225 years uranium and radioactivity cross-links around the Brocken – Klaproth, Elster and Geitel, Nazi research, Wismut prospection, and recent anomalies

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Abstract. When Martin Heinrich Klaproth, born in Wernigerode, presented the discovery of “uranite”, he could not know which role this new element would play. In 1823, Alexander von Humboldt published the idea of the “Classic Geological Square Mile” later used for parts of the Northern Harz area because of their geodiversity. Between 1880 and 1920, the two physicists Julius Elster and Hans Friedrich Geitel worked in the Harz area – Geitel coined the term “atomic energy” in 1899. A cluster of industry and the Mining Academy Clausthal developed in this area because of the diversity of mineral deposits and mining. In the Nazi era, this cluster conducted military research and partly also worked on the possible use of uranium (U) for weapon production. The German U reserve was placed in Staßfurt. After WW II, Wismut carried out prospection work in the Harz region, but no U deposits were found worth mining. The U source of an anomaly in surface waters within the agricultural landscape Magdeburger Börde is probably to be found in the intensive and long-time use of mineral phosphate fertilisers.

Introduction

Due to the high geodiversity of the Harz Mountains and their forelands, mining of different ore types and subsequent processing and smelting played a key role in this northernmost middle mountain range of Germany. Mining started as early as in the bronze age more than 3000 years ago (Monna et al. 2000) and lasted until 2007. For decades, U was also prospected for, beginning after WW II.

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1789: Martin Heinrich Klaproth discovers U

The chemist Martin Heinrich Klaproth was born in the Harz city of Wernigerode in 1743. When he discovered "uranite" in his Berlin laboratory following studies on pitchblende (Klaproth 1789) and presented it to the scientific community, he could not know how important this new element, later renamed "uranium", would become. The house where Klaproth was born still exists and can be found at Liebfrauenkirchhof No. 5 in Wernigerode.

1823: Classic Geological Square Miles

It was Klaproth's later university patron Alexander von Humboldt who coined the idea of the "Classic Geological Square Miles" for parts of Central Germany in 1823 (Humboldt 1823). The acreage of the classic geological square mile corresponds with the Prussian square mile and represents an area rich in geological diversity. This might be the reason why the term was used more and more for parts of the Northern Harz area northwest of the Brocken (Geopark Harz . Braunschweiger Land . Ostfalen 2013).

Right through the city of Klaproth's birth and near Wernigerode Castle runs the Northern Harz Boundary Fault, a deep tectonic structure. Extending to the north, the sediments of the Subhercynian Cretaceous Basin stretch out over an area where Wismut geologists in later years were prospecting for another U deposit of the Königstein type.

South of this fault, the Harz basement block crops out, consisting of rocks from the Paleozoic. Slates and greywackes with partly embedded flinty slates and limestones emerged from the oceanic sedimentation of the Devonian and the Lower Carboniferous, accompanied by volcanic activities in different phases. At the turn Lower/Upper Carboniferous, the Variscan collision folded these sediments and led to a regional metamorphosis. By this plate collision, rocks were pressed down in the deeper earth's crust, melted and rose upwards as magma, until the intrusive bodies became stuck and cooled down. In the course of these processes the Brocken Granite and Gabbro complex emerged. Wismut geologists hoped that the granite contains mineable U minerals, but failed.

The Harz fold-belt has then been partly eroded in the Lower Permian (the Rotliegend Formation bearing U minerals) and, in the Upper Permian, for a time was flooded by the Zechstein ocean (the Kupferschiefer Formation also containing U minerals).

In the course of the Mesozoic, massive sediment layers developed on the folded Harz rocks – for the most part under the cover of the sea – such as clay, sandstone and limestone of the Triassic, Jurassic and Cretaceous. Since the Upper Cretaceous and further in the Tertiary, the half-horst of the Harz basement has risen for more than 5000 m in the wake of the Saxonian tectonics along the Northern Harz

Boundary Fault. At the same time, mesozoic sediments north of the Harz basement were dragged upwards and turned locally in an upright position so that their once horizontal stratification planes now stand vertically, causing the high geodiversity in the region described by many authors. Following the pre-Saxonian and Saxonian tectonics, ore mineralisations partly filled the tectonic structures, mostly under Zechstein coverage (Franzke and Schwab 2011).

