Potential environmental impact resulting from inadequate remediation of uranium mining in the Karoo Uranium Province, South Africa

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Abstract. Inadequate remediation of uranium mining in the Karoo Uranium Province, South Africa led to a disused open pit and inclined shaft, uranium ore in stockpiles and barrels, as well as other mining related equipment.

Land owners were unaware of the potential threat of uranium and coherent heavy metals resulting in livestock grazing amongst ore stockpiles and drinking from contaminated water supplies. Land owners consequently used stockpiled uranium ore for gravel road maintenance and construction of farmhouse foundations. The concentration of the radioactive gas radon (^{222}Rn) was monitored in the aforementioned farmhouse and reached 835 Bq.m^{-3}, thus exceeding the concentration limitations (150 Bq.m^{-3}) for radon gas in dwellings set by the United States Environmental Protection Agency.

Lycium cinereum, *Fingerhuthia africana*, *Aristida congesta congesta* and *Phragmites australis* growing within these mining locations revealed high concentrations of uranium and molybdenum in leaves and roots. *Lycium cinereum* were found to accumulate molybdenum up to 650 ppm in some leaves. Uranium readily accumulates in the roots of some of the species, whilst only a fraction is translocated to the leaves. Plants were also subjected to protein profile studies revealing a general tendency that with an increase in uranium and molybdenum concentrations, protein concentrations in the leaves tend to decrease. These fauna serve not

only as a toxicological hazard for grazing livestock, but also as potential phytoremediators of polluted soils.

Xenopus laevis were found to reside within a water filled open pit where uranium and molybdenum concentrations reach 20 mg/l and 4 mg/l respectively. These aquatic organisms contain high hepatic, renal and bone concentrations of uranium and molybdenum. Histological sections of liver and kidney revealed anomalous levels of lymphocytes, indicative of infection or neoplasia, possibly as a result of heavy metal uptake.

Introduction

Traces of uranium mineralisation occur throughout the Karoo Supergroup, South Africa (Fig. 1). During the late 1970's the spot uranium price reached its zenith ($40 to $44/pound U) and the largest deposits became economically extractable (Wilson and Anhaeusser 1998). This report briefly describes the uranium trial mining operations within the Karoo Uranium Province, namely Ryst Kuil (on the farm Ryst Kuil 351) and Rietkuil (on the farm Rietkuil 307) as well as coherent inadequate rehabilitation and potential environmental impact.

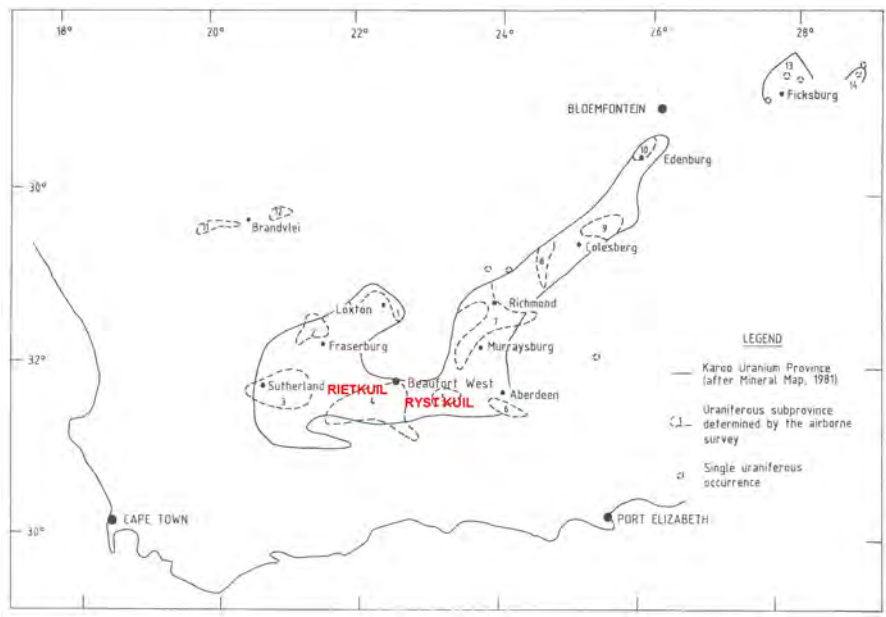

Fig. 1. Karoo Uranium Province showing mining locations (After Cole et al. 1991).

