Inaugural lecture

Uranium pollution of water

– a global perspective on the situation in South Africa

by

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1. Introduction

Of the 90 naturally occurring elements of the periodic table that make up the universe, uranium (U) is not only the heaviest one but also the one that most profoundly shaped the structure and face of planet earth. Containing the only natural isotope able to sustain a nuclear chain reaction, it was the radioactive decay of U that provided much of the energy used for melting rock into magma, creating and moving whole continents and causing devastating tsunamis and volcanic eruptions.

After U was discovered in 1789 by the German chemist Martin Heinrich Klaproth (born only a few kilometres away from where the author was born nearly 220 years later) it took one and a half centuries before mankind was able to tap into the unimaginable power of U following the groundbreaking research of Henri Becquerel, Marie Curie, Otto Hahn, Lise Meitner and others. Perhaps, not surprisingly, this power was first used by Man for killing hundreds of thousands of its own kind in Hiroshima and Nagasaki.

Ever since, uranium and radioactivity have evoked negative emotions in large sections of the general public, reinforced by nuclear accidents at Three Mile Island, Tchernobyl and recently, Fukushima. The use of depleted uranium for amour-penetrating ammunition in recent wars at the Gulf and Balkan regions – yet another destructive purpose – certainly did not improve the reputation of U, especially as its depleted, i.e. less radioactive, form was later linked to a new set of serious health problems collectively known as 'Gulf War Syndrome'.

The latter, however, has also provided a much needed boost for research into the health effects of uranium adding to a body of knowledge which now suggests that U displays a much wider spectrum of chemo- and radiotoxic properties than was previously thought to be the case.

In this lecture the risks associated with the exposure of humans to U-polluted drinking water is explored, using case studies from South Africa, assessed within a global context.

This includes an overview on general health risks associated with U and problems of setting appropriate limits, a brief history of research into U-pollution in South Africa, as well as an analysis of U-sources, pathways and human exposure in two case studies relating to mining-related and natural contamination respectively.

As an inaugural lecture marking the establishment of the first chair in Geography in the history of the Vaal Campus it also aims to illustrate the relevance of Geography for addressing current important environmental problems. Instead of adding to the confusing multitude of fragmented specialist studies Geographers, typically, adopt a generalist approach in order to sketch and understand the 'bigger picture'.

Environmental U-research covers an extremely wide spectrum of scientific disciplines ranging in scale from the sub-atomic and molecular level in biokinetics and geo-chemical issues, to more perceivable dimensions ranging from centi- to kilometres (e.g. from the sediment-water interface to whole catchments) as well as global processes such as U-exploration and trade. Generally, these studies are highly

specialised and whole scientific careers are often dedicated to understand tiny – albeit often crucial – pieces of the much bigger puzzle.

In this lecture a geo-ecological approach is applied that seeks to understand how U moves from natural and man-made sources of pollution through the aquatic environment and how it finally impacts on man.

Compared to the decades of state-sponsored, mostly confidential research into the uses of U for nuclear weapons and power generation from the early 1940s on to the end of the Cold War, investigations of environmental and health aspects are rather recent phenomena that started in earnest in the 1980s in the USA as part of the country's efforts to remediate the legacy of decades of uranium mining and milling activities (UMTRA[1]) (Waggit 1994).

The environmental research into U received a major boost in the 1990s with the reunification of Germany, as a condition of which the country assumed the responsibility to remediate the largest U-mining region in Europe – the formerly Soviet-controlled Wismut area that produced most of the U used in the atomic weapons programme of the Soviet Union. In an environmental rehabilitation programme of unprecedented scale in terms of resources and funding the German Government allocated over Deutschmark 15 bn (equivalent to € 7.5 bn and ZAR 88 bn in today's terms) to remediate Wismut legacy sites. This was scientifically complemented by a number of German universities and research institutions among them the oldest university on mining – the Bergakademie in Freiberg – which established an international conference series on '*Uranium Mining and Hydrogeology*' (UMH) that soon became the most renowned global forum on this topic.

It was also the Wismut environmental remediation programme that triggered the author's research interest in uranium 17 years ago. Selected results from research projects the author conducted ever since in mining areas of three continents will be presented in the lecture.

2. How dangerous is U to human health?

As a natural constituent of the earth whose radioactive energy ultimately drives the enormous forces that shape our planet through volcanic eruptions and plate tectonics, uranium – as the heaviest of all naturally occurring elements – is relatively common compared to other so-called 'heavy' metals (i.e. metals with a specific gravity of at least 6 g/cm^3). Displaying an average concentration of approximately 3 mg/kg in the continental crusts U is almost 1000 times more abundant than gold (Au), 40 times more than silver (Ag) and 10 times more common than the notoriously toxic metals cadmium (Cd) and mercury (Hg). While being a heavy metal with chemotoxic properties like Cd and Hg, uranium is also radioactive i.e. capable of spontaneously giving off energy and particles that can break chemical bonds and damage living cells. This property is termed 'radio-toxicity'. All three naturally occurring U-

[1] UMTRA – Uranium Mills and Tailings Remediation Act

isotopes (i.e. variants of the same chemical element which have the same number of protons but differ in the number of neutrons contained in the nucleus) emit relatively large alpha particles (consisting of two protons and two neutrons) which are considered to be 20 times more biologically damaging than the two other types of ionising radiation (beta and gamma radiation). However, the extreme low penetration depth of alpha radiation (it cannot pass through the outer skin of humans) renders alpha emitters (but not their progeny) harmless as long as they remain outside the body ('external source'). Once deposited inside the body, however, alpha emitters are potentially dangerous as adjacent tissue can be irreparably damaged by energy rich particles resulting in mutagenic defects and other malign transformations (ATSDR 2001, UBA 2008.).

Apart from the type of radiation, the potential for biological damage is also determined by the frequency at which emissions occur. The more decay events per time unit the higher is the 'activity' of an isotope and the shorter is its 'half life' (i.e. the time it takes for a radioactive substance until 50% of its mass disintegrated into daughter products). In case of U, however, the half life of its most abundant isotope (^{238}U) is extremely long, close to the age of the age of the earth resulting in relatively few decay events during an average human life span. The fact that the two other isotopes (^{234}U and ^{235}U[2]) are much more radioactive (especially ^{234}U accounting for nearly half of the total activity of U_{nat}) in terms of health risks is somewhat counterbalanced by the fact that they only comprise less than 1wt% of the total U_{nat} mass. For these reasons it is often assumed that the chemo-toxicity of U is of more concern that its radio-toxicity (ATSDR 2001, WISE 2001).

In line with the premise that U is mainly problematic as a heavy metal, the U-guideline limit for drinking water of the World Health Organisation (WHO) was based on the nephrotocicity of U as determined by exposing rats and rabbits to water spiked with uranyl nitrate during 28-91 days lab trials (Gilman et al. 1998 a/b). It was found that the limit for a tolerable daily intake (TDI) of U was at 6 µg/kg body weight (b.w.) and day. Divided by a safety factor of 10 the official TDI for U was finally set by the WHO at 0.6µg U/ kg b.w. x d. Assuming that 10% of the U is ingested by drinking water a guideline value of 2 µg/l U for drinking water was determined (WHO 1998). The same value is used by the German 'Mineral- and Tafelwasserverordnung' regulating the U-concentration in mineral waters used for the preparation of baby food (Konietzka et al. 2005,Von Soosten 2008, BfR and BfS 2006).

However, U-limits in major U-producing countries such as Canada, the USA and Australia were generally an order of magnitude higher ranging from 20 to 30 µg/l (WISE 2010, Von Soosten 2008). The highest value was set in SA at 70 µg/l equalling 3500% of the WHO value at the time (DWAF 1996 a) and at 10 µg/l for irrigation water (DWAF 1996 b). This represented already a significant reduction compared to the previous guideline which regarded 1000 µg/l U as safe (Class A-

[2] ^{235}U is the isotope used in nuclear reactors and weapons as it is the only natural isotope able to sustain a continued chain reaction of nuclear fission. In order to be usable in nuclear reactors it needs to be concentrated from 0.7 wt % in natural U to 3-5 wt% (low enriched uranium; LEU) and to >20 wt% for use in nuclear weapons (highly enriched uranium; HEU). At Oklo in Gabon geological conditions allowed for a natural enrichment of ^{235}U to about 3% in a natural nuclear reactor that formed some 1.7 billion years ago generating an average of 100 kW of power for a few hundred thousand years (Wikipedia, 19.01.2013 and 07.02.2013 http://teknologi2050.blogspot.com/2010/05/natural-nuclear-reactors.html).

water quality), 4000 µg/l as 'permissible' carrying 'insignificant risks' and 8000 µg/l as 'low' risk (DWAF 1993 based on recommendations of Kempster and Smith 1989).

Since 2011 the SABS adopted a value of 15 µg/l U for class I water (suitable for lifetime consumption) in line with the new value of the WHO making the analysis of U mandatory for water monitoring programmes in SA (SABS 2011) (Table 1).

Table 1: Recommended limits for the concentration of dissolved U in best quality drinking water according to different sources

Organisation/ Country	**Year**	**U concentration limit [µg/l]**
World Health Organisation (WHO) – drinking water	1998	2
	1/2003	9
	9/2004	15
Environmental Protection Agency of the USA (U.S. EPA) – drinking water	2006	30[a]
Health Canada – drinking water (Health Canada 2001)	2001	20
Germany; Federal Institute for Risk Evaluation - mineral water (*Mineral- und Tafelwasserverordnung*)	2008	2
Germany; Federal Environmental Bureau (UBA) - drinking water limit proposed by the to EU	2008	10
South Africa (Department for Water Affairs)	1993	
– Class A (no risk): domestic water		1000
– Class B (insignificant rik): domestic water		4000
TWQR[b]: – domestic water	1996	70
South Africa (South African Bureau of Standards) - domestic water	2011	15
Range		2 - 4000

[a] In the case of the US-EPA the originally determined limit of 20 µg/l was finally set at 30 µg/l to cater for cost considerations in water treatment (WISE 2008). [b] – Target Water Quality Range

Given that all guideline values ultimately aim to protect - universally identical - human health, it is difficult to scientifically explain the large differences in acceptable U-levels by relying on medical arguments.

Normally limits tend to get stricter over time as knowledge progresses, however, in the case of U the opposite happened. Within a span of les than a year the WHO twice raised the U-limit for drinking water from 2 µg/l to 9 µg/l (2003) and from 9 µg/l to 15 µg/l (2004) resulting in an overall increase by 750%. This was said to be done in response to findings indicating that the contribution of food and inhalation to the total intake of U in humans is much lower than the originally assumed 90%. Hence, it was argued, that a higher U-level in drinking water could be allowed as less U enters the body via other pathways. The increased value is now based on the assumption that drinking water accounts for 80% of the total daily U intake while 20% of the U enter the body via food ingestion and inhalation (UBA 2008, von Soosten 2008).

However, if the new guideline values are applied to children and babies with lower body weight than adults, the resulting U-intake associated with consuming water of the maximal allowable, U-levels exceeds the (unchanged) TDI value of the WHO by up to 250% (Winde 2011, Table 2).

Table 2: Daily intake of U through drinking water complying with the increased U-limits of the WHO for different (hypothetical) age groups in relation to the TDI for U set by the WHO (0.6 µg/kg bodyweight [b.w.] and day). Bold: Daily U intake via drinking water that exceed the TDI value) (WHO, 1998, 2003, 2005, 2006)

Year	U limit drinking water [µg/l]	Assumed proportion of U intake via drinking water [% of total U intake]	Resulting U intake [µg/kg b.w. per day] (% of WHO–TDI)*		
			Adult (60 kg; 2 l/d)	**Child** (20 kg; 1 l/d	**Baby** (5 kg /0.5 l/d - formula)
1998	2	10% (food/ inhal.: 90%)	0.07	0.01	0.02
2003	9	50% (food/ inhal.: 50%)	0.3	0.45	**0.9** (150% TDI)
2004	15	80% (food/ inhal.: 20%)	0.5	**0.75** (125% TDI)	**1.5** (250% TDI)

This is of particular concern as children are not simply small adults but show a higher uptake of U than adults adversely affecting their vulnerable developing organs. In view of this is it is not surprising that the original WHO limit of 2 µg/l is still applicable in Germany for all minerals waters advertising with the fact of being suitable for the preparation of baby food. This limit was thus also used as threshold value flagging potential problems in a German survey on U-levels in municipal tap waters (Foodwatch e.V. 2008).

Owing to such inconsistencies and the fact that an agreement exists between the WHO and the IAEA (as a lobby organisation for the peaceful use of nuclear energy) to adjust actions by mutual consent, doubts exists as to the true motivation behind the unusual increase of the U-limit. This especially, as the latter occurred when an increasing gap between U-supply and demand necessitated higher U-production needed for the continued expansion of nuclear power supply (Bertell 1999, WHO 1999). In order to stimulate U-production it is not inconceivable that less strict guidelines were seen as easing the commissioning on the needed new U-mines. The subsequent increase of U-production worldwide accompanied by renewed global exploration activities and the opening of new mines (especially in southern Africa) – termed 'U-renaissance' appears to confirm this assumption.

The profound changes in the global political power relations following the collapse of state socialism in Eastern Europe, perhaps somewhat unexpectedly, also boosted research in the toxicity of U. One reason was that large data sets on the health of Eastern European mine workers became available to researchers (e.g. Jacobi et al. 1997, Milacic et al. 2004, Tomášek and Malátová 2004, Rerecha et al. 2006). Among these records are those of 59 000 mine workers of the Wismut, providing the largest cohort of U-miners ever analysed (Jacobi et al. 1997).

Another reason was the access to a large body of mostly unpublished research on the transfer of radionuclides along the food chain conducted in the Soviet Union in the 1970s later used to review existing guidelines by the IAEA (e.g. Fesenko et al. 2007 a/b).

And lastly, the introduction of depleted uranium (DU) in armour-penetrating ammunition in ensuing regional wars in the Gulf and Balkan regions also indirectly sparked renewed research interest into the health effects of uranium as DU was subsequently linked to a new type of serious health problems known as 'Gulf War Syndrome'. In order to investigate the role of DU on human health and the environment research into depleted uranium internationally surged (e.g. McDiarmid et al. 2002, Milacic et al. 2004). Finally, and perhaps less related to political changes, the renaissance of nuclear power as a climate-neutral means to meet the growing energy demands of population "rich" countries such as China, Russia, India, Brazil, South Africa as well as the USA boosted research into environmental impacts associated with increasing mining and use of U. In France, where the majority of the energy comes from nuclear sources, an initiative by the Institute de Radioprotection et de Surete Nucleaire (IRSN) called 'Environhom', for the first time in radioprotection, specifically aimed at investigating the ecological effects of long-term low-dose exposure (IRSN 2005).

While it can certainly not be claimed that the research literature in the field of U-toxicity was exhaustively reviewed for this lecture, given the sheer volume, it is evident that a range of previously unknown toxic properties of U have subsequently been discovered which had not been considered when the current guidelines were determined.

Apart from kidneys it was found in tests on Gulf War veterans and laboratory animals that U attacks the brain (which may be as vulnerable as the kidneys), acts as endocrine disruptive compound by mimicking oestrogen (with possible consequences for the foetal development), compromises the immune system and damages the DNA (Vahrenholz et al, 1997, Zaire et al. 1997, McDiarmid et al. 2002, IRSN 2005, Raymond-Wish et al. 2007, Henner 2008). Epidemiological data from the Wismut cohort further suggest that existing models on effects of internally deposited alpha emitters underestimate the risk of contracting liver cancer by 20 to 70 times (Jacobi et al.1997).

Since most models currently used in radioprotection are derived from lifespan-studies of Hiroshima bomb survivors and nuclear accidents, which are high-dose short-term exposure events, little is known about effects of long-term low-dose exposure affecting many more people especially in mining areas. Thus far, it was believed that extrapolated data from extreme events such as a nuclear bomb explosion or nuclear accidents also cover less intense exposure in areas with naturally elevated radiation and U mining regions. The French Environhom initiative which specifically addressed this gap, demonstrated that this assumption is not correct and that the ingestion even of small amounts of radionuclides can lead to many deterministic adverse health effects (IRSN 2005).

In light of these new findings it is argued that current guidelines should be reviewed and revised in order to verify and justify their continued applicability. This is particularly the case for the repeatedly increased WHO limit for U in drinking water as this limit is globally used as a benchmark. In this process the ('tentative') DWAF guideline of 70 µg/l should also be revisited and adjusted as it is clearly at odds with

international limits as well as the revised SABS guideline (DWAF 1996 a, SABS 2011).

3. Investigating U-pollution of water: a geo-ecological approach

When John Snow, in 1849, finished a treatise titled *'On the mode of communication of cholera'* he was the first scientist to establish a link between water quality and human health and indeed to conduct what would later be referred to as an epidemiological study.

At a time, when everybody was convinced that epidemics such as cholera, plague and others are caused by the offensive smells riddling the overcrowded industrial towns (e.g. the very term 'Malaria' means 'bad air') his findings were, however, ignored and even ridiculed.

The *Lancet*, a renowned medical journal, suggested that Snow was in the pockets of industry that produces such smells and his suggested water pathway was just a "red herring"[3]. It was only after another devastating outbreak of cholera in Soho that killed 500 people in 10 days that Snow's suggestion to remove the handle from a public water pump in Broad Street was followed and cholera eventually subsided (Bryson 2004). The fate of Snow's work is one of the many examples for Schopenhauer's three stages of dealing with new insights:

at first they are ridiculed:
then fought against, and
eventually used by everybody as being self-evident.

The approach that allowed Snow to discover the link between polluted water and health problems was essentially geographic in nature and largely based on analysing spatial patterns. Investigating the geographic occurrence of cholera deaths which finally pointed to Broad Street as source of the problem, Snow's attention was drawn to two odd cases of deaths that occurred kilometres away from there. After hiking out to these sites and talking to relatives and neighbours (in today's terms conducting a reconnaissance study with semi-structured interviews – presumably without obtaining prior clearance from an ethical committee) he realised that in both cases water from Broad Street had been actually brought there – confirming results of his earlier spatial analysis (Bryson 2004).

As part of this lecture a very similar approach will be presented, used in the first and so far only large-scale epidemiological study in South Africa on health effects of uranium polluted drinking water.

Risk is commonly defined as the probability at which a hazard may translate into real damage. In the context of assessing health risks associated with U-pollution this, means quantifying the existing hazard (i.e. the health damage potential and extent of

[3] This reminds the author of a similar reaction by the Mail and Guardian on the 'Winde report' on the absence of flooding risks by underground gold mines in Johannesburg claiming that the report was a 'ruse' to downplay risks and polish the image of business in the city.

U-pollution) and the probability that humans will be adversely affected. Linking pollution sources to receptors by investigating the pathways along which a contaminant moves through the environment is at the core of geo-ecology also termed 'radio-ecology' when radioactive substances are concerned (Salbu 2009).

Using case studies from South Africa this lecture follows a typical geo-ecological approach considering the following three elements:

1. Characterising the pollution source (also termed 'source term' or 'hazard potential')
2. Understanding the movement of uranium from the source to the sink/receptor (also termed 'environmental transport', mobility' or fate')
3. Quantifying health impacts on exposed humans (also termed receptors)

The model underlying the radio-ecological approach is depicted in Fig. 1.

