THE ASSOCIATION OF GROUNDWATER CHEMISTRY AND GEOLOGY WITH ATYPICAL LYMPHOCYTES (AS A BIOLOGICAL INDICATOR) IN THE POFADDER AREA, NORTH WESTERN CAPE, SOUTH AFRICA

by

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Final report submitted to the Water Research Commission emanating from the Project : The correlation of high uranium, arsenic and other chemical element values in groundwater with abnormal haematological values related to the occurrence of leukaemia in the North Western Cape"

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ABSTRACT

A high prevalence of haematological abnormalities has been recorded in the Pofadder area of the Northern Cape Province. Hydrochemical analyses indicate that groundwater from certain aquifers in this area contain high concentrations of uranium, arsenic and fluoride, as well as elevated levels of radioactivity. Statistical analyses, using Geographic Information Systems (GIS), have shown that a correlation exists between the high concentrations of uranium and arsenic and the haematological abnormalities. As far as can be ascertained, this investigation represents the first documented evidence of a correlation of this nature. In terms of community health the recognition of the above is important, in that it enables the identification of potential areas where long-term residence and utilisation of groundwater could be regarded as undesirable, or alternatively, where the consumption of untreated groundwater should be avoided.

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

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- The Department of Community Health, Faculty of Medicine, University of Stellenbosch, for supplying the haematological data.
- Professor B. De Villiers, for his valuable advice concerning the interpretation of the medical data.

EXECUTIVE SUMMARY

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An executive summary of a report to the Water Research Commission on the Project

"The correlation of high uranium, arsenic and other chemical element values in groundwater with abnormal haematological values related to the occurrence of leukaemia in the North Western Cape"

WATER RESEARCH COMMISSION PROJECT NO: K5/839

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INTRODUCTION

In the early 1980's the Atomic Energy Corporation sampled groundwater from a number of boreholes in the Pofadder area as part of a geological programme. Relatively high uranium and arsenic values were noted, but as they were not considered to be of any economic significance, no further action was taken.

The Departments of Internal Medicine and Community Health of the Faculty of Medicine at the University of Stellenbosch have since noted that a number of cases of people suffering from haematological anomalies, related to leukaemia, have been reported from the area around Pofadder. These haematological values differ from the normal ranges to a significant extent.

Toens and Partners were approached by Dr B. de Villiers, Departmental Head: Community Health, Faculty of Medicine, University of Stellenbosch in October 1995 to assist his colleague, Dr J.G. Benade in his research pertaining to the above. Dr de Villiers has stated that: "In view of the high prevalence of haematological abnormalities in the study area, as well as the high uranium, chloride, nitrate, sulphate and fluoride levels in some borehole samples, we would like to discuss the possibility of doing a hydrochemical survey of those farms for which data is not available. It would be of tremendous medical value if a correlation could be shown to exist between the abnormal haematological values in the study population, and the water content, as information on this is very limited in the literature. Since a large number of people's health is at stake, it is paramount that this matter be further investigated".

The Community Water Supply and Sanitation programme, Department of Water Affairs and Forestry (DWAF), funded preliminary investigations during 1996. Funding to continue the project was made available by the Water Research Commission, as from January 1997 for a period 18 months.

OBJECTIVES

The main thrust of the investigation was to establish a statistically verifiable correlation between the various hydrogeological parameters and the high prevalence of haematological abnormalities of long-term residents in the Pofadder area.

A link of this nature would have far reaching implications, not only for the communities utilising such water, but also for regional and national authorities that are responsible for settlement planning and ensuring that new or existing settlements have an acceptable form of drinking water supply. In terms of community health, this type of investigation may make it possible to identify areas where the ingestion of certain types of water may be inadvisable in the long-term.

The rapid growth of the Geographic Information Systems (GIS) industry and related analytical and modelling techniques have made it possible to study the relationship of variables in new and more accurate ways, even in cases where these variables cannot be spatially referenced to each other. A further focus of the project is thus to develop a GIS based methodology which can be applied to investigations of a similar nature. This methodology would not only be limited to haematological and hydrogeological measurements, but could also be applied to further studies focussing on community health and environmental conditions.

The report is confined largely to the earth science aspects of the investigation. The significance of the haematological values and their relationship to the high incidence of leukaemia in the area fall beyond the scope of this report and should be dealt with by medical researchers.

GENERAL

The area under discussion is generally flat lying, with vegetation consisting of typical Karoo scrub and grass. With exception of higher lying areas, which reach altitudes of around 1100 m, the area is largely sand covered with an average altitude of between 900 and 1000 m. The annual rainfall is approximately 100 mm.

Geologically the area lies within the Namaqualand Metamorphic Complex. Towards the south these rocks are overlain by strata of the Karoo Super Group. The morphology over the entire area is extremely flat and has been described as conforming to a post-Karoo erosional surface, with gravel or float of the Dwyka Formation covering extensive areas in the south. Outcrop of the Namaqualand Metamorphic Complex is therefore obscured to a large extent, which is aggravated by the presence of a well-developed cover of calcrete. The calcrete is most common at the interface between the gneisses and the Dwyka Formation, as well as in topographic depressions such as drainage courses. Sand is widespread throughout and, in the northwest, permanent sand dunes have developed, typified by the red, well-rounded, Kalahari-type sand. The most frequently outcropping rocks are resistant quartzite and calc-silicate gneiss.

Over the last few decades, the area has been extensively explored for base metals and uranium. The most important mineral occurrence in the study area is located at Putsberg near the farm Houmoed, east of Pofadder. The main metal present at Putsberg is copper, with minor lead and zinc. Secondary uranium mineralisation in the form of carnotite, a uranium vanadium complex, is known to occur in the calcretes of the Koa River and other drainage systems. The younger, Karoo-aged Dwyka Formation is known to be radioactive in many areas.

The average concentration of arsenic in the earth's crust is 2 parts per million, in fresh water less than 1 μ g/l and in sea water approximately 4 μ g/l. Arsenic can be found as arsenate (e.g. Na₃AsO₄), with sulphides (e.g. arsenopyrite), associated with other metallic ores or occasionally in its native form. It is most often found as part of arsenic-bearing minerals, the most common being arsenopyrite (FeAsS sulphide and sulfosalt), which is associated with tin, tungsten, silver, copper and lead ores.

Practically the entire area underlain by the Namaqualand Metamorphic Complex yields groundwater with an electrical conductivity (EC) greater than 300 mS/m and a fluoride content greater than 2 mg/l. Aquifers in this area occur in fractured granitic gneiss rocks of the Namaqualand Metamorphic Complex, the Pofadder Lineament and related strata. The piezometric levels vary between 30 and 100 m below surface. The hydraulic gradient is generally flat (<1:200) and suggests a slow movement of groundwater towards the north. This is confirmed by the chemistry and age determinations of the water from the Vaalputs area, which vary between 920 and 14700 years Before Present. Aquifer recharge is very slow and localised as indicated by tritium analysis carried out on water samples from the Vaalputs area.

The high concentration of salts in the groundwater can be ascribed to the long residence time and the relatively high concentration of minerals in the host strata.

METHODOLOGY

Data Gathering

In the early 1980's the Atomic Energy Corporation (AEC) carried out a routine regional water-sampling programme over a large area of the Northern Cape. The results, which included comprehensive hydrochemical analyses, were made available by the AEC. From the range of chemical elements, only those that occur in excessive concentrations and are known to pose a health risk were considered for further analyses. The hydrogeological data was only available for the area south of 29°S and it was therefore necessary to collect water samples from farms in the Onseepkans area in order to consider the maximum area of overlap for the purposes of establishing a correlation.

The haematological data is the property of the Department of Community Health, Faculty of Medicine, University of Stellenbosch. The original population from whom blood samples were obtained included all persons aged 16 or older, residing in the area to the west of 20°E, bounded by the Kenhardt magisterial district. The data collected during 1993 represents a total population survey and consisted of blood samples taken from 630 individuals from 120 localities. For the purposes of this study the blood data of 418 individuals was considered. The remaining 212 individuals resided in areas where either (i) the population are not dependent upon groundwater for drinking purposes, or (ii) no accurate groundwater samples were available.

Atypical lymphocytes circulating in peripheral blood were identified morphologically and counted on blood films for each individual and recorded as one of the haematological variables. They were variable in appearance and similar to reactive lymphocyte transformation. For the purposes of this study they were used as a proxy to indicate a possible response of the haematological system to environmental factors.

It is important to emphasise that, in most cases, the exact locality of the haematological sample point is not known and that the centre of the relevant farm is usually taken as such. The ratio of the number of abnormal samples to the total number of samples at each locality was calculated. The population density at the sample localities varied between 10 and 100 persons, with Pofadder being the exception.

The nature of the data was not ideal, and it is important to take cognisance of the following constraints when evaluating the methodology used to process the data, as well as the results and conclusions:

- The water samples were relatively old and the analytical methods used did not pertain to drinking water specifically, i.e., the measurements for arsenic pertain to total As, and not just the component that is biologically available.
- The haematological data points were spatially recorded as the geographic centre-point of the relevant farm.
- The scope of the study did not provide for a control group.
- An anomaly can only be recognised in areas where the population has remained stable and stationary over a period of time. Although the rural population within the study area is relatively stable in terms of residence, internal migration does occur across farm boundaries.