Uranium is harmful to living organisms if the metal or its decay products enter the body (Ragnarsdottir and Charlet 2000). Deliberate overdose of uranium in mammals and fish resulted in, amongst other symptoms, acute renal failure, liver disfunction and paralytic ileum (Cooley et al. 2000).

Radiation exposure in the natural and urban environment is focused upon radon, the only radioactive gas that is formed by the decay of uranium and thorium. The gas is ingested respiratoraly where it can cause damage as severe as lung cancer (United States Department of Health 1990). In the open air, the concentration of radon is generally very low. Radon becomes a hazard in unventilated uranium mines or in homes, built in areas with a high natural uranium occurrence in the soil or bedrock.

Molybdenum is an essential micronutrient for life, but can be toxic when received in high doses. High levels of molybdenum can account for molybdenosis in livestock, a secondary copper deficiency (Ammerman et al. 1980).

Materials and methods

Environmental assessment included sampling and analysis of stockpiled ore and ore containing barrels, soil, surface water, water logged sediment, foliage, aquatic organisms and radon gas monitoring.

Sampling

Ore stockpiles were randomly sampled by removing the top layer of rock and sampling qualitatively from the top to the bottom of the stockpile. Equipment used included a stainless steel scoop and plastic sample bags.

The following techniques were used in sampling, storage and treatment of soils and water logged sediment as described by Djingova and Kuleff (2000) and Boulding (1994):

- Stainless steel scoop and plastic sample bags used.
- Sample taken on 30 cm depth and cooled to 4°C.
- Samples dried in laboratory at 110°C after removal of stones, roots and living organisms.
- Screening of samples through a 2 mm stainless steel mesh.

Water sample preparation and storage, as described by Djingova and Kuleff (2000) and Greenberg et al. (1985), were as follows:

- Use of 500 ml plastic water containers.
- Cleansing of water containers with Hydrochloric Acid (30% HCl) before sampling.
- Rinsing with sample medium before sampling.
- Filtering through ALBET low ash filter paper.

- Lowering pH to 2 with Nitric Acid (55% HNO_3) after sampling and storage at 4°C.

Radon gas was monitored within a farmhouse using etched track radon gas monitors over a period of two months in November and December 2003 as well as May and June 2004. This technique allows for potential ventilation fluctuations between summer and winter months, which may affect radon gas concentrations.

Xenopus Laevis samples were collected using a baited trap set in the water filled open pit. Samples were transported alive to the laboratory where dissection and removal of tissue took place.

Analysis

Whole rock, soil and sediment geochemical analysis were processed on a PHILLIPS PW 1404 X-ray spectrometer. Major element analyses were executed on fused glass discs, using the technique of Norrish and Hutton (1969). Trace elements were analysed for on pressed powder briquettes.

Water samples were analysed by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES). This technique is used for qualitative and quantitative determination of metals in solution.

Fig. 2. Un-rehabilitated uranium ore in stockpiles and barrels on Ryst Kuil.

Uranium and molybdenum concentrations in the roots and shoots of plants were extracted by acid digestion (Lamas et al. 2002) and the concentration thereof determined by ICP-OES.

Tissue from Xenopus Laevis were analysed using Inductively Coupled Mass Spectrometry (ICP-MS) after acid digestion.

Results

Ryst Kuil

Numerous mining related structures and equipment are located within the mining area, including the easily accessible, sub-horizontal Cameron Shaft, ore stockpiles (Fig. 2) and barrels, ore crushing - and ventilation structures as well as collapsed core storage (Scholtz 2003).

