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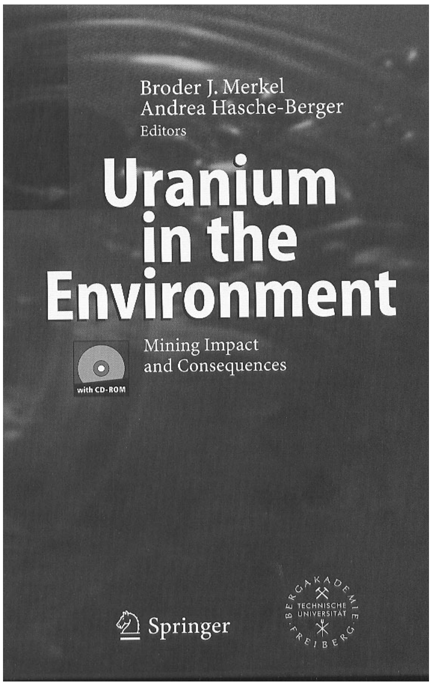

Changes in Uranium concentration in the Weisse Elster river as a mirror of the Remediation in the former WISMUT mining area

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Abstract. Since 1990 the research group „Pollutant Dynamics“ at the Saxon Academy has been investigating the process of self-purification of the former highly anthropogenically affected river Weisse Elster. This long term study, focusing on uranium, includes the components water (< 0.45 µm), suspended matter (SPM), sediment and floodplain soil (grain-size < 20µm,) from 1990 up to 2004.

Introduction

Eastern Germany was once one of the world's biggest uranium producers, mining 220,000 tonnes between 1945 and 1990 (see Tondorf 1994, Kämpf et al. 1995). The repercussions of uranium mining in eastern Thuringia and western Saxony continue to have a severe impact on the local environment (cf. Schultze and Knappe 2001 and Beleites 1992). For example, uranium still persists in the Weisse Elster and its tributary Pleisse, the receiving water of the Ronneburg mines near Gera, as well as in the Göltzsch, a tributary whose catchment area contained the uranium processing plants.

Since 1990 the research group „Pollutant Dynamics“ from the Saxon Academy of Sciences has been investigating the process of self-purification of a former highly anthropogenically affected river initiated by remediation on the example of the Weisse Elster river system. This study, focusing on heavy metals, includes the components water, suspended matter (SPM), sediment and floodplain soil (regional background) as well as using sequential speciation on selected sediments. This paper reports about a long-term decrease of uranium in the river downstream the uranium mining discharges at the Gera-Ronneburg-Area. These will be shown from data of river longitudinal profiles as well as monthly collected samples upstream the confluence with the Saale river at Halle/Saale.