Data Processing

The initial hydrogeological data was received in ASCII file format. The data was sorted according to the Local Origin (LO) co-ordinate system and converted to GIS coverages using the PC Arc/INFO GIS. The co-ordinate accuracy was checked by map number as well as using scanned, geo-referenced topocadastral maps as backdrops. The coverages obtained through this process were transferred to the ArcView 3.0a GIS for further processing and spatial analyses, including interpolation and contouring. The Inverse Distance Weighted (IDW) algorithm (within the Spatial Analyst extension) was used to create surfaces from the various chemical parameters. Each interpolated surface was then converted to a grid (200 x 200 m cell size). The haematological data was processed in a similar way and a 200 x 200 m grid was created. It is important to note that the cell size does not infer 200 x 200 m accuracy, but was merely intended to provide a unit that could be used in order to compare the surfaces to each other.

A series of maps were compiled illustrating the spatial distribution of the data. The preliminary results indicated a visual correlation between high uranium and arsenic and abnormal haematological values. It was necessary to establish whether this visual correlation was indeed statistically verifiable. As a result of the method that was used to record the spatial co-ordinates of the haematological data points, it was not possible to link a specific water drinking point to a specific haematological value, which necessitated an approach whereby spatially unrelated features could be correlated.

The approach decided upon entailed the procedure of correlating the interpolated grids with one another. This implies that every 200 x 200 m cell of, for instance, the uranium concentration grid, would be compared to every relevant (geographically identical) cell of the atypical lymphocyte grid. The Arc/INFO GIS 7.1.3 was used to perform this correlation. The relevant grids were grouped together in order to construct a "stack" (stacked grid layers). The statistical analyses were performed by using the STACKSTATS command.

The correlation coefficient was derived from the standard formula, which was applied to each of the roughly 500 000 cells in the grid being analysed. The following is a graphical illustration of the procedure employed:



Each grid cell of the first grid is compared to the geographically identical cell of the second grid. The statistical relationship is quantified by using the following formula:

 $\begin{aligned}
\rho_{XY} \text{ (correlation coefficient)} &= \frac{Cov(X,Y)}{\sigma_x \cdot \sigma_y} \\
\text{where } Cov(X,Y) &= \frac{1}{n} \sum_{i=1}^{n} (x_i \cdot \mu_x)(y_i \cdot \mu_y) \\
\sigma_x &= STANDARD \text{ DEVIATION of } X \\
\mu_x &= ARITHMETIC \text{ MEAN of } X
\end{aligned}$

RESULTS AND DISCUSSION

The hydrochemical analyses of groundwater indicated that uranium, arsenic and fluoride are present in excessive amounts that could pose potentially severe health risks. Electrical conductivity (EC) is included in the final results as a broad indicator of groundwater quality. Other elements such as copper, cobalt and cadmium and the anions nitrate and chloride were included in the original analyses, but do not occur in amounts that are known to pose a significant risk. The correlation matrix (Table 1) displays the results of the statistical analyses.

| TABLE 1: | STATISTICAL CORRELATION OF | CHEMICAL ELEMENT | VALUES WITH ATYPICAL |
|----------|----------------------------|------------------|----------------------|
| | LYMPHOCYTE COUNTS | | |

| | C | ORRELA | TION MAT | RIX | | 0 |
|-------------------------|----------|----------|----------------------|----------|----------|-------------------------|
| LAYER | ARSENIC | URANIUM | ARSENIC & URANIUM | FLUORIDE | | ATYPICAL LYMPHOCYTES |
| ARSENIC | 1.00000 | 0.30617 | 0.82138 | -0.16880 | -0.08021 | 0.40886 |
| URANIUM | 0.30617 | 1.00000 | 0.79412 | 0.20266 | -0.13309 | 0.57309 |
| ARSENIC & URANIUM | 0.82138 | 0.79412 | 1.00000 | 0.01367 | -0.13075 | 0.60387 |
| FLUORIDE | -0.16880 | 0.20266 | 0.01367 | 1.00000 | -0.13038 | -0.21590 |
| ELECTRICAL CONDUCTIVITY | -0.08021 | -0.13309 | -0.13075 | -0.13038 | 1.00000 | -0.22674 |
| ATYPICAL LYMPHOCYTES | 0.40886 | 0.57309 | 0.60387 | -0.21590 | -0.22674 | 1.00000 |

Electrical Conductivity

The upper limit for potability is regarded as 300 mS/m, while 210 mS/m is regarded as the upper acceptable limit for domestic water supplies. Groundwater with a conductivity of less than 300 mS/m only occurs in the area within a radius of \pm 30 km of Pofadder. The groundwater quality in the remainder of the study area varies between 300 and 700 mS/m, with values in excess of 1000 mS/m occurring in the north.

The generally high EC values within the study area are associated with the long residence time of groundwater, with the low annual precipitation (less than 100mm) resulting in very slow recharge and limited mixing of young and old water.

As to be expected of a broad indicator, there is no significant visual or statistical correlation between EC and the haematological values (Table 1).

Fluoride Concentrations

The maximum acceptable limit for fluoride is generally regarded as being 1,5 mg/l. Practically every water source within the study area exceeds this limit. Values in excess of 5 and as high as 10 mg/l are not uncommon. Long term exposure to concentrations above 8 mg/l is likely to cause crippling skeletal fluorosis and severe tooth damage.

As with EC, there is no meaningful visual or statistical correlation between fluoride

concentrations and haematological values (Table 1).

Arsenic Concentrations

The maximum permissible concentration for insignificant risk, and the maximum limit for low risk, are given by the Department of Water Affairs and Forestry (DWAF) as 100 μ g/l and 200 μ g/l respectively. Cancer or death could result from concentrations of more than 1000 μ g/l. Over large areas 500 μ g/l or even 1000 μ g/l is exceeded, posing a potentially severe health risk.

Arsenic accumulates in the body since it is excreted very slowly. Chronic arsenicosis is characterised by skin lesions, including hyperpigmentation, melanosis and/or keratosis, eventually progressing to gangrene or skin cancer.

The initial visual relationship between high arsenic concentrations and abnormal haematological values is supported by the statistical analyses, indicating a correlation coefficient of +0.41 (Table 1).

Uranium And Radioactivity

The tentative water quality guideline for drinking water in South Africa is currently 70 μ g/l or less. The United States Environmental Protection Agency (EPA) has proposed a Maximum Contaminant Level (MCL) of 20 μ g/l for uranium concentration in public water supplies. In accordance, 20 μ g/l has been adopted as a target water quality guideline for the purposes of this study. Values in excess of 70 μ g/l are regarded as posing a potential health risk if ingested over an extended period of time.

As was the case with arsenic, the visual correlation between high uranium concentrations and abnormal haematological values is supported by the statistical analyses, indicating a correlation coefficient of +0.57. This suggests that uranium is the single element that correlates the most significantly with haematological values (Table 1).

Combined Arsenic and Uranium Concentration

The above-mentioned correlation of arsenic and uranium with haematological values raises the question of what effect the combination of these substances might have. The arsenic and uranium grids were normalised and equally weighted before combining these layers to facilitate the construction of a new data set that could be correlated with the haematological data. A "percentage-of-total" grid was created and cross-correlated to the atypical lymphocyte grid.

From Table 1 it is clear that these two elements display a form of synergism in that the correlation of the combined data set with haematological values is higher than either of the elements individually. Statistical analyses produced a correlation coefficient of +0.60. This relationship is visually illustrated in Fig. 1, which was compiled by overlaying the geographical area where both uranium and arsenic pose a significant risk, with the greater area in which haematological abnormalities occur.



FIELD INVESTIGATIONS

Field investigations were carried out and a total of 126 boreholes were visited. At each borehole the general geology was noted and the levels of gamma-ray radioactivity of nearby outcrops measured. Ten groundwater samples were submitted to the AEC for analysis (Table 2). In particular, samples 1, 5, 6 and 8 were taken from farms with known cases of leukaemia.

| SAMPLE NUMBER | FARM NAME | ELECTRICAL CONDUCTI- VITY mS/m | GROSS a- ACTIVITY IN Bq/I | GROSS β- ACTIVITY IN Bq/I | URANIUM IN mg/l |
|------------------|---------------|--------------------------------------|---------------------------------|---------------------------------|-----------------|
| 835X001 | JORDAANSPOORT | 350 | 8.51 | 1.85 | 182 |
| 835X002 | NARUGAS | 265 | 1.43 | < 0.46 | 20.9 |
| 835X003 | KALKVLEI | 450 | 2.32 | 1.00 | 65.5 |
| 835X004 | OUBIP | 220 | 1.57 | <1.0 | 17.0 |
| 835X005 | LUBBESKOLK | 250 | 7.18 | 4.36 | 39.8* |
| 835X006 | SWANEPOELPUTS | 326 | 5.27 | < 0.88 | 221 |
| 835X007 | GRAPPIES | 615 | 8.16 | <1.2 | 105 |
| 835X008 | KEMPENSKRAAL | 332 | 3.86 | <1.5 | 72.3 |
| 835X009 | VALSVLEI | 395 | 16.9 | <1.7 | 89.0 |
| 835X010 | SPIEELPAN | 430 | 19.9 | <4.34 | 294 |

TABLE 2: RESULTS OF GROUNDWATER RADIOMETRIC ANALYSES

* newly drilled borehole

The results confirm that the uranium levels fall within the same range as the previous sampling programme (AEC data). The alpha activity was determined for the first time. Only two samples satisfy the accepted standard for low risk with respect to uranium concentration and gross alpha activity. The risk becomes significantly higher if the alphaor beta-activity is due to either radium (Ra-226 or Ra-228 respectively) or thorium-232.