The Ryst Kuil landowner's lack of knowledge regarding the potential toxicity of uranium and coherent metals (Table 1), led to the usage of water from the Cameron Shaft for crop irrigation. Stockpiled uranium ore was also used for gravel road maintenance and construction, including tennis court and concrete in housing foundations. Uranium ore used for the latter, led to possible accumulation of radon gas in the homestead.

The United States Environmental Protection Agency (EPA) has set an acceptable level of 150 $Bq.m^{-3}$ and 4 pCi/L for indoor radon in the domestic environment. Above these concentrations an action must be taken to reduce the radon levels (Table 2).

The uranium and molybdenum concentration in *L. cinereum* leaves from the mining area on Ryst Kuil were greater than the control leaf sample measurements (Fig. 3). The concentration of these metals in *L. cinereum* roots was not only greater than the control roots but also greater than the concentrations measured in the soil, indicating bioaccumulation and active uptake of uranium and molybdenum from the soil.

Uranium and molybdenum concentrations in leaves and roots from *A. congesta congesta* within the mining area are greater than those in the soil. These values were considerably higher than background leaf and root concentrations (Scholtz et al. 2005).

Table 1. Uranium and molybdenum in stockpiled ore and soil on Ryst Kuil. Normal concentratrions for soil from Alloway (1993).

		Stockpiled ore and barrels (ppm)	Soil (ppm)
Uranium	On site	4000	20
	Normal	N/A	1[a]
Molybdenum	On site	1500	3
	Normal	N/A	2[a]

N/A Not applicable
[a]Normal concentrations for metals in soils.

Table 2. Radon gas concentrations in farmhouse built with stockpiled uranium ore.

Monitor Location	NOV / DEC 2003			MAY / JUNE 2004		
	[Rn] (Bq m^{-3})	[Rn] (pCi/L)	Annual dose (mSv/a)	[Rn] (Bq m^{-3})	[Rn] (pCi/L)	Annual dose (mSv/a)
BEDROOM 1	835	22.57	14.19	797	21.00	13.55
BEDROOM 2	533	14.41	9.05	722	19.57	12.27
BEDROOM 3	502	13.57	8.54	610	16.53	10.37
KITCHEN	351	9.49	5.97	528	14.31	8.98
MOTOR GARAGE	N/A	N/A	N/A	108	2.93	1.84
STORE ROOM 1	N/A	N/A	N/A	207	5.61	3.52
STORE ROOM 2	N/A	N/A	N/A	269	7.29	4.57

Rietkuil

A test mining pit was excavated in 1977, covers an area of 5600m^2 and is approximately 15m deep (Scholtz 2003). A high concentration of uranium and molybdenum is present within the surface water and water logged sediment in the open pit (Table 3).

Soil uranium and molybdenum concentration from the mining area was less than concentrations in the leaves and roots of *L. cinereum* and *F. africana*, indicating possible bioaccumulation of these metals. The values for *L. cinereum* and *F. africana* roots and leaves, obtained from the mining area, were also greater than those from the control area (Fig. 4; Scholtz et al. 2005).

Phragmites australis, a reed growing in uranium-contaminated water (20 mg l^{-1}), accumulated uranium mostly in the roots (258 mg kg^{-1}) and to levels higher than its control counterparts (results not shown graphically).

Xenopus Laevis resides within the water filled open pit on Rietkuil, which contains high uranium and molybdenum concentrations. These organisms were analysed for uranium and molybdenum in various bodily tissues (Fig. 5) and revealed elevated values in comparison to control samples.

Discussion

A matter of concern is the fact that the farm owners were unaware of the possible toxic effects associated with uranium and molybdenum. The owner of Ryst Kuil consequently used the uranium ore for concrete in farmhouse foundations leading to accumulation of radon gas to above maximum allowed levels. The residents have been living in the aforementioned home without any knowledge of high levels of radon gas and its potential influence on health (Personal communication with Mr H.G. Scheün, owner of Ryst Kuil).