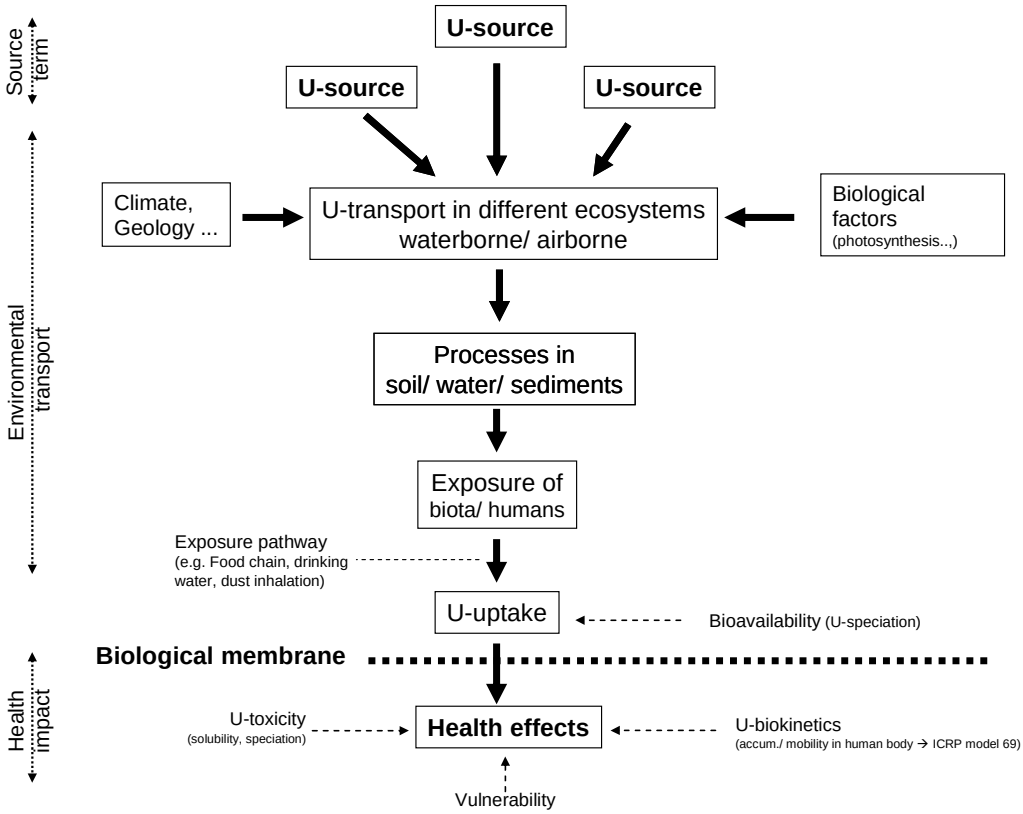

Fig. 1: Model of the radio-ecological approach to assessing risks associated with U-pollution (after an idea of Salbu 2009)

Source term characterisation

In order to quantify a contamination hazard, two major aspects are to be considered: the concentration of the pollutant and its quantity.

Concentration refers to the mass, or volume of contaminants relative to the mass/volume of the media in which they occur. Generally, associated risks increase the higher the concentrations since, for example, the ingestion of contaminated media

result in relative large quantities of the contaminant entering the body. Furthermore, large differences in concentration between the contaminated site and the surrounding environment create geochemical gradients promoting the dispersion of the contaminant.

Quantity of the contaminant refers to the total mass/volume of a contaminant in a certain area of concern. Considering this aspect is important as high contaminant concentrations in very minute quantities may not pose a problem, while less high concentrations occurring in very large quantities may.

In order to reduce environmental and health risks, maximum allowable concentrations are legislated for the various types of contaminants. If these limits are exceeded in certain material special precaution must be taken such as deposition in specially designed waste disposal sites.

Contaminant transport in the environment

This aspect is also termed 'environmental mobility', or 'environmental fate' and refers to the ability of a pollutant to move from a source through various environmental media such as water, soil or air towards a receptor or sink.

Since the concept of mobility is sometimes used ambiguously, a distinction between 'physical' and 'chemical mobility' is useful. While the former refers to the physical transport of U in whatever form - as particle, colloid, or dissolved - the concept of 'chemical mobility' commonly refers to differences between solid phases regarded as rather immobile [e.g. precipitates] and more mobile dissolved phases of a specific contaminant. Changes from dissolved to solid phases are thus referred to as 'immobilisation' with the reverse processes is termed 're-mobilisation'. If a contaminant is immobilised in a repository, for example as a solid phase contained in sediment, or soil, these repositories are termed 'sinks' – lowering the mobile amount of the contaminant in the environment.

Health impacts on exposed human

This aspect refers to the damage a contaminant causes that finally comes into contact with organisms such as humans ('receptors'). The degree to which organisms are affected depends on the actual health damage potential of the contaminant (*toxicity*), the extent of contact between the receptor and the contaminant (*exposure*), the ease at which the contaminant can enter the organism (*bio-availability*), and lastly, the *vulnerability* of the affected receptor.

Toxicity depends not only on the toxic properties of the actual element of concern but also on the various compounds, or species a contaminant may form that are able to alter its properties as well as its solubility changing its bio-kinetical behaviour and effects inside the organisms. Guidelines stipulating acceptable levels for certain contaminants are generally based on their toxicity. In many cases the toxicity was determined not by epidemiological studies on humans but by animal experiments using certain species such as small mammals like rodents, mice and rabbits and extrapolating the results. While such extrapolation of results from animals to humans is successful in many cases, medical history records cases where this failed and resulted in enormous human suffering (e.g. the Contergan scandal in Germany) .

The term ***exposure*** of receptors refers to the contact between contaminant and receptor while the various manners and media via which this occurs are referred to as '*exposure pathways*' and include, for example, the consumption/ingestion of contaminated water and food, the inhalation of dust etc. In many instances it is useful to distinguish between 'potential' and 'realistic' exposure pathways for quantifying associated risks. While 'potential' pathways refer to theoretical contact possibilities that cannot be excluded, 'realistic' pathways have a much higher *probability* to occur. In order to quantify the probability of realistic exposure statistical values, such as average water consumption etc., are often used in risk assessments.

Bio-availability generally increases with higher solubility of contaminants as dissolved phases are generally more readily absorbed than solid compounds. Solubility, in turn, is often governed by the type of chemical compound a contaminant forms ('speciation') as well as the physico-chemical conditions of the surrounding environment.

Vulnerability refers to the sensitivity of an organism to a particular toxin. While most guidelines aim to include differences in vulnerability of humans due to age, or gender when setting values for maximum allowable concentrations, all limits are generally designed for a healthy population (i.e. not considering the more vulnerable, or sick persons) and do not take individual hypersensitivity into account. Moreover, guidelines (for example for toxins in water) are commonly set based on statistically quantified behavioural pattern of the targeted society (such as the average water consumption) assuming a certain diet and 'normal' socio-cultural habits. Since such factors may vary significantly between societies of differing economic development levels (e.g. first vs. third world) and cultural identities (East vs. West vs. South) the frequently practised application of guideline values from other (mainly developed) countries in the West may not always be appropriate for the less developed or culturally different societies.

In the following section a brief overview on the history of research into U-pollution is provided before discussing the three main components of the radio-ecological approach this lecture follows, using examples from selected case studies in SA.

4. A brief history on research into U-pollution in SA

When, in 2007, the SA Government declared uranium a 'strategic mineral' and soon thereafter embarked on a nuclear expansion programme with intentions to become a global U-supplier, the country simply continued a tradition of several decades (Olivier 2007, Zhuwakinyu 2007). Initiated by the Manhattan Project to build the first atomic bomb, large-scale production of U as a much needed core component started in SA in 1951 after a government-sponsored large-scale U-programme provided the required infrastructure and resources (Taverner 1957). During the next decades of the ensuing Cold War, South Africa became the fourth largest U-producer world wide after the USA, Canada and the East Germany. However, dedicated U-mines remained the exception as most of the U in South Africa was produced as a by-product of deep level gold mines since the auriferous reefs often also contained significant U grades.

Falling U-prices in the 1980s, however, led to a significant drop in U-production and the radioactive heavy metal was again dumped along with tailings on the many slimes dams, as happened during the 65 years before the start of the South African U-programme. Since the discovery of gold in the Witwatersrand basin in 1886, gold mines produced well over 6 bn t of gold tailings covering some 400 km² (Robb and Robb 1998). With an estimated average of approximately 100 g U per ton these gold tailings contain higher U levels than many genuine U-tailings elsewhere and contain some 600,000 t U - about 3 times the amount exported during the Cold War. Given the huge amount of uraniferous tailings spread over large areas it is no surprise that many rivers and aquifers in mining regions are now affected by U pollution.

However, it was only recently that the mining industry at large acknowledged that there is, in fact, a problem and – at least partly – took on some responsibility. The fact that U-production in SA, since its inception, was surrounded by secrecy may have contributed to culture of denial well into the 1990s. This was despite the fact that suspicions on adverse effects of U-polluted mine water had been raised by farmers as early as 1967 (Stoch and Retief 1967) confirmed by even earlier internal investigations (Jordaan et al. 1960).

While the mining industry traditionally enjoyed a very close relationship with Government that often resulted in the rather soft policy on enforcing existing legislation, this changed, somewhat, after the democratic elections in 1994.

The first study on mining-related U-pollution publically available was a WRC project conducted by researchers of the Earth and Environmental Technology department of the Atomic Energy Corporation of SA Ltd. in cooperation with zoologists from the Rand Afrikaans University. In 1994, the researchers analysed several gold mining areas in the East, Central and West Rand for uranium and radium 226 pollution of water and soils as well as the associated migration of uranium along the food chain, sampling various vegetables, fish, cattle tissue, cow milk, cattle dung, aquatic vegetation and water birds. Apart from determining concentration ratios (also known as ‘transfer factors’) for the water-soil, fodder-cattle, fodder-milk and fodder-dung pathways the report also calculated the associated effective radiation dose for the various exposure pathways and found that none exceeded the 0.25 mSv/a limit applied for radiation from a single source (Bain et al. 1994).

Specifically addressing pollution by gold mining activities, two regional surveys of radioactive pollution of rivers in and around the Witwatersrand goldfields were conducted by the Department of Water Affairs and Forestry (DWAF) confirming that a number of rivers and streams were indeed significantly contaminated (Faanhof et al., 1995, Kempster et al. 1996).

Based on these surveys, in late 1996, the largest study on U-pollution to date was launched, focussing on the Wonderfonteinspruit catchment where the first complaints on adverse effects were reported in the 1960s (IWQS 1999).

Interestingly, at around the same time, a WRC study into health effects of naturally elevated U-levels was initiated in an arid sheep farming area of the Northern Cape (around the town of Pofadder) which finally established a geo-statistical link between U-levels in borehole water consumed by farmers and the occurrence of

haematological abnormalities serving as proxy for leukaemia from which an unnaturally high number of people suffered in the area (Toens et al. 1998).

Although some individuals served on the steering committees of both projects (i.e. the WFS and Pofadder study) there is no record of any cross-communication leave alone collaboration between the two research teams. This is puzzling as both studies investigated a nearly identical problem. Although the alarming findings of the Pofadder study were published a year before the IWQS report was released, the latter contains not a single reference to this highly relevant document. A contributing factor to this could have been that the title of the original Pofadder project was changed in such a way that crucial keywords such as 'uranium' and 'leukaemia' disappeared from the title of the final publication rendering any library search for these key topics (at the time) unsuccessful (Toens et al. 1998).

A letter of the principle researcher of the Pofadder project, Dr. Dennis Toens to Kader Asmal, then Minister of Water Affairs, requesting urgent intervention received a lukewarm response stating that no action can be taken as the prevalence of leukaemia in the region could also be caused by a range of other factors including toxic agro-chemicals, contaminated dust etc. not explored in the study. While three independent follow-up studies meanwhile confirmed the elevated levels of U in the region (Wullschleger et al. 1998, Sekoko et al. 2005, van Wyck and Coetzee 2006) no investigation into possible causes of the leukaemia cluster have been attempted in the past 16 years since the Toens report was published.

Despite being plagued by conflicting views of the various interest groups represented at the steering committee (Stoch EJ, McLaren A, personal communication 1999) the IWQS-study in the WFS marked the beginning of systematic scientific efforts to investigate the alleged pollution problem caused by gold mining. To this date the study remains the single most extensive and comprehensive investigation on the topic in South Africa based on the amount of analysed samples and time period covered.

Since then there have been over 40 related studies conducted in the Wonderfonteinspruit (WFS) catchment alone, many encountering similar obstacles caused by conflicting interests. One particular report (Coetzee et al. 2006) was for example embargoed by intervention of a particular mining house and a subsequent moratorium imposed on the report by the then Department for Minerals and Energy (DME). The report was published with a two year delay only after repeated requests from an environmental activist and a journalist (Liefferink M, personal communication 2006, Tempelhoff E personal communication 2011). When the report was eventually released it was hailed as first evidence of the 'democratisation of science in SA' (personal communication, Turton A 2006).

However, the National Nuclear Regulator (NNR) represented on the Steering Committee of the project insisted on including a disclaimer in the published version in which it disagrees with the risk assessment methodology and undertakes to conduct its own investigation. After considerable delay, this investigation finally got underway in 2006 when a German company (Brenk Systemplanung) conducted a radiological risk assessment on behalf of the NNR (Barthel 2007, Barthel 2011). The fact that this report (known as 'Brenk Report' even though its author was Dr. R. Barthel)

established even higher risks quotients triggered a public outcry accompanied by often sensationalising media reports in which headline phrases such as '*Death in the water', 'Lives at risks as mines coin it', 'Health nightmare* and '*Toxic tsunami'* were used, the latter latching on emotions the real tsunami in Indonesia had triggered just a few months before (Avni 2007, Tempelhoff 2007a, Beega 2008, Struijt 2008, Jordan 2009). The fact that the author of the Brenk report prevented from presenting an already accepted paper at a conference to be held in SA a few weeks later certainly did not help to instil public trust in open and transparent governance (Barthel et al. 2007).

After a shortened and edited version of the original report was finally released by the NNR mid 2007 on their website, a range of follow-up investigations started, concentrating on possible risk via the food chain as main exposure pathway identified in the Brenk Report (e.g. NNR 2007, McCrindle 2008, GFL 2008, Hamman 2012). Unfortunately, and this is stated with all due respect, many of these studies were conducted in an uncoordinated and *ad hoc* manner by activists, residents and academics that often were not familiar with this specialised field of science, nor with the study area resulting in contradictory and/or questionable findings that added to, rather than eliminated the confusion surrounding the issue. Associated newspaper articles claimed, for example, that thousands of cattle had to be shot because of pollution, that exposure to dust while driving through the area could lead to cancer and that extreme levels of U were found in vegetable grown in the area (Tempelhoff 2007c Tempelhoff 2008a/b, Botha 2008) triggering responses by readers and journalists questioning these statements and alleging deliberate distortion (e.g. Van Heerden 2007, Engelbrecht 2008, Martins 2008, Tempelhoff 2008c, Stoch 2008, Louw 2009a).

Following the Brenk report and the recommended need for intervention, a combined initiative by the DWAF and the NNR for rehabilitating the WFS catchment got underway with the aim of mitigating radiological health risks in the Wonderfonteinspruit catchment area (WCA).

In a first step a Specialists Task Team (STT) was appointed consisting of members from South Africa, Canada and the USA tasked to compile a so-called 'hot-spot map' indicating the sites needing most urgent intervention. The map was eventually produced by the author employing a specifically developed risk assessment methodology based on a combination of the extent of contamination and the associated probability of exposure to members of the general public (Winde 2008). However, instead of the STT jointly following up on how to address the identified 34 'hot spots' the consulting company that was originally tasked to merely facilitate easier project administration took over and, in its own capacity, effectively stopped further progress by compiling a final report (Remediation Action Programme, RAP) that later proved to be inadequate and was eventually rejected by the Steering Committee (Iliso 2008, Louw 2009b).

In order to overcome this set-back a new initiative was started, this time not only involving governmental representatives, as in the first attempt, but also the local mining industry (represented by the specially established Mining Interest Group,

MIG), environmental activists (FSE, WWF) as well as the affected municipalities. Having been appointed as expert advisor to the Steering Committee the author was tasked to develop a remediation strategy for the region. Unfortunately, progress stalled since mid-2011 following the involvement of newly appointed staff from NWU and resulting confusion on the role of the institution in the process (Phillips 2011, 2012).

Owing to the importance of the matter to the health of people as well as deliberate efforts of environmental activists to expose the matter, coverage of the U-problem in the media increased significantly. Apart from local weekly newspapers of the area concerned, national daily and weekly newspapers reported on the matter frequently on the front page allocating up to full double pages to the topic. Later the international news media, including well-known newspapers such as 'Die Welt' (Germany'), 'The Washington Post' (USA), 'Le Monde' (France), TV stations (German ZDF/ 3SAT; a Chinese TV station, Al Jazeera), South African prime-time TV show (Card Blanche, Fokus, Rooi Trok) as well as numerous online media and radio broadcasters from overseas (Canada, Belgium, Netherlands etc.) and South Africa covered the story.

The rise in public awareness following the Brenk report resulted, amongst other, in the formation of residents-driven interest groups such as the 'Wonderfontein Action Group' which – dissatisfied with the lack of governmental action – collaborated with parts of the mining industry in an effort to quantify the degree of sediment pollution (WAG 2007). The issues in the WFS catchment also initiated the foundation of NGOs such as the Public Environmental Arbiters (PEA) and the Federation for a Sustainable Environment (FSE) – the perhaps now most vocal, active and influential NGO of SA in this field. Largely owing to activities of the latter the WFS matter was repeatedly tabled in Parliament (e.g. Liefferink 2008.).

Although not located in the WFS catchment the Tlokwe municipality (formerly Potchefstroom) and the safety of its water supply soon became a major focal point in media reporting. While the local newspaper originally raised concerns about the problem in line with national media this later changed and increasingly the local paper assured residents that no further problem exists. This largely mirrored the changing attitude of City Council on the matter, which, in 2003, took legal actions against one of the upstream gold mines for polluting the town's water resources, but later claimed that no U-problem exists (Anonymous 2010a, Tempelhoff 2007e). In addition to a large (and presumably expensive) public-relation campaign with billboards and flags being placed across the entire town and full-page advertorials in local newspapers proclaiming the safety of its water supply, the City Council also organised a so-called 'Water Lekotla' aimed – according to the local newspaper – *…to get closure on the quality of Potchefstroom's water, once and for all'* while labelling the author as well as the head of research of the Cancer Association of South Africa (CANSA) as '*sceptics*' *…'whose comments have once again set the cat among pigeons*." (Anonymous 2010a). The overall tone of the announcement portrayed the forum as a kind of 'show-down' in which the '*sceptics*' would be put in their place.

Later media reports about the event, however, portrait a very different picture and generally agree that there are legitimate reasons for persisting concern (e.g. Anonymous 2010b). Following-up on results of a kettle-scale sampling campaign that indicated higher U-levels in calcite scales from Potchefstroom water kettles compared to nearby Ventersdorp where unpolluted dolomitic water is used (Winde 2003) CANSA is currently conducting a survey on U-levels in teeth of residents as possible bio-indicator for U-accumulation in exposed residents (Albrecht C personal communication 2012, Beega 2012).

The continued contradictions between reports in local and national news media further contributed to suspicions held in some quarters that efforts were still being made behind the scenes to obscure the true extent of the problem (Beega 2008b, Tempelhoff 2008d). The fact that recently released data of the water monitoring program of Potchefstroom indicate a significant rise in U-levels in raw and in tap water - a possibility predicted by the author on various occasions vindicates the '*sceptics*'.

In order to be inclusive on studies in SA relating to U-levels in the environment it should be mentioned that U-data for natural surface and groundwater bodies were provided by Kronfeld and Vogel (1991) surveying 20 rivers across the country and Sami and Druzinski (2003) using geological information to model possible U-levels in aquifers (unfortunately without providing concrete U-data). Neither of the two studies addressed the potentially associated pollution hazards.

5. Extent and risks of uranium pollution in SA: two case studies

The case studies covered here relate to two types of U-pollution:

(a) man-made (or anthropogenic) U-pollution caused by the century-old deep-level gold mining industry, and

(b) natural U-pollution associated with elevated U-concentrations in certain rocks.

While mining-related U-pollution occurs to different degrees across all gold fields of the Witwatersrand basin, the examples used refer to the Central Rand (CR), West Rand (WR) and Far West Rand (FWR) goldfields where U-related problems are particular pronounced. Two of the mentioned goldfields are located in the catchment of the Wonderfonteinspruit (WFS) which not only made national and international headlines because of radioactive pollution but also displays, in an exemplary manner, the wide variety and complexity of sources and pathways associated with mining-related U-pollution. As the water supply of the downstream municipality of Tlokwe is also affected by polluted water from the WFS, examples from this municipality are also included even though it falls outside the actual gold mining areas.

The effects of natural U-pollution are discussed based on results of an earlier mentioned study on the arid farming area near the town of Pofadder (Northern Cape) that suggested a link between U-levels in borehole water and leukaemia prevalent in residents of the area (Toens et al. 1997).

Case study 1: Mining-related U-pollution in the Witwatersrand goldfields

Pollution sources

In terms of their spatial dimension tailings are by far the most important source of pollution associated with gold mining. Tailings are the major waste product of mines and consist of milled ore from which the commodity of interest - in this case gold – is leached and which are then either backfilled into underground mine workings, or deposited on surface. As uranium is a natural constituent of the many auriferous ore bodies (termed 'reefs') in the Witwatersrand basin, gold mining inevitably also brought uranium to surface and into the biosphere.