The highest radioactivity was found to be associated with an orange weathered, pink, quartzo-feldspathic gneiss. The average radioactivity of these gneisses was 75 cps, almost three times the background level. In some cases the radioactivity was as high as 520 cps, almost eighteen times the background (min: 30; max 520).

CONCLUSIONS AND RECOMMENDATIONS

- The chemical analyses of groundwater samples taken from aquifers in the study area have shown that elevated levels of uranium, arsenic and fluoride exist. Geological fieldwork has indicated that higher levels of radioactivity (up to eighteen times the background radiation) are associated with the pink, quartzo-feldspathic gneisses of the Hoogoor Suite.
- Studies undertaken by the Departments of Community Health and Internal Medicine, Faculty of Medicine, University of Stellenbosch, have noted a number of cases of haematological abnormalities, related to leukaemia, from the area around Pofadder.
- A GIS-based methodology was used successfully to show that a positive correlation exists between the elevated levels of uranium and arsenic in groundwater and atypical lymphocytes (as a proxy of haematological abnormality), and hence the initial visual relationship was statistically verified. Arsenic (+0.41), uranium (+0.57) and the combination of uranium and arsenic (+0.60) returned positive correlation coefficients when cross-correlated with atypical lymphocyte counts. This methodology can be adapted and applied to other studies where it is necessary to quantify or correlate the effects of environmental conditions upon community health.
- From a community health aspect, the recognition of the above is important in that it makes it potentially possible to identify areas where long-term residence and utilisation of groundwater could be regarded as undesirable, or alternatively, where the consumption of untreated groundwater should be avoided.
- As far as can be established through the literature survey, this study represents the first documented evidence of such an association.
- The relevant authorities, planners etc. should be alerted to the potential health hazards related to long-term consumption of groundwater in the area. A programme of awareness and education should be implemented in order to inform the affected population of the risks and to advise with regard to mitigating measures and treatment options.
- The economic effects on the productivity of animals drinking water of poor quality, as well as the effect on human health resulting from the consumption of this meat and/or milk, should be researched.
- A number of known uranium deposits occur in the Northern Cape Province. These deposits are all associated with the Hoogoor Suite or similar granites and granitic gneisses. It is recommended that groundwater from these areas be analysed, particularly if this water is used for human consumption. A recent study for the Department of Water Affairs and Forestry (DWAF) has shown that elevated levels of uranium and/or radioactivity in groundwater used for domestic purposes occur within a wide area of Namaqualand.

- Groundwater with elevated uranium values is known to occur in other localities within South Africa. High concentrations occur specifically in the granitic basement rocks of the Northern Province and KwaZulu Natal and the uraniferous sediments of the Witwatersrand Basin, as well as in the Karoo sediments of the Cape Province and the Free State. It is recommended that similar studies be carried out in these areas, as yet unrecorded potential health hazards may be present.
- Groundwater is generally perceived as a safe source of domestic water supplies. Large-scale arsenic poisoning in India has highlighted the fact that millions of people in Developing Countries could be exposed to the effects of consuming potentially hazardous groundwater. It is of great importance that water sampling and analytical programmes be initiated in areas reliant on groundwater, specifically those areas where the geology suggests potential risks.

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| | South African Water Quality Guidelines. |
| | Volume 1. Domestic Use. |
| | DWAF. Sept. 1995 |

WATER RESEARCH COMMISSION PROJECT NO: K5/839

THE ASSOCIATION OF GROUNDWATER CHEMISTRY AND GEOLOGY WITH ATYPICAL LYMPHOCYTES (AS A BIOLOGICAL INDICATOR) IN THE POFADDER AREA, NORTH WESTERN CAPE, SOUTH AFRICA

1. INTRODUCTION

In the early 1980's the Atomic Energy Corporation sampled groundwater from a number of boreholes in the Pofadder area as part of a geological programme. Relatively high uranium and arsenic values were noted, but as they were not considered to be of any economic significance, no further action was taken.

The Departments of Internal Medicine and Community Health of the Faculty of Medicine at the University of Stellenbosch have since noted that a number of cases of people suffering from haematological anomalies, related to leukaemia, have been reported from the area around Pofadder. These haematological values differ from the normal ranges to a significant extent (B. De Villiers, *pers. com.*, 1995).

Toens and Partners were approached by Dr B. de Villiers, Departmental Head: Community Health, Faculty of Medicine, University of Stellenbosch in October 1995 to assist his colleague, Dr J.G. Benade in his research pertaining to the above. Dr de Villiers has stated that: "In view of the high prevalence of haematological abnormalities in the study area, as well as the high uranium, chloride, nitrate, sulphate and fluoride levels in some borehole samples, we would like to discuss the possibility of doing a hydrochemical survey of those farms for which data is not available. It would be of tremendous medical value if a correlation could be shown to exist between the abnormal haematological values in the study population, and the water content, as information on this is very limited in the literature. Since a large number of people's health is at stake, it is paramount that this matter be further investigated".

The Community Water Supply and Sanitation programme, Department of Water Affairs and Forestry (DWAF), funded preliminary investigations during 1996. Funding to continue the project was made available by the Water Research Commission, as from January 1997 for a period 18 months.

1.1 OBJECTIVES

The main thrust of the investigation is to establish a statistically verifiable correlation between the various hydrogeological parameters and the high prevalence of haematological abnormalities of long-term residents in the Pofadder area (Fig. 1).



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A link of this nature would have far reaching implications, not only for the communities utilising such water, but also for regional and national authorities that are responsible for settlement planning and ensuring that new or existing settlements have an acceptable form of drinking water supply. In terms of community health, this type of investigation may make it possible to identify areas where the ingestion of certain types of water may be inadvisable in the long-term.

The rapid growth of the Geographic Information Systems (GIS) industry and related analytical and modelling techniques has made it possible to study the relationship of variables in new and more accurate ways, even in cases where these variables cannot be spatially referenced to each other. A further focus of the project is thus to develop a GIS based methodology which can be applied to investigations of a similar nature. This methodology would not only be limited to haematological and hydrogeological measurements, but could also be applied to further studies focussing on community health and environmental conditions.

The report is confined largely to the earth science aspects of the investigation. The significance of the haematological values and their relationship to the high incidence of leukaemia in the area fall beyond the scope of this report and should be dealt with by medical researchers.

1.2 REPORT OUTLINE

The report can be outlined as follows:

Chapter One establishes the objectives of the study. Chapter Two deals with general information, including physiography, geology and hydrogeology. The literature survey is also discussed. The methodology employed to collate and analyse the data is explained in Chapter Three, while Chapter Four deals with the results of the analyses that were carried out, including a brief discussion of how the haematological data was incorporated. Chapter Five deals with the field verification of the relationship between the GIS-based analyses, geology, radioactivity and hydrogeology. The conclusions and recommendations are put forward in Chapter Six.

GENERAL

2.1 LITERATURE SURVEY

The literature survey comprised a review of material available in the WATERLIT database, scientific journals, the Internet and other publications. This survey yielded no results pertaining to the specific correlation of uranium and/or arsenic and haematological values. General studies investigating the chemistry of drinking water and the associated impact on human health are more common, although they deal mostly with drinking water and not groundwater specifically.

Studies focussing on the effects of arsenic in drinking water upon human health are the most prevalent. Jacobson (1998) summarises the disturbing occurrence of arsenic poisoning from groundwater in Bengal. He writes: "Arsenic poisoning, or arsenicosis, is emerging as the world's biggest environmental health disaster. As many as 200 000 people are estimated to have been poisoned in West Bengal (India) alone, and several thousand cases have emerged in Bangladesh."

Roberts (1996) surveyed the concentrations of naturally occurring uranium in groundwater in south-western North Dakota. Although the health impacts are only mentioned briefly, the study has great relevance as it deals specifically with the chemical toxicity of uranium.

In South Africa very few investigations of this nature have been undertaken. Hesseling, et. al. (1991) assessed the effect of elevated nitrate values in groundwater on infant health at Rietfontein in the Northern Cape Province. It is of interest to note that oesophageal cancer has been shown to be associated with the Beaufort Group of sediments in the Transkei (Marais and Drewes, 1962). At the time the reasons for this were unclear. The fact that uranium was subsequently found to occur in these sediments, however, might hold the key to this phenomenon. Hambleton-Jones (1982) investigated uranium occurrences in the surficial deposits of Southern Africa. Kempster et. al. (1996) published an overview of radioactivity in water sources.

