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....a classic real-world scientific problem. Weighing risks, weighing uncertainties. Most people never understood that the majority of scientific problems took this form. Acid rain, global warming, environmental cleanup, cancer risks - these complex questions were always a balancing act, a judgment call. How good was the research data? How trustworthy were the scientists who had done the work? How reliable was the Computer simulation? How significant were the future projections? These questions arose again and again. Certainly the media never bothered with the complexities, since they made bad headlines. As a result, people thought science was cut and dried, in a way that it never was. Even the most established concepts - like the idea that germs cause disease - were not as thoroughly proven as people believed ... (*From: "Timeline" by Michael Crichton 1999*)

Contribution of uranium in drinking waters to the daily uranium intake of humans - a case study from Northern Germany

Ewald Schnug, Heike Steckel and Silvia Haneklaus[1]

Abstract

Uranium (U) is a naturally occuring radioactive and toxic heavy metal, unavoidable taken in with solid and liquid food, accumulated preferably in kidneys, liver and bones where it has a potential for inducing cancer. The daily intake with solid food amounts to 2 - 4 µg U. The total U intake, however, is mostly dependent on the U content of the water consumed. In a case study the contribution of drinking water to the U intake was investigated. The median of U concentrations in tap and private well waters was beyond the detection limt of 15 ng l^{-1} U ("zero"), the maximum values were 1.44 and 8.95 µg l^{-1} U, respectively. In contrast, the median of 17 bottled mineral waters preferably consumed in the test area was 0.44, the maximum value was as high as 10.6 µg l^{-1} U. This means for the total daily U intake that by drinking tap water the risk for increased U uptake is near zero, but by drinking mineral water the U intake could be 10-fold higher in the worst case. Provenance and U content of bottled waters are defined. Therefore, selecting "zero" U waters is a most efficient tool to reduce health risks from U to a minimum. Prerequisite for this approach is, however, the compulsory labelling and information of U in bottled as well as in tap waters. The contribution provides also a discussion of critical and guideline values for U in drinking waters.

Key words: alimentation, critical values, drinking water, guideline values, health, radioactivity, risk, shallow ground water, uranium, uranium uptake

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Beitrag von Uran in Trinkwässern zur täglichen Uranaufnahme von Menschen – eine Fallstudie aus Norddeutschland

Zusammenfassung

Uran (U) ist ein natürliches, radioaktives und toxisches Schwermetall, das unvermeidbar mit fester und flüssiger Nahrung aufgenommen wird, sich bevorzugt in Nieren, Leber und Knochen anreichert und dort vorwiegend Krebserkrankungen auslösen kann. Mit fester Nahrung nimmt der Mensch täglich 2 - 4 µg U auf. Die Höhe der U-Aufnahme insgesamt wird jedoch wesentlich durch die U-Konzentrationen des konsumierten Trinkwassers bestimmt. In einer Fallstudie wurde der Beitrag verschiedener Wässer zur täglichen Uran-Aufnahme untersucht. Der Median der U-Gehalte von Leitungs- und Hausbrunnenwasser lag im Untersuchungsgebiet unterhalb der technischen Nachweisgrenze von 15 ng l^{-1} U, die Maximalwerte bei 1,44 bzw. 8,95 µg l^{-1} U. Demgegenüber lag der Median der U-Gehalte von 17 im Untersuchungsgebiet von den Konsumenten bevorzugten Mineralwässern bei 0,44, der Maximalwert bei 10,6 µg l^{-1} U. Für die tägliche U-Aufnahme bedeutet dies für die Konsumenten des Untersuchungsgebietes, dass durch Konsum von Leitungswasser kaum das Risiko einer erhöhten U-Aufnahme besteht, während sich die U-Aufnahme beim Genuss von Mineralwasser im ungünstigsten Fall verzehnfachen kann.

Herkunft und U-Gehalt von Trinkwässern sind eindeutig definiert und ermöglichen durch gezielte Wahl das individuelle Risiko im Hinblick auf gesundheitliche Schäden durch U wirkungsvoll zu minimieren. Voraussetzung hierfür ist jedoch eine obligatorische Informations- und Kennzeichnungspflicht der U-Gehalte, und zwar für kommunale Wasserversorger ebenso wie für die Hersteller von Flaschen- und Mineralwässern. Im Beitrag erfolgt darüber hinaus eine kritische Auseinandersetzung mit Grenz- und Leitwerten für U in Trink- und Mineralwässern.

