Journal of Environmental Radioactivity 64 (2003) 175–193

JOURNAL OF ENVIRONMENTAL RADIOACTIVITY

www.elsevier.com/locate/jenvrad

Uranium ores and depleted uranium in the environment, with a reference to uranium in the biosphere from the Erzgebirge/Sachsen, Germany

A. Meinrath [a,*], P. Schneider [b], G. Meinrath [c,d]

[a] *Klinikum, Bischof-Piligrim Str. 1, 94032 Passau, FRG, Germany*
[b] *Hydroisotop-Piewak GmbH Oberfrohnaer Str. 84 09117 Chemnitz, FRG, Germany*
[c] *RER Consultants, Schießstattweg 3a, 94032 Passau, FRG, Germany*
[d] *Technical University Mining Academy Freiberg, Institute of Geology, G.-Zeuner-Str. 12, 09596 Freiberg, FRG, Germany*

Received 26 June 2001; received in revised form 18 October 2001; accepted 22 October 2001

Abstract

The Erzgebirge (‘Ore Mountains’) area in the eastern part of Germany was a major source of uranium for Soviet nuclear programs between 1945 and 1989. During this time, the former German Democratic Republic became the third largest uranium producer in the world. The high abundance of uranium in the geological formations of the Erzgebirge are mirrored in the discovery of uranium by M. Klaproth close to Freiberg City in 1789 and the description of the so-called ‘Schneeberg’ disease, lung cancer caused in miners by the accumulation of the uranium decay product, radon, in the subsurfaces of shafts. Since 1991, remediation and mitigation of uranium at production facilities, rock piles and mill tailings has taken place. In parallel, efforts were initiated to assess the likely adverse effects of uranium mining to humans. The costs of these activities amount to about 6.5 10^9 Euro. A comparison with concentrations of depleted uranium at certain sites is given.

Keywords: Uranium; Depleted uranium; Uranium mining area remediation; Natural abundance; Health effects

---

[*] Corresponding author: Tel./fax: +49 851 70372.
E-mail address: meinrath@panet.de (A. Meinrath).

PII: S0265-931X(02)00048-6

1. Introduction

A connection was made in recent years between the release of depleted uranium (DU) during the war activities of the Gulf War and the Kosovo air strikes and certain adverse health effects observed in soldiers serving during these wars. During the bombardment of armoured installations, depleted uranium used in armour-breaking bullets was released upon impact or by corrosion of the bullet mantel materials. Uranium, however, is an almost ubiquitously occurring element. Therefore, adverse health effects were also assessed in the Saxonian mining area, a densely populated part of Eastern Germany, where uranium has been exploited on a large scale with little attention to possible long-term environmental and health effects (Gatzweiler and Mager, 1993).

After 1945, the Soviet government exchanged a sector of Berlin for Saxony, which had been under American control after the German surrender in World War II. The mining and treatment of uranium ores took place under the auspices of the Soviet Enterprise (SAG), which later became the Soviet-German Enterprise (SDAG) WISMUT. After uranium production activities were stopped, the open pit mines, subsurface mines, uranium extraction and leaching installations, as well as the enormous heaps of partially leached mill tailings, posed and still pose an eminent ecological and financial risk and a challenge to remediators and licensing authorities (Knoch-Weber, 1998; Meinrath et al., 2000).

The former German Democratic Republic (GDR) was the third largest producer of uranium until 1991 when uranium production was halted after the German reunification in 1989. Uranium exploited in the GDR (and Czechoslovakia) served the former Soviet Union’s strategic needs for uranium from 1945 until 1989. Consequently, protection of the workers and the environment was of secondary importance. The total amount of mined material (ores and tailings) was 1200 million tons. 200 million tons were transported to processing plants. The remaining 1000 million tons were dumped in nature, partly forming new landscapes. Dumping occurred without protection for ground and surface water, leaching by rain or distribution of uranium-bearing dust by wind. A total of 231,000 tons of uranium were produced and transported to the Soviet Union, from which 126,000 tons had been mined in Saxony.

Currently, some of the sites affected by uranium mining activities have been under the jurisdiction of the federal government since 1961 and in the process of remediation and mitigation (Gatzweiler and Mager, 1993; Flesch et al., 1998). Those sites transferred to the local authorities before 1961, remain under the jurisdiction of the local communities and their future is uncertain.

Of the 27 sites that have been exploited in total, 23 sites have been exhausted and four sites still contain some thousands of tons of uranium. The concentration of uranium in the ores was 0.04%–0.11%. The sandstone mines at Königstein contain about 0.08%. The characterization of the WISMUT mining areas in Saxony is summarized in Table 1. Central processing of saxonian (and thuringian) uranium ores was performed in Crossen.

Saxony comprises an area of 18.338 km^2 with currently about 4.7 million inhabitants. The population decreased after 1989 by about 1 million, due to demographic

Table 1
Characterization of Wismut mining areas in Saxony (Wolkersdorfer, 1996)

Location	Characterization	Uranium deposits	Amount UO_2 mined
Schlema	Hydrothemal vein deposit	Niederschlema/Alberoda	81 kt[a]
		Tellerhäuser, Oberschlema, “Weißer Hirsch”, Johanngeorgenstadt, Antonstal, Seiffenbach, Zobes, Bärenstein, Schneckenstein, Marienberg, Schneeberg, Pöhla, Geyer	21 kt
Königstein	Cretacious sandstone	Königstein, Pirna, Thürmsdorf, Rosenthal	19 kt
Freital	Permian hard coal	Heidenschanze, Bannewitz-Gittersee	4 kt

[a] kt: 1000 tons.

and migration effects. Due to the mining activities, the major mining areas of Aue, Schwarzenberg and Schneeberg are especially densely populated with about 300 inhabitants per km^2.

Fig. 1 gives a map of the German federal state of Saxony, together with the evaluated uranium contents from an aerogamma survey using the ^{238}U progeny ^{214}Bi (Saxonian Federal Agency of Environment and Geology, 1993). The local variation of uranium in the rock formations is clearly seen. The dark spots characterize mill tailings and production sites notably at Crossen (processing/sedimentation pond), Schneeberg and Aue (mill tailings), Dresden/Gittersee and Königstein. Note that the sites of Dresden/Gittersee (Junghans and Helling, 1998) and Johanngeorgenstadt are close to densely populated areas.