The lack of relevant data and the need for future research is emphasised by the fact that the U.S. Environmental Protection Agency (U.S. EPA) has not established final Maximum Contaminant Levels (MCL) for uranium or arsenic, although both have been classified as "Group A: Human carcinogen" (Office of Water, 1996). This classification indicates that sufficient evidence is available from epidemiological studies to support a causal association between exposure and cancer.

With reference to arsenic in drinking water the U.S. EPA states the following: "1994: EPA thoroughly reviewed the available information and determined that (1) there is evidence of an association between internal cancer and arsenic, (2) the risk of internal cancer cannot be quantified using the available epidemiological data...". (http://www.epa.gov/ogwdw000/ars/ars1.html)

The U.S. EPA is developing a research plan to reduce the uncertainty in assessing health risks from low levels of arsenic and has proposed that final regulations be promulgated by 1 January 2001. The U.S. congress has authorized \$2.5 million per year from 1997-2000 for the necessary research to be completed.

2.2 PHYSIOGRAPHY

The area under discussion is generally flat lying, with vegetation consisting of typical Karoo scrub and grass (Fig. 7). With exception of higher lying areas, which reach altitudes of around 1100 m, the area is largely sand covered with an average altitude of between 900 and 1000 m. The annual rainfall is approximately 100 mm. The main topographical features are the hills to the north of Pofadder, a range of hills stretching from Aggeneys to Houmoed, as well as a few scattered inselbergs close to the Koa Valley. The main centres are Pofadder, which has a population of ±5000 inhabitants, the mining town of Aggeneys, and Pella. The remainder of the area consists of large farms used mainly for extensive sheep and goat farming. At the time of the May 1996 and February 1997 field visits, a number of farms were found be occupied only by herdsmen and their families.

2.3 GEOLOGY

The geology of the larger area has been described by numerous authors, the most recent being Maclaren (1988), Viljoen *et al.* (1986), Blignault *et al.* (1983), and Paizes (1975). A generalised, pre-Kalahari geological map of the Northern Cape Province, indicating uranium occurrences in the Pofadder area, is shown in Fig. 2. The regional geology of the area is shown in Fig. 8.

The area lies within the Namaqualand Metamorphic Complex. Towards the south these rocks are overlain by strata of the Karoo Super Group. The morphology over the entire area is extremely flat and has been described as conforming to a post-Karoo erosional surface, with gravel or float of the Dwyka Formation covering extensive areas in the south. Outcrop of the Namaqualand Metamorphic Complex is therefore obscured to a large extent, which is aggravated by the presence of a well-developed cover of calcrete. The calcrete is most common at the interface between the gneisses and the Dwyka Formation, as well as in topographic depressions such as drainage courses. Sand is widespread throughout and, in the northwest, permanent sand dunes have developed, typified by red, well-rounded, Kalahari-type sand. Outcrop is greatest in the northwest of the area along the Mattheusgat Mountains (that mark a well-exposed portion of the Pofadder Lineament). The most frequently outcropping rocks are resistant quartzite and calc-silicate gneiss.

Over the last few decades, the area has been extensively explored for base metals and uranium. The most important mineral occurrence in the study area is located at Putsberg near the farm Houmoed, east of Pofadder (Fig. 2). The main metal present at Putsberg is copper, with minor lead and zinc. Secondary uranium mineralisation in the form of carnotite, a uranium vanadium complex, is known to occur in the calcretes of the Koa



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Figure 7 : Typical of landscape and vegetation in the more rugged northern region of the study area. The orange weathering pink gneiss forms the lower slope of the large ridge in the right foreground. Photo: N J Wullschleger.



Figure 8 : Water quality in the Northern Cape. This example of a salt encrusted water pipe was taken at Noncaip (2919BA2/2). The water has an electrical conductivity of 874 mS/m and is solely for animal use. Photo: N J Wullschleger River and other drainage systems. The younger, Karoo-aged Dwyka Formation is known to be radioactive in many areas (Le Roux and Toens, 1982).

The average concentration of arsenic in the earth's crust is 2 parts per million, in fresh water less than 1 μ g/l and in sea water approximately 4 μ g/l (DWAF, 1996). Arsenic can be found as arsenate (e.g. Na₃AsO₄), with sulphides (e.g. arsenopyrite), associated with other metallic ores or occasionally in its native form. It is often found as part of arsenic-bearing minerals, the most common being arsenopyrite (FeAsS sulphide and sulfosalt), which is associated with tin, tungsten, silver, copper and lead ores (Klein and Hurlbut, 1985).

2.4 HYDROGEOLOGY

From the generalised geological map of the Northern Cape (Fig. 2) it is apparent that the granites, schists and gneisses of the Namaqualand Metamorphic Complex and the Pofadder Lineament constitute a window about which is draped a variety of sedimentary formations. From Figs. 3, 4 and 5 it is clear that practically the entire area underlain by the Namaqualand Metamorphic Complex yields groundwater with an electrical conductivity (EC) greater than 300 mS/m and a fluoride content greater than 2 mg/l. The lower sedimentary members of the Karoo Super Group usually yield water with an electrical conductivity (EC) in excess of 300 mS/m. (Toens *et al.*, 1996)

Aquifers in this area occur in fractured granitic gneiss rocks of the Namaqualand Metamorphic Complex, the Pofadder Lineament and related strata. The piezometric levels vary between 30 and 100 m below surface. The hydraulic gradient is generally flat (<1:200) and suggests a slow movement of groundwater towards the north. This is confirmed by the chemistry and age determinations of the water from the Vaalputs area, which vary between 920 and 14700 years Before Present. Aquifer recharge is very slow and localised as indicated by tritium analysis carried out on water samples from the Vaalputs area (Levin, 1986).

The high concentration of salts in the groundwater (Fig. 8) can be ascribed to the long residence time and the relatively high concentration of minerals in the host strata.

METHODOLOGY

The following is a brief overview of the data sources used and the analytical techniques employed during the study. Where applicable, a more detailed description is included in the relevant sub-sections of the report.

3.1 DATA GATHERING

In the early 1980's the Atomic Energy Corporation (AEC) carried out a routine regional water-sampling programme over a large area of the Northern Cape. The results, which included comprehensive hydrochemical analyses, were made available by the AEC. From the range of chemical elements, only those that occur in excessive concentrations and are known to pose a health risk were considered for further analyses. The hydrogeological data was only available for the area south of 29°S and it was therefore necessary to collect water samples from farms in the Onseepkans area in order to consider the maximum area of overlap for the purposes of establishing a correlation.

The haematological data is the property of the Departments of Community Health and Internal Medicine, Faculty of Medicine, University of Stellenbosch. The original data set consisted of haematological samples that were taken from 630 individuals from 120 localities in the vicinity of Pofadder during 1993. A subset of this data was considered for the purposes of this study. Atypical lymphocytes were used as a proxy to indicate a possible response of the haematological system to environmental factors.

The nature of the data was not ideal, and it is important to take cognisance of the following constraints when evaluating the methodology used to process the data, as well as the results and conclusions:

- The water samples were relatively old and the analytical methods used did not pertain to drinking water specifically, i.e., the measurements for arsenic pertain to total As, and not just to the component that is biologically available.
- The haematological data points were spatially recorded as the geographic centre-point of the relevant farm.
- The scope of the study did not provide for a control group.
- An anomaly can only be recognised in areas where the population has remained stable and stationary over a period of time. Although the rural population within the study area is relatively stable in terms of residence, internal migration does occur across farm boundaries.

3.2 DATA PROCESSING

The initial hydrogeological data was received in ASCII file format. The data was sorted according to the Local Origin (LO) co-ordinate system and converted to GIS coverages using the PC Arc/INFO GIS. The co-ordinate accuracy was checked by map number as well as using scanned, geo-referenced topocadastral maps as backdrops. The coverages obtained through this process were transferred to the ArcView 3.0 GIS for further processing and spatial analyses, including interpolation and contouring. The Inverse Distance Weighted (IDW) algorithm (within the Spatial Analyst extension) was used to create surfaces from the various chemical parameters. Each interpolated surface was then converted to a grid (200 x 200 m cell size). The haematological data was processed in a similar way and a 200 x 200 m grid was created. It is important to note that the cell size does not infer 200 x 200 m accuracy, but was merely intended to provide a unit that could be used in order to compare the surfaces to each other.

A series of maps were compiled illustrating the spatial distribution of the data. The preliminary results indicated a visual correlation between high uranium and arsenic and abnormal haematological values. It was necessary to establish whether this visual correlation was indeed statistically verifiable. As a result of the method that was used to record the spatial co-ordinates of the haematological data points, it was not possible to link a specific water drinking point to a specific haematological value, which necessitated an approach whereby spatially unrelated features could be correlated.