Schlüsselworte: Ernährung, Gesundheit, Grenzwerte, Leitwerte, Mineralwasser, oberflächennahes Grundwasser, Radioaktivität, Risiko, Trinkwasser, Uran, Uranaufnahme

1 Introduction

Uranium (U) is a radio-nuclide and heavy metal, and part of our natural environment. Although the biological impact of radio-nuclides is mostly considered as being negative (Banks et al., 1995; BEIR III, 1980; BEIR IV, 1990), there are also sources which claim a positive effect of low radiation for life processes, called "hormesis" (Luckey, 1991). This was also proclaimed as being the active principle of a couple of spas and mineral waters (Akerblom, 1994; Franke et al., 1987; Hevesy and Paneth, 1938; Jianli et al., 1993; McNulty, 1991; West, 1954). Typical natural soil background values for U differ between 0.79-11 mg kg^{-1} U in relation to the parent material (Kabata-Pendias and Pendias, 2001).

U concentrations in environmental compartments may be elevated as a result of leaching from natural deposits or due to anthropogenic activities such as mining operations, nuclear industry, disposal of industrial and medical wastes, and last but not least the use of phosphate fertilisers in agriculture (Azuoazi et al., 2001; Barisic et al., 1992; Conceicao and Bonotto, 2000 & 2003; Jamal, 2004; Spalding and Sacket, 1972, Kobal et. al., 1990, Kratz and Schnug, 2006; Schnug et al., 1996, Zielinski et al., 1995 & 2000). Spalding and Sacket (1972) for instance attributed increased U concentrations in North American rivers, ~ 0.7 - 0.9 µg l^{-1}, compared to ~ 0.1 - 0.2 µg l^{-1} in South American rivers (Cothern and Lappenbusch, 1983) to the use of phosphate fertilisers in the region.

Hazardous effects of U on health may occur if humans and animals are contaminated by U compounds through inhalation, ingestion or skin contact. While skin contact with U is particularly a threat for persons working in the U industry, inhalation and ingestion are probable contamination pathways for broader levels of the population who live in U polluted regions. Ingestion of U occurs by drinking water, and through the food chain via crop plants, animal feed, and animal products.

Mammals have a particularly high sensitivity against U (Fellows et al., 1998). Once U enters the organism, it is transferred to the extra-cellular fluids and transported through the blood to others organs. The soluble form, uranyl (UO_2^{2+}) is transported and forms complexes with proteins and anions. The U that is not retained in the body is eliminated by urinary and faecal excretion. U tends to accumulate in the body, preferentially in kidneys, liver, spleen and bones. The risks related to the exposure to U are of radiological and chemical nature. U is certainly only a minor source of concern when considering the contribution of U to the overall level of radioactivity in the environment (Falck and Wymer, 2006). This assessment needs, however, to be re-considered once U is taken up by an organism and incorporated into tissues and bones. The most remarkable damage of U coming along with low and medium contaminations is cancer (Linsalata, 1994). The risk for contracting cancer from incorporated U is impressively highlighted by Gofman (1996): "By any reasonable standard of biomedical proof, there is no safe dose, … just one decaying radioactive atom can produce permanent mutation in a cell's genetic molecule. …citizens worldwide have a strong biological basis for opposing activities which produce an appreciable risk of exposing humans and others to plutonium and other radioactive pollution at any level. …Mutation is the basis not only for inherited afflictions, but also for cancer." This background is simply the reason why the Environmental Protection Agency of the US (EPA) has not set a limit for U in air, but it has set a goal of zero U in drinking water (ATSDR, 1999).

