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A fingerprinting method for the identification of uranium sources in alluvial aquifers: An example from the Khan and Swakop Rivers, Namibia

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ABSTRACT

A fingerprinting method for identifying sources of uranium in shallow groundwater alluvial aquifers of the Khan and Swakop Rivers was established using $^{234}U/^{238}U$ ratios and $^{235}U/^{238}U$ ratios in the areas that drain the Rossing Uranium mine and the Langer Heinrich Uranium mine, in Namibia. In most groundwater aquifers that drain basement granitoids enriched in uranium the contribution of the total uranium in the shallow alluvial aquifers may be significant. Another source of uranium in shallow alluvial aquifers maybe from anthropogenic sources associated with mining activities as is the case in our study area. The distribution of radionuclides in water depend on various factors that influence their solubility and mobility and control their concentration in water such as pH, Eh, O_2 and availability of ligands. The study identified a methodology that can fingerprint the two sources i.e., a natural source where $^{234}U/^{238}U$ ratios are above unity and a second one where this ratio is below unity implying that the source is anthropogenic. In the study area, $^{234}U/^{238}U$ activity ratio is above unity (1.3–1.7) and $^{235}U/^{238}U$ is 0.045 ± 0.015 that both identify a natural source for all elevated uranium and other radionuclides in groundwater of the study area. The uranium values in groundwater exceed the WHO guideline value of 15 μg/l and it increases in the lowest part of Swakop River; but there is no gradual or systematic change in uranium concentration thus indicating that concentration is related to local factors such as the geology and lithology of the aquifer material, Eh and pH for each borehole. The ^{238}U decay series exhibits disequilibrium due to different fractionation processes that include decaying of radioactive elements and alpha recoiling.

1. Introduction

Namibia is a dry country, whose population depends on groundwater. Close to 80% of the population depends on groundwater for their daily needs. The Uranium Province of Namibia is located some 60 km east of the towns of Walvis Bay and Swakopmund and about 300 km from Windhoek (Fig. 1). This area lies in the Namib desert part of Namibia where groundwater is scarce. According to Christelis and Struckmeier (2001), this area is classified as consisting of vulnerable aquifers. This province hosts two big uranium mines namely; Rossing Uranium and Langer Heinrich. Currently a number of exploration activities are going on this area. The Khan and Swakop River catchment (Figs. 1 and 2a) serve as source of water for the farming communities and the Rossing mine operating in the area.

It is known that uranium in drinking water should not exceed values of 15 μg/l due its chemical toxicity for the kidney (ICRP, 1991, WHO, 2008a). The ICRP (International Commission on Radiological Protection) (2000) established that uranium and its associated decay series radionuclides such as radon, radium, thorium can cause cancer when exposure is low to moderate over a long period of time. Thus for the populations along the Khan and Swakop rivers, exposure to radionuclides in water whose concentration is above 15 μg/l has inherent exposure problems. Studies done elsewhere e.g., Langmuir (1997), Wu et al. (2014) and Nriagu et al. (2012) have shown that uranium can be highly elevated in groundwater. Naturally occurring uranium in groundwater is common, for example; Banning et al. (2013); Chkir et al. (2009, 2012). Isotopes of uranium have been variously used to characterize how uranium behaves in groundwater and also identifying it as a pollutant (Arnold et al., 2011; Chkir et al., 2009; Christensen et al., 2004; Porcelli, 2008). Triapathi et al. (2013) used a similar approach to ours, in trying to establish whether secular equilibrium was attained in ground water where uranium was present.

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Associated with uranium is radon in water. The United States Environmental Protection Agency (2003) states that radon in groundwater is usually in small quantities compared to that found in air. As such more efforts are directed to the radionuclides radium, thorium, and uranium that may have severe hereditary effects for the population as it damages the reproductive organs. A study by Kurttio et al. (2002) showed that natural uranium when ingested in drinking water causes a reduced re-absorption in the kidney tubules. Uranium also when ingested in water with levels above 15 μg/litre may be harmful to bone tissue because it accumulates in the bones and causes excretion of phosphate and calcium, that are important ingredients in bone formation (Kurttio et al., 2005). Thus the significance of understanding the concentrations of uranium in groundwater cannot be overemphasized.

The contribution of anthropogenic uranium to the groundwater of the alluvial aquifers has not been known for a long time. This study is a first of its kind in Namibia to undertake a fingerprinting investigation for uranium, and it is hoped that other areas in Namibia and elsewhere will apply the method to differentiate uranium sources in shallow alluvial aquifers. Shallow alluvial aquifers are defined as those with a depth of not more than 20 m to the water table. In this paper a mechanism of identifying the two separate sources of uranium (natural and anthropogenic) is documented. This is important as it highlights the anthropogenic contribution from the big operating mines in the area.