The approach decided upon entailed the procedure of correlating the interpolated grids with one another. This implies that every 200 x 200 m cell of, for instance, the uranium concentration grid, would be compared to every relevant (geographically identical) cell of the atypical lymphocyte grid.

The Arc/INFO GIS 7.1.3 was used to perform this correlation. The relevant grids were grouped together in order to construct a "stack" (stacked grid layers). The statistical analyses were carried out by using the STACKSTATS command.

The correlation coefficient was derived from the standard formula, which was applied to each of the roughly 500 000 cells in the grid being analysed. The following is a graphical illustration of the procedure employed:



Grid X- Atypical Lymphocytes

Each grid cell of the first grid is compared to the geographically identical cell of the second grid. The statistical relationship is quantified by applying the following formula:

Grid Y- Uranium Concentration

 $\rho_{XY} \text{ (correlation coefficient)} = \frac{Cov(X, Y)}{\sigma_X \cdot \sigma_Y}$ where $Cov(X, Y) = \frac{1}{n} \sum_{i=1}^{n} (x_i - \mu_x)(y_i - \mu_y)$ $\sigma_X = STANDARD \text{ DEVIATION of } X$ $\mu_X = ARITHMETIC \text{ MEAN of } X$

RESULTS AND DISCUSSION

The hydrochemical analyses of groundwater indicated that uranium, arsenic and fluoride are present in excessive amounts that could pose potentially severe health risks. Electrical conductivity (EC) is included in the final results as a broad indicator of groundwater quality. Other elements such as copper, cobalt and cadmium and the anions nitrate and chloride were included in the original analyses, but do not occur in amounts that are known to pose a significant risk. The correlation matrix (Table 1) displays the results of the statistical analyses.

| | C | ORRELA | TION MAT | RIX | | |
|-------------------------|----------|----------|----------------------|----------|----------------------------|-------------------------|
| LAYER | ARSENIC | URANIUM | ARSENIC & URANIUM | FLUORIDE | ELECTRICAL CONDUCTIVITY | ATYPICAL LYMPHOCYTES |
| ARSENIC | 1.00000 | 0.30617 | 0.82138 | -0.16880 | -0.08021 | 0.40886 |
| URANIUM | 0.30617 | 1.00000 | 0.79412 | 0.20266 | -0.13309 | 0.57309 |
| ARSENIC & URANIUM | 0.82138 | 0.79412 | 1.00000 | 0.01367 | -0.13075 | 0.60387 |
| FLUORIDE | -0.16880 | 0.20266 | 0.01367 | 1.00000 | -0.13038 | -0.21590 |
| ELECTRICAL CONDUCTIVITY | -0.08021 | -0.13309 | -0.13075 | -0.13038 | 1.00000 | -0.22674 |
| ATYPICAL LYMPHOCYTES | 0.40886 | 0.57309 | 0.60387 | -0.21590 | -0.22674 | 1.00000 |

TABLE 1: STATISTICAL CORRELATION OF CHEMICAL ELEMENT VALUES WITH ATYPICAL LYMPHOCYTE COUNTS

4.1 ELECTRICAL CONDUCTIVITY

The upper limit for potability is regarded as 300 mS/m, while 210 mS/m is regarded as the upper acceptable limit for domestic water supplies (Kempster and Smith, 1985).

From Fig. 9 it is apparent that groundwater with a conductivity of less than 300 mS/m only occurs in the area within a radius of ±30 km of Pofadder. The groundwater quality in the remainder of the study area varies between 300 and 700 mS/m, with values in excess of 1000 mS/m occurring in the north.

The generally high EC values within the study area are associated with the long residence

time of groundwater, with the low annual precipitation (less than 100mm) resulting in very slow recharge and limited mixing of young and old water.

As to be expected of a broad indicator, there is no significant visual or statistical correlation between EC and the haematological values (Table 1).

4.2 FLUORIDE CONCENTRATIONS

The maximum acceptable limit for fluoride is generally regarded as being 1,5 mg/l. From Fig. 10 it is clear that practically every water source exceeds this limit. Values in excess of 5 and as high as 10 mg/l are not uncommon.

Long term exposure to concentrations above 8 mg/l is likely to cause "crippling skeletal fluorosis" and "severe tooth damage" (DWAF, 1995).

As with EC, there is no meaningful visual or statistical correlation between fluoride concentrations and haematological values (Table 1).

4.3 ARSENIC CONCENTRATIONS

The maximum permissible concentration for insignificant risk, and the maximum limit for low risk, are given by the Department of Water Affairs and Forestry (DWAF) as 100 μ g/l and 200 μ g/l respectively. Cancer or death could result from concentrations of more than 1000 μ g/l (DWAF 1995). Fig. 11 indicates that over large areas 500 μ g/l or even 1000 μ g/l is exceeded, posing a potentially severe health risk.

Arsenic accumulates in the body since it is excreted very slowly. Chronic arsenicosis is characterised by skin lesions, including hyperpigmentation, melanosis and/or keratosis, eventually progressing to gangrene or skin cancer. In Chapter 2 it was mentioned that arsenic poisoning from groundwater sources is a growing environmental health problem, which has reached disastrous proportions in India. Other countries severely affected include Mexico, Chile, Argentina, Mongolia, Ghana and Taiwan. (DWAF, 1995; Jacobson, 1998)

The initial visual relationship between high arsenic concentrations (Fig. 11) and abnormal haematological values (Fig. 14) is supported by the statistical analyses, indicating a correlation coefficient of +0.41 (Table 1).

4.4 URANIUM AND RADIOACTIVITY

Uranium in groundwater poses a health risk in terms of radioactivity as well as chemical toxicity. DWAF initially regarded uranium content of 4 mg/l (4000 μ g/l) as the maximum limit for insignificant risk and 8 mg/l as the maximum limit for low risk (DWAF, 1993). In this publication, DWAF regarded uranium as a heavy metal and ignored the radioactivity.












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This discrepancy was later rectified in publications such as Bain *et. al.* (1994), Faanhof *et. al.* (1995), DWAF (1995) and Kempster *et. al.* (1996). The publication by Kempster *et al.* (1996) states that the tentative water quality guideline for drinking water in South Africa is now 70 μ g/l or less.

The United States EPA has proposed a maximum contaminant level (MCL) of 20 μ g/l for uranium concentration in public water supplies. In accordance, 20 μ g/l has been adopted as a target water quality guideline for the purposes of this study. Values in excess of 70 μ g/l are regarded as posing a potential health risk if ingested over an extended period of time.

It must be borne in mind that radioactivity in water is due to the presence of radioactive nuclides emitting α and β particles, or γ rays. The main contributing nuclides to radioactivity in water are naturally occurring uranium-238, uranium-234, thorium-232, radium-226, radium-228, radon-222, potassium-40 and occasionally lead-210, polonium-210 and thorium-230.

Depending on the species and concentrations of the radioactive nuclides in groundwater, a water sample with an α -activity count of for instance 5 Bq/l, may have a potential risk rated as insignificant, moderate or even high.

From Fig. 12 it will be noticed that the maximum acceptable limit is exceeded over most of the study area, with values in excess of 200 μ g/l often occurring.

As was the case with arsenic, the visual correlation between high uranium concentrations (Fig. 12) and abnormal haematological values (Fig. 14) is supported by the statistical analyses, indicating a correlation coefficient of +0.57. This suggests that uranium is the single element that correlates the most significantly with haematological values (Table 1).

4.5 COMBINED ARSENIC AND URANIUM CONCENTRATION

The above-mentioned correlation of arsenic and uranium with haematological values raises the question of what effect the combination of these substances might have. The arsenic and uranium grids were normalised and equally weighted before combining these layers to facilitate the construction of a new data set that could be correlated with the haematological data. A "percentage-of-total" grid was created and cross-correlated to the atypical lymphocyte grid.

From Table 1 it is clear that these two elements display a form of synergism in that the correlation of the combined data set with haematological values is higher than either of the elements individually. Statistical analyses produced a correlation coefficient of +0.60. This relationship is visually illustrated in Fig. 13, which was compiled by overlaying the geographical area where both uranium and arsenic pose a significant risk, with the greater area in which haematological abnormalities occur.

4.6 HAEMATOLOGICAL DATA

The haematological data was made available by the Departments of Community Health and Internal Medicine of the Faculty of Medicine at the University of Stellenbosch. The original population from whom blood samples were obtained included all persons aged 16 or older, residing in the area to the west of 20°E, bounded by the Kenhardt magisterial district. The data represents a total population survey and consisted of blood samples taken from 630 individuals from 120 localities. For the purposes of this study the blood data of 418 individuals was considered. The remaining 212 individuals resided in areas where either (i) the population are not dependent upon groundwater for drinking purposes, or (ii) no accurate groundwater samples were available.