The overall dangers arising from the biochemical toxicity of U as a heavy metal are considered to be about six orders of magnitude higher than those from its radioactivity (Milvy and Cothern, 1990; NRC, 2005). Compared to other heavy metals, the chemical toxicity of U ranges between mercury and nickel, or christoballite and warfarin (Schnug et al., 2006). Hazards by U contaminations are related to the binding of U to organic molecules and are particularly high for kidneys because of peak concentrations during the excretion process. The U accumulation in the body is proportional to the concentration of U in inhaled air, solid and liquid food. The older an individual is, the higher will be the amount of U that is accumulated. This implies that the risk for contracting damages from U generally increases with the time of exposure and thus with age (WHO, 2004). But most experiments conducted to investigate hazardous effects of U alimentations were simply too short (30 days to 2 years; WHO, 2004) to unfold cancerogenic effects and thus are not suitable to draw reliable conclusions for real long-term exposures to U. The accumulation of U depends also on the internal pH in different compartments of an organism (WHO, 2004) and therefore is influenced by individual dietary habits, too. Negative effects of U on biological systems are only conditionally reversible (WHO, 2004). In animal experiments and in *in vitro* tests it was proven that U caused histological changes at all doses tested. Hereby, male individuals seemed to be more susceptible to damages from U exposure than female persons (WHO, 2004). Complex medical syndromes like the "Gulf War Syndrome" also seem to be related to exposures to U (Anon, 1997).

Considering the facts that firstly, U ingestion can not be avoided and secondly, that there is no lower limit for hazardous effects from U (ATSDR, 1999; WHO, 2004), minimising risks for contracting U related hazards imply to keep the daily U intake as low as possible.

With the increased consumption of bottled mineral waters, being advertised as the "better drinking water" (Misund et al., 1999) the relative importance of this source is increasing as is its share in the daily water supply of populations all over the world. Each German consumes statistically about 125 l of mineral water per year, top of

the list are Italians with a nearly 50 % higher consumption per person, followed by Mexico, Belgium and France (http://www.mineralwaters.org & http://www. mineral-wasser.com). Reasons for the increasing consumption of bottled waters are an unspecific fear about pollution of tap water for example by germs (NDR, 2005), nitrate from fertilisation in agriculture (UBA, 2004) and lead from over-aged plumbing (UBA, 1998). Nevertheless, the quality of daily drinking water belongs to the few constants in human life: taps in a household are connected to well-defined public suppliers or private wells, the mineral composition of bottled waters are highly specific and time constant for the individual sources (Dopychai, 2000; Sparovek et al., 2001; Stevenson, 2000), and consumers are fairly loyal to their favourite brands.

Cothern and Lappenbusch (1983) considered the U concentration in drinking water as the main variable factor affecting the daily U ingestion of humans. The objective of the research work reported here was to investigate the effect of different sources of drinking water on the daily U ingestion of urban individuals and to evaluate minimising strategies.

2 Materials and Methods

2.1 Origin of samples

In a region with the postal designation D-38xxx, roughly encompassed by the Northern German towns Braunschweig to the North, Salzgitter to the West and Goslar to the South, tap water and water from private wells with access to shallow ground water (0-3 m) were sampled in the first week of May 2005 (table 1). More than 98 % of the tap water in the region is derived from freshwater reservoirs in the Harz mountains (Harzwasserwerke, 2005). The private wells usually only supply water for watering of gardens and is only in a few cases used for human consumption. The different well waters have been investigated to give an idea on the quality of the water supply in a restricted area in case the supply from water works fails. In the text the sample set in table 1 is referred to as the "P38" survey. Information on the preferred mineral waters consumed in the area were collected by means of a telephone poll among 20 major retailers in the region.

Data for bottled mineral waters derive from a worldwide survey conducted by the Institute of Plant Nutrition and Soil Science of the Federal Agricultural Research Centre in Braunschweig, Germany including 485 different brands, of which 384 are from sources in Europe (EU of 25) of which 241 are from sources in Germany. The samples were bought from 2000 to 2005 in regular stores. The individual data are available under http://mineralwaters.org.

2.2 Determination of uranium

In this study U was analysed directly by means of inductively coupled quadropol plasma mass spectrometry (ICP-QMS) of ^{238}U (Lamas et al., 2002; Taylor, 2001) employing a VG-Elemental Plasmaquad 4 instrument.

The theoretical lower limit of detection for ^{238}U by ICP-QMS is 2 ng l^{-1} U (El-Himri et al., 2000), but practically the technical lower limit of detection (LLD) was found to be 15 ng l^{-1} U by Sparovek et al. (2001), which fits well with the 13 ng l^{-1} U reported by UNEP (2001). Accordingly the limit of quantitation accounted for 25 ng l^{-1} U.

