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Apatite weathering as a geological driver of high uranium concentrations in groundwater

Andre Banning *, Thomas R. Rüde

RWTH Aachen University, Institute of Hydrogeology, Lochnerstraße 4-20, 52064 Aachen, Germany

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ABSTRACT

Uranium is a heavy metal with potential adverse human health effects when consumed via drinking water. Although associated quality regulations have been implemented, geological sources and hydrogeochemical behavior of uranium in groundwater used for drinking water supply remain little understood. This study presents a hydrogeochemical and mineralogical characterization of a Triassic sandstone aquifer on a macro- and micro-scale, and an evaluation of uranium remobilization into groundwater, also considering the paleoenvironment and the distribution of the affected aquifer itself. Syndiagenetic uraniferous carbonate fluorapatite inclusions within the aquifer sandstones ("active arkoses") were found to show structurally (chemical substitution in the crystal structure) and radiatively (α-recoil damage from uranium decay) enhanced mineral solubility. Extraction experiments indicated that these inclusions release uranium to groundwater during weathering. In conclusion, apatite alteration was identified as the responsible mechanism for widespread groundwater uranium concentrations >10 $\mu g\ L^{-1}$ in the region representing Germany's most significant problem area in this respect. Therefore, results indicate that the studied sedimentary apatite deposits cause the regional geogenic groundwater uranium problem, and must be considered as potential uranium sources in comparable areas worldwide.

1. Introduction

1.1. Rationale

Uranium (U) is known to be a heavy metal with a nephrotoxic potential, possibly leading to adverse human health effects (Zamora et al., 1998; Kurttio et al., 2006). In order to limit public U exposure via drinking water, German authorities established a threshold value of 10 $\mu g\ L^{-1}$ in 2011, making Germany the only European Union member state to date with a binding legislation in this respect. Sources of U in groundwater can be natural or anthropogenic. While the former is mostly represented by uraniferous rocks like felsic magmatites (Banning et al., 2012; Frengstad et al., 2000) or fen peats (Read et al., 1993; Banning et al., 2013), the latter includes former U mining sites (Carvalho et al., 2005; Baborowski and Bozau, 2006), depleted U ammunition (Crançon et al., 2010; Dong et al., 2006) or phosphorus fertilizer (Zielinski et al., 2006; Schnug and Lottermoser, 2013). Drinking water supply in northern Bavaria is dependent on groundwater extraction from terrestrial Triassic (Keuper) sandstones. Therein, large areas with groundwater U concentrations >10 $\mu g\ L^{-1}$ were detected, making the region Germany's most significant U problem area known so far. The U sources and mobilization processes have been unknown. Consequently, in an effort to unravel U dynamics in the given area, this study focused on a geochemical and mineralogical characterization of aquifer materials, elemental distribution on different scales, and U mineralogical fractionation and mobility.

1.2. Study area and "active arkoses"

The study area around the city of Nürnberg in southeastern Germany (Figs. 1 and 3) is part of the epicontinental South German Keuper Basin filled with terrestrial and shallow marine sediments (see geological map, Fig. A1 in the Appendix). A medium to coarse grained, feldspathic sandstone ("Burgsandstein", Fig. 2) with clayey interbeddings from the terrestrial facies represents the major aquifer used for water extraction in the region.

Typical groundwater type in the "Burgsandstein" aquifer is Ca–Mg–HCO_3 (Heinrichs and Udluft, 1999). Hydrochemical data for 21 groundwater samples from this aquifer (kindly provided by the Bavarian Environment Agency, LfU) indicates a circumneutral pH milieu (mean: 7.1, ranging from 5.2 to 8.3). pH does not show

* Corresponding author at: Ruhr-Universität Bochum, Hydrogeology Department, Universitätsstraße 150, 44801 Bochum, Germany. Tel.: +49 (0)234 32 23298.
E-mail address: andre.banning@rub.de (A. Banning).

