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ON THE BEHAVIOUR OF RADIUM IN TAILINGS DAMS AND ENVIRONMENTAL WATERS IN THE WITWATERSRAND (SOUTH AFRICA) GOLD/URANIUM MINING AREA

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ABSTRACT

Natural radioactivity present in tailings impoundments from the gold/uranium mines on the Witwatersrand is a potential source of contamination of environmental waters. The reason is the presence of large activities, although at low concentrations, of radium-226, a long-lived radioactive decay product of uranium not recovered during the extraction of gold and uranium, which can lead to potential health hazards. An assessment of the situation on the Witwatersrand is made. Partition functions (k_d) in tailings impoundments are determined in field and laboratory studies, from which very low values for the mobility of radium are deduced. Results are supported by the measured migration of radium into the soil underlying 30-40 old impoundments. Clay materials, in particular pyrophyllite in the impoundments, are identified as being one of the main substances responsible for the retention of radium. The distribution of radium in surface and groundwaters is investigated. It is concluded that the spread of radium in the Witwatersrand environment is insignificant.

1. INTRODUCTION

Uranium mine waters contain primordial radioisotopes which have always been part of man's environment. As found in nature, these radionuclides contribute significantly to natural background radiation. When a more or less localised enhancement of such radionuclides is brought about by man's technological activities, some risk to man above that which would result from natural environmental concentrations alone may arise. This risk is largely caused by the presence of radium-226, a long-lived radioactive decay product of uranium not recovered during the extraction of uranium, which constitutes a potential source of contamination of environmental waters.

Obviously, there are many sites of natural high radium concentrations such as hot springs and outcrops of uraniferous materials, but the worldwide concern with the impact which radium may have on environmental waters is more specifically directed at activities concerned with the nuclear fuel

cycle, particularly the front end, i.e. the mining and milling of uranium. This concern is pertinently reflected in recent conferences and symposia (Man., 1978; Ura., 1980) and in research programmes organised and co-ordinated by the International Atomic Energy Agency, in which several countries participate (Beh., 1984).

South Africa is one of the main world producers of uranium, virtually all of it being recovered as a by-product in the gold mines of the Witwatersrand. This area has been mined for over 80 years and the total tailings, consisting of approximately 2 500 million tonnes of finely divided material, cover about 6 500 ha and make total containment of some 50 000 curies of radium-226 virtually impossible. The tailings dams are spread over an 80 km long strip (see Fig. 1), with 3.5 million people living within 20 km of this strip. Concern about the potential health effects of radium on the environment together with the fact that little information existed on the behaviour of radium in tailings dams and in surface and groundwaters constituted the motivation for this work.

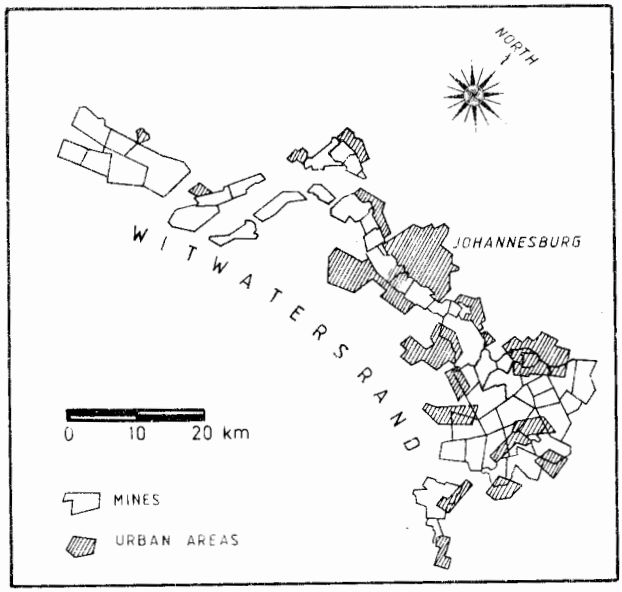

FIG. 1 - THE WITWATERSRAND MINING AREA

2. AN OVERVIEW OF RADIUM-226

2.1 The radionuclide radium-226

Radium was first discovered in 1896 by Marie and Pierre Curie (Curie, M. et al., 1898). Extensively used in medicine and industry as a source of radiation until the fifties, it was subsequently superseded by more suitable radioisotopes. Radium-226 has a high degree of radiotoxicity, is