The composition of mineral waters is considered to be site-specific and thus to be long-term stable (Dopychai, 2000; Stevenson, 2000), which again warrants the validity of analytical data also for subsequent samples. Sparovek et al. (2001) already showed a very good agreement between samples analysed for U by ICP-QMS and

Table 1:
Geographic coordinates for the origins of tap and well waters investigated in the P38 survey

	Geographical position (number of samples)		
Groundwater wells	10°14'E, 52°14'N (1);	10°05'E, 52°16'N (1);	10°33'E, 52°15'N (1)
	10°35'E, 52°16'N (1);	10°32'E, 52°15'N (2);	10°29'E, 52°18'N (6)
	10°28'E, 52°17'N (6);	10°30'E, 52°15'N (1);	10°28'E, 52°15'N (1)
	10°28'E, 52°13'N (2);	10°32'E, 52°13'N (1);	10°35'E, 52°14'N (3)
	10°48'E, 52°17'N (1);	10°22'E, 52°14'N (4);	10°41'E, 52°20'N (1)
	10°22'E, 52°20'N (5);	10°26'E, 52°10'N (1);	10°23'E, 52°03'N (3)
	10°18'E, 52°12'N (1);	10°13'E, 52°08'N (1);	10°19'E, 52°03'N (1)
	10°52'E, 52°06'N (1);	10°23'E, 52°31'N (2);	10°17'E, 52°00'N (1)
Tap water	10°32'E, 52°15'N (1);	10°32'E, 52°16'N (3);	10°34'E, 52°17'N (1)
	10°28'E, 52°18'N (2);	10°28'E, 52°16'N (8);	10°30'E, 52°15'N (6)
	10°28'E, 52°12'N (1);	10°31'E, 52°12'N (2);	10°34'E, 52°14'N (1)
	10°22'E, 52°14'N (2);	10°40'E, 52°16'N (1);	10°47'E, 52°06'N (1)
	10°38'E, 52°12'N (1);	10°22'E, 52°20'N (3);	10°36'E, 52°08'N (1)
	10°47'E, 52°25'N (1);	10°55'E, 52°21'N (1);	10°23'E, 52°31'N (1)
	10°10'E, 52°27'N (1);	10°26'E, 51°54'N (1)	

values which were determined by radiochemistry and that were published before. This finding was confirmed again by data from an analytical quality control scheme conducted by the authors' laboratory (Fig. 1).

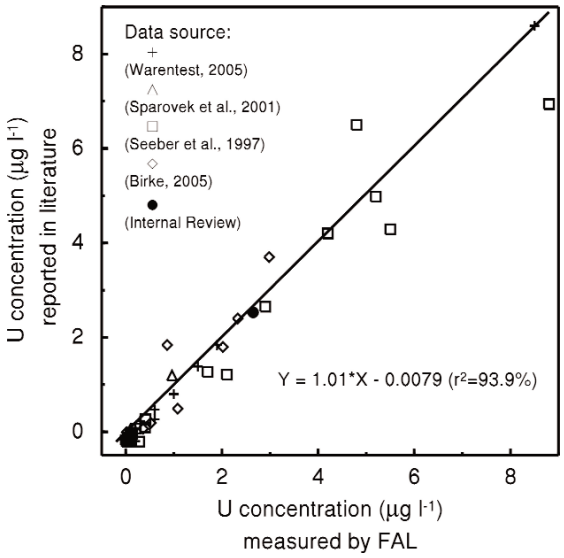

Fig. 1:
Re-measurement of uranium in mineral water samples of different origin

The quality of U measurements needs to be considered carefully when comparing data from older studies before the precise ICP-QMS equipment, which has a distinctly lower limit of detection than previously employed devices, became widely available. Thus, older data tend to have a characteristic trend towards too low U concentrations (e.g. Nozaki et al., 1970).

Statistical analyses were conducted employing the SPSS 10.0 statistical package.

3 Results and Discussion

3.1 Uranium concentrations in drinking waters available in the P38 area

Table 2 gives an overview of U concentrations in drinking waters available to the population in the P38 area. Surprisingly the "German" and "European" subsets of the world bottled water data had all nearly the same frequency distribution of U concentrations (table 1). Though the mean U concentration in the bottled waters preferred by the consumers in the P38 area was lower than those available in Germany, Europe and worldwide, a tendency towards higher U concentrations was observed. So, the median value for waters selected by the P38 population was between 3-4 times higher than in the larger sample sets (table 2).

In comparison to the bottled waters, the waters from wells and taps in the P38 area showed significantly lower U concentrations (table 2). While 75 % of the tap waters had U concentrations lower than the detection limit of ICP-QMS, only 10 % of the bottled waters met this concentration range.