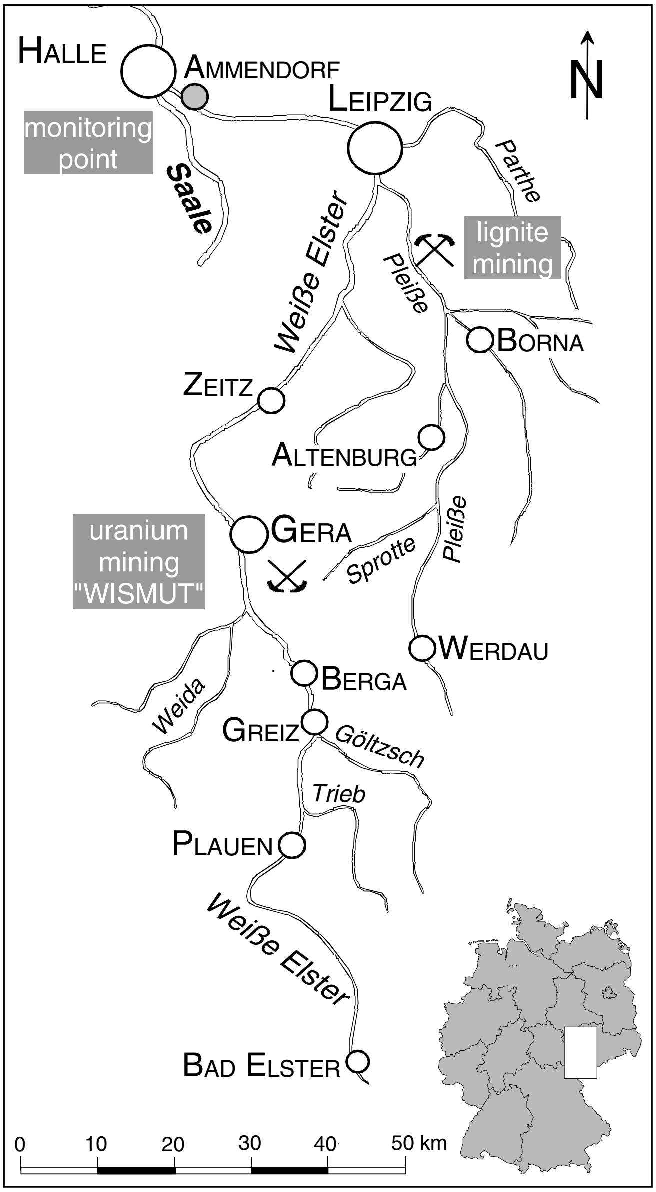

Fig. 1. Sketch of the Weisse Elster catchment area

Methods

Sampling

In order to determine the complete quantities of metals currently contaminating the Weisse Elster, mixed samples were taken from recent surface sediment. The suspended matter was separated from the water at the sampling site by filtration (celluloseacetate filter < 0.45 µm).

The recent sediment was pressed into bottles under water without being allowed to come into contact with the air. The idea was to enable the samples to be processed under both oxidizing and anoxic conditions. Pre-industrial, if possible layered and datable Holocene alluvial loam profiles were used to determine the geogenic background values. Dense vertical sampling enabled anthropogenically contaminated samples and samples which had been altered by chemical processes in the soil and water in the range of the ground-water level to be excluded. For more information on sampling and sample preparation, as well as the analytical methods used, see the detailed description in Müller et al. (2000).

Analyses

Wet sieving was used to obtain the <20 µm fraction from the mixed samples of recent sediment and soil samples. The fraction was then treated with aqua regia in accordance with DIN 38 414-S7. The small quantities of metals in the suspended matter obtained with the celluloseacetate filter and the residual fraction of sequential speciation were treated using a standardized aqua regia extraction technique developed together with the University of Leipzig's Institute of Analytical Chemistry in the microwave field. The results of this method correlate with the digestion process specified in DIN 38414-S7; cf. Dittrich et al. (1995).

ICP-MS (inductively coupled plasma mass spectroscopy), a reliable method for determining several different elements at once known as was applied detecting uranium. The ICP-MS measurements were used to determine the rare elements in all the analysis samples, as well as to determine the other metals owing to their low concentrations in the water, suspended matter and sequential extracts. Appropriate analysis techniques were developed for the task at hand and their reliability was tested. According to the results from inter-laboratory tests and AAS reference measurements, ICP-MS provided accurate results when used for the determination of metals in sediment samples and suspended matter with no significant differences from those obtained pursuant to DIN 38 414 (Lohse 1997). The accuracy of all the measurements was checked using standard reference materials.

Regional Frame – Regional Background of Uranium

The Weisse Ester river (mean anual waterflow 25 m^3/s) draining the saxothuringian industrial region was one of the most badly polluted tributaries in the

Elbe river system (Fig. 1). In terms of its uranium content, the catchment area of the Weisse Elster can be divided into three sections: the mainly geogenic upper course; the upper middle course dominated by the aureole of granite in the western Erzgebirge and the anthropogenically contaminated tributaries coming from the east; and the lower middle course together with the lower course influenced by uranium mining in Ronneburg. The samples used for the analyses of all metals in sediment, water and suspended matter were taken from a dense network (cf. Müller et al. 1998).

According to Müller, Zerling and Hanisch (2003), the catchment area of the Weisse Elster and the Pleisse can be divided in terms of geogenic background values into eight sub-regional areas (Table 1). The background values listed are means calculated from 12–20 samples containing background levels. Uranium's mean geogenic background in the Weisse Elster catchment is 3.3 mg/kg. This is below the generally accepted argillaceous rock standard according to Turekian and Wedepohl (1961) of 3.7 mg/kg, but which is based on the total extract. The background value for uranium in the upper course is far lower at just 2.0 mg/kg. However, it rises to 4.2 mg/kg in the area of Berga downstream of the inflow of tributaries (especially Göltzsch and Trieb) whose courses touch the west Erzgebirge granite complexes (Bergen and Kirchberg granite; cf. Förster and Tischendorf 1994). In the Gera region the mean background value drops back to 3.6 mg/kg. All other sub-regions – with the exception of the Leipzig district – have values which are around or below the argillaceous rock standard.