Due to this high geodiversity, ore mining, processing and smelting and later also rock quarrying have played a key role in the Harz area, for more than 3000 years now. Regional examples include the mining of copper, silver-bearing lead and zinc minerals (the mines of Rammelsberg, Clausthal-Zellerfeld, Sankt Andreasberg and Mansfeld being the internationally best known examples), quarrying of anhydrite, gypsum and dolomite along its southern edge, and paleobasalt, gabbro, granite, reef limestone, roofing slate and greywacke throughout the region's Paleozoic area. Originally, human impact was only small-scale, but over the course of mining history, alongside increase in technological possibilities, mining and smelting began to have increasingly serious effects on the surrounding nature and landscape (Knolle et al. 2011). Since the 1950s, the whole eastern part of the Harz Mts. was screened repeatedly by Wismut prospectors and geologists for radioactive anomalies (see below).

1899: The term "Atomic Energy" was formed in the Harz area

Both the teachers and physicists Julius Elster (*1854 in Blankenburg/Harz, † 1920 in Bad Harzburg) and Hans Geitel (*1855 in Braunschweig, † 1923 in Wolfenbüttel) were born in the Harz foreland and spent their working lives there. They faced very limited technical facilities, but provided important basic research in the field of radioactivity, atmospheric electricity, photoelectric effects and gas discharge physics (Fricke 1992). Elster's and Geitel's most important scientific achievement is the discovery of the photo cell. With the knowledge that photoelectricity produced was proportional to light intensity, they provided basic data for modern photoelectric photometry. A major focus of Elster and Geitel lay on atmospheric electricity – their theory of the electronic charging in thunder clouds through the friction of water droplets can still be found in physics textbooks. For use in describing the distribution of charges, Elster and Geitel developed the model example of a spherical capacitor. They introduced a new era of research when they applied the theory of ions to the field of atmospheric electricity. For a long time they hypothesised that sunlight induces the release of ions before discovering that environmental radioactivity is causative. To prove this theory, they carried out underground measurements in the Baumann's and Iberg Dripstone Caves in the Harz. In the end, they provided evidence of a higher radioactivity inside the natural caves than in the air outside. In 1907, the two scientists concentrated on the relationship of radioactivity and geothermal energy. Elster and Geitel were the first to view radioactivity as a phenomenon of atomic disintegration processes and to postulate a radioactive decay-chain of elements. They also analysed cathode rays and had glass cells produced, in which the electrode had the form of a filament which could be heated electrically. With that, Elster and Geitel created hot-cathode tubes which are nowadays irreplaceable in electronics. In recognition of their scientific accomplishments they were repeatedly honored and even nominated seven times for the Nobel Prize for Physics (Fricke 1992).

Since 1933: Nazi atomic research and development, U storage

The Harz area is known for having had the largest underground weapon production site of its time – the KZ Mittelbau-Dora installations near Nordhausen at the southern Harz rim. Less known is the fact that the diversity of ore deposits in this area brought a cluster of mining and metallurgy related industry to the area. Some mining facilities cooperated intensively with the Bergakademie Clausthal (later Technical University Clausthal) and other universities. During the Nazi regime, they also carried out military research, and several companies and institutes worked on the possible use of U in weapon production in WW II.

Only fragments of this research work are known so far. Prof. Dr.-Ing. Max Paschke, who was arrested after WW II due to his Nazi implications, carried out experiments to produce graphite-free U carbide in his Institute for Ferrous Metallurgy, Foundry and Enamel Technology of the Bergakademie Clausthal (Halstead Exploiting Centre 1945). U carbide (UC) can be used as nuclear fuel or as a substitute for tungsten and molybdenum in steels. The experiments were suggested by Gebr. Borchers A.G. in Goslar – their CEO Dr. Friedrich Borchers was under house arrest after WW II because of Nazi collaboration (Knolle and Schyga 2012). This document from 1943 was later found by the Allies within the secret reports from 1939 - 1945 of German atomic research („Haigerloch Documents“). The Bergakademie Clausthal also cooperated with other firms in the region on projects involving German atomic research – alongside the Grona GmbH (high pressure and hollow charge research), the Phywe AG (ultra-zentrifuges) was also involved, both situated in Göttingen (Baranowski 2013).

Besides this, in Staßfurt in the northeastern Harz foreland U supplies were stored. It was the stock of approximately 1,100 t of U ore and U oxide (yellow cake) which the US military secured in April 1945 during a commando action in Staßfurt before the Red Army occupied the storage area at the Friedrichshall shaft. This stock represented the Belgian and French U reserves which had been transported to the Harz region and which – under surveillance of the SS – were part of the raw materials stock of the Reich.