The reason that shallow ground and surface waters have most times a lower U content than mineral waters is related to the fact that these reservoirs are fed predominately by rain (Gellermann and Stolz, 1997). In deeper zones of the earth's crust heavy minerals containing radioactive nuclides are quite frequent. About 100 mineral species,

Table 2:
Mean, median (µg l^{-1} U) and percentiles (%) for uranium concentrations in different sets of drinking water samples

		P38-survey wellwaters	P38-survey tapwaters	P38-survey favourite bottled waters*	German bottled waters**	European bottled waters**	World bottled waters**
n		48	39	17	241	384	485
Mean		0.87	0.18	1.84	3.34	3.45	3.17
Median		< LLD	< LLD	0.44	0.11	0.18	0.17
Minimum		< LLD	< LLD	< LLD	< LLD	< LLD	< LLD
Maximum		8.95	1.44	10.6	474	474	474
Percentiles	10	< LLD	< LLD	< LLD	< LLD	< LLD	< LLD
	20	< LLD	< LLD	0.08	< LLD	< LLD	< LLD
	25	< LLD	< LLD	0.15	< LLD	< LLD	0.01
	30	< LLD	< LLD	0.18	0.01	0.01	0.02
	40	< LLD	< LLD	0.29	0.05	0.05	0.06
	50	< LLD	< LLD	0.44	0.11	0.18	0.17
	60	0.54	< LLD	0.64	0.26	0.47	0.43
	70	0.68	< LLD	1.65	0.72	0.86	0.82
	75	0.70	< LLD	2.27	1.29	1.18	1.15
	80	1.12	0.06	3.77	2.00	1.96	1.83
	90	2.32	0.95	7.63	5.64	5.22	4.47

Comments: LLD (15 ng l^{-1} U); * see table 6; ** U concentrations assembled in the FAL-PB database are available at http://www.mineralwaters.org

especially in granite rocks, contain ≥ 1 % U. Thus water from lower depths is more likely to have elevated U concentrations than surface and shallow groundwater (Casas et al., 1998; Drever, 1998; Harmsen and Haan, 1980; Higgo et al., 1989; Hodge et al., 1973; Hydroisotop, 2001).

When ascending to the earth's surface, water from extreme depths looses radioactivity due to a process described by Yoshida et al. (1994). In a simplified way this process involves a mechanism which also happens in chromatography, where in a stream flowing through a column heavy compounds stay behind lighter ones. However, increasing the flow through a column, which can be compared to forced pumping of mineral water in order to increase the yield of a source, weakens the separation process and the amount of heavier radio-nuclides transported to the surface consequently increases.

3.2 Contribution of drinking waters to the daily uranium intake of humans

Under normal conditions, the ingestion of U by humans occurs only through foods and drinks. Significant ingestion of U through inhalation occurs only if an individual is exposed to U containing dusts mainly in U mining, processing of U ores, and during or after military operations employing depleted uranium (DU) ammunitions (Brand and Schnug, 2005). Under non-exposed conditions the daily intake of U from air would be about 1 ng (WHO, 2004). In table 3, U concentrations in basic food components were assembled from various sources in literature (Rivas, 2005). Solid food has several magnitudes higher U concentrations than air (table 3). Lowest U concentrations were found in seeds, leaves and fruits, while approximately three times higher U contents were found in meat. For meat the ranking poultry < pork < beef reflects the higher U accumulation because of a longer lifespan. The highest concentrations of U occur in offal and shellfish.

Table 3:
Mean uranium concentrations in foods and drinks (µg kg^{-1} U fresh weight, extremes in brackets, data compiled from various sources listed in Rivas (2005))

Product	U concentration (µg kg^{-1})
Cereals & cereal products:	0.4 - 6 (15)
Vegetables:	
- leaves	0.5 - 5 (60)
- roots	0.9 - 8 (69)
Seeds	0.2 - 4
Milk & milk products:	0.1 - 4
- Eggs	0.2 - 10
- Fruits	0.7 - 2*
- Meat	0.1 - 20**
- Offal	20 - 70*
Fish:	0.4 - 1 (11)
- Shellfish	9 - 31
Beverages:	
- fruit juices	0.04 - 0.1
- tea & coffee	0.02
- tap water	<0.015 - 20
- bottled (mineral) waters	<0.015 - 30 (1925)

