Dose Limits and Maximum Concentration Limits (MCL´s) for Radionuclides – Implication on Remediation of Uranium Mining and Milling Facilities in Saxony, Germany

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Abstract. In 2000 and 2001 the EU framework for Community action in the field of water policy and simultaneously a new radiation protection regulation came into force in Germany. Both affect the remediation of uranium mining and milling activities in Saxony. Radionuclide emissions from sources like waste rock piles and flooding waters of mines contribute to increased levels of radionuclides in the Mulde River and in the groundwater. Drinking water supplies along the Mulde River have to guarantee a good potable water quality. The dose limit of the drinking water regulation has no real impact on the toxic importance of uranium. For the chemical toxicity of uranium no MCL exists. In Saxony the radiation protection and the water authorities are well aware of these circumstances. It is an urgent task for the German and the European authorities to set up a MCL for uranium in water.

Introduction

Saxony has a long tradition of ore mining. It is known, that it took place for at least 800 years. The region with the highest ore contents is the traditional mining area Erzgebirge, the German term for “ore mountains”.

From the early fifties until the late eighties of the past century uranium mining was going on intensively. Eastern Germany produced about 200.000 tons of uranium and therefore was worldwide number three in the field of uranium production. About 50% of it came from Saxony ore.

As a consequence of extensive mining radionuclides were mobilized by precipitation water from tailings and waste heaps as well as by flooding water from the underground mines.

This is the background why in Saxony radionuclides are of special interest as radiologically and chemically toxic elements.

Natural background in Saxon waters and man made contaminations

The natural background of radionuclide concentrations in ground and surface waters in Saxony depends on the geological situation on the one hand, on the other hand on the hydrogeochemical situation.

Rock formations with higher radionuclide contents

They are found in the Erzgebirge (hydrothermal vein deposits) in the southern part of Saxony. In the Permian Döhlen Basin (hard coal) west of Dresden some coal seams show high uranium contents. In some places of the lower Saxon Cretaceous we find increased uranium contents (Königstein sandstone deposit). In all these formations uranium was mined. In some tertiary lignite seams in north-western and south-eastern Saxony, which partly also show increased uranium concentrations, it was prospected but not mined.

Hydrogeochemical conditions controlling radionuclide migration

Different radionuclides have different chemical properties. Uranium for example is very well soluble under oxidising conditions, while radium can be found in increased concentrations under more reducing conditions. Both can be used as indicators for the presence of other natural radionuclides. If uranium and radium show very low concentrations, normally no other natural radionuclides can be found in waters.

If natural organic matter (NOM) is present uranium mainly is transported in organic complexes. In this case it is much more mobile. On the other hand if there is immobilized NOM in the water, uranium is much more immobile.

Rock surfaces as well can be very important for radionuclide mobility. Iron or manganese hydroxides on rock surfaces as well as carbonates can restrain radionuclide mobility in two ways: no radionuclides can escape from the rocks and the water soluble radionuclides can be bound at the large surfaces of the hydroxides.

From the above one can conclude, that areas

- with increased uranium concentrations in the rocks,
- where the uranium bearing rock is close to the surface (high redox)

- with increased contents of NOM

are prone to have naturally increased uranium concentrations in the ground and surface water.

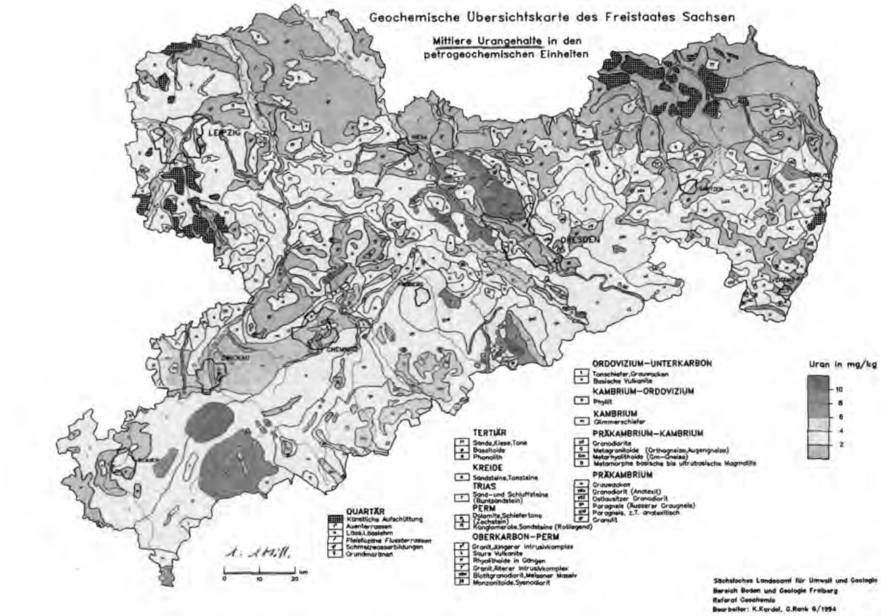

Fig. 1. Geochemical Map of Saxony. Mean uranium contents in Saxon rocks. The spots with the high uranium concentrations in the south west are part of the Erzgebirge.

