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Distribution of uranium in German bottled and tap water

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ABSTRACT

The results presented in this paper on uranium in bottled and tap water were determined within the scope of the project "European Groundwater Geochemistry: Bottled Water" of the Geochemistry Expert Group of EuroGeoSurveys. The analyses of bottled water provide an inexpensive approach to obtain information about European groundwater geochemistry. For this study, the uranium concentrations in 1785 European mineral water samples were analyzed by ICP–QMS in the BGR laboratories. The dataset is used to obtain a first impression about natural concentration levels and variation of uranium in groundwater (and bottled water) at the German and European scale.

A range of <0.0005–16.0 µg U/L (median 0.17 µg/L) was obtained in 908 bottled water samples in Germany. A range of 0.00115–9.0 µg U/L (median 0.073 µg U/L) was obtained for tap water from 163 municipal tap water systems. The regional distribution of uranium in the analyzed bottled and tap water is influenced primarily by lithological (Bunter and Keuper strata, crystalline basement rocks, Permian strata, and loess) and anthropogenic (old mining districts, phosphate fertilizers, and industrial emissions) factors. Elevated uranium concentrations (>2 µg U/L) in bottled water have geological sources and were found in Hesse, Thuringia, North Rhine–Westphalia, Saxony, southern part of Saxony–Anhalt and in Baden–Württemberg (Black Forest). Bunter and Keuper sandstone are the main source of uranium, followed by Permian and crystalline basement aquifers. The EU Guidelines for Drinking Water and the German Drinking Water Regulations give no upper limit for uranium; however, a provisional value of 15 µg U/L exists in the Guidelines for Drinking Water Quality of the World Health Organization. The only value for German bottled water is 2 µg U/L, above which the label may not say that the water may be used for the preparation of baby food. Nevertheless the uranium content of bottled and tap water needs to be monitored and controlled by governmental offices. In all, 14% of all the analyzed German bottled water samples contain >2 µg U/L and 15% of all analyzed European bottled water. Only one German bottled mineral water sample exceeded the guideline value of 15 µg U/L in 2008.

1. Introduction

Uranium is a lithophilic, naturally occurring trace element found in rocks and soils, but it is also a major anthropogenic contaminant that arises from activities such as mining, ore processing, agriculture, coal combustion, and nuclear fuel processing (Reimann and De Caritat, 1998). Sorption on mineral surfaces determine to a large degree the mobility of U(VI) in soils (Syed, 1999; Barnett et al., 2000). Organic material, especially humic and fulvic substances, also plays an important role owing to complexing, as well as sorption processes (Shen, 1999). Thermodynamic parameters are of prime importance in assessing the mobility of metals in the soil, especially the stability constants of metal–humate complexes, which largely determine the nature of the species being transported. The stability of humate and fulvate complexes is especially relevant in the case of elements with serious environmental consequences such as U(VI), notably in the form of the uranyl ion, UO_2^{2+}.

Uranium may enter bottled and tap water from naturally occurring deposits or as a result of human activity, such as mill tailings (Dreesen et al., 1982; Cothern and Lappenbusch, 1983), emissions from the nuclear industry, the combustion of coal and other fuels, and the use of phosphate fertilizers that contain uranium. Phosphate fertilizers sold in Germany contain an average concentration of 61.3 mg U/kg (Eckhardt and Schnug, 2008) and can also contribute uranium to groundwater (Spalding and Sackett, 1972; Birke et al., 2009).

Fulvic acids in the Stassfurt area show a significant uranium load (Gottschalch et al., 2007). The uranium input is ascribed to agricultural sources (phosphate fertilizers) rather than dumped materials. Franke et al. (2000), Bolea et al. (2006) and Gottschalch et al. (2007) found that chalcophilic elements as well as Ca, Cr and U were enriched in the low-molecular weight fractions of humic substances, corresponding to the molecular weight range of fulvic acid.

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The objective of the research presented here was to contribute to the knowledge of uranium in German bottled and tap water, and to investigate its natural variation from natural (geogenic) and anthropogenic sources.

2. Geological sources of uranium in water

Uranium is a metallic element that is ubiquitous in the environment. Its concentration in rocks is typically between 1 and 4 mg/kg (Bowen, 1979; Taylor and McLennan, 1985). Relatively high concentrations occur in granitic rocks (1.5–6.0 mg U/kg), argillaceous sediments (clays, shales) and Fe-rich rocks and metamorphic rocks derived from them. The uranium in granites is concentrated in primary accessory minerals, such as monazite, zircon and uraninite, as well as iron oxides (Plant et al., 1999, 2003; Bernhard, 2004). The uranium concentrations in the soils worldwide vary between 0.8 and 11.0 mg/kg (Kabata-Pendias and Pendias, 2001). In contrast to the heavy metals, e.g., Cd and Zn, under the oxidizing conditions of farmland soils, and especially in the presence of carbonate ions, uranium compounds are mobile (Baumann et al., 2008, Huhle et al., 2008, Schick et al., 2008). Mobile, extractible uranium concentrations were four times higher in such soils than in forest soil Huhle et al. (2008).

Uranium concentrations in acidic igneous rocks range from 3 to 4 mg/kg, in intermediate igneous rocks around 1.5 mg/kg, in basic igneous rocks about 0.6 mg/kg, and ultrabasic types about 0.03 mg/kg. High uranium concentrations have also been found in phosphate-bearing rocks. The uranium values in phosphate horizons vary from 30 to 120 mg/kg (Kratz and Schnug, 2006). Concentrations are significantly increased with increasing weathering.

Deposits rich in organic matter can also often have high uranium concentrations, presumably because reducing conditions favor the stabilization of U(VI) in the solid phase and the strong binding of uranium by humic substances. Relatively high uranium concentrations have been found in black shales (up to 250 mg/kg; Lee et al., 2001).

Uranium concentration in coal has been found as high as 2000 mg U/kg), but the average for uranium in brown coal in Germany is between 1 and 3 mg U/kg (Darbinjan, 1990). According to Smedley et al. (2006) in the Triassic Sandstone aquifer of the United Kingdom, uranium concentrations between 0.5 to 5.1 mg/kg (in rare cases up to 14 mg U/kg) have been measured. Uranium-rich bleached patches have also been described in the Triassic Sandstone which are of debatable origin but likely to be localized reduction zones, perhaps linked to the presence of organic matter.

Higher concentrations of uranium in groundwater coincide with low pH, low adsorption capacity, and presence of uranium ores (Merkel et al., 2002). The uranium level in groundwater varies considerably depending on the local geology (mineralized areas and granitic terrane). Especially granite and other crystalline rocks and its weathering soils, described as uranium-rich, produced also the uranium-richest vegetation (Anke et al., 2009).

3. Material and methods

About 908 bottles of mineral water sold all over Germany were collected and analyzed during 2008 and 2009 at the Federal Institute for Geosciences and Natural Resources (BGR) in Hannover. If the same water was available in carbonated and noncarbonated forms, both varieties were usually bought. Prior to analysis all water bottles were stored refrigerated. The mineral water bottles were opened in the laboratory. A 100 mL breaker was filled with water from the bottle. The cap was then left loosely on the mineral water bottle and the bottle was degassed for about 20 min in an ultrasonic bath.

Variation in uranium concentration in tap water was determined on 164 samples collected from residences in all parts of Germany in 2009.

