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The importance of radioactivity in geoscience and mining

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Abstract. Almost simultaneously with Roentgen rays, natural radioactivity was discovered. Its investigation led to important fundamentals of the geosciences: petrophysics, terrestrial heat flow, isotope geology, and absolute geological chronology. In applied geophysics and geology, exploration of radioactive ores and of tectonic faults, and radiometric well loggings, are used. Production of radioactive water and mining for uranium ores are discussed, including their development (especially in the Saxon ore mountains) and the consequences on health (radon in underground air and houses). Disposal of radioactive waste is touched on briefly.
Key words. Geophysics; geology; natural radioactivity; radiometry; mining; uranium ores; radon; disposal of radioactive waste.

Introduction

The discovery of the 'X-rays' by Wilhelm Conrad Roentgen in the year 1895 also encouraged the search for natural sources of radiation. In 1896 Henri Becquerel discovered the natural radiation of uranium salts and its relationship to the artifically generated 'Roentgen rays'. In 1898 the Curies succeeded in isolating the element radium from the uranium mineral pitchblende. Natural radioactive radiation consists not only of electromagnetic waves (gamma rays), but also of alpha rays (positively charged helium nuclei) and beta rays (negatively charged electrons).

General importance for the scientific fundamentals of geophysics and geology

During this century radioactivity became very important for the scientific fundamentals of geosciences and also for practical use in applied geology and geophysics and by mining[11]. On average the radioactivity of crystalline (magmatic) rock exceeds that of sedimentary rock. Radioactivity of magmatic rock increases with growing content of SiO_2, i.e. acid rock (e.g. granite) contains more radioactive elements than basic rock (e.g. basalt). Since rock generally becomes more basic with increasing depth, radioactive heat production most probably is concentrated in the upper parts of the earth's crust. On the continents the average geothermal degree of depth is 33 m/K (up to a depth of about 10 km, which is accessible by borehole logging). Locally and regionally there are important deviations from this mean value. The heat flow measured at the earth's surface amounts to 25–126 mW/m^2. Fifty to eighty percent of this value comes from radioactive decay in the earth's crust, and only 20–50% from heat flow out of the upper mantle of the earth[1].

The hypotheses about the development of the earth depend strongly on the significance of radioactive heat production (expansion if the influence of this heat source is high, and contraction if it is low). Modern plate tectonics depend on the assumption of subcrustal convection currents, and their causes are influenced by radioactive heat production.

Modern physical and chemical methods proved that chemically identical atoms often have different quantities of neutrons in their nuclei and therefore have different masses. Some of these elements – called isotopes (55 of 327 naturally occurring elements) – are radioactive, besides elements with atomic numbers ≥ 81, especially potassium and rubidium. By means of mass analysis (mass spectrometry) it is possible to obtain much geological information (origin of rock and deposits, paleo-temperatures of oceans, paleohydrogeology, migration of material, mapping of faults etc.).

An important geological use of radioactive decay (see Roth in this issue) is in the determination of absolute chronology. Unstable elements turn into stable ones directly or through several steps. If decay time (half life period) is known, it is possible to infer the time passed since origin of the rock sample from its content of disintegration products. The most important disintegration chains are:

uranium series $^{238}_{92}U \xrightarrow{4.47 \times 10^9 a} (-8\ He) \rightarrow ^{206}_{82}Pb$

actino-uranium series $^{235}_{92}U \xrightarrow{7.04 \times 10^8 a} (-7\ He) \rightarrow ^{207}_{82}Pb$

thorium series $^{232}_{90}Th \xrightarrow{1.41 \times 10^{10} a} (-6\ He) \rightarrow ^{208}_{82}Pb$

potassium series $^{40}_{19}K \xrightarrow{1.28 \times 10^9 a} ^{40}_{18}Ar$

If it is certain that no helium gas could escape, age determination is also possible from the quantity of He in rock. Besides the above-mentioned decay series the rubidium-strontium series is often used for age determination.

