



## Occurrence of uranium in Swiss drinking water

E. Stalder, A. Blanc, M. Haldimann\*, V. Dudler

Federal Office of Public Health (FOPH), Food Safety Division, Chemical Risks Section, CH-3003 Bern, Switzerland

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### ABSTRACT

The results of a nationwide survey of uranium in Swiss drinking water are reported. Elevated concentrations of uranium in groundwater are found mainly in the alpine regions and can be traced back to the geology of the bedrock. Water sources were systematically surveyed and analysed for the presence of Li, B, Si, Sc, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Sr, Cd, Sn, Sb, Ba, Tl, Pb and U and the results were analysed to determine if any correlation with uranium concentration was apparent. No correlation was found. The results are interpreted in relation to the current WHO guideline and those of other countries with a view to determining which areas would be affected if a maximum value were to be adopted and which areas require further investigation. Uranium content varied considerably, from below the limit of detection to almost  $100 \mu\text{g L}^{-1}$ . Of the 5548 data samples, 98% are below the 2004 WHO provisional guideline value of  $15 \mu\text{g L}^{-1}$  and 99.7% below the revised (2011) value of  $30 \mu\text{g L}^{-1}$ .

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### 1. Introduction

Uranium has been used since ancient times as a colourant in ceramic glazes and, later, in the glassmaking industry. Modern day usage of uranium centres on the nuclear industry where enriched uranium ( $\sim 3\%$   $^{235}\text{U}$ ) fuels nuclear power plants and highly enriched (between 20% and 90%  $^{235}\text{U}$ ) is used in naval reactors and nuclear weapons. Depleted uranium (the by-product of the enrichment process), with about 40–60% the radioactivity of uranium (US Department of Defense, 2000), is used in military branches for both armour plating and armour piercing ammunition. Such applications are somewhat controversial owing to the probability of environmental contamination and subsequent health implications (Craft et al., 2004). Other uses include as a shielding material for radiation protection and as counterweights in aircraft. On a much smaller scale, it has applications in catalysis and uranium salts are used as negative stains for biological imaging.

Uranium is a ubiquitous element, one of the densest metals and the second heaviest naturally occurring element, being present in the Earth's crust with an average abundance of 1.8 ppm (Mason and Moore, 1982). It occurs at higher concentrations in granites, shales and carbonates (Turekian and Wedepohl, 1961). Source minerals are uraninite ( $\text{UO}_2$ ), pitchblende ( $\text{U}_3\text{O}_8$ ), carnotite ( $\text{K}_2(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 3\text{H}_2\text{O}$ ), also phosphate minerals and lignite. There are three naturally occurring isotopes:  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{234}\text{U}$  all of which are radionuclides and decay via alpha emission. However, uranium has a low specific activity and, as such, its chemical toxicity

is of greater concern than its radiotoxicity and will therefore be the focus of this report (Health Canada, 1987; Kurttio et al., 2002; ATSDR, 2009; EFSA, 2009).

#### 1.1. Environmental distribution

Groundwater is in contact with the aquifer rock and so depending on the mineralogical and geological structure of this rock, the chemistry of the water and the physical conditions, the water will become mineralised to various extents. The dominant species present in primary minerals is the U(IV) state (Smedley et al., 2006), associated with hydroxides, phosphates and fluorides (Keith et al., 2007) and is relatively insoluble. U(IV) is, however, readily oxidised to U(VI) and since surface water and shallow groundwater are both oxidising media, it becomes readily soluble. It is found usually as complexes of the uranyl cation,  $\text{UO}_2^{2+}$ , and is known to exist in groundwater at much higher concentrations than is expected from the underlying rocks (up to an enrichment factor of 10) (Dall'Aglio, 1971). The uranyl cation tends to build complexes with carbonate, phosphate and sulphate anions (Murphy and Shock, 1999). Leaching studies have shown that uranium passes readily from uraninite in pegmatites (Dall'Aglio, 1971) and pitchblende in granites (Labhart and Rybach, 1974) to the water and that the primary factors determining the concentration of uranium in natural waters are the oxidation potential and the partial pressure of carbon dioxide (Osmond and Cowart, 1976). Just as it can be dissolved from the surrounding rocks, uranium can precipitate from the water if it enters a reducing environment and it can be scavenged from solution by complexation with humic acids (Dall'Aglio, 1971; Wanty and Nordstrom, 1993; Jacobs and Smalling, 2005).

\* Corresponding author. Tel.: +41 31 32 29558.

