstream from where Marievale mine releases its effluents. On the farm, vegetables such as lettuce, cabbage, cauliflower, beetroot and carrots are produced. Vegetables produced here are supplied to a local market as well as to a well-known supermarket group (Geral, 1990).

This investigation was preceded by a preliminary study on the $^{226}$Ra and uranium levels in a number of selected aquatic plants, algae, macro-invertebrate fauna as well as fish and water birds occurring at localities in the Blesbokspruit, Elsburgspruit and Natalspruit catchment areas on the East Rand. These results are also considered in the present report.

Permission was obtained to sample oats plants grown on a slimes dam near Marievale. These plants were experimentally cultivated by a mine to determine the potential survival of different plant species on slimes dams. The slimes dam was divided into approximately 30 field plots (4 X 6m) onto which various different plants, shrubs and crops were planted. The experiment was conducted by the mine and it lasted for approximately two years. No irrigation was applied to these plants. Rain provided the only moisture to these plants.

Table 3.2. $^{226}$Ra levels of dams and lakes in the study area accessible to the public (After de Jesus *et al.*, 1987).

| Sampling point | $^{226}$Ra Conc. Water (Bq.$\ell^{-1}$) | $^{226}$Ra Conc. Sediment (Bq.g-1) |
|---|---|---|
| EAST RAND | | |
| Middle Lake | 0,01 | N.S. |
| Apex Pan | 0,05 ± 0,02 | 0,17 ± 0,01 |
| Jan Smuts Dam | 0,01 | 0,06 ± 0,01 |
| Geduld Dam | 0,01 | N.S. |
| Cowles Dam | 0,03 ± 0,01 | N.S. |
| Vogelstruisbult Dam | 0,12 ± 0,04 | N.S. |
| Nigel Dam | 0,11 ± 0,04 | 0,24 ± 0,02 |
| Withokspruit | 0,01 | N.S. |
| Van Wyk Dam | 0,01 | N.S |
| Rooikraal | 0,05 ± 0,02 | N.S. |
| CENTRAL RAND | | |
| Hemmer Pan | 0,07 ± 0,02 | 0,24 ± 0,02 |
| Victoria Lake | 0,01 | N.S. |
| Elsburg Spruit | 0,05 ± 0,02 | N.S. |
| Angelo Pan | 0,13 ± 0,04 | N.S. |
| Boksburg Lake | 0,01 | 0,05 ± 0,01 |
| Elsburg Dam | 0,02 ± 0,01 | 0,22 ± 0,02 |
| WEST RAND | | |
| Florida Lake | 0,10 ± 0,01 | N.S. |
| Rand Leases Dam | 0,01 | N.S. |
| New Canada Dam | 0,01 | N.S. |
| KLERKSDORP | | |
| Vaal River (Downstream) | 0,02 ± 0,01 | 0,04 |
| Vaal River (Upstream) | 0,01 | 0,04 |
| Jagspruit | 0,24 ± 0,07 | 0,06 ± 0,01 |
| ORANGE FREE STATE | | |
| Sand River (Upstream) | 0,01 | 0,04 |
| Sand River (Adjacent) | 0,01 | N.S. |
| Sand River (Downstream) | 0,01 | 0,04 |
| Ooring Pan | 0,02 ± 0,01 | 0,32 ± 0,04 |

Abbreviations: N.S. : Not Sampled

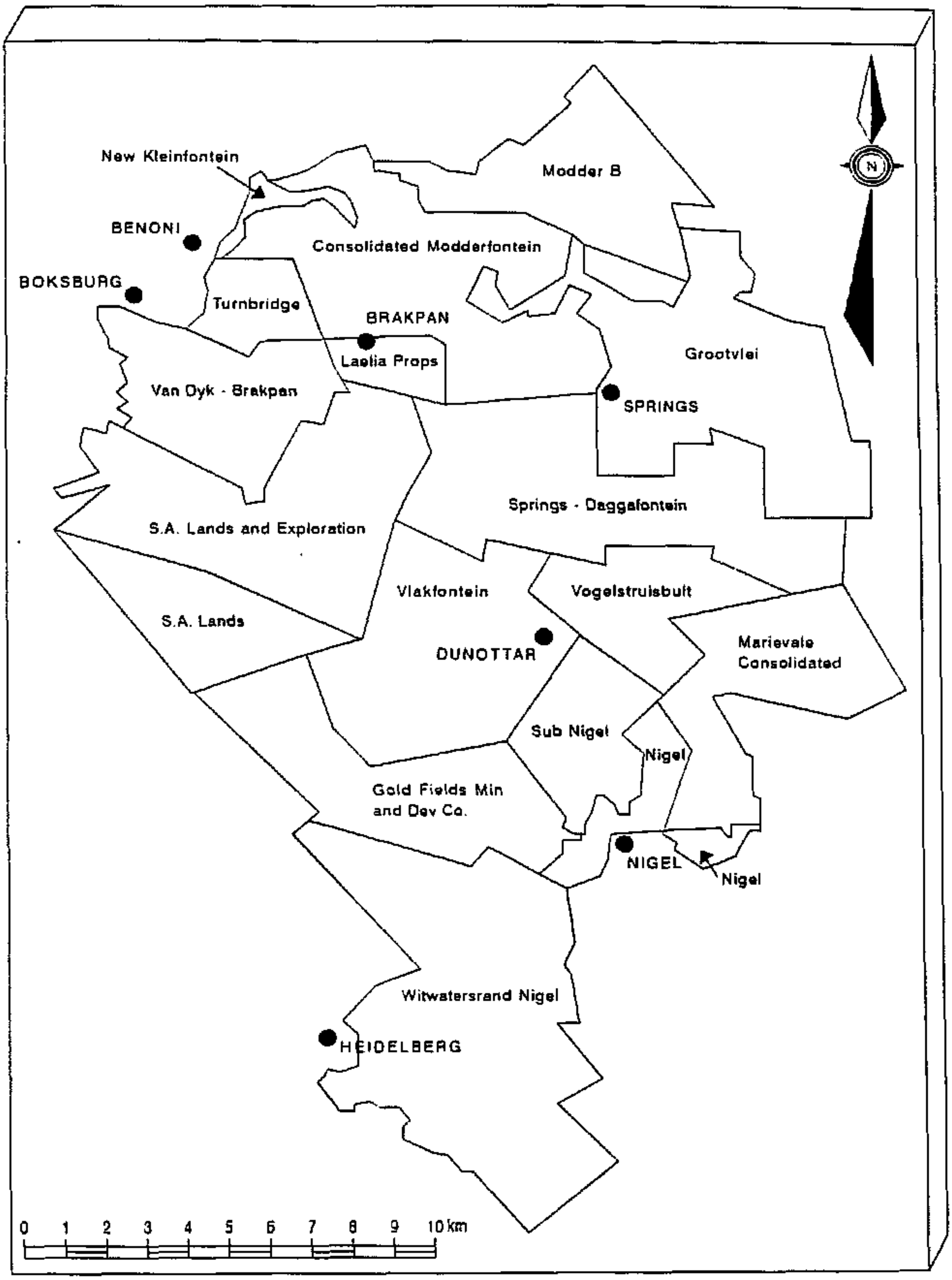

Figure 3.1 East Rand Goldfield locality map. After Antrobus (1986) and Engelbrecht (1989)

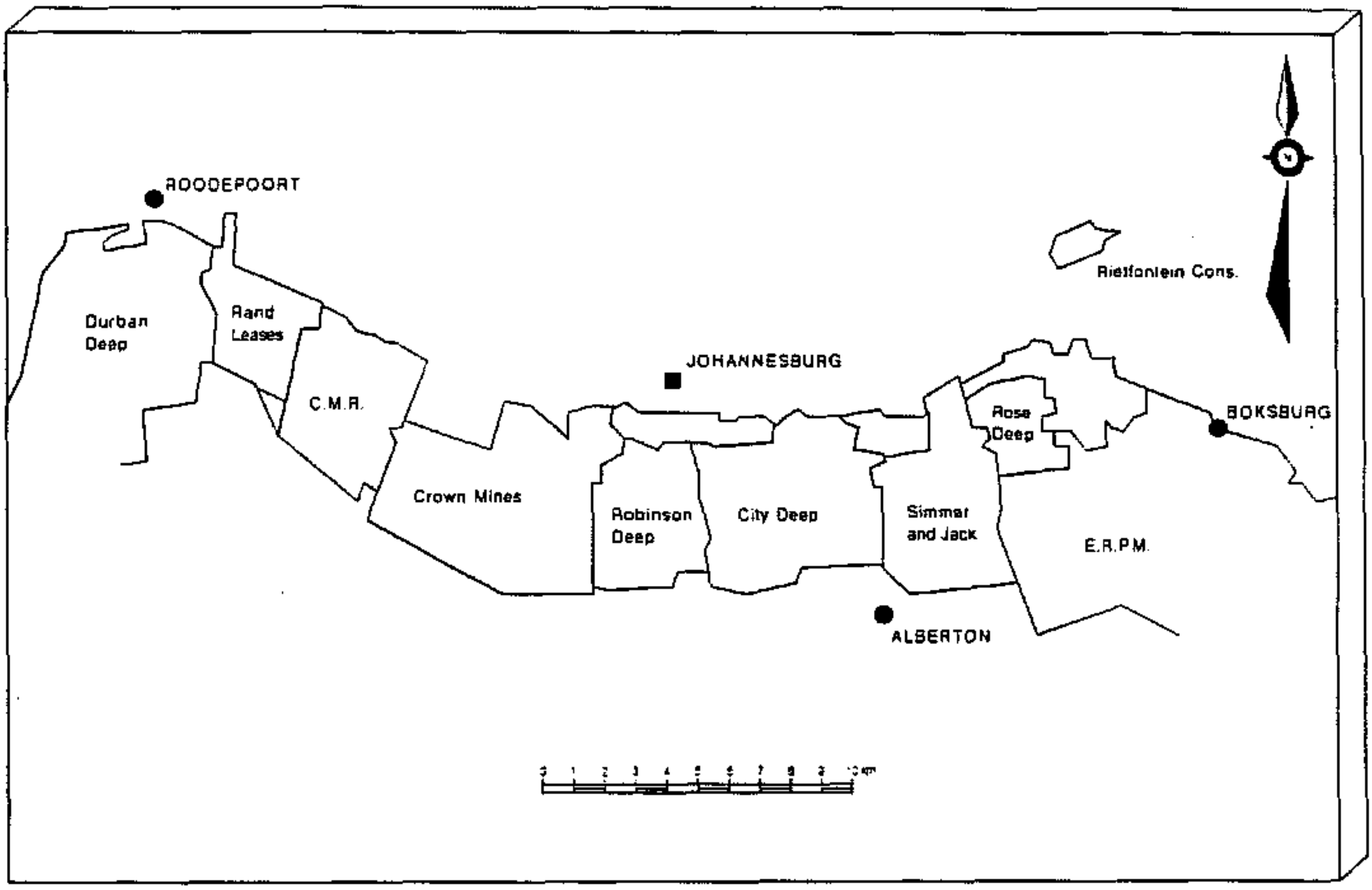

Figure 3.2 Central Rand Goldfield locality map. After Antrobus (1986) and Engelbrecht (1989)

### 3.2.2 Central Rand

This area is also known to be active in gold and uranium production (Figure 3.2), but results obtained by de Jesus *et al.* (1987) and Grundling & Schoeman (1989) have shown that $^{226}$Ra-activity to be the lowest of all the areas considered for the present investigation. Although extensive agricultural activities take place south of Johannesburg as well as Germiston, Alberton and Boksburg, nearly all the farmers utilize underground water for irrigation. As underground water is not representative of mining effluent, it was decided that these agricultural areas do not fall within the scope of this project. While, a few farmers were found to utilize water from the nearby wetlands, it was determined that $^{226}$Ra-levels were negligible due to the filtering effects of these waters. During reconnaissance investigations several incidents of political violence were encountered which often made sampling dangerous in these areas.

### 3.2.3 West Rand

Two agricultural areas which seemed to be of specific interest were identified:

#### 3.2.3.1 Luipaardsvlei

This agricultural area is situated south of Randfontein where it is surrounded by intensive gold and uranium mining activities (Figure 3.3). This area is characterized by vast underground aquifers which occur in dolomitic formations (Engelbrecht, 1989 ; & Funke, 1990). Significant amounts of water are pumped from underground compartments for mine dewatering as well as agricultural and domestic use.

A number of gold mines are active in this area, and $U_3O_8$, pyrite, silver and osmiridium are also produced as by-products (Engelbrecht, 1989). Some of these mines and a gold refinery are situated upstream along a river which drains through a series of dairy farms.

Most of the agricultural land is owned by farmers who specialize in milk production. Milk produced on these farms is delivered to a cheese factory (du Preez, 1991). The fodder produced is irrigated with water pumped from the nearby stream as well as with water received from a mine (du Plessis, 1991). Underground water is, however, unsuitable for human consumption (Coetzee, 1991).

#### 3.2.3.2 Vlakfontein

This agricultural area is situated approximately 3-4 km southeast of Luipaardsvlei (Figure 3.3) and also receives effluent and seepage waters from a nearby goldmine for irrigation purposes. This vegetable farm produces lettuce, cabbage, cauliflower, beetroot, carrots, and spinach. A vegetable shop, open to the public, is situated on the farm next to a main route and vegetables are also supplied to a local market. The farmer makes exclusive use of flood irrigation as spray irrigation appears to damage or burn the leaves of the irrigated crops (Inago, 1992).

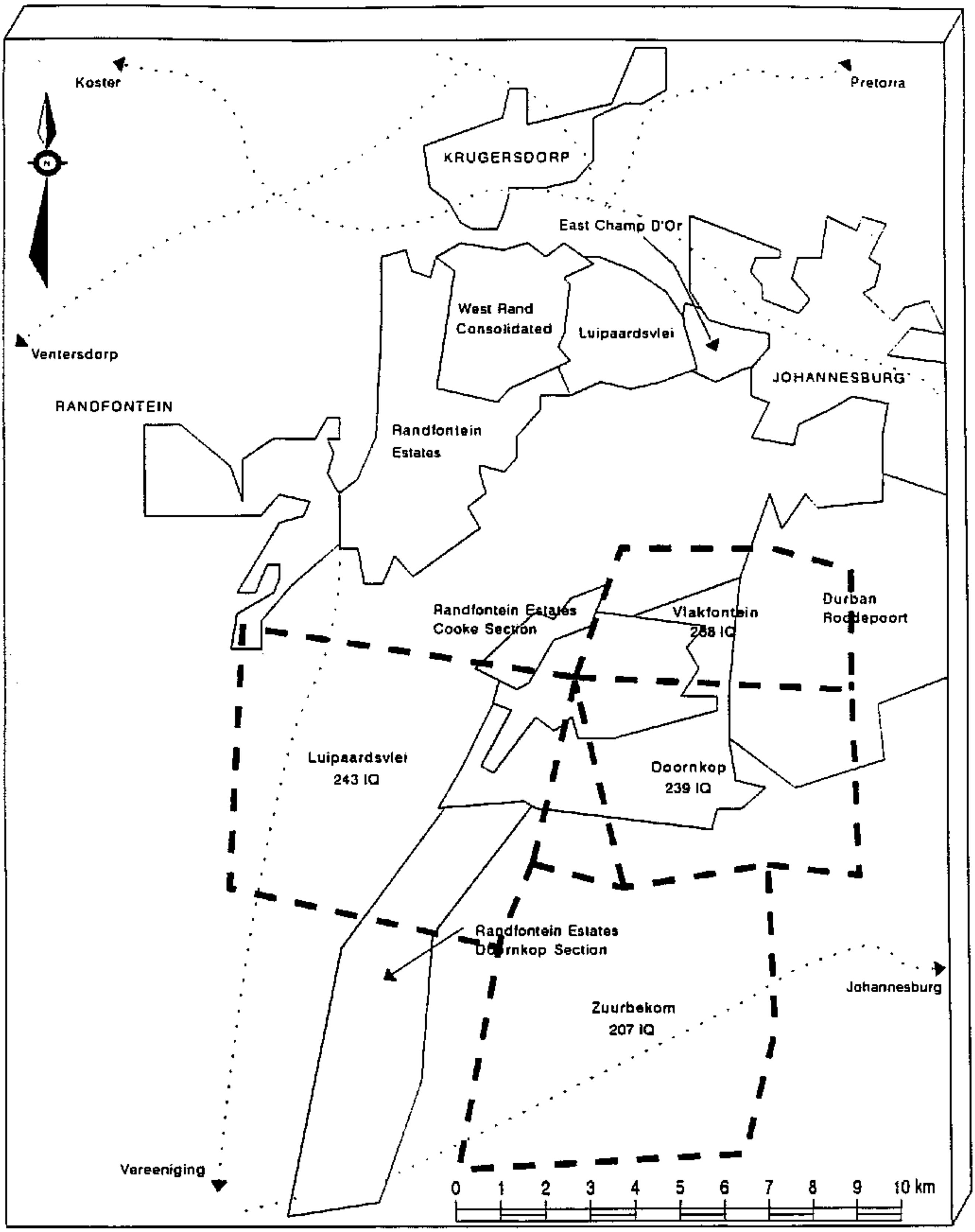

Figure 3.3 West Rand Goldfield locality map. After Antrobus (1986) and Engelbrecht (1989)

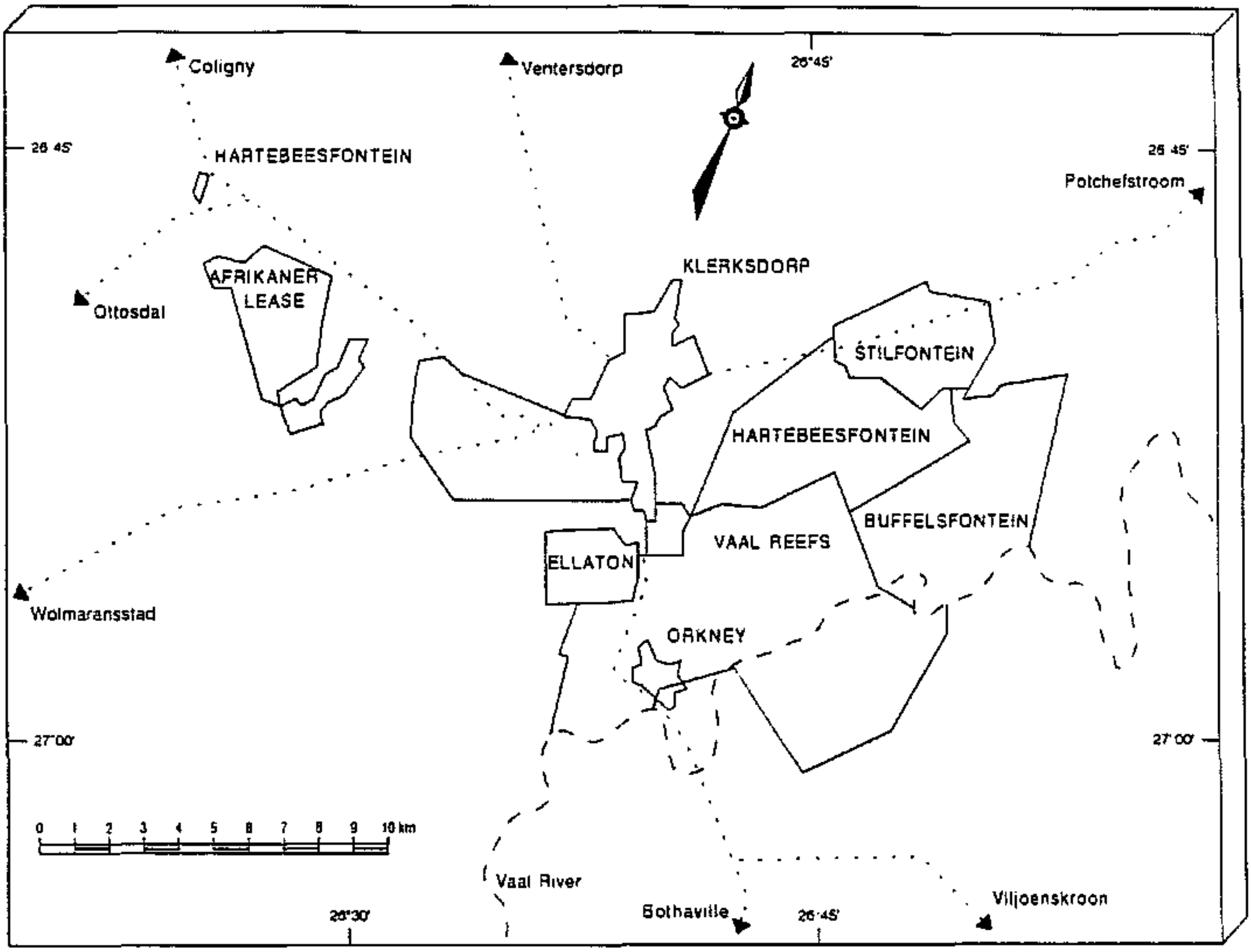

Figure 3.4 Klerksdorp Goldfield locality map. (After Antrobus (1986) and Engelbrecht (1989)

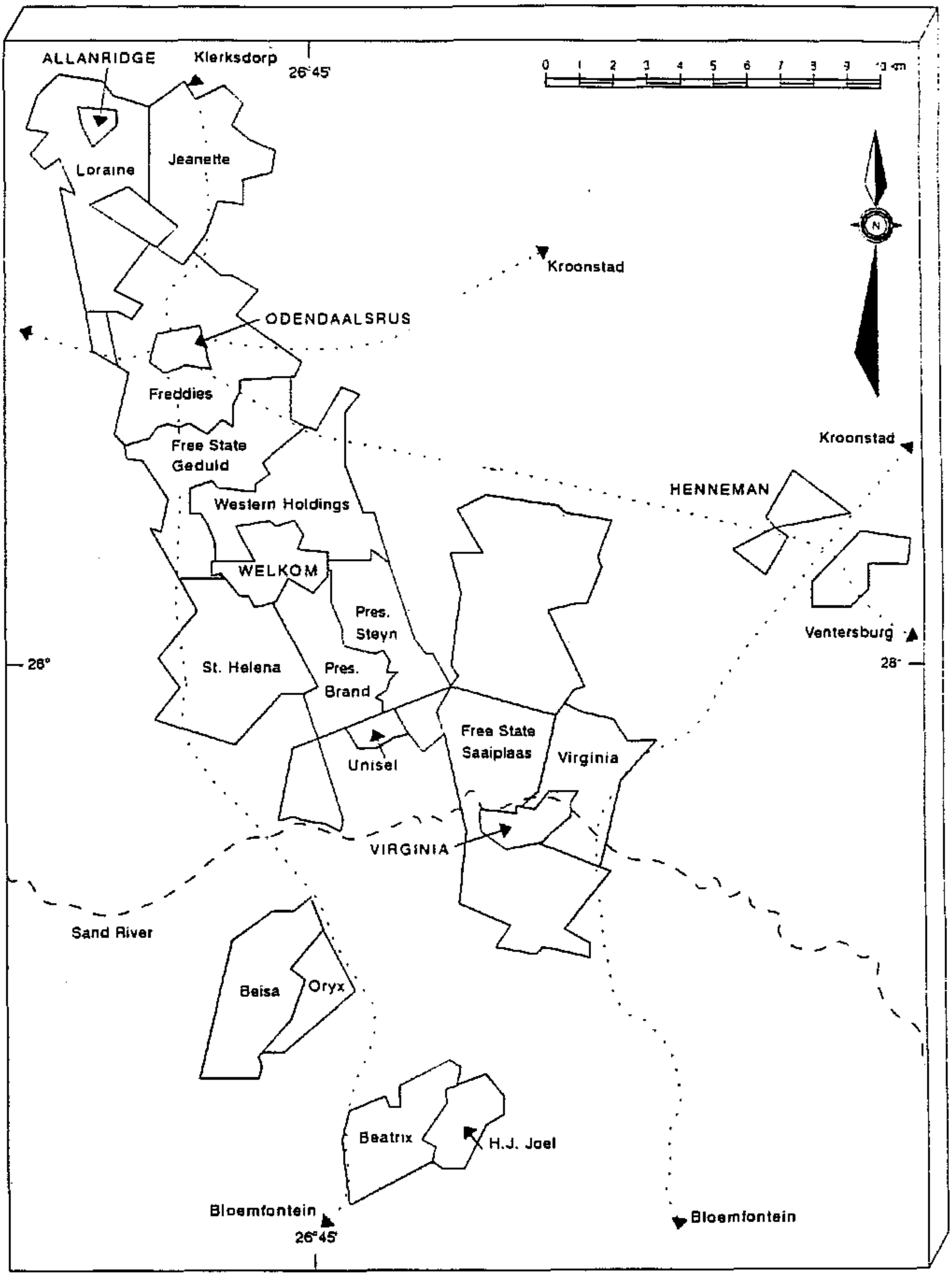

Figure 3.5 Orange Free State Goldfield locality map. After Antrobus (1986) and Engelbrecht (1989))

### 3.2.4 Orkney/Klerksdorp and OFS goldfields

Although the highest $^{226}$Ra-activity levels were reported in these areas (de Jesus *et al.*, 1987), a number of factors render these areas unsuitable for agricultural activities. Although no dramatic differences exist between the amount of precipitation on the Witwatersrand and the Orkney/Klerksdorp (Figure 3.4) and OFS (Figure 3.5) goldfields areas, the latter tend to be drier and unsuitable for the production of vegetable crops. The drought during the time of sampling has even made it difficult to sustain dry land crops such as maize. It was therefore decided to discontinue any investigations in these specific areas.