In 1953, a peak number of 115,405 workers in the Saxonian uranium industry were reported; the majority of whom worked in direct contact with uranium ores and processed uranium. The total number of workers will probably never be retraced precisely, but the estimated number ranges from 400,000 to 600,000 persons with

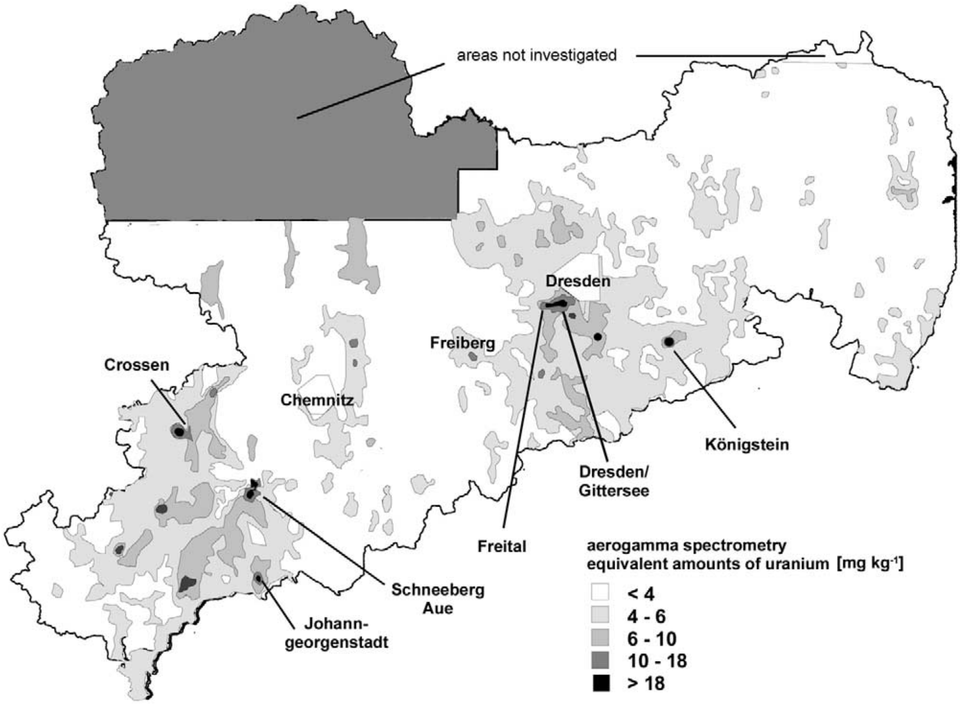

Fig. 1. Aerogamma survey of uranium contents in rocks and soil of the German federal state Sachsen (Saxony). The uranium is associated mostly with variszic granite rocks in the Erzgebirge region forming the political border with the Czech Republic to the south. To the east, Poland is located. The highest uranium concentrations shown by the dark areas are due to uranium mining and processing sites. The sites of Crossen, Johanngeorgenstadt, Schneeberg/Aue, Dresden/Gittersee and Königstein (subsurface sandstone deposit and surface uranium mill tailing) are shown together with the cities of Freiberg, Chemnitz and Dresden (Saxonian Federal Agency of Environment and Geology, 1993).

about 50% working in the mines or processing plants (Schröder et al., 1997). From 1963 to 1989, a constant average of 45,000 persons were employed with the majority working with uranium.

This communication collates data and facts from a century-long experience with increased uranium content in the geosphere. This experience will be compared to the scenarios developed concerning the use of depleted uranium (DU) during Gulf War (GW) and Kosovo Air Strikes, as well as to the application of DU in tank armour and in civilian airplanes (McDiarmid, 2001).

2. Natural uranium and depleted uranium

Uranium was discovered in 1789 by Martin Klaproth in an ore near Freiberg, Germany. This is in a mountainous region that comprizes a varizic geologic formation at the border between Germany's federal state, Sachsen (Saxony), and the Czech Republic to the south. This area is called 'Erzgebirge' (literally translated 'Ore Mountains') and is rich in minerals. Notably, in the 15th–17th century it was a center of silver mining. The city of Jachimov (Czech Republic) was known as Joachimsthal in the late 19th century and its name continues in the name 'dollar' ('Thaler') (Zeman and Benes, 1995). Other minerals, including nickel, cobalt, and germanium, were discovered during the analysis of the ore by the chemist Clemens Winkler in 1846 at the Freiberg School of Mines, the world's first mining academy. The intense mining activities around Freiberg took place in a granite formation with an abundance of uranium. Its radon progeny had taken its toll on the miners causing the so-called 'Schneeberg disease' (lung cancer) (Kreuzer et al., 2000).

Uranium (Z=92) is the heaviest element occurring naturally in weighable amounts. Despite its high atomic number, it is by no means a rare element. Its relative abundance compares to silver, gold and the light rare earths elements and it is more common than tin, mercury and lead. Its naturally occurring isotopes have masses 238, 235 and 234. All isotopes are radioactive (cf. Table 2) (Lieser, 1980). The average Uranium concentrations in the earth's crust are about 3 mg kg^{-1}. Calculations comparing the heat flow in oceans and continents show that the decay of uranium accounts for the majority of the continental heat production (Rogers and Adams, 1969). Concentrations of uranium in rocks are highest in continental-type rocks. However, uranium forms more than 160 mineral species and accounts for about 5% of all known minerals (Smith, 1984; Cejka and Urbanec, 1990). Very high uranium concentrations can be reached locally, e.g. in uranyl vanadate deposits (Garrels and Phoenix, 1966) or in phosphatic rocks. Apatites are known to hold up to 500 mg U kg^{-1} (Komura et al., 1985). There are no isotopic fractionation processes known in nature and natural materials show ^{238}U: ^{235}U ratios of 137.5±0.5 (Rogers, 1969).