E-mail address: [max.haldimann@bag.admin.ch](mailto:max.haldimann@bag.admin.ch) (M. Haldimann).

Uranium can be redistributed throughout the environment via anthropogenic activities. The mining process is, in the UK, probably the most significant source of anthropogenic remobilised uranium (Smedley et al., 2006). Tailings from mines have been shown to contaminate both surface water and groundwater at sites in the USA (Parkhurst et al., 1984; Goode and Wilder, 1987). The phosphate industry, both the processing of phosphate minerals (Boothe, 1977) and the use of phosphate fertilizers, is potentially a serious threat to the environment and water quality (Smedley et al., 2006; Birke et al., 2010). Release of particles into the atmosphere can occur via natural means, e.g. the observed increase of atmospheric uranium after the eruption of Mount St. Helens (Essien et al., 1985), or through man-made emissions such as the burning of coal although generally at such levels that are unlikely to be hazardous to the environment (Van Hook, 1979).

Traces of uranium are found in all foodstuffs and water. Whilst evidence from studies of uranium uptake in plants is conflicting (Ribera et al., 1996; ATSDR, 2009), it is present in the soil in varying concentrations and, although absorption is dependent on plant species and speciation, it is known to adsorb onto the roots of plants thus unwashed root vegetables are a dietary source (ATSDR, 2009).

### 1.2. Toxicology

Human exposure occurs via ingestion of food and water and inhalation. Exposure due to inhalation is considered insignificant ( $0.0015 \mu\text{g d}^{-1}$  except in the case of occupational exposure) and uranium intake from food and water is considered to be equal in the range of  $0.9\text{--}1.5 \mu\text{g d}^{-1}$  (ATSDR, 2009; EFSA, 2009). The WHO reckons with a total uranium intake via food of  $1\text{--}4 \mu\text{g person}^{-1} \text{d}^{-1}$ , a value supported by studies in Switzerland where an average consumption of  $3.7 \mu\text{g person}^{-1} \text{d}^{-1}$  (excluding drinks) was measured. A second study showed an average of  $1.1 \mu\text{g meal}^{-1}$  (FOPH, unpublished results).

Toxicity and bioavailability of uranium is determined by speciation, governing the mechanism by which it accumulates, is transported and transferred through the body. The most stable species in aqueous acidic solution is the uranyl ion and, in body fluids, is usually complexed with bicarbonate or citrate anions or plasma proteins (Keith et al., 2007). Uranium has been shown to bind to both the iron transport protein, transferrin, and to haemoglobin or red blood cells which are rich in bicarbonate (Ansolobro et al., 2006).

Human gastrointestinal absorption varies between 0.1% and 6% depending on speciation and the remainder is excreted in the faeces after a few days (Zamora et al., 1998; Kommission Human-Biomonitoring des UBA, 2005; Keith et al., 2007). Clearance of uranium in the blood is achieved (99%) within 24 h; 67% is excreted via urine and the remainder dispersed in the tissues: 22% to the skeleton and 21% to the kidneys (Ribera et al., 1996; ATSDR, 2009).

The main organ of concern is the kidney and most studies have concentrated on nephrotoxicity. Toxicological data has been obtained through both acute, subacute, chronic and subchronic exposure of various species to uranium. All have shown effects on the kidney, in particular damage to the proximal tubules and, at high doses, also to the glomerulus. Although the damaged cells regenerate if exposure is then limited, there seems to be slight differences in morphology with uncertain effect (Health Canada, 1987; COT, 2006; EFSA, 2009).

### 1.3. Epidemiological studies

Various studies have been carried out in places where there is a naturally high level of uranium found in drinking water, often in

cases where families are supplied by private wells rather than the public distribution system.

A study in Nova Scotia, Canada with a maximum exposure of  $700 \mu\text{g L}^{-1}$  showed no relationship between kidney disease or any other symptomatic complaint and uranium content. Indications of tubular defects were present. Those with the highest concentrations of uranium in their water did not follow the trend – by the time the tests were carried out, people in this group had already ceased drinking well water indicating that such defects could be reversible (Giddings, 2005). Another study carried out in Nova Scotia, Canada, (population 50,  $[\text{U}] < 1\text{--}781 \mu\text{g L}^{-1}$ ) showed a statistically significant correlation between glucose excretion and uranium concentration in water for both males and females (Zamora et al., 1998). The authors concluded that there were signs of interference with the proximal tubule but made no judgement on whether or not this would lead to progressive or irreversible renal injury.

