

## **REPORT ON THE RADIOACTIVITY MONITORING PROGRAMME IN THE MOOI RIVER (WONDERFONTEINSPRUIT) CATCHMENT.**

### **EXECUTIVE SUMMARY**

A radioactivity monitoring study was conducted by the Institute for Water Quality Studies (IWQS) of the Department of Water Affairs and Forestry (DWAF) in collaboration with a wide group of interested parties, in the Mooi River Catchment during 1997. The study served to establish the drinking water health risk, as well as the radiological status of the water resources, in the catchment from the viewpoint of drinking water. The intensive monitoring, both in time as well as in number of radionuclides measured served to clear up many areas of doubt, and has established with reasonable certainty the representative radiological status of the water resources in the catchment. The study covered surface streams and groundwater sources in the catchment. The evaluation of health risk was based on the levels of radioactivity in raw water samples that had been filtered prior to analysis, and on the use of such water for drinking purposes on a continuous basis. The relative contributions to the health risk from ingestion of the suspended solids in the water and from radiation exposure scenarios other than drinking water use were, with the possible exception of fish consumption, shown to be insignificant. The study did not consider radioactivity in sediments.

The radiological variables measured were all from the natural radioactive decay chains of uranium-238, uranium-235, and thorium-232. In addition to radiological variables, a full set of chemical variables was also monitored.

The radiation doses calculated in the study were based on the conservative assumption that the water at every sampling point was used continuously as the sole source of drinking water.

In view of the controversy surrounding the radiological status of water sources in the catchment, extensive efforts were made to validate the accuracy of the radiological measurements, as well as to cross check the validity of the total yearly doses calculated for each monitoring point. The total yearly dose was independently calculated by two different methods, which gave very similar radiation doses.

The set of dose criteria, used to evaluate the dose values found for drinking water, ranged from the ideal level of the World Health Organisation of 0,1 mSv/year, through the 0,25 mSv/year single facility dose limit used by the Council for Nuclear Safety, to the 1,0 mSv/year dose limit of the International Atomic Energy Agency for public exposure from anthropogenic sources. These dose criteria have been incorporated into proposed interim radioactivity water quality guidelines, with associated actions and interpretation.

The natural background radiation dose in drinking water in the catchment was estimated at 0,020 mSv/year. The great majority of sampling sites in the catchment showed a total drinking water radiation dose below 0,1 mSv/year, implying that no radiological problem exists from the viewpoint of drinking water. The general conclusion was that of the 41 sites monitored, 39 showed a water quality which is either ideal or acceptable for continuous lifetime use in terms of the proposed interim water quality guidelines for radioactivity in drinking water. Five sites had a dose between 0,1 and 0,25 mSv/year, showing a slightly larger increase above local natural background, but still fully acceptable for lifetime use with no significant detrimental effects to the user. Only two sites had significant elevation of the radiation dose which showed the need for planning to reduce the exposure over the course of time. Both these sites involved the discharge of mine water that had been pumped to the surface.

A highly relevant and comforting finding of the study was that the total radiation dose for both Potchefstroom untreated raw drinking water supply points was very low, and in fact not significantly different from the natural background dose value estimated for the study.

A valuable finding of the study was the good linear correlation between total radiation dose from all radionuclides and the uranium concentration. This will, in the future, make it possible to use the uranium concentration for screening and routine monitoring purposes within the catchment.

As regards the two classical screening parameters for radiation, viz., alpha and beta activity, the former showed a reasonably good correlation with total radiation dose, when compared on an annual average basis. The gross beta activity measurements were considered to be unreliable because of measurement difficulties at the low levels encountered. As regards chemical variables, while it was found that elevated radiation dose is usually associated with elevated sulphate concentrations, the converse was not true, consequently sulphate concentration cannot be used as an indicator of radioactivity in the water.

The water analysis technique involves filtering of the raw water samples prior to radiometric analysis, and the primary intention of the study was to measure only the radioactivity in the water passing through the filter. In the final month of the study, however, the radioactivity in the suspended solids trapped by the filter was also measured, as a preliminary indication of whether the suspended solids were of any significance as regard the possible radiation dose from ingestion of untreated water.

While an important aim of the study was to measure the concentrations of a large range of radionuclides in the natural uranium and thorium decay chains, it was not the intention to look at radon gas dissolved in the water. Dissolved radon, even at relatively high concentrations, does not contribute significantly to the drinking water health risk, and is generally considered to be of possible concern only where significantly elevated radon concentrations are associated with conditions that promote the dissolution and release of the gas into poorly ventilated enclosures. Such scenarios, which might include indoor spa baths and underground water treatment plants, are not known to exist in the Mooi River catchment.



## **1. INTRODUCTION**

### **1.1 Background**

Preliminary screening surveys of radioactivity in water sources was carried out by the Institute for Water Quality Studies in 1995 and 1996 [1,2]. The levels of the radioactive elements uranium and radium, found in streams in the vicinity of gold mining activities, were found to be elevated such that, in some cases, these streams might be regarded as unsuitable for continuous lifetime use as drinking water. Many radionuclides had not been measured, and there was no information on the variability of the radionuclide concentrations in the water sources. Due to the lack of detailed and definitive data on radionuclide concentrations, it was not possible to determine the safety or otherwise of the water sources when used for drinking water purposes without a more thorough and intensive monitoring programme. In order to obtain certain knowledge on the radiological status of the water sources to establish human health risk, it was essential that a more detailed investigation be conducted. This report summarizes the findings of an intensive radiological monitoring programme that was conducted in the Mooi River catchment during 1997.

### **1.2 Aims of the Study and Strategy Adopted**

The aims of the radioactivity monitoring programme were:

- (i) To measure and report on the most important radioactive components in surface streams and in groundwater at selected sampling locations, at regular intervals over a hydrological year.
- (ii) To establish the radiation dose from untreated water for the purposes of use as drinking water, the emphasis being placed on the dissolved component of the radionuclides present in the water samples, and not on the suspended component.
- (iii) To estimate, from such measurements, the incremental radiation doses above estimated background that could be received by users of the water.
- (iv) To establish, on the basis of international practice, guidelines for interpreting the significance of these incremental radiation doses with a view to identifying the need for remedial action at any particular location. The recommendations would be brought to the attention of the relevant authorities within DWAF.
- (v) To identify where further investigative work, beyond the scope of this study, was needed.
- (vi) To identify strategies for efficient monitoring.

For clarity it is important to note what the study aims did not address. The study focussed on the radioactivity status of raw water, filtered before analysis. Some preliminary work was done on the suspended solids. All the possible uptake routes were investigated in detail, of which only the drinking water route and possibly the fish consumption route were found to be significant. Sediments, dissolved radon gas and airborne gas and dust were outside the scope of the study.

The strategy adopted to achieve the aims comprised the following:

- (a) To focus on one catchment at a time, in order of priority - the Mooi River catchment (also known as and also containing the Wonderfonteinspruit catchment) was selected as the first catchment to be studied, and forms the basis of this report.
- (b) To undertake the monitoring programme in a coordinated, transparent manner with the participation of relevant governmental and non-governmental stakeholders.

### **1.3 Management and Co-ordination of the Programme**

The IWQS was responsible for the management and coordination of the radioactivity monitoring programme in the Mooi River catchment. The establishment of a Coordinating Committee and Technical Committee, involving representation from a wide spectrum of organizations interested or involved in monitoring of radioactivity, assured transparency and the involvement of a range of scientific opinion and decision making on the issue. The Coordinating Committee consisted of numerous individuals and role players including Rand Water, Goldfields Water, the Western Transvaal Water Company, the Lower Wonderfontein spruit Catchment Forum, AngloGold Limited, Gold Fields of South Africa and other representative mining companies such as Randfontein Estates, the School of Chemical Engineering of the University of Potchefstroom, the Directorate: Water Quality Management and the Gauteng Regional Office of DWAF, the Council for Nuclear Safety (CNS), the Atomic Energy Corporation (AEC), the Chamber of Mines, the Council for Geoscience and the Department of Minerals and Energy.

The Technical Committee included representation from the AEC, the Chamber of Mines, the Gauteng Regional Office, the Council for Geoscience, the Council for Nuclear Safety, and the IWQS.

## **2. MONITORING PROGRAMME**

### **2.1 Selection of Catchment**

The Mooi River Catchment (Figures 1 and 2), was selected as the first priority catchment for intensive radioactivity monitoring for reasons including the following:

- (i) Major gold mining activity is carried out in the region, with the potential for pollution of surface and ground water. The region has several large active gold mines which discharge fissure and process water into the aquatic environment.
- (ii) The upper section of the catchment has numerous diffuse sources from old and abandoned mine workings and mine residue deposits.
- (iii) There are many informal settlements within the region, giving rise to possible consumption of untreated surface and ground water.
- (iv) Formal townships, closely related to the mining activities, occur in the catchment. Carletonville municipality abstracts a small portion for water use from boreholes and Potchefstroom municipality abstracts water from the Boskop dam for domestic water use. During the course of the study, questions were raised regarding elevated levels of radioactivity in streams, within the catchment, that could have a negative impact on the quality of the untreated raw water supplied to Potchefstroom, located at the lower end of the catchment.