All analytical work was carried out using clean room procedures at the BGR laboratory. The uranium in bottled and tap water was analyzed by inductively coupled plasma quadrupole mass spectrometry (ICP–QMS) using an Agilent 7500ce instrument with a detection limit of 0.0005 µg U/L. The instrument is equipped with a standard peristaltic pump, a MicroMist concentric nebulizer, a Peltier-cooled spray chamber, plasma forward power, and the Shield Torch system.

For quality control purposes, the river water reference material SLRS-4 from the National Research Council Canada, and the low-level fortified standards for trace elements TM-27.2, TM-28.2 and TM-28.3 from the National Water Research Institute of Canada were used. All certified reference samples were obtained in 500 mL HDPE bottles. In addition, an internal laboratory mineral water standard (MinWat) was used.

In addition, duplicates were analyzed at regular intervals and a number of quintuple and ten-fold determinations were also carried out. Sample blanks were measured over the entire time span of the project.

SPSS for Windows was used for the univariate and multivariate statistical analyses. Prior to the principal component analysis the data was log-transformed to obtain a set of nearly normal distributed parameters. The factor scores were calculated by regression method after varimax rotation of the resulting factor matrix.

4. Results and discussion

4.1. Uranium concentrations in bottled mineral water

The distribution of uranium and its sources in surface water have been described by Birke and Rauch (2008) and Birke et al. (2009). Uranium concentration in bottled and tap water is determined to equal extents by the geological origin and anthropogenic uranium emissions (Fisenne, 1994; Schnug et al., 2005; Knolle, 2008; Schnug et al., 2008; Knolle and Schnug, 2009).

A recent survey of bottled water samples (EGS European Geochemistry Expert Group, 2010) found that uranium concentrations were less than the current US EPA (2003) drinking water limit value of 30 µg/L in all samples tested (908 samples) but a relatively large range was determined. Concentrations vary between <0.0005 and 16.0 µg U/L (median: 0.170 µg/L).

The uranium concentration of only one sample (Griesbacher Mineralquelle, from paragneiss in the Black Forest) was over the WHO guideline value of 15 µg U/L in 2008. A change in the method for removing iron from Griesbacher label bottled water has lowered the uranium content to less than 0.5 µg U/L since August 2009.

The bottle leaching can have a minor influence on the uranium concentrations determined in water sold in glass bottles, especially at low pH (EGS, EuroGeoSurveys Geochemistry Expert Group, 2010).

The next highest concentrations were from the Knüll Mts. (Windsberg) in Hesse and from Thuringia (Hessberger Mineralquelle). These two samples have concentrations of 12.1 and 13.3 µg U/L. The geology of the groundwater source is Bunter.

Of the 908 bottled water samples, 127 (14%) were found to have concentrations >2 µg U/L (Table 1). All bottled water with a content >2 µg U/L can be explained with high probability by geogenic factors. Elevated uranium concentrations are observed in the Black Forest, the Keuper area of southern Germany, the Weser Mts., the forelands of the Alps, and in the northeastern and southwestern forelands of the Harz Mountains (Fig. 1).

The elevated values at the north and southwest edges of the Harz Mts. are derived from the Zechstein formation water rising through the fault zones in these areas (Birke et al., 1995). The slightly elevated uranium concentrations at the north edge of the Harz are from the Subhercynian sediments there and are present in the bottled water from Bad Suderoder Mineralbrunnen (3.57 µg U/L), Blankenburger Wiesenquell (1.13 µg U/L) and Bad Harzburger Urquell (1.47 µg U/L(Fig. 1).

Permo-Triassic sediments in Hesse, Lower Saxony, Saxony–Anhalt, and southwestern Germany are related to the elevated and anomalous uranium concentrations >2 µg U/L (Schnug et al., 2008; Knolle

Table 1
Uranium in stream water, mineral water and tap water in Germany and the EU (in µg/L).

Statistical parameters	Stream water ($N=944$)	Mineral water ($N=908$)	Mineral water EU[a] ($N=1785$)	Tap water ($N=164$)	Tap water[b] ($N=476$)	Tap water EU[a] ($N=579$)
Minimum	< 0.002	< 0.0005	< 0.0005	0.0012	< 0.015	0.0010
Maximum	43.7	16.0	229	9.0	8.54	56.2
Arithmetic mean	0.902	0.938	1.31	0.506	0.430	0.996
Median	0.330	0.170	0.209	0.0801	0.135	0.304
Mode	0.009	< 0.0005	< 0.0005	0.0032	< 0.015	0.0045
Values >2.0 µg/L	113	127	270	9	19	60
Detection limit	0.002	0.0005	0.0005	0.0005	0.015	0.0005
Values below detection limit	3	11	17	0	198	0

[a] EGS (EuroGeoSurveys) Geochemistry Expert Group (2010).
[b] Schnug et al. (2008).

and Schnug, 2009). Bunter and Keuper sandstone are the main uranium source, followed by crystalline and Permian aquifers.

Slightly elevated values also occur in the Quaternary gravels that have a largely carbonate composition in the foreland of the Alps and in the Molasse (Fig. 1).

The known uranium enrichments in the Kupferschiefer of the Rotliegende are reflected in the elevated concentrations in bottled water from Friedrichsroda (Thüringer Waldquell: 3.81–4.72 µg U/L, Rennsteigsprudel: 0.496–7.11 µg U/L), Schmalkalden (Tau frisch Mineralbrunnen: 5.16 µg U/L), the Saale–Unstrut area, and Leissling (Leisslinger Mineralbrunnen: 8.45 µg U/L). The anomalous concentrations in the northern Black Forest (Fig. 1) are from the Precambrian paragneiss (Griesbacher Mineralquelle: 16 µg U/L) and can have been dissolved from the highly stressed gneiss by CO_2-rich water.

The elevated concentrations in the Weser Mts. (Fig. 1) are derived from the Keuper (Steinmergel and Gipskeuper).

Comparison of the median values for Germany and its neighbors (Fig. 2) shows that the medians from France, Italy and Switzerland are