bone-seeking, has a long half-life (1600 y) and, as an alpha emitter, has a high potential for biological damage when taken up in the human body. It was first identified as a potentially significant environmental pollutant only in the late fifties by Tsivoglou during his work in the uranium mining areas of the USA (Tsivoglou et al., 1958) and since then several other authors have reported similar findings (Havlik et al., 1968; Kirchmann et al., 1973).

2.2 Measurement of ^{226}Ra

Radium-226 can be determined by several methods. In the present work the emanometric method as described by Rushing was used (Rushing et al., 1967) as this is the method most widely employed, particularly with liquid samples. It is based on the measurement of the radon-222 emanated from radium-226 and it permits radium concentrations as low as a fraction of one picocurie per litre to be determined.

2.3 Source term

The main sources of radium contamination of environmental waters are plant effluents, which consist of process and mine waters enriched in dissolved and suspended radium, and solid wastes. Leaching and/or seepage through the solid wastes can carry appreciable amounts of radium to the aqueous environment.

2.4 Transport

Waste waters or portions of them are usually discharged into environmental streams or dams and the fate of the radium in the water depends on its physico-chemical form and on the handling and treatment of the water prior to discharge. The information available on the physico-chemical forms of radium in such waters is rather scant but it is presumed that, because of the high sulphate concentration remaining in the water even after its neutralisation, a significant part of the dissolved radium is present as neutral ion pair $Ra^{2+}SO_4^{2-}$. It should be stressed that formation of solid phase, consisting of radium sulphate only, is impossible at the radium concentrations typical for the waste waters, since the solubility product ($K_{so} = 10^{-10.37}$) of radium sulphate cannot be exceeded (Benes et al., 1982). Radium can, however, co-precipitate as $Ba(Ra)SO_4$ or $Ca(Ra)SO_4$.

Transport by seepage through the tailings dam is possible. Levins has reported seepages as large as 10 000 $m^3.d^{-1}$ from one tailings dam (Levins, 1979), but in general seepage flows are considerably lower. Several attempts have been made to mathematically analyse the transport of radium in tailings dams. Zettwoog for example, developed a model based on the one-dimensional diffusion equation while taking adsorption into consideration (Zettwoog, 1980), which produced good agreement between calculated and measured radium concentrations in an earthen dam after 20 years of tailings accumulation. A primary variable affecting the migration of radium in a tailings dam is the distribution of water content of the tailings as a function of time and depth. Klute and Heermann (1978) developed a mathematical model which characterises the movement and storage of water in tailings profiles and estimates future moisture distributions under various precipitation and evaporation conditions.

The leachability of radium from tailings has been extensively studied by

several workers (Shearer et al., 1964; Havlik et al., 1968; Levins et al., 1978) but these studies have been confined mainly to the laboratory as reliable results from field experiments are rather difficult to obtain. The main findings of these authors were that (a) the primary factor affecting the percentage of radium leached is the liquid/solid ratio; (b) the kinetics of radium released from the tailings is fairly rapid; (c) sulphates in solution tend to retard the release of radium; and (d) high concentrations of certain salts such as KCl and NaCl tend to promote the rapid leaching of radium.

2.5 Biological uptake

Owing to the similarity between the chemical behaviour of radium and calcium, the former is easily metabolised by biological systems although not to the same extent as Ca because Ca is a major nutrient and Ra is a non-nutrient. The presence or absence of major ions in solution has a strong influence on the biological uptake of non-nutrient substances (Polikarpov, 1968). Since uranium mill effluent usually contains high concentrations of dissolved Ca, SO_4, Cl, Mg, Fe and Mn, which will change the salinity of the water, radium uptake will be affected by the presence of such ions. According to McDowell-Boyer et al. (1979) most radium in soil is strongly fixed in the solid phase and only a small fraction of the mobile radium is directly available for root uptake. The entire mobile fraction, however, may be said to constitute the available pool, since the chemical forms are not static, i.e. previously acid-soluble or exchangeable radium may enter the water-soluble phase to re-establish equilibrium conditions as radium is depleted from the available phase.