Atypical lymphocytes circulating in peripheral blood were identified morphologically and counted on blood films for each individual and recorded as one of the haematological variables. They were variable in appearance and similar to reactive lymphocyte transformation. For the purposes of this study they were used as a proxy to indicate a possible response of the haematological system to environmental factors.(B. de Villiers, pers. comm., 1998)

It is important to emphasise that, in most cases, the exact locality of the haematological sample point is not known and that the centre of the farm is usually taken as such. The ratio of the number of abnormal samples to the total number of samples at each locality was calculated and the results were used to construct the interpolated grid displayed in Fig. 14. The population density at the sample localities varied between 10 and 100 persons, with Pofadder being the exception.

5. FIELD INVESTIGATIONS

Field investigations were carried out in order to

- measure and determine the levels of background radioactivity within the study area,
- assess whether the gamma-ray radioactivity of the specific rock types could be correlated with the higher uranium content and/or radioactivity of the groundwater, and
- confirm the nature and range of uranium and/or radioactivity in the groundwater as measured by the AEC.

The highly deformed metamorphic rocks of the Namaqualand Metamorphic Complex underlie most of the study area and are largely covered by varying thicknesses of calcrete, soil, aeolian sand and alluvium. The Namaqualand Metamorphic Complex consists of schists, quartzites, calc-silicate gneiss and quartz feldspathic gneiss of both sedimentary and igneous origin. Karoo-aged tillites and shales are exposed in the southeastern corner of the study area. A total of 126 boreholes were visited and at each borehole the general geology was noted and the levels of gamma-ray radioactivity of nearby outcrops measured. The position of each water source and gamma-ray measurement was recorded using a Global Positioning System (GPS). More than half of the boreholes sampled (56%) are being used to supply drinking water to people living in the area and 91% of the boreholes are for stock watering purposes.

The EC of the groundwater ranged from 54 mS/m to more than 2000 mS/m (2000 mS/m being the maximum possible range of the Conmet Conductivity Meter). The average EC was 479 mS/m. Of the boreholes used for human consumption, 56% (40/71) have EC's of more than 300 mS/m, which is, by definition, unsuitable for human consumption.

Ten groundwater samples were submitted to the AEC for analysis (Table 2). In particular, samples 1, 5, 6 and 8 were taken from farms with known cases of leukaemia.

| SAMPLE NUMBER | FARM NAME | ELECTRICAL CONDUCTI- VITY mS/m | GROSS α- ACTIVITY IN Bq/I | GROSS β- ACTIVITY IN Bq/I | URANIUM IN mg/ |
|------------------|---------------|--------------------------------------|---------------------------------|---------------------------------|----------------|
| 835X001 | JORDAANSPOORT | 350 | 8.51 | 1.85 | 182 |
| 835X002 | NARUGAS | 265 | 1.43 | < 0.46 | 20.9 |
| 835X003 | KALKVLEI | 450 | 2.32 | 1.00 | 65.5 |
| 835X004 | OUBIP | 220 | 1.57 | < 1.0 | 17.0 |
| 835X005 | LUBBESKOLK | 250 | 7.18 | 4.36 | 39.8* |
| 835X006 | SWANEPOELPUTS | 326 | 5.27 | <0.88 | 221 |
| 835X007 | GRAPPIES | 615 | 8.16 | <1.2 | 105 |
| 835X008 | KEMPENSKRAAL | 332 | 3.86 | <1.5 | 72.3 |
| 835X009 | VALSVLEI | 395 | 16.9 | <1.7 | 89.0 |
| 835X010 | SPIEELPAN | 430 | 19.9 | <4.34 | 294 |

TABLE 2: RESULTS OF GROUNDWATER RADIOMETRIC ANALYSES

* newly drilled borehole

The results confirm that the uranium levels fall within the same range as the previous sampling programme (AEC data). The alpha activity was determined for the first time. Only two samples satisfy the accepted standard for low risk with respect to uranium concentration and gross alpha activity. The risk becomes significantly higher if the alphaor beta-activity is due to either radium (Ra-226 or Ra-228 respectively) or thorium-232 (DWAF, 1995). The gamma-ray radiation of 73 rock outcrops was determined using the Scintrex GIS-3 spectrometer. The spectrometer was calibrated prior to every measurement using the TS-1 Calibration Source. Two different types of readings were performed:

- firstly only Broad Band radiation was measured to determine background radioactivity (usually for rocks with lower values), and
- secondly the Assaying Procedure, measuring Broad Band, K+U+Th, U+Th and Th, was applied to rocks displaying higher radioactivity.

The results show that there are three significant groups:

- A large variety of rocks (including quartzite, schist, leucocratic gneiss, calc-silicate gneiss, amphibolite gneiss, tillite, shale and calcrete) as well as soil and other superficial cover have low to intermediate radioactivity. This group forms the background radioactivity level of 29 counts per second (cps) (min : 5, max : 48).
- A second group consisting of vein quartz and pegmatites have intermediate radioactivity of 48 cps and a higher variance (min: 15, max : 110).
- The highest radioactivity is associated with an orange weathering, pink, quartzofeldspathic gneiss. The average radioactivity level measured is 75 cps, almost three times the background level. In some cases the radioactivity was as high as 520 cps, almost eighteen times the background level (min: 30; max 520).

In the field no distinct radioactive minerals could be identified in either the pegmatite or the pink gneiss. However, possible minerals responsible for the radioactivity, could include uraninite, thorianite, uranophane, torbernite, carnotite, gummite and uraniferous magnetite.

6. CONCLUSIONS

The chemical analyses of groundwater samples taken from aquifers in the study area have shown that elevated levels of uranium, arsenic and fluoride exist. This is probably due to factors such as the low rainfall and recharge, lack of drainage and the geochemistry of the rocks. Geological fieldwork has indicated that higher levels of radioactivity (up to eighteen times the background radiation) are associated with the pink, quartzo-feldspathic gneisses of the Hoogoor Suite.

Studies undertaken by the Departments of Community Health and Internal Medicine, Faculty of Medicine, University of Stellenbosch, have noted a number of cases of haematological abnormalities, related to leukaemia, from the area around Pofadder.

A GIS-based methodology was used successfully to show that a positive correlation exists between the elevated levels of uranium and arsenic in groundwater and atypical lymphocyte counts (as a proxy of haematological abnormality), and hence the initial visual





Figure 15 :

Exploitation of ground water at Mattheusgat close to outcrops of radioactive orange weathering gneiss. Photo: N J Wullschleger

Figure 16 :

Scintillometer reading of 130 counts per second (broad band) on an outcrop of radioactive orange weathering pink quartz feldspathic gneiss.

Photograph: N J Wullschleger

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relationship was statistically verified. Arsenic (+0.41), uranium (+0.57) and the combination of uranium and arsenic (+0.60) returned positive correlation coefficients when cross-correlated with atypical lymphocyte counts. As far as can be established through the literature survey, this study represents the first documented evidence of such an association.

When applying the GIS process to the total set of available hydrogeological data and considering the Water Quality Guideline limits, it is clear that potential health hazards exist to the west and southwest of the study area (Fig. 17). This methodology can be adapted and applied to other studies where it is necessary to quantify or correlate the effects of environmental conditions upon community health.

From a community health aspect, the recognition of the above is important in that it makes it potentially possible to identify areas where long-term residence and utilisation of groundwater could be regarded as undesirable, or alternatively, where the consumption of untreated groundwater should be avoided.

7. RECOMMENDATIONS

- The relevant authorities, planners etc. should be alerted to the potential health hazards related to long-term consumption of groundwater in the area. A programme of awareness and education should be implemented in order to inform the affected population of the risks and to advise with regard to mitigating measures and treatment options.
- The economic effects on the productivity of animals drinking water of poor quality, as well as the effect on human health resulting from the consumption of this meat and/or milk, should be researched.
- A number of known uranium deposits occur in the Northern Cape Province. These deposits are all associated with the Hoogoor Suite or similar granites and granitic gneisses. It is recommended that groundwater from these areas be analysed, particularly if this water is used for human consumption. A recent study for DWAF has shown that elevated levels of uranium and/or radioactivity in groundwater used for domestic purposes occurs within a wide area of Namaqualand and Bushmanland (Wullschleger et. al., 1998).
- Groundwater with elevated uranium values is known to occur in other localities within South Africa. High concentrations occur specifically in the granitic basement rocks of the Northern Province and KwaZulu Natal and the uraniferous sediments of the Witwatersrand Basin, as well as in the Karoo sediments of the Cape Province and the Free State. It is recommended that similar studies be carried out in these areas, as yet unrecorded potential health hazards may be present.

Groundwater is generally perceived as a safe source of domestic water supplies. Large-scale arsenic poisoning in India has highlighted the fact that millions of people in Developing Countries could be exposed to the effects of consuming potentially hazardous groundwater. It is of great importance that water sampling and analytical programmes be initiated in areas reliant on groundwater, specifically those areas where the geology suggests potential risks.

ANNEXURE 1

CHAPTERS ON RADIOACTIVITY AND ARSENIC TAKEN FROM SOUTH AFRICAN WATER QUALITY GUIDELINES. VOLUME 1. DOMESTIC USE.