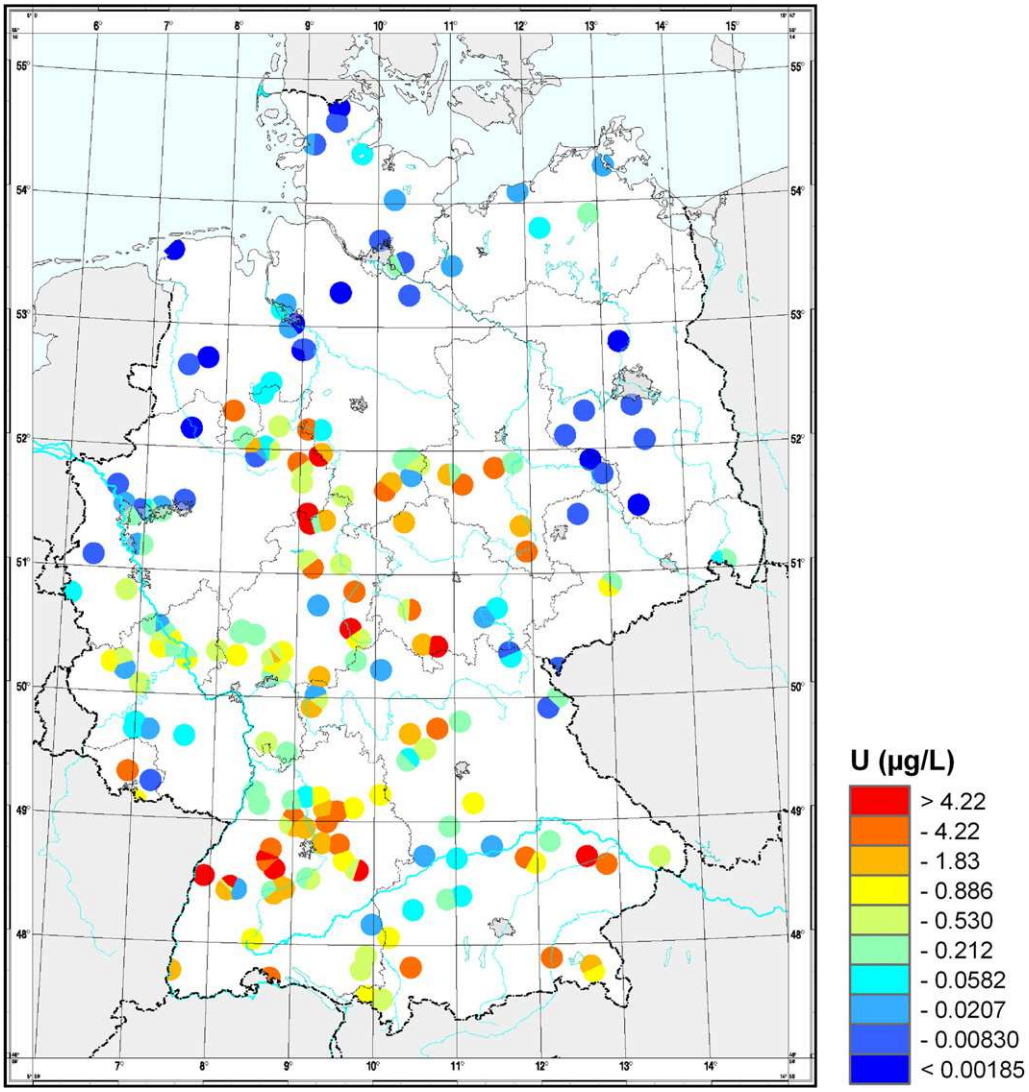

Fig. 1. Distribution of uranium in German mineral water as indicated by the median value at the locality (in µg/L).

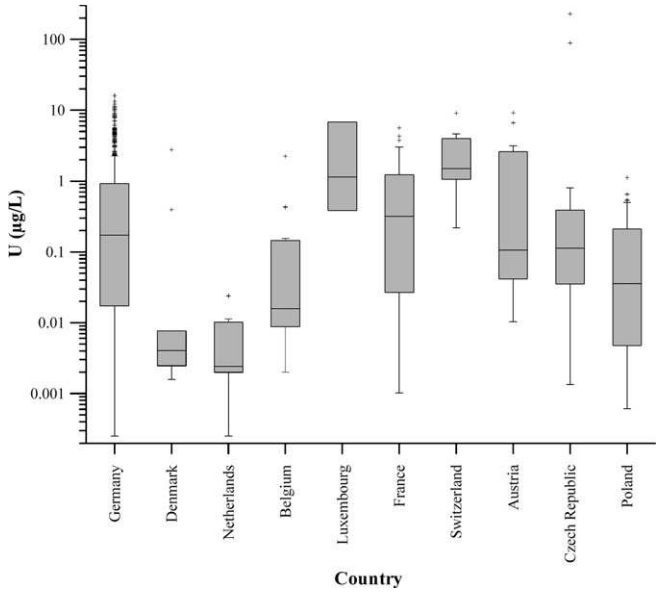

Fig. 2. Comparison of uranium concentrations in bottled water in Germany and neighboring countries (boxplot).

higher than the European and German median. The medians in bottled water from Austria, Denmark, The Netherlands, Belgium, the Czech Republic and Poland are significant lower. Up to date no agreement on a drinking water limit for uranium was reached by the EU. Germany operates with two "recommended" values, 10 and 2 µg U/L (Konietzka et al., 2005). The provisional upper limit given in the third edition of the Guidelines for Drinking Water is 15 µg U/L (WHO, 2004b). It is high time that the European Public Health Authorities agree on a upper limit value for U in mineral and drinking water.

Corresponding to the geological influence on groundwater, the highest median values for bottled water (Figs. 1–4; Table 2) are from Hesse, the Upper Rhine/Main regions, the catchment areas of the Danube, as well as in central Germany (northeastern and southwestern foreland area of the Harz Mountains). The lowest median values are for bottled water from northern Germany and the Lower Rhine/Maas region.

About 14% of the analyzed bottled water in Germany contained >2 µg U/L in 2008 and 2009. The strikingly high uranium concentration of the Griesbacher Mineralquelle (16 µg U/L) from the Black Forest is due to the natural uranium deposit in this region (Walther and Dill, 1995). The water from this spring flows through fine joints in the crystalline basement, which in the Black Forest consists of pre-Variscan and Variscan highly stressed gneiss interspersed with anatectic granite veins.