A study in southern Finland (population 325,  $0.001 < [\text{U}] > 1920 \mu\text{g L}^{-1}$ ) showed an association between uranium exposure and both increased fractional excretion of calcium (Kurttio et al., 2002) and tubular function. It was suggested that short-term exposure is most relevant for kidney injury and effects are unlikely to be cumulative. Despite these alterations in kidney function, there was no evidence of a risk of development of kidney disease. The authors noted that increased leakage of calcium into urine could increase susceptibility to osteoporosis and concluded that the safe level of uranium in drinking water is probably around the guideline values proposed by the WHO and US EPA.

A 2009 study of 398 subjects in Sweden showed strong correlation between urine uranium levels and well levels. There was however, no clear sign that uranium in drinking water was nephrotoxic at the levels measured ( $< 0.2\text{--}470 \mu\text{g L}^{-1}$ ), but there were signs of nephrotoxicity when urine uranium levels were used as a marker of exposure. The authors noted that the clinical relevance of their findings is unclear (Seldén et al., 2009).

### 1.4. Current guidelines

Overall uranium ingestion is considered low. However, if there is an increased concentration present in drinking water, then ingestion via water consumption becomes the main source of uranium and so the World Health Organisation allocates 80% of the tolerable daily intake (TDI) to water. The 2004 guidelines cite a TDI of  $0.6 \mu\text{g kg}^{-1}$  of body weight (b.w.)  $\text{d}^{-1}$  (based on a subchronic animal study), for a 60 kg adult drinking 2 L of water a day, giving a provisional guideline value of  $15 \mu\text{g L}^{-1}$  (WHO, 2004). Owing to an increasing database of results from human studies, a new provisional guideline value of  $30 \mu\text{g L}^{-1}$  has been published, based on a TDI of  $1 \mu\text{g kg}^{-1}$  b.w.  $\text{d}^{-1}$ , citing studies in Finland and Sweden (WHO, 2011).

Using the same TDI as the 2004 guidelines, but taking the average weight of an adult to be 70 kg, drinking 1.5 L of water a day and assigning 35% of the daily uranium intake to drinking water, Health Canada determines a health-based guideline value of  $10 \mu\text{g L}^{-1}$  and an interim maximum acceptable concentration (IMAC) of  $20 \mu\text{g L}^{-1}$  (Health Canada, 1987).

The US Environmental Protection Agency (EPA) publishes a maximum contaminant level (MCL) of  $30 \mu\text{g L}^{-1}$  and states that their maximum contaminant level goal for uranium is zero (EPA, 2011).

There is currently no EU legislation concerning uranium in drinking water (European Council, 2009). A report has been published by EFSA endorsing the TDI of  $0.6 \mu\text{g kg}^{-1}$  b.w.  $\text{d}^{-1}$  as described by the WHO but which notes that infants fed solely on infant formula may in some instances exceed this value (EFSA, 2009).

Germany has recently introduced a limit of  $10 \mu\text{g L}^{-1}$  (Bundesministerium für Gesundheit, 2011). Taking the 2004 WHO TDI and applying a safety factor of 3, they have derived a maximum permissible concentration of  $2 \mu\text{g L}^{-1}$  (Konietzka et al., 2005) for water permitted to be advertised as suitable for use in the preparation of infant formula (Bundesgesetz, 2006) and state that there is no increased risk for children drinking water with concentrations up to  $10 \mu\text{g L}^{-1}$  (BfR, 2009).

In the UK, there is no upper limit specified, neither for tap nor bottled water. Current advice from the Food Standards Agency is to avoid using natural mineral water to prepare infant food. Following a report by the Committee on Toxicity, legislation is in preparation to amend the Natural Mineral Water, Spring Water and Bottled Drinking Water Regulations to permit such waters to claim suitability for this use provided that they meet specific criteria (COT, 2006).

Uranium is not specified in the list of maximum permissible values of metals and metalloids in the Swiss Ordinance on Components and Contaminants in Foodstuffs. It is however specified in the list of maximum permissible concentrations of radionuclides that a maximum of  $10 \text{Bq kg}^{-1}$  is allowed in liquid foodstuffs (EDI, 2010). Taking the activity of natural uranium to be  $25.4 \text{Bq mg}^{-1}$  and assuming the isotopes to be in radiological equilibrium, this corresponds to a concentration of  $395 \mu\text{g L}^{-1}$  (IAEA, 2011). With this in mind, a survey of drinking water was undertaken in order to build a picture of the uranium levels present in drinking water and aiming to be as representative as possible of the water reaching the consumer. The results of the survey are presented in this work and help identify regions of low uranium concentrations, regions of concern (those which might be affected should such a maximum level be introduced) and areas for which no data is currently available, but nevertheless which have the potential to contain uranium and should be investigated.

