



Recent aspects of uranium toxicology in medical geology



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ABSTRACT

Uranium (U) is a chemo-toxic, radiotoxic and even a carcinogenic element. Due to its radioactivity, the effects of U on humans health have been extensively investigated. Prolonged U exposure may cause kidney disease and cancer. The geological distribution of U radionuclides is still a great concern for human health. Uranium in groundwater, frequently used as drinking water, and general environmental pollution with U raise concerns about the potential public health problem in several areas of Asia. The particular paleo-geological hallmark of India and other Southern Asiatic regions enhances the risk of U pollution in rural and urban communities. This paper highlights different health and environmental aspects of U as well as uptake and intake. It discusses levels of U in soil and water and the related health issues. Also described are different issues of U pollution, such as U and fertilizers, occupational exposure in miners, use and hazards of U in weapons (depleted U), U and plutonium as catalysts in the reaction between DNA and H₂O₂, and recycling of U from groundwater to surface soils in irrigation. For use in medical geology and U research, large databases and data warehouses are currently available in Europe and the United States.

1. Introduction

Uranium (chemical symbol U and atomic number 92) is a heavy metal belonging to the actinide series in the periodic table. In nature, ²³⁸U is the most diffused isotope of the element (> 99.27%), while a small percentage is represented by the radio-isotope ²³⁵U (0.7%) and to a much lesser extent by ²³⁴U (0.006%), yet all isotopes appear as weakly radioactive. Besides to neutrons, its atoms have 92 protons and 92 electrons, of which six are valence electrons. The primary oxidation states of U are 0, +2, +3, +4, +5, and +6, of which the most often occurring oxidative states present in nature as oxides or salts are +4 and +6. Geologically, U is described as a lithophile element, which would mean that it easily forms oxides and its compounds are concentrated in minerals of the Earth's crust, i.e. in granites as uraninite UO₂, previously called pitchblende. The *enrichment* process separates naturally occurring U into two fractions, one of them with a substantially increased fraction of ²³⁵U defined as *enriched* U and used in reactors and nuclear weapons, and the remaining portion termed *depleted* uranium (DU) is used in military armor and penetrating munition.

The population is frequently exposed to U. Although humans poorly

absorb U, it can easily enter the bloodstream. Uranium compounds with low pharmacokinetics are often resident in the lungs, where nearby lymph nodes capture them as uranium trioxide and uranium tetrafluoride for weeks or uranium dioxide and triuranium octaoxide for years. In body fluids, U is present in its more frequently saline associated hexavalent form, generally complexing with low molecular weight compounds such as citrate and bicarbonates, but also with proteins in plasma. Up to 70% of circulating U is filtered in the kidneys and leaves the body within 24 h (Keith et al., 2015).

The ground availability and turnover of U in soil are ruled fundamentally by the formation of the hydrated uranyl cation UO₂⁺, which is responsible for the solubility of U over a wide range of different soil pHs. This feature should allow U to be found in freshwater and thereby reach human life with water and plants using this polluted water. Organic acids might enhance U solubility in the soil, but its availability may also be restricted by the creation of marginally soluble precipitates, particularly in the form of phosphates, and by the adsorption on the organic matter of biomass and clay minerals. The solubility of U is generally lower under acidic reducing conditions than in oxidizing alkaline and carbonate-rich waters, although U is quickly soluble in those strongly acid and oxidizing waters that often are

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connected with acid mine drainage. Primary anthropogenic U sources include nuclear effluents, U mining and milling, military actions using DU, coal combustion, and phosphate fertilizers (Reimann and de Caritat, 1998; Banning et al., 2013).

This contamination represents a serious concern about health safety for human communities in these regions (Galhardi et al., 2017; Ielsch et al., 2017; Träber et al., 2015). Pollution coming from ^{238}U or DU, or other isotopes in groundwater and the environment, and also the U decay product radon (Rn), is a great concern for human health, as several forms of cancer such as leukaemia or lung tumors, may have their etiology just from such pollution (Drubay et al., 2014; Kreuzer et al., 2015; Winde et al., 2017; Krewski et al., 2006; Sahoo et al., 2015; Torres-Duran et al., 2016; Beck, 2017).

Exposure to U can cause both radiological and chemical toxicity (Kurttio et al., 2002, 2003; Konietzka, 2015). Among the main chemical effects related to exposure to U and its decay compounds are kidney disease and lung cancer (Drubay et al., 2014; Kreuzer et al., 2015). The main chemical toxic effect associated with high-level exposure to U is kidney toxicity (Kurttio et al., 2006; Seldén et al., 2009). Uranium can enter into the human body through inhalation of contaminated air (breathing), food and drinking water. In the body, soluble forms of U are taken up in the blood and are subsequently filtered by the kidneys, which may result in damage to tubular cells (Keith et al., 2015).

Uranium is particularly soluble as uranium hexafluoride, uranium tetrachloride, uranyl fluoride, and uranyl nitrate. These U compounds possess the highest renal toxicity. The less soluble compounds of U include ammonium diuranate, sodium diuranate, uranium tetrafluoride, as well as the insoluble U compounds uranium dioxide, uranium trioxide, uranium peroxide, and triuranium octaoxide. These are the less present U compounds in body fluids (Tannenbaum et al., 1951; Antunes et al., 2017; Kitahara et al., 2017).