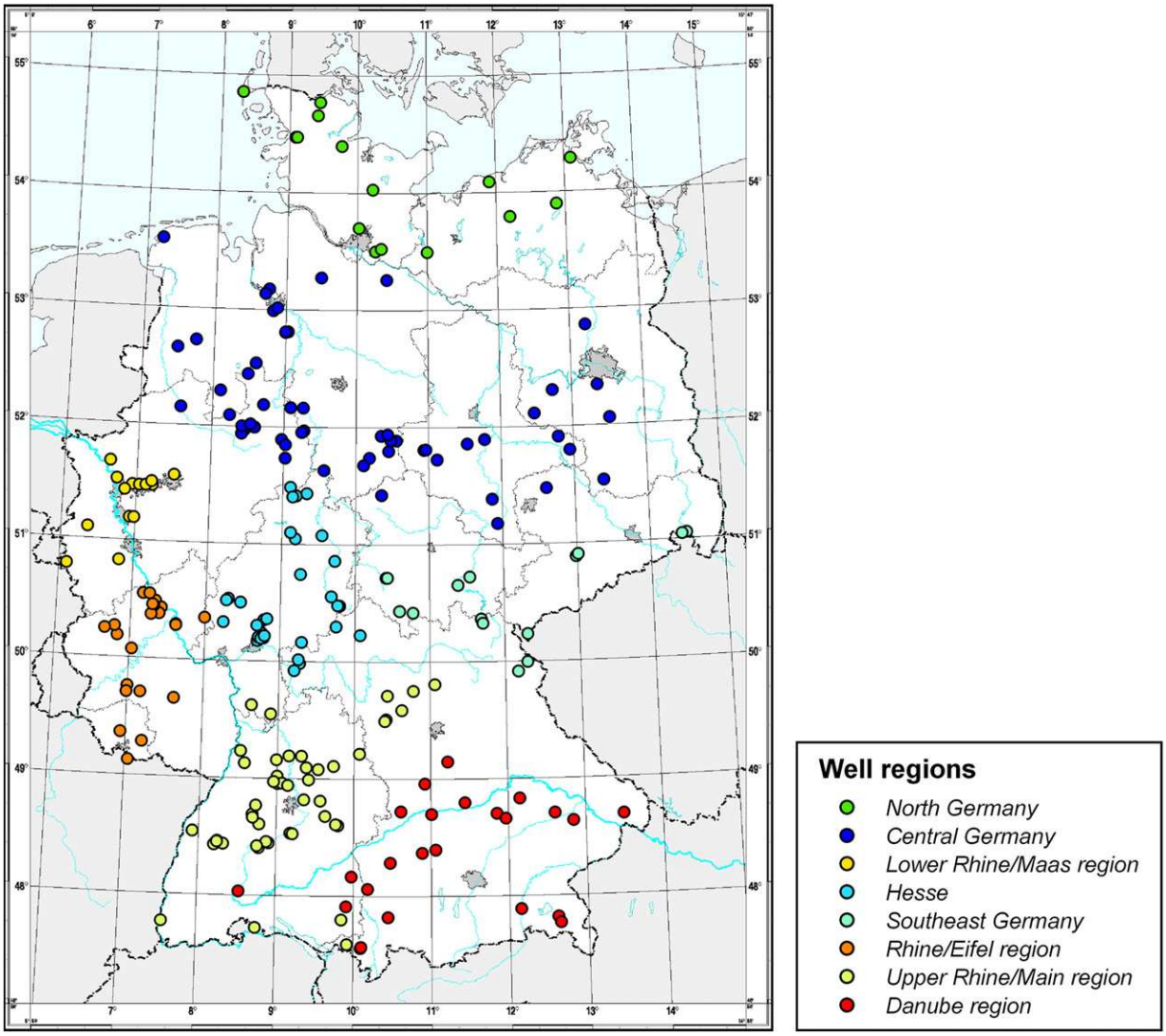

Fig. 3. Distribution of mineral water wells in the various regions of Germany.

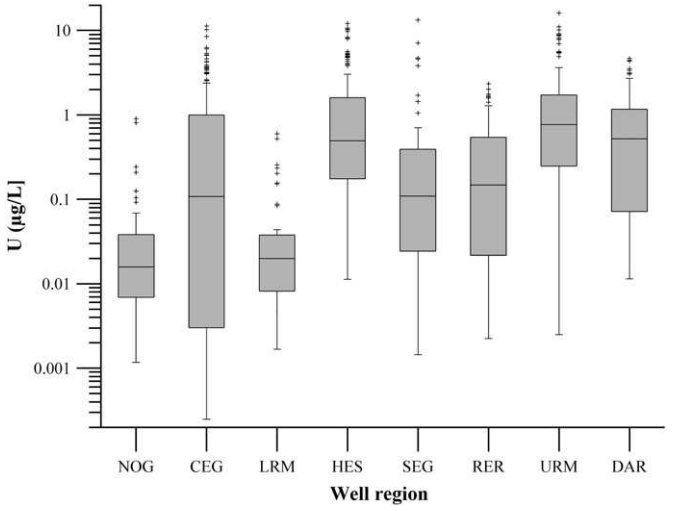

Fig. 4. Comparison of uranium concentrations in bottled water according to region in Germany (boxplot) (NOG — North Germany, CEG — Central Germany, LRM — Lower Rhine/Maas region, HES — Hesse, SEG— Southeast Germany, RER — Rhine/Eifel region, URM — Upper Rhine/Main region, and DAR — Danube region).

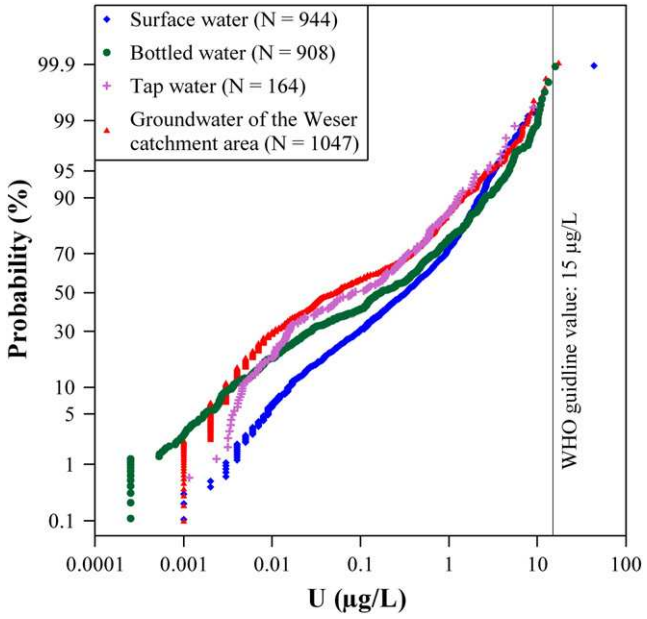

Fig. 5. Cumulative probability diagram for uranium in German surface, tap, bottled and groundwater.

The range of uranium concentration in bottled water is even much wider than that of tap water (Table 1). The cumulative probability diagram shows that the natural variation in uranium concentration in German bottled water covers at least five orders of magnitude (Fig. 5).

4.2. Uranium concentrations in tap water

The uranium concentration in tap water (Table 3, Fig. 6) has a similar distribution to that in bottled water (Fig. 1) and partly to that for surface water (Birke et al., 2009). This is not surprising since the uranium concentration in tap water is influenced to equal extents by the same geological origin and the same anthropogenic uranium emissions as for bottled and surface water (Fisenne, 1994; Birke et al., 2009).

The uranium concentration in tap water varies between 0.0012 µg U/L and 9.0 µg U/L in Germany and between 0.0010 µg U/L and 56.2 µg U/L in Europe as a whole (Tables 1 and 3). Recent studies confirm the median value reported by Schnug et al. (2008); for tap water in Europe as a whole the median value is double that for Germany (Table 1). The variation in uranium concentration of tap water in Europe is much wider than that in Germany.

The highest uranium concentrations in tap water were obtained in northern Hesse between the Knüll and Weser mountain areas, in the northern Black Forest, in Thuringia, in the Fichtel Mts., in northern Bavaria, and the catchment area of the Main River. Low elevated concentrations are observed in the eastern and northern parts of Mecklenburg–Vorpommern (Fig. 6). Elevated concentrations in northern Germany may be assumed to be derived from weathered till from Scandinavia (Birke et al., 2009), which is the source of uranium in the surface water there. In Hesse, Thuringia, Bavaria, and Baden–Württemberg the geogenic sources (Permo-Triassic sediments) are the same as those for bottled water.

The maximum values in German tap water are regionally very different, varying between 0.00115 and 9.0 µg U/L (Table 3). The uranium concentration in local tap water can considerably affect the human uranium intake via homemade and locally produced beverages. However, only 3% of the daily tap water consumption is in tea, coffee, etc. (Anke et al., 2009). The highest mean and median values were determined in Thuringia, followed by Bavaria and Baden Württemberg and Hesse (Table 3). The highest maximum values were in Bavaria, Thuringia, Hesse, Saxony and Lower Saxony.

The cumulative probability diagram shows that uranium concentrations are lower in German tap water than in German surface water (Fig. 5). The diagram shows that the natural variation in uranium concentration in German tap water covers at least four orders of magnitude.

4.3. Human exposure to uranium

Because uranium is present in numerous minerals in the Earth's crust, it is also present in plant and animal foods (Anke et al., 2009), groundwater, surface water, and tap water (Sparovek et al., 2001; Schnug et al., 2008; Schulz et al., 2008; Birke et al., 2009). The solubility, mobility, and bioavailability of uranium compounds in abiotic and biotic systems depends to a high degree on the uranium

Table 2
Statistical parameters of uranium in mineral water in the German well regions (in µg/L).