Since most of the mined Au and U ore was deposited billions of years ago in an oxygen-free atmosphere, the mining of reefs which preserved the reducing conditions, now exposes the ore to a chemically aggressive atmosphere with an abundance of extremely reactive oxygen and free water as one of nature's best leaching agents. Since 1886, when Au-mining started near present day Johannesburg, gold mines brought an estimated 800,000 t of uranium to surface of which only about a quarter was extracted and sold while the vast majority is left in the tailings. Since the dawn of the new U-renaissance in 2003 many uraniferous slimes dams are reworked by local mining companies as well as foreign investors originating mainly from traditional U-producing countries such as Canada and Australia.

Because gold is an elusive metal with low concentrations compared to other commodities – the very reason why it is so expensive – large quantities of rock have to be mined in order to produce the required quantities. On average there are only 5-10 grams of gold in a ton of South African reefs accompanied by about 100 grams of uranium. Thus to produce 1 kg of gold, 100 to 200 t of rock are brought to surface along with approximately 10 kg U (varying from 2 to 40 kg of U depending on the mined reef). Where U has not been leached from the ore it remains in the tailings and is dumped at original ore-grade on the so-called 'slimes dams'. With average concentrations of 100 ppm (reaching up to well over 200 ppm in some slimes dams) U in these gold tailings contain higher U-levels than tailings of many genuine U-mines for example in the Wismut region where concentrations of 70-80 ppm are common (Wymer 1999, Winde 2009). A map of the location of the various goldfields of the Witwatersrand and mining-polluted streams and rivers is displayed in Fig. 2.

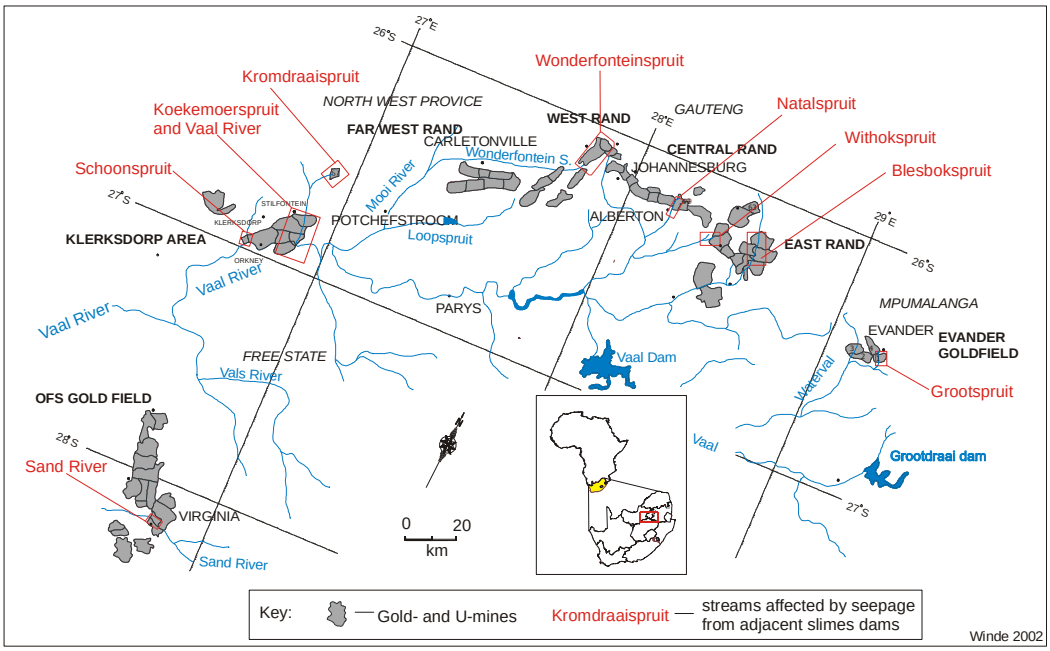

Fig. 2: Geographical distribution of gold mining areas (goldfields) in the Witwatersrand Basin including steams affected by mining-related pollution (Winde 2009a)

In addition to uraniferous tailings particles many inactive slimes dams develop another major U-source by forming extensive salt crusts that are readily dissolved by rainwater. Owing to the evaporation of contaminated porewater leaving dissolved salts behind, these crusts tend to enrich U to levels up to 10 times the concentration in underlying tailings (Winde and Sandham 2004, Winde et al. 2004, Winde and Erasmus 2011). In some instances, even waste rock dumps consisting of unmilled overburden, also act as U-source as examples from the Klerksdorp goldfield indicate (Labuschagne 2008).

Reefs with gold grades lower than expected are commonly left un-mined underground. Following the contact with oxygen and water U leaches into ingressing (fissure) water and thereby contaminates underground mine water. Acidic water decanting from the flooded mine-void system of the West Rand ('Western Basin') initially contained some 16,000 µg/l U (Coetzee et al. 2006).

The above average U-grades in the mined reefs of the WR and FWR resulted in a spatial focus of early U-production in the WFS catchment area, where at one stage 10 gold mines produced uranium for 5 U-plants (Winde 2010a). The very first pilot plant designed for extracting U from low grade ore was build at the Blyvooruitzicht Gold Mine near Carletonville while the first regular U-plant was commissioned at West Rand Consolidated in the upper part of the catchment (Taverner 1957). The produced 'yellow cake' (an U-concentrate) was further processed at the nearby Nuclear Fuels Corporation of SA (Nufcor) which sold over 240000 t of further concentrated U making it the largest continuous U-producer worldwide. Fig. 1 depicts countries with (at least temporary) U-production in 2003.

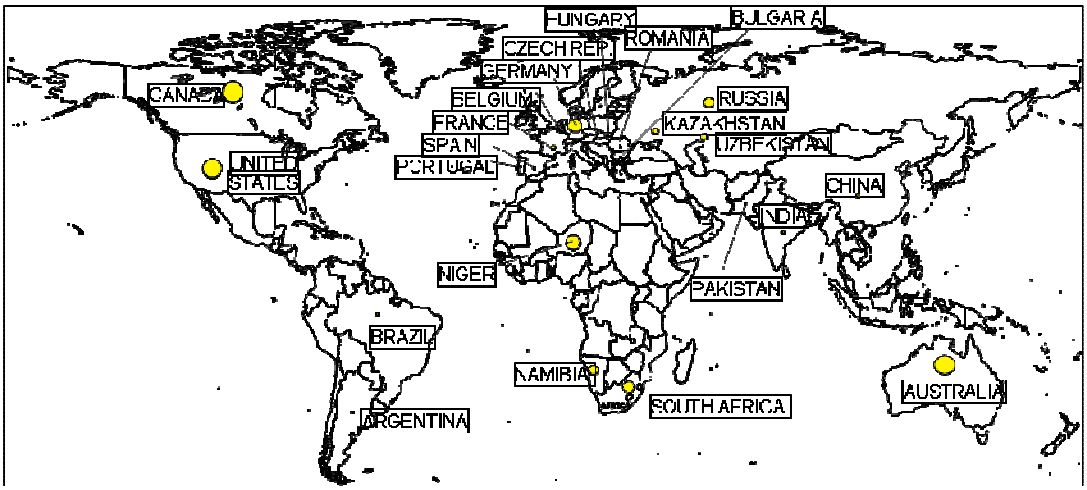

Fig. 3: Countries with at least temporary uranium production. The size of the yellow circles is a relative proxy for the extent of U-production (Winde 2009a)

Compared to U-levels in other mining areas worldwide, SA ore is regarded as low-grade displaying U-levels up to two orders of magnitude below Australia or Canadian ores. In fact, the tailings of Ranger Uranium mine in northern Australia, as a discarded waste product, contain more U (approx. 300 ppm) than most reefs from which U was produced in SA. Even in the Wismut area in Germany that also used mainly low-grade ore, the U-concentrations were on average 2-4 times higher than in SA. The difference is, however, that an average extraction efficiency of about 90% in dedicated U-mines left only a tenth of the original U contents in the tailings while in SA most mines for most of the time (for about 90 years out of 125 years) never extracted any U from the gold tailings (Winde 2009a). The extent to which U was accumulated on surface is perhaps best illustrated by the monetary value now placed on many the tailings deposits earmarked for re-processing reaching tens of millions of Rand per slimes dam.

Environmental transport

Given that U in natural soil surrounding the tailings in South Africa contains about two orders of magnitude lower levels of U, a steep concentration gradient between tailings and adjacent the environment exists that drives uncontrolled migration of U. Uranium migrates either in a solid phase – e.g. as eroded tailings particle - or dissolved in water. Both types and associated hazards are briefly discussed in the following section.

Particle-bound U-transport (solid phases)

The spreading of uraniferous tailings particles is either *deliberate* or *unintentional*. While the latter refers to wind and water erosion as well as failure of slimes dams, the former comprises the intentional disposal of tailings outside of dedicated tailings storage facilities, or underground mine workings. It was mainly practiced in dolomitic areas of the FWR where, for a limited period of time, tailings were pumped into natural caves on mine property in order to save disposal costs. Additionally, following the lowering of the groundwater table by mines, large amounts of tailings

were pumped into the many subsequently forming sinkholes to reduce infiltration of surface water into the underlying mine void.

In some cases, where sinkholes were connected to large underground cavities, even the continued injection of the full daily tailings production for several months on end could eventually not fill the sinkhole. In other cases successfully filled sinkholes re-opened after rainfall washing tailings into underlying karst receptacles. Seepage from tailings-filled sinkholes was found to be a significant potential source of U-pollution with modelled U-concentrations of up to 300 mg/l (Metago 2003).

Tailings also moved into the karst aquifer through slimes dams partly collapsing into sinkholes forming by the ongoing outflow of tailings seepage. While not posing a major threat to groundwater quality yet, this may change when the dewatered karst aquifers fill up again following the inevitable cessation of pumping by mines.

Many active and decommissioned slimes dams are also affected by wind erosion blowing off significant amounts of fine tailings dust especially during dry winter months. This not only affects nearby residents but also contributes indirectly to pollution of urban water courses as much of the blown-off dust settles on impervious surface from where it is flushed into stormwater drainage systems. Concentrating dust from relative large urban catchments the resulting tailings volumes finally being discharged into receiving streams are significant.

Apart from wind erosion, tailings are also eroded by rainfall affecting especially unvegetated slimes dams which often form deep gullies that accelerate the access of oxygen and the associated acidification of porewater. Once flushed into nearby streams the eroded tailings themselves act as source of acid mine drainage (AMD). However, the mixing of eroded tailings with unpolluted fluvial sediments and natural soil normally results in U-levels decreasing rapidly with increasing distance from the source of pollution (Winde & de Villiers 2002). This is very different for dissolved U which can be re-concentrated well outside of access-controlled mine properties to levels exceeding the concentration in the original pollution source.

Transport of dissolved U (water phase)

In many respects more problematic is the transport of dissolved U along with seepage or surface run-off from tailings dams. U is either leached from the milled ore into the slurry or from the deposited tailings into the tailings porewater. The porewater is recharged by infiltrating rainwater and excess slurry water in active slimes dams. A couple of metres below the tailings surface this creates a so-called piezometric surface, an elevated water table well above the natural water tables in adjacent streams and aquifers. The resulting hydraulic gradient drives porewater in form of seepage out of the slimes dams polluting nearby ground- and surface water bodies. The U-transport along the so-called ‘aqueous pathway’ occurs largely unabated since tailings dams in SA, for cost reasons, are generally not lined.

Owing to the limited transmissivity of the fine grained tailings, the vertical percolation through the slimes dams is slow resulting in a continuous outflow even in dry winter times. While many small streams in the gold mining areas stopped flowing during winter, this often is no longer the case as the continued outflow of seepage

from the large tailings dams generates artificial baseflow that feeds adjacent water courses all year round. Since many gold mining areas are located near watersheds formed by outcropping reefs, mining affects the relatively small headwater regions of streams - normally the most pristine areas along a river. Vast mine waste deposits in form of slimes dams, sand and rock dumps can cover a quarter and more of the total surface area in these small catchments. This results in heavy stream pollution displaying a typical mining signature of low pH-values and high salt loads often recognisable with the naked eye by the reddish colour of the water (Winde & Sandham 2004).

This signature is not due to the adding of leaching chemicals but caused by a natural process in which reduced sulphur contained in the mined ore (mainly in the form of iron sulphide – also known as pyrite or 'fools gold' because of its glittery appearance) is oxygenated and together with water forms sulphuric acid. The oxidised iron from the pyrite gives such water – then called acid mine drainage (AMD) or acid rock drainage (ARD) - its characteristic reddish colours. In streams affected by AMD aquatic life is often drastically reduced (although not altogether wiped out as crabs, for example, were observed by the author to live in pH 3 water) and display an overall poor water quality rendering stream water unfit for irrigation, animal watering, recreation etc. Since many heavy minerals that remain in the tailings are rapidly dissolved by acidic water, toxic metals are also liberated by AMD. While the oxidisation of reduced sulphur is an entirely natural process, mining drastically increases the extent and rate at which it occurs, firstly by exposing large volumes of sulphides to atmospheric conditions, and secondly, by expanding the reactive surface area of the mined rock through milling. Since the leaching of U occurs at the solid-water interface the area-size of the tailings particle surface is crucial for the rate and extent at which U and other toxic metals can be leached from the tailings. While milling increases the extraction efficiency for gold it also, inadvertently, promotes the unwanted release of U from the leached ore.

Topographical and climatic factors

Owing to the general shortage of naturally available water in and around Johannesburg, competition for water from the many small streams was fierce among early miners whose economic survival depended on it for running steam engines and other mining processes. In order to store water of the non-perennial streams for the dry period artificial reservoirs and dams were created and streams diverted to bring water close to the mines. Inversely, mine infrastructures, such tailing dams, also tended to be placed in the vicinity of water sources. As a consequence many mine waste deposits are now located near, or even in water courses, dams, wetlands and canals. In some instances return water dams for recycling slurry water from tailings dams were placed directly into water courses such as the Tudor and Lancaster dams in the upper WFS. As a consequence, the distance between potential pollution sources and receiving water courses was often considerably shortened promoting water pollution.

Apart from seepage emanating from tailings dams and rock dumps and diffusely entering adjacent streams, U is also directly introduced via discharged mine effluents. Most of this mine water is pumped from underground where it gets contaminated by

un-mined ore and dust. While in non-dolomitic mines the pumped mine water typically displays an AMD signature this is not necessarily the case in dolomitic area, where the large volumes of alkaline karst water are well-buffered. While dilution associated with the large volumes of ingressing dolomitic water (up to 100 Ml/d in the FWR mines) results in comparably low U-levels in pumped mine water (mostly 100– 400 µg/l), the associated U-loads can be significant. Winde (2010b) estimates that over 3 t/a of dissolved U are discharged into the WFS alone excluding the U-input associated with eroded tailings being washed into the stream.

Based on over 12 studies on U-pollution of sediments the Winde (2009b) estimated that fluvial sediments in the WFS catchment contain over 2200 t of U of which about a third consists of non-tailings material i.e. secondary U-accumulation in fluvial sediments suggesting an average rate of U-accumulation over the past 122 years of mining of slightly over 6 t/a. Together with the load of dissolved U of 3.5 t/a the WFS receive close to 10 t particulate and dissolved U of which approximately two thirds are retained in sediments. Even on a global scale such U-loads are significant. The effects of tailings deposits of the Central and East Rand on the pollution of Gauteng's largest River, the Vaal, were recognised decades ago and are well documented (e.g. Marsden 1986, SRK 1988). Based on an U-concentration of 7 µg/l measured at Orkney (Kronfeld and Vogel 1991) the Vaal River, at the time, carried an average U-load of approximately 7 t/a coming from the upstream goldfields.

Chemical mobility

In contrast to many heavy metals such as Cd, Pb or Ni, which are only mobile in acidic conditions, U displays a very complex geochemical behaviour and is soluble over a wide pH-range. As a result, leaching of U is not confined to acidic conditions but also occurs in neutral and alkaline waters. Furthermore, while many dissolved heavy metals precipitate at higher pH values, this is not necessarily the case with U that often forms soluble complexes that stay in solution. Much of the mobility of U is controlled by redox-conditions. Instead of forming insoluble precipitating sulphides like many other heavy metals, U - in reducing conditions - changes from a hexavalent to a tetravalent ion and forms insoluble oxides. Where this occurs, as for example in organic rich dam sediments, U may be immobilised. However, exposing such sediments again to atmospheric oxygen may release parts of the accumulated U back into the water (Wade et al. 2002). When such scenario seemed to be possible after a farmer in the WFS drained a dam with highly U-contaminated sediments in an attempt to extract the associated gold, the downstream municipality of Potchefstroom took legal action against the nearest gold mine fearing that the released U would pollute its drinking water supply (van Heerden 2003a/b).

Apart from redox-triggered immobilisation, which is uncommon under normal environmental conditions given the very low redox potential required, U can also be removed from the water phase by co-precipitation, i.e. the incorporation into insoluble compounds formed by other elements. Owing to the general abundance of dissolved iron (Fe) in mining waters, U often co-precipitates along with iron-hydroxides. As this process strongly depends on the pH, U-mobility is indirectly

affected by the natural diurnal and seasonal pH-oscillations which Winde (2002) introduced into the scientific discussion.

A major consequence of Winde's findings is that standard monitoring protocols, which sample only during day time, tend to underestimate the true extent of heavy metal pollution in the sampled stream as highest levels are reached at night when co-precipitation is at a minimum. In mining areas, the fluctuation of U-levels is further increased by varying rates of U input into receiving stream as most deep level mines try to reduce the massive pumping costs by pumping higher volumes at night (and weekends, or holidays) when electricity tariffs are lower. Since standard monitoring programmes do not take samples at night, weekends or holidays, they tend to systematically underestimate the extent of mining-related pollution. Owing to the superimposition of low natural U-immobilisation with increased U-input, the net U-levels are likely to be highest at night and in response to heavy rainfall (dissolving uraniferous salt crusts).

Since pH-oscillations are induced by the fluctuating carbon dioxide-calcium carbonate–equilibrium Winde (2006 b) suggests that the massive eutrophication of shallow farm dams in the WFS explains the extraordinary degree of U-accumulation in dam sediments. With U-levels of several hundreds of ppm the sediments exceed the U-concentration in ore and tailings as original source of the pollution (Wade et al. 2002).

Owing to the abundance of calcium and carbonate in dolomitic water U is also co-precipitated along with calcium-carbonate (calcite) (Winde et al. 2004). Since the CO_2–$CaCO_3$-equilibrium strongly depends on the level of dissolved CO_2 any changes in concentration caused by varying atmospheric CO_2 pressure and/or water temperature also affect the rate of calcite precipitation. Since dolomitic groundwater is often pumped by mines from depths of up to 3 km below surface the associated drop in atmospheric CO_2 pressure triggers the escape of dissolved CO_2 and results in the precipitation of uraniferous calcite. This is somewhat counteracted by the decrease in temperature of the water pumped to (a generally cooler) surface somewhat increasing the retention of dissolved CO_2. However, while uraniferous calcite layers line many mine water canals in dolomitic areas and cause elevated radiation signals in airborne surveys, the associated removal of U is only of academic importance and most likely not even detectable by standard analytical techniques.

Factors and processes affecting the waterborne transport of dissolved U along the aqueous pathway are summarised in Fig. 4:

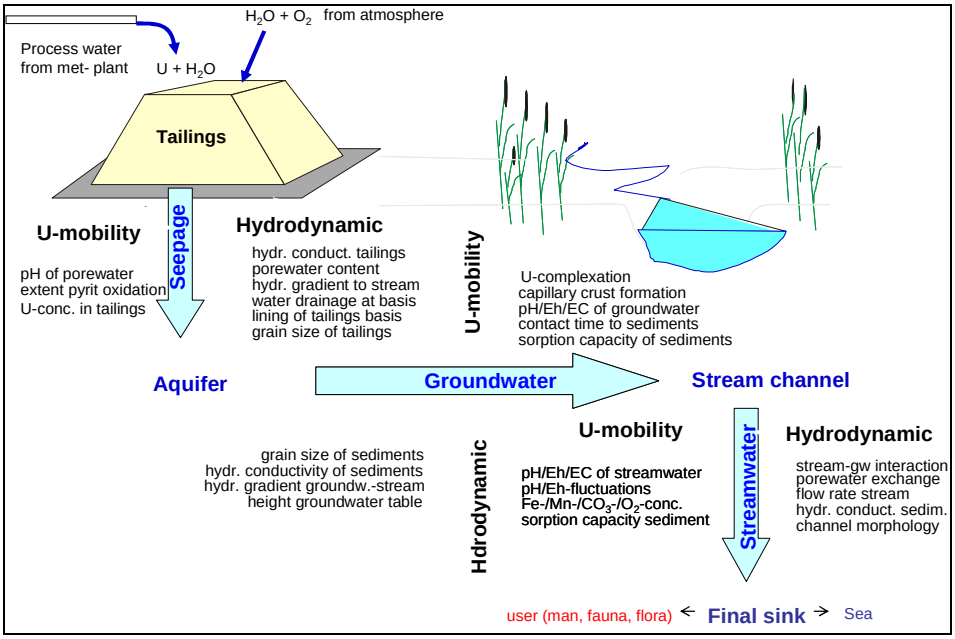

Fig. 4: Factors and process governing the physical transport and chemical mobility of dissolved uranium (U) along the aqueous pathway (Winde 2009a)

Extent of pollution (hazard potential)

By their sheer size and volume, gold tailings in SA are a major source of U-pollution releasing particulate as well as dissolved U uncontrolled into the surrounding environment. While the outflow of U-polluted seepage is generally a low-intensity, but continuous process (low magnitude – high frequency) primarily affecting shallow aquifers and adjacent streams, the dissolution of highly U-enriched salt crusts results in high but relatively short U-peaks (high magnitude – low frequency). As a result of the superimposition of both processes aquifers and surface waters in the Witwatersrand display elevated levels of U across the seasons. Table 3 provides on overview on uranium levels in natural and polluted surface and groundwater worldwide.