However, areas

- with increased uranium concentrations in the rocks
- where the uranium bearing rock is deep enough under the ground water table (low redox)

are prone to have naturally increased radium concentrations in the ground water.

In areas where uranium mining or mining of other ores with increased uranium concentrations took place the water naturally shows more increased radionuclide concentrations. The reason is an enlargement of the rock surface by the mining activities (tailings, waste rock) creating better conditions for the leaching agent (rain water) and alteration of the hydrogeochemical situation (e.g. flooding of underground mines to a lower level than the natural one).

The natural background concentrations of uranium in water of the three Saxon areas of former uranium mining are as follows:

a) In the Erzgebirge natural background samples could only be taken where no remnants from uranium mining exist. 6 samples from creekwater showed a mean uranium concentration of 0,17 µg/l. Groundwater samples showed concentrations between 1 and 2 µg/l.

b) Upstream samples of tailings ponds from the Döhlen Basin showed uranium concentrations between 0,7 and 0,8 µg/l.

c) Close to the ore body of the Königstein deposit uranium concentrations are generally close to or below the detection limit.

These generally low natural uranium concentrations in water from areas of ore deposits are not surprising. If the hydrogeochemical conditions had mobilized uranium, no ore deposits would exist.

Anyway, natural radium concentrations can be very high in the closer surroundings of uranium ore deposits.

In the tertiary lignite areas the situation is much more differentiated in regard to uranium concentrations. The concentrations vary between below detection limit and a maximum concentration of about 300 µg/L in a ground water well south of Leipzig. However, it is not clear if this very high concentration has been induced by a local lowering of the groundwater table. In this case, oxygen is transported into the groundwater and the solubility of uranium is increased. Most samples show uranium concentrations below 1 µg/l.

For other areas in Saxony only random samples exist. Surface waters show uranium concentrations of 0.11 to 4.5 µg/l. The very few ground water samples show uranium concentrations below 1 µg/l.

In contrast to these concentrations, uranium concentration is very high at all places where uranium mining was going on. In seepage waters from waste piles, in ground waters of the surroundings of tailings ponds and in flooding water from underground mines uranium concentrations of several mg/l are the rule. It can be observed, that during the flooding of mines uranium concentrations decrease and radium concentrations increase. On the other hand mine waters from Saxon locations flooded several decades ago show low uranium and low radium concentrations. But there are too few examples to prove that they are representative for the general geochemical development of mine waters.

Most former uranium mining objects in Saxony are located in the catchment area of the Mulde River. Today in the Mulde River concentrations between about 10 and about 80 µg uranium/L can be measured. Some creeks in the catchment area of the Mulde River show concentrations up to 600 µg uranium/L.

The catchment area of the Mulde river covers nearly a quarter of the area of Saxony. As long as uranium mining remediation activities are going on the Mulde water will show increased radionuclide concentrations.

German and international regulations and standards

§ 7 of the German Drinking Water Regulation of May 21, 2001 says that "…in water for human consumption indicator parameters of the standards in attachment 3 have to be followed". Attachment 3, number 20 says, that a total individual dose of 0,1 mSv/a is allowed. This regulation includes all radionuclides except of ^{3}H, ^{40}K, Rn and Rn daughters.

With this paragraph a corresponding regulation of the European Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption came into force for Germany.

The radiation protection regulation of July 20, 2001 gives standards for the ingestion of radionuclides with drinking water. Looking at the standards for infants from 0 to 1 year age one can calculate concentrations for different natural uranium isotopes from 1.59 to 1.73 Bq/L (128 to 130 µg/L).

In all these regulations uranium is only considered as a radionuclide, not as a heavy metal. Uranium as a heavy metal causes kidney diseases and is – if ingested - much more toxic than as a radionuclide (ATSDR 1999, Gilman et al. 1998a, Jacob et al. 1997 and Mc Donald-Taylor et al. 1992).