Half-lifes, specific activity and relative abundance of the naturally occurring uranium isotopes are given in Table 1. The isotopes ^{235}U and ^{238}U are primordial nuclides, while ^{234}U is a decay product in the 2n+2 series of ^{238}U. Depleted uranium (DU) is stripped from about 70% of ^{235}U and about 80% of ^{234}U (AEPI, 1995). A comparison

Table 2
Physical and radiation properties of uranium isotopes

Isotope	Isotopic composition [%]	Half-life [a]	Specific activity per gram [Bq]	Specific activity of natural or depleted uranium samples
Natural uranium				25446 Bq
238	99.275	$(4.47\pm0.02) \cdot 10^9$	12435	
235	0.720	$(7.04\pm0.01) \cdot 10^8$	79963	
234	0.0054[a]	$(2.455\pm0.006) \cdot 10^5$ a	$2.303 \cdot 10^8$	
Depleted uranium (DU)			specific activity per gram DU [Bq]	14880 Bq
238	99.79		12410	
235	0.21		170	
234	0.001		2300	

[a] ^{234}U is a daughter of ^{238}U. The isotopic composition in Table 2 is calculated on the basis of a secular equilibrium between ^{238}U and its progeny. However, due to dissolution, mobilisation and reprecipitation phenomena, natural samples may show considerable disequilibrium (Holden, 1989).

of specific activities of a uranium sample of natural composition and a DU sample is shown in Table 2. The calculated activities of natural uranium do not include the non-uranium progeny and the activity of natural uranium may triple for the β decay of ^{234}Th, and ^{234m}Pa and the α activity of ^{230}Th and ^{226}Ra. The contribution of the lighter progeny may depend on the rate of ^{222}Rn exhalation from the specific natural uranium sample. During the process of uranium refinement and isotopic enrichment, natural uranium is stripped from its daughters. Hence, DU has a considerably lower specific activity compared to uranium of natural composition in secular equilibrium. On the other hand, DU may contain traces of ^{236}U due to use of reactor-irradiated uranium and extremely low concentrations of other actinide isotopes, i.e. ^{239}Pu (Priest, 2001). Using the data from Table 1 (Holden, 1989), activity ratios $^{234}U{:}^{238}U=1$ and $^{235}U/^{238}U=0.048$ can be calculated, while the respective values for DU are $^{234}U{:}^{238}U=0.18$ and $^{235}U/^{238}U=0.013$.

^{235}U is separated from ^{238}U by processes taking advantage of the small mass differences of the both isotopes. There is no chemical way of separating the isotopes technically. Commonly, uranium is transferred into UF_6, one of the few non-uranyl compounds of hexavalent uranium. UF_6 is sufficiently volatile at elevated temperatures of about 60 °C and there is no other uranium compound with similar usability. UF_6 is highly corrosive and after isotopic separation, the material is stored as uranium metal. The enriched uranium fraction from this process is a highly valuable nuclear reactor fuel (3–6% ^{235}U) or weapon grade material ($>$90% ^{235}U). The DU fraction is about 70% depleted in ^{235}U and it might be used as reactor cladding in fast breeder reactors which transforms it to fissile ^{239}Pu by neutron capture. After the shut-down of almost all fast-breeder programs in the world, DU was used for other applications. Technical applications include X-ray shielding material for hospitals and radiologists' cabinets, counterweights in commercial aeroplanes, yacht keels, satellite ballasts, petroleum exploration drilling equipment, tank armour and the armour-piercing component in some ammunitions (Priest, 2001). In these applications, advantage is taken of the high density of uranium (18.97 g cm^{-3}) compared to iron (7.87 g cm^{-3}). Uranium chemicals are almost exclusively produced from DU. Some DU is also applied in dental porcelain (Sairenji et al., 1980). DU is also applied in military equipment by the USA, UK, Russia, Turkey, Saudi Arabia, Pakistan, Thailand, Israel and France (AEPI, 1994).

3. Chemistry of uranium in natural aqueous systems

There is ample literature describing in detail the chemistry of uranium under conditions of a natural aqueous system (Williams, 1994; Meinrath, 1997; Meinrath et al., 1999). Uranium corrodes rapidly in humid air forming yellow uranyl(VI) compounds, where the linear $[O{=}U{=}O]^{2+}$ entity forms the characteristic structural element. In presence of phosphate(V), vanadate(V), arsenate(V) and silicate, highly insoluble orthophosphate, orthovanadate, orthoarsenate or silicate compounds of hexavalent uranium may be formed, respectively. Representing phosphate and arsenate minerals are autunite $Ca(UO_2)_2(PO_4)_2\cdot 10–12\ H_2O$ and zeunerite $Cu(UO_2)_2(AsO_4)_2\cdot 12\ H_2O$.

A typical vanadate mineral is carnotite $K_2(UO_2)_2(V_2O_8)\cdot 1–3\ H_2O$. Vanadates are considered the most insoluble uranium minerals. Uranophane $Ca(UO_2)_2(SiO_3OH)_2\cdot 5\ H_2O$ is believed to be the most abundant hexavalent uranium mineral (Hostetler and Garrels, 1962).

In the absence of significant amounts of phosphates, arsenates, vanadates and silicates, the chemistry of hexavalent uranium is governed by its interactions with water and carbonate. A larger group of uranyl oxyhydrates, alkali/alkaline earth uranyl oxyhydrates and uranates are known. The affinity of hexavalent uranium to oxygen is extremely great. As a hard Pearson acid, uranium forms compounds preferentially with hard donors like oxygen. There are only very few oxygen-free U(VI) compounds, e.g. UF_6 (Denning, 1992).

While U(V) is known in the laboratory, it is not sufficiently stable for extensive investigations, and is very unlikely to play a significant role in nature. On the other hand, tetravalent uranium is well-known with a pitchblende UO_2 being the most abundant form of uraninite. Tetravalent uranium is strongly sorbing and highly insoluble. Freshly prepared U(IV) compounds show much higher solubility compared to uraninite minerals.

Fig. 2 shows a redox diagram of uranium under conditions typical for contaminated uranium mining areas in Germany (Meinrath et al., 1999). Fig. 2 highlights the pronounced importance of carbonated species in the understanding of the environmental behaviour of uranium. The correlation with redox potential of other environmentally relevant redox elements (Sigg and Stumm, 1996) indicates that U(VI) may be reduced to U(IV) at a high pH under a S(VI)/S(-II) redox regime. The unusual thermodynamic and kinetic stability of bonding between the central uranium and the axial oxygen atoms in the linear UO_2^{2+} entity require a high overpotential to reduce hexavalent uranyl(VI) molecules to tetravalent uranium. Studies on the immobilisation of uranium, e.g. by zero-valent iron, indicate that adsorption on reducing surfaces is an important step in the reduction process (Qui et al., 2000; Baas-Becking et al., 1960). Biological activity may play an important role in the immobilization process, too (Francis, 1999). These processes are subject to current research activities and cannot be answered clearly at present.