The exposure to humans and animals for radionuclides is measured mainly in four different units. The recommended dosage for one year regardless of age is 0.1 milli Seivert from consumption of groundwater, assuming a cumulative volume of 750 l of water per year (WHO, 2008b). 0.1 milli Seivert is considered safe by WHO (2008b). The Sievert is the equivalent radiation dose representing the stochastic biological effects of ionizing radiation, and therefore represents a measure of the biological effects. The other unit used is the Becquerel, which is defined as the "disintegration of a radioactive material per second". The activity of becquerels is measured in Bq/l or Bq/kg. The amount of deposited energy by radiation is called the Gray (Gy). 1 Gy is equivalent to 1 Joule per kg. In this study we will discuss only the concentration of a radionuclide in g/l or μg/l.

2. Geology of the study area

The study area is located in central zone of the Damara belt of Namibia (Figs. 2a and 2b). The geology of the Damara belt is made up of two main components: Mesoproterozoic basement rocks and the Damara Supergroup (Table 1). The basement rocks are mainly quartzofeldspathic gneisses, amphibolites and metasediments of the Abbabis Metamorphic Complex, whereas the Damara Supergroup in central zone is composed mainly basal Arkosic arenite with local alkaline volcanic and evaporites of the Nosib Group (Miller, 2008). The Nosib Group is overlain by the Swakop Group, which consists of the basal Chuos Formation succeeded by carbonates, shales and quartzites of the Rossing Formation (Table 1). The Rossing Formation is overlain by the carbonate rocks of the Arandis and Karibib Formations respectively. The top of the sequence is represented by greywackes of the Kuiseb Formation (Table 1). The Uranium mineralization is associated with the granitic bodies that intruded the Swakop Group as leucogranites (Fig. 3) which are late to post tectonic generally having high uranium grades (Bowden et al., 1995).

There are two mines operating in the vicinity of the Khan and Swakop River catchment, namely the Rossing Uranium mine and the Langer heinrich mine. The Rossing Uranium mine is hosted by alaskatic granitic bodies with a tonnage of up to 150 Mt and grades of 300–370 ppm. The Langer Heinrich Uranium mine is a calcrete hosted deposit with uranium resources of about 74 Mt and a grade of 0.06% U_3O_8 (Hartleb, 1988). The Khan and Swakop Rivers are made of alluvial aquifers that are not consistently uniform over their length. Bittner and Kuells (2010), divided the alluvial aquifers into compartments due to their different hydrologic characteristics such as volume and hydrochemistry. The Khan and Swakop rivers are both ephemeral rivers, that only flow for a short period each year, of circa two weeks during the rain season. In desert environments rainfall does not fall every year, such that some years are dry and the rivers do not flow. These alluvial aquifers are generally recharged once in two or three years. In certain years, they could be recharged every year as was the case in 2009–2011. Recharge is not systematic, but sporadic and

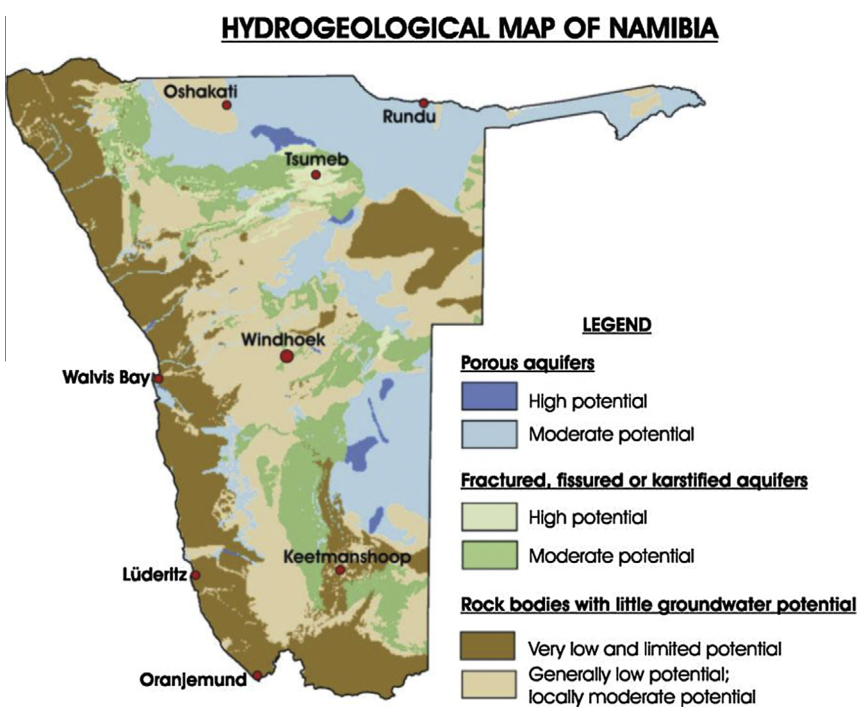

Fig. 1. Location of the study area.