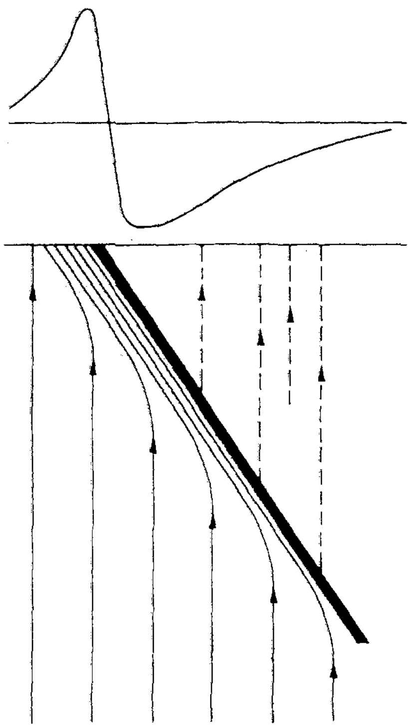


Figure 1. Radioactive anomaly above a tectonic fault (from ref. 1).

Radioactive dating methods give oldest rock an age of 3.6–3.8 billions of years, and the whole age of the earth is assumed to be 4–4.5 billions of years.

The radiocarbon dating method is important for young rock and organic substances up to 70,000 years old. ${}^{14}_{6}C$ results by cosmic radiation and disintegrates into ${}^{14}_{7}N$ with a half life period of 5730 years. Thereby a ${}^{12}_{6}C:{}^{14}_{6}C$-ratio of $10^{12}:1$ results. From radioactivity of tested samples it is possible to estimate their age.

Practical use in applied geology and geophysics

Methods of applied geophysics are very important for geological exploration of mineral deposits[11]. They are based on measurement of physical quantities changing with geological conditions. One of these quantities is natural radioactivity. Radioactive ores close to the surface are recognized by their gamma radiation. Measurements are possible with suitable radiometric instruments (Geiger counters, scintillation counters, semi-conductor detectors) at the earth's surface or from aeroplanes flying at low altitude. Particularly acid rock with a higher content of radioactive minerals generates by disintegration of radium gaseous emanation (radon). This emanation may reach the earth's surface from greater depth via joints and gaps. By this way localization of faults and mineral water becomes possible. As is shown in figure 1, normal vertical migration of volatile radioactive substances along jointed faults is promoted. This is why in the weathered zone above the outcrop of a concealed fault a maximum of radioactivity is generated, whereas above the fault itself, dipping in this example to the right, there is a minimum.

By irradiation of rock with neutrons some nuclides become artificially radioactive (neutron activation). The disintegration of these nuclides results in gamma radiation with characteristic energies. The chemical elements existing in the rock can be deduced from their spectrum (gamma spectrometry).

Radiometric methods are of special importance for well logging. They are also applicable to boreholes cased by iron tubes. Both neutrons and gamma radiation are able to penetrate iron. Therefore in contrast to electrical logging, rock characteristics and depth of boundaries of beds are available in cased boreholes. Of practical importance are gamma-log, gamma-gamma-log, neutron-gamma-log, tracer methods and markers.

Gamma-log: magmatic and sedimentary rock contain natural radioactive elements in variable concentration (e.g. U: 0.003 to 30 ppm). Some daughter-nuclides of these elements generate gamma-quanta of high energy, mostly during beta-ray emission, which are easily registered by gamma detectors in wells. Thickness measurements of layers containing uranium or potassium are of economic advantage. In this way it is possible to discover sand containing oil or water within impermeable clay, because clay beds generally have a higher content of natural radioactive nuclides.

Gamma-gamma-log: after introducing a gamma source (e.g. cobalt 60, 100 MBq) together with a gamma detector into a well, the emitted gamma-quanta act and react upon the surrounding rock. This so-called Compton scattering is proportional to the density of electrons and therefore approximately to the density of rock: density-log.

Neutron-gamma-log: a neutron source emitting neutrons of high energy (5–14 M eV) is introduced into a well, together with a gamma detector. The fast neutrons deliver their energy especially effectively to hydrogen atoms because of the equality of their masses. The excited nuclides emit prompt or delayed gamma quanta. So it is possible to estimate the content of hydrogen in the drilled-through layers.

Figure 2 shows an example of radiometric logging results, from the Zechstein subdivision of Thuringia[13].

Tracer methods: to investigate the flow within permeable layers or within the so-called annular space between rock and casing, radioactively labelled liquids are pressed into a borehole. The horizontal or vertical movement of the 'radioactive cloud' behind the casing is measurable by gamma logging over time.