Existing gaps in the knowledge of the hydrolysis and carbonate complex formation of U(IV) are shown in Fig. 2 by considering two limiting conditions: a) ignoring carbonate complex formation of U(VI) with $U(OH)_4^o$ as a dominate species, and b) including U(IV) carbonate complex formation with high values. The field '$U(OH)_4^\circ/UO_2^{+}$' gives the calculated stability field of U(V) against $U(OH)_4^\circ$ as relevant U(IV) species (U(IV) carbonato fields must be ignored). If carbonato species are considered as relevant U(IV) species the stability field of UO_2^+ is enlarged comprising both fields '$U(OH)_4^\circ/UO_2^{+}$' and 'carbonato species/UO_2^{+}'. The diagram immediately indicates at what E_H-pH region this knowledge gap takes effect. The relevance of different redox states under environmental conditions can be recognized by comparison with the E_H-pH boundaries of natural aqueous systems, shown in Fig. 2 as a dashed-line, boomerang-shaped field (Baas-Becking et al., 1960).

The redox diagram, however, is derived from thermodynamic data and can display only those information included in the construction of the diagram. It is necessary

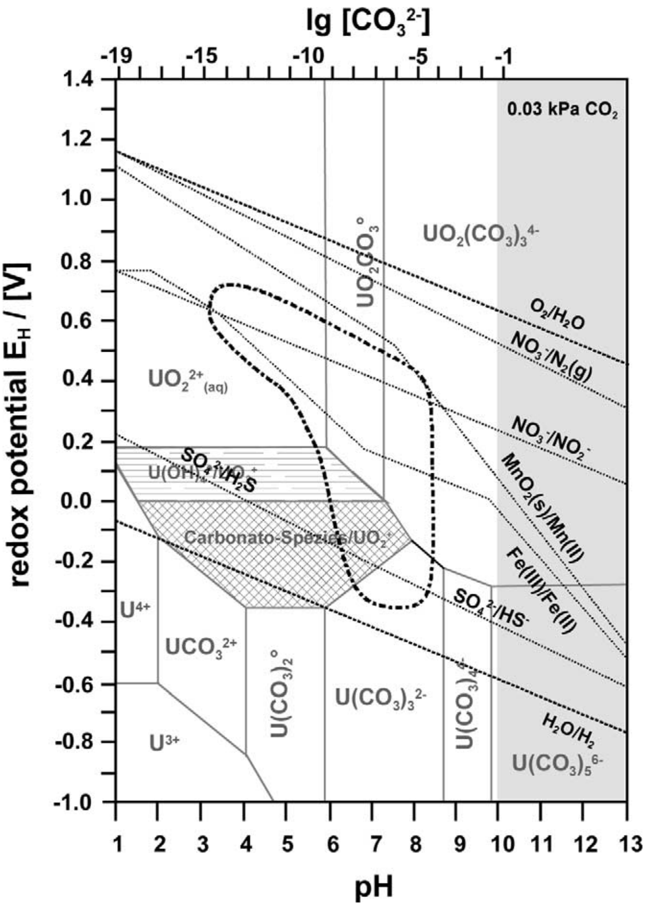

Fig. 2. E_H-pH diagram of uranium compared to the stability field of natural aqueous systems (Baas-Becking et al., 1960) and several environmentally relevant redox couples.

to note that the U(VI)/U(IV) redox process involves formation and breakage of two strong metal-to-oxygen bonds. This process leads to a considerable irreversibility of the redox process. Hence, a redox diagram can predict the presence of U(IV) in a respective E_H-pH region, whereas in nature the formation of this redox state is inhibited by the large overpotential required for breaking the strong bonds with the uranyl oxygen. The difficulty of accounting for the overpotential of bond breakage in the evaluation of redox standard potentials especially in a situation where the species under study (here U(V)) is at the detection limit may be a reasonable explanation for our lack of precise knowledge in this region of the E_H-pH diagram of uranium.

The diagram shows the instability of metallic uranium with respect to corrosion. Uranium metal will be transformed into U(VI) in most soils, sediments and aqueous systems. This process is usually rapid.

4. Uranium in natural aqueous systems: river, lakes and sea water

Uranium is introduced into natural aqueous systems by weathering of rocks (Palmer and Edmond, 1993) and by phosphate fertilizers (Mangini et al., 1979). Plutonic rocks hold higher concentrations, while sediments are usually lower. Due to the high tendency of uranium to be mobilized, i.e. by carbonate and phosphate, the uranium content in aquatic systems may vary considerably. From a comprehensive study using the Arsenazo III photometric method, uranium content ranging from $4{\cdot}10^{-3}$ % to $522{\cdot}10^{-3}$ % has been reported for ores and rocks (Singer and Matucha, 1962). Phosphate fertilizers may contain up to 200 mg U kg^{-1}. The U:P ratio is reported to be relatively constant at 1:1300 (Spalding and Sackett, 1972), although this ratio increases due to consumption of phosphate by plants. Uranium in the aerated region of soils is reasonably assumed to be in its hexavalent state as linear UO_2^{2+} ion. In the absence of other significant amounts of other ligands, i.e. arsenates, vanadates, silicates and phosphates, hydrolysis and carbonate complex formation are reasonably expected to be the predominant reactions of U(VI). Complex formation with carbonate can be spectroscopically observed at free carbonate concentration level of about 10^{-11} mol L^{-1} (Meinrath and Klenze, 1996). Taking the typical pH range of natural aqueous systems between pH 4 and pH 9 and the relative atmospheric CO_2 partial pressure of 0.03%, a highly mobile carbonated species of uranium is formed. Thus, uranium is eluted from soils to rivers and lakes. The total annual world-wide river run-off is estimated to be 20,000 tons. Sea water with an almost constant uranium concentration of 3.3 μg L^{-1} is a major deposit of uranium (Mangini et al., 1979; Bloch, 1980). Hence, a km^3 of sea water contains about 3 tons of uranium. The ocean seas contain a total of more than 4·109 tons of dissolved uranium.