### **2.2 Characterisation of the Mooi River Catchment and Water Use**

The Mooi River catchment consists of the Mooi River, Wonderfontein Spruit (Mooi River Loop) and Loop Spruit. The various dams situated in the catchment include the Donaldson, Klipdrift, Boskop and Potchefstroom (Lakeside) Dams. The catchment is situated on the Far West Rand with the upper section in the Gauteng Province and the lower part of the catchment in the North West Province. The Mooi River and its tributaries receive contamination from a wide variety of point and diffuse sources. The headwaters of the Wonderfontein Spruit originate around the mine residue deposits of several old and abandoned mines. These mine tailings dams, sand dumps and rock dumps are potentially significant contributors to diffuse contamination. Furthermore, numerous active gold mines are discharging fissure and process water into the water environment.

Most of the area is underlain by dolomite of which three of the dolomite compartments are dewatered by the gold mines. The water in the Wonderfontein Spruit is diverted into a one-metre diameter pipeline, which transports the water over two of the dewatered compartments. The Mooi River and its tributaries run through the magisterial districts of Potchefstroom, Westonaria, Oberholzer, Fochville and Carletonville. A number of growing communities are located in the catchment, including Kagiso, Mohlakeng, Toekomsrus, Rietvallei and Bekkersdal. These developments, as well as informal developments, contribute to the diffuse sources of pollution.

Rand Water supplies nearly all the water required for domestic use in the area, excluding Potchefstroom and the lower Mooi River area which is supplied by Potchefstroom municipality from the Boskop Dam. Carletonville Municipality sometimes extracts water for Welverdiend from a borehole in the Turffontein compartment.

Industrial use of water from the Mooi River is concentrated in and around Potchefstroom. Some water is abstracted by farmers along the lower reaches of the river for livestock watering and domestic supplies. The Mooi River is further used for angling and general recreational purposes.

Data on water usage by the various informal communities in the catchment were gathered primarily to establish usage for drinking water purposes (Appendix 1). This was important for determining the degree of conservatism inherent in assuming sole continuous use of the water for drinking purposes.

### **2.3 Selection of Monitoring Sites**

During the initial stages of the monitoring programme 39 sampling locations (28 surface water sites, and 11 groundwater sites) were selected on the recommendation of the Gauteng Regional Office (Figure 2). Sampling was started in January 1997. In addition to the sites selected initially, the two untreated, raw water abstraction points at the Potchefstroom purification works were added, some time after initiation of the monitoring programme.

Table 1 summarises the sampling site information and identifies the location of the sites.



**TABLE 1: Site, station numbers and monitoring point names, together with positional data.**

Station number	Site No	Monitoring Point Name	Waterbody Name	Latitude	Longitude
C2H152Q01	1	Luipaardsvlei (At rail bridge from Turk Shaft to 1st West Gm)	Wonderfontein Spruit [C2]	26°08'23"	27°46'00"
C2H153Q01	2	Rietvlei (Randfontein Azaadville bridge)	Wonderfontein Spruit [C2]	26°09'52"	27°46'02"
C2H154Q01	3	Luipaardsvlei (Doornkop Randfontein (R559) road bridge)	Wonderfontein Spruit [C2]	26°15'57"	27°41'58"
C2H025Q01	4	No 7 At Gembokfontein	Wonderfontein Spruit [C2]	26°17'18"	27°40'09"
C2H080Q01	5	Wonderfontein-End of 1m Pipe from Venterspost Gold Mine	Venterspost Gold Mine-Transfer [C2]	26°19'35"	27°24'38"
C2H030Q01	6	Oog Van Wonderfontein-Canal from Wonderfontein Eye	Wonderfontein Eye [C2]	26°18'47"	27°29'20"
C2H155Q01	6A	West Driefontein (down stream North Shaft Purification Works)	West Driefontein Gm-Fissure Water [C2]	26°21'49"	27°28'22"
C2H063Q01	7	Canal at Rooipoort	West Driefontein Gm-Transfer [C2]	26°20'26"	27°25'33"
C2H156Q01	7A	Carltonville West Driefontein Gm-C.Ville Cemetary Road Bridge	West Driefontein Gm-Process Water [C2]	26°21'31"	27°26'00"
C2H157Q01	8	Wonderfontein-Low water bridge to Abe Bailey Nature Reserve	Mooirivierloop [C2]	26°19'25"	27°21'15"
C2H069Q01	9	Blaauwbank	Mooirivierloop [C2]	26°22'32"	27°13'51"
C2H158Q01	10	Blyvooruitzicht Gold Mine-discharge To Doornfontein canal east of Pw	Blyvooruitzicht Gm-Fissure Water [C2]	26°23'15"	27°22'24"
C2H159Q01	11	Doornfontein Gm-Gold Plant discharge in canal upstream Doornfontein excess	Doornfontein Gm-Fissure Water [C2]	26°22'31"	27°20'12"
C2H160Q01	12	Doornfontein Gold Mine-Number 3 Shaft discharge	Doornfontein Gm-Fissure Water [C2]	26°25'29"	27°21'02"
C2H161Q01	13	Turffontein-gravel road bridge to Muiskraal	Mooirivierloop [C2]	26°26'05"	27°09'07"
C2H162Q01	14	Gerhard Minnebron-Rysmierbult road bridge upstream of Boskop Dam	Mooirivierloop [C2]	26°30'52"	27°07'29"
C2H163Q01	15	Western Deep Levels-farm bridge downstream of No 7 Shaft Slimes Dam	Varkenslaagte Spruit [C2]	26°26'06"	27°20'22"
C2H164Q01	16	Buffelsdoorn-Elandsrand Gold Mine	Elandsrand Gm-W Nursery Dam Overflow[C2]	26°26'44"	27°20'40"
C2H165Q01	17	Deelkraal Gold Mine recreational dam overflow	Deelkraal Dam-Outlet [C2]	26°27'18"	27°19'05"
C2H166Q01	18	Buffelsdoorn-Johannesburg/Potchefstroom road bridge	Buffelsdoorn Spruit [C2]	26°29'33"	27°22'24"
C2H167Q01	19	Elandsfontein-Johannesburg/Potchefstroom road bridge	Elandsfontein Spruit [C2]	26°27'24"	27°25'15"
C2H168Q01	20	Kraalkop-Old Johannesburg/Potchefstroom road bridge	Kraalkop Spruit [C2]	26°26'21"	27°29'56"
C2H169Q01	21	Weltevreden-Losberg/Bank road bridge	Loop Spruit [C2]	26°28'44"	27°32'22"
C2H170Q01	22	Klipdrift Dam-Outflow into concrete irrigation canal	Loop Spruit [C2]	26°37'01"	27°17'46"
C2H171Q01	23	Gempost-Venterspost Gold Mine Pipe from No 5 Shaft	Venterspos Gold Mine-Fissure Water [C2]	26°24'29"	27°10'42"
ZLUIPAAR1	24	Plot 40 Luipaardsvlei-35m south east of farm house	Borehole [C]	26°14'06"	27°44'49"
ZCARLTON1	25	Plot No 9 Carltonville	Borehole [C]	26°19'41"	27°22'24"
ZWELVER1	26	Plot at Welverdiend	Borehol [C]	26°22'13"	27°19'38"
ZWELVER2	27	Wolverdiend municipal water supply 2km south of Welverdiend	Borehole [C]	26°23'54"	27°17'16"
ZBLAAUB1	28	Blaaubank 100m east of house	Borehole]	26°23'03"	27°12'40"
C2H013Q01	29	Turffontein	Upper Turffontein Eye [C2]	26°24'29"	27°10'42"
C2H011Q01	30	Gerhardminnebron	Gerhardminnebron Eye [C2]	26°28'37"	27°09'09"
ZWONDER1	31	Oog Van Wonderfontein 110 between piggery buildings	Borehole [C]	26°17'41"	27°29'05"
ZDEPAN1	32	Plot 84 De Pan	Borehole [C]	26°15'38"	27°26'07"
ZKRAALK1	33	Plot Kraalkop	Borehole [C]	26°26'26"	27°28'40"
C2H172Q01	34	Bovenste Oog Van Mooirivier	Bovenste Oog [C2]	26°12'02"	27°09'45"
C2H173Q01	35	Mooi River: Potchefstroom Purification Works-Western abstraction point from canal	Boskop Dam-Outlet [C2]	26°39'37"	27°05'09"
C2H174Q01	36	Mooi River: Potchefstroom Purification Works-eastern abstraction point	Potchefstroom Dam-Outlet [C2]	26°39'42"	27°05'11"
C2H175Q01	37	Harry's Dam (Uitspanning at Wonderfontein)	Wonderfontein Spruit [C2]	26°20'10"	27°20'15"
C2H176Q01	38	Doringdraai Dam Welverdiend	Varkenslaagte Spruit [C2]	26°23'18"	27°16'27"
C2H033Q01	39	Doornfontein	Buffelsdoorn Spruit [C2]	26°26'12"	27°19'38"







Factors taken into account in the selection of the sites included:

- the potential for large-scale drinking water use,
- the identification of significant point-source discharges from mines,
- the need to establish, as far as possible, natural background levels.