## 3.3 Selection of radionuclides

Table 3.3 contains the radio isotopes which are formed in the important decay routes of $^{238}$U. Based on annual limit on ingestion intake , the isotopes of concern are $^{226}$Ra, $^{210}$Pb and $^{210}$Po.

**Table 3.3: Radioisotopes formed by the decay of $^{238}$U**

| Half-life(seconds) | Isotope | gram/ Becquerel | Annual limits on intake, ingestion in Becquerel (ICRP,1991) |
|---|---|---|---|
| 1.41e+17* | $^{238}$U | 8.05e-05 | 3e+06 |
| 2.08e+06 | $^{234}$Th | 1.17e-15 | 4e+06 |
| 2.43e+04 | $^{234}$Pa | 1.36e-17 | 4e+07 |
| 7.70e+12 | $^{234}$U | 4.32e-09 | 3e+06 |
| 2.43e+12 | $^{230}$Th | 1.34e-09 | 3e+05 |
| 5.05e+10 | $^{226}$Ra | 2.73e-11 | 9e+04 |
| 3.30e+05 | $^{222}$Rn | 1.74e-16 | - |
| 1.83e+02 | $^{218}$Po | 9.56e-20 | - |
| 1.61e+03 | $^{214}$Pb | 8.24e-19 | 1e+08 |
| 1.19e+03 | $^{214}$Bi | 6.09e-19 | 2e+08 |
| 1.64e-04 | $^{214}$Po | 8.41e-26 | - |
| 7.04e+08 | $^{210}$Pb | 3.50e-13 | 2e+04 |
| 4.33e+05 | $^{210}$Bi | 2.18e-16 | 1e+07 |
| 1.20e+07 | $^{210}$Po | 1.00e-14 | 9e+04 |

* 1.41e+17 = $1.41 \times 10^{17}$

In the absence of $^{226}$Ra, 1,0 Bq of $^{222}$Rn will decay to form only about 0,00047 Bq of $^{210}$Pb in about a month's time. $^{210}$Pb and $^{210}$Po can therefore only build up to appreciable quantities if $^{226}$Ra is present. Furthermore, due to the fact that radon is a noble gas with a short biological half-life relative to its radiological half-life, almost all of the radon will diffuse out of living tissue before it decays to form its daughter isotopes (Napier *et al.*, 1988). Hence, the possibility of intake of appreciable amounts of $^{210}$Pb and $^{210}$Po through farm produce irrigated with water contaminated with mine effluents is small. Evidence for this in the Witwatersrand area is confirmed by unpublished data of V. Ellerbeck (AEC Internal Report 1988) who investigated $^{210}$Po in the liver of cattle in mining areas and in a control area. The mean concentrations of $^{210}$Po were 6,2 ± 2,7 Bq/kg and 2,8 ± 0,5 Bq/kg fresh mass respectively. Such values are very low in relation to the annaul limit on intake given in Table 3.3. $^{226}$Ra could therefore represent the second part (last 9 daughter products) of the decay chain of $^{238}$U.

The literature on uranium is less abundant than that of $^{226}$Ra. The differences in the annual limit on intake of the first 5 isotopes in the $^{238}$U decay chain is not as big as between the last 9 isotopes. $^{238}$U itself can therefore represent the first part of the decay chain.

The other usually important natural decay chain is that of $^{232}$Th. However, the local gold-bearing reefs are associated with low concentrations of thorium, typically a factor 10 less than uranium concentrations and potential dose from the thorium decay chain can be neglected.

This study therefore concentrated on uranium and $^{226}$Ra as these would probably be adequate indicators of the extent of the possible effect of radionuclides on people living in the proximity of mining activities on the Witwatersrand.

## 3.4 Experimental uptake of $^{226}$Ra by two selected vegetable crops

### 3.4.1 Aim of the experiment

As spray irrigation is often used on vegetables at the selected localities, the aim of this experiment was therefore to determine the difference in uptake of a known concentration of $^{226}$Ra through the leaves and roots of two selected vegetable crops (one root and one leaf crop) under greenhouse conditions. Results obtained from the experiments were used to calculate concentration ratios (CR) for these crops and to assess effects of flood versus spray irrigation in agricultural production.

### 3.4.2 Preparation of $^{226}$Ra standard solutions for irrigation on experimental plants

A $^{226}$Ra standard solution of 4500 Bq (1 ℓ) was obtained from the AEC for the experimental study. Another stock solution for irrigation purposes was made by pipetting 5 mℓ from the original stock and making it up to 1 ℓ giving an irrigation stock of 22,5 Bq.$\ell^{-1}$ which, for comparison, is about 100 times higher than the drinking water limit. Therefore, a dosage of 0,0225 Bq was obtained per millilitre applied to the plants. A suitable irrigation scheme, giving a cumulative dosage of 5,67 Bq per plant at the end of the irrigation period was therefore calculated and used (Table 3.4).

### 3.4.3 Irrigation system and dose application

Four irrigation schemes were proposed for conducting the experiment:

1) Irrigation water enriched with $^{226}$Ra sprayed on the leaves only of the plants.

2) Irrigation water enriched with $^{226}$Ra applied to the roots only of the plants (via the soil).

3) Irrigation water enriched with $^{226}$Ra sprayed on the leaves and applied to the roots of the plants.

4) Irrigation water not enriched with $^{226}$Ra sprayed on the leaves and applied to the roots of the plants. These plants served as the control of the experiment.

Cabbage and beetroot seedlings were obtained from a farm remote from mining activities whereafter a certain number of these plants were kept for background analysis while the others were used for the experiment. For each irrigation scheme, ten plants of each type were used. Before the experiment commenced, all pots were cleaned with 10% formaldehyde to kill any pathogens. Plants used in the experiment were planted in these pots in fresh potting soil. These potted plants were kept in a greenhouse. After an acclimatization period of two weeks, the proposed irrigation schemes were commenced and application of the $^{226}$Ra-enriched water continued daily for 6 weeks. At the end of the exposure period the plants were carefully removed from the pots with roots intact and prepared for analysis as per section 3.5.3.

**Table 3.4: $^{226}$Ra irrigation scheme showing the daily dosage and calculated weekly-values for 6 successive weeks.**

| Week | $m\ell.day^{-1}$ | $Bq.day^{-1}$ | $Bq.week^{-1}$ |
|---|---|---|---|
| 1 | 5 | 0,1125 | 0,7875 |
| 2 | 5 | 0,1125 | 0,7875 |
| 3 | 6 | 0,135 | 0,945 |
| 4 | 6 | 0,135 | 0,945 |
| 5 | 7 | 0,1575 | 1,1025 |
| 6 | 7 | 0,1575 | 1,1025 |
| | | Total | 5,67 |

## 3.5 Collection, identification and preparation of water, sediment and biological samples for $^{226}$Ra and uranium analysis

### 3.5.1 Water samples

At least 5ℓ composite water samples were collected at the various sampling localities (Figures 3.1 - 3.6). These were individually filtered in the field with a Sartorius pressure filter through 8 and 0,45 micron membrane filters to remove any suspended solids. 10 mℓ concentrated nitric acid was added to each sample to retain any $^{226}$Ra present in solution.

$^{226}$Ra was determined in each water sample with the emanation method. In each case, a 400 mℓ sample was sealed in a bubble flask. After about three weeks, allowing for the build-up of $^{222}$Rn to equilibrium level with the $^{226}$Ra, the $^{222}$Rn-gas was emanated into a scintillating flask. The concentration of the $^{222}$Rn in the flask was then analysed on a photomultiplier tube, after a further period of three hours' storage. The scintillating flasks were calibrated using IAEA certified $^{226}$Ra solutions.

For uranium preparation and analysis, each water sample was evaporated on a thin plastic sheet. The sheet and solids on it were then ashed, and the resulting solid analysed with Neutron Activation Analysis (NAA). Alternatively a 250 mℓ water subsample was run through an ion exchange column which was subsequently analysed for uranium with NAA.

### 3.5.2 Sediment samples

After collection, all sediment samples were individually dried for 72h at 90 - 100 °C whereafter they were homogenized and sieved through a 1 mm sieve. Moisture, total organic content and pH of the sediment samples were also determined.

Each dried sample was sealed in a 30 mℓ glass bottle using epoxy glue. After about three weeks, allowing for the build-up of $^{214}$Bi to equilibrium level with $^{226}$Ra, the sample was analysed on a gamma spectrometer with a Ge-Li detector. The 609 keV gamma of $^{214}$Bi was used to determine the concentration. $^{226}$Ra and $^{60}$Co sources were used to do the energy calibration of the detector. An IAEA-certified $^{226}$Ra standard, sealed in the same geometry as the samples, was used as a reference standard.

Uranium was determined using a delayed neutron version of NAA. A small amount of homogenized sample (200 mg) was sealed in an ampoule and irradiated with neutrons. The delayed neutrons emanating from $^{235}$U were then quantitatively measured. The isotope distribution of natural uranium was then assumed to calculate the uranium in the sample.

### 3.5.3 Biological samples

Emergent, floating and submerged aquatic weeds were collected for $^{226}$Ra and uranium analysis (Table 3.5). Identification of the aquatic weeds was done using Cook *et al.* (1974) and Van Wyk & Malan (1988). Freshwater fish species, which included several

Table 3.5 Organisms collected for $^{226}Ra$ and uranium analysis.

| Species | Common name | Family |
|---|---|---|
| PLANTS<br>*Azolla filiculoides* Lam. | Water fern | Azollaceae |
| *Marsilea macrocarpa* (DC.) C. Presl | Water-clover | Marsileaceae |
| *Lemna major* L. | Duckweed | Lemnaceae |
| *Lagarosiphon muscoides* Harv. | Fine oxygen weed | Hydrocharitaceae |
| *Potamogeton pectinatus* L. | Fennel-leaved pondweed | Potamogetonaceae |
| *Typha capensis* (Rohrb.) N.E. Br. | Bulrush | Typhaceae |
| *Arundo donax* L. | Spanish reed | Poaceae |
| MACRO-INVERTEBRATES<br>*Potamonautes warreni* (Calman) | Freshwater crab | Potamonautidae |
| FISH<br>*Barbus aeneus* (Burchell, 1822) | Smallmouth yellowfish | Cyprinidae |
| *Cyprinus carpio* Linnaeus, 1758 | Common carp | Cyprinidae |
| *Clarias gariepinus* (Burchell, 1822) | Sharptooth catfish | Clariidae |
| *Tilapia sparrmanii* Smith, 1840 | Banded tilapia | Cichlidae |
| AQUATIC BIRDS<br>*Fulica cristata* Gmelin, 1789 | Redknobbed coot | Rallidae |
| *Phalacrocorax carbo* (Linnaeus), 1758 | Whitebreasted cormorant | Phalacrocoracidae |

edible fish (Table 3.5), were collected from localities in the Blesbokspruit, Elsburgspruit and Natalspruit. While gill nets were used to collect the bigger specimens, smaller fish were mainly caught using a modified Moore type shocker (Moore, 1968) as well as a beach seine net of 5 mm stretched mesh. Fish species were identified using the methods described by Le Roux & Steyn (1968) and Jubb (1967) and placed into taxonomical order as described by Jackson (1975). The freshwater crab, *Potamonautes warreni* (Calman), which commonly occurs in the waters of the East Rand, was selected as a representative of the macro-invertebrate fauna (Table 3.5). Aquatic birds including the weedeating redknobbed coot, *Fulica cristata* and the piscivorous white breasted cormorant, *Phalacrocorax carbo* (Table 3.5), were collected for analysis after being shot with a 0,22 rifle.

Biological material was placed in plastic bags during collection. All samples were carefully rinsed and cleaned at the laboratory. Adequate quantities of fresh material of all the biological samples were individually weighed, then dried for 5-6 days at 90 °C (Ellerbeck *et al.*, 1990) and the moisture content determined. The dried material was ashed in a muffle for 1 h at 200 °C whereafter the temperature was increased to 500°C. The duration of the ashing was about 2 hours or until no black fragments of carbon remained in the samples (Moolman *et al.*, 1990).

Following ashing, the samples were individually weighed into cleaned glass beakers and a mixture of 1:1 $HNO_3$ (180 mℓ) : $HClO_4$ (180 mℓ) was added to each of the samples and digested on a hotplate. After cooling, the samples were transferred to bubble flasks (100 mℓ or 400 mℓ, depending on the mass of the sample), sealed and stored for three weeks for $^{226}$Ra and $^{222}$Rn to reach equilibrium. The analysis of $^{226}$Ra in the digested samples were the same as for water.

During the preliminary study samples were dried and sealed in the same containers as mentioned in 3.5.2. The $^{226}$Ra concentrations proved to be below the detection limit of this method. During the main investigation, samples were digested as described above and the resulting solution/slurry was sealed in a bubble flask, and analysed as for $^{226}$Ra in water (3.5.1). For uranium analysis, subsamples of the ashed biological material were analysed using the NAA technique as described in 3.5.2.

## 3.6 Concentration Ratios

Concentration ratios are discussed in section 2.6.3. In this study two types of concentration ratios were calculated, namely CRs and CRw which were the ratios between the plant or animal and soil and water concentrations, respectively.

The CR values were calculated as following:

$$CRs = [Co]/[Cs]$$

Where: [Co] = Concentration of radionuclide in organism (U: $\mu g.g^{-1}$ (fresh) ; $^{226}$Ra: $Bq.g^{-1}$ (fresh))

[Cs] = Concentration of radionuclide in soil (U: $\mu g.g^{-1}$ (dry) ; $^{226}$Ra: $Bq.g^{-1}$ (dry))

$CRw = [Co]/[Cw]$

Where: $[Co]$ = Concentration of radionuclide in organism
(U: $\mu g.g^{-1}$ (fresh) ; $^{226}Ra$: $Bq.g^{-1}$ (fresh))
$[Cw]$ = Concentration of radionuclide in water
(U: $\mu g.m\ell^{-1}$ ; $^{226}Ra$: $Bq.m\ell^{-1}$)
or: (U: $\mu g.g^{-1}$ ; $^{226}Ra$: $Bq.g^{-1}$) as 1ml $H_2O$ = 1g

Note in Tables 4.5 - 4.18 that the concentration data are given for ashed mass and per litre of water.

## 3.7 Scenarios for dose assessment modelling

The GENII code allows several scenarios to be modelled. For this project the following options were chosen:

- Far field scenario
- Dose to a maximum individual adult from chronic exposure
- 1 year intake period
- 50 year committed dose period
- Exposure paths: a) drinking water
  b) consumption of irrigated foodstuffs
- Radionuclides, $^{238}U$ and $^{226}Ra$

Exposure pathways such as inhalation and swimming in contaminated water were not considered. The option for transport in environmental media was not used because the radionuclide concentrations in the water and foodstuffs consumed were assumed known as determined by field and pot experiment measurements.

The quantitative details of the input parameters used for radionuclide concentrations in water and food, volume or mass of food consumed annually all as fresh mass are given in Table 3.6. Concentration values chosen are meant to reflect realistic values found in the field. The consumption values chosen are conservative and reflect a possible extreme case for potential exposure. Usually the water found in streams near mining areas on the Witwatersrand are not used for drinking. It is assumed that vegetables consumed are all from contaminated irrigation water. Consumption of oats is separately evaluated assuming a daily breakfast intake and a high concentration value based on the Marievale sample.

**Table 3.6 Input parameters for GENII dose asessment code**

| Nuclide | Drinking water | | Vegetables | | | Cereals (oats) | | Fish | | Soil | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | Conc. Bq.$\ell^{-1}$ | Consumption $\ell$.y$^{-1}$ | Conc.* Bq.kg$^{-1}$ ($\mu$g.g$^{-1}$) | Consumption | | Conc.* Bq.kg$^{-1}$ ($\mu$g.g$^{-1}$) | Consumption kg.y$^{-1}$ | Conc.* Bq.kg$^{-1}$ ($\mu$g.g$^{-1}$) | Consumption kg.y$^{-1}$ | Conc.** Bq.kg$^{-1}$ ($\mu$g.g$^{-1}$) | Ingestion mg.y$^{-1}$ |
| | | | | Leafy kg.y$^{-1}$ | Roots kg.y$^{-1}$ | | | | | | |
| $^{238}$U | 0,5 | 730 | 0,3<br>(0,024) | 30 | 90 | 6,1<br>(0,494) | 80 | 0,12<br>(0,010) | 25 | 150<br>(12,2) | 410 |
| $^{226}$Ra | 0,04 | 730 | 0,25 | 30 | 90 | 8,6 | 80 | 2,5 | 25 | 25 | 410 |

* Concentration units per fresh mass ** Concentration units per dry mass

Abbreviation: Conc. - Concentration

# 4 RESULTS

Irrigation water and soil samples were collected for physical and chemical analysis at the same time as vegetable samples for radionuclide analysis. At the Marievale, Vlakfontein and Luipaardsvlei farms the same irrigation water samples, collected according to procedures described in Chapter 3, were used for both the physical-chemical and radionuclide analysis.

## 4.1 Physical and chemical characteristics of irrigation water and soil samples collected at the Marievale, Vlakfontein and Luipaardsvlei farms

No samples of the water and sediments were collected for uranium and $^{226}$Ra analysis during the preliminary survey of these radionuclides in selected aquatic organisms during 1989-90 (Table 4.5). However, during the main survey of 1990-92, irrigation water as well as soil samples were collected at the Marievale, Vlakfontein and Luipaardsvlei study areas for this purpose (Table 4.4). These are discussed in later sections.

### 4.1.1 Marievale farm

Results on the physical and chemical characteristics of the irrigation water and soil samples are summarized in Table 4.1. pH values for the six composite water samples were relatively constant, fluctuating between 7,5 and 7,9 (Table 4.1a). Electrical conductivity of the samples were high, reflecting the high amount of Total Dissolved Salts (TDS) in the irrigation water. Values for this parameter ranged between 1400 and 1700 $\mu$S.cm$^{-1}$ (Table 4.1a). Alkalinity values ranged between 72 and 102 mg.$\ell^{-1}$. Total hardness concentrations varied between 193 and 397 mg.$\ell^{-1}$ reflecting to some extent the effect of mining activities on the irrigation water. Ammonia, nitrite and nitrate values as well as concentrations of phosphates suggest some mild organic enrichment of the water (Table 4.1a).

Table 4.1a Water quality of the Marievale farm irrigation water

| Water Quality Parameter | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|
| pH | 7,5 | 7,6 | 7,6 | 7,6 | 7,7 | 7,9 |
| Electrical Conductivity ($\mu$S.cm$^{-1}$) | 1700 | 1700 | 1600 | 1600 | 1500 | 1400 |
| Alkalinity (mg.$\ell^{-1}$) | 79 | 72 | 79 | 100 | 75 | 102 |
| Total Hardness (mg.$\ell^{-1}$) | 215 | 397 | 234 | 311 | 193 | 387 |
| Ammonia (mg.$\ell^{-1}$) | <0,1 | <0,1 | <0,1 | <0,1 | <0,1 | <0,1 |
| Nitrite (mg.$\ell^{-1}$) | <0,01 | 0,01 | 0,02 | 0,01 | <0,01 | 0,01 |
| Nitrate (mg.$\ell^{-1}$) | 0,9 | 2,6 | 1,8 | 2,2 | 0,9 | 2,6 |
| Phosphate (mg.$\ell^{-1}$) | 0,3 | 0,2 | 0,6 | 0,3 | 0,6 | 0,3 |
| Sulphate (mg.$\ell^{-1}$) | 640 | 720 | 760 | 640 | 760 | 600 |

Soil pH values corresponded well with those of the irrigation water (Table 4.1b). A moderate percentage of organic material was present in the soils fluctuating between 0,43 and 0,70% (Table 4.1b).