Marker: for estimating the exact depth of layers the well cable is the 'measuring tape'. If the borehole is deep, the well cable may deliver the wrong depth due to differential stretching. A closed gamma emitter is shot into the rock near the interesting strata, and later on depth estimation is related to this fixed marker.

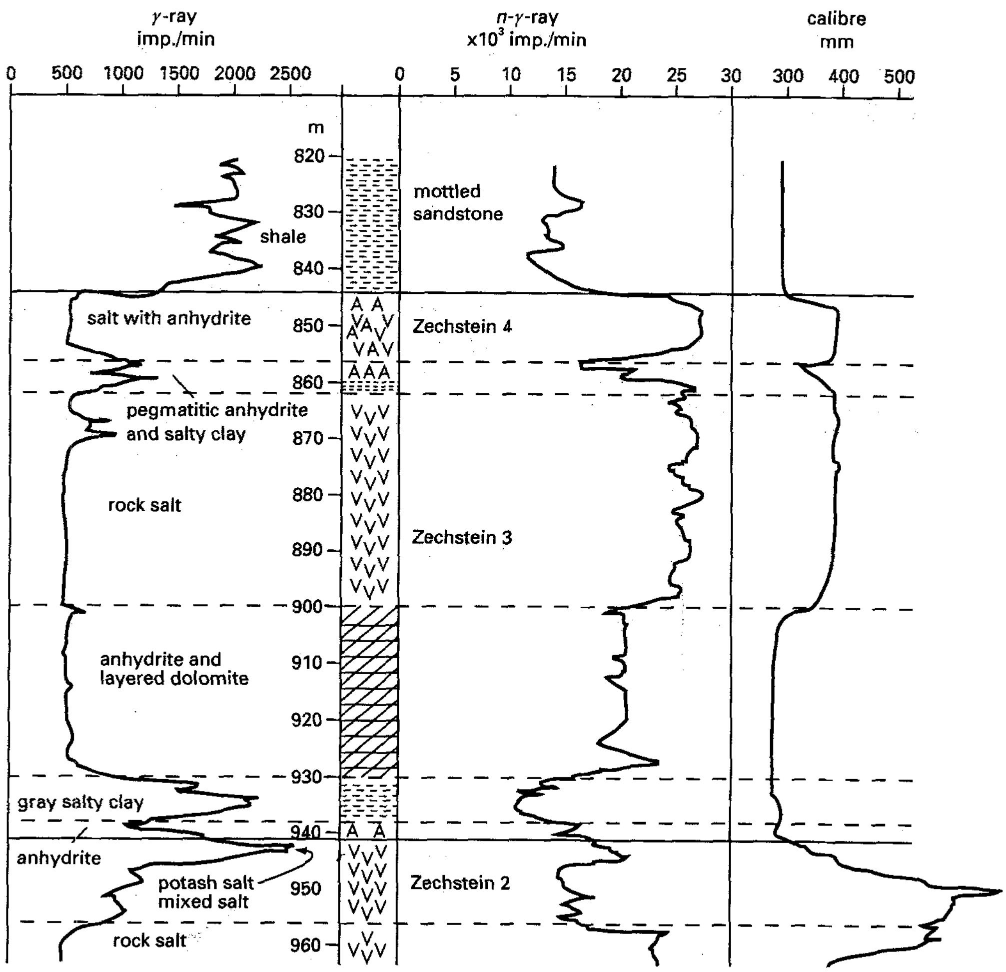


Figure 2. Radiometric logging graphs from the Zechstein subdivision of Thuringia (from ref. 13).

Radiometric well logging is highly technical and expensive, especially in very deep boreholes. But that is necessary, if loss of the drill core is not to be avoided.