## **2.4 Sampling Frequency and Duration**

Since, for chronic radiation exposures, it is the cumulative radiation dose that is important, doses to the public are normally integrated over a full year of exposure for the purposes of assessment. The exact yearly dose from environmental radioactivity, which varies over time, particularly in water sources, can only be determined with high frequency monitoring, ideally on a continuous basis. This was, however, not possible in practice due both to analytical capacity constraints and to budgetary constraints. A compromise had to be reached to ensure reasonable accuracy of the estimation of the integrated annual dose. Thus to achieve a reasonable estimate of integrated annual radiation dose, a weekly sampling frequency and a 25 week sampling duration was adopted for the first phase of the study (7 January to 25 June 1997). Preliminary analysis of the data from the first phase of the study showed that significant autocorrelation existed for the radioactivity data gathered at intervals of less than one month (see Appendix 3). This implied that the sampling frequency could be reduced to once a month without a significant loss in the ability to estimate the annual dose with a reasonable degree of accuracy. Thus, during the second phase of the study (July to December 1997), data was gathered on a monthly rather than on a weekly basis.

## **2.5 Geological and Radiological Characteristics of the Catchment**

Because gold mining was established in the Mooi River catchment long before radioactivity measurements were made, it was not possible to establish unequivocally the true natural background level, especially as the natural ground water recharge constitutes a significant proportion of the base flow of the river. Recent gamma ray spectrometric surveys and a large body of radioactivity measurements on geologically similar areas for airborne radiometric mapping of the environmental impact of gold and uranium mining in Gauteng Province, South Africa, were also reported by Coetzee, H, (1995) [10]. The pertinent geological factors are as follows:

- The dolomitic areas (most of the Mooi River catchment is underlain by dolomite) have very low (~10% of crustal average) radio-element contents. These dolomites also constitute the major groundwater source in the area.
- The quartzites and shales in the area tend to be enriched in potassium, uranium and thorium and consequently, the daughter nuclides of uranium and thorium reach levels generally at 1.5-3 times the crustal average.
- The granites tend to contain slightly elevated uranium concentrations and elevated potassium and thorium concentrations.

The highest naturally occurring uranium series activities in the area are found in the gold reefs of the Witwatersrand Supergroup. These, however, are extremely limited in outcrop, generally sub-outcropping below hundreds or thousands of metres of younger cover rocks.

## **2.6 Variables Measured and Data Collected**

### **2.6.1 Radionuclides**

The three natural radioactive decay series of relevance are those headed by the radionuclides uranium-238, uranium-235 and thorium-232. Details of these decay series and an explanation of terms are given in Appendix 2. The radiological variables originally requested from the AEC for analysis were gross alpha activity and the individual activities of uranium-238, radium-226 and thorium-232. The AEC contributed significantly to the study by determining, in addition, gross beta activity and the individual activities of radium-223, radium-224 and uranium-235. During the second phase of the study the number of radiological nuclides measured was increased to include polonium-210, lead-210, thorium-230, thorium-227, uranium-234, and radium-228. This was done in order to clarify uncertainties in the dose

calculated, relating to the non-equilibrium of nuclides with the parent nuclides in the water phase. It was also decided that the protactinium-231 and actinium-227 in the water samples had to be determined on a limited set of samples.

Additional analyses on the last batch of samples were also performed. These analysis included radiological variables on the suspended solids that were left on the filter in the samples.

The use of gross beta measurements for estimating the contributions of beta emitters to the total radiation dose could not be considered, because the measurements were deemed to be unreliable owing to analysis problems caused by the effects of water chemistry. The AEC concurred that the well-established gross beta measurement techniques used by them could not be regarded as suitable for the determination of very low beta activity concentrations in waters characteristic of those sampled in this study. It was accordingly decided not to accept the gross beta data set, but rather to measure those beta emitters likely to contribute significantly to the total ingestion dose, in phase two of the study. Beta emitters measured included lead-210, radium-228, and actinium-227

The methods used for radiological analysis of the samples are given in Appendix 2.

### **2.6.2 Chemical Variables**

Chemical variables, both major inorganic and trace metal constituents, were measured by the IWQS laboratories. The primary reason for collecting chemical variables was to establish whether a relationship could be found between dose and the chemical variables, so as to answer the question as to whether any of the chemical variables could be used as surrogate parameters.

The chemical variables measured were:

- a. The following metals (dissolved fraction): aluminium, barium, bismuth, iron, manganese, lead, yttrium and germanium.
- b. The following major inorganic determinands: pH, electrical conductivity, total alkalinity, sodium, potassium, calcium, magnesium, ammonium, chloride, fluoride, sulphate, nitrate + nitrite (as N), phosphate as P, and silicate as Si.

The most significant of the chemical variables measured was possibly sulphate, which is formed by the oxidation of pyrite in the mine residue deposits, leading to acidic conditions conducive to the mobilization of some radionuclides into water.

### **2.6.3 Other Data**

Although the radiological data gathered in this study related primarily to radioactivity in the dissolved constituents of the water, limited data were gathered also on radioactivity in the suspended solids. No data on environmental levels of radioactivity in sediments, river banks, vegetation or other possible elements of the human food chain were gathered. Instead, potential radiological impacts from exposure pathways other than drinking water were estimated on an order-of-magnitude basis through the use of screening models.

Other data collected were flow and rainfall data where available. From the very limited river flow and rainfall data that was available for the catchment, no correlation could be established with the radiological data. Unfortunately very few radiation monitoring sites corresponded with flow gauging sites. In the few sites that did correspond, the flow was heavily influenced by man made structures such as dams, weirs, canals and treatment works. This resulted in a highly modified pattern of flow which displayed little or no correlation with radioactivity.

### **2.6.4 Access to Analytical Data**

Analytical results collected during the study can be obtained from the Hydrological Information System (HIS) of the Department of Water Affairs and Forestry. Requests for data from the HIS can be sent directly to:

Directorate: Hydrology  
 Department of Water Affairs and Forestry  
 Paterson 536  
 Private Bag X313  
 Pretoria 0001  
 Tel: (012) 338 7500, ask for the Data Supply Section in Directorate: Hydrology  
 Fax: (012) 326 1488

The official departmental station numbers, provided elsewhere in the report (example C2H073) should be provided with all data requests. Data can be provided in an ASCII format and files can be provided via e-mail.

## 2.7 Quality Control

A number of actions were taken to address quality control. As a quality control measure, split samples were analyzed by three laboratories, as part of phase two (Appendix 5). These confirmed the accuracy of the radiological analyses.

The AEC conducted the radiometric analyses of the water samples for the study. As a CNS-recognised laboratory, the AEC adopts approved methods and procedures for analysis, and incorporates specific quality control methods. The quality control and validation done by the AEC's Radioanalytical Laboratory is shown in Appendix 6.

Measurements of uranium by both radiochemical and ICP-MS techniques, during the second phase of the study, allowed comparisons to be made as an additional quality control check. The following good correlation for uranium concentration in mg/l was obtained by linear regression from the 98 samples analysed:

$$[U]_{\text{ICP-MS}} = 0,993 \times [U]_{\text{Radiochemical}} - 0,563 \quad (r^2 = 0,906)$$

Thorium-232 was also measured by both techniques in the second phase, but a correlation between the two techniques could not be established because the ICP-MS measurements were frequently at the lower limit of detection and therefore inapplicable.