Table 3: Average and maximum uranium (U) concentrations in natural uncontaminated waters and in polluted water for selected streams and aquifers in South Africa and international examples

Type of water	Location/name	Year	Ref.	U [µg/l]		
				n	av	max
Natural background						
global freshwater average	world rivers, discharged-unweighted	1996	DWAF (1996a)		0.4	
sea water		1997	DWAF (1996a)		3	
regional background dolomitic water	upper Mooi River	2009-2012	Potch Municp (2012)	*162*	0.2	
	Kromdraaispruit	1999	own data	*4*	0.1	
Unpolluted streams South Africa	whole of SA except Witwatersrand	1990	Kronfeld & Vogel (1991)	*18*	1	3
unpolluted (dolomitic) groundwater SA	Bovenste Oog	1997	IWQS (1999)		0.2	0.8
Rainwater	*Klerksdorp gold mining area*	19 Oct.1999	own data	*1*	<0.01	
Mining-polluted water						
Rain	*West Rand, Westonaria*	14 Jan. 1988	Dorling (2008)	*1*		27
Streams						
SA: Witwatersrand	CR + FWR + KOSH (Klip, Vaal, WFS)	1990	Kronfeld & Vogel (1991)	*5*	29	120
SA: Witwatersrand	Wonderfonteinspruit	1997-2010	Winde (2010b)		79	759
SA: Witwatersrand	whole Witwatersrand	1995-1996	Faanhof et al. (1995), Ken			4100
SA: Witwatersrand	whole Witwatersrand	1999-2000	own data	*20*		500
Germany (Wismut)	Lerchenbach at Culmitzschaue	1991-1994	TLU Jena (1994)	*24*	455	1690
Groundwater						
SA: shallow alluvial aquifers	Klerksdorp goldfield, Koekemoerspruit	1999	own data	*7*	286	540
SA: dewatered dolomitic aquifers	FWR: pumped water Driefnt. - Doornfnt.	1996	IWQS (1999)	*54*	282	2576
Germany (Wismut)	Lerchenbach at Culmitzschaue	1996-1999	own data	*4*	1600	2100
Naturally contaminated water						
SA: N-Cape	Pofadder area	1996	Toens et al. (1997)	*10*	101	294

(TLU – Thüringische Landesanstalt für Umwelt – unpublished data)

Table 3 indicates that U-levels in the WFS exceeds the regional background level for uranium on average by 400 times reaching over 4000 times in the upper reaches of the stream. Compared to the average in rivers of the whole of SA (which includes some areas with elevated U-levels in rock) the U-level in the WFS is between 80 and 250 times higher. However, compared to the Wismut region this is relatively low with U-levels in the Lerchenbach (at the beginning of remediation programme) exceeding those in the WFS by up to 500%. On the other hand, smaller streams in the Central Rand, such as Russels' stream, where less clean water is diluting the U-input display even higher peak concentrations than the Lerchenbach.

Polluted groundwater in SA's mining areas displays a generally higher average U-level than surface water but lower than levels found in groundwater of the (humid) Wismut region despite higher recharge rates of the latter. Natural groundwater in the arid Northern Cape Province contains on average somewhat less U than contaminated groundwater in the mining areas but exceeds the average level in the WFS (Table 3).

While stream pollution by diffuse and point sources occurs in many mining areas worldwide the intentional deposition of tailings into karst aquifers is perhaps unique to SA – especially given the potential role these large groundwater resources could play for the nearby water-stressed metropolitan area.

Regarding the concentration of U in mine water and environmental media, South Africa generally falls into the range of values reported for other mining areas. For tailings seepage the highest U-concentration of 30 000 µg/l was found in a toe dam of a decommissioned slimes dam in Boetrand (Klerksdorp goldfield) (own data). In dolomitic areas, non-acidic tailings seepage commonly display much lower levels of around 1000 µg/l, however maxima of over 20 000 µg/l have been reported. Pumped dolomitic fissure water contains 100 to 400 µg/l U but may also reach 3000 to 5000 µg/l (Pulles 1991, IWQS 1999). This compares to U-levels in seepage of the humid Wismut area of 1000 – 2500 µg/l. Fig. 5 depicts U-levels in selected tailings as well as in associated seepage of different U-mining areas in SA, Namibia, Germany and Australia.

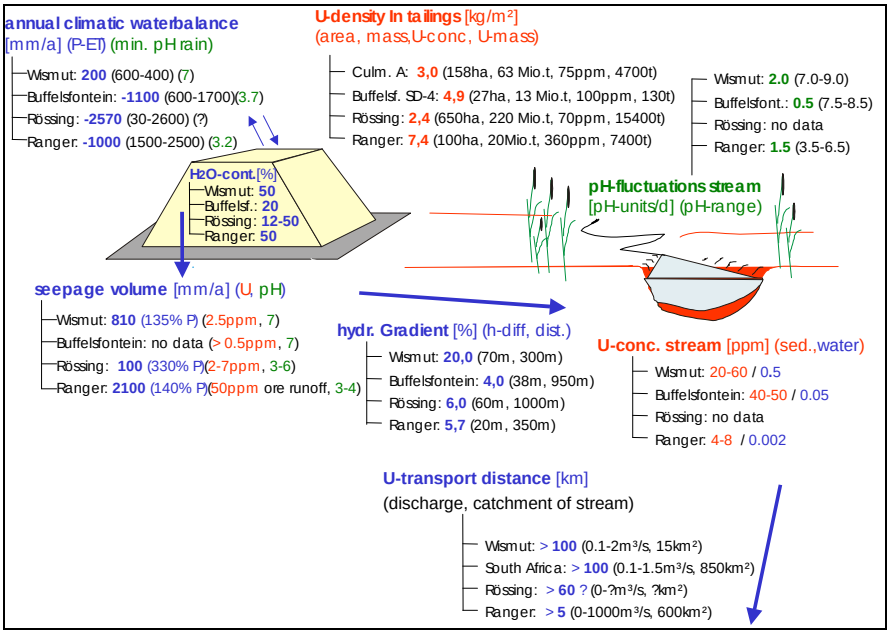

Fig. 5: Potential and extent of waterborne uranium pollution in mining areas of different climatic conditions (Winde 2009a)

Apart from comparable U-levels in the mine waste deposits the tailings deposition techniques practised elsewhere are also often not more environmentally friendly than in South Africa as the disposal of U-tailings in deep lakes in Canada, or rivers in Indonesia suggest. U-tailing in the USA were even deposited in the middle of towns and offered to local residents as building sands (Grand Junction, Colorado, USA) (Waggit 1994). The same option was practiced in SA where tailings have also been used as termite deterrent in basements of wooden-floor houses (Erasmus E, personal communication 2010), as well as for brick production.

A major difference, however, is the fact that many U-mines world wide are located in remote and sparsely populated areas such as the Saskatchewan lakes area in northern Canada near the arctic circle, the Alligator River Region in tropical northern Australia, or many desert-type regions in the USA. While mining in the Wismut area of Germany occurred in relatively densely populated regions (although less dense than in SA) the associated exposure of the local population is considerably smaller than in SA given the general access to safe water.

Human exposure and health risks

For assessing potential health risks associated with mining-related U-pollution four distinct groups of receptors are distinguished:

(a) Mine workers directly exposed by drinking U-polluted cooling water

(b) Poor, mostly black people living in informal settlements at the urban fringes and next to mines which often lack access to basic water infrastructure as well as poor rural dwellers relying on small-scale subsistence farming and gardening using untreated water from polluted streams

(c) Commercial farmers living in rural parts of the mining areas relying on polluted stream water for irrigation and animal watering, as well as consumers of contaminated agricultural products

(d) Residents in municipalities receiving locally sourced water potentially affected by mining-related U-pollution

(a) Underground mine workers

Based on urine samples from 484 randomly chosen mine workers of Deelkraal Gold Mine it was found that the high U levels in cooling water corresponded with higher urine concentrations of U in miners working below 9 level, some displaying concentrations as high as over 50 µgU/l. At NUFCOR workers are removed from the work place at such levels (Deelkraal GM, post 1991). Compared to U-levels reported for urine samples taken from 1518 German citizens between 2001 and 2003, ranging from 0.001 to a maximum of 0.32 µg/l (UBA 2005), U-levels in urine of underground mine workers were up to 100 times higher. The exposure of mine workers mainly results from the (illegal) consumption of chilled service water used as a cooling agent underground, which can contain as much as 4000 µg/l U at Deelkraal GM and 20000 µg/l U at W-Driefontein (COMRO 1990, 1991; Pulles 1991).

While it was first assumed that consumption of such water may have contributed to very high U-levels of 1300 - 13000 µg/l found in toilet drains of changing rooms at several gold mines (Pulles 1991) this is unlikely to be the case given the exceedingly high U-levels. According to E. Erasmus (pers. communication 2010), in some mines process water from the metallurgical plant is run through sanitary systems including toilet drains – i.e. no flushing required – before eventually flowing into the sewage plant and being discharged. This is more likely to be the actual cause of the high U-levels found in toilet drains and also explains why Slabbert (1996) considers sewage effluents as a major source of U-pollution in discharged mine effluents.

In some instances whole mining communities have been supplied with underground water pumped by the mines, as was the case at the Blyvooruitzicht village near Carletonville. The water was apparently of such poor quality that visitors to the mining village found the water unpalatable even when used for coffee. According to a teacher working at a nearby school for slow learners in Carletonville (Goudwes School) most of the pupils in this special school came from the Blyvooruitzicht village (EJ Stoch, pers. communication 2008) sparking speculations on possible links to recent findings that U adversely affects cognitive performance. Since U-levels in Blyvooruitzicht were of such grade that the very first U-extraction pilot plant was build at this gold mine in 1949 chances are that the pumped groundwater supplied to the mining community did contain elevated U-levels.

(b) Residents of urban and rural informal settlements

Resulting from an unfortunate combination of natural physico-geographical factors and low-cost oriented tailings-disposal as well as compounding climatic and

topographical factors, stream pollution in many goldfields of the Witwatersrand, especially in and around Johannesburg, reached extraordinary proportions already decades ago and appears to continue unabated. While many informal settlements at the urban fringes are supplied with safe municipal tap water (stand pipes) the use of polluted stream water, especially for small scale urban gardening, or to augment domestic water as well as recreational use such as swimming, bathing, playing (mostly children) and fishing constitute a realistic exposure pathways in such areas (IWQS 1999, Du Toit 2007).

The associated exposure to contaminated soil, water, food and air (incl. inhalation of the radioactive gas, radon, released from the uraniferous tailings as well as dust) is often exacerbated by tailings being ingested either unintentionally by children as part of hand-to-mouth activities (pica behaviour), or by adults due to deliberate geophagy. The latter refers to the practice of pregnant women who eat soil as a means of obtaining mineral supplements. Moreover, cakes made of tailings are reportedly sold as muti (traditional African medicine) and eaten as cure for certain ailments, while a tailings paste directly applied on the skin is used as a cure for acne (Beega 2011).

In some case uraniferous tailings have also been used as building sand for residential housing as well as for producing building bricks, a practice apparently still ongoing in a brick factory near Krugersdorp (M Liefferink – personal communication 2012).

While a number of campaigns aimed at raising awareness were conducted, generally low levels of education, the prevalence of more pressing problems associated with the extreme poverty prevalent in the area (including crime and food insecurity etc.) seem to prevent a lasting change of risk-taking behaviour. At the same time the prevailing lack of medical care, effects of malnutrition, wide-spread abuse of drugs and compromised immune systems due to the high prevalence of HIV infections especially among migrant workers, render informal settlers a very vulnerable population which is ill equipped to tolerate additional toxic stress. As a consequence the most vulnerable population is often also the most exposed.

A water user survey in the WFS in 1997 indicated that many informal settlers not only use untreated water from polluted streams, but also highly contaminated mine water taken directly from canals for all domestic purposes incl. cooking, drinking, personal hygiene, washing of clothes etc. as well as for watering gardens and small domestic animals (chicken, goats etc.) (IWQS 1999). In addition many households also consume fish caught in the polluted WFS. An informal settlement in the upper part of the catchment was even located on top of an old tailings dam (which also occurs in other areas of the Witwatersrand) where the planting of vegetables in the tailings material was found to occur. In Gauteng alone an estimated 1.6 million people live on, adjacent, or near tailings dams (FSE 2012).

While the extent of pollution in the target area is comparable to that found in mining areas worldwide, the combination of a multitude of partly very intense exposure pathways with an increased vulnerability results in exceptionally high risk potential.

So far, however, there are no supporting epidemiological data available. Apart from the rather short research time in SA, which did not allow for long-term studies as well as the methodological challenges, lack of resources and apparent disinterest of the

authorities and the mining industry combine to explain the lack of relevant data. Many adverse health effects such as specific cancers take several years, even decades, to manifest and become diagnosable. However, given the high mobility of many migrant workers that often populated these areas for relatively short periods of time and the low degree of medical care they are exposed to in the hinterland where they reside, it may prove difficult to detect any mining-related health burden as affected residents may have left the area before symptoms occurred, or being diagnosed and put on record by local doctors. The high prevalence of premature deaths associated with the HIV pandemic of which the mining areas with theire single sex hostels for migrant workers appears to be the epicentre, may further obscure the diagnosis of related diseases.

(c) Commercial farmers, consumers of contaminated agricultural products

The survey of the WFS catchment by Barthel (2007) as part of a radiological risk assessment on behalf of the NNR, identified a range of realistic and potential pathways relating to the use of contaminated water for the production of food (irrigation, cattle farming) and found that radiation exposure exceeded the limit of 1 mSv/a (by up to a factor of 100) at over 50% of the sampled 47 sites. As a pathway of particular concern Barthel (2007) identified the consumption of contaminated meat resulting in the highest dose rates especially for 12-17 year olds. Barthel (2007 and 2011) also criticised IWQS (1999) for underestimating the radiological health risks in the WFS due to methodological shortcomings in the dose-based risk assessment.

As Barthel's study was only based on a limited number of grab samples and relied on locally untested transfer coefficients taken from internationally accepted models, many of the predicted contaminant radiation levels for components of the food chain are hypothetical and need verification. As a direct consequence of the Brenk Report, the NNR issued a Directive that ordered the gold mines in the catchment to analyse a range of food samples and furnish the NNR with the results (NECSA 2007a-e). These data were augmented by a number of studies that also analysed selected vegetables, as well as milk and meat of animals from the areas. Selected results of these studies compared to international findings are shown in Table 3.

Table 4: Uranium concentrations in selected food sampled in the lower Wonderfonteinspruit catchment compared to international data from U-contaminated mining sites as well as from uncontaminated control sites (all original units have been converted into µg/kg U238, U-levels given per wet weight have been converted to dry weight based on the estimated water contents of samples to allow for better comparison)

Data source	Study area	Analysed material	^{238}U-conc. [µg/kg]*** min	max	*fresh weight correction* [b] *water contents [wt %]*	*conv. U-conc. [ppb dry weight]*	Sample with max U
NECSA (2003)	Deelkraal recerational dam	fish (n=1),	**771**				
NECSA (2007a-d)	lw WFS, Dfnt . GM	fish (n=4),	**<21.3**[a]	**1130**	*90%*	***213-11300***	NECSA 2007b - not specified
(all samples ashed)	lw WFS, Dfnt . GM	chicken (n=1)	**<1700**[b]				
	lw WFS, Dfnt . GM	meat (type not specified, n=3)	**<230**		*80%*	***<1840***	
	GMB, vegetable garden Potgieter	sping onion, oats, asparagus (n=3)	**3**	**27.4**	*90%*	***274***	spring onion
McCrindle (2008)	lw. WFS incl. Visser farm	cattle tissue (bulls, n= 45)	**<0.006**	**9**			bone
	and upper Mooi River **	cow milk (n=1, dry weight)	**<0.006**				
Hamman (2012)	lw. WFS - Visser farm	cattle tissue (cows, 6 data sets)	**220**	**31810**			liver
	lw. WFS - Visser farm	cattle fodder (grass, 3 data sets)	**2980**	**350000**			gras from river banks
	control site: upper Mooi River	cattle tissue	**4**	**50**			liver, spleen, muscle
Fisenne (1987)	New York City, USA	vegetables, cereals, fish	**1**	**15**	*85%*	***10-150***	
Anke et. Al (2009)	Wismut U-mining area (Ronneburg, Germany)	12 species of vegatables, wild clover (n=121)	**1.2**	**78**			lettuce
	Control site	13 species of vegatables, wild clover (n=121)	**0.1**	**18**			lettuce
Neves et al. (2011)	Cunha Baixa U-mine (Portugal)	potatoes irrigated with U-cont. water (n=4)	**7**	**590**			unpeeled potatoes
Roivainen et al. (2011)	U-exploration mine, in boreal forest (Finland)	4 forest flowers and ferns (n=100)	**0.01**	**0.17**			narrow buckler fern
Thomas (2008)	U-mining site Saskatchewan, Canada	9 types of vegetables, cereals, gras (n=9)	**<160**	**12098**			Barley (Eston)
		14 types of small mammals (n=40), wet weight	**242**	**656**	*85%*	***2420-6560***	deer mice

** lw. WFS - lower Wonderfonteinspruit*
n - number of analyses, not specified in Hamman (2012) where only aggregated data are given
*** no clear distinction between contaminated and control samples possible*
a - U conc. relates to fresh weight, in order to make results comparable data are converted to dry weight based on estimated water contents of the analysed sample
b - LDL 7 x higher than for other samples
< below lower detection limit (LDL)
**** all values rounded to one decimal except for LDL*

(lw. WFS – lower Wonderfonteinspruit; Dfnt. GM – Driefontein Goldmine, GMB – Gerhard Minnebron wetland)

Table 4 illustrates that U-levels in food sampled in the lower WFS catchment shows a wide range of U-levels spanning a total of 9 orders of magnitude.

By far the highest levels are reported by Hamman (2012) for samples from the lower WFS, where not only the maxima but also a minimum exceed all levels so far reported in pertinent literature as reviewed by Miaullis (2012) who, in a global literature survey, found that 37 Bq/kg (equalling 1400 µg/kg) was never exceeded in any food sample, suggesting this to be the maximum level to which U can accumulate in organisms. Compared to this, the 350000 µg/kg U (350 ppm) reported by Hamman (2012) for grass growing next to the polluted WFS are over two orders of magnitude higher (250 times) by far exceeding all values reported for biological samples from contaminated sites listed in Table 4.

Apart from the fact that the reported U-concentration in grass would exceed U-levels in most South African ore, it is also higher than that of the soil in which the grass grows, resulting in transfer factors exceeding values reported internationally by several orders of magnitude (e.g. Rodriguez et al. 2006, Schick et al. 2008, Anke et al, 2009, Shtangeeva 2010, Roivainen et al. 2011). Although the U-contents of soils and sediments in the WFS frequently exceeds the guideline value of 23 ppm (dry weight) stipulated by Canadian authorities for agriculturally used land (CCME 2007) their U-levels are comparable to soils in U-mining regions elsewhere.

Apart from the misfit with international results, the data in Hamman (2012) are also in stark contrast with results of studies conducted in the very same area. The maximum U-levels in meat reported by NECSA (2007a-d) and McCrindle (2008) are between two and four orders of magnitude lower. The Necsa-data are in line with U-levels reported for potatoes grown in a former U-mining area in Portugal (Neves et al. 2011) (Table 4). This is particular relevant as the U-levels in soil at the Portuguese site are comparable to those reported by Hamman (2012) (43–134 mg/kg dry wt.) while the water used for irrigating the potatoes in Portugal was much higher polluted than the WFS (1035 µg/l vs. 233 µg/l). Yet, the average U-level in (unpeeled) potatoes is over 70 times lower than the average U-level in irrigated grass reported by Hamman (2012) (218 vs. 15850 µg/kg dw). Even the highest U-level in potatoes from the U-mining site in Portugal (590 µg/kg dw) is still nearly 30 times lower than in

grass irrigated with stream water[4] containing on average 20 times lower U-levels (Table 4).