The uranium concentrations in river water has been studied several times. The average uranium concentration in river waters is given as 0.2 μg kg^{-1} (Palmer and Edmond, 1993), 0.3 μg kg^{-1} (Bertine et al., 1970; Turekian and Chan, 1971) and 0.6 μg kg^{-1} (Sackett et al., 1973). These values also indicate variability due to differences in the river samples, the time of sampling and sample treatment. In particular, the Brahmaputra River has a high uranium content, and the inclusion or exclusion into/from a sample gives rise to a variation in the mean value of 50% (Palmer and Edmond, 1993). Some data on natural background values in Saxonian rivers are summarized in Table 3. In Fig. 3, the uranium run-off in some selected rivers of the world is given where the extreme position of the large rivers Brahmaputra and Orinoco are clearly evident. However, attention is directed to the fact that even in the smaller European rivers, an hourly runoff of several kilograms of uranium takes place.

Measurements of uranium contents in Saxonian rivers and lakes are scarce for the uranium production period. A sampling in 1977 (under the auspices of the former GDR government and the Soviet Union authorities) reported an average of 2.9 mg U L^{-1} (Schröder et al., 1997). These concentrations are much higher compared to the average concentrations in worlds' rivers (cf. Fig. 3) and illustrate the long-term release of uranium from production sites and tailings to the unprotected environment.

Table 3
Natural uranium background of surface and ground waters in Saxony (Freistaat Sachsen, 1997)

Location	Type	^{234}U [Bq L^{-1}]	^{238}U [Bq L^{-1}]
Klipphausen	surface water	0.061–0.096	0.045–0.065
Markersdorf	surface water	0.003–0.0014	0.0014–0.003
Reichenbach (near Siebenlehn)	surface water	0.098	0.071
Bad Schandau	surface water	0.024–0.035	0.013–0.025
Dommitsch	surface water	0.022–0.027	0.022–0.027
Zehren	surface water	0.028–0.03	0.023–0.024
Deschka	ground water	$<$0.0005	$<$0.0005–0.0008
Radebeul	ground water	0.0011–0.0012	0.0011

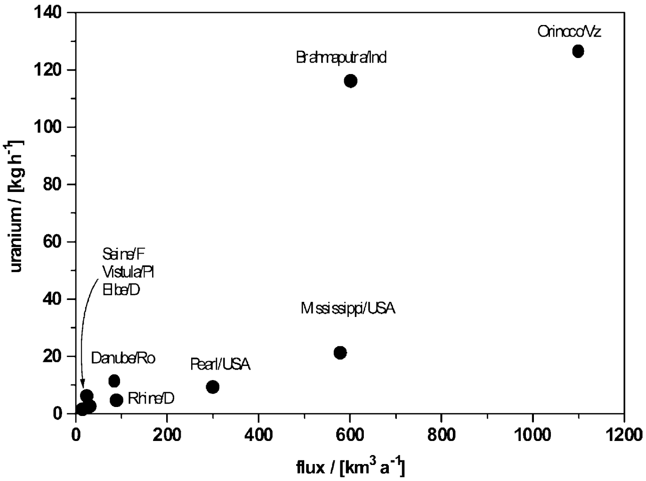

Fig. 3. Amount of uranium per hour transported by some selected rivers (Palmer and Edmond, 1993).

However, seasonal variations are expected to be considerable. Some data showing uranium in rivers after 1989 are summarized in Table 4. Table 5 gives uranium contents observed in tailings, mine waters and dump waters.

Close to sedimentation ponds and tailings, small rivers may carry up to 300 kg uranium per year. It should be kept in mind that these amounts are measured after the shut-down of the facilities and the population in the areas next to the rivers has diminished since 1989.

5. Health effects of uranium

It may be surprising to see that the contamination of a densely populated area with the radioactive and toxic heavy metal uranium did not have greater health effects. Notwithstanding more detailed systematic health studies currently in process, the health effects observed to date are attributed to radon and silicosis (lung cancer), dust, smoke and noise (the mining machinery was driven by pressurized air), ammonium and diesel engine exhausts. The reason for this observation may be at least, partly understood by the metabolism of uranium in the human body.

The human body contains about 100 μg uranium from the ingestion of food and drinking water; with a daily intake of about 1.5 μg (Dang et al., 1995). Up to 2% of orally administered uranium is resorbed, while 90% is excreted within 24 h (Priest, 2001). The remainder is excreted in the following days and only a small percentage is retained in the body. The form with highest bioavailability is UO_2^{2+}. Uranium circulates bound to erythrocytes and forms a series of compounds with biomolecules and proteins (Taylor, 1989). It behaves similarly to the alkaline earth elements and is co-deposited with calcium in bones. Bone marrow, however, is free of uranium. There is no confirmed evidence of mutageneity due to uranium.

Table 4
Uranium concentrations in rivers and lakes of Saxonian uranium areas after 1989 (Beuge et al., 1995; Helling et al., 1996)

River	Flow/volume [m^3/s]	Dissolved uranium [µg L^{-1}]	Particle bounded uranium [mg kg^{-1}]	Sediment
Zwickauer Mulde (ZM)	1200	23		124 mg kg^{-1}
ZM near Albernau	1.2	6.9	231	
Schwarzwasser	2	≈15	16	
Lößnitzbach	0.16	≈15	145	
Schlemabach	0.12	25.1	114	
Markus-Semmler Stolln	0.005	32.3	2008	
ZM near Niederschlema	3.29	77.4	1068	
Kohlungbach	0.02	600	106	
ZM near Hartenstein	3.33	27.5	673	
Wildenfelser Bach	0.13	69	39	
ZM near Wiesenburg	3.6	36.4	677	
Rödelbach	0.56	≈15	38	
ZM near Zwickau-Pölbitz	4.47	13.5	280	
ZM near Crossen	4.6	16.2	337	
Oberrothenbacher Bach	0.03	354	706	
ZM near Oberrothenbach	4.67	24.4	502	
Zinnbach	0.02	579	45	
Mülsenbach	0.23	62	64	
ZM near Glauchau	4.98	23.7	373	
Lungwitzbach	0.96	16.2	82	
Reservoir Glauchau		30–80[a]		78 mg kg^{-1}
ZM near Jerisau	5.69	15.6	202	
Kaitzbach (Dresden/Gittersee)		0.12–0.015 Bq L^{-1} ^{238}U		

[a] Uranium concentrations in water are correlated with plankton biomass.