**Table 4.1b Soil pH and organic content of several field plots on the Marievale farm**

| Field plot | pH | Organic content (%) |
|---|---|---|
| Carrots | 7,67 | 0,48 |
| Beetroot | 7,71 | 0,43 |
| Lettuce | 7,80 | 0,61 |
| Cabbage | 7,66 | 0,70 |
| Cauliflower | 7,73 | 0,68 |

### 4.1.2 Vlakfontein

At the Vlakfontein farm, the pH of the irrigation water was slightly alkaline with values for electrical conductivity (1400 $\mu S.cm^{-1}$), hardness (1419 $mg.\ell^{-1}$) as well as sulphates (950 $mg.\ell^{-1}$) clearly reflecting the effects of mining activity on the water quality of irrigation water used on this farm. The alkalinity of the water was low (16 $mg.\ell^{-1}$) with values for ammonia (0,22 $mg.\ell^{-1}$), nitrite (0,01 $mg.\ell^{-1}$), nitrate (2,4 $mg.\ell^{-1}$) and phosphate (<0,01 $mg.\ell^{-1}$) being generally lower than those at Marievale (Table 4.2a).

The soil pH was alkaline at all the randomly selected sites on the farm where samples collected varied between 7,26 and 7,68 (Table 4.2b). The organic content of the soils were on the average slightly higher than that of the Marievale farm.

Table 4.2a **Water quality of the Vlakfontein irrigation water**

| Water Quality Parameter | Irrigation water pump sample |
|---|---|
| pH | 7,1 |
| Electrical Conductivity ($\mu S.cm^{-1}$) | 1400 |
| Alkalinity ($mg.\ell^{-1}$) | 16 |
| Total Hardness ($mg.\ell^{-1}$) | 1419 |
| Ammonia ($mg.\ell^{-1}$) | 0,22 |
| Nitrite ($mg.\ell^{-1}$) | 0,01 |
| Nitrate ($mg.\ell^{-1}$) | 2,4 |
| Phosphate ($mg.\ell^{-1}$) | <0,01 |
| Sulphate ($mg.\ell^{-1}$) | 950 |

Table 4.2b **Soil pH and organic content of randomly collected soil samples at Vlakfontein farm**

| Sample No. | pH | Organic Content (%) |
|---|---|---|
| 1 | 7,68 | 0,55 |
| 2 | 7,47 | 1,32 |
| 3 | 7,31 | 0,76 |
| 4 | 7,26 | 0,98 |
| Mean | | 0,90 |

### 4.1.3 Luipaardsvlei

The water quality parameters at the three adjacent farms as well as the soil conditions (Tables 4.3a &b) showed prevailing conditions to be mildly acidic to alkaline. Water pH values fluctuated between 6,4 and 8,1 (Table 4.3a). In all three cases, the effect of mine acid waters on the electrical conductivity, hardness and, to a lesser extent, sulphates are reflected in the results. The alkalinity of the irrigation water at all three farms remained below 50 mg.$\ell^{-1}$ (Table 4.3a).

There was a considerable difference in the pH of the soils at the three farms fluctuating between 4,34 and 8,23 (Table 4.3b). These differences in soil pH has a marked effect on the solubility, speciation and bio-availability of uranium, $^{226}$Ra and other trace elements at these sites.

**Table 4.3a Water quality of the Luipaardsvlei irrigation water**

| Water Quality Parameter | LP1 | LP2 | LP3 |
|---|---|---|---|
| pH | 8,1 | 6,4 | 6,9 |
| Electrical Conductivity (µS.cm$^{-1}$) | 800 | 1100 | 900 |
| Alkalinity (mg.$\ell^{-1}$) | 47 | 22 | 35 |
| Total Hardness (mg.$\ell^{-1}$) | 219 | 148 | 185 |
| Ammonia (mg.$\ell^{-1}$) | 0,039 | 0,116 | 0,142 |
| Nitrite (mg.$\ell^{-1}$) | 0,01 | 0,01 | 0,01 |
| Nitrate (mg.$\ell^{-1}$) | 0,6 | 1,1 | 0,9 |
| Phosphate (mg.$\ell^{-1}$) | 0,05 | 0,16 | 0,10 |
| Sulphate (mg.$\ell^{-1}$) | 140 | 165 | 125 |

**Table 4.3b Soil pH and organic content of the three Luipaardvlei farms**

| Farm | pH | Organic Content (%) |
|---|---|---|
| LP1 | 8,23 | 0,67 |
| LP2 | 4,34 | 0,65 |
| LP3 | 5,30 | 0,88 |

## 4.2 Uranium and $^{226}$Ra in the water and soils of the Marievale, Vlakfontein and Luipaardsvlei study areas

According to the results obtained, concentrations for both uranium and $^{226}$Ra were significantly lower than their respective standards laid down for drinking water at both the Marievale and Vlakfontein farms. At Luipaardsvlei, however, the composite irrigation water samples obtained from three adjacent localities yielded variable results with two values for uranium of more than 50 µg.$\ell^{-1}$. The highest of these values exceeded 56 µg.$\ell^{-1}$ (Table 4.4a). In both cases the drinking water guideline value of 44 µg.$\ell^{-1}$ for uranium (Funke, 1990) was exceeded. Somewhat different conditions existed for $^{226}$Ra, where the mean and individual concentrations obtained for the various composite water samples at both Marievale and Vlakfontein were less than one tenth of that laid down for drinking water standards (Table 4.4a). Even at the Luipaardsvlei localities, where comparatively high uranium concentrations were recorded (Table 4.4a), the concentrations for $^{226}$Ra were on the average still at least one tenth of that of the drinking water guideline (Funke, 1990).

Table 4.4a Concentrations of uranium and $^{226}Ra$ in water samples collected at the Marievale, Vlakfontein and Luipaardsvlei farms.

| Irrigation cycle (h) | Marievale | | Water sample No. | Vlakfontein | | Water sample No. | Luipaardsvlei | | Location | Drinking water guideline values | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | Uranium $\mu g.\ell^{-1}$ | Radium $Bq.\ell^{-1}$ | | Uranium $\mu g.\ell^{-1}$ | Radium $Bq.\ell^{-1}$ | | Uranium $\mu g.\ell^{-1}$ | Radium $Bq.\ell^{-1}$ | | Uranium $\mu g.\ell^{-1}$ | Radium $Bq.\ell^{-1}$ |
| 1 | 16,8±0,6 | <0,007 | Farm ave. | 28,2±0,8 | 0,075±0,014 | 1 | 52,6±1,4 | 0,018±0,006 | Dr. water[1] | 44 | 0,19 |
| 2 | 17,3±0,7 | 0,02±0,01 | | | | 2 | 10,6±0,3 | 0,022±0,008 | | | |
| 3 | 16,5±0,6 | 0,01±0,007 | | | | 3 | 56,1±1,5 | 0,018±0,007 | | | |
| 4 | 15,6±0,6 | 0,02±0,01 | | | | Mean | 39,8 | 0,019 | | | |
| 5 | 16,2±0,6 | 0,016±0,007 | | | | | | | | | |
| 6 | 17,2±0,7 | 0,012±0,003 | | | | | | | | | |
| Mean | 16,6 | 0,014 | | | | | | | | | |

[1] Uranium and radium in drinking water (South Africa) (Funke, 1990)

**Table 4.4b** Concentrations of uranium and $^{226}$Ra in soil samples collected at the Marievale, Vlakfontein and Luipaardsvlei farms.

| Field plot | Marievale | | Field plot No. | Vlakfontein | | Field plot No. | Luipaardsvlei | | Location | Sediment guideline values | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | Uranium $\mu g.g^{-1}$ | Radium $Bq.g^{-1}$ | | Uranium $\mu g.g^{-1}$ | Radium $Bq.g^{-1}$ | | Uranium $\mu g.g^{-1}$ | Radium $Bq.g^{-1}$ | | Uranium $\mu g.g^{-1}$ | Radium $Bq.g^{-1}$ |
| Lettuce | --- | 0,025±0,004 | 1 | 5,63±0,16 | (0,001) | 1 | 3,1±0,1 | (0,03) | Nat. soil[1] | --- | 0,004-0,126 |
| Cabbage | --- | 0,019±0,012 | 2 | 7,75±0,19 | 0,032±0,016 | 2 | 13,6±0,3 | (0,02) | U-mine[2] | 2,8-5,6 | --- |
| Cauliflower | --- | 0,019±0,012 | 3 | 6,40±0,17 | (0,005) | 3 | 40,2±0,8 | 0,066±0,012 | U-control[3] | 1,5 | --- |
| Beetroot | --- | 0,029±0,006 | 4 | 4,86±0,17 | 0,023±0,009 | Mean | 19,0 | (0,039) | U-core[4] | 3,8-760 | --- |
| Carrots | --- | 0,034±0,003 | Irr. pump | 5,77±0,16 | 0,03±0,01 | | | | | | |
| Mean | --- | 0,025 | Mean | 6,08 | 0,028 | | | | | | |

[1] Radium in natural soils (IAEA, 1990)

[2] Uranium nearby U-mine in agricultural soil (Morishima *et al.*, 1977)

[3] Uranium in control soil (Tracy *et al.*, 1983)

[4] Uranium in soil core 15-30 cm underground (Tracy *et al.*, 1983)

Concentrations of uranium and $^{226}$Ra per dry mass soil.

No specific standards are available for uranium and $^{226}$Ra in soils. However, concentration values were obtained for soils in the vicinity of uranium mines in other countries (Morishima *et al.*, 1977 ; Tracy *et al.*, 1983) as shown in Table 4.4b. In the case of $^{226}$Ra, concentrations for radium in natural soils are provided by the IAEA (1990). In uranium soil core samples an extremely high concentration for this radionuclide (3,8 - 760 $\mu$g.g$^{-1}$ : Table 4.4b) was recorded in the USA (Tracy *et al.*, 1983). Concentration values for uranium was only obtained from the field plots at the Vlakfontein and Luipaardsvlei areas. In the case of Vlakfontein, individual and mean concentrations (6,08 $\mu$g.g$^{-1}$) all exceeded those found for soils in Japan (2,8 - 5,6 $\mu$g.g$^{-1}$ : Morishima *et al.*, 1977) and in some cases the USA (3,8 - 760 $\mu$g.g$^{-1}$ : Tracy *et al.*, 1983) (Table 4.4b). At the Luipaardsvlei farms, a considerable variation in uranium concentration was found in the soils, which varied from 3,1 to 40,2 $\mu$g.g$^{-1}$. Uranium concentrations for these soils exeeded in all three cases those recorded in Japan and USA (Table4.4b)

Mean and individual concentrations for $^{226}$Ra in the soils at Marievale, Vlakfontein and Luipaardsvlei were all in the range of concentrations quoted for natural soil by the IAEA (1990) (Table 4.4b).

### 4.3 Uranium and $^{226}$Ra concentrations in some aquatic organisms

The concentrations of uranium and $^{226}$Ra in selected aquatic plants, macro-invertebrates, fish and aquatic birds collected during the preliminary survey are listed in Table 4.5. Variable but relatively high concentrations were obtained for the emergent, floating and submerged aquatic weeds collected at four different localities from the mining areas of the East Rand. Concentrations for uranium fluctuated between a highest of 111 $\mu$g.g$^{-1}$ (*Typha capensis* : Blesbokspruit) and a lowest of 12,5 $\mu$g.g$^{-1}$ (*T.capensis* :Germiston lake) (Table 4.5a). In the case of $^{226}$Ra, values for *T. capensis* were again the highest in the Blesbokspruit (0,153 Bq.g$^{-1}$) followed by *Arundo donax* with a $^{226}$Ra concentration of 0,195 Bq.g$^{-1}$ (Germiston lake) (Table 4.5a). Comparatively high concentrations of $^{226}$Ra were recorded for submerged and rooted floating weeds. The $^{226}$Ra concentration for *Marsilea macrocarpa* was 0,082 Bq.g$^{-1}$ and for *Lagarosiphon muscoides* and *Potamogeton pectinatus* 0,305 Bq.g$^{-1}$ and 0,178 Bq.g$^{-1}$, respectively (Table 4.5a).

During the main survey, uranium and $^{226}$Ra analysis were also made on *Spirogyra* sp. and *P. pectinatus* from a stream at Luipaardsvlei. Concentrations for uranium was comparatively high for both *Spirogyra* sp. (52,6 $\mu$g.g$^{-1}$) and *P. pectinatus* (85,9 $\mu$g.g$^{-1}$) (Table 4.5b). In both cases (*Spirogyra* sp.: 1,27 Bq.g$^{-1}$ ; *P. pectinatus:* 0,76 Bq.g$^{-1}$) values obtained for $^{226}$Ra were, however, relatively low (Table 4.5b). Compared to the aquatic plants, the concentration of uranium in the freshwater crab, *Potamonautes warreni*, was considerably lower than in the plants (1,37 $\mu$g.g$^{-1}$ : Natalspruit). Its concentration of $^{226}$Ra was 0,033 Bq.g$^{-1}$ (Table 4.5a).

Comparing both the uranium and $^{226}$Ra concentrations obtained for the different fish species analyzed, both radionuclides appeared in both instances to be also distinctly lower than those found for the associated aquatic plants. This particularly applies to

Table 4.5a Aquatic plants, macro-invertebrates, fish and aquatic birds collected for uranium and $^{226}$Ra analysis during 1989-1990.

| Organism | Blesbokspruit | | Elsburgspruit | | Germiston lake | | Natalspruit | |
|---|---|---|---|---|---|---|---|---|
| | Uranium ($\mu g.g^{-1}$, ash) | $^{226}$Ra ($Bq.g^{-1}$, ash) | Uranium ($\mu g.g^{-1}$, ash) | $^{226}$Ra ($Bq.g^{-1}$, ash) | Uranium ($\mu g.g^{-1}$, ash) | $^{226}$Ra ($Bq.g^{-1}$, ash) | Uranium ($\mu g.g^{-1}$, ash) | $^{226}$Ra ($Bq.g^{-1}$, ash) |
| AQUATIC PLANTS | | | | | | | | |
| *Azolla filiculoides* | 47,9 | <0,099 | --- | --- | --- | --- | --- | --- |
| *Marsilea macrocarpa* | 37,9 | <0,092 | 58,4 | 0,082 ± 0,011 | --- | --- | --- | --- |
| *Lemna major* | --- | --- | --- | --- | --- | --- | 31,7 | <0,109 |
| *Lagarosiphon muscoides* | --- | --- | --- | --- | --- | --- | 26,4 | 0,305 ± 0,017 |
| *Potamogeton pectinatus* | --- | --- | 7,7 | <0,039 | --- | --- | 18,0 | 0,178 ± 0,009 |
| *Typha capensis* | 111 | 0,153 ± 0,031 | 18,5 | <0,037 | 12,5 | 0,097 ± 0,013 | 15,3 | <0,076 |
| *Arundo donax* | 37,7 | <0,162 | 15,8 | <0,071 | 18,1 | 0,195 ± 0,015 | 14,2 | <0,087 |
| MACRO-INVERTEBRATES | | | | | | | | |
| *Potamonautes warreni* | --- | --- | --- | --- | --- | --- | 1,37 | <0,033 |
| FISH | | | | | | | | |
| *Barbus aeneus* | --- | --- | --- | --- | --- | --- | 0,52 | <0,064 |
| *Cyprinus carpio* | 0,89 | <0,045 | --- | --- | --- | --- | --- | --- |
| *Clarias gariepinus* | 4,99 | <0,036 | --- | --- | 2,42 | <0,065 | 0,60 | <0,063 |
| *Tilapia sparrmanii* | 1,33 | <0,081 | --- | --- | --- | --- | --- | --- |
| AQUATIC BIRDS | | | | | | | | |
| *Fulica cristata* | --- | --- | --- | --- | --- | --- | 0,56 | <0,034 |
| *Phalacrocorax carbo* | --- | --- | --- | --- | --- | --- | <0,17 | <0,074 |

Table 4.5b Aquatic plants collected at Luipaardsvlei during 1991-1992 and analyzed for uranium and $^{226}$Ra.

| Aquatic plants | Uranium ($Bq.g^{-1}$) | Radium ($Bq.g^{-1}$) |
|---|---|---|
| *Spirogyra* sp. | 52,6 ± 0,9 | 1,27 ± 0,01 |
| *Potamogeton pectinatus* | 85,9 ± 0,8 | 0,76 ± 0,01 |

uranium, where values fluctuated between 4,99 $\mu g.g^{-1}$ (*Clarias gariepinus*: Blesbokspruit) and 0,52 $\mu g.g^{-1}$ (*Barbus aeneus* : Natalspruit) (Table 4.5a). The concentration data given here are based on complete fish samples which gives a higher value than for the edible portion on its own. This is because radium accumulates in the bone and invariably the gut contains sediment and higher concentrations than the flesh.

In the the two aquatic birds analyzed for uranium, levels were distincly lower for *Fulica cristata* (0,56 $\mu g.g^{-1}$) and *Phalacrocorax carbo* (<0,17 $\mu g.g^{-1}$) than for the fish collected. However, concentrations for $^{226}$Ra in both these birds remained about the same as for the crab and the fish (Table 4.5a).

## 4.4 Uranium and $^{226}$Ra in selected vegetable crops and cattle fodder from the three different farms at Marievale, Vlakfontein and Luipaardsvlei

During the growing season of 1989-1992 collections were made of vegetable crops and cattle fodder at the three farms where water and soil samples were also analyzed for uranium and $^{226}$Ra. The first investigation into the occurrence of these radionuclides in the vegetable crops took place at the Marievale farm. At this farm, carrots used in the analysis were pooled from different areas of the land from which it was cultivated. The other crops were randomly collected from the various field plots and individually-analyzed to determine possible variation in concentrations (Table 4.6 - 4.9).

All material collected of the different crops were first dried and ashed and then sealed in bottles for later analysis according to procedures described in Chapter 3. This procedure lead to values obtained for both uranium and $^{226}$Ra which in most cases were below the detection limit for this particular analytical procedure. During the subsequent sampling and analysis of vegetable crops at Vlakfontein and Luipaardsvlei, all biological samples were ashed but the materials were then acid digested and analyzed as liquid samples according to procedures described in Chapter 3. This approach enabled the detection of lower concentrations of both radionuclides and a considerable increase in the accuracy of the results.

Note: ***Concentration results are given for ash mass for plants, dry mass for soil and liquid mass for water while concentration ratios use the fresh mass concentrations for plants.***

### 4.4.1 Marievale farm

Uranium values for the different vegetable crops cultivated at the Marievale farm are listed in Tables 4.6 - 4.9. The root vegetables yielded the highest values for uranium namely 0,23 $\mu g.g^{-1}$ for carrots, and 0,21 $\mu g.g^{-1}$ to 0,36 $\mu g.g^{-1}$ for beetroot. This was followed by cauliflower with uranium concentrations varying between 0,1 $\mu g.g^{-1}$ and 0,17 $\mu g.g^{-1}$. Lowest concentrations occurred in cabbage where concentrations fluctuated between 0,02 $\mu g.g^{-1}$ and 0,027 $\mu g.g^{-1}$ (Table 4.8). Where values were obtained for $^{226}Ra$, the concentration levels for the different crops were similar, fluctuating around 0,01 $Bq.g^{-1}$ for both the root and leafy vegetable crops (Table 4.8). As shown in Tables 4.6 - 4.9, only the water/vegetable ratio for uranium was calculated whereas both water and soil concentrations of $^{226}Ra$ were used to determine the concentration ratios (CR) for the different crops. Highest CRw values for uranium were recorded for beetroot fluctuating between 0,33 and 0,46 (Table 4.6). This was followed by carrots (0,16) with variable ratios obtained for cauliflower (0,07 - 0,12), lettuce (0,03 - 0,06) and cabbage (0,0095 - 0,12) (Tables 4.6 - 4.9). Although the results were limited, indications are that the highest CR-values for both radionuclides occurred in the root vegetables.

With the exception of beetroot, CR-values for $^{226}Ra$ for both the water and soil were not only variable in both cases, but did not display any distinct tendencies. In the case of beetroot the CRw values were approximately double that of the soil (Table 4.6). In all the other cases, CR-values for both water and soil were extremely low, varying between 0,002 (Lettuce : CRs) and 5,6 (Cabbage : CRw) (Tables 4.6 - 4.9).

From the CR-values obtained for both uranium and $^{226}Ra$, it can be deduced that these crops were not very efficient in accumulating these radionuclides from either the irrigation water or the soils in which they were grown.

### 4.4.2 Vlakfontein farm

The concentrations of uranium and $^{226}Ra$ in selected vegetable crops cultivated on this farm are summarized in Table 4.10. In addition to carrots, beetroot and lettuce, spinach, leeks and turnips were additional vegetables investigated for their ability to accumulate uranium and $^{226}Ra$ from the soil.

Concentration values for uranium and $^{226}Ra$ in the different vegetable crops grown at the Vlakfontein farm revealed interesting results. Where the root vegetables at Marievale farm generally had higher concentrations of both uranium and $^{226}Ra$ compared to the leafy vegetables, this situation did not apply to the spinach grown at Vlakfontein. Spinach leaves had the highest concentrations of all the crops with 2,5 $\mu g.g^{-1}$ (ash) for uranium and 0,023 $Bq.g^{-1}$ (ash) for $^{226}Ra$ (Table 4.10). Values obtained for leeks and lettuce were correspondingly lower than those for beetroot and carrots, but turnips yielded intermediate concentrations of 0,13 $\mu g.g^{-1}$ for uranium and 0,0014 $Bq.g^{-1}$ for $^{226}Ra$, which in both cases were between those for leafy and root vegetables (Table 4.10).

Table 4.6 Concentrations of uranium and $^{226}$Ra in root vegetables (carrots and beetroot) collected at Marievale farm with an indication of CRs (soil) and CRw (water) values.