The World Health organisation (WHO) has established a Tolerable Daily Intake (TDI) for uranium of 0.6 µg/kg body weight and day. This is based on adverse affects observed by Gilman et al. (1998b) with kidneys of rats at uptakes of 60 µg U per kg and day. The WHO provisional guideline for drinking water quality recommends 2 µg uranium per litre as a standard.

Zamorra (1998) was the first to publish a study on the effects of chronic ingestion of uranium with drinking water on humans. He found that kidney function is affected by uranium uptakes considered to be safe in the publications based on animal studies.

International standards and recommendations are taking the chemical toxicity of uranium into consideration:

- Health Canada recommends an interim maximum acceptable concentration (IMAC) of uranium in drinking water of 20 µg per litre.
- The U.S. Environmental Protection Agency (EPA) Rule on Radionuclides in Drinking Water prescribes a maximum contaminant level for naturally occurring uranium of 30 µg per litre. EPA determines a safe level of 20 µg per litre assuming that an adult with a body mass of 70 kg drinks 2 litres of water per day and that 80% of exposure to uranium is from water. For cost considerations, however, EPA established a standard of 30 µg per litre rather than 20.
- The U.S. EPA Preliminary Remediation Goals for Superfund objects recommend 2.22 µg per litre for uranium in tap water while the U.S. EPA Groundwater Standards for Remedial Actions at Inactive Uranium Processing Sites recommend 30 pCi/L for U-234 and U-238 (44 µg per litre where secular equilibrium obtains).
- The U.S. Nuclear Regulatory Commission (NRC) made occupational annual limits on intakes (ALI) for oral ingestion of 10 µCi (=14.8 g).
- The Australian Drinking Water Guidelines say that the concentration of drinking water should not exceed 0.02 mg/L (= 20 µg/L). (http://www.antenna.nl /wise/uranium/utox.html)

As mentioned above in federal German regulations no standards for uranium as a chemical toxic exist. But e.g. the environmental department of the state Hessen in 1998 laid down a standard of 2 µg/L for uranium in drinking water.

Due to increased natural concentrations in drinking waters in northern Bavaria the Bavarian water authorities are presently discussing to recommend a standard of 5 µg/L for drinking water.

Also the German State Commission on Water Issues (Länderarbeitsgemeinschaft Wasser) presently is discussing a level of insignificance (Geringfügigkeitsschwellenwert) of 1 µg/L for natural uranium in surface and ground water.

It is to state, that the standards for the radiation doses and for the chemical toxicity are not comparable. Additionally the residual risk from chemical toxicity regarded acceptable usually is orders of magnitude lower than from radiation. Unfortunately there are still not sufficient data available for the chemical toxicity of uranium. These may be the reasons for the relatively wide spreading values in the different international regulations.

Proposal for an EC regulation

A standard for uranium as a heavy metal, i. e. as a chemical toxic in drinking water is not only needed for Germany. The European Commission should take care for a common regulation for Europe.

The toxicity of uranium is comparable to that of Arsenic. The EC standard for arsenic is 10 µg/L. For that reason it would be plausible to discuss a standard of 10 µg/L for uranium in drinking water.

Taking into consideration the facts shown above by the responsible persons in the EC it should be only a question of time until a standard for uranium is part of a European drinking water directive.

We do not have data of uranium concentrations in waters of other European countries. But regarding the situation in Germany one can guess, that more than 90% of all drinking water supplies in Europe have waters with uranium contents below 1 µg uranium/L. Drinking water supplies with uranium concentrations higher than 10 µg/L in their waters will be far below 1%. They should – in case of the establishment of a standard - get a special support by the water authorities to minimize economic disadvantages.

Impact of a regulation on uranium mining remediation

Drinking water regulations definitely can have a substantial influence on the demands of the regulator concerning the way and quality of the remediation method. In the case of a precautionary uranium standard for example, water authorities have to demand water treatment methods that keep back higher contents of uranium. This can lead to higher expenses for the deposition of the solid remnants of the treatment. Additionally it can lead to longer water treatment times.

Therefore it is very important to work out optimized remediation concepts from the beginning.

In the first place such concepts should take into account that the hydrogeochemical development of radionuclide migration strongly depends on the specific on-site conditions. Especially if prognoses let expect long treatment times one should develop and apply passive treatment methods at the earliest possible time.

However, drinking water standards are no remediation standards. In Saxony remediation decisions are made on the base of the respective present utilization of waters. For this reason in most cases a standard for uranium as proposed above would have no direct consequences on remediation decisions.

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