In view of these large discrepancies with local and international results the "alarming" data reported by Hamman (2012) should be interpreted with caution. It is unfortunate that Hamman (2012) fails to provide the required technical detail of the analytical method used, an unusual omission for an academic thesis, and even mistranslates the acronym ICP-MS which suggests a degree of unfamiliarity with the intricacies of trace analyses in general.

However, given that Hamman (2012) indicates nowhere that he doubts the accuracy of his own data, his conclusion that there is no health risk associated with the consumption of such highly contaminated meat is disturbing given the serious nature of the matter at hand. Using the data produced by Hamman (2012) for the most highly contaminated part of the cows, the liver, as well as for meat (muscle) for calculating the average daily intake of U associated with the consumption of such meat (based on consumption parameters specified in NECSA 2007a) the resulting U-intake for all age groups dramatically exceeds the applicable TDI limit of the WHO by orders of magnitude (Table 5).

Table 5: Average meat consumption for different age groups and the intake of U associated with the consumption of contaminated cattle meat as reported by Hamman (2012) compared with the TDI-value for U of the WHO of 0.6 µg/kg bw x day (WHO 2006).

Exposed humans		**Average**	Associated **U-ingestion**							
Age	**Body weight**	**meat**	**cow liver**				**cow muscle meat**			
		consumption*	**max** U-conc.	**31810**	**av.** U-conc.	**5070**	**max** U-conc.	**4630**	**av.** U-conc.	**1580**
[a]	[kg]	[kg meat/d]	[µg/kg bw x d]	% of TDI (WHO) (0.6 µg U/kg bw x d)	[µg/kg bw x d]	% of TDI (WHO) (0.6 µg U/kg bw x d)	[µg/kg bw x d]	% of TDI (WHO) (0.6 µg U/kg bw x d)		
<1	5	0.027	174	29030%	28	4627%	25	4225%	9	1442%
1 - 2 a	15	0.055	116	19354%	19	3085%	17	2817%	6	961%
2 - 7 a	25	0.137	174	29030%	28	4627%	25	4225%	9	1442%
7 - 12 a	40	0.205	163	27216%	26	4338%	24	3961%	8	1352%
12 - 17 a	50	0.274	174	29030%	28	4627%	25	4225%	9	1442%
>17 a	60	0.274	145	24192%	23	3856%	21	3521%	7	1202%

* based on annual consumption of meat for different age groups as given in NECSA (2007a)

The consumption of cattle liver by members of the various age groups would exceed the WHO TDI limit between 193 and 290 times posing a serious chemotoxic health risks, with babies (<1a), 2-7 years-old children and 12-17 years- olds youth incurring the highest risks (Table 5).

Related to the average U-intake of 1–1.5 µg U per day observed in numerous studies (Harley 1988, UBA 2005) covering the total diet including vegetables and drinking water, the resulting U-intake for adults only via consuming contaminated liver from cattle is 8589 µg/day - about 8000 times higher – and that for one particular type of food. Together with contaminated vegetables and drinking water the U-intake would be much higher which fact Hamman does not bring into consideration. While U-intake in uranium mining areas is generally elevated above the average (ranging from 13 to 18 µg/day, (Wrenn et al. 1985, UBA 2005) these values are still orders of magnitudes below the intake resulting from U-levels in meat as reported by Hamman (2012).

[4] Assuming that the water was indeed coming from the WFS which at the farm in question is not necessarily the case as irrigation water was normally drawn from a borehole with much better quality water.

The consumption of liver, with *maximum* U levels is a worst-case scenario required for arriving a conservative risk assessment. The ingestion of liver with an *average* U-concentration as well as beef ('muscle') of average and maximum U-contamination results, for all age groups, in unacceptable risks exceeding the guideline value by orders of magnitude (Table 5).

Apart from exceeding the WHO-value for *chemotoxic health risks* the consumption of meat from the WFS would also pose a serious *radiological* risk if the Hamman (2012) data are correct. Based on the maximum U-level in meat and consumption parameters by the German Radiation protection guideline as well as the ingestion dose model of the International Commission on Radioprotection (ICRP 72) used by the WISE age-depended dose calculator (WISE 2013), the effective radiation dose rate for all age groups caused by eating beef from the WFS area exceeds the international limit of 1mSv/a by up to a factor of 155, confirming the earlier model results of Barthel (2007) who predicted the highest exposure for youth (Table 6).

Table 6: Calculated effective dose rate associated with the consumption of meat containing 31810 µg/kg dry weight (dw) U (Hamman 2012) for different age groups

U.conc. meat: 31810 µg/kg dw

Age	meat consumption* [kg/a]	Dose rate ** [mSv/a]	[% IAEA limit]***
3 months	5	91.1	9110%
1 year	13	73.16	7316%
5 years	50	155.2	15520%
10 years	65	148.9	14890%
15 years	80	172.3	17230%
Adult	90	90.89	9089%

* StrlSchV 2001 (Germany)

*** 1 mSv/a

** calculated by Uranium Radiation Age-specific Dose Calculator (WISE 2013)

In terms of realistic exposure scenarios the sale of contaminated meat from the WFS farm to nearby markets is a concern especially in cases where customers habitually rely on the same type of meat for extended periods. While this is unlikely to be the case in large mass markets that use a multitude of suppliers, a risk may occur in local butcheries/shops selling such meat routinely, or where purchased directly of the affected farms.

While doubts exists that the U-data in Hamman (2012) are correct, the possibility cannot be excluded especially as uranyl carbonate complexes, assumed to form in the WFS, are highly soluble and more bioavailable than other U-species perhaps occurring elsewhere which could lead to higher rates of bio-accumulation. If the data are assumed to be correct than the consumption of meat from the WFS area poses an extremely high chemotoxic and radiological health risk which exceeds relevant guideline values for both types of risks by two orders of magnitude apart from being 8000 times above the average U-intake worldwide.

Yet, Hamman (2012) concludes that '*no toxic risk exists*' based on an assumed (age-independent) meat consumption rate of 0.13 kg per day. Apart from giving no detailed explanation how this consumption rate was determined (it is only half of the average consumption by adults and youths and more than 5 times of babies as

stipulated by NECSA 2007a) its application, for all age groups, result in exceeding acceptable levels of U-intake by orders of magnitude (Table 5)[5].

How such serious misjudgement of associated risks can occur in a thesis that, according to its author, was guided by a total of three supervisors, where one is the normal practice, is unclear. However, given the implications possibly wrong data and ill-informed conclusions and recommendations could potentially have for consumers as well as meat producers in the affected area, it is proposed that a review of the thesis takes place before any results, or findings are published. Unfortunately some information has, meanwhile, already been released to the news media further unsettling the general public and affected farmers alike (Tempelhoff 2012, SAPA 2012, Beega 2012).

Apart from consuming contaminated meat also drinking polluted drinking water is a potential exposure pathway for commercial farmers often relying on their own borehole water. In this regard the recent drastic increase of groundwater pollution in the area reported by E.J. Stoch to the DWA (personal communication 2013) is of major concern. Erasmus (personal communication 2013) identified a mine discharging pumped groundwater from a flooded neighbouring mine void as the likely cause of the recently detected groundwater pollution.

(d) Urban residents using municipal tap water

Many towns in the Witwatersrand goldfields, including the metropolitan area of Johannesburg and Ekurhuleni, receive their water from SA's largest water service provider, Rand Water (RW). This water is sourced from the Vaal Dam (located upstream of the gold mining areas) and augmented by water imported from Lesotho via the Lesotho-Highland Water Project (LHWP). For some mining towns in the West Rand Rand Water provides groundwater from the Zuurbekom compartment south of Johannesburg from where it is abstracted for over a hundred years.

However, there are also municipalities that source their drinking water from local streams and rivers which are potentially affected by mining-related U-pollution. This includes the Tlokwe Municipality (formerly known as Potchefstroom), Randfontein and Westonaria (supplied by water from the Zuurbekom pumping station of Rand Water) and the Midvaal water company supplying water sourced from the Vaal River to various towns in the Klerksdorp goldfield.

Table 7 lists U-levels observed in raw and tap water as well as sewage effluents of selected municipalities in South Africa and worldwide.

Table 7: Average and maximum U-concentrations reported for raw and tap water as well as sewage effluents of selected municipalities in South Africa and worldwide

[5] To add to the confusion, the meat consumption rate was recently changed by the student in the library copy from 0.13 kg/d to 0.38 kg/d, effectively tripling the daily U-intake. Apart from the fact that the associated U-intake now exceeds the applicable WHO-limit even more (in the worst case by close to 900 times), there was also no justification provided for this crucial change. Furthermore, this change was only made in the Conclusion section, while the original value of 0.13 kg/d remained unchanged in the remainder of the document including the section on risk assessment. The fact that the allowable meat consumption was suddenly raised by 300% without any corresponding changes in the preceding argument where the original value is still used for caclculating associated risks discredits the entire study.

Type of water	Location/name	Year	Ref.	U [µg/l]		
				n	av	max
Municipal water						
Potchefstroom (raw water)	Mooi River at Boskop dam outflow	2009-2012	Potch Munic (2012)	*165*	1.5	5.1
Potchefstroom (tap water)		1999-2000	Winde (2000)	*2*	5	6
		2009-2012	Potch Munic (2012)	*159*	1.3	5.5
Potchefstroom sewage effluent)			Potch Munic (2012)	*58*	1.3	3.6
Midvaal Water Company Klerksdorp (raw)	Vaal River at Vereeniging and Orkney	1990	Kronfeld & Vogel (1991)	*2*	7	11
	Vaal River at Koekemoerspruit mouth	1999	own data	*1*	5	
Rand Water - Randfontein (raw)	Zuurbekom dolomtic aquifer	1988	Dorling (2008)	*3*	26	44
Rand Water - Randfontein/ Westonaria (tap)		1988	Dorling (2008)	*4*	11	19
Rand Water - Randfnt/Weston. (sewage effluer	Flip Human	2002-2007	Dorling (2008)	*63*	9	29
Germany-wide survey of tap water	150 samples > 2µg/l	2008	Foodwatch e.V. (2008)	*8200*		8.5
German Environmental Survey	German water works	1990-1992	Becker et al. (1997)		0.3	48.4
Canada			von Sosten (2008)			780
Finland			von Sosten (2008)			1500
Britain, Italy , Germany	tap water		Werner et al. (1999)			40
Borehole water						
SA: N-Cape	Pofadder area	1996	Toens et al. (1997)	*10*	101	294

The data in Tab. 7 indicate that U-levels in tap water span a wide range even in developed countries such as Germany, Italy, UK, Finland and Canada with (isolated) maxima exceeding levels in South African tap water by far.

However, compared to the average of 0.3 µg/l U (despite including a maximum value of nearly 50 µg/l) indicates that, in general, levels of U in most tap water systems of Germany are lower than in South African municipalities affected by mining. Of 8200 German municipal tap waters surveyed in 2008 by Foodwatch e.V. only 150 exceeded the 'Baby-safe level' of 2 µg/l and showed a maximum of 8.5 ppb, which is only slightly higher than that of Potchefstroom and considerably below the maxima of Randfontein and Westonaria (Table 7).

The elevated U-levels in tap water of the latter two towns could perhaps be linked to the diffuse pollution of the Zuurbekom dolomitic compartments by stream loss from the WFS and other mining-polluted water such as tailings seepage found in earlier studies. Furthermore, the Western Area gold mine was obliged to recharge (potentially U-polluted) groundwater pumped from underground mine workings back into the compartment. And lastly, the U-enrichment plant of Nufcor is also located on the outcropping dolomite of the Zuurbekom compartment constituting a potential source of U-pollution. Whether or not this pollution was behind the recent decision of Rand Water to stop water abstraction from the Zuurbekom aquifer after more than a 100 years could not be established.

Uranium in sewage effluents and sludge

Monitoring data from the Potchefstroom municipality suggest that a link exists between U-concentration in tap water and U-levels in the municipal waste water (sewage effluent) (Fig. 6)

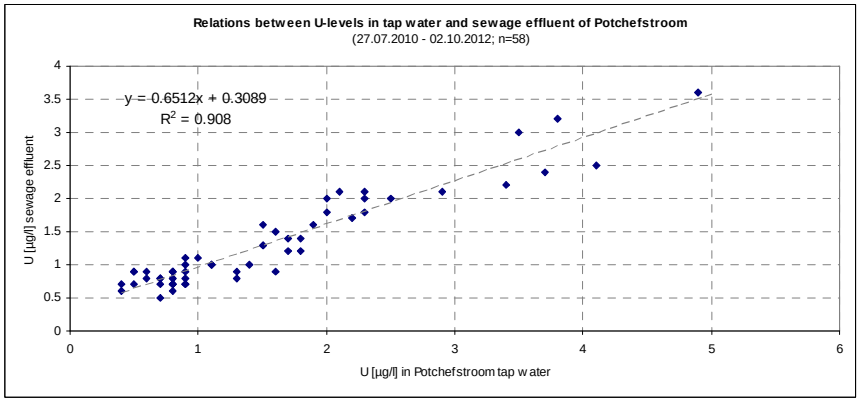

Fig. 6: Relationship between U-levels in tap water and sewage effluents from the municipal sewage works for a 17 months period (data source: Potchefstroom municipality 2012)

Since elevated U-levels in water are also likely to be reflected in the sewage sludge a WRC survey on the U-contents of sewage sludge of 78 industrial and domestic sewage plants across South Africa conducted between June 2001 and August 2003 may provide clues for possibly existing water pollution problems (Snyman et al. 2005). The survey indicates an average of nearly 5.2 ppm U (total conc.) ranging from 0.2 to 84.4 ppm and to 116 ppm for leachable U. (Snyman et al. 2005). Concentrations appeared to be generally higher in the wet summer season (Nov.-Febr. 2002) than in the dry winter period (June-Sep. 2001 and July –Aug. 2003) possibly indicating the impact of stormwater-related inputs flushing of tailings dust from impervious areas into the municipal waste water works of mining areas. The following U-levels are averages of the three sampling campaigns.

The highest U-levels by far (47.3 and 41.5 ppm U) occurred in 2 waste water treatment plants in Upington (Northern Cape) coinciding with the high levels of U in borehole water in that arid area (Toens et al. 1997, Wullschleger et al. 1998 , Sekoko et al. 2005, van Wyk and Coetzee 2006). With 6.9 ppm sewage sludge in Kakamas – another town in this arid region also showed above average U-levels as did Beaufort West (7.9 ppm) in the arid Karoo which is underlain by uraniferous sandstones (Snyman et al. 2005).

A second cluster of sewage works with elevated U-levels links towns in gold mining areas including Welkom (Free State goldfield: 14.7 – 20.8 ppm, n=3), Orkney (Klerksdorp goldfield: 14.9 ppm U) and Germiston (Central Rand goldfield: 20.2 ppm) (Snyman et al. 2005).

Less high but still above the median concentration of 2.4 ppm are U-levels in sewage works of Krugersdorp (now Mogala City; West Rand: 3.8-5 ppm), Johannesburg Olifantsvlei (Central Rand: 5.9 ppm) and Klerksdorp (Klerksdorp goldfield: 7.3 ppm). This compares to 3.7 ppm in sewage sludge of Potchefstroom measured in 2005 when U-levels in tap water were comparatively low (own data).

Generally the sewage sludge survey seems to indicate that many more municipalities have potential problems with elevated U-levels in their drinking water, with problems in towns of the arid Northern Cape being potentially more serious than in towns of gold mining areas.

The case of Potchefstroom

Being well aware of the potential problem, Potchefstroom City Council for the past 9 years or so runs a monitoring programme that not only analyses U-levels and other constituents, of tap water but also of selected raw water sources and Mooi River tributaries. However, given the extremely dynamic nature of fluvial U-levels discussed in detail by Winde (2006 a/b) the interval of one sample per week (even if taken from a number of different sites along the river) is unlikely to adequately capture possible short-term U-peaks moving downstream towards Potchefstroom in a matter of days, or hours.

The fact that average U-levels in calcite scales from water kettles in Potchefstroom were 20 times higher than in kettle scales from a nearby town indicates that U does indeed get into the tap water system. Since water kettles are commonly used for long periods of time several times a day for boiling tap water, the deposited calcite scales are a better indicator of long-term trace metal pollution than weekly water samples that only reflect water quality at the moment of sampling.

Of particular concern is the fact that U-levels in raw water sources of Potchefstroom recently started to show drastic increases (from 1 to >30 µg/l equalling a 30-fold rise) accompanied by a slightly delayed rise in tap water U-levels from <0.2 to over 5 µg/l equalling a >25-fold rise (Fig. 7).

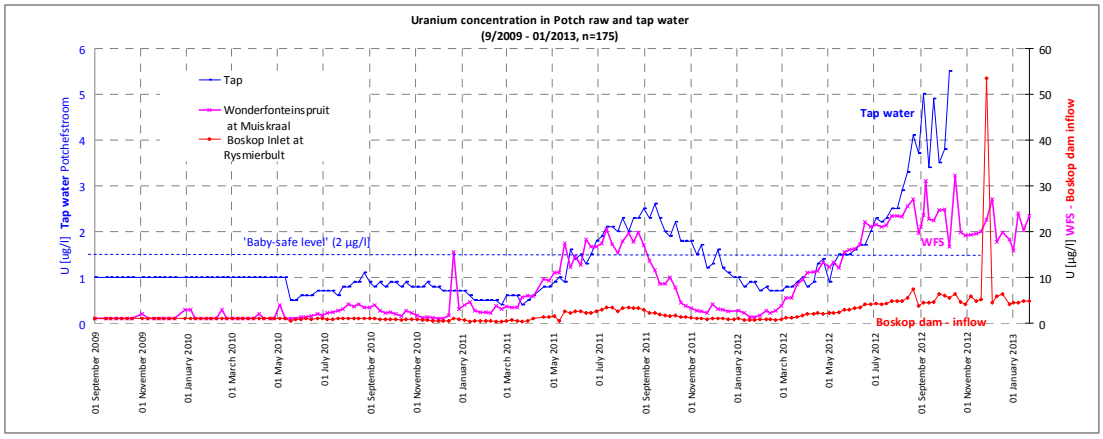

Fig. 7: U-concentration in raw water and tap water of the Potchefstroom municipality between Sep. 2009 and 22 Jan. 2013 indicating a cyclic increase as well as isolated U-peaks

The current levels of U in tap water are, however, not unprecedented as similar high levels were found by the author in 1999 and 2000 (4-6 µg/l U) when the WFS were still reaching the Mooi River on surface (Table 7). The period of low U–levels reported by the Potchefstroom municipality (2003-2010) coincides with the drying-up of the WFS well before it reaches the Mooi River owing to a sudden, and still unexplained, decrease in stream flow between October 2002 and April 2003 from 60 to 20 Ml/d (Winde 2011c, d). However, since winter 2010 the WFS is flowing again (EJ Stoch, personal communication 2012) discharging U-polluted water directly into the Mooi River and Boskop Dam from where Potchefstroom sources its drinking water. The impact of polluted surface water from the gold mining area is indicated by the cyclic rise in U-concentration reaching ever higher and longer lasting levels in dry

winter periods when no dilution takes place (Fig. 7). However, while dilution increases in the rainy season resulting in a levelling out of U-levels in the WFS, frequent thunderstorms during this time trigger isolated and short peaks of U resulting from highly polluted surface runoff. While the actual peak concentrations are likely to be missed most of the time by the fixed schedule monitoring programme employed (sampling takes place mostly in the mornings and early afternoons while thunderstorms generally occur in the late afternoon) on 22 November 2012 a peak concentration of 53 µg/l U was detected at the inflow of Boskop dam exceeding normal levels by a factor of 10.

The potentially associated risk for tap water users is compounded by the fact that the technology employed to purify the Tlokwe water is not able to remove significant amounts of U as the linear relationship with a gradient of nearly 1 between U-levels in raw and tap water illustrates (Fig. 8).