1949 - 1973: Wismut prospection during the Cold War

After WW II, SAG, later SDAG, Wismut exploited the U deposits of the Erzgebirge, and also carried out intensive prospection work in the whole GDR, including the eastern Harz area. Here, they worked from 1949 to 1973. There were clear geochemical indications for possible larger U mineralisations in this area, like red bed sediments, copper shale and Königstein-type Cretaceous sediments at the rim of the Harz Mts. and a mineralisation of the Bi-Co-Ni formation at the Brocken granite complex near Wernigerode (Fig. 1). However, no U deposits were found worth mining. The largest concentration of U minerals was found in a small deposit bound to the Hornburg Anticline, northeast of Sangerhausen (Stedingk 2002; Runge 2006; see below).

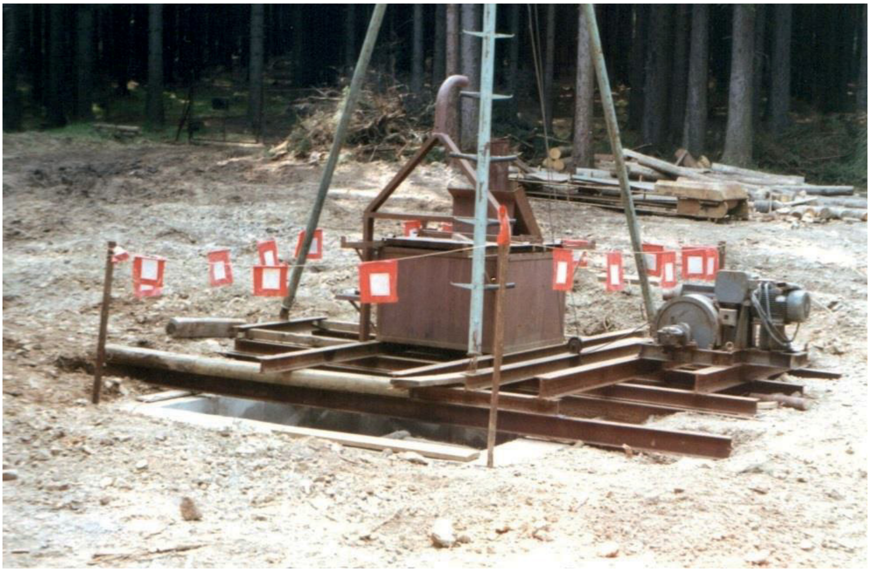

Fig.1. Wismut shaft safekeeping near Drei Annen-Hohne, Wernigerode (photo: Dieter Schulz)

U rock and water anomalies in the Harz forelands

Within the inner Harz Mts., only in the Roter Bär learning mine (Sankt Andreasberg, Lower Saxony), a small uraninite mineralisation is known. The situation is different in the immediate Harz forelands. As mentioned above, alongside the eastern and south-eastern Harz rim a sedimentary complex of Permian rocks crops out in Lower Saxony, Thuringia and primarily in the Mansfeld and Sangerhausen area of Saxony-Anhalt. Especially the copper shales and the Rotliegend red sands contain U minerals. This results in regionally increased U and radioactivity values in runoff water of historic mine galleries and groundwater (Landesamt für Umweltschutz Sachsen-Anhalt 2007). Examples for mine run-off water are Erdeborner Stollen (0.41 Bq/l U-238) and Schlüsselstollen (0.37 Bq/l U-238). The highest groundwater concentrations were found in the regions Mansfeld (30 µg/l U), Hettstedt (100 µg/l U), Eisleben/Wimmelburg (90 µg/l U) and Hornburg (120 - 170 µg/l U).