Table 5
Uranium contents in selected tailing ponds, mine waters and processing wastes

Site	Type	Amount	Reference
Dresden/Gittersee	Tailings	0.04–1 mg L^{-1}	(Helling, 2000)
Dresden/Gittersee	Tailings	9.44 Bq L^{-1}	(Helling et al., 1996)
Dresden/Gittersee	Tailings	0,53–11,86 Bq L^{-1}	(Helling et al., 1998)
Crossen	Tailings	^{238}U: 1630 Bq kg^{-1} ^{235}U: 154 Bq kg^{-1}	(Schimmanck et al., 1998)
Königstein	dump water	20–30 mg L^{-1}	(Schneider et al., 1999; Schneider et al., 2001)
Shaft Pöhla	mine water	0.22–0.26	(Fleischer et al., 1998)
Shaft Schlema-Alberoda	mine water	5.6 – 6.4 mg L^{-1}	(Schimmanck et al., 1998)
Shaft 216 Niederschlag	mine water	1.5–1.9 mg L^{-1}	(Schneider et al., 2001)
Glückauf shaft Johanngeorgenstadt	mine water	0.011 mg L^{-1}	(Schneider et al., 2001)
Johanngeorgenstadt Steinsee	Tailings	275 mg L^{-1}	Junghans and Helling, 1998
Schneckenstein	Tailings	193 mg L^{-1}	Junghans and Helling, 1998

The target organs are the kidneys, as for other heavy metals like cadmium, lead and mercury. The kidney toxicity in mammals is caused by the precipitation of hexavalent U mostly in the kidney tubules during the urinary blood clearing processes, but also in the glomerulous and other areas of the tubules (Hursh and Spoor, 1973). The resulting tissue damage leads to kidney failure with all typical pathologic findings in the blood and urinary system. However, terminal renal failure demanding lifetime dialysis has not been reported in humans. No exact dose-effect relationship is known for transient or permanent renal insufficiency. It is estimated that inhalation of about 40 mg soluble uranium is the threshold for inducing permanent renal injury, and about 8 mg is required for transient renal damage (Gilliland et al., 2000).

DU dust may be formed when DU ammunition hits a hard target, and the heat of combustion or weathering may oxidize these small particles. Depending on the size of the particles, uranium can be inhaled. The uranium present in the air is of no importance, less than 1% of the inhaled uranium reaches the kidneys, as a result of the body's efficient clearing system (ICRP, 1995). Absorption in the lung is rapid for soluble compounds, but may take years for oxides in particulate form.

Despite being a toxic heavy metal comparable to lead, no drinking water regulations exist for uranium in Germany and many other countries. Due to its use in nuclear reactors and strategic weapons, uranium is almost exclusively seen according to its strategic importance. Other aspects, i.e. its chemical toxicity as a heavy metal, play a lesser role. German regulations for radiation protection set the limit for the handling of uranium without license to an activity of $5 \cdot 10^5$ Bq; this corresponds to 0.197 kg of uranium in the natural isotopic ratio. For a daily intake, no regulation exist in Germany. The World Health Organization (WHO) has established a tolerable daily intake for uranium of 0.6 $\mu g\ kg^{-1}$ body weight. For drinking water, WHO suggests 2 μg uranium L^{-1}. Uranium mill tailings in Saxony may carry up to 30 mg U L^{-1} (Knoch-Weber, 1998). Those extreme values however occur only in small water samples due to concentration effects by evaporation etc.

The major concern of health effects due to uranium is its chemical toxicity. The radioactivity is not strong enough to cause health effects. With respect to chemotoxicity, uranium and DU are indistinguishable. No evidence has been found showing an association between uranium uptake and the overall death rate (Brown and Bloom, 1987; Checkoway et al., 1988), adverse effects on the nervous system (Brown and Bloom, 1987; Carpenter et al., 1988), hepatic effects (Spencer et al., 1990), body weight (Dygert, 1949), immune system (Cragle et al., 1988), haematological change (UNSCEAR, 1986) or reproductive disorders (Vich and Kriklava, 1970). No data exist concerning effects on the gastrointestinal tract, the skin, and the musculoskeletal and cardiovascular systems. However, a 7-year follow-up of Gulf War veterans exposed to DU, showed significant changes in serum prolactin levels and uranium concentrations in semen. In addition, a statistically significant relationship was found between elevated uranium urine concentrations and reduced performance on automated tests assessing performance, efficiency and accuracy (McDiarmid et al., 2001). Birth defects have not been reported for the DU exposed group.

Radiological toxicity of uranium and DU results from its α-, β- and γ-emission. The long-term effects include a risk for cancer and effects on descendants. Negative effects resulting from radiation have not been found in humans. There is proof of lung cancer in underground miners, resulting from a combination of exposure to radon, short-lived decay products and other inhaled toxic agents, such as silica dust, diesel fumes and cigarette smoke (Poldenak et al., 1982). Uranium mill workers have not shown an increase of lung cancer. There is a theoretical risk of osteosarcoma, as uranium is deposited in the bones, however no association was reported (Cragle et al., 1988; Poldenak et al., 1982). Irradiation doses for a soldier in a DU armed tank would reach the annual limit for the general public after 70 days of 8-h shifts. The dose to flight attendants and passengers in an aircraft containing DU counterweights is less than a thousandth of that, due to cosmic radiation.

6. Conclusions

As a contribution to the discussion of possible health effects of DU ammunition after short term exposure during war activities, attention is directed to two aspects of uranium impact on man: a) the natural, almost ubiquitous distribution of uranium in the environment, often enhanced by application of phosphate fertilizers and b) the exposure of workers and the general public to uranium released, in part, by careless exploitation and processing of uranium in Saxony, Germany. Furthermore, experience with 500 years of mining in the crystalline rocks with elevated geogenic uranium background is available in Saxony. Health effects due to radon are well-known but prevalent health effects due to uranium itself have not been described.