In natural uranium, the activity ratio between uranium-238 and uranium-235 is 21,719. The following good correlations, between the data for the two isotopes, were obtained by linear regression:

Radiochemical (phase 2), 98 data:	$^{238}\text{U} / ^{235}\text{U} = 21,341 \pm 0,115$	$(r^2 = 0,996)$
ICP-MS (phase 1), 570 data:	$^{238}\text{U} / ^{235}\text{U} = 20,785 \pm 0,030$	$(r^2 = 0,999)$
ICP-MS (phase 2), 63 data:	$^{238}\text{U} / ^{235}\text{U} = 22,171 \pm 0,571$	$(r^2 = 0,860)$

## 3. BASIC RADIOLOGICAL CONSIDERATIONS

### 3.1 Exposure from Natural Background Radioactivity and Medical Procedures

Most of the ionizing radiation to which people are exposed comes from sources which are natural features of the environment. These sources include radon gas and its decay products in the atmosphere (originating from natural uranium in soil and rocks), gamma rays from the ground, cosmic rays from outer space, naturally-occurring radioactivity in foodstuffs and drinking water, derived from radionuclides in the soil, as well as inhalation of respirable airborne dust. The total radiation dose received by an individual, from these natural sources, is typically about 2,4 mSv/a (millisieverts per annum), but geological and geographical factors can cause doses from any one of such sources to be elevated by a factor of 10 in high-background regions [3].

In addition to radiation from natural sources, man is exposed to radiation during medical treatment (X-rays, radiotherapy and nuclear medicine). Internationally, average doses to individuals from all medical sources range from 0,07 mSv/a to 1,8 mSv/a [3].

Thus, a typical member of the public will receive, as a matter of course, a radiation dose of between 2,5 and 4,2 mSv/a. In regions with high natural background, doses of 10 mSv/a are not uncommon.

### 3.2 Exposure Pathways

Exposure of humans, to ionizing radiation, may occur via various routes or 'pathways' that can be grouped simply as:

- exposures to penetrating radiation from sources external to the body, and
- exposures to both penetrating and non-penetrating radiation from radioactive substances taken into the body by ingestion, inhalation, or absorption through the skin.

Exposures from water containing radioactive contaminants essentially occur internally through ingestion, either by direct consumption or indirectly by consumption of animal or vegetable products that have themselves taken up the water.

A detailed study of the potential major ingestion pathways, relevant to the Mooi River catchment, revealed only two pathways with potential for giving rise to significant exposures ( Appendix 7).

- direct ingestion resulting from regular and continuous use of the water for drinking purposes, and
- regular consumption of fish obtained from contaminated water bodies.

With respect to the latter, there is very little information on the bioaccumulation rates of radionuclides in local fish species, and international experience shows that bioaccumulation can vary by as much as three orders of magnitude. The fish pathway therefore requires more research, and could not be addressed in the present study. Accordingly, the decision was taken to address only the drinking water pathway in this study.

### 3.3 Health Effects of Ionizing Radiation

The process of ionization changes atoms and molecules. In cells, such changes may result in damage which, if not adequately repaired, may:

- prevent the cell from surviving or reproducing, or
- result in a viable but modified cell.

The two outcomes have profoundly different implications for the organism as a whole.

In the case of the former, the loss of large numbers of cells in a tissue can result in a loss in tissue function. Such effects are known as deterministic effects, and are characterized by a dose threshold above which the probability of causing harm increases steeply from zero to 100%. Above the threshold, the severity of harm also increases with dose. Threshold doses are generally two or three orders of magnitude above background doses, and deterministic effects are thus only now seen in the case of accidents or as a side effect of medical radiation therapy.

The outcome is very different if the irradiated cell is modified rather than killed. It may then be able to produce a clone of modified daughter cells which, in spite of the highly effective defence mechanisms within the body, may cause, after a prolonged and variable delay, a malignant condition - a cancer. The probability, but not the severity, of the cancer increases with dose. This effect is called stochastic (meaning of random or statistical nature).

Epidemiological studies have shown, with good statistical significance, that this dose-response relationship is linear for accumulated doses of more than about 200 mSv. It is widely assumed that this linear relationship, with certain corrections, holds true also at lower doses, all the way down to zero - that is, there is no dose threshold for stochastic effects. This linear relationship yields, for low doses and dose rates, a nominal probability of fatal

cancer induction of  $5 \times 10^{-5}$  per mSv. Due to the high incidence of cancer induced by other carcinogens, it will be difficult, if not impossible, to obtain conclusive epidemiological evidence supporting this linear relationship at low doses. Some evidence suggests the opposite, in that there is actually a beneficial effect.

Stochastic effects can also take the form of hereditary effects which may be of many different kinds and severity, and are expressed in the progeny of the exposed person. Although the existence of hereditary effects in man is not in doubt, the risk estimates appear to be so small that it is not surprising that epidemiology has not yet detected hereditary effects of radiation in humans with a statistically significant degree of confidence.

Notwithstanding the fact that there is no evidence of statistically significant health effects associated with exposure to low levels of radiation, the internationally accepted principle is to keep radiation exposures as low as reasonably achievable.

### 3.4 Radiation Protection Principles and the System of Radiation Protection

Internationally a system of radiation protection has been agreed upon, based on the health effects described in section 3.3. This system has been recommended by the International Commission on Radiological Protection (ICRP), which is a non-governmental scientific organization that has been publishing this and related recommendations for over half a century. Different countries evaluate and implement the recommendations in a manner that is appropriate to their circumstances.

The following recommendations of the ICRP [4] are based on the assumption that there is indeed a linear non-threshold relationship between radiation dose and the probability of contracting cancer. Central to the system of radiation protection for proposed and continuing human activities that increase exposure to radiation are the following general principles:

1. No activity, which results in the exposure of persons to radiation, should be adopted unless the activity produces a net positive benefit.
2. All radiation doses should be kept as low as reasonably achievable (ALARA), taking economic and social factors into account.
3. The radiation doses should not exceed limits recommended by the ICRP.

For situations where the sources of exposure are already in place and radiation protection has to be considered retrospectively, remedial action to reduce the exposures should be based on the following general principles:

- a) The remedial action should be justified in the sense that the costs, including social costs, should be more than offset by the reduction in radiation dose likely to be achieved.
- b) The form, scale and duration of the remedial action should be optimized so that the net benefit to society is maximized.

To apply the above principles to, for instance, radioactivity in water, it is necessary to calculate the radiation doses which result from the use of the water.

### 3.5 Calculation of Dose for the Drinking Water Ingestion Pathway

The annual radiation dose from any given radionuclide and for any given age group is expressed as:

$$\begin{array}{ccccccc} \text{Annual} & & \text{Activity} & & \text{Annual} & & \text{Dose} \\ \text{dose} & = & \text{concentration} & \times & \text{consumption} & \times & \text{Conversion} \\ \text{(mSv/a)} & & \text{(Bq/}\ell\text{)} & & \text{(\ell/a)} & & \text{Factor} \\ & & & & & & \text{(mSv/Bq)} \end{array}$$

The total radiation dose for that age group is, then, the sum of the doses from individual radionuclides. This implies that the activity concentration of every radionuclide must be known. However, it was not feasible to measure every radionuclide, and this had to be taken into account in the calculation of age group specific doses. The method used to calculate lifetime average doses in this report is given in Appendix 8.

Two methods (IWQS and AEC) are presented in the Appendices for calculating the dose. Both need to address the problem that fewer nuclides were measured in the first phase than in the second. The so-called IWQS method handled this problem in two ways :-

1. Where a suitable set of predictor variables (chemical or radioactive) could be found, the missing nuclides were regressed onto a set of predictor variables. The multilinear regression was used to predict what the value of the nuclide was during the first phase.
2. Where no set of predictor variables could be found that performed better than just using the average, the average value was used.

The so called AEC method handled this problem by regressing, for the period of Phase II, those nuclides measured in Phase I onto the dose calculated from all the nuclides measured in Phase II. This regression was used to predict the dose for Phase I. The IWQS and AEC methods differed in the assumptions used of how to deal with unmeasured nuclides.

The second problem that needed to be addressed by both methods was the fact that even in Phase II, not all the nuclides in the decay chains were measured. The so-called IWQS method took a simpler approach to this for the purpose of estimating the uncertainty in the dose arising from not measuring these nuclides. The IWQS method simply assumed that all the unmeasured nuclides had the same value. This implied that the uncertainty remaining in the dose due to the unmeasured nuclides was about 0,003 mSv/a. The AEC method had a more advanced model, based on which nuclide was related to which other via a decay chain of the shortest half-life.

### **3.5.1 Dose Conversion Factor**

Each of the radionuclides in the three decay chains of interest has its own 'dose conversion factor' (DCF) for the ingestion pathway, relating the dose received, in mSv, to the amount of radioactivity ingested, in Bq (becquerels, or number of nuclear disintegrations per second). The DCFs used are those published by the International Atomic Energy Agency (IAEA) [5]. The IAEA gives different dose conversion factors for the various age groups. There are various ways in which the exposure dose per year for the various age groups can be combined. Investigation into the possible ways in which to combine the age groups specific doses showed that differences for the various ways of determining lifetime exposure were in fact trivial, and a "lifetime average" method was adopted for the purposes of this study.

### **3.5.2 Activity Concentration**

In many solid materials such as rocks and soil, the mobility of the elements in the decay chains is limited, even over long periods of time, and the mixture of radionuclides is therefore relatively undisturbed. In such cases, the radionuclides may be said to be in secular equilibrium, meaning that all the radionuclides in a given decay chain have similar activity concentrations.