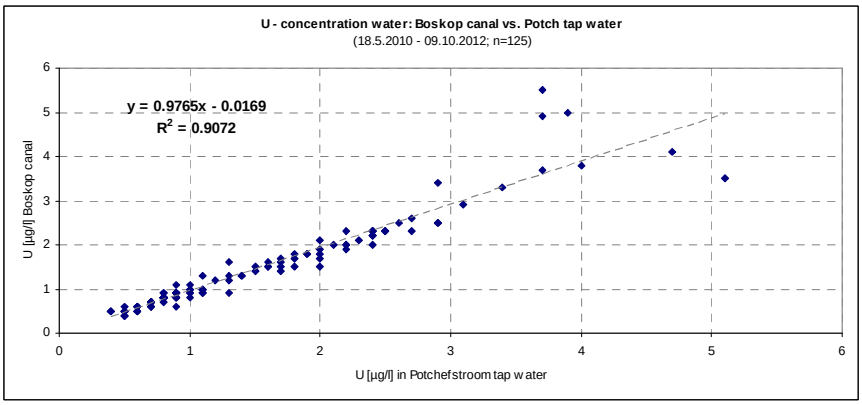

Fig. 8: Relationship between U-concentrations in **raw water** at the inlet of Potch water purification works ('Boskop canal') and the **tap water** of Potchefstroom for a 30-months period from May 2010 to October 2012 indicating that no U is removed by the water purification techniques (Data: Potchefstroom Municipality 2012)

Winde (2010b) pointed to this low removal efficiency at a time when U-levels in raw water were still low, based on findings that calcium, carbonate and bicarbonate ions, each reduce the removal rate of standard potabilisation processes (i.e. filtration, flocculation and chlorination) for dissolved U (Baeza et al. 2008). Since all three ions are abundantly present in dolomitic water used in Potchefstroom it is not surprising that the conventional treatment technologies applied by the local water works are unable to reduce U-levels. This, in turn, means that the U-peak of 20 November 2012 (53 µg/l), that was detected by chance, is likely to have caused an U-peak in tap water. In order to estimate the level of U-peak in tap water the long-term ratio between U-levels at the inflow and outflow of Boskop dam can be used (Fig. 9).

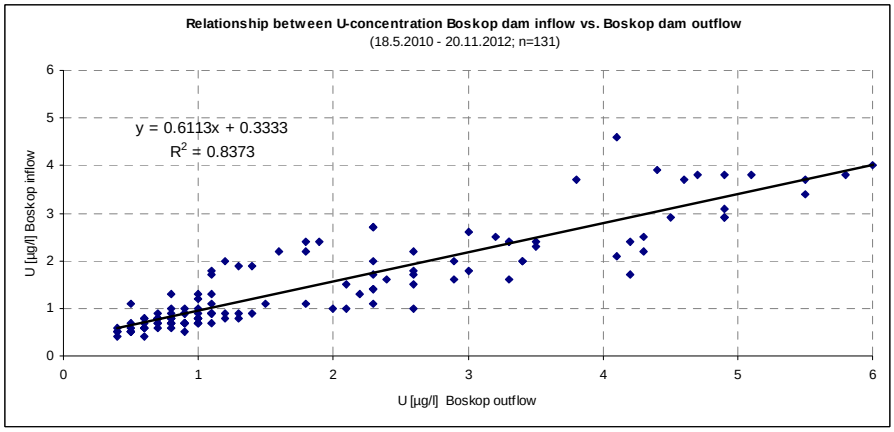

Fig. 9: Ratio of U-levels in water entering and leaving Boskop dam based on 131 samples covering a period of 2.5 years

As U is unlikely to be immobilised in the dam to any significant extent due to the formation of soluble carbonate complexes, the 39% decrease in concentration from inflow to outflow is probably due to dilution by unpolluted tributaries and less polluted dam water (Fig. 9). Assuming that the U-peak of 20 Nov 2012 was reduced by 39% the peak U-level in tap water is likely to have been around 21 µg/l as no further reduction occurs during the treatment of raw water (Fig. 8). While this exceeds the SANS limit of 15 µg/l it is probably an underestimate since the true maximum of the event was most likely missed. The event does, however, confirm the event-driven nature of pollution indicated by Winde (2002, 2006 and 2011d).

According to the long-term relationship between U-concentrations in the WFS and tap water in Potchefstroom, U-levels in tap water are on average 10 times lower than in the WFS due to dilution from the Mooi River and the Gerhard Minnebron eye as well as a few smaller tributaries (Fig. 10).

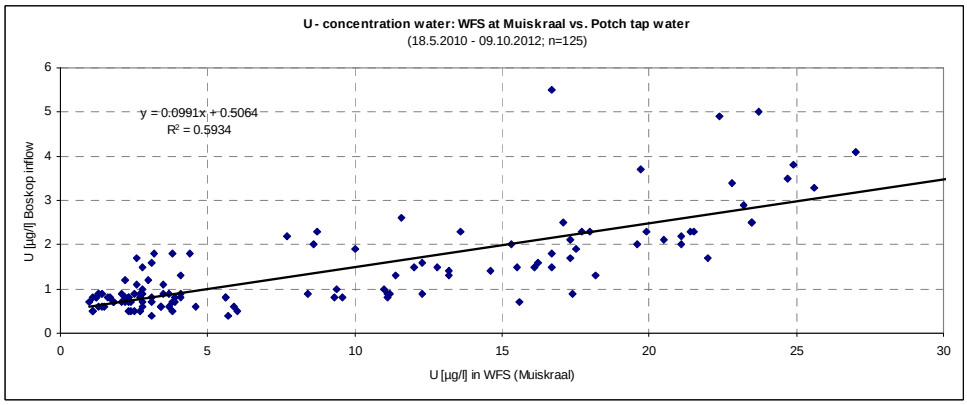

Fig. 10: Relationship between U-concentration in the WFS at Muiskraal and tap water in Potchefstroom based on 125 samples (raw data: Potchefstroom municipality 2013)

This dilution ratio suggest that the WFS supplied on average 10% of Potchefstroom's tap water over the monitored period. However, increasing deviations of recent data suggest that the contribution of the WFS has increased. The linear relationship between the U-levels of the last samples taken between May and October 2012

(thereafter no tap water data are available) suggests that the WFS contribution has meanwhile increased to 15%.

A further factor that, potentially, renders U-removal from dolomitic water particularly difficult is the possible formation of highly soluble uranyl-carbonate complexes. Owing to their extremely high solubility, such complexes are also readily bioavailable increasing associated health risks.

Responding to the extensive coverage of the issue in news media many concerned residents chose to use bottled water instead (at least those who are able to afford it), increasing sales and the number of businesses selling filtered tap water or mineral water sourced elsewhere.

The use of mineral water may, however, not in all case be a safe alternative as some geological formations tend to give off elevated U-levels into surrounding groundwater (e.g. Becker et al. 1997, Werner et al. 1999, Rosborg et al. 2005, Schnug et al. 2005, Anke et. Al 2007, UBA 2008, von Soosten 2008, Krachler and Shotyk 2009).

Generally it appears that U-levels in municipal tap water are still low compared to water used by informal residents but, in some instance, do exceed the original WHO limit of 2 µg/l which in Germany marks the 'baby-safe' level. Since the exposure appears to be not continuous but confined to short-term events, the overall exposure is comparably low. However, the high bioavailabity and toxicity of U complexes likely to form in dolomitic water may be a potential reason for concern.

In order to assess the extent to which consumption of drinking water has resulted in biological accumulation in local residents CANSA recently started the co-called 'tooth study' aiming at determining U-levels in teeth of residents (Winde 2011b).

Tab. 8 lists U-levels in biological samples from humans and animals in the WFS area and other polluted sites in South Africa as well as results of international studies.

Table 8: Uranium concentrations in biological samples from humans and animals including hair, bone, teeth and urine taken from U- polluted areas and uncontaminated control in South Africa and other countries.

Type of sample	Sampling area	Ref.		U [ppb]	
			n	min	max
Hair					
cows	lw WFS - Blaaubank farm	CANSA (2011)	5	349	1465
	lw WFS - Visser farm	Hamman (2012)	?	220	4630
	control site - Mooi River	Hamman (2012)	?	10	10
sheep	Pofadder area	own data	2	310	998
ground squirrel	lw WFS - Blaaubank farm	CANSA (2011)	1	549	
humans	lw WFS - Blaaubank farm residents	CANSA (2011)	3	385	500
	Carletonville area	CANSA (2011)	20	107	951
	control	CANSA (2011)	1	345	
	upper WFS	MicroTrace GmbH (2010)	1	70	
Bones					
cows	lw WFS - Visser farm	Hamman (2012)	?	410	1080
	lw WFS + Buffelsvlei	McCrindle (2008)	10	4	9
	control Mooi River	Hamman (2012)		10	10
frogs	Karoo, Ruitskul U-exploration site	Scholtz et al (2006)	13	82000 (av.)	
Teeth					
cows	lw WFS - Blaaubank farm	CANSA (2011)	1	895000	
sheep	Pofadder area	own data	1	260	
humans	Carletonville area	CANSA (2011)	2	1142	2115
	control SA	CANSA (2011)			
	Potch, milk tooth	own data	1	<10	
	Brazilian U-mining area	Prado et al. (2007)	41	10	55
Urine					
Humans	mine workers Deelkraal GM	Deelkraal GM (1995)	100		>50
	Germany	UBA (2005)	1518	0.0008	0.3

(? – number of samples not specified)

The results listed in Table 8 indicate that a number of mostly grab-type of samples was taken in the WFS as well as in other areas of South Africa. The perhaps most striking differences to international data pertains to U-levels in human teeth where CANSA (2011) reports up to 100 times higher U-levels in two grab samples than found in a large-scale study in a Brazilian U-mining area (Prado et al. 2007) (Table 8).

The U-content of a tooth from an ‘old’ cow from the lower WFS (CANSA 2011) is over three orders of magnitude higher than that of almost all other samples including a tooth of 7-old sheep exposed to polluted water of the Pofadder area. The next highest U-value was measured in bone of frog from a highly polluted water-filled open pit of an abandoned U-exploration site in the Karoo (Scholtz et al. 2006) (Table 8). Since both samples appear to be far above values reported from similar contaminated sites the possibility of a unit-reporting error cannot be excluded.

In any event, U-levels in biological samples only allow assessing the extent to which U accumulated in exposed organisms without indicating a manifest health problem. For detecting the latter dedicated epidemiological studies are required. Apart from an earlier attempt to survey the health of people in the lower WFS catchment by the Potchefstroom municipality sampling blood and urine from 59 residents aged between 6 and 72 years that unfortunately failed to include U in the analyses (Potchefstroom municipality 2002), there is, to the author’s knowledge, only one published study in SA that used epidemiological data for assessing health risks associated with the consumption of U-polluted drinking water namely the earlier mentioned Pofadder study (Toens et al. 1997).

Case study 2: Natural U-pollution in the Northern Cape

Initiated and promoted by local medical doctors as well as a doctor from the Unit for Communal Health at Stellenbosch University who noticed that many of the leukaemia patients came from this particular area, in 1995, a study was initiated aimed to investigate a possible link between the quality of borehole water and the concentrated occurrence of leukaemia on farms in and around Pofadder. The report was finally published in 1997 indicating a significant geo-spatial correlation between abnormally high counts of lymphocytes (as indicator for leukaemia) and the concentration of dissolved U in the often very salty borehole water consumed on the farms. The findings were based on two independently generated data sets. The water quality data for 126 boreholes (including U-concentration data) came from a geological survey of the Atomic Energy Commission (AEC) as part of their U-exploration programme in the early 1980s while the haematological data were generated during a communal health survey that sampled blood from over 600 local residents from 120 localities (of which finally only 418 were used as the balance did not drink borehole water) (Toens et al. 1997). The geo-spatial approach to correlating the above discussed parameters as employed by Toens et al. (1997) is illustrated in Fig. 11.

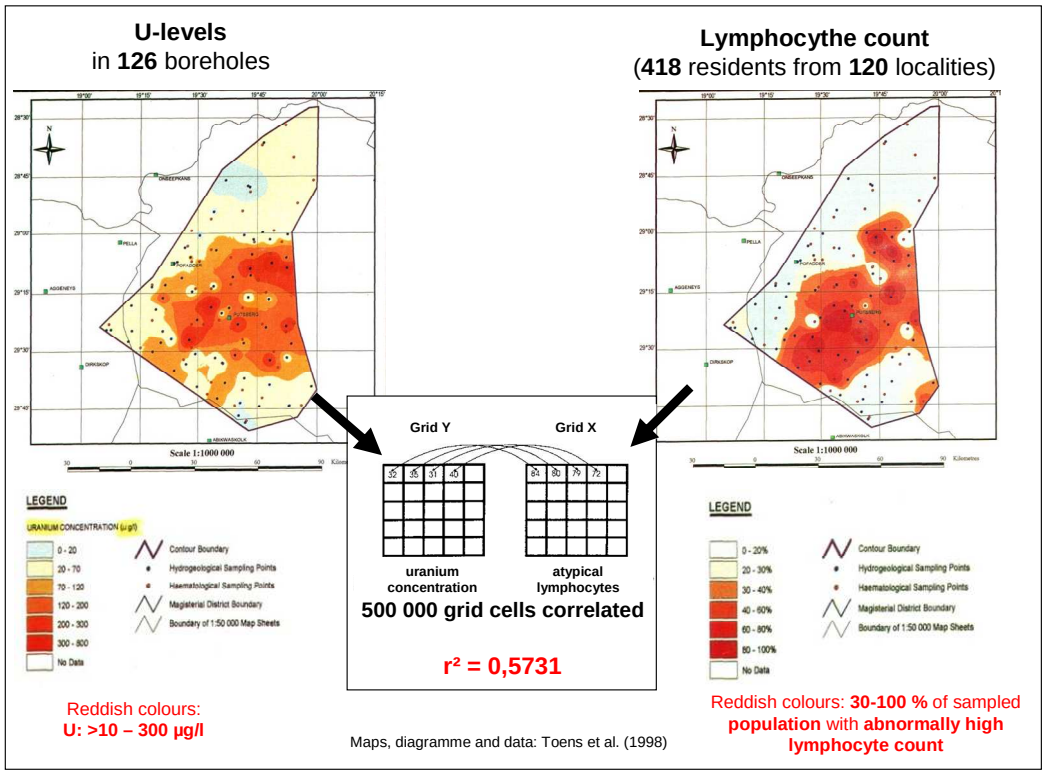

Fig. 11: GIS-based spatial correlation of the percentage of residents with atypical lymphocyte counts and the U concentration in borehole water according to Toens et al. (1997) (Winde 2011a)

Given the error margins associated with using secondary and partly aggregated data, generated at different times for different purposes, the uncertainty of trace analyses, and geographical inaccuracies as well as with the employed GIS interpolation techniques etc. it is rather amazing that a geospatial correlation between the percentage of people suffering from abnormal lymphocyte counts and the U-

concentration in boreholes was detected. The only other water quality parameter that showed a higher correlation than U was the combined U-As concentration. A follow-up study by the Council for Geoscience established, however, that As was wrongly determined at the time while confirming the correctness of the U-measurements (Van Wyk & Coetzee 2006). The correctness of the U-data was also confirmed by Wullschleger et al. (1998) and Sekoko et al. (2005) indicating that U-pollution of borehole water is not confined to the Pofadder area but affects large parts of the Northern Cape Province.

Since Toens et al. (1997) relied almost entirely on secondary data no other possible pathways of U-exposure than drinking polluted borehole water were explored. In order to assess exposure via the inhalation of dust, handling of agrochemicals and the ingestion of mutton produced on these farms, the Mine Water Research Group (MWRG), in July 2012, initiated a follow-up study in collaboration with the Institute for Radiochemistry in Dresden-Rossendorf (Germany) taking samples from the various media of concern, which are currently in the process of being analysed in Germany.

Since U in groundwater originates from uraniferous host rock from where it leaches into the aquifers (in the Pofadder area this is mainly gneiss) similar water pollution problems in other uraniferous rock provinces in SA cannot be excluded. This is particular true for large areas in the semi-desert Karoo region which are underlain by U-containing rock (Beaufort Group) that were even mined at small scale (Scholtz et al. 2006) (Fig. 12)

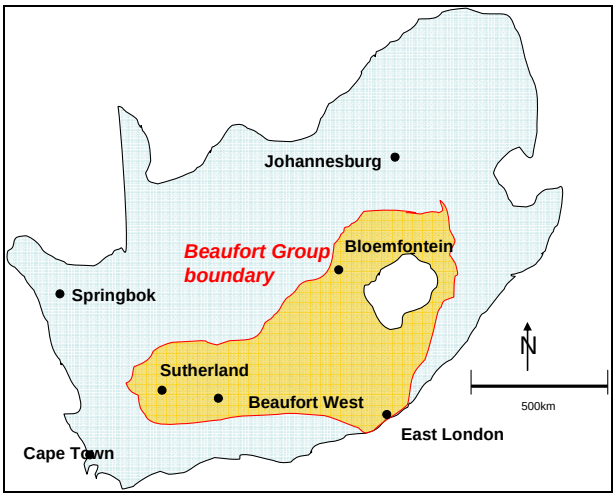

Fig. 12: Occurrence of the uraniferous Beaufort group in South Africa (based on map of Africa Uranium Ltd. www.africauranium.com, 29 Jan. 2013)

Owing to the low population density in this semi-desert area and the absence of a central reporting point like Stellenbosch in the Pofadder case, existing health problems may simply be more difficult to detect especially in the absence of aggregated medical records for the rural population. It is therefore suggested that the potentially affected areas be mapped and a reconnaissance study be conducted into the U-pollution of borehole water and possible associated health problems of the farming community.

6. Summary and conclusions

Although uranium is a common natural constituent of the earth crusts, it is generally toxic to humans and displays no beneficial properties like the other, so-called, essential heavy metals such as zinc, copper, or iron. Exposure to U can cause serious health damage to kidneys, the brain and many other vital systems in the human body at relatively low concentrations.

While mining and production of U-concentrate partly removes the radioactive element from a certain area (especially where shallow ore deposits are mined) it also often brings U from a 'safe' geological underground many hundreds or thousands of meter below surface into the biosphere and dramatically increases its environmental mobility and the associated exposure of local residents.

Even though uranium, in SA, was only produced as a by-product of deep-level gold mining, the associated hazards are now exceeding those in dedicated U-mining areas worldwide. This is caused by a range of factors creating *a significant pollution potential*, *promoting water pollution* and resulting in *above-average exposure* of an often very *vulnerable population.* These factors are summarised in the following:

(I) Factors resulting in a high pollution potential (source term)

1. Although gold ore contains relatively low U-levels compared to high-grade ore mined elsewhere uranium was, for most of the time in the history of gold mining, *left at its original ore-concentration* in the tailings resulting in U-levels of average gold tailings that are comparable to genuine U-tailings in areas such as the Wismut region, where over 90% of the U were leached from the ore.

2. Gold mining produced *exceptionally high volumes of tailings* owing to the typically *low concentration of gold* necessitating the mining of large volumes of rock and the *long period of time*, deep level gold mining operations have taken place in the Witwatersrand basin as the world's largest known gold deposit. In over a quarter and a century SA gold mines produced well over 6000 million t of uraniferous tailings (Wymer 1999). This is nearly 20 times more than in the USA and Canada combined, as the two largest U-producers (based on data in Waggit 1994). Compared to the annual global production of U tailings of some 20 million t in 1992 (IAEA cited in Waggit 1994) the amount of gold tailings deposited in the Witwatersrand basin is equivalent to 300 years of global production. Since large-scale U-production only commenced some 60 years ago (and then at considerably lower rates) it can be safely assumed that over five times more uraniferous tailings are deposited in the goldfields of South Africa than U-tailings worldwide.

(II) Factors aiding easy water pollution

1. The semi-arid climate in the Witwatersrand region allowed for low-cost tailings disposal in the form of uncovered slimes dams resulting in significant wind and water erosion. This is in contrast to mines in humid climates where tailings are commonly contained in water-covered storage facilities.

2. Arguing that the strongly negative climatic water balance will result in more water evaporating than infiltrating, slimes dams in SA were generally not lined (also

for cost reasons) promoting the outflow of highly contaminated seepage into nearby water resources.

3. An extended dry season and generally high evaporation rates allow for the formation of extensive salt crusts on tailings that concentrate U to high levels. Being instantly dissolved by rainfall these crusts are a major source of rapid stream pollution via stormwater runoff.

4. In dolomitic areas, U-migration into the aquatic environment was further accelerated by placing unlined slimes dams on cavernous ground and by injecting tailings directly or indirectly into the underlying karst aquifer at large scale.

5. Being generally located in water-scarce areas polluted rivers and streams have a relatively low dilution capacity compared to water-rich mining areas elsewhere resulting in relatively high U levels in receiving streams compared to humid areas. This also applies to the arid farming areas of the southern Kalahari and the Karoo where very low rates of groundwater recharge allow for significant concentrations of U in the slow moving groundwater as a result of ongoing leaching from the uraniferous host rocks.