Comments:
* muscle, liver, kidney = 14, 26, 70
** poultry < pork < beef
*** infusions with 5g l^{-1} dry matter

Table 4:
Contribution of different foods to the daily uranium uptake of humans – a simplified alimentation plan

	U concentration µg kg^{-1}	U uptake µg day^{-1}
100g bread	3.5	0.35
200g meat	10	2.0
300g vegetables	4	1.2
1 l coffee & tea *	0.02	0.02
300g fruits	1	0.3
Total (from solid aliments):		3.87 µg day^{-1} U
- additionally (from 2 l waters (Heseker, 2005))		0 - 40 µg day^{-1} U

Comments: * prepared from drinking water with U concentration < 15 ng l^{-1} U (LLD)

Based on the data in table 3, values given in table 4 present the daily ingestion of U through a simplified alimentation which was calculated by a plain diet scheme for human nutrition. The total amount of U ingested with solid food is in good accordance with values given in literature (Pais and Benton Jones jr., 1997; WHO, 2004). Tables 3 and 4 reveal also that even basic changes in alimentation habits, such as becoming a vegetarian, has no big influence on the daily U intake of an individual as long as solid food as a source of U is concerned. Far stronger influence on the daily U intake has the U concentration in the source of drinking water (table 3). With U concentrations in non-detectable ranges up to ≥ 20 µg l^{-1}, waters are the most significant factor for the amount of U taken up by an individual. Assuming a daily water consumption of 2 l (40 ml kg^{-1} body mass; Heseker, 2005), the intake by U through waters can exceed the intake through solid foods in extreme cases by factor 10.

Table 5 shows this model applied to the P38 survey and reveals that a preference of tap waters over bottled waters would distinctly reduce the risk of higher exposures to U. In addition, table 6 displays the U concentrations in the most popular bottled waters in the P38 area. The range of concentrations in these brands exceeds with a factor of nearly 1000 by far the ones of the tap waters in the same region. Depending on which brand of a bottled mineral water is consumed by an individual, the contribution of this source of U to the daily U intake varies between <1 % up to 84 %. Tables 4 and 5 reveal clearly that selecting the

Table 5:
Percentiles for the contribution of drinking water from different sets of sources to the daily uranium intake of a human (%) assuming a basic daily intake by solid food of 3.87 µg day^{-1} U and a water consumption of 2 l day^{-1} *

Percentiles	P38-wellwaters	P38-tapwaters	P38-favourite bottled waters	German bottled waters	European bottled waters	World bottled waters
10	0	0	0	0	0	0
20	0	0	2	0	0	0
25	0	0	4	0	0	<1
30	0	0	4	<1	<1	1
40	0	0	7	1	1	2
50	0	0	10	3	4	4
60	12	0	14	6	11	10
70	15	0	30	16	18	18
75	15	0	37	25	23	23
80	22	2	49	34	34	32
90	37	20	66	59	57	54

Comments: * Heseker (2005)

source of drinking water is the most efficient means to minimise the U intake of an individual.

3.3 Relationship between uranium concentration and radioactivity of drinking waters

The main source for radioactivity in drinking waters are ^{226}Ra and ^{222}Rn (Akerblom, 1994; Banks et al., 1995). Natural U is a mixture of three isotopes ^{238}U, ^{235}U, and ^{234}U in the weight proportions 99.27 %, 0.72 % and 0.006 %, respectively. Because ^{226}Ra and ^{222}Rn are members of the ^{238}U decay series it would be expected to find a straightforward positive correlation between the radioactivity and the U content of waters. In Fig. 2 the U concentrations measured in 166 mineral waters of mainly German origin were plotted against radioactivity expressed as dose in micro Sievert (µSv) per year for 0 - 1 year old nurslings. The data originate from BfS (2002). BfS calculated the radioactive dose from the activity concentrations of ^{226}Ra, ^{228}Ra, ^{234}U, ^{235}U, ^{238}U, ^{210}Po, ^{210}Pb and ^{227}Ac and for a consumption of 170 l drinking water per year. However, the scatterplot of U content against radioactivity reveals no significant relationship between both parameters (Fig. 2). This confirms the findings in a much smaller sample set by Sparovek et al. (2001). Banks et al. (1995) concluded from the weak or missing correlation between the radio-nuclides that "hydrodynamic factors, complexing, pH and redox conditions and solution recoil phenomena are the major controlling factors for radio-element concentrations, often masking the effect of mere radio-element concentrations in the bedrock".