In water systems, however, the dissolution and precipitation characteristics of the various decay chain elements may differ significantly, leading to a high degree of disequilibrium. Assumptions of equilibrium are, therefore, no longer valid. On the other hand, measurement of the activity concentration of every single radionuclide is neither economically feasible nor necessary in order to obtain a reasonable estimate of the ingestion dose. Certain radionuclides will contribute very little to the overall radiation dose because they have very small DCFs and / or their parents may be present only at very low activity concentrations.

In the first phase of the study, the parent radionuclides of the three decay series, plus the three radium isotopes radium-226, radium-223 and radium-224 that occur near the mid-points



of each series, were measured. In addition, uranium-234 was assumed to be in equilibrium with uranium-238 on the basis of results from other studies [6]. From the results of this first phase, it was established that only three radionuclides of major importance remained unaccounted for: thorium-230, lead-210 and polonium-210. These were measured in phase 2 of the study, together with three radionuclides of lesser importance: actinium-227, protactinium-231 and radium-228, and, therefore, made it possible to calculate the estimated annual dose with a high degree of certainty.

Consideration was initially given to the use of gross alpha measurements for estimating the dose contributions from the radionuclides that were not individually measured. In practice, however, the uncertainties inherent in the determination of gross alpha activity, typically around 20% to 30%, lead to unacceptably large uncertainties in the final dose determination.

The use of gross beta measurements for estimating the contributions of beta emitters to the total radiation dose could not be considered, because the measurements were deemed to be unreliable owing to elevation of the beta measurements caused by water chemistry. The AEC concurred that the well-established gross beta measurement techniques used by them could not be regarded as suitable for the determination of the very low beta levels in the waters characteristic of those sampled in this study. It was accordingly decided not to use the gross beta data in dose calculation, but rather to directly measure the more important beta emitters, with the highest dose conversion factors during the second phase of the study.

#### **4. DRINKING WATER QUALITY CONSIDERATIONS**

From the preliminary screening surveys [1,2] uranium was found to be the main radioactive element present, and has both a potential for a chemical toxicity and a radiological hazard. Current DWAF Water Quality Guidelines [7] give criteria for uranium-238 concentrations in drinking water. These criteria are based on the chemical toxicity of uranium to the kidney rather than its radiological toxicity.

From a radiological perspective, it is the total radiation dose from all radionuclides in the water that is important, and the Technical Committee has proposed interim guidelines in this regard, taking into account the following:

- (i) The World Health Organization (WHO) recommends a reference level for radiation dose, received from the continuous consumption of drinking water for a full year, of 0,1 mSv/a [8]. This value is only about 5% of the dose from the total natural background, and can therefore be regarded as an ideal situation. From section 3.3 it can be deduced that, on the conservative basis of the linear non-threshold theory, a radiation dose of 0,1 mSv/a represents a probability of attributable fatal cancer of 4 in 10 000 over a 70 year lifetime. By contrast, cancer from all causes is responsible for about 2 000 in 10 000 deaths, it thus being evident that, for the WHO reference level of 0,1 mSv/a, the increase in the probability of cancer induction, if it exists at all, is insignificantly small.
- (ii) The dose limit to members of the public due to all anthropogenic sources is currently recommended internationally at 1 mSv/a [4,5], and this has been implemented in several countries. This value is based on acceptance of the linear non-threshold theory, and can therefore be regarded as conservative.
- (iii) The dose limit for members of the public, recommended internationally, was previously 5 mSv/a, and many countries still adopt this limit. It is common practice in uranium mining remedial action programmes to design the programmes such that compliance with the 5 mSv/a limit is achieved in the short term, and with the 1 mSv/a limit in the longer term.
- (iv) Dose limits to members of the public relate to the combined effect of all exposures from human activities. It is common practice to place a dose constraint on releases from individual facilities. Such a constraint is normally set at some fraction of the dose limit of 1mSv/a - commonly of the order of 0,25 mSv/a, i.e., allowing for the combined dose from up to four separate facilities on a single individual not exceeding the 1mSv/a limit. Although this approach is intended for new rather than existing operations, it may have some relevance to water systems in gold mining areas in that

it embodies the concept of allowing for doses from other sources of exposure without causing the 1 mSv/a dose limit to be exceeded. The reference value of 0,25 mSv/a is the dose limit already imposed by the CNS on individual mines in the Mooi River catchment.

Table 2 gives information on the DWAF guidelines for uranium, while Table 3 embodies the proposed guidelines on radiation dose in drinking water. The DWAF guidelines have taken into consideration all the above limits for the protection of the public from anthropogenic sources of radiation.

The basis for the colour coded classification system was chosen to be in line with the approach used in the joint Assessment Guide, published by DWAF, the Department of Health, and the Water Research Commission [9]. The meaning of the colour classes for chemical constituents given in this DWAF/DOH/WRC guide are as follows:

- **Blue, class 0 = Ideal water quality. Suitable for lifetime use.**
- **Green, class I = Good water quality. Suitable for use, rare instances of negative effects.**
- **Yellow, class II = Marginal water quality. Conditionally acceptable. Negative effects may occur in some sensitive groups.**
- **Red, class III = Poor water quality. Unsuitable for use without treatment. Chronic effects may occur.**
- **Purple, class IV = Dangerous water quality. Totally unsuitable for use. Acute effects may occur.**

The practical meaning intended for interpretation of the classes is that “blue” or “green” water is fit for lifetime use without any further questions. Yellow class or marginal water, is however, only fit for interim use, and should not be used for a lifetime if at all possible. Red and purple class water are seen as unfit for use.

**Table 2: Current DWAF 1996 guideline [7] on uranium-238 in domestic water, with colour classes**

Uranium-238 (Bq/ℓ)	Uranium-238 (mg/ℓ)	Effects	Colour Class
Target water quality range 0 to 0,89 Bq/ℓ	Target water quality range 0 to 0,070 mg/ℓ	No significant effects. Annual cancer risk less than 1 in 4 000 000.	Blue, Ideal(<0,25 Bq/ℓ) and Green (0,25 to 0,89 Bq/ℓ)
0,89 to 3,6	0,070 to 0,284*	Annual cancer risk less than 1 in 1 000 000. May potentially be a slight risk of renal toxicity in sensitive individuals where renal function is impaired, but unlikely to have demonstrable renal toxicity in healthy individuals.	Yellow
3,6 to 18	0,284 to 1,42	Annual cancer risk less than 1 in 200 000, but significant risk of chemical toxicity with renal damage.	Red
>18	>1,42	Increasing cancer risk in long term. Increasing risk of renal damage in short term.	Purple

\* If 0,284 mg/ℓ is exceeded, human health may be at risk due to chemical toxicity.

**Table 3: Proposed interim water quality guidelines for the radiation dose in drinking water**

Radiation dose (mSv/a)	Suitability	Action required	Colour Class
≤ 0,1 (WHO reference level)	Ideal, suitable for lifetime use	Water complies fully with radioactivity guideline. No further action necessary	Blue (ideal)
>0,1 and ≤ 0,25	Water acceptable for lifetime use, subject to confirmation of dose.	Confirm dose level in respect of specific nuclide analysis	Green (acceptable for lifetime use).
>0,25 and ≤ 1	Water acceptable for short term use. Use in longer term (lifetime) requires further investigation.	An environmental impact assessment may be necessitated	Yellow (acceptable for short term use)
>1 and ≤ 5	Unacceptable for lifetime use	Remediation required over a reasonable time period.	Red (Unacceptable for lifetime use)
>5	Unacceptable even for short term use	Immediate remediation required	Purple (Unacceptable even for short term use).

## 5. DISCUSSION OF RESULTS

### 5.1 Annual Doses within the Mooi River Catchment for the Drinking Water Exposure Route

The annual doses for the drinking water route of consumption, for the Mooi River Catchment, are shown in Table 4 and Figure 3.