The water within the Rietkuil open pit yields molybdenum and uranium concentrations of up to 4 and 20 mg/l respectively. The concentrations of these metals

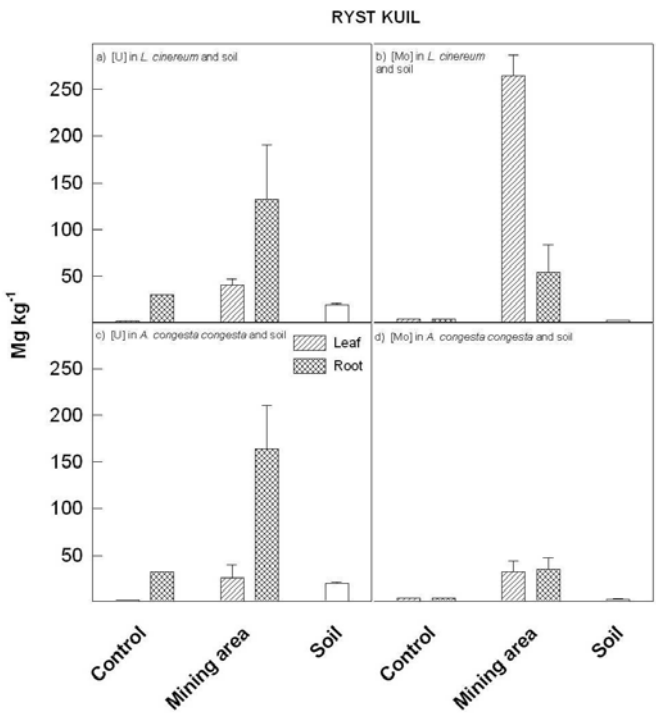

Fig. 3. Uranium and molybdenum concentrations in Ryst Kuil foliage and soil.

Table 3. Uranium and molybdenum in Rietkuil soil, sediment, water and stockpiled ore. Normal concentrations for soil from Alloway (1993).

		Stockpiled ore and barrels (ppm)	Soil (ppm)	Sediment (open pit) (ppm)	Surface water (open pit) (mg/L)
Uranium	On site	3500	15	700	20
	Normal / Max. allowed	N/A	1[a]	N/A	0.5[b]
Molybdenum	On site	1000	2	600	4
	Normal / Max. allowed	N/A	2[a]	N/A	0.2[b]

N/A Not applicable

[a]Normal concentrations for metals in soils

[b]Canadian Environmental Quality Guidelines (CEQG) for livestock watering. Maximum allowed concentrations (derived from CEQG) for aquatic ecosystems are 0.07 mg/L for molybdenum; no concentrations are available for uranium.

exceeds the maximum allowed levels suitable for aquatic life (Canadian Environmental Quality Guidelines 1999). *Xenopus Laevis*, an amphibian species, resides within this water, concentrating the metals into various bodily tissues. Although histological sections through *X. Laevis* organs revealed anomalous concentrations of lymphosytes, possibly indicative of neoplasia, no other deformities of any kind were visible, indicating a possible tolerance of the organism towards the specific metals and associated radiation.

On all four trial mining locations livestock are allowed to graze amongst the ore stockpiles and drink from the water filled open pit. Certain foliage types were also identified as uranium and molybdenum accumulators. The potential effects on livestock drinking from the Rietkuil open pit and grazing on metal accumulating foliage needs urgent investigation..

This research suggests that, *L. cinereum*, *F. africana*, *A. congesta congesta* and *Phragmites australis* can serve as bio-indicators for uranium and molybdenum pollution in contaminated soil and water. Differences between heavy metal concentrations in soil and plants collected within the mining area, indicate that the above mentioned plants bio-accumulate uranium and molybdenum. These plants also exhibit phytoextraction and rhizofiltration (*P. australis*) possibilities of these elements from contaminated soils and water. Harvested plants can be incinerated to reduce volume and the heavy metals either extracted from the ash or stored as hazardous waste.