2.6 Health hazards

Mme. Curie was probably the first victim of a health hazard caused by radium-226 when she succumbed to the effects of the then new substance. Although the external radiation dose to the human body can be high if it is exposed to a strong radium source, the radium concentrations usually prevailing in mine wastes are so small from the point of view of external irradiation that the resulting doses can be neglected. The true health hazard lies in ingestion of radium, even in very small quantities, since when metabolised, it will necesssarily result in biological damage to the tissues surrounding the site in the human body where it happens to become localised. Another health hazard which is currently receiving worldwide attention is that brought about by the daughter of radium-226, radon-222 which, being a radioactive gas that decays into other radioactive products, can have detrimental effects when inhaled. However, environmental waters can hardly be regarded as a significant source of radon-222, and the health hazards arising from it will not be discussed any further in the context of this work.

The volume of solid wastes, particularly tailings, is usually large, which severely limits the options available to isolate the tailings from the environment over very long time spans. The concentration of radium in tailings is very small - about 50 g of radium per ton of tailings for a 0.02 % ore. This extremely low concentration of radium and its progeny contained in tailings makes separation of the radioisotopes from the bulk of the tailings impracticable, at least with today's technology and costs. Since the precise dimensions of the radiation risk from mill tailings is difficult to assess and control measures costly, management of

mill tailings has been and continues to be controversial. Present trends are to use cost-benefit methodologies in which the detriment from radiation doses below the dose limit is balanced by the cost of achieving this level of protection.

3. MINE WASTE MANAGEMENT IN THE WITWATERSRAND

Although the gold/uranium association in most South African gold mines has been known since 1923 (Travener, 1957), it was not until the late forties that uranium extraction started in earnest. The main uraniferous constituent of Witwatersrand ores is uraninite (Liebenberg, 1972), which normally occurs in a complex matrix of conglomerates. In the reefs of the West Rand the most common uranium mineral is brannerite, a uranium-bearing titanate (Feather, 1957).

All mining is done by underground methods and the wastes which originate from the milling stage consist mainly of the solid fraction of the tailings, liquid effluents from the leaching plant plus smaller volumes of other liquids, decant solutions and seepage from waste retention systems as well as from contaminated runoff from the plant area. Treatment of the effluents by barium chloride to precipitate dissolved radium is normally not practised in South Africa as the radium concentrations in the effluents are considered by the mining community to be too low to justify such treatment.

Uranium is usually extracted by sulphuric acid leaching, the sulphuric acid being manufactured from the iron pyrite which is present in the gold ore. The principal chemical form in which radium is present in the tailings is $RaSO_4$, but a strong likelihood exists that the radium ion is also adsorbed onto the surface of particulate material and that chemical dissolution or desorption from the surface could play an important role in the transport and distribution mechanisms of radium.

South Africa currently produces some 5 500 tonnes of uranium per annum from ore ranging from 0.015 % to 0.05 % uranium as a by-product of gold and copper. Much of this "ore" is the previously milled gold tailings; hence both the "ore" and the subsequent tailings are currently located in tailings impoundments on the surface. This contrasts greatly with the situation in areas primarily mined for uranium, such as in Canada (Wiles, 1978) and Australia (Levins, 1978), where ore grades of between 2 % and 4 % uranium are mined. There is thus a difference of about two orders of magnitude in the radium concentrations in these wastes compared with those in South Africa.

The accepted practice in most South African mines is to minimise the water discharge to the tailings impoundment by diverting as much water as possible to a separate return-water dam. Water on and in the impoundment is limited by using thickened discharges, rapid decanting to the return water and some underdrainage (Blight, 1979).