The environmental and geological concern associated with U-related health issues expanded a debate about the widespread presence of U isotopes and radon in rural areas, due to their diffusion into water, beverages and food (Fathi et al., 2013; Al-Shammari, 2016; Kibria et al., 2016; Kosior et al., 2017). This debate escalated principally due to DU pollution of dusty sand and soil following the Gulf War and other military expeditions and conflicts (Briner, 2010). Uranium toxicity has been suspected to cause neurotoxicant-derived disorders and cancers in regions with presumably contaminated rural areas or freshwater sediments (Crawford et al., 2017). This paper highlights the link between the geological distribution of U and its impact on human health, particularly in regions where geographical distribution of U sediments is critical for water sources.

2. Uranium in soil: a role of fertilizers?

Higher oxidation states of U is found in soils and living organisms as the uranyl (UO_2^{++}) ion. This water soluble form of U is found in oxidized geological environments, like oxidized soils and seawater (Garrels and Christ, 1965; Lashley et al., 2016).

The extensive use of mineral fertilizers based on phosphorus also introduces U as a contaminant into the environment. If the rate of leaching is not high enough, this can cause that U accumulates continuously in soils (Liesch et al., 2015; Hegedűs et al., 2016; Bigalke et al., 2017). When agrochemical processes cause that U mobilizes from the upper soil layers, this may result in an increased concentration of U in groundwater. This is a particularly serious concern in those regions where the availability of U due to geogenic mechanisms are enhanced in the soil. For example, some bituminous shales, like the Cambrian alum shales found in parts of Scandinavia and Central Asia, contain much more U than igneous rocks and may represent a public health problem, also because of their high levels of cadmium and vanadium (Kabata-Pendias and Muherjee, 2007; Solodukhin et al., 2015).

Atmospheric transport and deposition might also play some role as a

source of U in the topsoil because of high U concentrations in coal (Karangelos et al., 2004; Mahur et al., 2008; Papastefanou, 2010a, 2010b), causing emission from coal-fired power plants. However, the contribution of atmospheric deposition to U concentrations in mosses and topsoils in areas affected by polluted rain in Europe, such as the Southern part of Norway, has as far as we know not yet been adequately studied.

Uranium is only soluble in water in the presence of molecular O_2 (Garrels and Christ, 1965). Under anoxic conditions, it is precipitated in the form of virtually insoluble UO_2 , as happened when the Cambrian alum shales were formed. Uranium might be present in drinking water due to the water content of dissolved O_2 . Nevertheless, uranyl (UO_2^{++}) is more easily reduced than e.g. chromate (CrO_4^{2-}) or vanadate (VO_4^{3-}) (Christophersen and Haug, 2011) since its redox potential is different from these compounds. Thus, a partial reduction to the oxidation number +4 can be performed by organic matter in the soil, even in moderately aerated conditions, provided that the concentration of organic reducing matter in the soil is high enough. Quadrivalent U is presumably poorly available for uptake in the plant roots (Christophersen and Haug, 2011).

Recycling of uranyl from groundwater to surface soil can take place through irrigation of agricultural areas with U-rich groundwater. When soluble U from the groundwater reservoir reaches the surface soils, it is expected to be available for uptake by the plant roots, similarly as happens with arsenic (As), when As-rich groundwaters are used for irrigation e.g. during the winter monsoon season in Bangladesh (Ahmed et al., 2011; Rahman and Hasegawa, 2011; Talukder et al., 2012; Halder et al., 2013; Pollizotto et al., 2013). The high solubility of U under highly oxidizing conditions in soils containing little organic matter, and in O_2 -containing groundwater, should be considered in those semiarid parts of Central Asia where bedrock concentrations of U are high. The reason for this is that bedrock is bituminous shales of Cambrian that is comparable to the U-rich alum shales of Scandinavia and the Baltic countries, which are of equal age. They have been formed because of salinity stratification in the prehistoric oceans due to the large-scale formation of very dense, hypersaline waters in epicontinental seas with a surplus of evaporation over precipitation (Christophersen, 2012, 2013). This involves that U-contaminated groundwater, when used as drinking water, might represent a significant public health problem, i.e. in some Siberian and Iranian regions, as it is in Northern parts of India. Whether or not fertilizers are an important source of U, compared to natural sources, given the relatively high estimate (of about 2.7 ppm) for the average U concentration in the Earth's continental crust, has been discussed in the past and is still a controversy (Krauskopf, 1982; Liesch et al., 2015; Bigalke et al., 2017; Schnug and Lottermoser, 2013).

The decrease of the pH of soil waters because of fertilizer application is presumed to lead to mobilization of the soluble UO_2^{++} from cation exchangers in the soil. Such cation traps exist both in the form of soil organic matter and clay minerals. The uranyl mobilization may secondarily result in raised levels of U in groundwater that may be used as a source for drinking water.

It is expected that manganese (Mn) also may be partly solubilized when the pH value of the topsoil is reduced following chemical fertilizer application. However, Mn is probably much less mobilized, compared to U. And if Mn leaches downwards, it might be immobilized by minerals in the groundwater reservoir (Garrels and Christ, 1965).

Due to redox causes and the microenvironment pH and partial oxygen pressure, different levels of solubility for Mn^{++} and UO_2^{++} are expected, as well as for arsenic (As). Arsenic is only highly soluble when the water is anoxic (Guo et al., 2008, 2011; Xie et al., 2009). In oxygen-rich environments, As will instead be removed from the water phase by high adsorption of arsenate and arsenite ions to ferric oxide/hydroxide and sulfide minerals (Guo et al., 2008, 2011; Xie et al., 2009). Thus, oxygen status of water will modify the presence of dissolved U. These aspects are particularly stressed here to comprehend the abundant

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