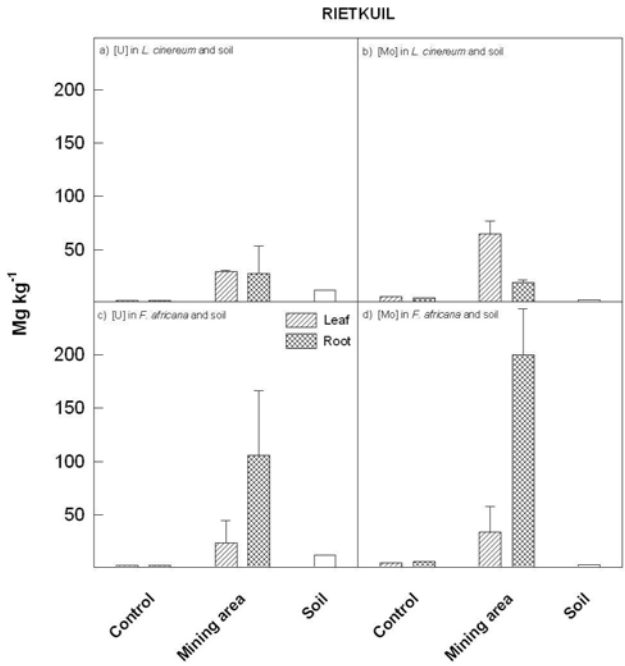

Fig. 4. Uranium and molybdenum concentrations in Rietkuil foliage and soil.

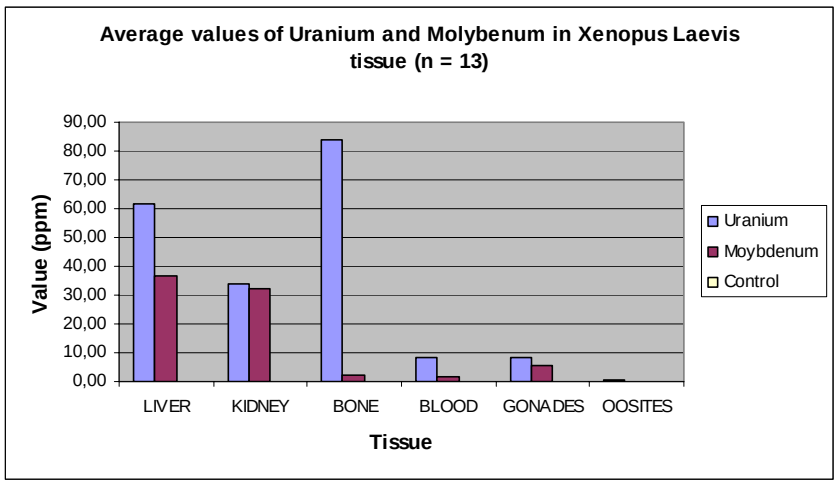

Fig. 5. Uranium and molybdenum concentrations in *Xenopus Laevis*.

Molybdenum transport mechanisms are evident in *L. cinereum*. The molybdenum concentrations are greater in the leaves than in the roots. The latter, together with *L. cinereum*'s high biomass productivity, makes it a strong candidate for the phytoextraction of molybdenum from contaminated soils.

Uranium accumulation in *L. cinereum* was generally higher in the roots compared to the leaves, but uranium still accumulates in reasonably high concentrations in the leaves. This suggests that fractions of the uranium in the roots are translocated to the above-soil biomass, making *L. cinereum* a possible phytoextractor of uranium as well.

Uranium and molybdenum accumulation in the grasses *F. africana* and *A. congesta congesta* appear to be greater in the roots than in the leaves. Comparing the uranium and molybdenum concentrations in the soil to those in the leaves indicate the translocation of uranium and molybdenum from roots to shoots. The low biomass yield of these grass species does not, however, allow them to be successful phytoremediators of heavy metal contaminated areas.

Conclusions

The failure to properly remediate the uranium trial mining areas in the Karoo, through removing ore stockpiles, waste rock and mining structures as well as preventing entry into open pits and shafts leads to the existence of a largely overlooked and potentially very serious environmental hazard.

The results obtained from the present investigation are of a disturbing nature, revealing previously underestimated and overlooked sources of localised pollution of hazardous substances in soil, surface water, foliage and aquatic organisms as well as anthropogenic sources of radiation.

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