DWAF SECOND EDITION (DRAFT, SEPT. 1995)

Arsenic

Background Information

| Introduction | Arsenic is a greyish, semi-metal and occurs in three oxidation states, namely, (0), (III) and (V). In solution arsenic can exist as arsenite, As(III); arsenate, As(V); and as various organic complexes. Inorganic arsenates form arsenate salts with cations of calcium or iron. Soluble arsenic compounds are readily taken up by living organisms and at elevated concentrations can exert toxic effects. Ingestion of arsenic in drinking water is most likely to lead to chronic effects, principally, different types of skin lesions. |
|------------------------|---|
| Occurrence | Arsenic is fairly widespread in the environment, the average concentration in the earths crust being about 2 mg/kg. It is found as arsenates, sulphides and in association with many other metallic ores, and occasionally in the elemental form. Typically, the concentration of arsenic in fresh water is less than 1 μ g/ ℓ and in sea water, 4 μ g/ ℓ . Elevated concentrations of arsenic occur where there is pollution from industrial sources, or where geological outcrops of arsenic minerals occur. For example, new borehole water supplies in areas where arsenic minerals are known to occur, should be tested for arsenic content. |
| | Arsenic is used in metallurgy, in the manufacture of glassware and ceramics, and as a pesticide and wood preservative. |
| | Once absorbed by living organisms, arsenic is slowly excreted, and hence accumulation easily occurs. |
| Interactions | To a large extent pH and redox potential determine the type of inorganic arsenic species present in the aquatic environment. Metabolically, arsenic interacts with many elements, among them selenium and iodine. |
| Measurement | The reference type method for the determination of arsenic is atomic absorption spectrometry. The various forms of arsenic are converted to $As(V)$ in an acid digestion and then reduced to $As(III)$, prior to the generation of arsine gas with borohydride. If other methods are used to determine arsenic, their characteristics relative to the above reference type method should be known. |
| Data Interpretation | Once absorbed arsenic is not readily lost and accumulates in the body and therefore a single, once off exposure to a high concentration thereof can have serious effects. The criteria given should be treated as a maximum values not to be exceeded. If mean values are used they must be 5 times less than the non-exceedance values. |
| Treatment Options | Arsenic is most effectively removed from water in its pentavalent form. Trivalent arsenic is first converted to the pentavalent form using an appropriate oxidising agent such as chlorine or potassium permanganate. Pentavalent arsenic is effectively removed from water using conventional coagulation and flocculation processes followed by settlement and filtration. Suitable coagulants include aluminium sulphate, ferric salts and lime. |

The process requires monitoring to ensure the arsenic is being effectively removed. The process will also generate a watery sludge rich in arsenic which may present disposal problems.

The Effects of Arsenic

| Norms | The norm used in the guideline for arsenic is human health. |
|------------|---|
| Effects | Arsenic is slowly excreted from the body, hence it can easily accumulate. Poisoning can be both chronic and acute. Chronic poisoning is characterised by skin lesions including hyperpigmentation and cancer, whilst acute poisoning may result in death from upper respiratory, pulmonary, gastro-intestinal and cardiovascular failure. Nerve damage, characterised initially by sensory loss in the peripheral nervous system, is a primary symptom of arsenic poisoning. Effects, with the exception of cancer, are often reversible if exposure is discontinued. |
| Mitigation | The chronic effects of ingesting arsenic excluding cancer can be reversed by discontinuing the exposure from contaminated water. Acute effects may be reversed by administering chelation therapy using 2,3-dimercaptopropanol (British Anti-Lewisite). If arsenic poisoning is suspected medical advice should be sought immediately. |

Criteria

Effects of Arsenic on Human Health.

| Arsenic Range $(\mu g/l)$ | Effects |
|---|---|
| Target Water Quality Range 0 - 10 | No health effects expected; ideal concentration range |
| 10 - 200 | Tolerable concentration, but low risk of skin cancer in highly sensitive individuals over long term. |
| 200 - 300 | Increasing possibility of mild skin lesions over long term. Slight possibility of induction of skin cancer over. |
| 300 - 600 | Possible adverse, chronic effects in sensitive individuals; brief exposure has no effect; skin lesions, including hyperpigmentation, will begin to appear on long-term exposure. |
| 600 - 1000 | Symptoms of chronic poisoning such as skin lesions, including hyperpigmentation, will appear on long term exposure. |
| 1000 - 10 000 | Cancer or death will result from chronic poisoning. |
| > 10 000 | Death will result from acute poisoning. |

Note: It is recommended that the concentration of arsenic in potable water should never exceed 200µg/f, but ideally should not exceed the TWQR.

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Radioactivity Tentative Guideline

Background Information

Introduction Radioactivity in water is due to the presence of radioactive nuclides emitting α and β particles, or γ rays. These arise from radioactive nuclides found naturally in the environment. The main contributing nuclides to radioactivity in water are uranium-238, uranium-234, thorium-232, radium-226, radium-228, radon-222, potassium-40 and occasionally lead-210, polonium-210 and thorium-230.

Occurrence Of the radioactive nuclides normally found in water, potassium-40 is by far the most common, as potassium-40 is found together with all other potassium deposits, is an essential intracellular mineral, and is found in all living organisms and in all water supplies. Potassium-40 is a β - emitter. β - activity measurements in water are thus of lesser consequence, unless the potassium-40 activity is first subtracted. In ground water supplies the only other β -emitters which are usually important are radium-228, from the thorium-232 decay series, and possibly lead-210, from the uranium-238 decay series.

 γ -activity is not normally considered of health significance in water, since γ -rays pass through the body with very little absorption.

From a health perspective, the risk of radioactivity in water is mainly centred around α -emitting nuclides. These are uranium-238, uranium-234, thorium-232, radium-226 radon-222 and possibly polonium-210. Internationally the radioactive elements that are normally monitored in water are mainly uranium, thorium, radium and radon. The occurrence of uranium, thorium, radium and radon is as follows:

Traces of uranium are found in all soils, typically in the concentration range of 0.7 -9 mg/kg (world mean value), although relatively higher concentrations may be found in South African soils. Of the naturally occurring uranium, 99.27% consists of the U²⁹⁸ nuclide, the remainder being, U²⁹³ and a trace of U²⁹⁴. From a water quality viewpoint the uranium nuclides U²⁹⁸ and U²⁹⁴ are of practical importance. Uranium-238 decays by α -emission with a half-life of 4.468 x 10° years, and further decay yields various daughter products, such as uranium-234, thorium-230, radium-226, radon-222, lead-210 and polonium-210. Examples of uraniferous minerals are uraninite and carnotite. The world mean concentration of uranium in fresh surface water is 0.4 $\mu g/\ell$, and in sea water 3.2 $\mu g/\ell$. Substantially higher concentrations of uranium concentrations of up to several hundred $\mu g/\ell$ may be found in ground waters in such areas.

The inert gas radon-222, has a half-life of 3.83 days and occurs as an emanation from rocks and soil containing uranium minerals. Radon is quite soluble in water and thus groundwaters very often contain substantial concentrations of radon-222 particularly in association with uranium bearing mineral deposits.

Thorium-232 has a half life of 1.41 x 10^{10} years and decays by α -emission. The world median concentration of thorium in soils is 9 mg/kg. Thorium is found in the minerals monazite, thorianite and thorite. The world mean concentration of thorium in water is 0.03 μ g/ ℓ .

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| | Examples of commercial uses of naturally radioactive elements include: the nuclear industry, in the case of U²⁰⁵. the glass industry (natural uranium) for the manufacture of yellow glasses and glazes. thorium, in the manufacture of incandescent mantles for portable gas lamps, and in special optical lenses. radium-226 in medicine, for the treatment of cancer. (Radium was formerly used in luminescent paints on watch dials). |
|------------------------|---|
| Interactions | The radio-active elements found in water differ considerably in their chemical interactions. Uranium is quite reactive, and can form a number of water soluble species and complexes, and thus tends to be quite mobile in the aquatic environment. Thorium on the other hand is generally far less soluble and less mobile. The mobility of radium, lead and polonium are limited by the insolubility of their sulphates. Radon, on the other hand, being chemically inert but quite soluble in water, is very mobile in the aquatic environment. |
| Measurement | The measurement of radioactive nuclides is by the detection of emissions from α and β particles, or γ rays. The criteria given for radioactivity are measured on the liquid portion of the sample. To achieve adequate detection limits, concentration procedures are needed in some cases. Measurements may be expressed either as disintegrations per second, i.e., Becquerels per litre of water, or as the equivalent mass of the emitting nuclide. In the case of uranium-238, 1 Bq/t of α radioactivity is equivalent to 80.9 μ g/t uranium. |
| Data Interpretation | As α - or β - activity in water is each caused by a number of different nuclides, measurement of the gross α - or β -activity will not indicate specific nuclide emissions. Therefore it is necessary to do a full radioanalytical investigation to determine which nuclides are responsible for the radioactivity present. |
| | In the interpretation of radioactivity levels in water it is necessary to have some information on the ambient levels of radioactivity normally found in a given area or water supply. |
| Treatment Options | Treatment options need to be tailored to the chemical nature of the major nuclide causing the radioactivity. In the case of radon, which is chemically inert, the radon may be removed by physical processes, such as aeration of the water. For the chemically reactive radio-nuclides of uranium, thorium and radium, removal processes may range from optimisation of conventional flocculation processes, through to processes such as concentrated lime treatment and ion-exchange or reverse osmosis where high removal efficiencies are required. The radioactivity concentrated in the sludges or waste streams may present disposal problems. Treatment options for lead-210 are the same as for natural lead, (see lead), while the treatment options for polonium are similar to those for group VI elements in the periodic table (see selenium). |

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Guidelines

The Effects of Radioactivity

Norms

The norms used in the guideline for radioactivity in water are based on long term health effects.