6. Often taking place near the outcrop of watershed-defining ore bodies, gold mining in the Witwatersrand frequently affects the small headwater catchments of streams, further reducing the potential for diluting pollution loads in affected streams.

7. Generally, the chemical mobility of U in mining-polluted streams is relatively high. In non-dolomitic streams this is due to the low pH-values keeping rates of natural immobilisation low while in dolomitic streams the formation of highly soluble complexes reduces natural U-removal by co-precipitation.

8. The chemical composition of dolomite drastically lowers the U-removal efficiency of standard water purification techniques allowing possible U-peaks in raw water to affect tap water quality unabated.

(III) Factors increasing U-exposure of the population

1. Owing to their enormous volume, tailings affect an unusually large surface area of some 400 km².

2. In contrast to the major U-producers like Canada, the USA and Australia, where U-mining takes place in remote barely populated regions, the gold mining areas of the Witwatersrand are among the most densely populated in SA attracting migrant labour and rural poor in search for jobs. The associated uncontrolled growth of informal settlements often encroaching right next to the many mine dumps, or even onto them, results in an unusual large number of people being directly exposed via exceptionally short pathways to toxic mining waste. It was estimated that in metropolitan areas of Gauteng alone some 1.6 million people live next to slimes dams.

3. Windblown dust and contaminated seepage from uncovered and unlined tailings dams often directly affect nearby residents who live next to – and in some cases even on top of – slimes dams.

4. Many informal settlers have no access to clean water and use untreated polluted stream and mine water for domestic purposes (including drinking, gardening etc).

5. This is frequently exacerbated by additional exposure via ingesting contaminated food (grown with polluted water), inhalation of tailings dust and radon, as well as geophagy, pica behaviour – intentional and unintentional ingestion of contaminated soil by pregnant woman and children (hand-to-mouth contact) – who are the most exposed and vulnerable groups

6. A peculiarity in South African townships may be the ingestions of 'slimes cakes' as 'traditional' medicine and the application of tailings onto the skin as acne treatment.

7. Lack of awareness and education and more pressing problems arising form the wide-spread poverty affecting informal settlers often result in an indifference to pollution-related risks.

8. Apart from informal settlers, there are also other user groups affected who live in formal suburbs and towns. This includes whole mining communities, such as the Blyvooruitzicht village, which was supplied with pumped mine water, but also municipalities whose raw water sources are affected by U-pollution.

9. These residents are commonly very well informed by media on health threats associated with U-pollution and take precautionary measures to reduce exposure including the installation of household filters, buying of bottled water, changing water supply to borehole water etc.

10. In such communities isolated cases of exaggerated associated risks, in order to substantiate compensation, claims from the mines also occurred. This is counteracted by parties who stand to lose out on spreading rumours of water pollution and tend to downplay the matter (including City Councils). This often results in a highly emotional and antagonistic public debate exacerbated by contradicting media reports, which appear to have a general bias towards the coverage of these problems compared to the more serious ones in informal settings.

11. In situations where no one is to blame for the naturally occurring pollution and no real alternative water supply options exists – like in the arid sheep farming area of the Northern Cape, affected farmers tend to deny the existence of any health problems looking for alternative explanation for the many cancers affecting family members and neighbours.

(IV) Above-average vulnerability of exposed population

1. Among the affected population, mainly poor black people living in urban and rural informal settlements are the most exposed group displaying the highest vulnerability. This results in the unfortunate situation that the population with the lowest capacity to handle additional toxic stress is the very same one that is by far the most exposed to it.

2. In terms of exposed population groups other than informal settlers U-ingestion via drinking water and consumption of contaminated food – particular meat from cattle and sheep are main exposure pathways.

3. Since the 'baby-safe' level of U in drinking water is frequently exceeded in tap water of affected municipalities babies and infants are at higher risks than adults. This may also apply to pregnant women. This is somewhat different for the consumption of contaminated meat where the youth may be at highest risks owing to similar consumption patterns like adults but generally lower body weights.

4. In sheep farming areas like the Northern Cape where mutton is a staple food and include the consumption of inner organs such as kidneys and liver known to accumulate U, this exposure pathway may be as significant as the consumption of polluted borehole water.

7. Recommendations

As a short-term response to the established extent of U-exposure in mining areas as well as in certain geological provinces it is recommended that the actual health burden resulting from exposure to mining-related as well as natural U-pollution be investigated through high-confidence epidemiological studies.

As a consequence of the adverse effects U-pollution has on humans and many of the major U-producers such as Germany, Canada, Australia, the USA, France dedicated research centres and institutes aimed at scientifically supporting the systematic remediation of mining legacy sites within large governmental programmes such as the UMTRA project in the USA and the German environmental rehabilitation programme of the Wismut have been established.

However, despite the large-scale problems associated with U in South African gold mining areas there is still no dedicated institute able to, scientifically, address the matter. So far the problem has been left to isolated enthusiastic scientists and NGO's largely lacking governmental support. It is therefore recommended that the establishment of similar institute in South Africa focussing on the rehabilitation of polluted mining sites and mitigation of associated health risks be explored.

Given the current problems arising from uncoordinated and unprepared mine closures in the West, Central and East Rand and their dire environmental consequences, the brief of such institute should also include the development of a pro-active integrated strategy to prepare for closure of the many mines which are still active. This is of particular importance as many mines operate in dolomitic areas with the possible of risk of polluting large water resources for generations to come if the closure process is not properly managed. Experience in the past clearly show that tasks of national importance cannot be left to a heterogeneous industry with competing players who espouse different environmental ethics. This would also include the need to proactively combat the rapid loss of institutional memory affecting Governmental departments and the mining industry alike through collection and archiving of relevant data, documents and information in a central data base. The linking of the

institute to similar institutions worldwide and creating virtual networks of international experts would support local expertise and capacity building while keeping staff requirements at reasonable levels.

As one of the most important mining nations worldwide in which mining – active and inactive - will continue to play a significant role for the foreseeable future, SA would be well advised to establish a dedicated institute that scientifically and in a coordinated and systematic manner addresses the pressing issues of mining-related health risks and water security which already affect many, often volatile, communities.

8. References

Anke M, Seeber O, Müller R, Schäfer U, Zerull J (2007): Uranium transfer in the food chain from soil to plants, animals and man. Chemie der Erde/ Geochemistry, 69, 75-90.

Anonymous (2010a): Yes, you can drink it. Potchefstroom Herald, 1 October 2010, p.3.

Anonymous (2010b): Drinkwater veilig, inwoners nie oortuig. (*Drinking water safe, residents not convinced*.) NorthWest Independent, 2 February, front page.

ATSDR (Agency for Toxic Substances and Diseases Registry) (2001): Toxicological profile for Uranium – September 1999. http:// www.atsdr.cdc.gov, pp. 398

AVNI J (2007) Death in the water. Millions of people drink and bathe in poisoned water flowing from the mines. *Sowetan 24 July 2007*, Cover page and p.4.

Baeza A, Salas A, Legarda F (2008) Determining factors in the elimination of uranium and radium from groundwaters during a standard potabilization process. *Sci. Total Environ.* **406** 24-34.

Bain CAR, Schoonbee HJ, de Wet LPD, Hancke JJ (1994): Investigation into the concentration ratios of selected radionuclides in aquatic ecosystems affected by mine drainage effluents with reference to the study of potential pathways to man. WRC report no. 313/1/94, Pretoria, 108 pp.

Barthel R (2007) Assessment of the Radiological Impact of the Mine Water Discharges to Members of the Public Living around Wonderfonteinspruit Catchment Area. BSA Project No. 0607-03, BS Associates, Consulting Engineers and Scientists. Report to the National Nuclear Regulator (NNR), Contract No. RRD/RP01/2006, Bedfordview, pp. 225, unpublished

Barthel R, Deissmann G, Leotwane W (2007): Radiological impact assessment of mining activities in the Wonderfonteinspruit catchment area. Accepted abstract, Environmin Conference, Pilanesbertg, July 2007, South Africa.

Barthel R (2011): Radiological impact assessment of mining activities in the Wonderfonteinspruit catchment area, South Africa. In: Merkel BJ, Schipek M [eds.]: The new uranium mining boom. DOI 10.1007/978-3-642-22122-4, Springer Berlin-Heidelberg, 517-528.

Becker K, Müssig-Zufika M, Hoffmann K, Krause C, Meyer E, Nöllke P, Schulz C, Seiwert M (1997): Umwelt-Survey 1990/92. Band IV: Trinkwasser. Deskription der Spurenelementgehalte im Haushalts- und Wasserwerks-Trinkwasser der Bevölkerung in Deutschland. WaBoLu-Heft, 5/97, Umweltbundesamt, Berlin, Eigenverlag.

Beega S (2008a): Living in fear of a toxic tsunami. Far West Rand residents claim poisoning. Saturday Star, 12 April, full page article.

Beega S (2008b): Pollution problem played down. Saturday Star, 27 Sep.

Beega S (2011): Residents use radioactive mud as an acne cure. Experts warn old mine dumps could cause birth defects and brain disorders. Saturday Star, 12 November, p. 8.

Beega S (2012a): Teeth hold the clues to Potchefstroom's future. Saturday Star, 22 December, p. 4.

Beega (2012b): Toxic mining metals in cattle. Study finds high concentrations of elements in grass, water and livestock in main area for gold. Saturday Star, 15 December 2012

Bertell R (1999): Conflict of interest between IAEA and WHO. WISE News Communique on November 19, www.antenna-wise.nl, accessed 9 Feb. 2009

BfR (Bundesanstalt für Risikobewertung), BfS (Bundesanstalt für Strahlenschutz) (2006): Gemeinsame Stellungnahme Nr. 014/2006 des BfS und des BfR vom 16 Januar 2006. BfR korrigiert Höchstmengenempfehlung für Uran in Wässern zur Zubereitung von Säuglingsnahrung. (Joint statement Nr. 014/2006 of the BfS and the BfR dated 16 January 2006: BfR corrects recommendation regarding highest permissible uranium quantities in water used for preparing baby food). Bundesanstalt für Risikobewertung, Berlin, Germany.

Botha C (2008): Toxic shock. Onions from Twitcher's Eden were tested by Necsa and found to contain levels of radioactivity above acceptable levels. Potchefstroom Herald, 8 February, p.3

Bryson B (2010): At home – a short history of private life. ISBN 978-0-552-77255-6, Transworld Publishers, London, p. 514-516.

CANSA (Cancer Association of South Africa) (2010): Analytical results for U in two sweet potatoes and maize from the Wonderfonteinspruit catchment, by C. Albrecht, 1.p, unpublished.

CANSA (Cancer Association of South Africa) (2011a): Analytical results for U in hair and teeth of humans, cattle and other animals sampled at the farm Blauubank at the lower WFS, by C. Albrecht, unpublished.

CANSA (Cancer Association of South Africa) (2011b): Analytical results for U in water, soil, tailings, gutter sediment, vacuum cleaner dust, geyser scales, peat, vegetables, teeth of humans, and other materials from the lower Wonderfonteinspruit and Gerhard Minnebron wetland areas, by C. Albrecht, unpublished.

CCME (Canadian Council of Ministers of the Environment) (2007). Canadian soil quality guidelines for uranium: environmental and human health. Scientific supporting document. Winnipeg; 2007 http://www.ccme.ca/assets/pdf/uranium_ssd_soil_1.2.pdf.

Coetzee H, Winde F and Wade P (2006): An Assessment of Sources, Pathways, Mechanisms and Risks of Current and Potential Future Pollution of Water and Sediments in Gold Mining Areas of the Wonderfonteinspruit Catchment (Gauteng/ North West Province, South Africa). WRC Report No. 1214/1/06**.** Water Research Commission, Pretoria, South Africa. 202 pp.

COMRO (Chamber of Mines Research Organisation) (1990): Untitled sheet with analytical results for samples taken between 26.11.1990 and 20.6.1991 by Deelkraal GM and COMRO. 11 pp. (Unpublished).

COMRO (Chamber of Mines Research Organisation) (1991): Unpublished COMRO Underground Environment Water Laboratory Analytical Report: 9 samples submitted by A McLaren on 12 December 1991. 1 p.

Deelkraal Gold Mine (post-1990)*:* Pilot study to determine uranium levels in urine of underground mine workers at Deelkraal Gold Mine. pp.2, unpublished

Du Toit JS (2007): Background report on communities risk within Mogale City Local Municipality affected by mining-related activities, with special reference to radiation & toxicity. September, unpublished, 83 pp.

DWAF (1993): South African Water Quality Guidelines. Volume 1: Domestic Water Use. First edition, Pretoria

DWAF (1996a): South African Water Quality Guidelines. Volume 1: Domestic Water Use. Second edition, Pretoria

DWAF (1996b): South African Water Quality Guidelines. Volume 4: Agricultural use: Irrigation. Second edition, Pretoria, http://www.orangesenqurak.org/UserFiles/File/National%20Water%20Departments/DWEA-DWAF/MiddleVaalISPJul04.pdf

DWAF (2004): Internal strategic perspective for the Middle Vaal water management area (WMA no 9). Directorate National Water Resource Planning, DWAF report no. P WMA 09/000/00/0304, DWAF, Pretoria,

Engelbrecht P (2008): Leuens, se boere (*Lies, say farmers*). Potchefstroom Herald, 8 February, p.2

FAANHOF A, VAN VEELEN M and PULLES W (1995) Radioactivity in water sources: a preliminary survey. DWAF Report No. N/0000/00/REQ/0695. Department of Water Affairs and Forestry, Pretoria.

FSE (Federation for a sustainable environment) (2012): Tour of the West Rand Goldfields, 12 November 2012, Agenda, prepared for visitors of the Heinrich Böll Stiftung Germany, 17 pp., unpublished.

Fesenko S, Isamov N, Howard BJ, Voigt G, Beresford NA, Sanzharova N (2007a): Review of Russian language studies on radionuclide behaviour in agricultural animals: part 1. Gut absorption. Journal of Env. Radioactivity, 98, 85-103.

Fesenko S, Howard BJ, Isamov N, Voigt G, Beresford NA, Sanzharova N, Barnett CL (2007b): Review of Russian language studies on radionuclide behaviour in agricultural animals: part 2. Transfer to milk. Journal of Env. Radioactivity, 98, 104-136.

Foodwatch e.V. (2008): Uranbelastung von Trinkwasser in Deutschland (Messung FAL-PB August bis November 2006, Stand 27.3.2008). Analytical results, pp. 10

Fisenne, I.M., P.M. Perry, K.M. Decker, H.K. Keller (1987): The daily intake of 234,235,238U, 228,230,232Th, and 226,228Ra by New York City residents. Health Phys 53:357-363.

UBA (German Federal Environment Agency) (2005): Uranium and Human Biomonitoring. Opinion of the Human Biomonitoring Commission of the German Federal Environment Agency. 13 pp. (in English) published in: Bundesgesundheitsblatt – Gesundheitsforschung – Gesundheitsschutz 48 (7), (2005), 822-827 (in German)

Gilman AP, Villeneuve DC, Secours VE, Yagminas AP, Tracy BL, Quinn JM, Valli VE, Willes RJ, Moss MA (1998 a): Uranyl Nitrate: 28 day and 91 day toxicity studies in the Sprague Dawley Rat. *Toxicol Sci*, **41**, 117-128

Gilman AP, Moss MA, Villeneuve DC, Secours VE, Yagminas AP, Tracy BL, Quinn JM, Long J, Valli VE, Willes RJ (1998 b): Uranyl Nitrate: 91 day exposure and recovery studies in the male New Zealand white rabit. *Toxicol Sci*, **41**, 138-151

Hamman D (2012): A holistic view on the impact of gold and uranium mining on the Wonderfonteinspruit. Dissertation for the degree Master of Science in Environmental Science, North-West University, Potchefstroom campus, Faculty of Natural Sciences, April 2012, 147 pp., unpublished.

Harley JH (1988): Naturally occurring sources of radioactive contamination. Radionuclides in the food chain. In: Carter MW [ed.] International life sciences Institute monographs. Heidelberg: Springer-Verlag, p. 58-71

Health Canada (2001): Guideline for Canadian Drinking Water Quality: supporting documentation – Uranium, pp. 10.

Henner P (2008): Bioaccumulation of radionuclides and induced biological effects in situations of chronic exposure of ecosystems - an uranium case study. In: De Kok LJ, Schnug E [eds]: Loads and fate of fertilizer derived uranium. Backhuys Publishers Leiden, The Netherlands

Iliso (Consulting Ltd.) (2008) Wonderfonteinspruit catchment area remediation plan. Report to Department of Water Affairs and Forestry and National Nuclear Regulator, 1st draft, November 2008.

IRSN (Institute de Radioprotection et de surete nucleaire) (2005): Envirhom: Bioaccumulation of radionuclids in situations of chronic exposure of ecosystems and members of the public. Progress report 2 covering the period June 2003 – September 2005. Report DRPH 2005-07 & DEI 2005-05. pp. 64

IWQS (Institute for Water Quality Studies) (1999): Report on the radioactivity monitoring programme in the Mooi River (Wonderfonteinspruit) catchment. Report no N/C200/00/RPQ/2399, April 1999, Department of Water Affairs and Forestry, Pretoria, pp. 26, plus appendices, unpublished.

Jacobi W, Roth P, Noßke D (1997): Mögliches Risiko und Verursachungs-Wahrscheinlichkeit von Knochen- und Leberkrebs durch die berufliche Alphastrahlen-Exposition von Beschäftigten der ehemaligen WISMUT AG [Possible risk and probability of causation of bone and liver cancer due to the occupational alpha ray exposure of workers at the previous WISMUT Uranium Mining Company], 57 pages in German, Forschungsbericht, GSF- Forschungszentrum für Umwelt und Gesundheit, Oberschleißheim, July 1997

Jordaan JM, Enslin JF, Kriel JP, Havemann AR, Kent LE, Cable WH (1960): Final Report of the Interdepartmental Committee on Dolomitic Mine Water: Far West Rand. DWA report. DWA, Pretoria, unpublished, pp. 22,000 incl. supporting information.

Jordan B (2009): Toxic water poses health nightmare. Sunday Times, 19 July.

Kempster PL, Smith R (1985): Proposed aesthetic/ physical and inorganic drinking water criteria for the Republic of South Africa. CSIR Research Report No. 628. National Institute for Water Research, CSAIR, South Africa.

Kempster PL, van Vliet HR, Looser U, Parker I, Silberbauer MJ, du Toit P (1996): Overview of radioactivity in water sources: uranium, radium and thorium. Final report. IWQS-No: N/0000/00/RPQ/0196. Pretoria

Konietzka R, Dieter HH, Voss J-U (2005): Vorschlag für einen gesundheitlichen Leitwert für Uran im Trinkwasser. *Umweltmed Forsch Prax*, **10** (2), 133-143

Krachler M, Shotyk W (2009): Trace and ultratrace metals in bottled waters: Survey of sources worldwide and comparison with refillable metal bottles. *Sci Total Environ*, 407, 1089-1096

Kronfeld J, Vogel JC (1991): Uranium isotopes in surface waters from southern Africa. Eart and Planetary Science Letters, 105, 191-195.

Labuschagne C (2008): Investigation of the acid mine drainage potential of the Kopanang rock dump, Vaal Reefs. Mini-dissertation, North-West University, Potchefstroom Campus (South Africa), pp. 54, unpublished

Liefferink M (2008): Oral submission on behalf of the Federation of sustainable Environment to the Parliamentary Portfolio Committee on Water Management and Pollution, 4 June 2008.

Louw A (2009a): 'There is no reason for panic'. Mine pollution is possibly decreasing. Carletonville Herald, 17 July, p.5.

Louw A (2009b): Wonderfontein cleanup plan 'seriously flawed'. Carletonville Herald, 14 August, p.5.

Marsden DD (1986): The current limited impact of Witwatersrand gold-mine residues on water pollution in the Vaal River system. J S Afr Inst Min etall, 86, 12, 481-504

Martins M (2008): Moet inwoners maskers dra? Carletonville Herald, 23 May, p. 11.