With a precipitation of approximately 700 mm/annum, the climate of the Witwatersrand is relatively dry. Most of the rain falls in summer, when occasional torrential rains could give rise to considerable wash-off. On the other hand, winters are dry and cool to cold. Thus most of the water discharged onto the tailings dams will evaporate, resulting in drying out of the tailings, and radon may become the radionuclide of concern.

4. RADIUM IN TAILINGS

An extensive survey aimed at assessing the prevailing radium-226 levels in the tailings impoundments of the Witwatersrand was conducted. These levels were found to vary between 6 and 15 pCi/g for the West Rand and 15 and 85 pCi/g for the East Rand, with the overall mean at around 15 pCi/g. Ordinary soil contains 1-2 pCi/g. Many of the impoundments from which the samples originated have already been decommissioned for periods which vary between a few years and a few decades. Consequently, their state of conservation varies considerably from site to site. While some of the impoundments have been subjected to intensive revegetation programmes or covered with protective layers of crushed stone or slag to prevent erosion by wind and water, there are impoundments showing clear signs of erosion and resultant deposition of tailings, particularly the finer fraction, in the vicinity of the impoundment.

In spite of the relatively dry climate of the Witwatersrand, phreatic zones can nevertheless be formed in the impoundments, with resulting seepage into the underlying soil continuing for many years. Because seepage rates are largely dependent on the hydraulic conductivity of the tailings, a series of in situ and laboratory experiments was conducted to determine the prevailing seepages. The in situ measurements were performed in two 30-50 year old tailings impoundments which contained phreatic zones in spite of having been decommissioned for many years. The partition functions (k_d), defined as the ratio of the concentration sorbed per gram of tailings to the equilibrium concentration in the water saturating the tailings, were determined with the aid of tailings samples augered out of the phreatic zone. Using the k_d values thus obtained in expression (Rahn, 1978)

$$V_i = \frac{V}{1 + \frac{k_d B_d}{\theta}}$$

where V_i = radium-ion velocity;
V = water velocity;
B_d = bulk density of tailings; and
θ = volumetric moisture content of the tailings

it was possible to calculate the velocity of the radium relative to that of water. The results are shown in Table 1 below.

In the case of the laboratory measurements use was made of 1 250 mm long columns, each packed with a different type of tailings. A small amount of tailings (50 g) onto which approximately 8×10^5 pCi of radium-226 had been adsorbed was introduced at the top of each packed column, which was then subjected to a constant water head feeding the percolation through the column. In this way it was possible to measure the migration of the radium pulse as well as its velocity relative to that of water under very conservative conditions, i.e. saturated flow conditions. All columns were continuously operated for well over a year. Fig. 2 shows a typical set of results, from where it may readily be seen that after 550 days of continuous flow through the tailings in the column the radium had moved only 2.5 cm. It is also worth noticing that very little of the original radium in the bulk of the tailings was leached away, as deduced from the difference between the original and final radium concentrations below a depth of about 5 cm. The results for all columns, also shown in Table 1, are consistent amongst themselves and point to an extremely low mobility

of radium in the tailings impoundments studied. They further show fairly good agreement between in situ and laboratory results. The findings that the radium in the impoundments moved 1 300-2 900 times more slowly than the water could explain the rather low radium contents (< 2 pCi/ℓ) of seepage waters measured at the base of some impoundments.

Having established that the radium-ion adsorbs strongly onto the particulate material, the study was pursued one step further. It was assumed that a fair possibility existed that the radium was associated not

TABLE 1
MOBILITY OF RADIUM IN TAILINGS

	Tailings type					
	T1	T2	T3	T4	T5	T6
Column measurements:						
Duration of percolation (days)	540	525	460	540	380	550
Radium migration in the column (cm)	0.4±0.1	0.4±0.1	1.8±0.2	2.1±0.2	3.0±0.2	2.5±0.2
Partition function (k_d)	1 400 ± 400	700 ± 180	870 ± 130	400 ± 50	430 ± 50	640 ± 80
Velocity of water relative to that of radium	4 500 ±1 200	2 300 ± 600	3 200 ± 420	1 400 ± 160	1 300 ± 130	2 200 ± 220
In situ measurements:						
Partition function (k_d)	840 ± 70	630 ± 60	-	-	-	-
Velocity of water relative to that of radium	2 700 ± 510	2 070 ± 380	-	-	-	-