## 2. Experimental

### 2.1. Reagents

High purity nitric acid 65% Suprapur<sup>®</sup>, ICP Standards of  $1 \text{g L}^{-1}$  Rh–Sc–Sb–Sn–Si CertiPUR<sup>®</sup> and an ICP multi-element standard solution VI CertiPUR<sup>®</sup>  $10 \text{mg L}^{-1}$  were obtained from Merck. A stock solution of 1 ppm was prepared from the multi-element standard solution VI and the ICP standards and then further diluted to prepare a stock solution of 2 ppb used to prepare the solutions used for calibration.

### 2.2. Water sample collection

Over the period of 2003–2011, samples of water were collected from some cantons in Switzerland. Groundwater and surface water sources were sampled at the first available extraction point (well/spring source, pumping station, reservoir, water supply network). Most data for the canton of Graubünden are taken from a previous publication and were collected in 2002 (Deflorin, 2004).

Local food inspectors or official representatives of the communes collected the water samples. 50 mL polypropylene tubes were filled and subsequently acidified with 0.5 mL 65% nitric acid at the cantonal laboratory before being sent to the laboratory in Bern. The samples were not filtered since the aim of the study is to document the total elemental content of the water that reaches the consumer and so released into the body once consumed. After acidification, no turbidity was visible in any of the samples. The samples were stored at  $4^\circ\text{C}$  in darkness until they were measured.

### 2.3. Preparation of standards, references and samples

Analysis was carried out using external calibration for quantification in the range of 0.1 ppb–1 ppm. The analysis was part of a multi-element method (Li, B, Si, Sc, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Sr, Cd, Sn, Sb, Ba, Tl, Pb and U). A blank of 25 mL 1%  $\text{HNO}_3$  with 100  $\mu\text{L}$  rhodium solution (250 ppb) was used. The samples were measured after 3-fold dilution with 1.2% nitric acid and addition of 18  $\mu\text{L}$  of rhodium standard (250 ppb). Any samples showing erratic measurements of the internal Rh standard were re-measured. Blank measurements were repeated after each series of 30–50 samples to check for contamination or memory effects. If any deviation was found for the blank measurements, the samples were re-measured. The measured values of the certified reference materials were plotted on a probability plot and those not fitting the normal distribution were classified as outliers (<5%). The values of the 95% confidence interval of the measured means overlap with the 95% confidence interval of the certified values, indicating good agreement. The standards used were: Surface Water SPS-SW1-B110, Surface Water SPS-SW2-B108 and Simulated Rain Water TMRain-95. Unused sample containers were tested as a control and no uranium was observed.

### 2.4. Instrumentation

The ICP-MS instrument used was a sector field Element2 (Thermo, Bremen, Germany) equipped with a CETAC 260 autosampler. It was operated in low resolution mode. Details of the instrument and measurement parameters are given in Table 1. The instrument was tuned daily.

In this survey, simply U-238 was measured, no information was derived about other isotopes, nor about the species present. The limit of detection (LOD) was calculated from a linear calibration line (with upper and lower confidence limits) generated from 10 measurements at each concentration ( $0\text{--}1 \mu\text{g L}^{-1}$ ). The LOD is the concentration value ( $x$ ) of the upper confidence limit corresponding to the  $y$  value (ICP-MS response of uranium-238) at which the lower confidence limit intercepts the  $y$  axis and was determined to have a value of  $0.05 \mu\text{g L}^{-1}$ . In accordance with US EPA

**Table 1**  
Typical instrument and measurement settings.

<i>Instrument settings</i>	
Instrument	Element2 (Thermo)
Rf power	1.2 kW
Gas flows: plasma gas	15 L min <sup>-1</sup>
Auxiliary	0.2–0.5 L min <sup>-1</sup>
Sample gas	0.8–0.9 L min <sup>-1</sup>
Additional gas	0.2–0.4 L min <sup>-1</sup>
Injector	Sapphire
Nebuliser	Microflow ESI-PFA-ST-1045
Spray chamber	ESI-PFA
Cones	Ni
Sample uptake	2 min
Instrument tuning	<sup>115</sup> In and <sup>238</sup> U
Wash time	3 min
<i>Measurement parameters</i>	
Resolutions utilised	$m/\Delta m = 300$
Scan type	Electrical scan
No. of sample sweeps for each resolution	4
Mass range	237.80–238.15
Dwell time per acquisition point	0.01 s
No. of acquisition points per mass segment	50
Segment duration per scan per resolution	0.25 s
Magnet settle time	0.01 s
Detector mode	Pulse counting
Integration type	Average

