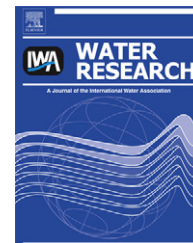


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Enrichment processes of arsenic in oxidic sedimentary rocks – From geochemical and genetic characterization to potential mobility

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ABSTRACT

Sedimentary marine iron ores of Jurassic age and Tertiary marine sandy sediments containing iron hydroxides concretions have been sampled from boreholes and outcrops in two study areas in Germany to examine iron and arsenic accumulation processes. Samples were analyzed for bulk rock geochemistry (INAA/ICP-OES), quantitative mineralogy (XRD with Rietveld analysis), element distribution (electron microprobe) and arsenic fractionation (sequential extraction). Bulk Jurassic ores contain an average arsenic content of $123 \mu\text{g g}^{-1}$ hosted in mainly goethite ooids which slowly formed in times of condensed sedimentation. Enrichment occurred syndepositionally and is therefore characterized as primary. Iron concretions in Tertiary sediments mainly consist of goethite and yield arsenic up to $1860 \mu\text{g g}^{-1}$. The accumulation process is secondary as it took place in the course of oxidation of the originally reduced marine sediments under terrestrial conditions, leading to element redistribution and local enrichment in the near-surface part. The scale of enrichment was assessed calculating Enrichment Factors, indicating that arsenic accumulation was favoured over other potential contaminants. In spite of higher bulk arsenic contents in the oxidic rocks, the mainly pyrite-hosted As pool within the reduced deeper part of the Tertiary sediments is shown to have a higher potential for remobilization and creation of elevated arsenic concentrations in groundwater.

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1. Introduction

1.1. Background and objectives

During the past two decades, the intensity of arsenic (As) research has increased substantially. Reasons for that are the recognition of As toxicity and its initially underestimated impact on human health on a global scale, focussing on large problem areas, especially in Southeast Asia (e.g. Ravenscroft et al., 2005; Zahid et al., 2009). Consequently, drinking water threshold values were broadly lowered in the 1990ies, mostly down to $10 \mu\text{g L}^{-1}$. This confronted water suppliers with the

problem of an increased need to process raw water in order to match drinking water requirements. In recent years, it was also found that in the large majority of cases, naturally occurring As is responsible for elevated groundwater concentrations. Thereby, mobilization from As-enriched minerals is the dominating process (e.g. Lowers et al., 2007; Armienta et al., 2001). Showing characteristics of both chalcophile and siderophile behaviour, arsenic tends to be preferably hosted by sulphide minerals like pyrite or (hydr)oxidic Fe phases like goethite, both of which can contain As up to several wt.% (Smedley and Kinniburgh, 2002). In spite of its low average abundance in the upper earth's crust

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(1.5–2 $\mu\text{g g}^{-1}$; Matschullat, 1999), arsenic can accumulate in rocks to concentrations several orders of magnitude higher than this value. The fate of As in the environment is controlled by the prevailing physico-chemical conditions and the presence of other ions. Redox potential, pH and ionic competition are crucial parameters governing As behaviour (adsorption, desorption, transport, redox transformation). Reducing conditions can lead to As mobilization from oxides while oxidizing conditions may mobilize As bound to sulphides. High groundwater pH constrains As adsorption to mineral surfaces and may therefore be responsible for elevated concentrations in solution. Ions competing with As species for surface binding sites, especially phosphate, can lead to the same result (Stollenwerk, 2002; Smedley and Kinniburgh, 2002).

The basis to understand geogenic As cycling and the impact of water-rock-interaction leading to groundwater contamination is the comprehension of the mechanisms that led to those accumulations, i.e. arsenic enrichment processes. These often took place millions of years ago, but still have a major influence on the recent occurrence and thus, hydrogeochemical behaviour, of As and other potential groundwater contaminants.

While As accumulation in Southeast Asia is subject to extensive research and ongoing lively discussions (e.g. Acharyya et al., 2000; Stanger, 2005), little is known about the origin and development of geogenic As accumulations in

Germany which have been studied sporadically and on a rather local to regional scale (e.g. Bayer, 1997; Heinrichs and Udluft, 1999; Rüde and Königskötter, 2009; Banning et al., 2009), although elevated As concentrations have been detected in many parts of the country, an overview is given by Heinrichs and Udluft (1996).