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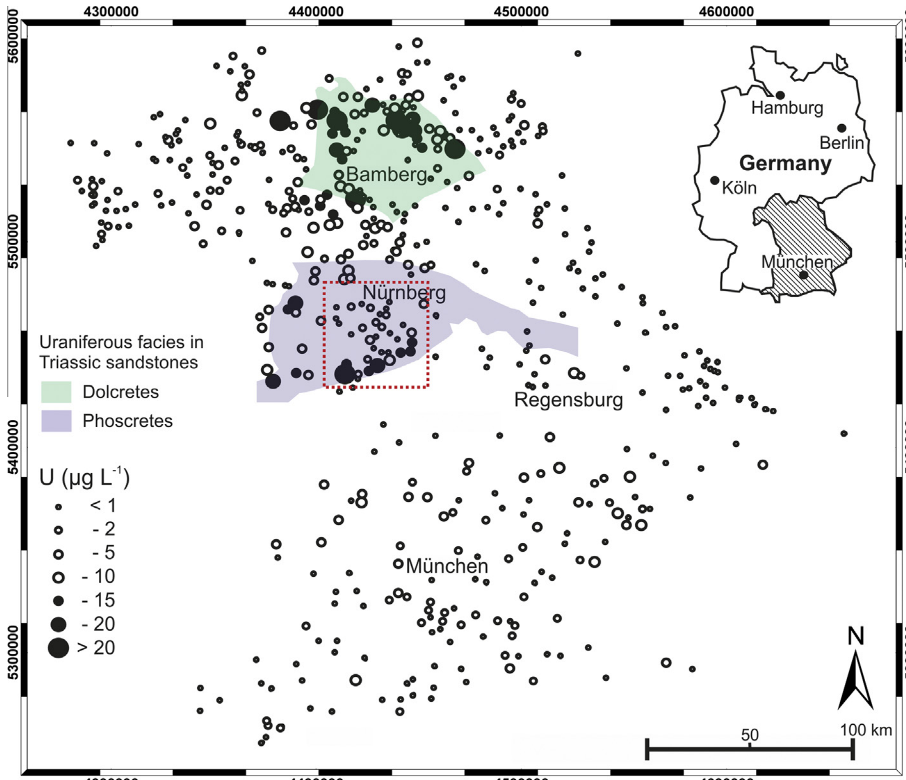

Fig. 1. Uranium concentrations in Bavarian drinking water and distribution of uraniferous facies in Triassic sandstones (the latter after Dill, 1988). The dashed red box indicates the study area. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

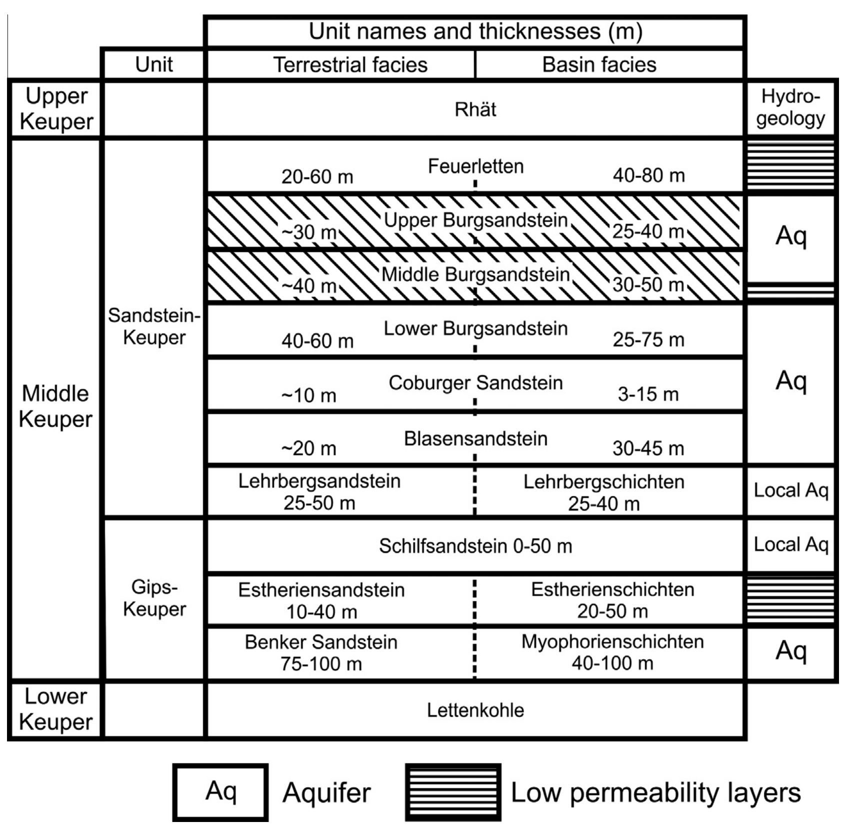

Fig. 2. Keuper stratigraphy of the study area with sampled units highlighted by hachures, approximate unit thicknesses and basic hydrogeology (modified after Heinrichs and Udluft, 1999). "Active arkoses" only occur in Middle and Upper "Burgsandstein" aquifers, parts of the terrestrial Norian "Sandsteinkeuper".