Well region	*N*	Minimum	Maximum	Arithmetic mean	Median	Modal	Values below DL[a]	Values >2 µg/L
Germany	908	<0.0005	16.0	0.938	0.170	<0.0005	11	136
Danube region	50	0.0114	4.65	1.04	0.455	0.0167	0	10
Hesse	142	0.0113	12.1	1.62	0.493	0.0118	0	41
Central Germany	287	<0.0005	11.3	0.831	0.112	<0.0005	11	48
North Germany	70	0.00117	0.900	0.0531	0.0134	0.00636	0	0
Upper Rhine/Main region	155	0.00250	16.0	1.57	0.736	0.00799	0	30
Rhine/Eifel region	104	0.00225	2.33	0.363	0.129	0.0122	0	2
Southeast Germany	48	0.00144	13.3	0.894	0.110	0.0280	0	5
Lower Rhine/Maas region	52	0.00168	0.601	0.0593	0.0194	0.00723	0	0

[a] Detection limit (DL) = 0.0005 µg/L.

Table 3
Uranium concentrations in German tap water (in µg/L).

Germany state	Statistical parameters				
	N	Arithmetic mean	Minimum	Maximum	Median
Schleswig–Holstein	7	0.0904	0.00115	0.561	0.00373
Lower Saxony	49	0.284	0.00339	4.42	0.0162
Mecklenburg–Western Pomerania[a]	25	0.28	0.05	0.94	
Mecklenburg–Western Pomerania	8	0.335	0.00409	0.769	0.288
Brandenburg[a]	22	0.31	0.08	1.0	
Brandenburg	8	0.276	0.00234	0.124	0.00675
Berlin	6	0.200	0.0501	0.413	0.196
North Rhine–Westphalia	13	0.314	0.00447	1.08	0.178
Saxony–Anhalt[a]	19	0.76	0.09	2.8	
Saxony–Anhalt	18	0.133	0.00459	0.593	0.311
Rhineland–Palatinate	5	0.0332	0.0108	0.0769	0.0235
Hesse	10	1.31	0.0127	5.54	0.289
Thuringia[a]	55	2.4	0.11	8.6	
Thuringia	3	2.56	0.898	3.85	2.94
Saxony[a]	31	1.1	0.09	4.5	
Saxony	8	0.198	0.00689	0.591	0.101
Baden–Württemberg	7	0.955	0.429	1.84	0.759
Bavaria	21	1.36	0.00834	9.00	0.537

[a] Anke et al. (2009).

species (Merkel et al., 2002; Dobler et al., 2007a,b; Bernhard and Geipel, 2007). The regular consumption of bottled water and/or tap water that contains uranium is often the main source of uranium in the body.

Within the scope of the current bottled water survey (EGS, EuroGeoSurveys Geochemistry Expert Group, 2010), uranium concentrations in Germany were determined from <0.0005 to 16.0 µg/L in bottled water and between 0.0012 and 9.0 µg U/L in tap water. The mean concentrations were 0.938 µg/L and 0.506 µg/L, respectively (Table 1).

WHO gives a guideline value for tap water of 15 µg U/L (WHO, 2004b), replacing the provisional value of 9 µg U/L of 2003, which had replaced the previous value of 2 µg U/L (WHO, 1998a,b, 2006). Independent of this, the TDI (Tolerable Daily Intake) of WHO remains unchanged at 0.6 µg/kg body weight. The guideline value for daily intake of uranium consumed in tap water, however, was raised from 10% of the TDI (WHO, 1998a,b) to 50% (WHO, 2004b, 2006) and then 80% (WHO, 2004a). This relatively high value is based on the assumption that the 2 L/day water consumption by a person with 60 kg body weight represents the principal uranium intake. Only about 2–4 µg U/day are consumed in solid foods, independent of the foodstuff (Schnug and Schnug, 2004). This is doubled by an average intake of 2 L/day of water with a concentration of 1–2 µg U/L or increased ten-fold by water with a concentration of 10–20 µg U/L.

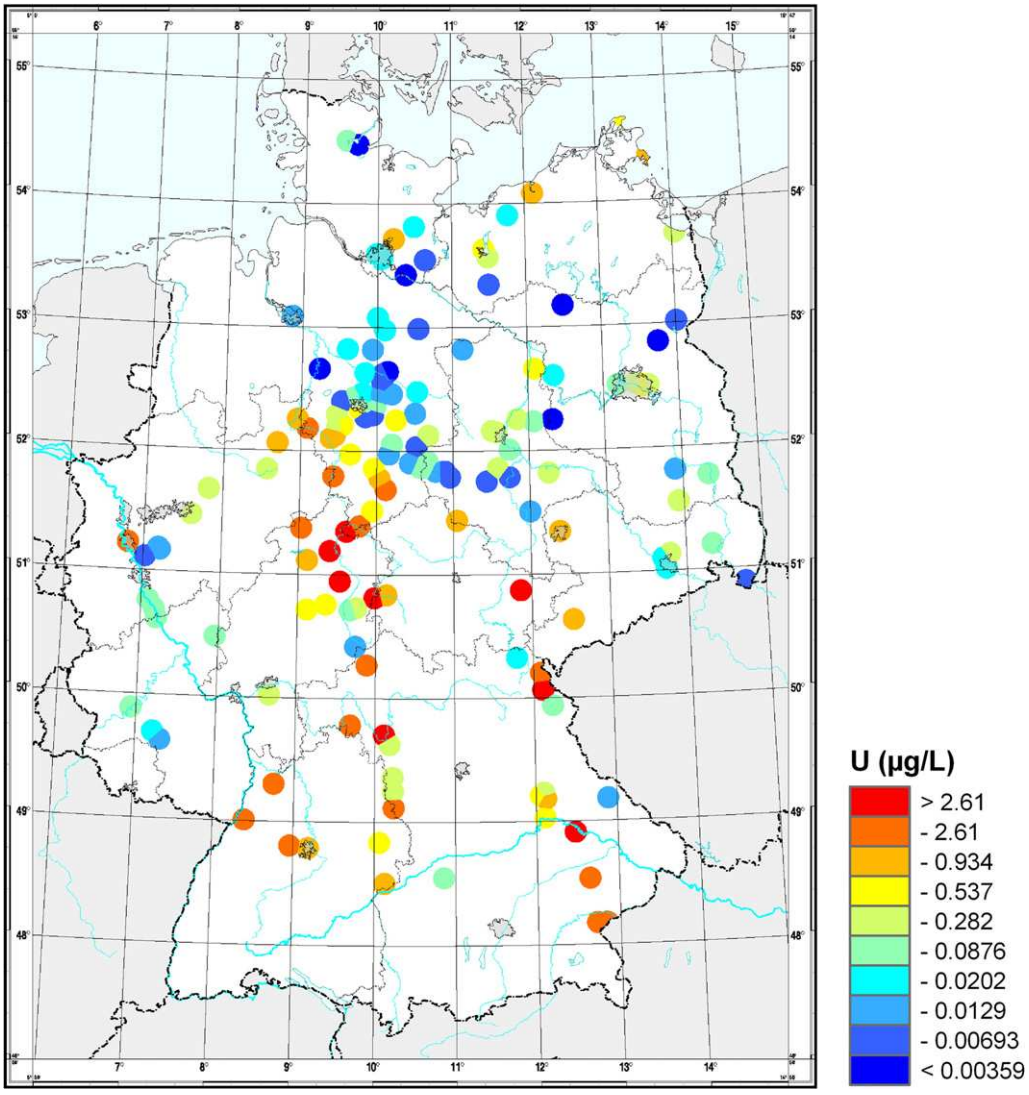

Fig. 6. Distribution of uranium in tap water in Germany (in µg/L).