Table 6:
Uranium concentrations in the most favourite bottled waters consumed by the population in the P38 area, and their estimated contribution to the daily U intake of a human assuming a basic daily intake by solid food of 3.87 µg d^{-1} U and a drinking water consumption of 2 l day^{-1} *

Product	U concentration (µg l^{-1})	Contribution of U in mineral water to the daily U intake (%)
Graf Rudolf, St. Willehad, Vilsa	< LLD	<1
Bad Harzburger, Bad Pyrmonter	0.13	6
Apollinaris, Harzer Grauhof	0.18	9
Regensteiner, Rhönsprudel	0.30	13
Frische Brise, Volvic	0.47	20
Vittel	0.68	26
Gerolsteiner	1.90	50
Extaler	2.65	58
Fonte Randa (Mühlenquelle)	5.44	74
Leislinger	6.95	79
Saskia	10.4	84

* Heseker (2005); Uranium concentrations of more bottled water brands are available from Lindemann (2005 a), Seeber et al. (1997), Stellpflug (2005), Warentest (2005) or http://www.mineralwaters.org

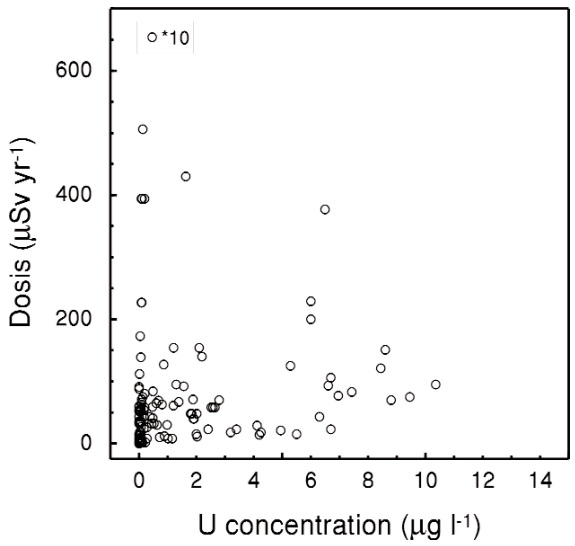

Fig. 2:
Relationship between uranium concentration and radioactivity of mineral waters

This result has strong implications for the evaluation of the quality of waters. Waters with a high radioactivity may only contain low concentrations of U and vice versa, waters with a low radioactivity may still contain higher amounts of U. The importance of this issue is adressed by the fact that German law permits expressively to advertise waters with a dose lower than 100 µSv yr^{-1} as "suitable for the preparation of baby food" (BfS, 2002). Fig. 2 reveals that even waters with the highest U concentrations can easily meet this treshold. On the other hand waters with only low-medium U concentrations, like in fig. 2 the Portugese brand "Pedras Salgas" with 0.46 µg kg^{-1} U can still show high readings for radioactivity (6539 µSv yr^{-1}; BfS, 2002).

3.4 Critical and guideline values for uranium in drinking waters

Consumers reserve the right to be protected from hazardous compounds in foods and drinks. U is such a hazardous compound with the feature that U is in all foods and all drinks in certain amounts and that U has two modes of damaging, by its radioactivity and by its chemical toxicity. The question arises as to which amounts can be safely tolerated and under what circumstances. From scientific point of view, and especially in the case of U, hazards depend very much on the circumstantial parameters assumed and never consider interactions between individual susceptibilities (e.g. diseases, low immunity, genetic conditions) or other hazardous substances and influences on the organism in question.

Doses and risks from internal radiation are still burdened with many uncertainties (Fairlie, 2005). Obviously considering Gofman´s (1996) statement that there is no safe dose for radioactivity, EPA has set a goal of no U in drinking water (ATSDR, 1999) and "calls this the "Maximum Contaminant Level Goal (MCLG)", but recognizes that, currently, there is no practical way to meet this goal. Because of this, EPA proposed in 1991 to allow up to 20 µg of U per liter (20 µg l^{-1}) in drinking water, and states began to develop regulations to achieve this level. EPA calls this the Maximum Contaminant Level (MCL). The MCL for U is based on the calculation that if 150,000 people drink water that contains 20 µg l^{-1} of U for a lifetime, there is a chance that one of them may develop cancer from the U in the drinking water. Important to mention here that low probabilities are subliminally connected to large time scales. But it is part of nature that there is also an, admittedly faint, chance that just one U atom spontaneously causes cancer in an organism and in this case the risk is always 100 % for the victim of circumstances!