T1 = ± 40-year-old tailings (East Rand - mine A)
T2 = ± 40-year-old tailings (East Rand - mine B)
T3 = ± 2-year-old oxidised tailings (East Rand)
T4 = fresh tailings before neutralisation (West Rand)
T5 = fresh tailings after neutralisation (West Rand)
T6 = ± 3-year-old non-oxidised tailings (West Rand)

with the bulk tailings material but with one or more of the minor ion-exchanging components (clay materials) which may be present at low concentrations. Examination of the mineral composition of the Witwatersrand ore readily revealed that two of such clay minerals, namely pyrophyllite and sericite, are present at levels of up to 16 % and 2 %, respectively. The bulk component consists of 80-90 % quartz. The adsorption characteristics of quartz and pyrophyllite in relation to radium were determined by separately contacting (shaking) particulate material, of these minerals in various pH solutions containing radium-226 and measuring the k_d values. The results, for contact periods of one

month, are displayed graphically in Fig. 3, from where it may readily be seen that adsorption onto both quartz and pyrophyllite increases rapidly with increasing pH and that adsorption onto pyrophyllite is about two orders of magnitude greater than onto quartz. The latter is indeed quite small as deduced from the very low values for k_d, even at high pH. Lack of opportunity did not permit repeating the experiment with sericite.

Virtually all uranium extraction in the Witwatersrand is done by means of acid leaching, with lime being used to neutralise the tailings prior to their discharge to the impoundments. In practice the mines aim at a pH of 8 or 9. In the presence of air and moisture, oxidation of the pyrite in

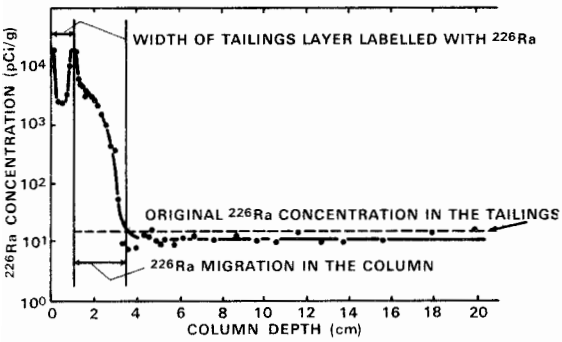

FIG. 2 – 226Ra DISTRIBUTION IN A PACKED COLUMN AFTER 550 DAYS.

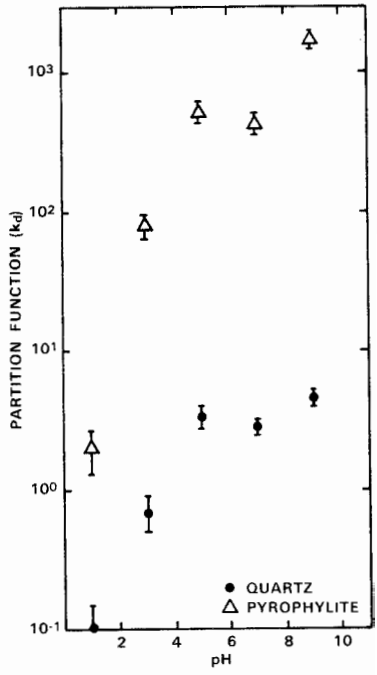

FIG. 3 – ADSORPTION OF 226Ra IN QUARTZ AND PYROPHYLITE.

the tailings through the action of the bacterium Thiobacillus ferrooxidans gives rise to the production of H_2SO_4 which enhances leaching of residual uranium as soluble $(UO_2)SO_4$. As a result of the high pH the initial oxidation is slow but as the pH decreases bacterial activity increases, with corresponding speeding up of the oxidation process. It is mainly the top 30 cm which will eventually become strongly oxidised although bacterial activity has been noticed as deep as 3 m (Mrost, 1971).