The uranium load in the human body depends essentially on the uranium content of ingested food and drink. Depending on the solubility of the uranium compound, only 0.2–2% of the ingested uranium is taken up in the stomach and intestines (Dobler et al., 2007a), the remainder is not absorbed and is excreted in the feces. The absorbed uranium, mainly as the carbonate or bicarbonate (Bernhard and Geipel, 2007), is excreted via the kidneys, which are the most sensitive organ of the body to the chemical toxicity of uranium. Studies have shown a slight increase in uranium concentration in urine with increasing age (Kemper et al., 2006), which in comparison to regional geogenic differences places only a secondary role (Schnug et al., 2008).

The uranium dissolved in bottled water and tap water is toxic for the human kidney regardless of the uranium compound (Bernhard and Geipel, 2007). Studies in Finland (Auvinen et al., 2002; Kurttio et al., 2002, 2006a,b) and Canada (Limson-Zamora et al., 1998, 2003) confirm the nephrotoxicity of uranium for humans. Exceptionally high uranium concentrations have been measured in private drilled wells in granite areas in southern Finland (Muikku et al., 2007). In an epidemiological study on renal effects of uranium in southern Finland the uranium concentration in urine varied from 1 to 5650 ng/L (Kurttio et al., 2002). Consequently, elevated concentrations of uranium in the hair of Finnish subjects were also observed in the continuation of the study (Muikku et al., 2007), and have also been confirmed in human-biomonitoring program in Germany (Dobler et al., 2007a). Kurttio et al. (2002, 2006a,b) observed that a daily intake of very small amounts of uranium in tap water (2–1920 µg U/L) can lead to kidney damage. Recent research in France has demonstrated brain damage by the deposition of uranium in the hypothalamus (IRSN, 2005). Especially babies and small children are vulnerable.

The Federal Environmental Agency (UBA), therefore, has recommended a guideline value of 10 µg/L uranium in tap water since 2004 (UBA, 2005; Dobler et al., 2007a). A health-based action value as might be derived from this guideline value on the basis of an official recommendation from the Federal Environmental Agency (UBA, 2003). This value, which would be an effect when the guideline value is temporarily exceeded, could be about 20 µg U/L. It would be safe for exposure periods of up to 10 years (Konietzka et al., 2005).

The Bundesrat of the Federal Republic of Germany passed an upper limit for uranium in bottled water. Bottled water used for the preparation of baby food may contain no more than 2 µg U/L (MinTafWV, 2006).

The uranium concentration in blood, blood plasma, and urine in humans has been monitored in a countrywide program (Werner et al, 1999; Kemper et al., 2004; Dobler et al., 2007a). The results show values between 0.62 ng/L and 173.44 ng/L in urine, between 2.01 and 28.04 ng/L in blood (Dobler et al., 2007a). For all three bodily fluids, the highest mean values were found in persons from the Halle region (Dobler et al., 2007a). For most of the world population the mean concentration in urine is about 10.0 ng U/L (Li et al., 2005). For Germany the mean uranium concentration in urine is 8.71 ng/L (Dobler et al., 2007a). The mean value is 1.5 times higher than in blood (4.02 ng U/L; Dobler et al., 2007a, 2008).

The data show that the range of uranium concentration in blood and urine (Dobler et al., 2007a) is only two orders of magnitude in contrast to the natural variation in bottled water and tap water (Fig. 5). The data from the human-biomonitoring program was linked with data from environmental monitoring to investigate the relationship between uranium concentrations in the urine of young adults and environmental factors (Dobler et al., 2008; Pesch et al., 2009; Schmidt et al., 2009). The results show that the concentrations in surface water correlate best with those in urine (Schmidt et al., 2009).

Anke et al. (2009) analyzed the uranium content of 116 foodstuffs and beverages and observed that it varies also greatly. Vegetable foodstuffs contain concentrations as low as 0.8 µg U/kg DM (dry matter) (in margarine), to >100 µg U/kg DM (mixed mushrooms, Anke et al., 2009). For animal foodstuffs they found lower uranium concentrations (0.7 µg U/kg DM–16 µg U/kg DM). The uranium content in plants in the immediate area of old mining and waste dumps, e.g., Ronneburg, is significantly elevated. Anke et al. (2009) found that leafy vegetables accumulated much uranium, whereas tubers, onions, fruits and grains stored less uranium. They also observed the uranium content decreases significantly with increasing age of the vegetation. So it is clear that the uranium transfer to the food chain of humans is significantly affected by the geological origin of the soils and the groundwater as well as the regional environmental conditions of the flora and tap water reservoir. Phosphate fertilizers (Haneklaus and Schnug, 2008; Huhle et al., 2008; Jacques et al., 2008; Rogasik et al., 2008; Birke et al., 2009) and industrial emissions cannot be completely excluded as a source of uranium, especially in areas of intensive agriculture and in industrial areas.

Principal component analysis of the data for human body fluids (whole blood, blood plasma, 24 h-urine, Dobler et al., 2007b) clearly shows that uranium accumulates in the body via intake of food and especially drink. The principal component analysis yielded a REE factor (Nd, La, Ce, Pr, Yb, Y, Sm, Er, Tm, Gd, and Dy) which also included uranium and thorium was determined for whole blood and blood plasma (Table 4). A similar factor was also obtained for both German and European bottled and tap water as well as for surface water in Germany; this factor had the highest eigenvalue of the analysis, however without uranium. The REE factor results mainly from the very similar migration behavior of the rare earth elements in surface water and groundwater. In whole blood and blood plasma, uranium shows a distribution similar to that of the REE. The preliminary results of principal component analysis for human body fluids confirm that it is possible to characterize biological and anthropogenic multi-element associations (factors) also in these biological media as well as the geogenic and anthropogenic associations in water.

4.4. Results of the principal component analysis in bottled and tap water

Principal component analysis was used to investigate the correlation between the 71 inorganic parameters measured on the bottled and tap water samples. Complex multivariate relationships among variables are revealed by this statistical method that are not normally shown by a simple correlation analysis. This method was used to determine the areal distribution patterns of the high

Table 4
Uranium patterns in German surface, bottled and tap water in comparison to human body fluids (summary of preliminary principal component analysis results).

Sample media	Factor number	Source	Pct of var	Element association of factor
Surface water (Birke et al., 2009)	Factor 5	Geogenic	3.39	HCO_3^-–Ca–pH–Mg–EC–(U–Se)
	Factor 14	Anthropogenic	1.69	U–Se–Mg–Li–(SO_4^{2-}–Mo)
Mineral water	Factor 11	Geogenic	2.75	Be–Bi–(Si–U)
	Factor 10	Anthropogenic	2.92	NO_3^-–Se–Cu–U–(Mo–V)
Tap water	Factor 6	Geogenic	5.30	Mg–Sr–HCO_3^-–Ca–EC–U
	Factor 8	Anthropogenic	4.07	Ba–Eu–As–(U–Sm–NO_3^-)
Blood plasma (Dobler et al., 2007b		Biogenic		Nd–La–Ce–Th–U–Pr–Yb–Y–Sm–Er–Tm–(Gd–Dy)
Whole blood (Dobler et al., 2007b)		Biogenic		Nd–La–Ce–Y–Sm–Er–Th–Yb–Pr–U–Gd–Dy–(Bi–V–Eu–Zr)
24 h-urine (Dobler et al., 2007b)		Anthropogenic		U (single element component)

correlated element associations in the analyzed bottled water. Classification of the factors permits the examination of the complex causes of geochemical anomalies and their influence on element enrichment and migration (Figs. 7 and 8).