The limits set for radioactivity in water are based on the possible stochastic risk of cancer induction in the long term through exposure to radioactivity. The lifetime fatal cancer risk of exposure to radiation has been estimated as 5×10^{-2} per Sievert. The absorbed tissue dose in Sieverts may be calculated from the tissue dose to radioactivity conversion factors for each radio-nuclide, assuming a mean water intake of 2 ℓ /day. Normally a conservative approach is used which assumes that 100% of the ingested radio-nuclide is absorbed. In practice, this is usually not the case.

Applicable tissue doses to radioactivity conversion factors for water ingestion are given below:

| | 7.6 | x | 104 | mSv/Bq | for | 23*U. |
|---|-------|---|-----------|--------|-----|--------|
| ٠ | 8.3 : | x | 10^{-6} | mSv/Bq | for | 235U. |
| ٠ | 8.3 : | x | 10* | mSv/Bq | for | 234U. |
| | | | | mSv/Bq | | |
| ٠ | 2,2 : | х | 10^{-4} | mSv/Bq | for | =10Th. |
| | 1.4: | х | 104 | mSv/Bq | for | ="Th. |
| | | | | mSv/Bq | | |
| | | | | mSv/Bq | | |
| | | | | mSv/Bq | | |
| ٠ | 8.3 | x | 10-4 | mSv/Bq | for | 210Pb. |

Effects

In general the effects of exposure to elevated levels of radioactivity in water is an increase in the cancer risk. The organ at risk for cancer will depend on the site in the body where the given radionuclide tends to accumulate. This in turn, depends on the chemical nature of the radionuclide. In the case of radon and its short lived daughter nuclides, the primary target organ is the lungs, and the risk is presented via inhalation of air contaminated with radon. This may occur on showering with radon rich water in a poorly ventilated area. The radon in the air may arise from contaminated building materials, from the underlying ground, or be transported via the water supply used, particularly if this is ground water in an area rich in uraniferous deposits.

In the case of radium and thorium, the target organ is the bony skeleton, because of the insolubility of the phosphate salts. Uranium on the other hand tends to accumulate in the kidneys and the liver. In the case of uranium, chemical toxicity is of more concern than the radiological risk of cancer.

Most of the effects of natural radioactivity described in the literature are for the elements uranium, radium, radon and thorium. There is little known on the specific effects of lead-210 and polonium-210, other than it is presumed that the major effect is as a consequence of exposure to the mother nuclide radon-222.

Mitigation

The carcinogenic risks presented by radioactivity in water are of a stochastic nature and are not easily mitigated.

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Criteria

α-radioactivity:

The α -emitters usually encountered in water are radon-222, uranium-238, uranium-324, thorium-232 and radium-226. By far the most common which may be present in water are radon-222 uranium-238 and uranium-234, these nuclides usually accounting for the major portion of the gross α -activity in water. Occasionally situations may be found where thorium-232 and radium-226 are major contributors, as the insolubility of the sulphate and phosphate salts of radium and thorium in water normally limits their presence in solution in water. However, occasionally, they may be found in the insoluble suspended matter and clay fraction.

The health effects of gross α -activity are variable, because of the difference in the effects of the main α -emitters which may be present in water. Gross- α activity should be used for screening purposes only. The significance of the screening for gross α -activity is given in Table 1. The criteria for the specific α -emitting nuclides are given in Table 2 for uranium-238, in Table 3 for thorium-232, in Table 4 for radium-226, and in Table 5 for radon-222.

| Gross α -activity (Bq/ ℓ) | Interpretation Under the usual situation where uranium or radon are the main α -emitters then no health effects (Annual cancer risk less than 1 in 7 million). However, if radium-226 or thorium-232 are present as the major contributor to the gross α -activity, then there may be an increase in the cancer risk to around 1 in 100 000 per annum. | | |
|--|---|--|--|
| Target water quality range 0 - 0.5 | | | |
| 0.5 - 11.0 | No health effects if radon gas is the main α - emitting species present. Sample should be analyzed for specific α - emitters. Moderate annual cancer risk of 1 in 200 000 if uranium-238 is the main α -emitter. High risk if thorium-232 or radium-226 are the main α -emitters. | | |
| 11.0 - 33.0 | If radon gas is the major α -emitter present, then no significant risk for drinking water. Sample should be analyzed for specific α -emitters. High risk to health if uranium, thorium or radium are present as major components of the α -emission. | | |
| >33.0 | Increasing likelihood of the presence of uranium, thorium or radium at levels which would damage heath. If radon is the main emitter present, however, then the risk is mainly due to inhalation of radon gas, with a subsequent increase in lung cancer risk, on showering. | | |

Table 1: Interpretation of gross α -activity measurements in domestic water.

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Guidelines

Table 2: Effects of uranium-238 on human health.

| Uranium | ²³⁸ U range | Effects | |
|---|--|--|--|
| Bq/ℓ | mg/ℓ | | |
| Target water quality range 0 - 0.89 | Target water quality range 0 - 0.070 | No significant effects. Annual cancer risk less than I in 4 million. | |
| 0.89 - 3.6 | 0.070 -0.284 | Annual cancer risk less than 1 in a million. 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 | |
| | | RATION IS EXCEEDED, HUMAN HEALTH CHEMICAL TOXICITY. | |
| 3.6 - 18 | 0.284 - 1.42 | Annual cancer risk less than 1 in 200 000, but significant risk of chemical toxicity with renal damage. | |
| >18 | >1.42 | Increasing cancer risk in long term. Increasing risk of renal damage in short term. | |

Table 3: Effects of thorium-232 on human health.

| Thorium -232: range (Bq/l) | Effects No significant effects. Annual cancer risk less than 1 in 100 000 | | |
|--|--|--|--|
| Target water quality range 0 - 0.228 | | | |
| HUMAN HEAL CONCENTRATION IS E | TH MAY BE AT RISK IF THE ABOVE XCEEDED | | |
| 0.228 - 2.28 | Annual cancer risk less than 1 in 10 000. | | |
| >2.28 | Annual cancer risk greater than 1 in 10 000. | | |

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Guidelines Radioactivity

Table 4: Effects of radium-226 on human health.

| Radium-226 (Bq/l) | Effects No significant effects. Annual cancer risk less than 1 in 200 000. The typical background radium concentration in water of 0.015 Bq/l has an associated cancer risk of 1 in 5 million per year. | | |
|--|--|--|--|
| Target water quality range 0 - 0.42 | | | |
| HUMAN HEALT CONCENTRATION IS EX | TH MAY BE AT RISK IF THE ABOVE CEEDED. | | |
| > 0.42 | Increasing risk of bone cancer in the long term. | | |

Table 5: Effects of radon-222 on human health.

| Radon-222 Range (Bq/ℓ) | Effects No significant effects either with drinking water, or on showering. | | |
|---|--|--|--|
| Target water quality range 0 - 11 | | | |
| 11-33 | No risk on drinking water, slight risk in showering in a non-ventilated area. | | |
| 33-100 | No risk on drinking water. Moderate risk in showering in a non-ventilated area. | | |
| >100 | Increasing risk on showering of inhalation of radon gas, leading to an increased risk of lung cancer. | | |

Guidelines

β radioactivity:

The most common β -emitter present in all water samples is potassium-40. This is associated with all living organisms. One gram of potassium contains an amount of potassium-40 with a β -activity of 27.6 Bq. For example, a water sample with 50mg/ ℓ potassium will have a β -activity of 1.38 Bq/ ℓ . There is consequently little sense in setting a criterion for gross β -activity. One β -emitter of potential concern that may be present is radium-228, for which the same criterion as for the α -emitter radium-226 may be set, because of the similarity of their tissue absorbed dose to Becquerel conversion factors.

The criterion for the β -emitter, radium-228 is given in Table 6. Note that the gross β -activity, which is usually largely due to potassium-40 is of little health significance and must be subtracted from any measured gross β -activity prior to interpretation.

Table 6: Domestic water criterion for radium-228 :

| Radium-228 Range (Bq/ℓ) | Effects |
|---|---|
| Target water quality range 0 - 0.42 | No significant effects. Annual cancer risk less than I in 200 000. |
| HUMAN HI CONCENTRATION | EALTH MAY BE AT RISK IF THE ABOVE IS EXCEEDED. |
| > 0.42 | Increasing risk of bone cancer in the long term. |

γ radioactivity:

 radioactivity in water is not normally of health concern, and no criterion is specified for this type of radioactivity.

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Guidelines

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