As the pyrite is oxidised the acid so formed could solubilise "acid soluble" radium and also, if radium is adsorbed on the pyrite, its oxidation could liberate this radium and make it available for leaching. To test this hypothesis two samples of fresh tailings were taken simultaneously and sterilised. One sample was kept sterile while the

other was inoculated with the abovementioned bacterium and growth stimulated through the addition of small amounts of nutrients. After virtually all the pyrite had been oxidised, both samples were leached with water and the leached radium-226 was measured. Contrary to expectations, the results seem to indicate that more radium was leached from the sterile than from the inoculated sample. However, due to some discrepancies in the radium accountancy, the results must be regarded as inconclusive.

The opportunity afforded by the removal of five 30-40 year old tailings impoundments (at different localities in the Witwatersrand) for reprocessing was used to establish the degree of radium contamination of the underlying soil. One-metre deep auger holes were drilled and the radium profile determined. As shown in Fig. 4, it was found that although the pH of the soil increased with increasing depth from values of 3 to 6.5, thus reflecting the influence of the overlying impoundments, the radium concentrations (1-2 pCi/g) remained fairly constant with depth and compared well with those of control samples collected from areas far removed from mining activities.

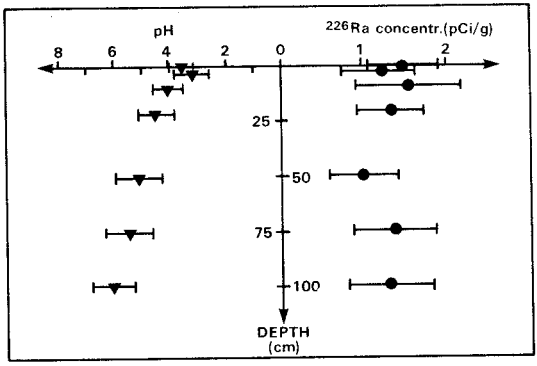

FIG. 4 – MEAN ^{226}Ra CONCENTRATION AND pH IN PROFILES OF SOILS UNDERLYING OLD TRAILINGS DAMS.

5. RADIUM DISTRIBUTION IN ENVIRONMENTAL WATERS

5.1 Surface waters

The impact which radium in surface waters may have on the environment of the Witwatersrand mining area depends on how that environment relates to the people living there. Such waters are usually more accessible to people and other biological systems and, in the case of rivers, can potentially lead to widespread contamination of adjacent areas. The environment concerned contains major metropolitan areas (see Fig. 1), and rural regions around and between mines carry a considerable amount of crop, sheep and cattle farming.

Contamination of surface waters can occur virtually only during mine

operations as it is caused mainly by discharge of milling effluents (process waters) into water courses. From all the mines in the Witwatersrand approximately 30 % have their liquid wastes totally contained within the mine property while the remaining 70 % discharge variable portions of their liquid effluents into channels or streams which eventually reach the environment. In the West Rand, where there are large underground aquifers contained in dolomitic formations, considerable amounts of water (mine waters) are pumped out of the underground works for de-watering purposes as well as from boreholes (dolomitic waters) for irrigation and domestic use. Approximately 60 % of the latter waters are discharged into public streams and lakes.

Fairly comprehensive tests were carried out at many mines, whereby radium concentrations were measured at the point of effluent release at the mine and at several points downstream through to the public streams. It was found that in general no significant enhancement of radium concentrations was occurring. In a few isolated cases, where radium concentrations of up to 30 pCi/ℓ in the waste channel were measured, these fell to levels of about 1 pCi/ℓ within 3-4 km of the release point. Some cases were observed where visible chemical contamination of the water had clearly taken place. However, the radium content of the water seldom exceeded 3 pCi/ℓ. Process, mine, and dolomitic waters, measured at the points of release, were found to average 15, 8 and 0.8 pCi/ℓ, with maximum values of 30, 13 and 1 pCi/ℓ, respectively. Dams and lakes used for recreation purposes, some of them in very close proximity to old tailings dams, gave radium concentrations analogous to those of natural waters.