Generally, it should be kept in mind that 40 fired DU ammunitions of about 10 kg DU result in soil uranium concentrations equivalent to the natural uranium concentrations (Meineke, 2001). The theoretical dose resulting from 20 years of use of phosphate fertilizers is equivalent to the dose from DU ammunitions dropped in the same 20,000 km^2 area during Gulf War. Claimed health effects must be compared to the long-term exposure of the population and workers in the uranium mining areas in Saxony, Germany during the past 50 years. Due to the high geogenic uranium background, all subsurface mining activities prevailing in the Ore Mountain area for almost 500 years, leads to exposure to uranium. However, no adverse effect is known to be caused by uranium; lung cancer (‘Schneeberg disease’) is a result of radon inhalation.

DU is less radioactive than natural uranium. Hence, DU cannot be claimed to have more severe radioactive impact on the human body than natural uranium. With respect to chemotoxicity, natural uranium and DU are indistinguishable. Claims of adverse health effects explicitly caused by short-term exposure to depleted uranium must be weighted against the 500 years of experience with subsurface mining activities in an area with increased geogenic background concentrations of uranium and a rather careless and partly irresponsible exploitation and processing of uranium ores which affected one hundred of thousand subsurface miners and inhabitants of a densely populated area over almost 60 years. The human has an efficient clearing system for uranium. Only minor quantities are retained in the human body, and an overwhelming percentage is excreted rapidly. This also includes uranium quantities that remain in the muscles of those soldiers hit through friendly fires (McDiarmid et al., 2000). During the Gulf War, 24 American armoured vehicles were hit by friendly fire with a total of 29 exposed veterans. Nine veterans have embedded DU shrapnel.

References

AEPI, 1994. Health and Environmental Consequences of Depleted Uranium Use by the US Army. Summary Report to Congress, US Army Environmental Policy Institute, June 1994.

AEPI, 1995. Health and Environmental Consequences of Depleted Uranium Use in the US Army. Technical Report, US Army Environmental Policy Institute, Atlanta/USA June 1995.

Baas-Becking, L.G.M., Kaplan, I.R., Moore, D., 1960. J. Geol. 68, 234–284.

Bertine, K.K., Chan, L.H., Turekian, A., 1970. Geochim Cosmochim Acta 34, 641.

Beuge, P., Hoppe, Th., Klemm, W., Kluge, A., Knöchel, A., Starke, R., 1995. Der Einfluß des Uranbergbaus auf die Spurenelementgehalte der Zwickauer Mulde, In: Merkel, B., Hurst, S., Löhnert, E.P., Struckmeier, W., Uranium Mining and Hydrogeology. GeoCongress Verlag Sven von Loga Köln/FRG 1, 25–34

Bloch, S., 1980. Geochim. Cosmochim. Acta. 44, 373–377.

Brown, D.P., Bloom, T., 1987. Mortality Among Uranium Enrichment Workers. Report to the National Institute for Occupational Safety and Health, Cincinnati/USA.

Carpenter, A.V., Flanders, W.D., Frome, E.L., 1988. Am. J. Ind. Med. 13, 351–362.

Cejka, J., Urbanec, Z., 1990. Secondary Uranium Minerals. Transactions of the Czechoslovak Academy of Science. Math.-Natur. Sci. Series 100, pp. 93. Academia Prague/CZ.

Checkoway, H., Pearce, N., Crawford-Brown, D.J., 1988. Am. J. Epidemiol. 127, 255–266.

Cragle, D.L., Burnett, W.C., Qualters, J.R., 1988. Am. J. Ind. Med. 14, 379–401.

Dang, H.S., Pullat, V.R., Sharma, R.C., 1995. Health Phys. 68, 328–331.

Denning, R.G., 1992. Structure Bond. 79, 215–276.

Dygert, H.P., 1949. Pharmacology and Toxicology of Uranium Compounds. McGraw-Hill, New York, NY, pp. 673-675.

Fleischer, K., Knappik, R., Mende, A., Meyer, J., 1998. Bestimmung von partikulären und gelösten Stoffanteilen im Flutungswasser der Gruben Pöhla und Schlema-Alberoda. In: Merkel, B., Helling, C. (Eds.), Uranium Mining and Hydrogeology. GeoCongress Verlag Sven von Loga Köln/FRG 5: 95–103.

Flesch, K., Nitzsche, O., Merkel, B., 1998. In: Merkel, B., Helling, C. (Eds.), Uranium Mining and Hydrogeology. GeoCongress II Verlag Sven von Loga Köln/FRG 5, 449–458

Francis, A.J., 1999. Agronomy Monograph 37, 239–271.

Freistaat Sachsen, Sächsisches Staatsminiserium für Umwelt und Landesentwicklung: Umweltradioaktivität in Sachsen, 1997. Annual Report 1996, Materialien zu Strahlenschutz und Umweltradioaktivität 4/1997.

Garrels, R.M., Phoenix, D.A., 1966. Geochemistry and Mineralogy of the Colorado Plateau Uranium Ores. US Gological Survey Prof. Papers 320, 55–64.

Gatzweiler, R., Mager, D., 1993. Geowiss 11, 164–172.

Gilliland, F.D., Hunt, W.C., Archer, V.E., Saccomanno, G., 2000. Health Phys 79, 365–372.

Helling, C., 2000. Isotopes Environ Health 36, 211–222.

Helling, C., Nitzsche, O., Ullrich, B., Merkel, B., 1998. Mine Water Environ 17, 41–51.

Helling, C., Nitzsche, O., Hebert, D., Merkel, B., 1996. Hydrogeologische Untersuchungen an einem müllüberlagerten Tailing der Uranerzaufbereitung. In: Merkel, B., Dietrich, P.G., Struckmeier, W., Löhnert, E.P. (Eds.), Grundwasser und Rohstoffgewinnung. GeoCongress Verlag Sven von Loga Köln/FRG 2, 173–178.

Holden, N.E., 1989. Pure Appl Chem 61, 1483–1504.

Hostetler, P.B., Garrels, R.M., 1962. Econ Geol 57, 137–167.

Hursh, J.B., Spoor, N.L., 1973. Data on man. In: Hodge, H.C., Stannard, J.N., Hursh, J.B. (Eds.), Handbook of Experimental Pharmacology, vol 36. Springer, Berlin.