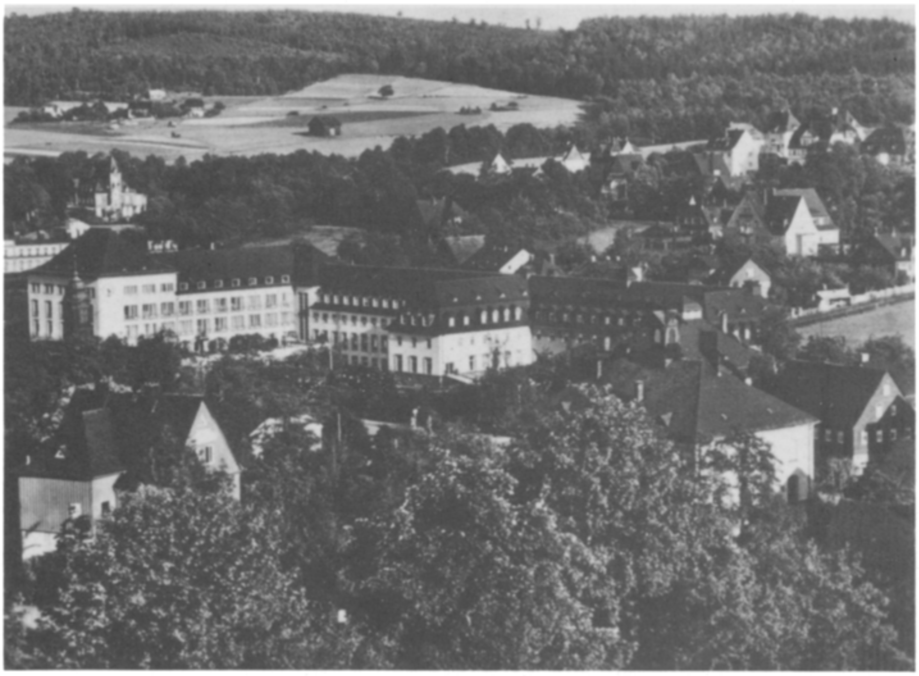
Radioactivity and mining

In the middle German ore mountains, exploitation of precious metals was already taking place in the 12th century[21]. However mining of radioactive ores – at least for use of their radioactivity – only began in this century. During the 19th century uranium ores were only a by-product of mining silver, lead, zinc, bismuth, cobalt and nickel, and they were used for production of colours.

After the discovery of radium from residues of dressing uranium ores in Joachimsthal, there was a growing interest in radioactive water for curative purposes. Joachimsthal in the Bohemian ore mountains was the first radium health resort in the world, in 1906. In consequence in 1908 the Saxon government declared all discoveries of radioactive materials to be the property of the state. From 1908 to 1911 the professors Schiffner and Weidig of the mining academy of Freiberg investigated the radioactive water of Saxony. From 1911 to 1948 in Freiberg a special radium institute existed. Especially successful were many years of measurements by Richard Friedrich, foreman in the blue-colour plants of Oberschlema, who found the highly radioactive waters in the Markus Semmler gallery in the area of Oberschlema. This is a centuries-old culvert of the Schneeberg silver mining area, and has a length of about 44 km (with some branches). It was due to Friedrich that even before World War I to a minor extent, and more officially since 1918 radium cures were possible. At that time radium- or radon-bathing was considered as a miracle cure for many diseases. In 1924 Oberschlema was given the official designation 'radium health resort' and later on it was named 'strongest radium health resort of the world'. Waters of up to 4000 Mache-units (about 54,000 Becquerel of activity by the new nomenclature) were available. The ore mining in the Schneeberg area stopped almost completely during World War II.

After the construction and use of the first atomic bombs in 1945 the need for uranium ores increased rapidly. The Soviet Union intended to break the atomic bomb monopoly of the USA, and therefore in the year 1946 in the already well-known uranium ore districts of the Saxon ore mountains began a new 'Berggeschrei' (demand for mining). Under the name 'Wismut-AG' a mining company of the Soviet state was founded, which later on as 'Soviet-German joint-stock company' (SDAG Wismut), a 'state in the state', executed intensive mining of uranium ores in Saxony and Thuringia. During more than four and a half decades a total of 250,000 tons of uranium were mined[3].