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | Uranium ($\mu g.g^{-1}$ ash, soil) ($\mu g.\ell^{-1}$ water) | $^{226}$Ra ($Bq.g^{-1}$ ash, soil) ($Bq.\ell^{-1}$ water) |
|---|---|---|---|---|---|
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,034 ± 0,0003 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Carrots : $\bar{X}$ ± S.D. | 224,56 | 23,32 | 2,54 | 0,23 ± 0,04 | (0,01) |
| CRs | --- | --- | --- | --- | (0,003) |
| CRw | --- | --- | --- | 0,16 ± 0,03 | (7,10) |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,029 ± 0,006 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Beetrt. 1 : $\bar{X}$ ± S.D. | 95,34 | 16,29 | 1,88 | 0,36 ± 0,05 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,43 ± 0,07 | N.D |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,029 ± 0,006 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Beetrt. 2 : $\bar{X}$ ± S.D. | 81,09 | 14,08 | 1,18 | N.D | (0,01) |
| CRs | --- | --- | --- | --- | (0,005) |
| CRw | --- | --- | --- | N.D | (10,5) |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,029 ± 0,006 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Beetrt. 3 : $\bar{X}$ ± S.D. | 110,07 | 19,15 | 3,96 | 0,21 ± 0,04 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,46 ± 0,1 | N.D |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,029 ± 0,006 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Beetrt. 4 : $\bar{X}$ ± S.D. | 82,6 | 14,68 | 2,28 | 0,2 ± 0,04 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,33 ± 0,07 | N.D |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,029 ± 0,006 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Beetrt. 5 : $\bar{X}$ ± S.D. | 104,9 | 18,55 | 2,68 | N.D | (0,01) |
| CRs | --- | --- | --- | --- | (0,006) |
| CRw | --- | --- | --- | N.D | (12,0) |

° Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}$Ra in soil expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for a dried sample.
Uranium and $^{226}$Ra in water expressed as $\mu g.\ell^{-1}$ and $Bq.\ell^{-1}$ respectively.
Uranium and $^{226}$Ra in vegetables expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1m$\ell$)

For $^{238}$U: 1Bq = 80,9 µg

Table 4.7 Concentrations of uranium and $^{226}$Ra in lettuce collected at Marievale farm with an indication of CRs (soil) and CRw (water) values.

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | Uranium ($\mu g.g^{-1}$ ash, soil) ($\mu g.\ell^{-1}$ water) | $^{226}$Ra ($Bq.g^{-1}$ ash, soil) ($Bq.\ell^{-1}$ water) |
|---|---|---|---|---|---|
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | --- | 0,025 ± 0,004 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Lettuce 1 : $\bar{X} \pm$ S.D. | 238,33 | 9,64 | 1,36 | 0,09 ± 0,03 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,03 ± 0,01 | N.D |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | --- | 0,025 ± 0,004 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Lettuce 2 : $\bar{X} \pm$ S.D. | 321,70 | 12,98 | 1,81 | 0,17 ± 0,04 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,06 ± 0,02 | N.D |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | --- | 0,025 ± 0,004 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Lettuce 3 : $\bar{X} \pm$ S.D. | 326,01 | 12,84 | 2,09 | N.D | (0.01) |
| CRs | --- | --- | --- | --- | (0,0025) |
| CRw | --- | --- | --- | N.D | (4,5) |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | --- | 0,025 ± 0,004 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Lettuce 4 : $\bar{X} \pm$ S.D. | 309,51 | 12,42 | 1,61 | N.D | (0,01) |
| CRs | --- | --- | --- | --- | (0,002) |
| CRw | --- | --- | --- | N.D | (3,7) |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | --- | 0,025 ± 0,004 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 16,6 ± 0.6 | 0,014 ± 0,007 |
| Lettuce 5 : $\bar{X} \pm$ S.D. | 162,09 | 6,63 | 0,97 | 0,08 ± 0,03 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,03 ± 0,01 | N.D |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}$Ra in soil expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for a dried sample.
Uranium and $^{226}$Ra in water expressed as $\mu g.\ell^{-1}$ and $Bq.\ell^{-1}$ respectively.
Uranium and $^{226}$Ra in vegetables expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1m$\ell$)

For $^{238}$U: 1Bq = 80,9 µg

Table 4.8 Concentrations of uranium and $^{226}$Ra in cabbage collected at Marievale farm with an indication of CRs (soil) and CRw (water) values.

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | Uranium (µg.g$^{-1}$ ash, soil) (µg.ℓ$^{-1}$ water) | $^{226}$Ra (Bq.g$^{-1}$ ash, soil) (Bq.ℓ$^{-1}$ water) |
|---|---|---|---|---|---|
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,019 ± 0,012 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Cabbage 1 : $\bar{X}$ ± S.D. | 219,33 | 18,28 | 1,87 | (0,02) | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | --- | N.D |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,019 ± 0,012 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Cabbage 2 : $\bar{X}$ ± S.D. | 229,07 | 18,47 | 1,65 | 0,022 ± 0,02 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,0095 ± 0,009 | N.D |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,019 ± 0,012 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Cabbage 3 : $\bar{X}$ ± S.D. | 224,1 | 18,63 | 1,63 | 0,027 ± 0,02 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,012 ± 0,009 | N.D |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | --- | 0,019 ± 0,012 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,007 |
| Cabbage 4 : $\bar{X}$ ± S.D. | 232,38 | 18,58 | 1,82 | N.D | 0,01 ± 0,005 |
| CRs | --- | --- | --- | N.D | 0,004 ± 0,003 |
| CRw | --- | --- | --- | N.D | 5,6 ± 3,2 |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}$Ra in soil expressed as µg.g$^{-1}$ and Bq.g$^{-1}$ respectively for a dried sample.
Uranium and $^{226}$Ra in water expressed as µg.ℓ$^{-1}$ and Bq.ℓ$^{-1}$ respectively.
Uranium and $^{226}$Ra in vegetables expressed as µg.g$^{-1}$ and Bq.g$^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1mℓ)

For $^{238}$U: 1Bq = 80,9 µg

Table 4.9 Concentrations of uranium and $^{226}$Ra in cauliflower collected at Marievale farm with an indication of CRs (soil) and CRw (water) values.

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | Uranium ($\mu g.g^{-1}$ ash, soil) ($\mu g.\ell^{-1}$ water) | $^{226}$Ra ($Bq.g^{-1}$ ash, soil) ($Bq.\ell^{-1}$ water) |
|---|---|---|---|---|---|
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | --- | 0,019 ± 0,012 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,0074 |
| Caulif. 1 : $\bar{X} \pm$ S.D. | 190,56 | 18,24 | 2,18 | 0,1 ± 0,03 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,07 ± 0,02 | N.D |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | --- | 0,019 ± 0,012 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,0074 |
| Caulif. 2 : $\bar{X} \pm$ S.D. | 146,32 | 13,96 | 1,65 | 0,17 ± 0,04 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,12 ± 0,03 | N.D |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | --- | 0,019 ± 0,012 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,0074 |
| Caulif. 3 : $\bar{X} \pm$ S.D. | 188,77 | 18,08 | 2,11 | N.D | (0,01) |
| CRs | --- | --- | --- | --- | (0,006) |
| CRw | --- | --- | --- | N.D | (0,008) |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | --- | 0,019 ± 0,012 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 16,6 ± 0,6 | 0,014 ± 0,0074 |
| Caulif. 4 : $\bar{X} \pm$ S.D. | 191,02 | 19,28 | 2,22 | 0,13 ± 0,03 | N.D |
| CRs | --- | --- | --- | --- | N.D |
| CRw | --- | --- | --- | 0,09 ± 0,02 | N.D |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}$Ra in soil expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for a dried sample.
Uranium and $^{226}$Ra in water expressed as $\mu g.\ell^{-1}$ and $Bq.\ell^{-1}$ respectively.
Uranium and $^{226}$Ra in vegetables expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1mℓ)

For $^{238}$U: 1Bq = 80,9 µg

Table 4.10 Concentrations of uranium and $^{226}$Ra in selected vegetable crops collected at Vlakfontein farm with an indication of CRs (soil) and CRw (water) values.

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | Uranium ($\mu g.g^{-1}$ ash, soil) ($\mu g.\ell^{-1}$ water) | $^{226}$Ra ($Bq.g^{-1}$ ash, soil) ($Bq.\ell^{-1}$ water) |
|---|---|---|---|---|---|
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 6,1 ± 0,2 | 0,027 ± 0,001 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 28,2 ± 0,80 | 0,075 ± 0,014 |
| Spinach: $\bar{X}$ ± S.D. | 2593 | 227 | 51,1 | 2,5 ± 0,1 | 0,023 ± 0,001 |
| CRs | --- | --- | --- | 0,008 ± 0,0006 | 0,02 ± 0,008 |
| CRw | --- | --- | --- | 1,75 ± 0,01 | 6,0 ± 0,01 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 6,1 ± 0,2 | 0,027 ± 0,001 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 28,2 ± 0,80 | 0,075 ± 0,014 |
| Leek: $\bar{X}$ ± S.D. | 880 | 131 | 8,3 | 0,07 ± 0,02 | (0,0011) |
| CRs | --- | --- | --- | 0,00011 ± 0,00003 | (0,00037) |
| CRw | --- | --- | --- | 0,023 ± 0,008 | (0,14) |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 6,1 ± 0,2 | 0,027 ± 0,001 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 28,2 ± 0,80 | 0,075 ± 0,014 |
| Turnip: $\bar{X}$ ± S.D. | 1613 | 130 | 19,5 | 0,13 ± 0,02 | 0,0014 ± 0,0002 |
| CRs | --- | --- | --- | 0,00026 ± 0,00005 | 0,00063 ± 0,00042 |
| CRw | --- | --- | --- | 0,056 ± 0,008 | 0,23 ± 0,006 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 6,1 ± 0,2 | 0,027 ± 0,001 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 28,2 ± 0,80 | 0,075 ± 0,014 |
| Lettuce: $\bar{X}$ ± S.D. | 2369 | 96,9 | 18,5 | 0,17 ± 0,03 | 0,0017 ± 0,0004 |
| CRs | --- | --- | --- | 0,00022 ± 0,00005 | 0,00048 ± 0,00036 |
| CRw | --- | --- | --- | 0,047 ± 0,001 | 0,18 ± 0,010 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 6,1 ± 0,2 | 0,027 ± 0,001 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 28,2 ± 0,80 | 0,075 ± 0,014 |
| Beetroot: $\bar{X}$ ± S.D. | 1487 | 256 | 24,8 | 1,7 ± 0,1 | 0,003 ± 0,0005 |
| CRs | --- | --- | --- | 0,0047 ± 0,0004 | 0,0019 ± 0,0012 |
| CRw | --- | --- | --- | 1,0 ± 0,005 | 0,67 ± 0,014 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 6,1 ± 0,2 | 0,027 ± 0,001 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 28,2 ± 0,80 | 0,075 ± 0,014 |
| Carrot: $\bar{X}$ ± S.D. | 1456 | 171 | 20,7 | 1,7 ± 0,20 | 0,0041 ± 0,0005 |
| CRs | --- | --- | --- | 0,004 ± 0,0006 | 0,0022 ± 0,0013 |
| CRw | --- | --- | --- | 0,86 ± 0,009 | 0,78 ± 0,017 |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}$Ra in soil expressed as $ug.g^{-1}$ and $Bq.g^{-1}$ respectively for a dried sample.
Uranium and $^{226}$Ra in water expressed as $\mu g.\ell^{-1}$ and $Bq.\ell^{-1}$ respectively.
Uranium and $^{226}$Ra in vegetables expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1mℓ)

For $^{238}$U: 1Bq = 80,9 μg

The CR-values for the soil and water were roughly reflected by the actual concentrations of both radionuclides in the soil and water. Highest CRw values of 1,75 for uranium was recorded for spinach with a corresponding 0,008 for the CRs. The second highest CRw value, namely 0,86 was obtained for carrots. The CRs of uranium in carrots was, however, very low (0,004) and of the same order as for beetroot (0,0047) (Table 4.10). In the case of leeks, turnips and lettuce, the CR-values remained low fluctuating between 0,023 and 0,056 for water and 0,00011 to 0,00026 for soil (Table 4.10).

CRs-values for uranium were low, with a highest CR of 0,008 recorded for spinach, followed by beetroot (0,0047) and carrots (0,004). CRs-values for leeks, turnips and lettuce were of a similar order of magnitude, fluctuating between 0,00011 and 0,00026, respectively (Table 4.10).

The CRw-values for $^{226}$Ra for leeks, turnips, lettuce, beetroot and carrots were of the same order of magnitude, fluctuating between 0,14 and 0,78. The only exception was spinach, which yielded a value of 6,0 which was one order of magnitude higher than the latter. Nearly a similar picture emerged from the CRs-values for $^{226}$Ra. A highest CRs of 0,02 was recorded for spinach, while the other CRs-values ranged between 0,00037 (leeks) and 0,0022 (carrots).

There was a difference in the types of irrigation used at these farms. Where flood irrigation was practised at Vlakfontein, spray irrigation was used at Marievale. Factors such as soil-pH and species of these radionuclides involved may well have affected their concentrations in the different crops.

### 4.4.3 Luipaardsvlei

At Luipaardsvlei, spray irrigated fodder produced on the three adjacent farms as well as the milk from cows which were fed on this fodder, were analyzed for uranium and $^{226}$Ra. Cow dung was also analyzed to determine any possible declines in radionuclide concentrations during the digestion process. All results are summarized in Tables 4.11 - 4.13.

Considerable variation existed in the concentrations of uranium in the fodder from the three adjacent Luipaardsvlei farms varying from 5,0 $\mu g.g^{-1}$ to 107 $\mu g.g^{-1}$ (Table 4.11). Concentration values obtained for $^{226}$Ra varied much less with values fluctuating between 0,055 to 0,094 $Bq.g^{-1}$ (Table 4.11).

Calculating the CR-values for fodder from the soil and water for both uranium and $^{226}$Ra, comparatively high values were found in all cases. The CRw values at all three farms fluctuated between 0,46 and 92,2 for uranium and 22,9 and 41,0 for $^{226}$Ra. CRs-values on the other hand were much lower for both uranium (0,0025 - 0,072) and $^{226}$Ra (0,00011 - 2,5), being a reflection in the first instance of the actual concentration of these radionuclides in the soil (Table 4.11).

Milk produced by the cows on two of these farms was analyzed for the presence of uranium and $^{226}$Ra (Table 4.12). In both cases the uranium and $^{226}$Ra concentrations

Table 4.11 Luipaardsvlei (Localities 1-3 (LP1 - LP3): Water and sediment transfer to cattle fodder)

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | Uranium ($\mu g.g^{-1}$ ash, soil) ($\mu g.\ell^{-1}$ water) | $^{226}Ra$ ($Bq.g^{-1}$ ash, soil) ($Bq.\ell^{-1}$ water) |
|---|---|---|---|---|---|
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 3,1 ± 0,1 | (0,003) |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 52,6 ± 1,4 | 0,018 ± 0,006 |
| Fodder(LP1): $\bar{X}$ ± S.D. | 2917 | 285 | 14,2 | 5,0 ± 0,2 | 0,094 ± 0,029 |
| Crs | --- | --- | --- | 0,0079 ± 0,0006 | (1,5) |
| CRw | --- | --- | --- | 0,46 ± 0,03 | 25,4 ± 0,02 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 13,6 ± 0,3 | (0,02) |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 10,6 ± 0,3 | 0,022 ± 0,008 |
| Fodder(LP2): $\bar{X}$ ± S.D. | 1744 | 334 | 16 | 107 ± 1,5 | 0,055 ± 0,003 |
| Crs | --- | --- | --- | 0,072 ± 0,003 | (2,5) |
| CRw | --- | --- | --- | 92,2 ± 0,004 | 22,9 ± 0,01 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 40,2 ± 0,8 | 0,066 ± 0,012 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 56,1 ± 1,50 | 0,018 ± 0,007 |
| Fodder(LP3): $\bar{X}$ ± S.D. | 2032 | 283 | 24,6 | 8,15 ± 0,25 | 0,061 ± 0,018 |
| Crs | --- | --- | --- | 0,0025 ± 0,01 | 0,00011 ± 0,00005 |
| CRw | --- | --- | --- | 1,8 ± 0,0001 | 41,0 ± 0,03 |

Table 4.12 Luipaardsvlei (Localities 1-2 (LP1 - LP2): Cattle fodder to milk transfer)

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | Uranium ($\mu g.g^{-1}$ ash, fodder, milk) | $^{226}Ra$ ($Bq.g^{-1}$ ash, fodder, milk) |
|---|---|---|---|---|---|
| Fodder(LP1): $\bar{X}$ ± S.D. | 2917 | 295 | 14,2 | 5,0 ± 0,2 | 0,094 ± 0,029 |
| Milk (LP1): $\bar{X}$ ± S.D. | 5726 | 559 | 19,4 | (0,21) | 0,05 ± 0,02 |
| CRmilk/fodder | --- | --- | --- | (0,029) | 0,37 ± 0,0013 |
| Fodder(LP2): $\bar{X}$ ± S.D. | 1744 | 334 | 16 | 106,5 ± 1,5 | 0,055 ± 0,029 |
| Milk (LP2): $\bar{X}$ ± S.D. | 1364 | 147 | 25 | 0,22 ± 0,2 | 0,0051 ± 0,020 |
| CRmilk/fodder | --- | --- | --- | 0,044 ± 0,00003 | 0,185 ± 0,008 |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}Ra$ in soil expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for a dried sample.
Uranium and $^{226}Ra$ in water expressed as $\mu g.\ell^{-1}$ and $Bq.\ell^{-1}$ respectively.
Uranium and $^{226}Ra$ in vegetables expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1mℓ)

For $^{238}U$: 1Bq = 80,9 µg

Table 4.13 Luipaardsvlei (Localities 1-3 (LP1 - LP3): Cattle fodder to faeces transfer)

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | Uranium ($\mu g.g^{-1}$ ash, dung, fodder) | $^{226}Ra$ ($Bq.g^{-1}$ ash, dung, fodder) |
|---|---|---|---|---|---|
| Fodder(LP1): $\bar{X} \pm S.D.$ | 2917 | 285 | 14,2 | 5,0 ± 0,2 | 0,094 ± 0,029 |
| Dung (LP1): $\bar{X} \pm S.D.$ | 4031 | 820 | 20.3 | 22,5 ± 0,35 | 0,19 ± 0,02 |
| CRdung/fodder | --- | --- | --- | 4,66 ± 0,001 | 2,09 ± 0,004 |
| Fodder(LP2): $\bar{X} \pm S.D.$ | 1744 | 334 | 16 | 106,5 ± 1,50 | 0,055 ± 0,029 |
| Dung (LP2): $\bar{X} \pm S.D.$ | 2242 | 847 | 25,4 | 6,4 ± 0,2 | 0,04 ± 0,003 |
| CRdung/fodder | --- | --- | --- | 0,07 ± 0,00002 | 0,898 ± 0,005 |
| Fodder(LP3): $\bar{X} \pm S.D.$ | 2032 | 283 | 24,6 | 8,15 ± 0,25 | 0,061 ± 0,018 |
| Dung (LP3): $\bar{X} \pm S.D.$ | 2163 | 2051 | 34,8 | 2,3 ± 0,1 | 0,032 ± 0,011 |
| CRdung/fodder | --- | --- | --- | 0,375 ± 0,00034 | 0,697 ± 0,005 |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}Ra$ in soil expressed as $ug.g^{-1}$ and $Bq.g^{-1}$ respectively for a dried sample.
Uranium and $^{226}Ra$ in water expressed as $\mu g.\ell^{-1}$ and $Bq.\ell^{-1}$ respectively.
Uranium and $^{226}Ra$ in vegetables exoressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1mℓ)

For $^{238}U$: 1Bq = 80,9 μg

were considerably lower in the milk than the concentrations recorded in the fodder and the dung. Concentrations of both uranium and $^{226}Ra$ in milk were extremely low varying between 0,21 to 0,22 $\mu g.g^{-1}$ (U) and 0,0051 to 0,005 $Bq.g^{-1}$ ($^{226}Ra$) (Table 4.12). The milk/fodder CR-ratios were low in both cases, in particular uranium, where the CR-values fluctuated between 0,029 and 0,044. In the case of $^{226}Ra$, the CR values were higher, fluctuating between 0,185 and 0,37 (Table 4.12).

Uranium and radium concentrations obtained from dung samples from the three adjacent Luipaardsvlei farms as well as the dung/fodder ratios for these radionuclides are listed in Table 4.13. Variation was found to exist for uranium between the concentrations of the cow dung samples from the three adjacent farms. This also applied to the $^{226}Ra$ concentrations. Uranium concentration in the cow dung samples fluctuated between 2,3 and 22,5 $\mu g.g^{-1}$ whereas the concentrations for $^{226}Ra$ varied between 0,032 and 0,19 $Bq.g^{-1}$ (Table 4.13).

Comparing the CR dung/fodder values for uranium and $^{226}Ra$, considerable variation was obtained for the different farms. There was no relation in values for these two radionuclides, suggesting a number of factors which may have affected the bio-avialabilty of both these radionuclides (Table 4.13).

## 4.5 Experimental uptake of $^{226}$Ra by cabbage and beetroot

The experimental setup and application procedures for $^{226}$Ra during the pot plant experiment are described in Chapter 3. Background data on the $^{226}$Ra concentrations of the seedlings are listed in Table 4.14 whilst those on the $^{226}$Ra application on beetroot and cabbage are listed in Tables 4.15 - 4.18. Natural $^{226}$Ra concentrations in the beetroot and cabbage from unpolluted soils differed slightly being the highest in the beetroot again reflecting the better ability of the roots of these vegetables to accumulate this radionuclide from the soils.

**Table 4.14 Background values for vegetables used in experimental uptake of $^{226}$Ra**

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | $^{226}$Ra (Bq.g$^{-1}$ ash)(sed) |
|---|---|---|---|---|
| Beetroot background | 873 | 79,6 | 27,6 | 0,0091 ± 0,0006 |
| Cabbage background | 610 | 56,2 | 6,8 | 0,007 ± 0,001 |

In the case of beetroot (Table 4.15 - 4.16) there were some slight differences in the concentrations of $^{226}$Ra depending on the site of application of the radionuclide. In the case where the roots were irrigated, the concentrations of $^{226}$Ra in the root tissue (0,063 Bq.g$^{-1}$) was slightly higher than in the leaves (0,056 Bq.g$^{-1}$). Transfer of this radionuclide from the irrigated leaves to the roots were very effective, leading again to the higher concentration of $^{226}$Ra in the roots (0,111 Bq.g$^{-1}$). Where both roots and leaves were irrigated with the radionuclide containing water, the roots again were better able to bioconcentrate $^{226}$Ra. In the control plants, the levels of the natural $^{226}$Ra were also higher in the roots than in the leaves (Tables 4.15 - 4.16).

Where the roots were irrigated, the CRs-values showed no significant difference between the roots and the leaves. In the cases where the leaves and roots and leaves were irrigated, the CRs (roots analyzed) were distinctly higher than the CRs (leaves irrigated) ratios. There was also a marked difference between the CRs (roots analyzed) and CRs (leaves analyzed) for the controls, which cannot be explained (Tables 4.15 - 4.16), except to say that the soil concentration was below the detection limit.