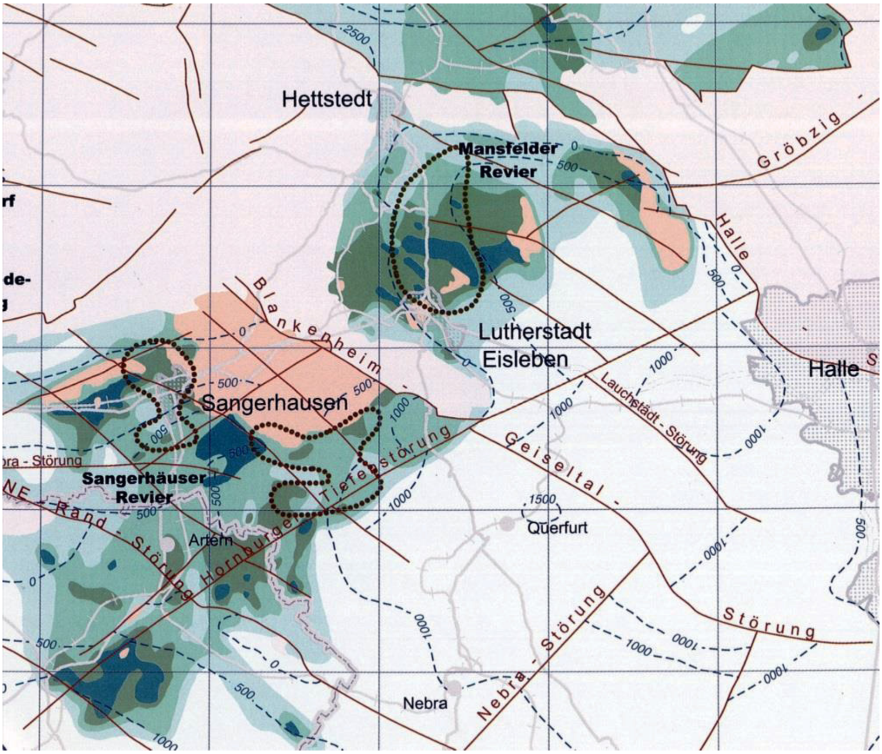

Fig.2. Overview of Mansfeld und Sangerhausen copper shale mining areas showing details of the orebody parameters like copper dissemination, oxidation zones („Rote Fäule"), tectonic structures and position of Zechstein basis; the dotted lines indicate natural radiogene anomalies (from Stedingk and Rentzsch 2003)

These groundwater anomalies are connected to natural radiogene anomalies in copper shales and red molasse sandstones of Permo-Carboniferous age in the Mansfeld and Sangerhausen mining areas (Fig. 2). In the case of the Hornburg orebody, Stedingk (2002) reports for the copper shale a concentration of 180 ppm U and for the molasse sandstone 170 - 260 ppm U resulting in a 400 t U deposit in the sandstone.

The mapping work for the Geochemical Atlas of Germany reveals a U anomaly in surface waters located in the agricultural landscape of the Magdeburger Börde north of the Harz Mts. with a peak value of 7.84 µg/l in the Beber River, north-west of Magdeburg (Fig. 3; Birke and Rauch 2008).

According to Birke and Rauch (2008) and Schnug and Haneklaus (2014), the source of this U is probably to be found in the intensive and long-term use of mineral phosphate fertilisers in the intensively used agricultural areas in the northern Brocken foreland – see also Birke et al. (2009).

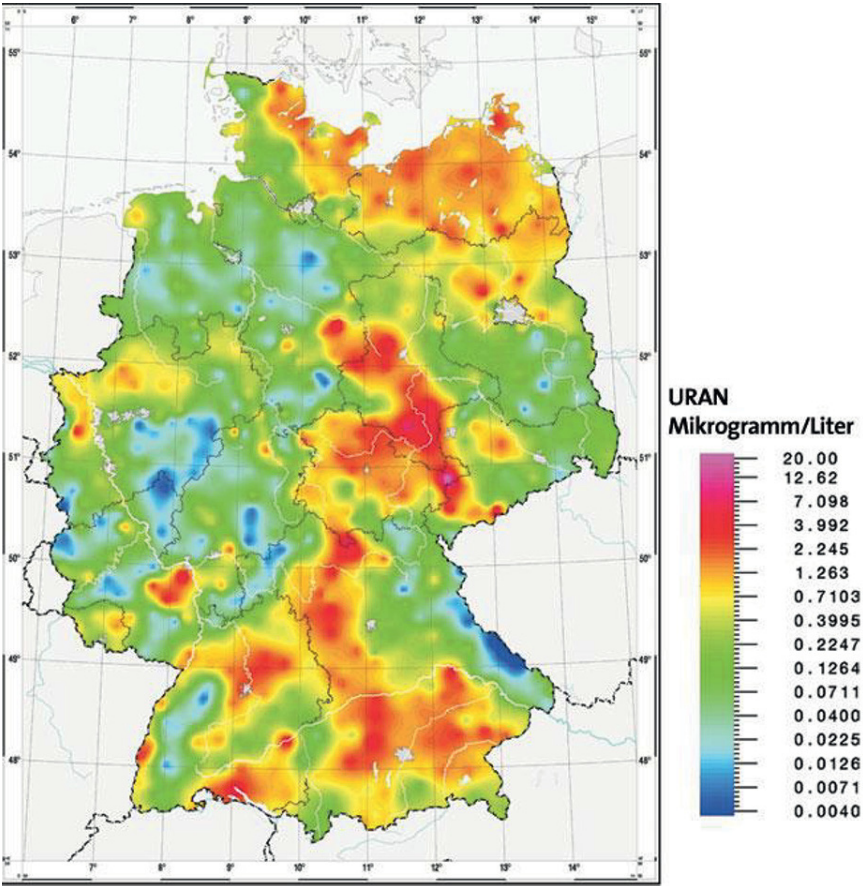

Fig.3. U content in surface waters of the Federal Republic of Germany (from Birke and Rauch 2008)

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