During mining of the ores existing in veins (infilled fissures) and for sampling of the mining product, gamma radiation is measured by Geiger counters or by scintillometers. During the first years of mining, sam-



a

b

Figure 3. *a* View of the well-known radium spa Oberschlema/Erzgebirge (Saxony) with the new cure hotel in 1938 (Photo archives Hans-Guenther Landgraf, Aue and Schlema/Saxonia, Germany, 1938.)
b Devastation of the mountain scenery, the small charming spa and Schlema Valley following uranium mining ('Wismut-AG') and dumping since 1946. (Photo archives H. G. Landgraf, 1952.)

a

b

Figure 4. *a* View from Oberschlema to the famous old silver town of Schneeberg/Erzgeb. before the uranium campaign. (Photo archives H. G. Landgraf, 1937.)
b Same view after the uranium rush between 1946–1959 and restoration efforts. (Photo archives H. G. Landgraf, 28.8.1994.)

ples of the crude ore were taken at fixed intervals and finely crushed. By use of ionization chambers it is possible to determine the radiation intensity, and that from the velocity of electroscopic discharge by the ionized air above the ore sample compared with the same above a standard preparation (etalon). One of the authors (Reinhardt) worked from 1947 to 1949 as a young graduate from a secondary school in an ore-geophysical laboratory of the 'Wismut-AG' with such equipment for measurement of radiation. As a reward he got 'privileges for underground workers' (more food, which was essential for survival in this area at that time!). The absolute values of radiation of course were 'top secret'.

The landscape was markedly altered by the period of ore mining after 1945. From the centuries of silver and cobalt mining nurmerous little waste heaps and buildings for mining use (shaft heads, shaft towers, adit entrances) resulted. They fit into the landscape relatively harmoniously, and those that are preserved are of importance for technical and cultural history[19]. On the other hand during the period of uranium mining big cone-shaped and flat waste heaps grew up within a few years, which totally changed the landscape (figs 3 and 4). In figure 4b they are mostly overgrown.

By the end of the 1950s the ore stocks in the area of Schneeberg and Oberschlema were exhausted, and following the ore veins the remnants of mining shifted in a north-eastern direction (Niederschlema, Hartenstein, Alberoda near Aue). There and in eastern Thuringia (Gera, Ronneburg) mining ceased about 1990 owing to exhaustion of reserves and decreasing need of uranium due to disarmament[3].

After the reunification of Germany the 'Bundesamt für Strahlenschutz' (BfS) began an extensive working project, and 'Wismut GmbH' as the successor of the former 'SDAG Wismut' took part in it[5,14]. The project consists of the following investigations:

1) radon within buildings ('radon programme')
2) radioactive contamination by mining waste ('waste register')
3) consequences of uranium mining on health.

During the last few years the radioactivity of numerous suspicious areas has been measured, and the first steps of recultivating took place. Many dumps have been removed, and others covered by soil and planted. As a basis for detailed investigations a comprehensive data bank with respect to environmental radioactivity caused by mining has been built up.

In this connection the health aspects of mining radioactive ores are of special interest. As early as the end of the 15th century it was recognized, that in the mines of Schneeberg 'underground air is very unhealthy', and the often fatal sickness of young miners was named 'Bergsucht'[17]. It is not known if in those days a connection to the black uranium oxide was made, or if the name 'pitch-blende' was derived only from the black colour of pitch and related to the meaning of 'bad luck' owing to the economical worthlessness of this ore. Later on the 'pulmonary disease of Schneeberg' ('Schneeberg lung cancer') was recognized as a carcinoma of the lungs and as a typical professional disease of miners in the ore mountains (refs 7, 8, 15, 16; see Fritz-Niggli in this issue). But only in this century was the relation between this sickness and the special geological conditions of this region, i.e. the radioactivity of the vein ores, explored. Intensive investigations of these conditions took place in the 1930s[16,17]. During World War II the first boundary values for admissible concentration of radon in mine air were fixed. But during the first years of forced uranium mining in the ore mountains after 1945 they were disregarded.

Except for sporadic X-ray examinations of thorax and blood tests in these years no measurements or checks of radiation risks to miners took place. Of course underground miners, inhaling gaseous radium emanation (radon) and radioactive dust for a long time, were especially endangered[8-10]. In spite of instruction for wet drilling, at the beginning rock was often drilled dry in order to reach higher norms[3]. At first air flow in the mines was also not quite what one would wish. Nowadays it is estimated that 80% of the collective radiation load at the 'Wismut-AG' took place in the first ten years of mining from 1946 to 1955. In all the years of uranium mining more than 6000 deaths by cancers of the lungs were registered, and annually some hundred new applications for acknowledgement of professional disease are received[17]. Within the 'Hauptverband der gewerblichen Berufsgenossenschaft' the 'Zentrale Betreuungsstelle Wismut' (ZeBWis) was founded in 1992, which organizes medical measures for former employees of the uranium mines. Therefore data from about 600,000 persons are registered. The effects of uranium mining on the health of these employees are to be investigated thoroughly by means of these voluminous data.