Table 1. The local geogenic background values of uranium (in mg/kg) in fine-grained river sediment of the Weisse Elster catchment area

River	Sampling area	Geogenic background for uranium	No. of samples
Weisse Elster	Pirk-Geilsdorf downstream of Oelsnitz	2.0	20
	Eula upstream of Berga	4.2	12
	Ahlendorf between Gera and Zeitz	3.6	16
	Großstorkwitz (near Pegau)	2.9	17
	Cospuden and Leipzig-Süd	4.6	18
	Rassnitz and Zöschen, SE of Halle	3.9	15
Pleisse	Münsa and Kraschwitz (near Altenburg)	2.5	12
	Markkleeberg-Ost	3.1	14
Mean for the area of the Weisse Elster		3.3	124

Characterization of Uranium in recent Sediment

The uranium levels in the sediment of the upper course of the Weisse Elster in 1991, 1992 and 1994 were between 2.8 and 6.9 mg/kg, approximately tallying with the geogenic background (Table 1, Fig. 2). The tributaries Trieb and Göltzsch, whose upper courses from the aureole of the west Erzgebirge granite already contained higher geogenic background levels (6-18 mg/kg), were enriched with uranium levels up to 30 mg/kg (and as much as 60 mg/kg in 1992) by the legacy of ore-mining and processing operations. This was caused in particular by the Plohnbach creek near Lengenfeld, and even prompted slight uranium contamination (6–18 mg/kg) in the Weisse Elster in the area of Greiz. The sudden uranium contamination of the sediment in the Weisse Elster, starting downstream of Berga, resulted from the influx of water from the Ronneburg uranium-mining district operated by the East German-Soviet Wismut AG, especially from the Gessenbach creek, which contained levels of dissolved uranium of as much as 268 mg/kg in the sediment and 76 µg/l in the water (see Müller et al. 1998). Other small creeks contained up to 400 mg/kg uranium in suspended matter (SPM) and up to 300 µg/l dissolved uranium. The high concentrations in the sediment of the Weisse Elster downstream of Gera gradually declined owing to dilution down to 30–50 mg/kg (in 1994 15-25 mg/kg) until it flowed into the Saale. Part of the water from uranium processing was at times discharged eastwards, reaching the Pleisse via the Sprotte. As a result, the uranium contamination in the Pleisse downstream of the influx of the Sprotte creek more than doubled despite the high dilution. In 2000 clearly a tremendous decrease of uranium in the recent sediments can be seen in fig 2. Never the less the uranium content in the sediment sampled in 2000 ist enriched to factor 3-6 compared to the regional geogenic uranium background.

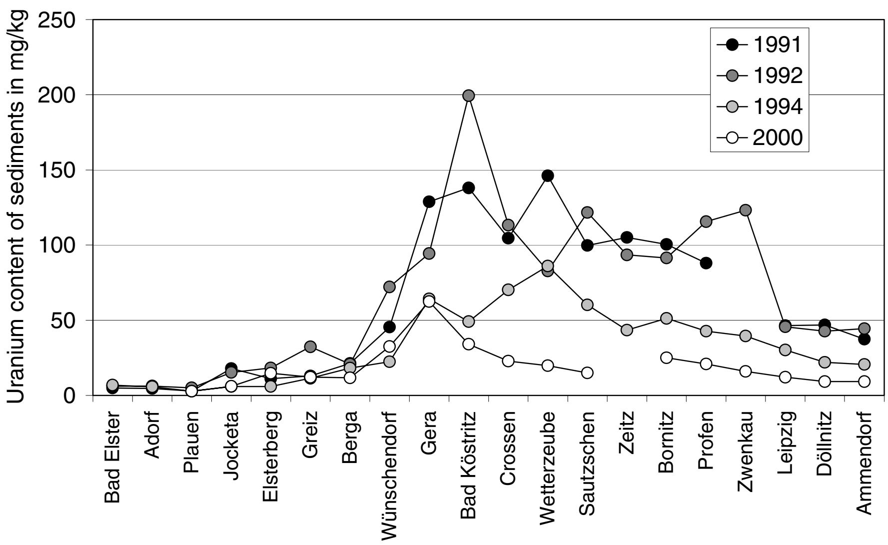

Fig. 2. Uranium content of recent sediments in 1991, 1992, 1994, and 2000 from upstream (Bad Elster) to downstream (Halle-Ammendorf)

Comparison over time (Fig. 2) shows, that until 1994 the uranium levels in the Weisse Elster upstream of Berga remained almost constant, but greatly declined in the contaminated area till 2000. Even more striking is the decline in the Pleisse (cf. Müller et al. 1998).