ICRP, 1995. Age Dependent Doses to Members of the Public from Intake of Radionuclides: Part 4 Inhalation Dose Coefficients. Ann. ICRP vol. 25.

Junghans, M., Helling, C., 1998. Proc Tailings Mine Waste 1998, Fort Collins/USA Balkema, Amsterdam/NL.

Knoch-Weber, J., 1998. Braunkohle Surface Mining 50, 347–356.

Komura, K., Yanagisawa, M., Sakurai, J., Sakanoue, M., 1985. Radioisotopes 34, 529–536.

Kreuzer, M., Müller, K.M., Brachner, A., Gerken, M., Grosche, B., Wiethege, T., Wichmann, H.E., 2000. Cancer 89, 2613–2621.

Lieser, K.H., 1980. Einführung in die Kernchemie. Verlag Chemie, Weinheim, FRG.

Mangini, A., Sonntag, C., Bertsch, G., Müller, E., 1979. Nature 278, 337.

McDiarmid, M.A., Keogh, J.P., Hooper, F.J., McPhaul, K., Squibb, K., Kane, R., DiPino, R., Kabat, M., Kaup, B., Anderson, L., Hoover, D., Brown, L., Hamilton, M., Jacobson-Kram, D., Burrows, B., Walsh, M., 2000. Health Phys Environ Res Sect 82, 168–180.

McDiarmid, M., 2001. Brit Medical J 322, 123–124.

McDiarmid, M.A., Engelhardt, S.M., Oliver, M., 2001. Health Phys 80, 270–273.

Meineke, V., 2001. Bay Ärztebl 5, 220–223.

Meinrath, G., 1997. Wiss. Mitt. Geol. Freiberg 4, 100 p.

Meinrath, G., Klenze, R., 1996. Radiochim Acta 74, 81–86.

Meinrath, G., Volke, P., Helling, C., Dudel, G.E., Merkel, B., 1999. Fresenius J Anal Chem 364, 191–202.

Meinrath, G., Hurst, S., Gatzweiler, R., 2000. Fresenius J Anal Chem 368, 561–566.

Palmer, M.R., Edmond, J.M., 1993. Geochim Cosmochim Acta 57, 4947–4955.

Poldenak, A.P., Keane, A.T., Beck, W.L., 1982. Environ Res 28, 313–328.

Priest, N.D., 2001. Lancet 357, 244–246.

Qui, S.R., Lai, H.F., Roberson, M.J., Hunt, M.L., Amrhein, C., Giancarlo, L.C., Flynn, G.W., Yarmoff, J.A., 2000. Langmuir 16, 2230–2236.

Rogers, J.J.W., Adams, U.K., 1969. Uranium-isotopes in nature; Heat Production in Common Rocks. Handbook of Geochemistry 92-B, 1–3.

Sackett, W.M., Mo, T., Spalding, R.F., Exner, M.E., 1973. A reevaluation of the marine geochemistry of uranium. In: Proc Int Symp Interaction of Radioactive Contaminants with the Constituents of the Marine Environment. IAEA, Wien/A, pp. 757–769.

Saxonian Federal Agency of Environment and Geology, 1993. Annual Report.

Sairenji, E., Moriwaki, K., Shimizu, Noguchi, K., 1980. J Dent Res 59, 1136–1140.

Schimmanck, W., Klotz, D., Schramel, P., Bunzl, K., 1998. Leaching of natural Radionuclides and heavy metals from uranium mill tailing material as observed in laboratory and outdoor column experiments, In: Merkel, B., Helling, C. (Eds.), Uranium Mining and Hydrogeology. GeoCongress II Verlag Sven von Loga Köln/FRG 5: 310–317.

Schneider, P., Osenbrück, K., Nindel, K., Voerkelius, S., Forster, M., Schreyer, J., 1999. Current and future impact of an uranium mine waste disposal site on groundwater. In: Proceedings of "Isotope Techniques in Water Ressources Management", Vienna. International Atomic Energy Agency.

Schneider, P., Neitzel, P.L., Osenbrück, K., Noubactep, C., Merkel, B., Hurst, S., 2001. Hydrochimica et Hydrobiologica Acta 29, 1–10.

Schneider, P., Voerkelius, S., Nindel, K., Forster, M., Schreyer, J., 2001. Release of Contaminants from Uranium Mine Waste—Laboratory and Field Experiments. Mine Water Environ, 20, 30–38.

Schröder, C., Friedrich, K., Otten, H., Willmann, G., 1997. Materialien zu Strahlenschutz/Umweltradioaktivität 3/1997 Ministry for Environment and Development, Dresden/FRG [in German].

Sigg, L., Stumm, W., 1996. Aquatische Chemie. Teubner, Stuttgart, FRG.

Singer, E., Matucha, M., 1962. Z. Anal. Chem 191, 248–253.

Smith, D.K., 1984. Uranium mineralogy. In: de Vito (Ed.), Uranium Geochemistry, Mineralogy, Geology, Exploration and Resources. The Institute of Mining and Metallurgy, London UK, pp. 43–88.

Spalding, R.F., Sackett, W.M., 1972. Science 175, 629–631.

Spencer, H.S., Osis, D., Fisenne, I.M., Perry, P., Harley, N.H., 1990. Radiation Res 24, 90–95.

Taylor, D.M., 1989. Sci Total Env 83, 217–225.

Turekian, K.K., Chan, L.H., 1971. In: Brunfeldt, Steines (Eds.), Activation Analysis in Geochemistry and Cosmochemistry. Stockholm University Press, 311.

UNSCEAR, 1986. Genetic and Somatic Effects of Ionizing Radiation. Report to the General Assembly New York, NY.

Vich, Z., Kriklava, J., 1970. Br J Ind Med 27, 83–85.

Williams, P.A., 1994. Oxide Zone Geochemistry. Ellis Horwood, Chichester, UK.

Wolkersdorfer, C., 1996. Hydrogeochemische Verhältnisse im Flutungswasser eines Uranbergwerks—Die Lagerstätte Niederschlema/Alberoda, Clausthaler Geowiss. Diss., Heft 50.

Zeman, A., Benes, P., 1995. Radiochim Acta 70/71, 23–29.