For purposes of comparison it is appropriate to mention that the derived limit for the concentration of dissolved radium-226 in drinking water is 10 pCi/ℓ, according to a previous recommendation of the International Commission for Radiological Protection.

5.2 Groundwaters

Owing to the relatively large amounts of radium which the impoundments contain, they are perhaps the most significant single contributors to possible enhancement of radium in groundwater. Even when an impoundment is decommissioned, seepage into the underlying soil could continue for many years (Nelson, 1980), the quantity of seepage release being dependent not only on the seepage control system but also on the properties of the tailings and foundation soil or rock beneath the impoundment and the nature of the groundwater in the area.

In Section 4 it was shown that the migration of radium in the Witwatersrand tailings is so small that possible contamination of groundwaters through seepage from the tailings impoundments is in all probability equally small. This has largely been verified by the consistently low radium contents measured in the dolomitic waters of the mining areas of the West Rand, as pointed out in Section 5.1. Observations based on a few dozen boreholes spread throughout the Witwatersrand, whose waters were tested for radium, are very similar. In one specific case where water clearly contaminated with manganese was flowing through underground channels into a major river in the Western Transvaal, the origin of the manganese was traced to seepage from the tailings impoundment of a gold/uranium mine situated at approximately 1 km from the river. Yet, the radium content of such water was below 1 pCi/ℓ, as was that of a borehole drilled on top of the impoundment to reach the

water table. Several boreholes drilled close to this impoundment and on the downwards slope of the water table produced similar results.

Mine waters originating at the underground works of various mines, often at depths in excess of 2 000 m, sporadically showed radium values above 10 pCi/ℓ.

5.3 Biota

Streams and rivers into which effluents from the Witwatersrand rivers are discharged supply irrigation water for the considerable farming activity of the area. Also, those water courses often develop into fairly large marshy areas used by cattle and sheep for grazing. The survey was therefore extended to include some biological materials. Plant life samples were collected in the vicinity of well-established tailings impoundments and streams while samples of milk and bones of cattle raised in those areas were also analysed for their radium content. The results, shown in Table 2, clearly indicate that no significant difference exists between such specimens and control specimens collected in areas with no mining activities. Where no values for control samples are mentioned (fresh vegetables, green fodder and maize) the results still compare well with those of similar samples in other countries.

TABLE 2
RADIUM-226 CONCENTRATIONS IN BIOLOGICAL MATERIALS

Material	^{226}Ra concentration (pCi/g ashed material)			
	Mining areas		Control areas	
	Range	Mean	Range	Mean
Milk	0.20–0.80	0.35	0.38–0.68	0.54
Grass	0.47–0.98	0.70	0.50–0.98	0.83
Cattle bones	0.26–1.27	0.64	0.58–0.78	0.64
Reeds	–	0.78	0.54–0.60	0.57
Fresh vegetables	0.53–0.89	0.65	–	–
Green fodder	–	0.22	–	–
Maize	0.58–1.18	0.96	–	–

6. SUMMARY AND CONCLUSIONS

In this study an empirical approach supported by collection, quantification and evaluation of environmental data regarding radium was used to gain long overdue information concerning its behaviour both in tailings impoundments and in the environmental waters of the Witwatersrand. Because gold/uranium mining activities have existed for many decades - and thus with their effects on the environment reasonably crystallised - it was possible to assess the magnitude of one important facet of the radiological impact of mining on the Witwatersrand environment. The other facet, also related to uranium, is that caused

by radon and is currently being investigated.

The evidence gathered shows no significant enhancement of radium concentrations in the environmental waters in and around the Witwatersrand mining area. In spite of the vast quantities of total radium contained in the tailings impoundments, its relatively low concentration together with the verification of its extremely low mobility indicate that its spread in the environment, at least in the short to medium term, is unlikely to reach proportions that could cause concern.

7. ACKNOWLEDGEMENTS

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