Examination of the ratio between dissolved and particulate uranium in the course of the river revealed that dissolved uranium was enriched upstream relative to particulate uranium by a factor of 2.5. The concentration of dissolved uranium also declined upstream, albeit not as rapidly as that of the particulate phase (Fig. 3). Uranium is one of the elements with a low tendency to bind to particulate substances (Fig. 4). One reason for this is the high volatility of uranium in acidic milieus and under reducing conditions in sediment (where it forms highly soluble complexes). Since the sulphate and chloride concentration is important for the solubility of uranium, let us turn our attention to the sources of the two main anions present in the water of the Weisse Elster: sulphate and chloride. In 1992 and 1994 the sulphate levels increased considerably from the upper course until the point where the Weisse Elster flows into the Saale. A sharp increase in the sulphur input is to be observed in the Berga area. Furthermore, the level of the sulphate load was found to be higher in 1994 than in 1992. Due to the reduction of mining water input caused on the remedion of lignite mines south of Leipzig there was a strong decrease of sulphate rich mining water up to 2000 (Czegka et al 2003).

Uranium in Suspended Matter (SPM)

We use three graphics to present the development of uranium in suspended matter (SPM) and dissolved uranium in the Weisse Elster river.

The spot check samples data from our sampling sites from 1992, 1994 and 2000 are presented in a longitudinal river section (from source to mouth) can be seen in Fig. 3. Sampling during the years 1992 and 1994 took part at a normal water level, in 2000 during a decaying flood event.

The uranium content lay in the longitudinal section in the upper courses of the Weisse Elster (till Berga) in the SPM within the years 1992, 1994 and 2000 between 3.5 and 14 mg/kg uranium. In the middle course the content of uranium in SPM increases on 12-22 mg/kg. Here the results from 2000 lie clearly over those of the years 1992 and 1994, caussed by the different water levels during sampling. In the lower course the values in 2000 are decreasing, in 1992 and 1994 they were increasing. This effect can primarily connected the change of water chemistry (e.g. pH, SO_4^{2-} content) in the late 1990ies by reducing the input of sour lignite open pit waste-waters. (see above)

The second illustration (fig. 3) shows the development of the uranium in SPM

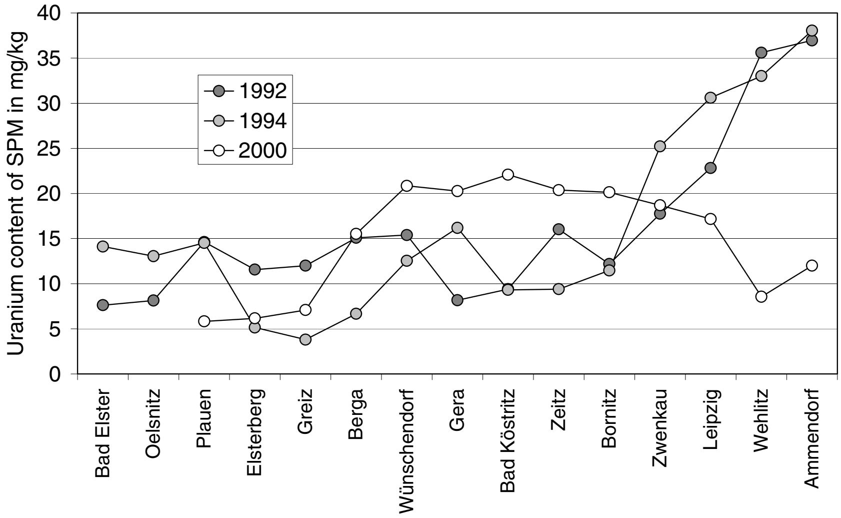

Fig. 3. Uranium contend in SPM 1992,1994 and 2000

as well as the dissolved uranium at our monitoring sampling site at the confluence of the Weisse Elster with the Saale at Halle. The graphics resulted from monthly sampling between 1991 and 2004. It is very obvious that the uranium contents, both in SPM and solution, decrease rapidly after 1996. Additionally a stepwise reduction can be seen especially in the data of uranium in solution. (comp. Geletneky, J., Büchel, 1999; Geletneky, J.: 2002). Nevertheless as shown in Fig. 4 the total amount of transported uranium in the Weisse Elster is realted to the soluted phase but not to SPM. Generally the content of disolved uranium is up to 3-6 times more than in SPM.