**Table 4: Final annual total dose [a] in mSv/a for the drinking water route, in the Mooi River catchment, arranged according to ascending dose. Also given is the incremental dose [b] above the estimated background of 0,02 mSv/a.**

Site No	Dose[a]	Dose[b]		Site No	Dose[a]	Dose[b]
29	0,02	0,00		16	0,04	0,02
30	0,02	0,00		13	0,04	0,02
14	0,02	0,00		3	0,05	0,03
35	0,02	0,00		10	0,06	0,04
27	0,02	0,00		4	0,06	0,04
34	0,02	0,00		2	0,06	0,04
6	0,02	0,00		39	0,06	0,04
31	0,03	0,01		5	0,06	0,04
26	0,03	0,01		23	0,08	0,06
20	0,03	0,01		37	0,08	0,06
25	0,03	0,01		8	0,08	0,06
28	0,03	0,01		17	0,08	0,06
32	0,03	0,01		9	0,11	0,09
19	0,03	0,01		11	0,14	0,12
33	0,03	0,01		7	0,16	0,14
22	0,03	0,01		15	0,18	0,16
18	0,03	0,01		1	0,24	0,22
24	0,03	0,01		7a	0,27	0,25
21	0,03	0,01		12	0,52	0,50
6a	0,03	0,01				
36	0,03	0,01				
38	0,03	0,01				

Applying the proposed interim water quality guidelines to the mean annual doses calculated for the radionuclides, an annual dose map for drinking water was produced. The dose map (Figure 3) shows that the radiological quality of the water, at the majority of the sampling sites in the Mooi River Catchment, is either in the ideal (blue,  $\leq 0,1$  mSv/year) or acceptable for lifetime use (green;  $>0,1$  to  $\leq 0,25$  mSv/year) class.

Two sites were in the yellow class ( $>0,25$  to  $\leq 1,0$  mSv/year), implying suitability for interim use, including the need to establish the origin, and consumption rate of the water at the site. No sites were in the red class ( $>1,0$  mSv/year), implying that there were no sites which were unsuitable for use, and thus which needed active intervention.

In summary the following may be stated:

- All sites had an associated annual radiation dose less than 1 mSv/year, implying that at no site was the radiation dose at a level that would necessitate consideration of immediate intervention, such as the necessity of immediately providing an alternative water supply.
- Two sites had a radiation dose level in the yellow class of  $>0,25$  to  $\leq 1$  mSv/year. These were:
- Site 7a (West Driefontein mine process water before settling dams). This implies that the water is radiologically suitable for drinking water use for an interim period, but that a site specific investigation should be done, including the collection of information on drinking water consumption.

- Site 12 (Doornfontein gold mine service water). It was determined that this site dried up, and that water was no longer being discharged. The radionuclide input to the surface water from this site ceased for the further duration of the 1997 monitoring survey.
- Five of the sites were in the green class (acceptable for lifetime use), with radiation dose levels between  $>0,1$  and  $\leq 0,25$  mSv/year.
- The large majority of the sites monitored (34 sites) had insignificant radiation dose levels, and complied fully with the World Health Organization's ideal screening guideline for radioactivity in drinking water of  $\leq 0,1$  mSv/year. With respect to those sites at which there was no radiation problem from a drinking water point of view, it was noteworthy that such sites included:
  - (i) The two raw water intakes for drinking water treatment to the town of Potchefstroom.
  - (ii) Most of the groundwater sites, including the Gerhardminnebron, and the Turffontein eye.
  - (iii) The drinking water supply borehole of Welverdiend in the municipality of Carletonville.
  - (iv) All but two of the mine water discharge points.

## 5.2 Discussion of Predominance of Uranium

The results of the monitoring in the Mooi River catchment have shown that of the radionuclides measured, the parent radioactive element uranium, is responsible for the major portion of the measured alpha activity.

A map representing the measured uranium-238 chemical toxicity values is given in Figure 4, with the proposed colour classes. It can be seen immediately from this map that at the lower end of the catchment the sites are all in the ideal (blue) class, and that specifically the water of Potchefstroom is in the ideal class. The great majority of the sampling sites in the catchment were acceptable as far as uranium is concerned, with only 7 sites requiring further investigation from the viewpoint of uranium chemical toxicity (6 in the yellow class and 1 in the red class). The six sites in the yellow class for uranium chemical toxicity were:

- Site 1: Luipardsvlei.
- Site 7a: West Driefontein process water.
- Site 7: West Drienfontein transfer water.
- Site 11: Doornfontein Gold plant discharge in canal, upstream Doornfontein
- Site 15: Western Deep levels farm bridge down stream, no 7 Shaft Slimes dam.
- Site 9: Mooirivierloop at Blaubank.

The single site in the red class for uranium chemical toxicity was site 12: Doornfontein Gold Mine no 3 shaft discharge.

It is noticeable from Figure 4, that the majority of sites of elevated uranium concentration occur around the centre of the Mooi River catchment, with the concentrations again decreasing as the river flows further west on course to Boskop Dam. It is debatable what the reasons are for the decrease in uranium concentration after the initial increase around the middle section of the Mooi River. It is noticeable that the sites with elevated uranium concentrations almost all have contributions from mine water. Important attenuating mechanisms downstream of the points of contamination are probably a combination of sediment adsorption and dilution effects.

The majority of the sites not complying with the chemical drinking water criterion for uranium are associated directly with discharges from gold mining activities.

### 5.3 Annual Radiation Dose from Background Radiation Levels in Water

The radiation dose arising from the ingestion of the water at the various sampling locations is made up of two components, the dose attributable to background radioactivity in the water and the dose attributable to the additional radioactivity originating from mining activities in the region. As explained in section 2.5, it is not possible to establish unequivocally the background radioactivity levels in the water. However, for some sampling locations the radioactivity levels were very low, and the dose corresponding to these levels was about 0,02 mSv/year. For one of those sampling locations (C2H172Q01, site no. 34), there is no possibility of upstream mining influence. It can be assumed, therefore that a value of 0,02 mSv/year represents an upper bound value for the annual ingestion dose arising from background radioactivity in water.

World wide reference values for non-elevated levels of naturally occurring radionuclides in water [3] correspond to an annual ingestion dose of between 0,01 and 0,02 mSv/year.

It was therefore assumed for the purposes of this investigation that the annual radiation dose attributable to background radioactivity in water was 0,02 mSv/year. It will be seen from the results presented in Table 4 that this value is so small that the uncertainty in its estimation is not critical to the outcome of the investigation.

### 5.4 Relationship between Uranium Concentration and the Annual Dose

The IWQS (Appendix 8) and AEC (Appendix 9) methods of calculating mean annual dose at each site, while they differed in the assumptions used to deal with unmeasured nuclides, nevertheless gave very similar results, and essentially verified one another.

As shown in Appendices 8 and 9, an excellent linear correlation exists between the annual mean uranium concentration at a site and the annual radiation dose for the drinking water route at that same site. This correlation holds for the Mooi River catchment, but it should be noted that it may not hold equally well for other catchments due to possible differences in radiochemical water quality characteristics.

For all sites in the Mooi River catchment, the following correlation between uranium in  $\mu\text{g}/\ell$  and the total average annual lifetime dose in mSv/a was found:

$$D = 0,0012895 C_u + 0,02128 \quad (r^2 = 0,98)$$

Where D is the annual radiation dose from continuous drinking water use in mSv/year,

And  $C_u$  is the uranium concentration in the water in  $\mu\text{g}/\ell$ .

The implication of the existence of this correlation is that for further monitoring purposes in the Mooi River catchment, only the uranium concentration need be measured, from which the all nuclide dose can be accurately estimated. To illustrate the high degree of accuracy with which the total annual radiation dose from drinking water may be estimated from the uranium concentration alone, a comparison of the total dose calculation from the full nuclide analyses (dose[a]) as compared to the all nuclide dose as estimated from the uranium concentration alone (dose[c]) is shown in Table 5.

**Table 5: Annual Doses calculated for the Mooi River catchment sites for drinking water. Annual doses [a] in (mSv/a) for drinking water route, in the Mooi River catchment, for the 1997 sampling year arranged according to ascending dose. Also given is the comparative dose obtained from the mean U-238 concentration (mg/l) using the linear regression; dose [c] = 0,0012895 x U + 0,02128 found for the study.**

Site no.,	Dose[a]	Dose[c]		Site no.,	Dose[a]	Dose[c]
29	0,02	0,02		16	0,04	0,05
30	0,02	0,02		13	0,04	0,04
14	0,02	0,02		3	0,05	0,07
35	0,02	0,02		10	0,06	0,05
27	0,02	0,02		4	0,06	0,07
34	0,02	0,02		2	0,06	0,05
6	0,02	0,03		39	0,06	0,05
31	0,03	0,02		5	0,06	0,06
26	0,03	0,02		23	0,08	0,05
20	0,03	0,02		37	0,08	0,10
25	0,03	0,02		8	0,08	0,10
28	0,03	0,02		17	0,08	0,10
32	0,03	0,02		9	0,11	0,12
19	0,03	0,02		11	0,14	0,13
33	0,03	0,02		7	0,16	0,18
22	0,03	0,03		15	0,18	0,17
18	0,03	0,03		1	0,24	0,22
24	0,03	0,02		7a	0,27	0,30
21	0,03	0,03		12	0,52	0,50
6a	0,03	0,02				
36	0,03	0,03				
38	0,03	0,04				

### 5.5 Relationship between Gross Alpha Activity and the Annual Dose

Comparison of the measured gross alpha activity results with the alpha activity calculated from individual radionuclide measurements gave a reasonable linear correlation but with strongly scattered individual data, indicating that individual gross alpha activity measurements should only be used as a screening tool to identify whether the activity is high or low, and not as a decision tool for determining the acceptability of radiological water quality.