In the case of the CRw-values for beetroot a similar picture emerged, showing a slightly higher CRw (leaves analyzed) ratio (0,032) than the CRw (roots analyzed) (0,029). In the case of the irrigated leaves and roots and leaves, the ratio again was in favour of the roots.

A comparison of the ability of the roots and leaves of the cabbage plants to accumulate $^{226}$Ra clearly showed the roots more capable of bioconcentrating this radionuclide. In all three types of irrigation systems the concentration ratio from soil to the roots were clearly the highest, varying between 0,044 (leaves irrigated) and 0,052 (roots irrigated) to 0,117 for roots and leaves irrigated (Tables 4.17 - 4.18). Control values also showed some difference, with the concentration of $^{226}$Ra being low in both cases.

## Table 4.15 Experimental uptake of $^{226}$Ra by the roots of beetroot

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | $^{226}$Ra (Bq.g$^{-1}$ ash, soil) (Bq.$\ell^{-1}$, water) |
|---|---|---|---|---|
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | 0,04 ± 0,015 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 22,5 |
| Beetroot (Roots irr.) : $\bar{X} \pm$ S.D. | 253 | 38,3 | 2,65 | 0,063 ± 0,006 |
| CRs | --- | --- | --- | 0,016 ± 0,008 |
| CRw | --- | --- | --- | 0,029 ± 0,003 |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | 0,084 ± 0,015 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 22,5 |
| Beetroot (Leaves irr.) : $\bar{X} \pm$ S.D. | 215 | 34,1 | 2,9 | 0,111 ± 0,007 |
| CRs | --- | --- | --- | 0,018 ± 0,004 |
| CRw | --- | --- | --- | 0,066 ± 0,004 |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | 0,082 ± 0,018 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 22,5 |
| Beetroot (Rt + Lvs irr.) : $\bar{X} \pm$ S.D. | 144 | 22,9 | 2,6 | 0,15 ± 0,009 |
| CRs | --- | --- | --- | 0,033 ± 0,009 |
| CRw | --- | --- | --- | 0,12 ± 0,007 |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | (0,022) |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | --- |
| Beetroot (Control) : $\bar{X} \pm$ S.D. | 375 | 51,9 | 4,9 | 0,02 ± 0,003 |
| CRs | --- | --- | --- | (0,012) |
| CRw | --- | --- | --- | --- |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}$Ra in soil expressed as µg.g$^{-1}$ and Bq.g$^{-1}$ respectively for a dried sample.
Uranium and $^{226}$Ra in water expressed as µg.$\ell^{-1}$ and Bq.$\ell^{-1}$ respectively.
Uranium and $^{226}$Ra in vegetables expressed as µg.g$^{-1}$ and Bq.g$^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1m$\ell$)

For $^{238}$U: 1Bq = 80,9 µg

## Table 4.16 Experimental uptake of $^{226}Ra$ by the leaves of beetroot

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | $^{226}Ra$ ($Bq.g^{-1}$ ash, soil) ($Bq.\ell^{-1}$, water) |
|---|---|---|---|---|
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 0,04 ± 0,015 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 22,5 |
| Beetroot (Roots irr.) : $\bar{X}$ ± S.D. | 777 | 77,1 | 9,9 | 0,056 ± 0,004 |
| CRs | --- | --- | --- | 0,018 ± 0,008 |
| CRw | --- | --- | --- | 0,032 ± 0,002 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 0,084 ± 0,015 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 22,5 |
| Beetroot (Leaves irr.) : $\bar{X}$ ± S.D. | 991 | 92,1 | 11,4 | 0,057 ± 0,003 |
| CRs | --- | --- | --- | 0,0078 ± 0,002 |
| CRw | --- | --- | --- | 0,029 ± 0,002 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 0,082 ± 0,018 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 22,5 |
| Beetroot (Rt + Lvs irr.) : $\bar{X}$ ± S.D. | 1063 | 95,4 | 12,2 | 0,09 ± 0,004 |
| CRs | --- | --- | --- | 0,013 ± 0,003 |
| CRw | --- | --- | --- | 0,046 ± 0,002 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | (0,022) |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | --- |
| Beetroot (Control) : $\bar{X}$ ± S.D. | 1033 | 88,4 | 8,9 | 0,012 ± 0,001 |
| CRs | --- | --- | --- | (0,0047) |
| CRw | --- | --- | --- | --- |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}Ra$ in soil expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for a dried sample.
Uranium and $^{226}Ra$ in water expressed as $\mu g.\ell^{-1}$ and $Bq.\ell^{-1}$ respectively.
Uranium and $^{226}Ra$ in vegetables expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1m$\ell$)

For $^{238}U$: 1Bq = 80,9 µg

Table 4.17 Experimental uptake of $^{226}Ra$ by the roots of cabbage

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | $^{226}Ra$ ($Bq.g^{-1}$ ash, soil) ($Bq.\ell^{-1}$, water) |
|---|---|---|---|---|
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 0,034 ± 0,015 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 22,5 |
| Cabbage (Roots irr.) : $\bar{X}$ ± S.D. | 96,8 | 15,1 | 1,5 | 0,113 ± 0,014 |
| CRs | --- | --- | --- | 0,052 ± 0,03 |
| CRw | --- | --- | --- | 0,078 ± 0,01 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 0,063 ± 0,014 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 22,5 |
| Cabbage (Leaves irr.) : $\bar{X}$ ± S.D. | 134 | 21,5 | 3,5 | 0,106 ± 0,007 |
| CRs | --- | --- | --- | 0,044 ± 0,01 |
| CRw | --- | --- | --- | 0,123 ± 0,08 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | 0,076 ± 0,014 |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | 22,5 |
| Cabbage (Rt + Lvs irr.) : $\bar{X}$ ± S.D. | 156 | 24,8 | 5,3 | 0,262 ± 0,01 |
| CRs | --- | --- | --- | 0,117 ± 0,03 |
| CRw | --- | --- | --- | 0,396 ± 0,02 |
| Soil: $\bar{X}$ ± S.D. | --- | --- | --- | (0,014) |
| Water: $\bar{X}$ ± S.D. | --- | --- | --- | --- |
| Cabbage (Control) : $\bar{X}$ ± S.D. | 94 | 13,3 | 1,1 | (0,02) |
| CRs | --- | --- | --- | (0,017) |
| CRw | --- | --- | --- | --- |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}Ra$ in soil expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for a dried sample.
Uranium and $^{226}Ra$ in water expressed as $\mu g.\ell^{-1}$ and $Bq.\ell^{-1}$ respectively.
Uranium and $^{226}Ra$ in vegetables expressed as $\mu g.g^{-1}$ and $Bq.g^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1mℓ)

For $^{238}U$: 1Bq = 80,9 µg

## Table 4.18 Experimental uptake of $^{226}$Ra by the leaves of cabbage

| Sample | Fresh mass (g) | Dry mass (g) | Ash mass (g) | $^{226}$Ra (Bq.g$^{-1}$ ash, soil) (Bq.$\ell^{-1}$, water) |
|---|---|---|---|---|
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | 0,034 ± 0,015 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 22.5 |
| Cabbage (Roots irr.) : $\bar{X} \pm$ S.D. | 909 | 90.3 | 9,0 | 0,057 ± 0,003 |
| CRs | --- | --- | --- | 0,017 ± 0,008 |
| CRw | --- | --- | --- | 0,025 ± 0,001 |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | 0,063 ± 0,014 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 22,5 |
| Cabbage (Leaves irr.) : $\bar{X} \pm$ S.D. | 976 | 96,5 | 11,3 | 0,028 ± 0,003 |
| CRs | --- | --- | --- | 0,0051 ± 0,002 |
| CRw | --- | --- | --- | 0,014 ± 0,002 |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | 0,076 ± 0,014 |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | 22,5 |
| Cabbage (Rt + Lvs irr.) : $\bar{X} \pm$ S.D. | 828 | 82,1 | 9,9 | 0,097 ± 0,012 |
| CRs | --- | --- | --- | 0,015 ± 0,005 |
| CRw | --- | --- | --- | 0,052 ± 0,006 |
| Soil: $\bar{X} \pm$ S.D. | --- | --- | --- | (0,014) |
| Water: $\bar{X} \pm$ S.D. | --- | --- | --- | --- |
| Cabbage (Control) : $\bar{X} \pm$ S.D. | 593 | 53,7 | 6,8 | 0,008 ± 0,001 |
| CRs | --- | --- | --- | (0,0066) |
| CRw | --- | --- | --- | --- |

* Concentrations below the detection limit are indicated in brackets.
Standard deviations were calculated on counting statistics and not on replicate analysis.

---: = Not Determined N.D.: = Below detection limit but no estimated value could be given.

Uranium and $^{226}$Ra in soil expressed as μg.g$^{-1}$ and Bq.g$^{-1}$ respectively for a dried sample.
Uranium and $^{226}$Ra in water expressed as μg.$\ell^{-1}$ and Bq.$\ell^{-1}$ respectively.
Uranium and $^{226}$Ra in vegetables expressed as μg.g$^{-1}$ and Bq.g$^{-1}$ respectively for an ashed sample.

CRs: Concentration Ratio of plant (fresh mass) (per gram) concentration over soil concentration (per gram)
CRw: Concentration Ratio of plant (fresh mass) (per gram) concentration over water concentration (per 1m$\ell$)

For $^{238}$U: 1Bq = 80,9 μg

In the case of beetroot and cabbage the CRs and CRw values again confirmed the superior ability of the roots over the leaves to accumulate $^{226}$Ra, being true for all three irrigation types. The difference in the concentration ratios from the soil to roots and leaves varied from a fraction to about 3 standard deviations. The differences were more apparent in the CRw to roots and leaves, due to the smaller standard deviations on the CRw's. The CRw values obtained were in all cases higher than those for CRs.

Another interesting observation is that the soil surrounding the roots of the plants in the leaves-only irrigation scheme afterwards also had an enhanced $^{226}$Ra concentration. This concentration was in the case of both beetroot and cabbage higher than the soil from the roots-only irrigation scheme, but comparable to the roots and leaves irrigation scheme.

## 4.6 Dose assessment results

The dose results are interpreted in terms of the International Commission on Radiological Protection (ICRP, 1991) recommendations on dose limits. The effective dose limit for members of the public is 1 mSv in a year but in special circumstances a higher value of effective dose could be allowed in a single year, provided that the average dose over 5 years does not exceed 1 mSv per year. The limit applies to the sum of the doses from external exposure and 50 year committed dose from ingestion and inhalation.

For a single practice resulting in a radiation dose to the public the annual dose is further restricted to 250 μSv. This is to allow for additional doses from other sources such that the combined dose does not exceed the 1000 μSv per year limit. The exposure resulting from mining activities should be subject to this lower limit. Table 4.19 gives the annual effective dose D for $^{238}$U and $^{226}$Ra for the first year of exposure and the annual committed effective dose, $D_{50}$, over a committed period of 50 years, for ingestion of drinking water, vegetables, cereals and fish, from the concentrations and volumes mentioned in Table 3.6.

### 4.6.1 Drinking water and vegetable pathway

Field observations show that the two most likely significant ingestion pathways for this study are drinking water and vegetables. In these cases the first year exposure values are all very low being individually less than 1% of the 250 μSv limit. The annual committed effective dose provides higher values, but still well within the limit. The committed dose due to ingestion of radium is much higher than for uranium because radium is a bone seeker and has a much longer biological half-life than uranium.

The doses from drinking water pathway are greater than for vegetable consumption.The dose from the drinking water is, however, exaggerated by deliberately assuming consumption values of 730 l per year, the value for standard man. In practice, surface water is not routinely used for drinking and cooking. The dose from the vegetable pathway assumes that all vegetable intake annually (30kg from leafy and 90kg from root crops) is from a contaminated source. In practice this is very unlikely. The model dose results will therefore decrease with lower expected consumption habits.

The conclusion is that the potential effective dose received from the most likely pathway of drinking water and consuming vegetables irrigated with contaminated water is well within limits set internationally.

### 4.6.2 Cereal pathway

This pathway was included because of an isolated occurrence of oats being experimentally cultivated directly on a mine dump. The concentrations of uranium and radium in the oats was the highest of any produce measured in this study and in fact ranks with the maximums encountered elsewhere in the world. For modelling purposes an annual consumption of 80 kg was assumed. The resulting effective dose for uranium is low being about 1% of the 250 µSv limit. The effective dose from radium is, however, much higher with the committed effective dose being 180 µSv.

At this particular site the area under cultivation was small, about 24 $m^2$ and took place for only 2 years and has been discontinued now. The occurrence, however, serves as a warning that any crop production on mine dump land should be controlled.

### 4.6.3 Fish pathway

In this study a limited number of fish were sampled in the field and the concentration data are for complete fish samples. However, from a previous confidential report other data for edible part of fish samples exists and for the sake of completeness a fish pathway was included in the modelling study. The assumed consumption of 25 kg fish annually is based on that of a sports fisherman. Probable species include mixed portions of the common carp, *Cyprinus carpio*, the african catfish, *Clarias gariepinus* and the smallmouthed yellowfish, *Barbus aeneus*. Once more the dose due to uranium can be neglected while the committed effective dose from radium is 16 µSv, which is considerably below the limit. The amount of fish caught and consumed is, however, small compared to the amount of vegetables produced, and therefore the significance of this pathway should be neglegible.

### 4.6.4 Miscellaneous pathways

By definition this project has excluded some pathways of public exposure such as the inhalation of radon gas and radioactive dust. Other components of aquatic pathways that are usually modelled are direct exposure due to swimming, boating and sunbathing on contaminated sediments. In this study the latter pathways were excluded since in any case they are usually a very small fraction of the different pathways. Other possible pathways are direct exposure from the contaminated irrigated lands and internal exposure from inadvertent soil ingestion. For the latter the model assumes a default value of 410 mg per day and for a soil radium concentration of 25 $Bq.kg^{-1}$ the committed effective dose is 10 µSv. For direct exposure from such soil for 2000 hours per year the annual effective dose is 2,3E-2 µSv (Table 4.19).

### 4.6.5 Summary

In summary, the summation of doses for an assumed maximum exposed individual due to ingestion of water, vegetables and soil plus direct exposure from soil will give an overall committed effective dose significantly less than 250 μSv and even more reduced for a more average individual. With the addition of the very unlikely pathway of either cereal (oats) grown on mine dump land or fish consumption from contaminated water the limit of 250 μSv/a may, however, be approached.

The collective dose for the population in general has not been estimated but for the aquatic pathway it is considered to be low. Further downstream from mines the radionuclide concentrations decrease even more and the public at large is even less likely to consume any open surface water. The relatively small mass of potentially contaminated food products will be greatly diluted in the large open market place and any possible dose will be greatly fractionated.

Note: The GENII code uses the organ weighing factors of the ICRP 26 and 30 publications. The more recent ICRP 60 of 1990 recommends changes in these factors. For radium being a bone seeker the important change is the reduction in the bone surface weighing factor from 0,03 to 0,01. This has the effect that the results reported here over estimate the committed effective dose. For uranium the kidney and stomach weighing factors changes of 0,06 to 0,025 and 0,06 to 0,12 respectively, are relevant. These compensate for each other and significant changes in effective dose are not expected.

Table 4.19 Annual Effective dose (D) and Annual Committed Effective Dose (D50) from various ingestion pathways - Units: µSv.

| Nuclide | Drinking water | | Vegetables | | | | Oats | | Fish | | Soil ingested | | Soil external | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Leafy | | Root | | | | | | | | | |
| | D | $D_{50}$ | D | $D_{50}$ | D | $D_{50}$ | D | $D_{50}$ | D | $D_{50}$ | D | $D_{50}$ | D | $D_{50}$ |
| $^{238}U$ | 1,7 | 2,3 | 0,042 | 0,057 | 0,12 | 0,17 | 2,3 | 3,1 | - | 0,02 | - | - | 1,3E-2 | - |
| $^{226}Ra$ | 0,88 | 7,8 | 0,22 | 1,95 | 0,65 | 6,0 | 20 | 180 | 1,8 | 16,0 | 1,3 | 10 | 2,3E-2 | - |

# 5 DISCUSSION AND RECOMMENDATIONS

## 5.1 Radium and uranium in effluents and surface waters

Discharge of uranium milling effluents or process waters during mining operations is the main cause of radionuclide contamination of surface waters in South Africa and elsewhere. Approximately 30% of Witwatersrand mines retain their total liquid wastes within the mine property whilst variable portions of the remaining 70% of the liquid effluents are eventually discharged into streams and thus reach the aquatic environment (de Jesus *et al.*, 1987). In the West Rand, large underground aquifers are contained in dolomitic formations. Vast amounts of such underground water are pumped by mines out of underground works, mainly for dewatering purposes. Numerous boreholes also extract water from these compartments for irrigation and domestic purposes. Of the latter, about 60% is discharged into public streams and lakes (de Jesus *et al.*, 1987).

From the point of release, the transport of $^{226}Ra$ in mining wastewater depends on the hydrodynamic conditions of the wastewater stream, the chemical and physical composition of the carrier water and the physico-chemical forms of $^{226}Ra$. The $Ra^{2+}$ ion is commonly the prevailing dissolved form of radium, except in the case of mining wastewater which mainly contains significant concentrations of free sulphates, carbonates and some organic chelating agents (Benes, 1990). Therefore, in uranium mill wastewater with high concentrations of sulphates, the radium sulphate neutral ion pair will predominate amongst the dissolved forms of radium (Benes, 1982). Radium can also be co-precipitated with several other solids present in mining wastewater. These include barium sulphate, calcium sulphate and carbonate, ferric hydroxide, etc. However, the process of co-precipitation always depends on the chemical composition and/or pH of the water as well as the concentration of the precipitate (Benes, 1990).

Uranium milling wastewater containing $^{226}Ra$ is usually discharged from the property through a wastewater channel. Experience has shown that the wastewater channel not only acts as a stream conducting wastewater to a natural recipient, but also as a site where significant changes in the concentration and chemical form of $^{226}Ra$ take place.

Little data exists on the nature and behaviour of dissolved and particulate forms of radium in rivers and lakes. The common assumption amongst researchers is that dissolved radium exists in water as hydrated cations, $Ra^{2+}$. However, this view seems to be incorrect as the presence of sulphates in most surface waters may lead to the formation of radium sulphate ion pairs (Benes *et al.*, 1982). By utilizing selective dissolution (Benes *et al.*, 1981), Benes *et al.* (1983) was able to analyze the particulate forms of radium in river water. Four different forms of particulate radium were clearly identified by this author, namely "loosely bound" (LB), "acid soluble" (AS), radium co-precipitated with barium sulphate, $Ba(Ra)SO_4$, and radium in "crystalline detritus" (CD). Results showed that particulate radium in river water contaminated with mining effluent predominantly consisted of barium-radium sulphate. It is, however, important to realise that in mining effluent with a high sulphate concentration, a significant portion of the dissolved radium occurs as the neutral ion pair $Ra^{2+}SO_4^{2-}$ (de Jesus *et al.*, 1987). The formation of a solid phase consisting only of radium sulphate is usually impossible at radium concentrations typical for uranium waste waters. Under these conditions, the solubility product of radium sulphate cannot be exceeded ($K_{so} = 10^{-10,37}$), as radium is usually co-precipitated as $Ba(Ra)SO_4$.

Several authors have noticed a marked decrease in radium concentrations along the length of a wastewater channel or flow. In a few cases, de Jesus *et al.* (1987) noticed that where radium concentrations of 4,2 $Bq.\ell^{-1}$ were measured in a wastewater channel near a mine, levels declined to about 0,04 $Bq.\ell^{-1}$ within a few kilometres from the point of release. The United States Environmental Protection Agency (USEPA) conducted a similar study of radium migration in mine drainage water in the arid New Mexico where the only water flow in an otherwise dry channel originated from uranium mine mill discharges. Dissolved $^{226}Ra$ concentration decreased from 1,1 $Bq.\ell^{-1}$ at the point of release to 0,03 $Bq.\ell^{-1}$, 9,2 kilometres downstream. This decrease in concentration is therefore due to the rapid removal of $^{226}Ra$ by strong adsorption or precipitation onto the stream sediments (USEPA, 1975 ; USEPA, 1983).

In surface streams, two basic forms of $^{226}Ra$ migration can be found, namely migration of $^{226}Ra$ in solution and migration of $^{226}Ra$ in suspended solids. These two migratory forms are closely interrelated and are connected with the bottom sediments of streams via sedimentation of suspended solids and resuspension of bottom sediments (Benes, 1990). The adsorption of dissolved radium on stream suspended solids and the dissolution or desorption of radium from stream sediments determine the ratio in which the migration forms of $^{226}Ra$ may occur in streams.

Since the average uranium concentration in South African ores (0,01 %) is at least an order of magnitude lower than that mined in the rest in the world (Funke, 1990), a similar tendency can be expected for $^{226}Ra$ concentrations in locally affected waters. Consequently, roughly the same estimates might be expected when comparing South African data on radionuclide concentrations of biota subjected to these isotopes in nature compared to those found in some overseas countries. It can also be expected that levels of uranium and $^{226}Ra$ in aquatic plants, macro-invertebrates, fish, aquatic birds and agricultural crops irrigated with such water could also be correspondingly lower.

## 5.2 Radium and uranium in algae and aquatic vegetation

Comparing results obtained for uranium and $^{226}Ra$ concentrations in aquatic plants collected in mine polluted rivers on the East Rand of the Transvaal, overall lower concentrations for both radionuclides were recorded than concentrations measured for related species in other countries. For example, *Spirogyra* sp. collected at Luipaardsvlei contained 1,27 $Bq.g^{-1}$ $^{226}Ra$ and 52,6 $\mu g.g^{-1}$ uranium whilst a mixture of the green filamentous algae, *Cladophora glomerata*, *Oedogonium capillare* and *Rhizoclonium* sp. from the uranium mine polluted Nedvedieka and Mze rivers in Moravia and Bohemia (Czechoslovakia) yielded values of 3,73 $Bq.g^{-1}$ for $^{226}Ra$ and 600 $\mu g.g^{-1}$, uranium, respectively (Justyn & Stanek, 1974). In both these cases, values were more than double those measured in *Spirogyra* sp. from the mine polluted rivers in South Africa. However, Tsivoglou *et al.* (1960), monitoring the Animas river in the vicinity of uranium mills at Colorado, recorded a concentration of 0,148 $Bq.g^{-1}$ for $^{226}Ra$ in algae from unpolluted sections of the river. However, mine polluted sections of the same river yielded algae with a $^{226}$Ra concentration of 129,5 Bq.g$^{-1}$. *Spirogyra* sp. from uranium mine polluted waters in Jaduguda, India (Markose *et al.*, 1978), also showed relatively high $^{226}$Ra concentrations, ranging between 2,51 and 89,3 Bq.g$^{-1}$, which in all cases (6 sampling localities), were generally higher than those recorded for this genus in South Africa.