Additionally, the EPA statement shows clearly a much more serious dilemma than just the lack of a scientific background: critical as well as guideline values are the product of the trade-off between the consumer's claim for safety and the interest of suppliers of drinking water in making profits. With often only a faint scientific background left, critical values suggest consumers that products passing the value are safe, which is in case of U not justified. Based on the same scientific reports, just by recalculations, the guideline value for U in drinking water raised within only six years from 2 to 15 µg l^{-1} U (table 7) which has a very significant effect on the percentage of brands and suppliers remaining as "acceptable" in the market. Pretty perfect world ...!

Based on an analysis of an undisclosed data set investigated by the German Federal Institute for Risk Assessment (BfR, 2005) the German Ministry for Consumer Protection, Food and Agriculture (BMVEL, 2005) stated in a recent press release that "less than 3 % of the mineral waters available in Germany had a U content of lower than the guideline value of 15 µg published by the WHO (2004)".

Without further information about the structure of the data set (time of sampling and analysis, number of brands, frequency distribution of different brands, analytical methods employed) such a statement is of no value and feigns a level of safety which is not justified. Worried about possible hazards the BfR claimed already "U free" waters for nurslings (BfR, 2005). Apart from the fact that the U concentration of a water can only be lower than the technical detection limit of contemporary state of the art analytical techniques and never zero, the claim does also not specify the target group in question: according to ICH guidelines there are "newborn infants (0 to 27 days old) and infants and toddlers (28 days to 23 months old)" (ICH, 2000). Shortly after BfR defined "free of U" as con-

Table 7:
Percentage of bottled mineral water brands (%) meeting different critical values for uranium

Critical value (µg l^{-1} U)		factor	Data source (mineral waters)			
			World (n = 485)	Europe (n = 384)	German (n = 240)	P38 (n = 17)
< technical LLD ICP-QMS	(0.015)	1	30	33	35	18
< BfR (2005), LLD	(0.200)	13	46	51	58	41
< WHO (1998)	(2.000)	133	81	80	80	76
< UBA* (2005)	(10.000)	667	96	96	98	94
< WHO (2004)	(15.000)	1000	98	98	99	95

* Konietzka et al. (2005)

centrations < 0.2 µg l^{-1} U, which is 13 times more than the technically LLD. The discussion becomes preposterous with the release of a guideline value of 10 µg l^{-1} U by the same authors who pleaded just 5 years earlier, and still based on the unchanged facts supplied by the WHO (1996 & 2004), for 1 µg l^{-1} (Dieter, 2000; Konietzka et al., 2005). Striking result of this metamorphosis: all brands on the market hit the guideline value (table 7). In this context Reiman and Banks wrote in 2004 a paper with the very comprehensive title: "Setting action levels for drinking water: Are we protecting our health or our economy (or our backs!)?".

4 Conclusions

The content of radio-nuclides and radioactivity are a characteristic features of drinking waters. As the significance of natural mineral waters has developed from an occasionally administered remedy (Albu et al., 1997) towards a common thirst quencher replacing tap water as "the better drinking water" (Misund et al., 1999) its impact on the intake of U and radioactivity increased, too. Although there seems to be no hard evidence for immediate health risks, the coincidence of the increasing consumption of mineral waters (VDM, 2005) with increasing numbers of kidney cancers (Fischer, 2005) attracts attention. Based on available facts, a causal relation to an increased U intake has to be taken into consideration. There is definitively more evidence for an approval of this hypothesis than the proverbial "stork delivers babies" theory (Höfer et al., 2004).

No matter if someone favours the idea that radioactivity is a threat or a benefit for life, in both cases information on the concentrations of radio-nuclides and radioactivity are essential for the consumer and should be provided with consumer information and become part of the "Codex Norms for Mineral Water" (CCA, 1983).

As source and quality of drinking waters are easy to maintain parameters in the daily diet, selecting waters according to their U concentration is a highly efficient means to keep the U ingestion at an unavoidable level supplied together with solid foods and thus to keep the risk of hazardous health effects of U at minimum.

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