Nowadays the causal relationship between a high concentration of radon (see Roth and Fritz-Niggli in this issue) in the underground air and carcinoma of lungs of miners has been statistically proved. Not only the rare gas Rn-222 itself, but above all its solid daughter-nuclides of polonium, lead and bismuth (often fixed to particulate aerosols), are responsible for the high radiation load in air ways of miners[9,10]. The mean annual equivalent dose of an adult by inhalation of air with an average content of radon at the earth's surface amounts to about 1 mSv, that is a little more than half of the total load[20].

The German commission for radiation protection proposed that a mean value of 250 Bq/m^3 for the indoor activity of radon shall not be exceeded in living- and sleeping rooms. For comparison: the mean value in German dwellings is 50 Bq/m^3, free air has 10–20 Bq/m^3, while the radon activity of underground air in the 'Wismut' mines from 1946 to 1955 was estimated at 150,000 Bq/m^3 (refs 12, 17).

For radiation protection, mining and geophysics release of radon from rock containing the mother substance radium into the pore space filled by water or soil air is of great importance. The emanation ability of rock is the relation of radon arriving at the pore space to the total radon escaping from the rock. Rock near the earth's surface has the emanation ability of <1–50%.

The transport of radon into mining caverns, houses or atmospheric air is influenced by two physical factors: diffusion and convection. Diffusion of Rn into soil gas is the reason why Rn-222 available within the pore space at a depth of 0 to 0.2 m crosses the rock/air interface into the air of mines, houses or the atmosphere. Convection causes transport of radon together with soil gas or soil liquid. Convection accounts for the high content of Rn in some spring-waters: Bad Brambach 30 000 Bq/l, Elizabeth spring of Bad Gastein 1580 Bq/l[17].

In mining caverns or in houses the concentration of radon can effectively be reduced by aeration (convection!). Moreover the sealing of cellars in houses against the underground is possible, especially in old mining areas. In such areas radon concentration in buildings with especially high radioactive contamination in many cases could be considerably reduced by these steps[5,12].

Convection also causes transport of radon into higher layers of the atmosphere up to altitudes of some kilometers. The representation of uranium ore bodies or of salt diapirs with high radioactive rim waters at depths of more than 100 m within the soil air is also explained by convection.

Disposal of radioactive waste

With regard to disposal of mining waste, the participation of geosciences is necessary for the final disposal of radioactive waste from nuclear reactor power plants, scientific research, industry, and medicine. Storage times of 10^3 to 10^6 years are to be taken into consideration. Host rock of such final disposals may be salt, anhydrite, mudstone, granite, basalt and tuff. Disposal at the deep-sea bottom also seems to be possible. All these possibilities have been investigated worldwide for some time past, and they are discussed heatedly, sometime even emotionally[2].

In Germany rock salt is proposed for the final disposal. It has favourable mechanical properties (stable cavities without support, plastic response to tectonic stress), it is dense and impermeable, and is a good heat conductor. This is important to divert heat emission resulting from radioactive disintegration. Local research took place principally at the salt diapirs of Gorleben (Niedersachsen) and Morsleben (Sachsen-Anhalt), but the final results have not yet been presented[2]. Salt rock deposited at a depth of 500 to 1000 m suitable for mining also seems to be favourable, if it is layered without strong deformation by diapirism and only slightly influenced by salt tectonics[18]. Especially important is a good seal against ground water near the surface by an impermeable overlayer of sufficient thickness.

For many decades the geosciences, mining and radioactivity have acted and reacted upon each other. By the intensive utilization of radioactive conversion processes many problems have resulted: disarmament of the enormous potential of nuclear weapons, protection against dangers resulting from industrial use of nuclear energy, disposal of radioactive technical and mining waste of all kinds. Besides politics, economy, technology and medicine, the geosciences must also contribute to solving these vital problems of human existence.

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