In the case of the aquatic macrophytes, both the concentrations of uranium and $^{226}$Ra in *Potamogeton pectinatus* from Luipaardsvlei, Elsburgspruit and Natalspruit were lower than those recorded for this species at Langley Bay, Canada, which is affected by uranium mine mill tailings (Waite *et al.*, 1988). While the concentrations of uranium and $^{226}$Ra in this aquatic weed at the Langley Bay site was 5130 $\mu$g.g$^{-1}$ and 56,5 Bq.g$^{-1}$ respectively, values were in both cases higher than those recorded for a nearby control locality (U: 70 $\mu$g.g$^{-1}$ ; $^{226}$Ra: 0,37 Bq.g$^{-1}$). It is however interesting to note that the uranium and $^{226}$Ra concentrations obtained for *P. pectinatus* from the mine polluted Elsburgspruit and Natalspruit rivers (Table 4.5a) were even lower than those obtained for the Langley Bay control values.

Although $^{226}$Ra and uranium concentrations of *Spirogyra* sp. collected in mine polluted rivers in South Africa were found to be relatively low, a number of factors should be taken in consideration when determining the actual concentrations bio-accumulated within the cells. Results from algal analysis may on occasions seem to be high, but do not necessarily reflect the actual amount of metal or element bioaccumulated. In most cases, an unknown portion of these elements may be bound to the outer surface of the cell wall of the plant. The cell wall, consisting of a porous matrix of gelatinous polysaccharide fibres surrounding the cell membrane is the first barrier between an aquatic plant and the radium entering it from the surrounding water column (Alberts *et al.*, 1983). The kinetics of accumulation of several cationic, anionic and neutral metal ions forming complexes with freshwater algal cell walls have been demonstrated by Story *et al.* (1983). Their results showed that separate sites on the cell walls were selective to the three ionic groups. Of the three groups, the metal cations were the most strongly accumulated while radium was the best accumulated amongst the alkaline and alkaline earth elements (Story *et al.*, 1984). By chemically removing loosely bound $^{226}$Ra from the cell walls of a range of algae, Havlik (1971) and Havlik & Robertson (1971) could demonstrate that algal cell walls play a major role in $^{226}$Ra capture which could therefore not be considered as bioaccumulation.

Another factor which also may have an important influence on the radionuclide bioaccumulation ability of aquatic weeds is the extensive development of epiphytes or aufwuchs (algae and bacteria) attached to their submerged roots and shoots. The presence of these epiphytes may strongly influence the "ability" or capacity of these plants to accumulate elements from the water column (Emerson & Hesslein, 1973). It is therefore important that during the collection and preparation of submerged algae and aquatic weeds, a standardized approach be followed to remove the epiphytes and ensure a more correct measurement of concentration ratios of $^{226}$Ra and uranium.

Uranium and $^{226}$Ra concentrations from the stems (U: 5 $\mu$g.g$^{-1}$ ; $^{226}$Ra: 1,3 Bq.g$^{-1}$) and roots (U: 85 $\mu g.g^{-1}$ ; $^{226}Ra$: 18,7 $Bq.g^{-1}$) of *Typha* sp. collected at Langley Bay, Canada, were generally higher than concentrations from whole composite (roots + stems) samples for the same species found in the Elsburgspruit, Germiston lake and the Natalspruit. The only exceptionally local high uranium value for *T. capensis* was recorded in the Blesbokspruit where a concentration of 111 $\mu g.g^{-1}$ was recorded.

No data exists in the literature on the radionuclides uranium and $^{226}Ra$ accumulated by aquatic macrophytes such as *Lemna* sp., *Azolla filiculoides*, *Marsilea macrocarpa*, *Lagarosiphon muscoides* and *Arundo donax*. In the present study, all values found for these plants seemed to fall within the same order of magnitude for uranium and $^{226}Ra$, respectively. In the case of the floating aquatic weeds, uranium (47,9 $\mu g.g^{-1}$) and $^{226}Ra$ (<99 $Bq.kg^{-1}$) recorded in *Azolla filiculoides* (Blesbokspruit) showed slight differences with those of *Lemna major* from the Natalspruit (U: 31,7 $\mu g.g^{-1}$ ; $^{226}Ra$: <109 $Bq.kg^{-1}$) (Table 4.5a). The rooted submerged aquatic fern, *Marsilea macrocarpa*, showed values ranging between 37,9 (Blesbokspruit) and 58,4 (Elsburgspruit) $\mu g.g^{-1}$ for uranium while $^{226}Ra$ fluctuated between 82 (Elsburgspruit) and 92 (Blesbokspruit) $Bq.kg^{-1}$. The other rooted submerged aquatic weed analysed, *Lagarosiphon muscoides*, showed a somewhat different tendency. While the uranium (26,4 $\mu g.g^{-1}$) concentration seemed to be within range of the other aquatic weeds, this plant possessed a marked ability to accumulate $^{226}Ra$ (305 $Bq.kg^{-1}$).

There is a widespread occurrence of *Arundo donax* in lakes and wetlands of South Africa. It was the only plant collected at all the sampling localities. A lowest uranium concentration for this plant was measured at Natalspruit (14,2 $\mu g.g^{-1}$) while the higest concentration occurred at the Blesbokspruit (37,7 $\mu g.g^{-1}$). Other than uranium, a highest $^{226}Ra$ value was recorded at the Germiston lake (195 $Bq.kg^{-1}$) with a lowest of 71 $Bq.kg^{-1}$ at the Elsburgspruit.

The main difference between submerged algae and aquatic macrophytes and rooted emergent aquatic macrophytes is that the latter partially obtain nutrients from the water column as well as from aquatic sediments. Sediments are generally more anoxic as they are saturated with water most of the time. Usually the levels in sediments are below that in the water which would allow some oxygenation to occur. Under anoxic conditions the major sediment chemical processes tend to go into a reduced state (Ponnamperuma, 1972). Under such conditions, radium remains a divalent cation, but its availability could indirectly be influenced by other sediment constituents such as iron, manganese and sulphates which also go into a reduced state (Cooper *et al.*, 1981 ; Hesslein & Slavicek, 1984).

Radium coprecipitated with calcium or barium, $Ba(Ra)SO_4$, present in South African stream sediments, may be released under reducing conditions because the sulphides of calcium and barium are much more soluble than their sulphates (Linke, 1958). Under these conditions, and especially those which are characteristic for local rivers and wetlands, increased uptake of $^{226}Ra$ by aquatic weeds is therefore highly possible as has been demonstrated by data obtained from the Blesbokspruit, Elsburgspruit, Germiston lake and Natalspruit rivers and their wetlands.

The degree of mobility, activity and bioavailability of elements in soil can be influenced by a number of factors, namely soil pH, temperature, redox potential, cation exchange capacity of the solid phase, competition with other metal ions, ligation by anions and composition and quantity of the soil solution (Trangmar *et al.*, 1985 ; Wopereis *et al.*, 1988). In a normal soil system under equilibrium, the solid and the liquid phases of the soil are in close contact. The solid phase is mostly composed of clays, metal oxides and organic matter and carries in most cases an excess of negative charge. Water, which is a polar solvent interacts with the surface structure of the solid (soil) and is therefore capable of enveloping charged sites on the soil particles (Schmitt & Sticher, 1991) which has a number of important consequences: the pH of the water affects the charge pattern of the solid surface, and water acts as a transport medium for ions in exchange and adsorption processes (Schmitt & Sticher, 1991).

### 5.3 Radium and uranium in aquatic fauna

No information is available on the presence and concentration of uranium and $^{226}$Ra in freshwater crabs elsewhere in the world. In the case of the freshwater crab, *Potamonautes warreni* which was the only macro-invertebrate organism analyzed in the present study, uranium concentrations were more than double those obtained for fish whilst $^{226}$Ra concentrations were slightly lower but of a similar order to those found for fish and aquatic birds.

Fish are considered to be the principal link in the food chain by which water-borne contamination can reach man. In this way, it is also believed to affect the extent of radiological exposure of a population (Justyn & Havlik, 1990). In uncontaminated water, the $^{226}$Ra content of fish seem to be very low (thousandths to hundredths of Bq.g$^{-1}$ dry weight) while in surface waters contaminated by uranium milling effluent, the $^{226}$Ra content can be from 3 to 6 orders of magnitude higher in fish muscle (Paul *et al.*, 1980 ; Anderson *et al.*, 1963 ; Martin *et al.*, 1969 ; de Bortoli & Gaglione, 1972 ; Martinec, 1973 ; Davy & Conway, 1974 ; Davy & O'Brein, 1975 ; Ettenhuber *et al.*, 1975).

Since there are not many similarities between the fish fauna of the different countries, data obtained for South African fish species were compared with each other. It seems that physiological differences between species do not have as great an influence on the bioaccumulation of radionuclides as regional or geographical differences might have. In all cases, concentrations of $^{226}$Ra for all fish species collected during the present survey were lower than 0,1 Bq.g$^{-1}$ with values ranging between 0,036 Bq.g$^{-1}$ (*C. gariepinus*) and 0,081 Bq.g$^{-1}$ (*T. sparrmanii*). A comparison of these results with those obtained for other freshwater fish collected in Lake Athabasca, Canada (Swanson, 1985), showed that all values for the local fish species analyzed were between one and two orders of magnitude lower than those for Canada. Concentration values of $^{226}$Ra in fish collected in Lake Athabasca ranged between 0,15 Bq.g$^{-1}$ (Lake Chub: *Covesius plumbeus*) and 4,0 Bq.g$^{-1}$ (Spottail shiner: *Notropius hudsonius*). A similar tendency was observed for uranium uptake by fish in South Africa with values ranging between 0,60 $\mu$g.g$^{-1}$ (*C. gariepinus* : Natalspruit) and 4,99 $\mu$g.g$^{-1}$ (*C. gariepinus* : Blesbokspruit). The uranium content of the Lake Athabasca fish were again one order of magnitude higher, fluctuating between 25,5 $\mu$g.g$^{-1}$ (*Covesius plumbeus*) and 78,3 $\mu$g.g$^{-1}$ (Small white sucker: *Catostomus commersoni*) (Swanson, 1985). Czechoslovak researcers who investigated the $^{226}$Ra-concentrations in fish from reservoirs in the vicinity of uranium milling operations in Northern Bohemia also obtained values for radium which were at least one order of magnitude higher than those for fish from different mine polluted streams in South Africa. Justyn & Lusk (1976) recorded $^{226}$Ra in fish varying between 0,185 Bq.g$^{-1}$ (*Esox lucius*) and 1,41 Bq.g$^{-1}$ (*Perca fluviatilis*). Other fish analyzed by these authors yielded more intermediate results with $^{226}$Ra values for *Anguilla anguilla* (0,28 - 0,5 Bq.g$^{-1}$), *Gobio gobio* (0,3 - 0,425 Bq.g$^{-1}$) and *Leuciscus cephalus* (0,225 - 0,55 Bq.g$^{-1}$) falling within this range. Justyn *et al.* (1979) obtained nearly similar results for *Perca fluviatilis* (0,18 - 1,41 Bq.g$^{-1}$) and *Salmo trutta* (0,016 - 0,045 Bq.g$^{-1}$) while values of 1,75 Bq.g$^{-1}$ and 1,25 Bq.g$^{-1}$ was obtained for *Cyprinus carpio* and *Perca fluviatilis* respectively, by Justyn *et al.* (1985).

It is important to evaluate the $^{226}$Ra content in fish in relation to the species of radium in the water as well as the presence and species of other chemical substances in the water, namely calcium and barium. The pH, alkalinity and acidity of the water is also important as it influences the mobility, toxicity and bioavailability of radium to fish. Two pathways normally exist whereby fish and other aquatic animals can assimilate and bioconcentrate radium or non-nutritional elements, namely: direct uptake from the water through the skin or gills during water filtration ; uptake from food (Justyn & Havlik, 1990). However, uptake and bioconcentration of radium and other elements by fish are mainly dependent on a number of conditions: it has been found that differences exist between fish species and the extent of the concentration and uptake of radium from contaminated water. However, these differences can largely be related to the food and feeding habits. A low radium content and therefore a low concentration ratio was found in *Coregonus lavaretus maraena* Bloch, which is known to live in the water column and to feed mainly on zooplankton organisms. Other fish such as *Gobio gobio* L., *Leuciscus cephalus* L., *Thymallus thymallus* L. and *Anguilla anguilla* L. also showed relatively low radium contents. In the benthophagous and phytophagous *Tinca tinca* L., *Cyprinus carpio* L., *Rutilus rutilus* L. and *Carassius carassius*, a relatively high uptake of radium was observed. In predatory fish, the CR was higher in *Salmo trutta* m. fario L. than in *Esox lucius* L. (Martinec, 1973 ; Justyn & Lusk, 1976 ; Justyn *et al.*, 1979 ; Justyn *et al.*, 1985). These differences can, according to Justyn & Havlik (1990) primarily be ascribed to lifestyle, food preference (zoophagous, phytophagous, benthophagous or predatory), rate of metabolism, growth and sexual maturity. In field tests with *Cyprinus carpio* stocks, Justyn *et al.* (1979) found that radium uptake in younger fish was almost double that in older fish. With a rise in water temperature a more rapid increase in $^{226}$Ra volume activity was observed. Justyn *et al.* (1985) also noted that higher $^{226}$Ra concentrations were observed in fish in the summer months than in winter. According to these authors, $^{226}$Ra concentrations in fish can be related to the intensity of food intake during the active feeding summer period. Therefore, interpretation of results obtained for fish in this study should take the above factors into account.

Although the previously mentioned organism groups are extensively covered in the literature, no data could, however, be found for uranium and $^{226}$Ra concentration in aquatic birds. However, data collected from the Natalspruit on the coot *Fulica cristata* and the cormorant *Phalacrocorax carbo* seems to be a first definitive contribution to the investigation of the concentrations levels of these radionuclides in these species of birds. From the results for uranium it is interesting to note that the phytophagous *F. cristata* contains more uranium (0,56 $\mu g.g^{-1}$) than the fish eating *P. carbo* (<0,17 $\mu g.g^{-1}$). This tendency can most likely be ascribed to the feeding habits of these two bird species as well as the uranium content of the food. However, this tendency is in the case of $^{226}Ra$ not so definite with *F. cristata* containing <0,034 $Bq.g^{-1}$ and *P. carbo* <0,074 $Bq.g^{-1}$.

## 5.4 Radium and uranium in vegetables

As no definite standards are available for uranium and $^{226}Ra$ content for vegetable crops, results obtained from the Marievale, Vlakfontein and Luipaardsvlei farms were compared with research findings in the literature which mainly originate from Germany, Japan, India and USA. Uranium concentrations in carrots collected at Marievale (0,23 $\mu g.g^{-1}$) and at Vlakfontein (1,7 $\mu g.g^{-1}$) were in both cases higher than those recorded for the same crop (0,033 $\mu g.g^{-1}$) by Morishima *et al.* (1977) in Japan (Table 5.1). The same applied to beetroot (Marievale: 0,2 - 0,36 $\mu g.g^{-1}$ ; Vlakfontein: 1,7 $\mu g.g^{-1}$) which in both cases yielded higher concentrations than uranium found in root vegetables (0,026 - 0,033 $\mu g.g^{-1}$) by Welford & Baird (1967). However, in the case of cabbage from the Marievale farm (0,02 - 0,027 $\mu g.g^{-1}$) values were clearly less than those recorded by Morishima *et al.* (1977), being 0,067 $\mu g.g^{-1}$ (Table 5.1).

In the case of $^{226}Ra$, the concentration obtained for carrots collected at Vlakfontein (0,0041 $Bq.g^{-1}$) was intermediate between those recorded by Tracy *et al.* (1983) (0,0035 $Bq.g^{-1}$) and Muth *et al.* (1960) (0,0055 $Bq.g^{-1}$).

Beetroot from the Vlakfontein farm (0,003 $Bq.g^{-1}$), were of the same order of magnitude as those recorded by Tracy *et al.* (1983) (0,0067 $Bq.g^{-1}$). Results from the lettuce crop from Vlakfontein (0,0017 $Bq.g^{-1}$) appeared to be much lower in radium than the 0,0095 $Bq.g^{-1}$ found for lettuce by Straub *et al.* (1961) in th USA (Table 5.1).

Unfortunately the $^{226}Ra$ concentrations in the vegetables from the Marievale farm were below the detection limit of the analytical method used at the time. During later stages of the study a more elaborate method was used, which yielded the results for the Luipaardsvlei and Vlakfontein localities, as well as the experimental study.

## 5.5 Concentration Ratios

CRw values for the vegetable crops and fodder from the Marievale, Vlakfontein and Luipaardsvlei farms and the irrigation water were consistently higher than those of the corresponding CRs values. This is, however, not an indication that more $^{226}Ra$ or uranium is taken up from the water, but just that the concentrations in the water, expressed as activity/mℓ for the calculation of the concentration ratio's, is lower than the activity/g of soil. It is unlikely that the concentrations in the plants were in any of the cases in equilibrium, with the concentrations in the irrigation water which is a

Table 5.1 Concentrations of uranium and $^{226}$Ra in vegetable crops from different countries of the world including South Africa

| Crop | Uranium (µg.g$^{-1}$) (ash) | Reference (U) | $^{226}$Ra (Bq.g$^{-1}$) (ash) | Reference (Ra) |
|---|---|---|---|---|
| Carrots | 0,033 | Morishima *et al.* (1977)<br>Japan | 0,0035 | Tracy *et al.* (1983)<br>USA |
| | | | 0,0055 | Muth *et al.* (1960)<br>Germany |
| | 0,23-0,70 | This study | 0,0041-0,010 | This study |
| Beetroot | --- | --- | 0,0067 | Tracy *et al.* (1983)<br>USA |
| | 0,20-1,70 | This study | 0,003-0,010 | This study |
| Root vegetables | 0,026-0,033 | Welford & Baird (1967)<br>USA | 0,0026 | Morse & Welford (1971)<br>USA |
| | | | 0,0018 | Hallden & Fisenne (1961)<br>USA |
| Cabbage | 0,067 | Morishima *et al.* (1977)<br>Japan | 0,0032 | Muth *et al.* (1960)<br>Germany |
| | 0,020 | This study | 0,010 | This study |
| Lettuce | --- | --- | 0,0095 | Straub *et al.* (1961)<br>USA |
| | 0,080-0,170 | This study | 0,0011-0,0025 | This study |
| Vegetable mix | 0,078-0,22 | Welford & Baird (1967)<br>USA | 0,0024 | Klement (1982)<br>USA |
| Milk | 0,026-0,153 | Lal *et al.* (1982)<br>India | 0,00046 | Morse & Welford (1971)<br>New York average |
| | 0,21-0,22 | This study | 0,0081-0,0500 | This study |

prerequisite to calculate the concentration ratio. The same apparent observation was made in the experimental part of this study.

The CRw-values were generally higher than the CRs-values. This was true for cabbage and beetroot for all the different irrigation schemes used. Again this might just be that the soil is the dominating medium for transfer of $^{226}$Ra to the vegetables. Cawse and Turner (1982) state that higher levels of radionuclide activity in plants can usually be associated with plants with hairy leaf surfaces, hairy convoluted fruiting inflorescences as well as plants with broad "open" leaves such as spinach. Romney *et al.* (1976) is of the opinion that much of the radionuclide contamination to plants may be ascribed to direct deposition of resuspended soil particles (possibly from wind blown sand particles from nearby slimes dams in the vicinity of all of these farms). Corey *et al.* (1977) also ascribed the major Pu uptake by food crops to foliar deposition of this isotope and that only 10% may be due to root uptake. This assumption is also confirmed by Adriano *et al.* (1980).

The $^{226}$Ra concentrations [Q] in the vegetables are probably described by the relationship:

[Q] = CRs[Cs] + CRw[Cw]

If [Cs] and [Cw] are respectively the $^{226}$Ra concentrations in the soil and in the irrigation water, and [Cw] is less than [Cs], then this relationship indicates that CRw has to be bigger than CRs for the irrigation water to contribute to the $^{226}$Ra concentration in the plant.

In the experimental part the $^{226}$Ra concentration in the irrigation water [Cw] was kept constant at 0,0225 Bq.m$\ell^{-1}$. If one assumes that CRw stays constant, then the equation becomes:

[Q] = CRs[Cs] + k : **Where:** k = CRw[Cw] and is constant.

If one then plots [Q] against [Cs], then the slope is the concentration ratio from soil to the vegetable plant, and the intercept, k, divided by the concentration ratio of the $^{226}$Ra in the irrigation water is the concentration ratio from water to the vegetable plant. If the $^{226}$Ra in the irrigation water is not in equilibrium with the plant, it means that the ratio is valid only under similar circumstances (eg. irrigation method). This was done, and the best correlation that could be found was that between soil and beetroot $^{226}$Ra concentration, and even in this "best" case the correlation coefficient was only 0,57. The reasons for this low coefficient is probably the lack of sufficient data points (6 in this case) and that the $^{226}$Ra concentration in the independent variable (soil $^{226}$Ra concentration) did not cover a wide range of values. The soil $^{226}$Ra concentrations varied only between 0,04 and 0,08 Bq.g$^{-1}$ which is in the range of normal soil concentrations of 0,004 to 0,125 Bq.g$^{-1}$ given by Iyengar (1990).

If one nevertheless accepts that these calculations do give a rough indication of the concentration ratios, the slope is 1 ± 0,7 and the intercept is 0,02 ± 0,05. The concentration ratio from soil to beetroot is therefore of the order of 1 and for the concentration ratio of water to beetroot the order of magnitude is also about 1 (0,02 / 0,0225). The concentration of $^{226}$Ra in water has therefore to be of the same order as the concentration in the soil to contribute equally to the transfer of $^{226}$Ra to the plant. This again is only valid according to the irrigation method used. If one has, for example, a soil $^{226}$Ra concentration of 0,03 Bq.g$^{-1}$, then an irrigation water concentration of 0,03 Bq.m$\ell^{-1}$ will contribute equally to the $^{226}$Ra in the plant. Concentrations of 0,03 Bq.m$\ell^{-1}$ or 30 Bq.$\ell^{-1}$ was never found in this study. The highest concentration used for irrigation by a farmer was of the order of 0,2 Bq.$\ell^{-1}$, which is a hundred times lower than concentrations normally found for $^{226}$Ra in soil for this study. Even if there is a factor 10 error in the calculation of the CRs and CRw, and CRs is really 10 times lower and CRw is really 10 times higher than calculated, the uptake of $^{226}$Ra from the irrigation water will still only be of the same order as the transfer from soil.