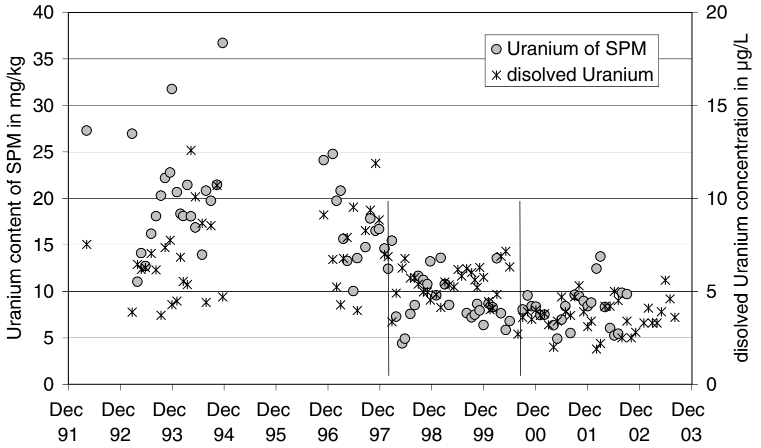

Fig 4. Uranium content of suspended matter (SPM) and dissolved uranium concentration at the Weisse Elster at Halle Ammendorf between 1991 and 2004

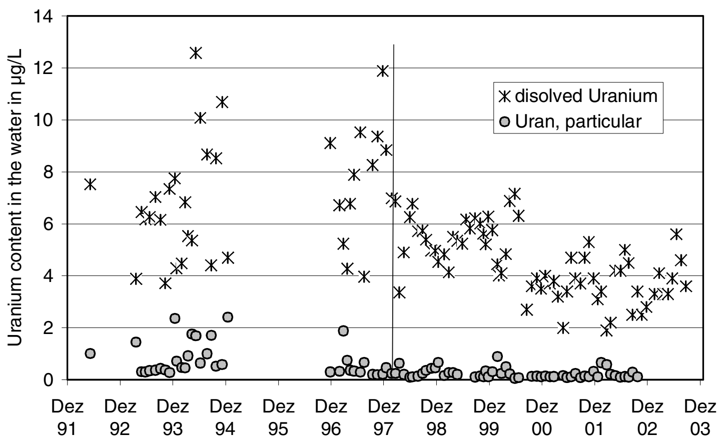

Fig. 5. Uranium content in water (dissolved and particular) at Halle Ammendorf between 1991 and 2004

Conclusions

Caused by the appearance of different uranium ore leading rocks in the catchment area of the Weisse Elster river, the regional geogenic uranium background is increased compared to the clay standard of Turekian and Wedepohl (1961). This fact is particularly defining the uranium content in the upper course of the Weisse Elster.

On the other hand the middle and lower course of the Weisse Elster are dominated by the anthropogenic uranium input caused by the uranium ore dressing of the former Wismut AG.

Long time examinations at recent sediment, SPM and water documents altogether a decrease in the anthropogenic uranium supply in the time period from 1991 to 2004.

This is justified as a result of the remedial actions which were carried out in the Wismut abandoned hazardous sites in the Gera-Ronneburg area during this time period. The decrease in the uranium contents was carried out gradually, according to the way, effectiveness and time of the remedial actions. This fact is extremely obvious with the putting into service of the water conditioning plant at the Ronneburg/ Culmitzsch uranium site in spring 1998. As of this time no more uranium loaded leachate reached into the Weisse Elster, which is documented by the erratic decrease of uranium in the different uranium-compartments (SPM, dissolved uranium, sediment). On reason of the low particular binding ability of uranium the gradual decrease shows themselves amplified at the dissolved uranium fraction in the water of the Weisse Elster. In site of the remedial actions the uranium load of the sediments of the middle und lower course are up 3-6 times over the regional geogenic background.

Acknowledgements

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