The annual radiation dose (calculated from individual radionuclide activities) was found to be linearly related to the annual average gross alpha activity in the following way:

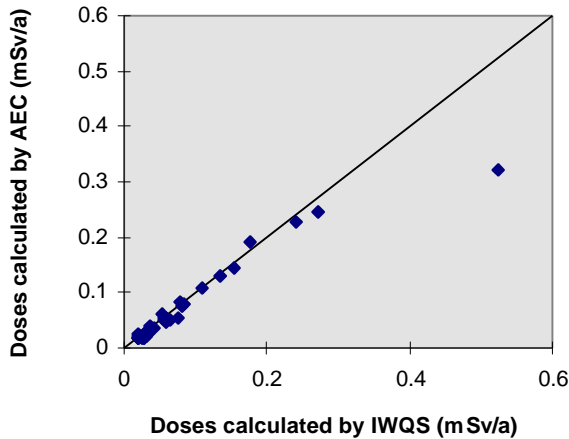
$$D = 0,02835 A_{\alpha} + 0,021 \quad (r^2 = 0,856)$$

where D is the lifetime average annual radiation dose from continuous drinking water use (mSv/a), and  $A_{\alpha}$  is the gross alpha activity (Bq/l).

### 5.6 Verification of Dose Calculations

To verify the doses calculated by the IWQS according to the methodology described in section 3.5 above, the AEC performed an independent dose calculation using, for the unmeasured radionuclides and unsampled sites, different assumptions from those used by the IWQS. Details of the AEC's assumptions and calculation methodology are given in Appendix 8. A comparison of the results from the two calculation methods is shown in Figure 6. It can be seen that, apart from one site (site 12), where the AEC calculation gave a significantly lower dose than the IWQS calculation, the results were in good agreement. The discrepancy with respect to site 12 can be explained by the fact that limited data were obtained from this site, because the flow ceased during the course of the study; in the AEC calculation, 6 months of data were represented by only one data point.

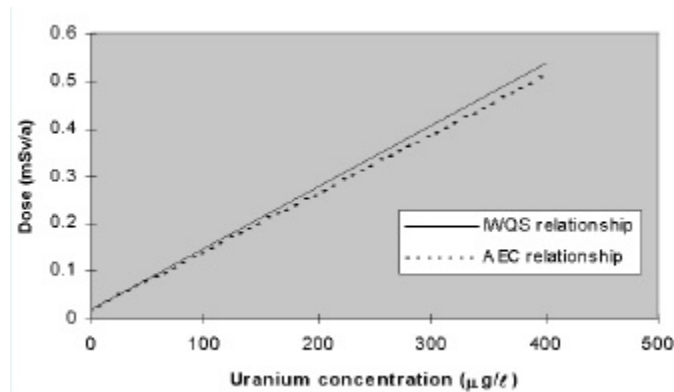
**Figure 6: Comparison of doses calculated by the IWQS (method 1) and the AEC (method 2), using different assumptions with respect to the unmeasured radionuclides and sites not sampled in the second phase**



The linear relationship between uranium concentration and annual dose, derived using the AEC's assumptions with respect to unmeasured radionuclides and unsampled sites, was:

$$D = 0,00124 C_U + 0,017 \quad (r^2 = 0,97)$$

This is very close to the relationship derived using the IWQS assumptions (see section 5.4), as can be seen from Figure 7.



**Figure 7: Comparison of IWQS and AEC relationships between uranium concentration and dose, using different assumptions with respect to the unmeasured radionuclides and sites not sampled in the second phase.**

## 5.7 Possible Uncertainties in Dose Calculations

Possible uncertainties in the estimation of the lifetime average annual dose are examined in Appendix 10. The results can be summarized as follows:

- Analytical uncertainties, based on a comparison between the radiochemical and ICP-MS techniques, are estimated to be about 1,5%.
- Uncertainties in projecting the present results to future years, assuming that the variations in radionuclide concentrations observed during the year of study are purely random, are estimated to be typically 20%. If there is a true seasonal component to the variations, then the uncertainty will be less than this value.



- Uncertainties in future dose estimations based only on uranium measurements, arising from the derived linear relationship between uranium and dose, are estimated to be less than 10%.
- Uncertainties due to monthly rather than weekly sampling, based on uranium data obtained in the first phase of the study, were estimated to range up to a factor of 3, but such estimates arise as a result of the short (six month) sampling period. The uncertainties would progressively decrease over longer sampling periods.
- By far the greatest uncertainty is that arising from the assumption of sole continuous use of the water for drinking purposes, which represents a 'worst case' scenario. For an individual falling within any given age group, the dose received will be directly proportional to the amount of water consumed while in that age group.

## 5.8 Suspended Solids

It was not the intention of this study to measure radioactivity in the solids suspended in the water. However, in order to obtain an indication of the possible contribution of suspended solids to the radiation dose from ingestion, concentrations of individual alpha-emitting radionuclides in the suspended solids were measured in samples obtained from the 15 sites sampled in the final month of the monitoring programme (December 1997).

In calculating the annual radiation doses from ingestion of the suspended solids, it was assumed that the uptake factors were the same as those for the dissolved constituents. The doses, expressed as percentages of the doses from filtered water, were found to be very low:

Average over 15 sites: 2,3% ± 2,1%

Median: 1,9%

Minimum: 0,1%

Maximum: 7,6%

Contributions to the dose from the suspended solids were found to originate mainly from the radionuclides thorium-230, polonium-210, actinium-227, protactinium-231 and thorium-232. This contrasts with the situation for filtered water, where the main contributors to dose were uranium-238, uranium-234 and radium-226.

## 5.9 Chemical Results: Sulphate

A summary of the sulphate concentrations found in the study is shown in Figure 5. The reason for collecting chemical data in this study, was to enable correlations with radiological data to be explored. An extensive search was made for meaningful correlations between the radiological variables and the total dose, but no statistically significant meaningful correlations were found.

The correlation between mean annual sulphate concentration and mean annual uranium concentration was investigated, but found to be poor ( $r^2 = 0,394$ ). This implies that sulphate levels can, therefore, not be used as a surrogate for indicating the possible presence of radioactivity. Sulphate in water is, however, important from the viewpoint of drinking water in that it gives rise to traveller's diarrhoea in individuals not used to drinking high levels of sulphate. Sulphate also accelerates corrosion in distribution systems and appliances.

## 6. The Gauteng Regional Office Water Quality Management Strategy for the Mooi River

### 6.1 Source Directed Controls

The Gauteng Regional Office has for some time identified the need to control, monitor and audit all point sources in the Mooi River catchment more effectively. The method used is to instruct all direct impactors to complete a strategic water management plan to ensure their effective management of the activities total water balance. The water quality management plans should include, amongst other aspects, the following:

- Water quality management measures in order to minimise pollution should be implemented at source. The fundamental principle is to prevent, inhibit, retard or stop the hydrological, chemical, microbiological, radioactive or thermodynamic processes, which result in the contamination of the water environment.
- If the water/waste water problems cannot be solved by the above water quality management measures at source, water/waste water reuse and minimisation measures should be implemented. This includes the prevention of the inflow of ground and surface water into the industry and mining related activities.
- If the water/waste water problems cannot be solved by reuse and minimisation measures, then water/waste water treatment applications should be implemented.

It should be appreciated that all of the above entails intensive negotiations between the relevant role players including catchment forums, consultants and specialists where necessary. This ensures participation, collaboration and transparency in decision making.

## **6.2 Water User Assessments**

To assess the extent of water use in the catchment, impactors were requested to initiate, in collaboration with the Region, water user assessments in the catchment. The results are captured in Appendix 1.

## **6.3 Actions taken at Sites 7a and 12**

The application of water quality management measures has resulted in specific actions being taken to address the sites showing elevated levels of radioactivity. These are as follows:

- West Driefontein Gold Mine (Site 7a)

The source of the water at Site 7a is excess mine water which is a mixture of recycled underground water, stormwater and final sewage effluent. The mine is currently investigating all options to control at source, reuse and minimise their excess water. This investigation includes the quantification of their total impact on the water environment.

- Doornfontein Gold Mine (Site 12)

The source of the water at Site 12 is excess mine water which is a mixture of recycled underground water and fissure water. The mine has investigated all options to control at source, reuse and minimise their excess water. This investigation included the quantification of their total impact on the water environment. The permit application was lodged at Department of Water Affairs and Forestry in December 1998.

Both these sites have been identified as currently not posing a threat for use as drinking water in the short term. Further investigation will be required to establish whether the water quality is radiologically acceptable in the long term.