The principal component analysis of surface water (Birke et al., 2009) revealed two factors with high factor loads of uranium (Table 4). Elevated and anomalous factor scores of the factor U–(Se–Mg) in surface water are correlated with agricultural use of phosphate fertilizer in the eastern and northern foreland areas of the Harz Mountains, the Thuringian basin, the hill area of Saxony, the northern part of Upper Rhine Valley, and the area north of Stuttgart. The second factor in surface water HCO_3^-–Ca–pH–Mg–EC–(U–Se) correlates with the presence of uranium in soluble stable carbonate complexes in the Weichselian moraine landscapes in northern Germany, in the boulder clay soils of southern Germany, and the areas of marly rocks in the Franconian and Swabian Jura regions (Birke et al., 2009).

The results for bottled and tap water were similar to those for surface water. The results of the principal component analysis for tap water, however, should be viewed with reservation owing to the low number of samples relative to the high number of analyzed parameters. For a principal component analysis, at least three times as many samples should be available as analyzed parameters.

The results of the principal component analysis of the data for bottled water are given in Tables 5 and 6. The distribution of the factor NO_3^-–Se–Cu–U in the bottled water is very close to the distribution of the factor U–(Se–Mg) for surface water, confirming the influence of fertilizer application in agriculture. The presence of uranium together with NO_3^-, Se and Cu of factor 10 (Tables 4 and 5) also points to the influence of fertilizers. Other researchers have also determined a significant correlation between anthropogenic origins and uranium together with the parameters NO_3^-, SO_4^{2-}, K, Cl^-, and EC (Schäf et al., 2007; Knolle, 2008; Lindemann, 2008) in bottled and tap water in Germany (Knolle et al., 2008).

The anomalous scores of the factor NO_3^-–Se–Cu–U in bottled water (Fig. 7) occur in the areas of Keuper sandstone in the Swabian and Frankonian Jura and in the areas of Bunter in the Weser Mts. This factor explains 2.92% of the total variance (Table 5).

Bottled water from areas of crystalline rock (especially gneiss and granite) in the Black Forest, and the Franconian Forest yielded anomalous factor scores of the factor Be–Bi–Si–(U) (Fig. 8). This factor explains 2.75% of the total variance.

In the principal component analysis of all European bottled water (EGS, EuroGeoSurveys Geochemistry Expert Group, 2010) was determined the factor U–SO_4^{2-}–Mg–EC–Na, which explains 4.2% of the total variance. There are anomalous scores of this factor in the Czech Republic, Slovakia and Hungary, as well as Lithuania, Belarus, and Finland. In Germany, slightly elevated scores of this factor are observed in the Swabian and Franconian Jura, in Thuringia, and the

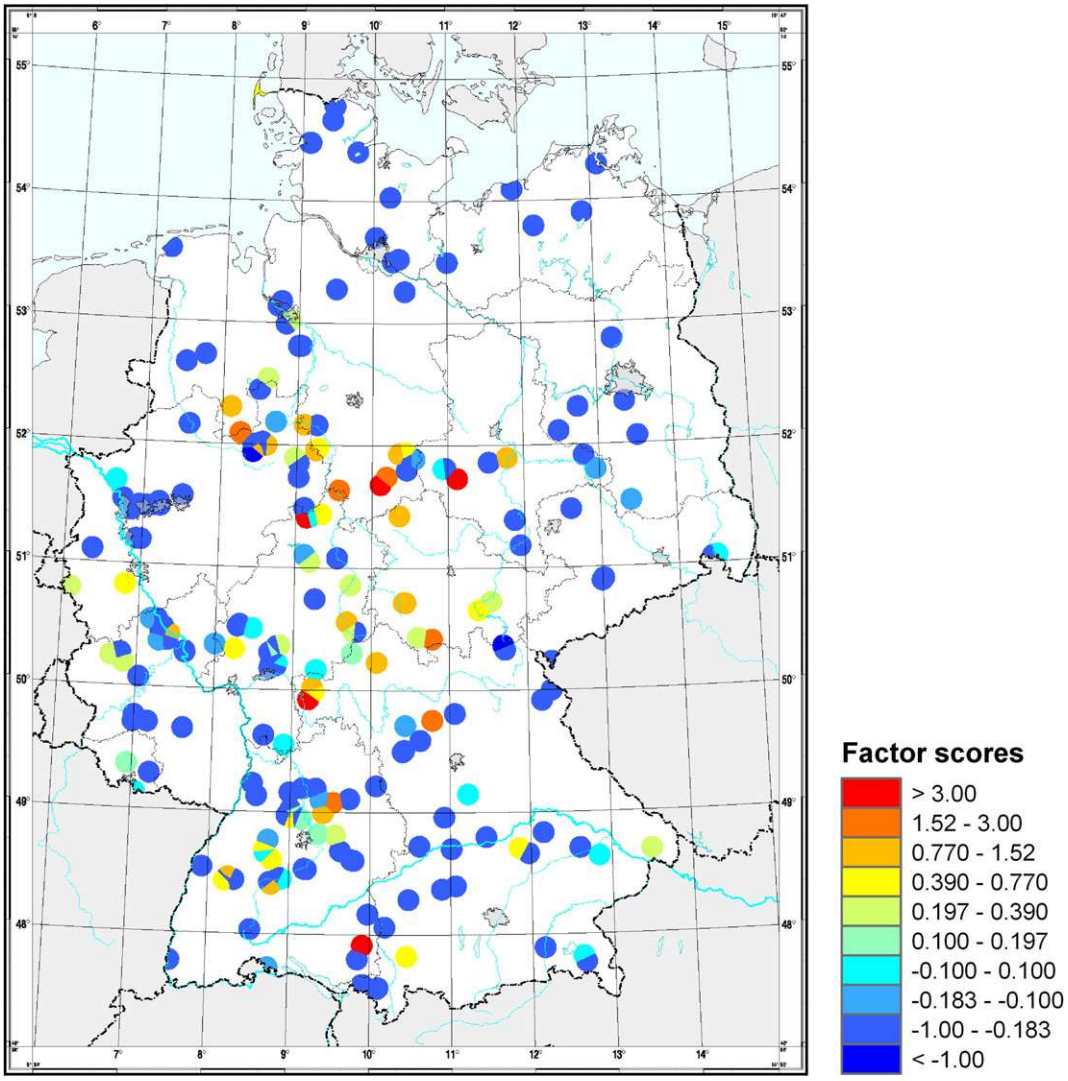

Fig. 7. Distribution of the factor values for the factor NO_3^-–Se–Cu–U in German bottled water.