McCrindle (2008): Preliminary results from analysing water, sediments, milk and cattle tissue for uranium and other metals in the Wonderfonteinspruit catchment. 45 tissues samples,

McDiarmid MA, Keogh JP, Hooper FJ, McPhaul K, Squibb K, Kane R, DiPinoR, Kabat M, Kaup B, Anderson L, Hoover D, Brown L, Hamilton M, Jacobson-Kram D, Burrows B, Walsh M (2002): Health Effects of Depleted Uranium on Exposed Gulf War Veterans. *Environm Research*, 82, 2, 168 - 180

Metago (Environmental Engineers Pty. Ltd.) (2003): The assessment of the impacts on groundwater quality associated with the backfilling of dolomitic cavities with gold mine tailings. Final report to the Water Research commission (WRC), WRC proj. nr. 115-002, pp. 110, Pretoria, unpublished

Milacic S, Petrovic D, Jovicic D, Kovacevic R, Simic J (2004): Examination of the health status of populations from depleted-uranium-contaminated regions Examination of the health status of populations from depleted-uranium-contaminated regions. *Environm Research*, **95**, 1, 2-10

Miaullis AP (2012): Uranium contamination values and limits. Master of Science thesis, Colorado State University, Fort Collins, Colorado, USA, pp. 98.

Neves MO, Figueiredo VR, Abreu MM (2011): Transfer of U, Al and Mn in the water–soil–plant (*Solanum tuberosum L.)* system near a former uranium mining area (Cunha Baixa, Portugal) and implications to human health. Science of the Total Environment, 416, 156-163.

NECSA (Nuclear Energy Corporation of South Africa) (2003): Report of radioactivity analysis of fish from Deelkraal recreational dam. Job code RA 05295, 4. Dec 2003, for Harmony Randfontein Operations, 4 pp. unpublished.

NECSA (Nuclear Energy Corporation of South Africa) (2007a): Report of radioactivity analysis of fish and chicken. Job code RA 08190, 28 Aug. 2007, for Driefontein GM, 4 pp. unpublished.

NECSA (Nuclear Energy Corporation of South Africa) (2007b): Report of radioactivity analysis of vegetables and fish. Job code RA 08424, 11 Sep. 2007, for NNR, 10 pp. unpublished.

NECSA (Nuclear Energy Corporation of South Africa) (2007c): Report of radioactivity analysis of meat and fish. Job code RA 08303 (B), 25 Sep. 2007, for Driefontein GM, 4 pp. unpublished.

NECSA (Nuclear Energy Corporation of South Africa) (2007d): Report of radioactivity analysis of meat. Job code RA 08365, 10 Dec. 2007, for Driefontein GM, 4 pp. unpublished.

NECSA (Nuclear Energy Corporation of South Africa) (2007e): Report of radioactivity analysis of meat. Job code RA 08407, 10 Dec. 2007, for Driefontein GM, 4 pp. unpublished.

Olivier M (2007): DME finalises national nuclear strategy. Mining Weekly online, published 14 February 2007

Phillips O (2011): Letter of the chairman of the Remediation Steering Committee Mr. O. Phillips (National Nuclear Regulator) to the Vice-chancellor of the North-West University (Dr. T. Eloff): Request for collaboration by Remediation Steering Committee. 30 June 2011, 1 p., unpublished.

Phillips O (2012): Minutes of the Regulators meeting regarding Wonderfonteinspruit catchment area remediation. Date of meeting: 20 February 2012, National Nuclear Regulator, Centurion, 4 pp., unpublished

Potchefstroom municipality (2002): Analytical results of concentrations of Al, Cu, Pb, Cd, Cr, Pb, Hg, Zn and V in blood and urine samples of 59 residents on the farms Blaaubank and Muiskraal in the lower WFS/ upper Mooi River catchment, August 2002, unpublished.

Potchefstroom Municipality (2012): U-levels in raw and tap water of Potchefstroom, analytical data for the period May 2010 to Dec. 2012, unpublished.

Prado GR, Arruda-Neto JDT, Sarkis JES, Geraldo LP, Müller RML, Garcia F, Bittencourt-Oliviera MC, Manso Guavera, MV, Rodrigues G, Mesa J, Rodriguez TE (2007): Evaluation of uranium incorporation from contaminated areas using teeth as bioindicators – a case study. Radiation Protection dosimetry, doi:10.1093/rpd/ncm489, scientific note, 1-4.

Pulles W (1991): Radionuclides in South African gold mining water circuits: an assessment of licensing, health hazards, water and waste water regulations and impact on the environment and

workforce. Restricted report no. 17/91, Programme reference GE1C, April. Chamber of Mines of South Africa, unpublished

Raymond-Wish S, Mayer LP, O'Neal T, Martinez A, Sellers MA, Christian PJ, Marion SL, Begay C, Propper CR, Hoyer PB, Dyer AC (2007): Drinking water with uranium below the U.S: EPA water standard causes estrogen receptor-dependent responses in femal mice. *Environ Health Persp*, 1**15**, 12, 1711-1716

Rerecha V; Kulich M; Rericha R; Shore DL; Sandler DP (2006): Incidence of leukemia, lymphoma, and multiple myeloma in Czech uranium miners: A case-cohort study. *Environm Health Perspect*, 114, 6, 818 - 822

Robb VM, Robb LJ (1998): Environmental impact of Witwatersrand gold mining. In:Wilson MGG, Anhaeusser CR [eds.] (1998): The mineral resources of South Africa. Sixth edition. Handbook 16, Council for Geoscience, Pretora (Silverton), 14-17.

Rosborg I, Nihlgard B, Gerhardson L, Gernersson M-L, Ohlin R, OlssonT (2005): Concentrations of inorganic elements in bottled waters on the Swedish market. *Environ Geochem Health*, **27**, 217-227

SABS (2011): South African National Standard (SANS) 241-1:2011 for drinking water. Part 1: microbiological, physical and chemical determinants. South African Bureau of Standards, Pretoria, South Africa

Salbu B (2009): Challenges in radioecology. J Env Radioactivity, 100, 1086-1091

Sami K, Druczinski AL (2003): Predicted spatial distribution of naturally occurring arsenic, selenium and uranium in groundwater in South Africa – reconnaissance survey. Water Research Commission, Pretoria, WRC report no. 1236/1/03, 84 pp.

SAPA (South African Press Agency) (2012): Cattle contaminated by uranium. http://www.bizcommunity.com/Article/196/475/87268.html, 19 Dec 2012

Schick J, Schroetter S, Lamas M, Rivas M, Kratz S, Schnug E (2008): Soil to plant interface of uranium. In: De Kok LJ, Schnug E [eds.]: Loads and Fate of fertilizer-derived uranium. Backhaus Publishers, Leiden, 157-167.

Schnug E and Hahneklaus S (2008): Dispersion of uranium in the environment by fertilization. In: Merkel BJ, Hasche-Berger A (2008): Uranium, mining and hydrogeology. ISBN 978-3-540-87745-5, Springer, Berlin – Heidelberg, 45-52

Schnug E, Steckel H, Haneklaus S (2005): Contribution of uranium in drinking waters to the daily uranium intake of humans – a case study from Northern Germany. *Landbauforsch Volkenrode*, **55**, 227-236

Scholtz N, Scholtz OF, Potgieter GP (2006): Potential environmental impact resulting from inadequate remediation of uranium mining in the Karoo Uranium Province. South Africa. In: Merkel BJ, Hasche-Berger A [eds.] Uranium in the environment. ISBN 10 3-540-28363-3, Springer Berlin Heidelberg New York, 789-799.

Sekoko I, Mafejane A, Potgieter D, Conradie B, Kempster P, Kühn A, Diefenbach A (2005): A survey on the radiological and chemical quality of water resources in selected sites of the Northern Cape Province. Resource Quality Service Report No. N/0000/GEQ0603, Final Report May 2005 (unpublished). 61 pp.

Shangeeva I (2010): Uptake of uranium and thorium by native and cultivated plants. Journal Environ Radioactivity, 101, 458-463

Snyman HG, Herselman JE, Kasselman G (2005): A metal content survey of South African sewage sludge and evaluation of analytical methods for their determination in sludge. WRC project no. 1283, WRC, 'Pretoria, South Africa, 215 pp.

SRK (Steffen, Robertson & Kirsten) (1988): Research on the contribution of mine dumps to the mineral pollution load in the Vaal Barrage. WRC-Report-No: 136/1/89. Pretoria. Pp. 114

Stoch EJ, Retief J (1967): Memorandum of farmers on adverse effects associated with mine water on the health of farm animals. Private archive of EJ Stoch, Welverdiend, South Africa, unpublished, 3 November, 5 pp.

Stoch EJ (Les) (2008): Don't believe everything you read. Published letter in Carletonville Herald, 30 May.

Stuijt A (2008): SA radioactive stream – 400,000 at high risk. http://www.rense.com/general77/sarad.htm, accessed 5.5.2008

Taverner L (1957): An historical review of the events and developments culminating in the construction of plants for the recovery of uranium from gold ore residues. In: Uranium in South Africa 1946-1956. Volume 1. Proceedings of a joint symposium of the Associated Scientific & Technical Societies of South Africa. Hortors ltd., Johannesburg, 1-19

Tempelhoff E (2007 a): Mynmonopolie se gru-moeras. Beeld, 2 February 2007, p. 11 (newspaper article)

Tempelhoff E (2007 b): Potch se water 'skoonste in SA dalk die wreld' (Potch's water 'cleanest in SA and perhaps the word'). Beeld, 3 August, p. 6.

Tempelhoff E (2007 c): Duisende beeste dalk geskiet oor besoedeling ('*Thousands of cattle shot because of pollution'*). Beeld, 29 November 2007

Tempelhoff E (2008a): 'Dra maskers in Wonderfontein' ('*Use masks in Wonderfontein'*). Beeld, 7 May, p. 5.

Tempelhoff E (2008b): 'Giftstof in kos skok – radioaktiewe stowwe in voedselketting. ('*Poison in food shock – radioactive susbtances in foodchain'*). Beeld, 2 February, frontpage.

Tempelhoff E (2008c): 'G'n radioaktiewe stowwe in kos, reken reguleerder. *('No radioactive substances in food, reckons regulator'*). Beeld, 8 February, p. 10.

Tempelhoff E (2008d): Regering mislei publiek' oor Wonderfronteinspruit se veiligheid *(Afrikaans, 'Government misleads public over Wonderfonteinspruit's safety*. Beeld, 23 Sep.

Tempelhoff E (2012): Bees-organe is besmet – studie (Afrikaans, '*Cattle organs are polluted – study*'). Beeld newspaper, frontpage, 18 December.

Thomas PA (2008): Food chain transfer of uranium series nuclides. De Kok LJ, Schnug E [eds.]: Loads and Fate of fertilizer-derived uranium. Backhaus Publishers, Leiden, 169-178.

Toens PD, Stadler W, Wullschlege NJ (1998): The association of groundwater chemistry and geology with atypical lymphocytes (as biological indicator) in the Pofadder area, North Western Cape, South Africa. WRC report 839/1/98.

Tomášek L, Malátová I (2004): Leukaemia among Czech uranium miners. Proceedings of the 9th International Conference on Health Effects of Incorporated Radionuclides Emphasis on Radium, Thorium, Uranium and their Daughter Products - HEIR 2004, GSF-National Research Center for Environment and Health, Neuherberg, Germany, Nov 29 - Dec 1, 2004, U. Oeh, P. Roth, H.G. Paretzke (Editors), Institut für Strahlenschutz, GSF-Bericht 06/05, 128-135

UBA (Umweltbundesamt) (2005): UBA (UMWELTBUNDESAMT) (2005) Uran und Human-Biomonitoring: Stellungnahme der Kommission Human-Biomonitoring des Umweltbundesamtes. *Bundesgesundheitsbl – Gesundheitsforsch– Gesundheitsschutz 7-2005* ***48*** *822-827. DOI: 10.1007/s00103-005-1069-7, Springer Medizin Verlag.*

UBA (Umweltbundesamt) (2008): Uran im Trinkwasser. Kurzbegründung der gesundheitlichen Leit- und Grenzwerte, 19. August 2008, http://www.umweltdaten.de/wasser/themen/trinkwasserkommission/kurzbegründung_uran_leitwert.pdf

Vahrenholz C, Popp W, Wust T, Rietschel F, Bauer P, Konietzko N, Norpoth K (1997): DNA damage in blood lymphocytes of former uranium miners. *European Journal of Cancer*, 33, 8, 14 - 14 (1)

Van Heerden D (2003a): Potch Council takes gold mines to court. Potchefstroom Herald, 10 January 2003, page 2

Van Heerden D (2003b): Everyone stirred into action – farmer. Potchefstroom Herald, 10 January 2003, page 2

Van Heerden D (2007): Experts say water is safe, for now. Potchefstroom Herald, 14 December.

Van Wyk, Coetzee H (2008): The distribution of uranium in groundwater in the Bushmanland and Namaqualand areas, Northern Cape Province, South Africa. In: Merkel BJ, Hasche-Berger A [eds.]: Uranium. Mining and hydrogeology. Springer Berlin Heidelberg, Germany, 639-644.

Von Soosten C (2008): Stellungnahme der Beratungskommission der Gesellschaft für Toxikologie e.V. (GT) zur aktuellen Diskussion über eine mögliche gesundheitliche Gefährdung durch Überschreitungen des Trinkwasser-Leitwertes für Uran. http://idw-online.de/pages/de/news279231, accessed 22.9.2008

Wade PW, Woodbourne S, Morris WM, Vos P and Jarvis NV (2002) Tier 1 risk assessment of radionuclids in selected sediments of the Mooi River. WRC-Project No K5/1095, Pretoria.

WAG (Wonderfonteinspruit Action Group) (2007): Concentrations of u and other heavy metals in sediments of the lower Wonderfonteinspruit catchment – results from a grid based sampling exercise funded by Goldfields Ltd., unpublished

Werner, E., P. Roth, U. Heinrichs, P. Schramel, I. Wendler, J. Mc Aughey, R. Hearn, M.C. Cantone (1999): Uranium concentration in tap water and urine of unexposed subjects in Britain, Italy and Germany. IRPA Regional Congress on Radiation Protection in Central Europe. Budapest 23.-27.08.99

WHO (1999): Agreement between the International Atomic Energy Agency (IAEA) and the WHO, ratified by the 12th WHO-General Assembly, 28th May 1959, Resolution WHA 1240, Organisation mondiale de la santé, "Documents fondamentaux", 42. edition, WHO, Geneva

WHO (World Health Organization) (1998): Guidelines for drinking water quality. Health criteria and other supporting information, Addendum to Vol. 2 WHO/EOS/ 98.1 Geneva: World Health Organization

WHO (World Health Organization) (2003): Uranium in drinking water. Summary statement – extract from chapter 12 – chemical fact sheets of WHO guidelines for drinking-water quality, 3rd edition, 2004. WHO/SDE/WSH/03,04/118, http://www.who.int/water sanitation health/

WHO (World Health Organization) (2005): Uranium in drinking-water. Background document for development of WHO guidelines for drinking water quality. WHO/SDE/WSH/03.04/118

WHO (World Health Organization) (2006): Guidelines for drinking water quality [electronic resource]: incorporating first addendum. Vol. 1, Recommendations, 3rd ed. Electronic version for the web

Winde F (2002): Uranium contamination of fluvial systems. Mechanisms and processes. Part III: diurnal and event-related fluctuations of stream chemistry. Pitfalls from mining affected streams in South Africa, Germany and Australia. *Cuadernos de investigación geográfica*, ISSN 0211-6820, **28**, 75-100.

Winde F (2003): Comparative study on U-levels in calcite scales of water kettles in Potchefstroom and Ventersdorp. Analytical data, unpublished.

Winde F (2006a): Challenges for sustainable water use in dolomitic mining regions of South Africa – a case study of uranium pollution, Part I: sources and pathways. *Physical Geography*, ISSN 0272-3646, **27**, 2, 335-346.

Winde F (2006b): Challenges for sustainable water use in dolomitic mining regions of South Africa – a case study of uranium pollution, Part II: Spatial patterns, mechanisms and dynamics. *Physical Geography,* ISSN 0272-3646, **27**, 2, 379-395.

Winde F (2008): Development of a map ranking sites with known radioactive pollution in the Wonderfonteinspruit catchment according to the urgency of required intervention ('Intervention site map') - Underlying methodology and results. Confidential report to Joint Coordinating Committee of the Department of Water Affairs and Forestry (DWAF) and the National Nuclear Regulator (NNR), DWAF Pretoria, unpublished, pp. 17.

Winde F (2009a): Urankontamination von Fließgewässern – Prozessdynamik, Mechanismen und Steuerfaktoren. Untersuchungen zum Transport von gelöstem Uran in bergbaulich gestörten Landschaften unterschiedlicher Klimate. *(Uranium contamination of streams – process dynamics, mechanisms and governing factors. Investigations into the transfer of dissolved uranium in mining-impacted landscapes of different climatic conditions)*; ISBN 978-3-86866-087-6, Taunusstein, pp. 657.

Winde F (2009b): Uranium pollution of water resources in mined-out and active goldfields of South Africa - A case study in the Wonderfonteinspruit catchment on extent and sources of U-pollution and associated health risks. International Mine Water Conference, 19-23 October 2009, Pretoria (South Africa), Conference proceedings, CD, ISBN 978-0-9802623-5-3, 772-782.

Winde F (2010a): Uranium pollution of the Wonderfonteinspruit: 1997 – 2008. Part I: U-toxicity, regional background and mining-related sources of U-pollution. *Water SA*, **36** (3), 239-256.

Winde F (2010b): Uranium pollution of the Wonderfonteinspruit: 1997 – 2008. Part II: U in water – concentrations, loads and associated risks. *Water SA*, **36** (3), 257-278.

Winde F (2011a): Challenges in assessing uranium-related health risks: two case studies from South Africa – Part 1: guideline and toxicity issues and the Pofadder case study. In: Merkel B, Schipek M (2011): The new Uranium mining boom. ISBN 978-3-642-22121-7, Springer Verlag Berlin Heidelberg, 529 – 538.

Winde (2011b): Email to Dr. Carl Albrecht on a possible design of the tooth study and a follow-up study on the Pofadder project, 20 October, 15:04

Winde (2011c): Uranium pollution in the Wonderfonteinspruit and Mooi River system with special reference to the situation in Potchefstroom. Presentation at the Water Quality Lekotla, 28th January, Old City Hall, Tlokwe Municipality, 65 min.

Winde F (2011d): Challenges in assessing uranium-related health risks: two case studies from South Africa – Part 2: Case study Potchefstroom. In: Merkel B, Schipek M (2011): The new Uranium mining boom. ISBN 978-3-642-22121-7, Springer Verlag Berlin Heidelberg, 539 – 546

Winde F, de Villiers AB (2002): Uranium contamination of streams by tailings dams – case studies in the Witwatersrand gold mining area (South Africa). In: Merkel BJ, Planer-Friedrich B, Wolkersdorfer C [Eds]: Uranium in the Aquatic Environment. Proceedings of the International Conference Uranium Mining and Hydrogeology III and the International Mine Water Association Symposium, Freiberg, Germany, 15-21 September 2002, Springer, Berlin, Heidelberg, New York, 803-812.

Winde F, Wade P, van der Walt IJ (2004): Gold tailings as a source of waterborne uranium contamination of streams – the Koekemoerspruit (Klerksdorp goldfield, South Africa) as a case study. Part I: Uranium migration along the aqueous pathway. *Water SA*, 30 (2), 219-226

Winde F, Sandham LA (2004): Uranium pollution of South African streams – an overview of the situation in gold mining areas of the Witwatersrand. Supplement volume, *GeoJournal*, **61**, 139-149

Winde F, Erasmus E (2011): Peatlands as filters for polluted mine water? – A case study from an uranium-contaminated karst system in South Africa. Part I: Hydrogeological setting and U-fluxes. *Water*, **3**, 291-322, doi:10.3390/w3010291

WISE (2001): Uranium toxicity. http://www.antenna.nl/wise/uranium/utox.htm, acc. 1.6.2002

WISE (2013): Uranium Radiation Age-specific Dose Calculator. http://www.wise-uranium.org/rdcua.html, accessed on 28 January 2013.

Wrenn ME, Durbin PW, Howard B, Lipsztein J, Rundo J, Sill ET, Willis DL (1985): Metabolism of ingested U and Ra. Health Phys, 48, 601-633

Wullschleger NJ, Visser D, Stadler W (1998): Groundwater health hazards of selected settlements in the province of the Northern Cape. Report to the DWAF, pp. 16, unpublished, Toens and Partners, Capetown.

Wymer D (1999): Table of Uranium concentrations in slimes dams of South African goldmines - based on the uranium concentration in ore, the amount of milled ore and the extend of extracted uranium. Johannesburg, unpublished

Zaire R, Notter M, Riedel W, Thiel E (1997): Unexpected rates of chromosomal instabilities and alterations of hormone levels in Namibian uranium miners. *Radiation Research*, 147, 5, 579-584

Zhuwakinyu M (2007): Minister envisages uranium mining, enrichment, fuel making and even processing of spent fuel elements in SA. Mining Weekly online, published 5 October 2007