In this study, we analyzed and compared As-bearing sedimentary rocks in two study areas in Germany and deduced the relevant accumulation processes, taking hydro-geochemical, mineralogical and paleogeographical information into account, evaluating the scale of enrichment and estimating the consequences for potential As remobilization and output into groundwater.

1.2. Study areas

Two investigation areas in Germany were selected for rock sampling in the Upper Rhine Graben and in the Lower Rhine Embayment, respectively (Fig. 1).

1.2.1. Upper Rhine Graben (URG)

The URG is an approximately NE-SW-striking rift structure in the border area between Germany and France. It is situated in between the crystalline basement areas of the Black Forest (Germany) and the Vosges mountains (France) and mainly filled with sediments of Tertiary age (Fig. 1). Along the fringes, within the contact zone between crystalline basement and

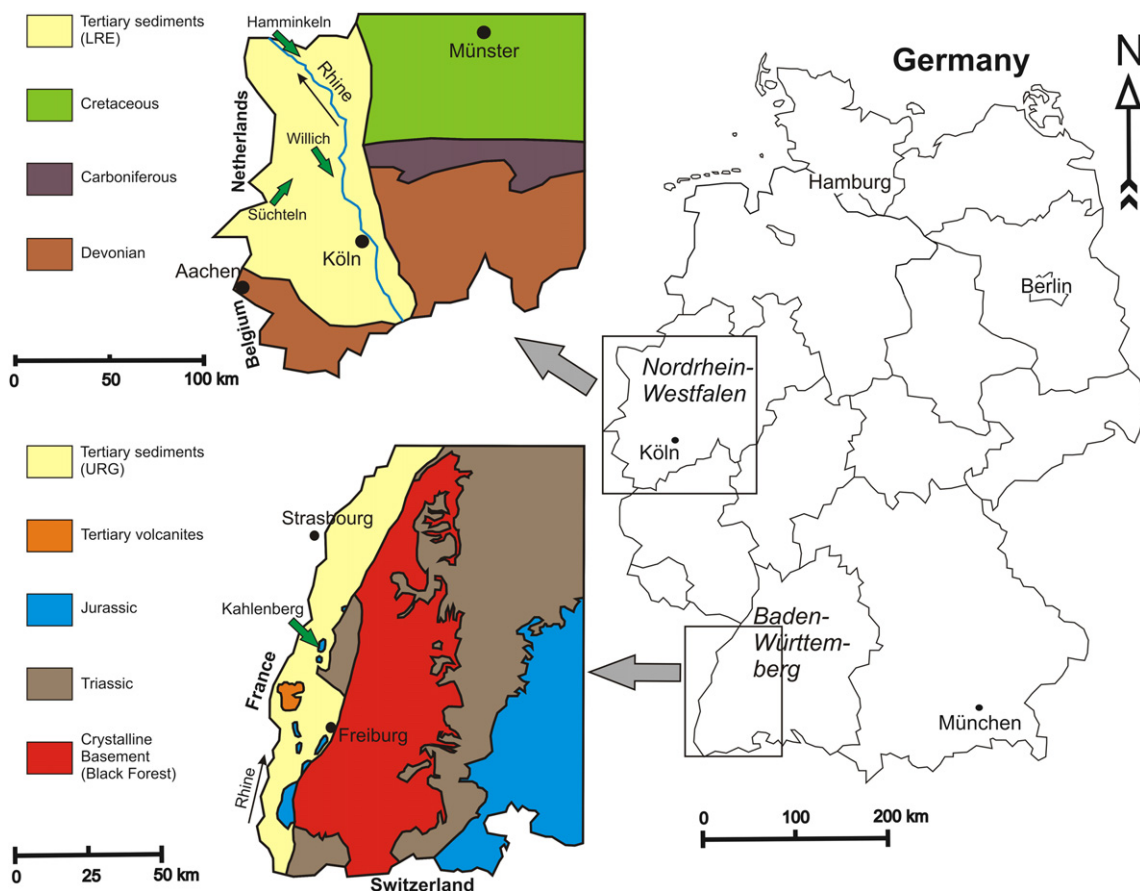


Fig. 1 – Location of study areas in Germany (right side) and geological overview maps with sampling locations in the Lower Rhine Embayment (LRE; upper left side) and the Upper Rhine Graben (URG; lower left side).

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