One of the most important factors which may have had an influence on the CR values in the present study, is whether the plant and soil are in equilibrium. If in equilibrium, the CR will remain predictable (Whicker & Schultz, 1982). Another important factor is the choice of location and the soil sampling method. This especially applies to areas where soil contamination is non-uniformly distributed. In the case of spray and flood irrigated agricultural lands, the distribution of contaminated water should be relatively homogenous which most likely ensures a relatively uniform distribution of elements. During the collection of soil samples, particular attention was given to the randomness of the sampling programme. All subsamples were pooled, thoroughly mixed and homogenized to ensure an even better distribution of elements.

Even though the concentration ratio of $^{226}$Ra from irrigation water to vegetables might be bigger than the same ratio from soil to vegetables, the biggest contribution to the $^{226}$Ra concentration in vegetables will probably be from the soil. This is due to the fact that the $^{226}$Ra concentration in the soil is usually a few orders higher than in the water used by farmers in the vicinity of the mines. For the same reason the buildup of $^{226}$Ra in soils irrigated with mine water will be insignificant.

The concept of CR has a serious weakness, namely the lack of an easily interpreted physical meaning which makes the direction and degree of change if circumstances vary, difficult to predict. These circumstances may include soil type, physical and chemical characteristics of soil (pH, organic content etc.), physical and chemical characteristics of applied irrigation water, plant species and climate (Van Dorp *et al.*, 1979). Therefore, if the CR value is specified with respect to one or more of the variables which may have an influence on it, a huge amount of data will need to be stored which will make a database for radiological assessment very extensive.

It is suggested that although the concept of CR is a useful tool in predicting food chain transport, it needs further evaluation, modification and refinement. It is recommended that a CR is calculated for each main crop type or if sufficient data exists, together with each associated main soil type (Ng *et al.*, 1990). An alternative approach for predicting concentrations of radionuclides in plants via uptake was suggested by Van Dorp *et al.* (1979). This proposed approach utilizes a selectivity coefficient, a transpiration coefficient as well as the annual production of dry matter. It seems, however, that the actual equation for the calculation of a CR which takes all environmental factors in consideration, will be open for discussion long into the foreseeable future. However, Simon & Ibrahim (1990) stresses that at a minimum, the CR should be treated as a function of substrate concentration rather than a constant value.

## 5.6 Dose assessment

The dose assessment results for reasonably realistic yet conservative assumptions for water and vegetable consumption show that doses to a maximum exposed group will be of the order of 1% of the annual limit of 250 µSv. Radium as expected has a higher contribution than uranium. If the drinking water radium concentration were a factor 5 higher, i.e. equal to current EPA(USA) limit of ≈ 0,2 $Bq.\ell^{-1}$ then the annual committed effective dose would be 39 µSv, still well below the dose limit. Only infrequently and then very close to mine properties have such concentrations been observed locally. Similarly a factor 5 or 20 increase in vegetable concentrations will still leave a good margin below the limit. Obviously production of crops on mine dumps should be prohibited or strictly controlled as evidenced by the individual results from the oats production.

There is no obvious data to confirm the model's default value of 410 mg per day for soil ingestion. The associated committed effective dose value for radium of 10 μSv is the same order as the dose from drinking water and vegetable consumption. The control value for radium contamination of soil set by the Council for Nuclear Safety (CNS) is 200 $Bq.kg^{-1}$ or a factor eight greater than the concentration used in the model. Such a limit would result in an ingestion dose of 40 μSv.

The data collected in this study in the Witwatersrand area shows that no member of the public is expected to be exposed via aquatic pathways to any significant dose. If mine effluents are controlled to less than 0,2 $Bq.\ell^{-1}$, radium and less than 0,5 $Bq.\ell^{-1}$ uranium the doses received from various aquatic pathways should be well within dose limits set. The concentration values quoted here are not intended to imply these should be concentration limits. Control limits are set by CNS based on site specific assessments.

### 5.7 Comments and recommendations

A few comments relating to this study are highlighted below:

- Due to analytical uncertainty, low level measurements suffer from large errors and as demonstrated by some samples obtained from the Marievale farm can be due to the analytical technique used below the detection limit. More sensitive techniques are also more costly.

- It is often found with older literature that difficulties arise when different units of measurement and different terminologies are encountered. Considerable time is usually spent in converting such data to a more recent and acceptable format. Mass units (g,kg), mass basis (fresh, dry and ash) and the conversion factors are all very important components in report format. In this report, it was decided to use basic SI units of measurement. Therefore, in the case of $^{226}Ra$ content in solid matter (ash), concentrations were expressed in $Bq.g^{-1}$ while concentrations in liquid samples (water) were expressed in $Bq.\ell^{-1}$. This approach in measurment could therefore explain the differences often encountered in the CRs and CRw values. The difference in CRs and CRw values would therefore have been smaller if concentrations were expressed in $Bq.kg^{-1}$, or when $Bq.g^{-1}$ and $Bq.m\ell^{-1}$ was used. For the sake of uniformity, a few sacrifices had to be made and the use of basic SI units of measurement proved to be the best.

- Data are always expressed as $Bq.g^{-1}$ ash mass, but wet, dry and the ash mass of each sample were also supplied. In the literature, conversion factors are often supplied, but in this report these figures were provided to facilitate calculations and make it more accurate.

- To further improve reporting, environmental conditions such as water quality (physical and chemical) as well as soil characteristics were included. Often, these data proved to be helpful in explaining certain phenomena. Also included were the sources of radium contamination which gave more information on its possible species, the analytical procedure used and the estimated lower detection limit.

- The term CR (Concentration Ratio) was also accepted as a univeresal term as suggested by Simon & Ibrahim (1990). The term CR gives clarification on its meaning, i.e. implies a ratio, while terms such as bioconcentration factor (BF) etc. could sometimes be cumbersome and misleading and does not specifically refer to a ratio.

Certain areas which justify additional research are recommended as follows:

- The differences in $^{226}$Ra and uranium uptake between different plants species.
- The kinetics of the movement of $^{226}$Ra across root membranes and within plants.
- The degree of foliar absorption of $^{226}$Ra by plants. The influence of wax and hair layers on leaf surfaces on $^{226}$Ra absorption.
- The influence of calcium and other alkaline earth elements on radium uptake by plants, fish, etc.
- Determination of the species of radium in soil, water and plants.
- The effect of organic matter, soil pH etc. on the mobility, activity and bioavailability of $^{226}$Ra.
- Preparation and cleaning methods of algae and aquatic macrophytes prior to analysis.
- Additional consideration of the concept that the CR may be treated as a function of substrate concentration rather than a constant should be followed.

Steps are taken to bring the results obtained in the present investigation to the attention of the relevant mining, municipal and governmental authorities. A total of four publications covering the various aspects of the present investigation are in the process of preparation.

## 6 REFERENCES

ADRIANO, D.C., COREY, J.C. & DAHLMAN, R.C. 1980. Plutonium contents of field crops in the South Eastern United States. In: **Transuranic Elements in the Environment.** (HANSON, W.C. Ed.) NTIS. Springfield. Virginia. pp. 381 -

AINSLIE, L.C. 1993. Atomic Energy Corporation of South Africa. Personal communication

ALBERTS, B., *et al.*, 1983. **Molecular Biology of the Cell.** Garland Publishing. New York.

ANDERSON, J.B., TSIVOGLOU, E.C., SHEARER, S.D. 1963. Effects of uranium mill wastes on biological fauna of the Animas River (Colorado-New Mexico). **Radioecology.** (Proc. 1st Natl. Symp. Fort Collins, CO, 1961) (SCHULTZ, V., KLEMENT, A.W. Eds.). Rheinhold an American Institute of Biological Sciences, Washington, DC 373 - 383.

ANTROBUS, E.S.A. (Ed.). 1986. **Witwatersrand Gold - 100 Years.** Geological Society of South Africa. Kelvin House. Johannesburg.

BENES, P. 1982. Physico-chemial forms and migration in continental waters of radium from uranium mining and milling. In: **Environmental Migration of Long-Lived Radionuclides.** (Proc. Int. Symp. Knoxville. TN.) IAEA. Vienna. pp. 3 - 23.

BENES, P. 1990. Radium in (continental) surface water. In: **The Environmental Behaviour of Radium.** Technical Reports Series No. 310. Vol. 1. pp. 373 - 418. IAEA (INternational Atomic Energy Agency). Vienna.

BENES, P., SEDLACEK, J., SEBESTA, F., SANDRIK, R. & JOHN, J. 1981. Method of selective dissolution for characterization of particulate forms of radium and barium in natural and waste waters. **Water Res.,** 15(12): 1299 - 1304.

BENES, P., OBDRZALEK, M. & CEJCHANOVA, M. 1982. The physicochemical forms of traces of radium in aqueous solutions containing chlorides, sulfates and carbonates. **Radiochem. Radioanal. Lett.,** 50(4): 227 - 242.

BENES, P., SEBESTA, F., SEDLACEK, J., OBDRZALEK, M. & SANDRIK, R. 1983. Particulate forms of radium and barium in uranium mine waste waters and receiving river waters. **Water Res.,** 17: 619 - 624.

BERNARD, S.R. & STRUXNESS, E.G. 1957. **A Study of the Distribution and Excretion of Uranium in Man.** ORNL-2304. Oak Ridge National Laboratory. Oak Ridge. Tennessee.

BIDDULPH, O. 1960. Radioisotopes in plants: Foliar entry and distribution. In: **Radioisotopes in the Biosphere.** (CALDECOTT, R.S. & SNYDER, L.A. Eds.). pp. 73 - 85. University of Minnesota Press. Minneapolis. MN.

BLIGHT, C.E. & CLADWELL, J.A. 1984. The abatement of pollution from abandoned gold-residue dams. **J. S. Afr. Inst. Min. Metall.**, 84(1): 1-9.

BRAY, R.H. 1954. A nutrient mobility concept of soil-plant relationships. **Soil Sci.**, 78: 9 - 22.

BURKART, W. 1991. Uranium, Thorium and Decay Products. In: **Metals and Their Compounds in the Environment: Occurrence, Analysis and Biological Relevance.** (MERIAN, E. Ed.). pp.1275 -1287. VCH Verlagsgesellschaft. Weinheim.

CARTWRIGHT, F.D. 1983. Some seventy years in the shrouding of mining residue deposits for environmental protection. I.A.W.P.R. Conf., **Wat. Sci. Tech.**, 15(2): 85-102. "Mine Water Pollution", (P.E. ODENDAAL Ed.).

CATALDO, D.A. & WILDUNG, R.E. 1978. Soil and plant factors influencing the accumulation of heavy metals by plants. **Environ. Health Perspect.**, 27: 149 - 159.

CAWSE, P.A. & TURNER, G.S. 1982. **The Uptake of Radionuclides by Plants: A Review of Recent Literature.** Rep. AERE-R-9887. UKAEA Atomic Energy Research Establishment. Harwell.

CHAMBER OF MINES OF SOUTH AFRICA. 1979. The design, operation and closure of residue deposits. **Handbook of Guidelines for Environmental Protection.** 1.

COETZEE, N.J.C. 1991. Personal communication

COGGLE, J.E. 1983. **Biological Effects of Radiation.** Taylor & Francis Inc. New York.

CONWAY, N.F., DAVY, D.R., GILES, M.S., NEWTON, P.J.F. & POLLARD, D.A. 1974. **The Alligator rivers area fact finding study.** Four AAEC Reports. Australian Atomic Energy Commission. Research Establishment Lucas Heights.

COOK, W. 1973. Green grow the mine dumps. **Mining Survey**, 73: 21-23.

COOK, C.D.K., GUT, B.J., RIX, E.M., SCHNELLER, J. & SEITZ, M. 1974. **Water plants of the world: A manual for the identification of the genera of freshwater macrophytes.** Dr. W. Junk b.v, Publishers. The Hague.

COOPER, M.B., STANNEY, K.A. & WILLIAMS, G.A. 1981. **An Investigation of the Speciation of Radionuclides in Sediments and Soils.** Rep. ARL/TR-039. Australian Radiation Laboratory. Melbourne.

COREY, J.C., BONI, A.L., ADRIANO, D.C., PINDER, J.F. & McLEOD, K.W. 1977. Summary of plutonium terrestrial research studies in the vicinity of a nuclear fuel reprocessing plant. In: **Savannah River Laboratory Annual Report.** Dp-1489, pt.2. Aiken. S.Carolina. pp.. 25 - 28.

COUGHTREY, P.J. & THORNE, M.C. 1983. **Radionuclide Distribution and Transport in Terrestrial and Aquatic Ecosystems: A Critical Review of Data.** A.A. Balkema. Rotterdam.

CURIE, M., CURIE, P. & BEMONT, G. 1898. Another new radio-active element. **C.R.,** 127: 1215 - 1217.

DAVY, D.R., CONWAY, N.F. 1974. Environmental studies, Northern Territory Uranium Province 1971-73. **The Alligator Rivers Area Fact Finding Study.** Four Reports (CONWAY, N.F., DAVY, D.R., GILES, P., NEWTON, P.J.F, POLLARD, D.A. Eds). Rep. AAEC/E-305, Australian Atomic Energy Commission, Lucas Heights Research Laboratories, III>

DAVY, D.R., O'BRIEN, B.G. 1975. Radiological Aspects. In: **Rum Jungle Environmental Studies** (DAVY, D.R., Ed.). Rep. AAEC/E-365, Australian Atomic Energy Commission, Lucas Heights Research Laboratories, Chapter 9.

DE BORTOLI, M., GAGLIONE, P. 1972. Radium-226 in environmental materials and foods, **Health Phys.,** 22: 43 - 48.

DE JESUS, A.S.M., MALAN, J.J., ELLERBECK, V.T., VAN DER BANK, D.J., MOOLMAN, E.W. 1987. **An assesment of the radium-226 concentration levels in tailings dams and environmental waters in the gold/uranium mining areas of the Witwatersrand.** PER-159. Pelindaba.

DEPARTMENT OF WATER AFFAIRS. 1986. **Management of the Water Resources of the Republic of South Africa.** CTP Book Printers. Cape Town.

DREESEN, D.R., WILLIAMS, J.M., MARPLE, M.L., GLADNEY, E.S. & PERRIN, D.R. 1981. **Mobility and Bioavailability of Uranium Mill Tailings Contaminants.** Rep. LA-UR-3733. Los Alamos National Laboratory. Los Alamos. NM.

D'SOUZA, T.J. & MISTRY, K.B. 1970. Comparative uptake of thorium-230, radium-226, lead-210 and polonium-210 by plants. **Radiat. Bot.,** 10: 293 - 295.

DU PLESSIS, L.J. 1991. Personal communication

DU PREEZ, W.J.S. 1991. Personal communication

EISENBUD, M. *et al.* 1964. Naturally occurring radionuclides in foods and waters from the Brazilian areas of high radioactivity. In: **Natural Radiation Environment.** (Proc. Int. Symp. Houston. 1963.) (ADAMS, J.A.S. & LOWDER, W.M. Eds.) pp. 837 - 854. University of Chicago Press. Chicago. IL.

ELLERBECK, V.T., SMIT, M.C.B & GERMISHUYS, G. 1990. **Voorbereiding van plant materiaal (Omgewingstudies).** Atoom Energie Korporasie van Suid-Afrika. Inligting en Handleidings: Verwysing 2/5/04.

EMERSON, S. & HESSLEIN, R. 1973. Distribution and uptake of artificially introduced radium-226 in a small lake. **J. Fish. Res. Board Can.** 30: 1485 - 1490.

ENGELBRECHT, C.J. 1989. **Operating Gold Mines and Gold Recovery Plants in the RSA.** Department of Mineral and Energy Affairs. Minerals Bureau. Braamfontein.

EPSTEIN, E. & LEGGETT, J.E. 1954. The absorption of alkaline earth cations by barley roots: Kinetics and mechanisms, **Am. J. Bot.**, 41: 785 - 791.

ETTENHUBER, E., CLAJUS, P. & ROHNSH, W. 1975. Bestimmung der **Akkumulationfatoren Fisch/Wasser einiger fur die Strahlenbelastung des Menschen Wichtiger Radionuklide.** Rep. SAAS-175, Staatliches Amt fur Atomsicherheit und Strahlenschutz, Berlin.

FEATHER, C.E. & KOEN, G.M. 1975. The Mineralogy of the Witwatersrand reefs. **Miner. Sci. Engng.**, 7, 189-224.

FISHER, D.R. 1988. Uranium. In: **Handbook on Toxicity of Inorganic Compounds.** (SEILER, H.G. & SIGEL, H. Eds.). Marcel Dekker, Inc. New York.

FLINN, F.B. 1926. Radioactive material an industrial hazard? **J. Am. Med. Assoc.**, 87: 2078 - 2081.

FRIED, M. & SHAPIRO, R.E. 1961. Soil-plant relationships in ion uptake. **Annu. Rev. Plant Physiol.**, 12: 91 - 112.

FUNKE, J.W. 1990. **The water requirements and pollution potential of South-African gold and uranium mines.** WRC Report No. KV 9/90. Water Research Commission. Pretoria.

GERAL, J. 1990. Personal communication

GRZYBOWSKA, D. 1974. Uptake of radium-226 by plants form contaminated soils. **Nukleonica**, 19(1): 62 - 68.

GUNN, K.B. & MISTRY, K.B. 1970. The effect of chelating agents on the absorption of radium by plants. **Plant Soil**, 33: 7 - 16.

HALLDEN, N.A., FISENNE, I.M. 1961. **Fallout Program Quarterly Summary.** Rep. HASL-112, United States Atomic Energy Commission, Washington, DC.

HALLDEN, N.A., HARLEY, J.H. 1964. Radium-226 in diet and human bone from San Juan, Puerto Rico, **Nature**, (London) 204: 240 - 241.

HAMILTON, E.I. 1972. **Health Phys.**, 22: 149 - 153.

HARRISON, G.E.; CARR, T.E.F. & SUTTON, A. 1967. Distribution of radioactive calcium, strontium, barium and radium following intravenous injection into a healthy man. **Int. J. Radiat. Biol.**, 13: 235 -247.

HAVLIK, B. 1970. Radioactive pollution of rivers in Czechoslovakia. **Health Phys.**, 19: 617 - 624.

HAVLIK, B. 1971. Radium in aquatic food chains: Radium uptake by freshwater algae. **Radiat. Res.** 46: 490 - 505.

HAVLIK, B., ROBERTSON, E. 1971. Radium uptake by freshwater algae. In: **Radionuclides in Ecosystems** (Proc. 3rd Natl Symp. Oak Ridge, TN, 1971) (NELSON, D.J. Ed.). CONF-710501-P1. United States Atomic Energy Commission. Washington, DC. 372 - 380.

HESSLEIN, R.H. & SLAVICEK, E. 1984. Geochemical pathways and biological uptake of radium in small Canadian Shield lakes. **Can. J. Fish. Aquat. Sci.** 41: 459 - 468.

HODGE, H.C. 1973. A History of Uranium Poisoning (1824 - 1942). In: **Uranium, Plutonium, Transplutonic Elements.** (HODGE, H.C., STANNARD, J.N. & HURSH, J.B. Eds.). pp. 5 - 68. Springer-Verlag. New York.

HOFFMANN, F.O., MILLER, C.W., SHAEFFER, D.L. & GARTEN, C.T. 1977. Computer codes for the assessment of radionuclides released to the environment. **Nucl. Safety**, 18: 343 - 354.

HOLTZMAN, R.B., URNEZIS, P.W., PADOVA, A. & BOBULA, C.M. 1979. **Contamination of the Human Food Chain by Uranium Mill Tailings Piles.** Rep. NUREG/CR-0758. ANL/ES-69. Argonne National Laboratory. Argonne. IL.

IAEA (International Atomic Energy Agency). 1990. Technical Report No. 310.

IBRAHIM, S.A., CHURCH, S.L. & WHICKER, F.W. 1984. The effects of barium chloride treatment of uranium ore on Rn-222 emanation and Ra-226 leachability from mill tailings. In: **Management of Uranium Mill Tailings, Low-Level Waste and Hazardous Waste.** (Proc. 7th Symp. Fort Collins. CO. 1984). (NELSON, J.D. Ed.). pp. 327 - 334. Colorado State University. Fort Collins.

ICRP (International Commission on Radiological Protection). 1973. **Alkaline Earth Metabolism in Adult Man.** ICRP Publication No. 20. Permagon Press. Oxford.

ICRP (International Commission on Radiological Protection). 1975. **Report of the Task Group on Reference Man.** Publication No. 23. ICRP. Permagon. Oxford.

ICRP (International Commission on Radiological Protection). 1977. **Recomendations of the International Commission on Radiological Protection.** Publication No. 26. ICRP. Pergamon. Oxford.

ICRP (International Commission on Radiological Protection). 1979.. **Radionuclide Release in the Environment: Assessment of Doses to Man.** ICRP Publication No. 29. Annals of the ICRP 2, 2C.

ICRP (International Commission on Radiological Protection). 1979. **Limits on Intakes of Radionuclides by Workers.** ICRP Publication No. 30. Part 1. Vol 2. No. 3/4/ Permagon Press. Oxford.

ICRP (International Commission on Radiological Protection). 1979. **Limits for Intakes of Radionuclides by Workers.** ICRP Publication No. 39. Part 1 - 3. Permagon Press. Oxford.

ICRP (International Commission on Radiological Protection). 1991. **1990 Recommendations of the International Commission on Radiological Protection.** ICRP Publication 60. Annals of the ICRP, 21 No. 1-3. Permagon Press, 1991.

ICRP (International Commission on Radiological Protection). 1991. **Annual Limits on Intake of Radionuclides by Workers Based on the 1990 Recommendations.** ICRP Publication 61. Annals of the ICRP, 21 No. 4. Pergamon Press, 1991.

INAGO, F. 1992. Personal communication

IYENGAR, M.A.R., 1990. The natural distribution of radium. In: **The Environmental Behaviour of Radium.** Technical Reports Series No. 310. IAEA (International Atomic Energy Agency). Vienna.

JACKSON, P.B.N. 1975. **Common and scientific names of the fishes of Southern Africa. Part II. Freshwater Fishes.** Special Publication No 14. J.L.B. Smith Institute of Ichthyology. Rhodes University. Grahamstown.

JENNE, E.A. & LUOMA, S.N. 1977. Forms of trace elements in soils, sediments and associated waters: An overview of their determination and biological availability. In: **Biological Implications of Metals in the Environment.** (Proc. 15th Annu. Hanford Life Sciences Symp. Richland. WA. 1975). pp. 110 - 143. CONF-750929. Technical Information Center. United States Energy Research and Development Administration. Oak Ridge. TN.