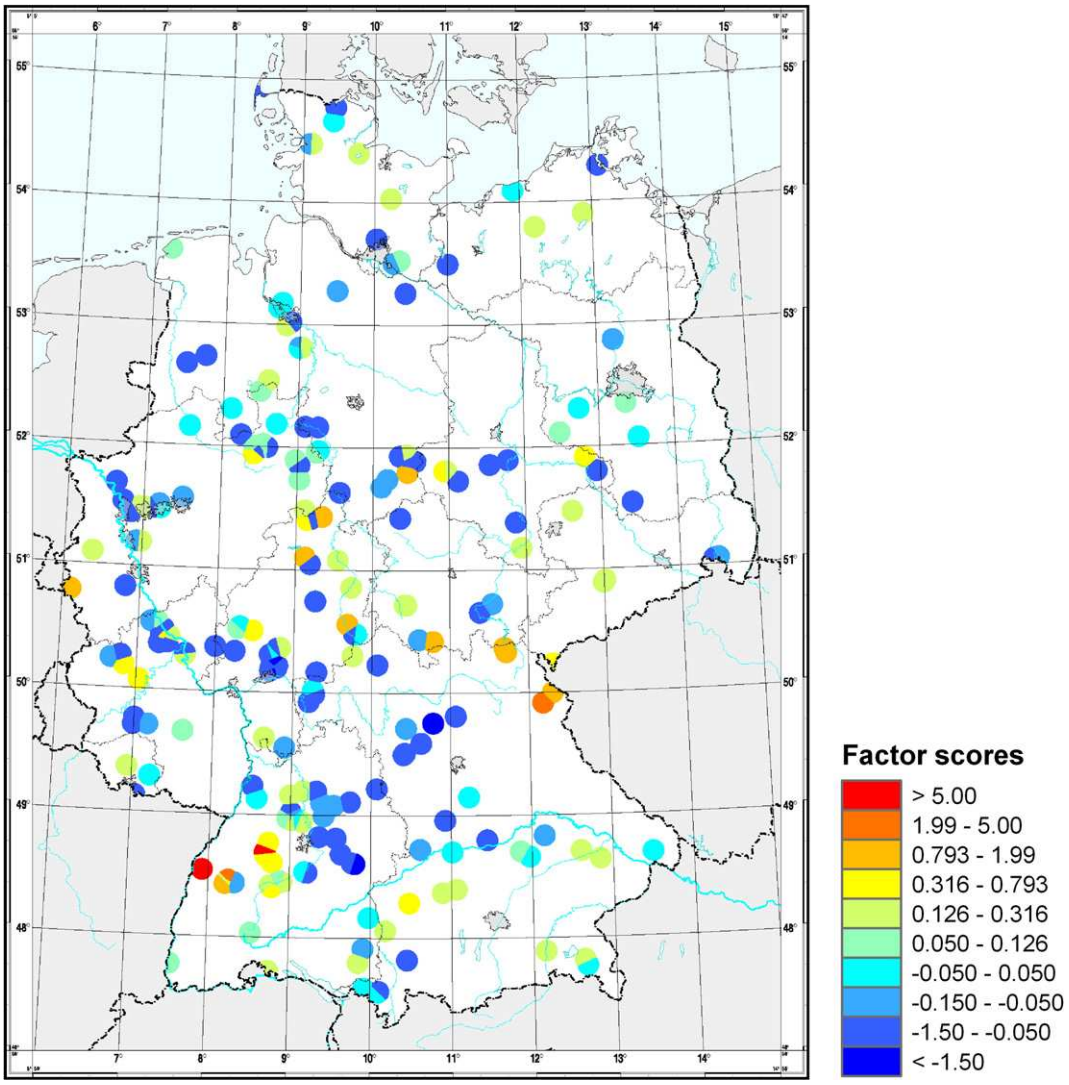

Fig. 8. Distribution of the factor values for the factor Be–Bi–(Si–U) in German bottled water.

Weser Mts., correlating with the known geogenic sources of uranium in those areas (Bunter and Keuper strata, crystalline basement). An anthropogenic influence (e.g., fertilizers) in this factor cannot be excluded, however, owing to the high factor loading values.

Anthropogenic influence (fertilizers) as well as a typical geogenic factor can also be seen in the results of the principal component analysis in all researched sample media (surface, bottled and tap water; (Table 4). The anthropogenic influence is indicated in all three water types by the element association U ± Se, Mg, NO_3^-, Mo, and Li. The geogenic factors indicate various lithological sources: gneiss, granite, Bunter and Keuper, as well as the boulder clay and marly rocks of Weichselian moraine landscapes.

5. Conclusions

Elevated uranium concentrations (>2 µg U/L) in German bottled and tap water can be explained with high probability by geogenic factors. They are related to the (Permo-)Triassic sediment complexes observed in Hesse, Lower Saxony, Saxony–Anhalt and southwestern Germany. The main uranium source rocks are Bunter and Keuper sediments, only secondarily crystalline basement rocks (paragneiss in the Rench River area, Black Forest) and Permian (Thuringia: Friedrichroda, Schmalkalden) aquifers. The geology of the bedrock of a source is the predominant factor influencing the uranium content of bottled water.

The risk of accumulation of uranium in agricultural soils and in groundwater by application of phosphate fertilizers is a serious threat for environment and the quality of bottled and tap water.

The anthropogenic influences determined by the principal component analysis show that bottled and tap water in Germany have a significant anthropogenic (mainly phosphate fertilizers) uranium

Table 5
Principal component analysis results for bottled water.

Factor	Eigenvalue	Pct of var	Element and factor loading
10	2.046	2.92	NO_3^- (0.703), Se (0.694), Cu (0.649), U (0.546)
11	1.924	2.75	Be (0.825), Bi (0.734), Si (0.330), U (0.299)

Table 6
Communalities for geochemical elements and parameters resulting from principal component analysis of bottled water.

Parameter	Communality
Be	0.735
Bi	0.581
Cu	0.557
Se	0.578
Si	0.665
U	0.666
NO_3^-	0.617

load. The distribution of high factor scores of the anthropogenic uranium factors prove that the groundwater circulation is associated for anthropogenic pollutions such like phosphate fertilizers and other industrial emissions. The situation for tap water is somewhat more complicated than for bottled water in that in many regions tap water is derived from surface water or shallow groundwater sources.

In agreement with the published uranium data for untreated water and tap water in Mecklenburg–Vorpommern at more than 80 waterworks values lie between 0.5 and 5.0 µg U/L with a maximum of 26.4 µg U/L (Baumann and Puchert, 2007). Regions in Germany where increased uranium content in surface water could not be sufficiently explained by geological factors and which are supposedly related to agricultural activities (phosphate fertilizers) have been determined by Birke and Rauch (2008).

The natural variation in uranium in bottled and tap water is much like that in ground and surface water. The variation in bottled water, groundwater and surface water covers at least five orders of magnitude, in tap water only four orders of magnitude.

The intake of uranium via water can be reduced very effectively by a conscious selection of the water used for drinking. Removal of uranium from bottled and tap water is technically possible and quite effective (Jekel et al., 2007; Hagen, 2008) and therefore can be required by law.

Revision of the German Drinking Water Regulations of 2001 and the Mineral Water Regulations of 2006 is overdue, because they do not include the monitoring of uranium. There is international agreement that there can be no upper limit for radioactive elements below which no consequences to health can be assumed. For this reason a provisional guideline value (15 µg U/L) has been given by the World Health Organization (WHO, 2004b). Though it is not of a legally binding nature in the European Community (EU Guidelines for Drinking Water, 1998) nor in Germany. It could be the starting point for EU and national legislation. Using the limiting value for water for baby food (MinTafWV, 2006), a value of 2 µg U/L is in discussion for tap water, because the main source of uranium for humans is tap water and bottled water. For this reason, it should be made obligatory for uranium to be included on the label of bottled water throughout the EU as a measure for consumer protection.

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