## **6.4 Monitoring**

As part of their functions in the catchment the Gauteng Region undertakes river and audit monitoring at point sources. The analysis of chemical uranium at specifically identified sites was initiated during phase 1 of the study. Following phase 1 of the study the routine monitoring program has been extended to include the sites of elevated activity.

## 7. CONCLUSIONS AND RECOMMENDATIONS

### 7.1 General Conclusions

The aim of the radioactivity monitoring programme in the Mooi River was to address the risk of radioactivity in water to human health, and to determine the total dose from surface and some ground water sources that are, or could be, used potentially as drinking water supplies. After a year of data collection, the results showed that of the 41 monitoring sites covered by the study, 39 sites exhibited a water quality that is ideal or acceptable for continuous lifetime use in terms of proposed interim water quality guidelines for radioactivity in drinking water. At the two remaining sites, both of which are associated with the discharge of water from gold mining activities, the elevation of radionuclides is such that the water is still radiologically acceptable for use as drinking water in the short term, but further investigation would be required to establish whether the water quality is radiologically acceptable in the longer term. It should be noted that at no site was the radiological quality such that immediate remediation was called for.

### 7.2 Municipal Water Supplies

The only water in the Mooi River catchment used as a source of municipal drinking water is that supplied to Potchefstroom from the Mooi River and the ground water supplied to Welverdiend in the municipality of Carletonville. In both cases, there was no significant elevation of radioactivity above background levels, and the water falls into the 'ideal' classification in terms of proposed interim water quality guidelines for radioactivity in drinking water.

### 7.3 Indicators of Radiological Water Quality

The search for simple indicators of radiological quality revealed the following:

- (a) Some correlation exists between annual mean gross alpha activity and annual radiation dose from ingestion, but is not particularly strong.
- (b) The correlation between gross beta activity and radiation dose is poor.
- (c) Although waters with elevated radioactivity levels generally exhibit elevated sulphate concentrations, the converse is not always true. Consequently, sulphate concentration is not a reliable indicator of radiological water quality.
- (d) An excellent linear correlation exists between annual mean uranium concentration and the annual radiation dose from ingestion. The best-fit linear relationship for the data gathered in this study is given by:

$$D = 0,0012895 C_U + 0,02128 \quad (r^2 = 0,98)$$

where: D = average lifetime annual radiation dose from continuous drinking water use (mSv/a)

$C_U$  = uranium concentration in the water ( $\mu\text{g}/\ell$ )

The estimated uncertainty arising from the use of this relationship to predict annual radiation doses arising from the use of waters in the Mooi River catchment for continuous drinking purposes is less than 10%.

It remains to be seen at this stage whether different relationships will apply to different catchments or whether a single, more general relationship can be established that will be applicable across several catchments. The validity of the correlation between total dose and uranium concentration for the Mooi River catchment should be checked periodically where used for long term monitoring.

#### **7.4 Suspended Solids**

Preliminary indications were that the additional radiation dose resulting from the ingestion of the suspended solids in the water is minimal ( $\approx 2\%$  on average) and can therefore be ignored.

#### **7.5 Guideline Development**

Provisional guidelines for evaluating the significance of the drinking water pathway of radiation exposure were developed, based on a synthesis of the dose limits given by the World Health Organisation, the Council for Nuclear Safety, and the IAEA recommendations.

#### **7.6 Recommendations**

The following recommendations can be made from the results of the study:

(i) Action

No immediate action is required to reduce radioactivity levels in surface and ground waters in the Mooi River catchment.

(ii) Management approach

The future monitoring and control of radioactivity in surface and ground waters should be integrated into the existing approach used in the management of the catchment, in terms of which all pollutants of concern are addressed.

(iii) Application of relevant International Commission on Radiation Protection (ICRP) principles

The ongoing regulation, at source, of intended radioactive discharges from mining operations affecting water quality in the catchment should be conducted in accordance with relevant ICRP principles, which are as follows:

Firstly, radiation doses should be kept as low as reasonably achievable (ALARA), economic and social factors being taken into account. This should involve a periodic review of existing practices to see that they conform to the ALARA principle.

Secondly, the exposure of individuals should not exceed 1 mSv in a year, taking into account other radiation sources subject to control. To enable regulation to be applied at source, this will require that the optimization of protection according to the ALARA principle be constrained by source-related dose constraints of less than 1 mSv in a year. Discharges of radioactivity in water should accordingly be subject to appropriate limitation on a mine-specific basis.

(iv) Monitoring strategy

The experience and knowledge gained in this study should be used as input to the formulation of a national strategy and action plan for routine and follow-up monitoring of radioactivity in public water streams, as part of an integrated approach to water quality management. The principal approach to radioactivity monitoring should be as follows:

Where no data exists, then a full nuclide analysis is advisable. For the Mooi River catchment, monitoring of uranium on a monthly basis only may be used, with use of the relationship between uranium and running average annual dose as a monitoring and evaluation tool. Within this catchment, monitoring of chemical uranium concentrations (or surveillance in the absence of water flow) should continue on a monthly basis at those sites associated with radiation doses greater than 0,1 mSv/a (sites 1, 7, 7A, 9, 11, 12 and 15) and at those sites associated with municipal water abstraction (sites 27, 35 and 36), as part of the integrated catchment management

approach referred to in (ii) above. The average uranium concentrations over a year should be used to estimate the annual radiation dose at each site from sole continuous use of the water for drinking purposes, using the relationship derived in this study. Site 12 needs to be kept under surveillance if and when water flow recommences at this point.

(v) Chemical uranium as an indicator

The use of chemical uranium as a monitoring parameter for radioactivity should be investigated for other catchments to determine its applicability outside the Mooi River catchment.

(vi) Mine closure

The potential for ongoing radiological impacts, after mine closure, on water sources in the Mooi River catchment should be taken into account in the site-specific mine decommissioning plans that are required as part of the mines' Environmental Management Programme (EMP) obligations. Such plans address all potential sources of environmental pollution, such as acid mine drainage, in a holistic manner, so that interdependencies are taken into account. Radioactivity should be included in this holistic approach. Decommissioning plans will specify the nature and duration of any aftercare arrangements that might be required, and these will include appropriate ongoing monitoring requirements with respect to chemical and radiological pollutants.

(vii) Radioactivity in sediments

Since this study was concerned only with radioactivity in water sources, the question of radioactivity in the sediments in the Mooi River catchment remains largely unexplored. An investigation of radioactivity in sediments needs to be undertaken, with a view to understanding the role played by water chemistry, and it should be noted that this is indeed the subject of a project funded by the Water Research Commission starting in 1999 (Project No. K5/1095: Tier 1 Risk Assessment of Radionuclides in Selected Sediments of the Mooi River).

(viii) Radionuclides in fish

Because of the huge uncertainties in the uptake of radionuclides in fish, studies on the fish consumption exposure pathway should be conducted. It needs to be established whether potential radiation dose from this route in the first instance is likely to be significant or not.

(ix) Continuation of Technical Committee

The current Technical Committee should continue in order to ensure continuity of the monitoring efforts in catchments other than the Mooi River catchment as part of the national radiological monitoring programme.

## 8. REFERENCES

1. Institute for Water Quality Studies, Department of Water Affairs and Forestry, Radioactivity in Water Sources: A Preliminary Survey, Department of Water Affairs & Forestry, Project Report No P554001, April, 1995.
2. Institute for Water Quality Studies, Department of Water Affairs and Forestry, Overview of Radioactivity in Water Sources: Uranium, Radium and Thorium, Department of Water Affairs & Forestry, Report No N/0000/00/RPQ/0196, April 1996.
3. Sources and Effects of Ionizing Radiation, United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), Report to the General Assembly with Scientific Annexes, United Nations, New York, 1993.
4. Recommendations of the International Commission on Radiological Protection, Annals of the ICRP, 1990, Vol. 21, no. 1-3, ICRP Publication 60, Pergamon Press, Oxford, 1991.
5. International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources, Safety Series No. 115, International Atomic Energy Agency, Vienna, 1996.
6. Measurement and Application of Uranium Isotopes for Human and Environmental Monitoring, Goldstein, S J, Rodriguez, J M and Lujan, N, Health Phys. 72(1):10-18, 1997.
7. South African Water Quality Guidelines Department of Water Affairs and Forestry - Domestic Use, Department of Water Affairs and Forestry, Pretoria, 1996.
8. World Health Organization Guidelines for Drinking Water Quality, Geneva, 1993.
9. Department of Water Affairs and Forestry, Department of Health, Water Research Commission, Quality of Domestic Water Supplies, Volume 1: Assessment Guide, Water Research Commission number TT 101/98, Pretoria, 1998.
10. Coetzee, H. in Proceedings of the Symposium on the Application of Geophysics to Engineering and Environmental Problems, Orlando, Florida, 23-26 April 1995, p860-863.