JONES, G.A., BRIERLY, S.E., GELDENHUIS, S.J.J. & HOWARD, J.R. 1989. **Research on the contribution of mine dumps to the mineral pollution load in the Vaal Barrage.** Report to the WRC by Steffen Robertson and Kirsten (PTA) Inc. WRC Report No. 136/1/89. Water Research Commission. Pretoria.

JUBB, R.A. 1967. **Freshwater fishes of Southern Africa.** A.A. Balkema. Cape Town.

JUSTYN, J. & STANEK, Z. 1974. Accumulation of natural radionuclides in the bottom sediments and by aquatic organisms of streams. **Int. Revue ges. Hydrobi ..**, 59(5): 593 - 609.

JUSTYN, J., LUSK, S. 1976. Evaluation of natural radionuclide contamination of fishes in streams affected by uranium ore mining an milling, **Folia Zool.**, 25(3): 265 - 274.

JUSTYN, J., MARVAN, P., ROSOL, J. 1979. Radioactive wastes and aquatic organisms, Vydal Vyzkumny ustav vodohospodarsky ve Statnim zemedolskem nakladatelstvi. Prague. **Prace a Studie, S.**, 151: 43 - 214.

JUSTYN, J. & HAVLIK, B. 1990. Radium uptake by freshwater fish. In: **The Environmental Behaviour of Radium.** Technical Reports Series No. 310. Vol. 1. pp. 529 - 543. IAEA (International Atomic Energy Agency). Vienna.

KALIN, M. & SHARMA, H.D. 1982. Radium-226 and lead-210 uptake in Typha latifolia from inactive uranium mill tailings in Canada. In: **Environmental Migration of Long-Lived Radionuclides.** (Proc. Int. Symp. Knoxville. TN. 1981.). pp. 247 - 262. IAEA. Vienna.

KHADEMI, B., ALEMI, A.A. & NASSERI, A. 1980. Transfer of radium from soil to plants in an area of high natural radioactivity in Ramsar, Iran. In: **Natural Radiation Environment III.** (Proc. Int. Conf. Houston. 1978). (GESELL, T.F. & LOWDER, W.M. Eds.). Vol. 1. pp. 600 - 610. CONF-780422. Technical Information Center. United States Department of Energy. Oak Ridge. TN.

KIRCHMANN, R., BOULENGER, R. & LAFONTAINE, A. 1966. Uptake of Ra-226 by crop plants. **Health Phys.**, 12: 1805 - 1806. (abstract only).

KIRCHMANN, R., LAFONTAINE, A., CANTILLON, G. & BOULENGER, R. 1973. **Etude du cycle biologique parcouru par la radioactvite.** Rep. BLG-477. Belgian Nuclear Centre (CEN/SCK). Mol, Belgium.

KLEMENT, A.W., Jr. 1982. **Natural sources of environmental radiotion.** CRC Handbook of Environmental Radiation (KLEMENT, A.W., Jr., Ed.). CRC Press. Boca Raton.

KLINGENBERG, M. 1981. Membrane protein oligomeric structure and transport function, **Nature**, (London) 290: 449 - 454.

LAL, N., SHARMA, Y.P., SHARMA, P.K., TALWAR, I.M., NAGPAUL, K.K. & CHAKARVARTI, S.K. 1982. Uranium assay in milk. **Health Phys.**, 43(3): 425 - 428.

LAPPENBUSH, W.L. & COTHERN, C.R. 1985. **Health Phys.**, 48: 535.

LE ROUX, P.J. & STEYN, L. 1968. **Fishes of the Transvaal.** Cape & Transvaal Printers Ltd. Cape Town.

LIEBENBERG, W.R. 1955. The occurrence and origin of gold and radioactive minerals in the Witwatersrand system, the Dominion reef, the Ventersdorp Contact and the Black reef. **Trans. Geol. Soc. S. Africa,** 58, 101-227.

LIEBENBERG, W.R. 1972. Mineralogical features of gold ores in South Africa. In: **Gold Metallurgy in South Africa.** Adamson, R.J. (Ed.). chap 11, pp. 352-426. Chamber of Mines of South Africa, Johannesburg.

LONG, S.E. 1987. **Uranium and human diet: a literature review.** Chemical Analysis Group. Environmental and Medical Sciences Division. United Kingdom Atomic Energy Authority. Harwell Laboratory. Oxforshire.

MALAN, J.J. 1981. **Die bepaling van radium-226 in die uitskot van goudmyne en hulle omgewings.** M.Sc. Tesis. Universiteit van Pretoria.

MARKOSE, P.M., EAPPEN, K.P., VENKATARAMAN, S. & KAMATH, P.R. 1978. Distribution of radium and chemical toxins in the environment of a uranium complex. In: **Natural Radiation Environment III.** (Proc. Symp. Houston, 1978). Technical Information Centre. Springfield. CONF-780422 (1980). 1655 - 1667.

MARTIN, S.S., HELM, W.T., SIGLER, W.F. 1969. Accumulation of 226-Ra in two aquatic ecosystems. **Radioecology.** (Proc. 2nd Natl. Sump., Ann Arbor, MI, 1967) (NELSON, D.J., EVANS, F.C., Eds.) CONF-670503, United States Atomic Energy Commission, Washington, DC, 307-318.

MARTINEC, M. 1973. Vliv tezby uranu na obsah 226-Ra v povrchove vode, rakosu a sladkovodnich rybach, **Cesk. Hyg.,** 18 (10): 475 - 478 (in Czech).

MARTLAND, H.S., CONLON, P. & KNEF, J.P. 1925. Some unrecognized dangers in the use of and the handling of radioactive substances, with especial reference to the storage of insoluble products of radium and mesothorium in the reticulo-endothelial system. **J. Am. Med. Assoc.,** 85: 1769 - 1776.

McMICHAEL, A.J. 1989. **The Contribution of Epidemiology to Understanding the Mechanisms of Action of Carcinogens.** Proceedings Vth International Congress of Toxicology. Brighton. pp. 25 - 35. Taylor & Francis. London-New York-Philadelphia.

MENZEL, R.G. 1965. Soil-plant relationships of radioactive elements. **Health Phys.,** 11: 1325 - 1332.

MOOLMAN, W., SMIT, M.C.B. & LABUSCHAGNE, M. 1990. **Verassing van dierlike weefsel.** Atoom Energie Korporasie van Suid-Afrika. Inligting en Handleidings: Verwysing 2/5/03.

MOORE, W.H. 1968. A light-weight pulsed d.c. fish shocker. *J. appl. Ecol.*, 5, 205 - 208.

MORISHIMA, H., KOGA, T., KAWAI, H., HONDA, Y. & KATSURAYAMA, K. 1977. **J. Radiat. Res.**, 18: 139 - 150.

MORROW, P.E., GIBB, F.R. & LEACH, L.J. 1966. **Health Phys.**, 23: 273.

MORROW, P.E., LEACH, L.J., SMITH, F.A., GELEIN, R.M., SCOTT, J.B., BEITER, H.D., AMATO, F.J., PICANO, J.J., YUILE, C.L. & CONSLER, T.G. 1982. **Metabolic Fate and Evaluation of Injury in Rats and Dogs Following Exposure to the Hydrolysis Products of Uranium Hexafluoride.** NUREG/CR-2268. New York.

MORSE, R.S. & WELFORD, G.A. 1971. Dietary intake of $^{210}$Pb. **Health Phys.**, 21: 53 - 55.

MUTH, H., RAJEWSKY, B., HANTKE, H.-J., AURAND, K. 1960. The normal radium content and the Ra-226/Ca ratio of various foods, drinking water and different organs and tissues of the human body, **Health Phys.**, 2: 239 - 245.

MYTTENAERE, C., BOURDEAU, P & BITTEL, R. 1969. Importance relative de l'eau et du sol dans la contamination indirecte en radiocesium et radiocobalt de rivieres irriguees. In: **Environmental Contamination by Radioactive Materials.** (Proc. Seminar. Vienna). pp. 175 - 182. International Atomic Energy Agency. Vienna.

NAPIER, B.A., PELOQUIN, R.A., STRENGE, D.L., RAMSDELL, J.V. 1988. **GENII - The Hanford Environmental Radiation Dosimetry Software System.** Pacific Northwest Laboratory - BATTELLE. PNL-6584 UC-600. Vol 1. December 1988.

NATHWANI, J.S. & PHILLIPS, C.R. 1978. Adsorption of Ra-226 by soils in the presence of Ca(+2) ions. Specific adsorption (II). **Chemosphere**, 5: 293 - 299.

NATIONAL TECHNICAL PLANNING GROUP. 1981. Report of the National Technical Planning Group on Uranium Tailings Research. Canada Centre for Mineral and Energy Technology (CANMET). Department of Energy, Mines and Resources. Ottawa.

NCRPM (National Council on Radiation Protection and Measurements). 1975. **Natural Background Radiation in the United States.** Rep. 45. NCRPM. Washington DC.

NG, Y.C., COLSHER, C.S. & THOMPSON, S.E. 1979. Transfer factors for assessing the dose from radionuclides. In: **Biological Implications of Radionuclides Released from Nuclear Industries.** Vol. 2. IAEA. Vienna. pp. 295 - 318.

OTAKE, M., YOSHIMARU, H. & SCHULL, W.J. 1987. **Severe Mental Retardation among the Prenatally Exposed Survivors of the Atomic Bombing of Hiroshima and Nagasaki: A Comparison of the Old and New Dosimetry Systems.** Radiation Effects Research Foundation RERF Technical Report. pp. 16 - 87. Hiroshima.

PALLY, M. & FOULQUIER, L. 1983. **Synthese bibliographique sur la capacite et les modalites de la fixation du radiostrontium par les vegetaux aquatiques.** Rep. CEA-BIB-238. CEA. Centre d'etudes nucleaires de Saclay. Gif-sur-Yvette.

PAUL, A.C., LONDHE, V.S., PILLAI, K.C. 1980. Radium-228 and radium-226 levels in a river environment and its modification by human activities. **Natural Radiation Environment III** (Proc. Int. Conf. Houston, 1978) (GESELL, T.F., LOWDER, W.M., Eds.), Vol. 2, CONF-780422, Technical Information Center, United States Department of Energy, Oak Ridge, TN 1633 - 1654.

PONNAMPERUMA, F.N. 1972. The chemistry of submerged soils. **Adv. Agron.** 24: 29 - 96.

PONTIUS,F.W. **Journal AWWA**, Vol 83 no 4, April 1991.

POPOVA, O.N., KODANEVA, R.P. & VAVILOV, P.P. 1964. Distribution in plants of radium absorbed from the soil. **Sov. J. Plant Physiol.**, 11: 371 - 375.

POPOVA, O.N. & KYRCHANOVA, A.N. 1974. Forms of Ra-226 deposition in plants. **Agrokhimiya**, 2: 110 - 115. (in Russian). English translation. Chem Abs. 81. Abstr. No. 74116m.

ROMNEY, E.M., WALLACE, A., GILBERT, R.O. & KINNEAR, J.E. 1976. Pu-239 and 240, and Am-241 contamination of vegetation in aged fallout areas. In: **Transuranium Nuclides in the Environment.** IAEA. Vienna. pp. 479 - 491.

RUNDO, J. & HOLTZMAN, R.B. 1976. Comparison of the late excretion of $^{226}$Ra and $^{239}$Pu by man. In: **Health Effects of Plutonium and Radium.** (JEE, W.S.S. Ed.). pp. 497 - 504. J.W. Press. salt Lake City. UT.

RUSANOVA, G.V. 1962. Study of leaching and migration of radium in soils. **Sov. Soil Sci.**, 9: 962 - 964.

SCHINDLER, P.W. 1990. Co-adsorption of metal ions and organic ligands: Formation of ternary surface complexes. In: **Mineral-Water Interface Geochemistry.** (HOCHELLA, M.F. & WHITE, A.F. Eds.). Vol. 23. Chp. 7. pp. 281 - 307. Reviews in Mineralogy. Mineralogical Society of America. Washington DC.

SCHMITT, H.W. & STICHER, H. 1991. Heavy Metal Compounds in the Soil. In: **Metals and Their Compounds in the Environment: Occurrence, Analysis and Biological Relevance.** (E. MERIAN Ed.). Weinheim. New York.

SCHÜTTELKOPF, H. & KIEFER, H. 1982. Radium-226 contamination of the Black Forest and the radioecological behaviour of radium. In: **Environmental Migration of Long-Lived Radionuclides.** (Proc. Int. Symp. Knoxville. TN. 1981). pp. 345 - 352. IAEA. Vienna.

SCOTT, L.M. 1973. Environmental Monitoring and Personnel Protection in Uranium Processing. In: **Uranium, Plutonium, Transplutonic Elements.** (HODGE, H.C., STANNARD, J.N. & HURSCH, J.B. Eds.). pp. 271 - 294. Springer - Verlag. New York.

SHEPPARD, M.I., SHEPPARD, S.C. 1985. The plant concentration ratio concept as applied to natural uranium, **Health Phys.**, 48(4): 494 - 500.

SIMON, S.L. & IBRAHIM, S.A. 1990. Biological Uptake of Radium by Terrestrial Plants. In: **The Environmental Behaviour of Radium.** Technical Reports Series No. 310. IAEA (International Atomic Energy Agency). Vienna.

SIMPSON, D.B. & McGILL, B.L. 1980. **Users Manual for LADTAP II. A computer program for calculating Radiation Exposure to man from routine release of nuclear reactor liquid effluents.** NUREG/CR-1276. ORNL 1980.

SMIT, D.S. 1989. Uranium. In: **South Africa's Mineral Industry.** (VAN ZYL, L., SCHREUDER, C.P. & VAN ZYL, W.J. Eds.). pp 61 - 66. Minerals Bureau. Department of Mineral and Energy Affairs. Braamfontein.

STARY J., ZEMAN, A., HAVLIK, B. 1983. Radionuclides in the investigarion of cumulation of toxic elements on algae and fish. **Isotopenpraxis**, 19(7): 243 - 244.

STATHER, J.W. 1990. The Behaviour, Effects and Radiation Dosimetry of Radium in Man. In: **The Environmental Behaviour of Radium.** Technical Reports Series No. 310. Vol. 2. pp. 297 - 343. IAEA (International Atomic Energy Agency). Vienna.

STEVENSON, F.J. & ARDAKANI, M.S. 1972. Organic matter reactions involving micronutrients in soils. In: **Micronutrients in Agriculture.** (MORTVEDT, J.J., GIORDANO, P.M. & LINDSAY, W.L. Eds.). Soil Science Society. Madison. Wisconsin.

STRAUB, C.P., MURTHY, G., CAMPBELL, J.E. 1961. Radionuclides in foods, **J. Am. Diet. Assoc.**, 38: 15 - 21.

SWANSON, S.M. 1985. Food-chain transfer of U-series radionuclides in a northern Saskatchewan aquatic system. **Health Phys.**, 49(5): 747 - 770.

TASKAEV, A.I., OVECHENKOV, V.Ysa., ALEKSAKHIN, R.M. & SHUKTOMOVA, I.I. 1976. Effect of pH and cation composition of the liquid phase on the extraction of radium-226 from soil. **Pochvovedenie**, 12: 46 - 50.

TASKAEV, A.I., OVCHENKOV, V.Y., ALEKSAKHIN, R.M. & SHUKTOMOVA, I.L. 1977. Uptake of Ra-226 by plants and change in its state in the soil-plant tops-litter fall system. **Sov. Soil Sci.**, 2: 79 - 85.

TERMINE, J.D. & POSNER, A.S. 1967. Ar[illegible]ous/crystalline interrelationships in bone mineral. **Calcif. Tissue Res.**, 1: [illegible] - 23.

THOMAS, D.C., McNEILL, K.G. & DOUGHERTY, C. 1985. Estimates of lifetime lung cancer risks resulting from radon progeny exposure. **Health Phys.**, 49: 825 - 846.

TILL, J.E. & MEYER, H.R. 1983. **Radiological Assessment: A textbook on Environmental Dose Analysis.** U.S. Nuclear Regulatory Commission. Washington DC. NUREG/CR-332 ORNL-5968.

TRACY, B.L., PRANTL, F.A. & QUINN, J.M. 1983. Transfer of $^{226}$Ra, $^{210}$Pb and uranium from soil to garden produce: assessment of risk. **Health Phys.**, 44(5): 469 - 477.

TRANGMAR, B.B., YOST, R.S. & UEHARA, G. 1985. Application of geostatistics to spatial studies of soil properties. **Adv. Agron.**, 38: 45 - 94.

TRAVENER, L. 1957. An historical review of the events and developments culminating in the construction of plants for the recovery of uranium from gold ore residues. In: **Uranium in South Africa 1946-1956.** Vol 1, p. 1. The Associated Scientific and Technical Societies of South Africa, Johannesburg.

TSIVOGLOU, E.C., BARTSCH, A.F., RUSHING, D.E. & HOLADAY, D.A. 1958. Effects of uranium ore refinery wastes on receiving waters. **Sewage Industrial Wastes**, 30: 1012 - 1027.

TSIVOGLOU, E.C., STEIN, M. & TOWNE, W.W. 1960. Control of radioactive pollution of the Animas River. **J. Water Poll. Cont. Fed.**, 32: 262 - 287.

UNITED NATIONS. 1977. **Sources and Effects of Ionizing Radiation** (Report to the General Assembly). Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). UN. New York.

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY. 1975. **Water Quality Impacts of Uranium Mining and Milling Activities in the Grants Mineral Belt, New Mexico.** Rep. EPA-906/9-75-002. USEPA. Washington. DC.

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY. 1983. **Potential Health and Environmental Hazards of Uranium Mine Wastes.** EPA-520/1-83-007. USEPA. Washington. DC.

USNRC (U.S. Nuclear Regulatory Commission). 1980. **Final Generic Environmental Impact Statement on Uranium Milling.** Vol. 3. NUREG-0706. Washington.

USNRC (U.S. Nuclear Regulatory Commission). 1982. Calculation Models for Estimating Radiation Doses to Man from Airborne Radioactive Materials Resulting from Uranium Milling Operations. Regulatory Guide 3.51. March. USNRC. Washington. DC.

VAN DORP, F., ELEVELD, R., FRISSEL, M.J. 1979. **A new approach for soil-plant transfer calculations, Biological Implications of Radionuclides Released from Nuclear Industries** (Proc. Int. Symp. Vienna, 1979), Vol. 2, IAEA, Vienna: 399-406.

VAN WYK, B. & MALAN, S. 1988. **Veldgids tot die veldblomme van die Witwatersrand- en Pretoria gebied.** Struik Uitgewers. Kaapstad.

VASCONCELLOS, L.M.H., AMARA, E.C.S., VIANNA, M.E. & PENNA FRANCA, E. 1987. Uptake of Ra-226 and Pb-210 by foodstuff cultivated in a high natural radioactivity region in Brazil. **J. Environ. Radioact.**, 5(4): 287 - 302.

VAVILOV, P.P., POPOVA, O.N. & KODANEVA, R.P. 1964. On the behaviour of radium in plants. **Sov. J. Plant Physiol.**, 157(4): 992 - 994.

VERKHOVSKAYA, I.N., VAVILOV, P.P. & MASLOV, V.I. 1967. The migration of natural radioactive elements under natural conditions and their distribution according to biotic and abiotic environmental components. In: **Radioecological Concentration Processes** (Proc. Int. Symp. Stockholm. 1966). (ABERG, B & HUNGATE, F.P. Eds.)..pp. 313 - 328. Permagon Press. Oxford.

VERKHOVSKAYA, I.N., *et al.* 1969. The content and translocation of natural radioactive elements in the system "soil-plants-animals" under natural and experimental conditions. In: **Radioecology.** (Proc. Int. Symp.). pp. 781 - 832. Departement de protection sanitaire. CEA. Centre d'etudes nucleaires de Fontenay-aux-Roses.

WAITE, D.T., JOSHI, S.R. & SOMMERSTAD, H. 1988. The effect of uranium mine tailings on radionuclide concentrations in Langley Bay, Saskatchewan, Canada. **Arch. Environ. Contam. Toxicol.**, 17: 373 - 380.

WEATHER BUREAU. 1992. Pretoria. Personal Communication.

WELFORD, G.A. & BAIRD, R. 1967. Uranium levels in human diet and biological materials. **Health Phys.**, 13: 1321 - 1324.

WILLIAMS, A.R. 1981. **Biological uptake and transfer of $^{226}$Ra: a review. Migration in the Terrestrial Environment of Long-lived Radionuclides from the Nuclear Fuel Cycle.** Proc. Int. Symp. Knoxville. International Atomic Energy Agency. Vienna. IAEA-SM-257.

WILLIAMS, A.R. 1982. Biological uptake and transfer of radium-226: A review. In: **Environmental Migration of Long-Lived Radionuclides.** (Proc. Int. Symp. Knoxville. TN, 1981). pp. 175 - 20[illegible]. IAEA. Vienna.

WILLIAMS, A.R. 1984. Biological Uptake and Transport. In: **The Behaviour of Radium in Waterways and Aquifers.** pp. 177 - 202. IAEA-TECDOC-301. IAEA (International Atomic Energy Agency). Vienna.

WILLIAMS, A.R. 1990. Radium uptake by freshwater plants. In: **The Environmental Behaviour of Radium.** Technical Reports Series No. 310. Vol. 1. pp. 487 - 507. IAEA (International Atomic Energy Agency). Vienna.

WILLIAMS, A.R. & KIRCHMANN, R.J. 1990. Radium: A historical introduction. In: **The Environmental Behaviour of Radium.** Technical Reports Series No. 310. Vol. 1. pp. 3 - 10. IAEA (International Atomic Energy Agency). Vienna.

WITTMANN, G.T.W. & FöRSTNER, U. 1977. Heavy metal enrichment in mine drainage. III. The Klerksdorp, West Wits and Evander Goldfields. **S. Sfr. J. Sci.,** 73: 53 - 57.

WOPEREIS, M.C., GASCUEL-ODOUX, C., BOURRIE, G. & SOIGNET, G. 1988. Spatial variability of heavy metals in soil on a one-hectare scale. **Soil Sci.,** 146 : 113 - 118.

WRENN, M.E., DURBIN, P.W., HOWARD, B., LIPSZTEIN, J., RUNDO, J., STILL, E.T. & WILLIS, D.L. 1985. **Health Phys.,** 48: 601.

YUILE, C.L. 1973. Animal Experiments. In: **Uranium, Plutonium, Transplutonic Elements.** pp. 165 - 196. (HODGE, H.C., STANNARD, J.N. & HURSH, J.B. Eds.). Springer-Verlag. New York.