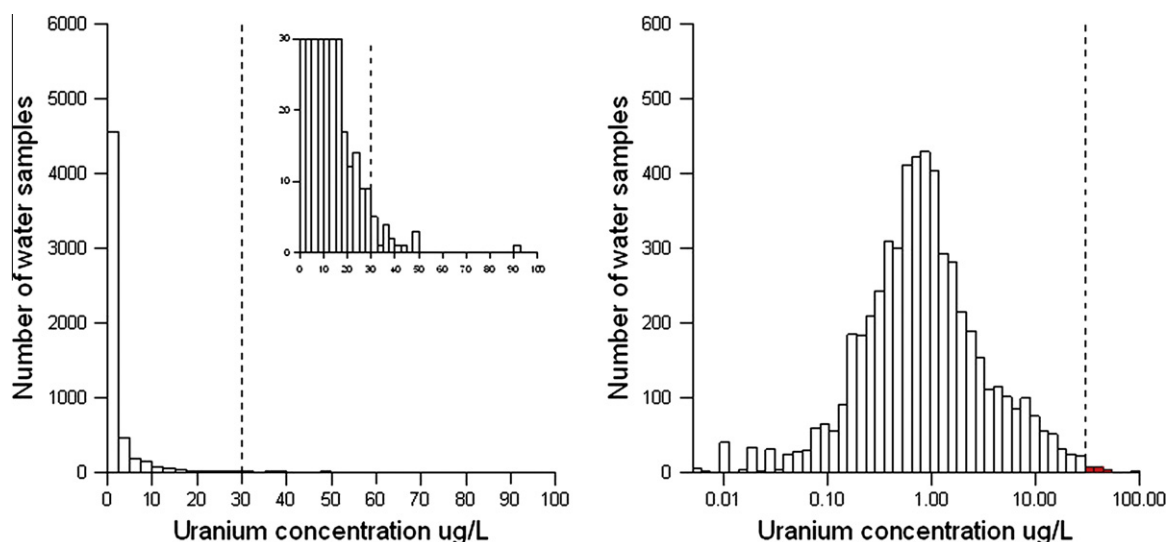


Fig. 1. Histograms showing distribution of uranium in Swiss waters. Dashed line indicates WHO guideline (WHO, 2011).

guidelines for analysing data with non-detects, any samples giving a reading lower than the LOD were assigned a concentration of  $0.025 \mu\text{g L}^{-1}$  (EPA, 2006). No interference from polyatomics is expected.

### 3. Distribution of uranium in Swiss water

Five-thousand five hundred and forty eight data points were collated and uranium concentrations in the range of  $0.05\text{--}92.02 \mu\text{g L}^{-1}$  were observed, with a median of  $0.77 \mu\text{g L}^{-1}$  and a (geometric) mean of  $0.78 \mu\text{g L}^{-1}$ . Of the samples, 2% were higher than  $15 \mu\text{g L}^{-1}$ , 0.3% higher than the WHO provisional guideline value of  $30 \mu\text{g L}^{-1}$  (see Fig. 1, Table 2). These high values are linked to the underlying geology of the region rather than any sources of pollution, the high uranium concentrations being attributable to various rock types (sandstone, conglomerate, schist, gneiss and granite) (Gilliéron, 1988). At the other end of the scale, 78% had uranium concentrations less than  $2 \mu\text{g L}^{-1}$  and 3% of the samples were below the limit of detection (LOD). The majority of samples, 92%, are from groundwater (47% direct from source and 45% from network), 1% from surface water and 7% of the samples were unassigned.<sup>1</sup> Many sources, especially in the mountainous regions, are in very remote areas and are not readily accessible for sample collection. For such sources, samples were taken from the first available point in the network.

An overview of the geographic distribution can be seen in Fig. 2 and a breakdown by canton is shown in Table 3. The graduated colouring of the communes shows the differing extent of uranium distribution. This map is generated using the average values of the samples collected within a commune and is not necessarily representative of individual water networks within the commune. Nor does it mean that all of the networks or sources within the commune are affected, as can be seen in Table 4; in those communes with the highest concentrations, samples were also measured with low concentrations. Some communes receive their water supply from neighbouring communes thus no water source was analysed originating from this commune; for example all communes in

Geneva are supplied from Lake Geneva. These are marked in light grey on the map to distinguish them from areas from which no samples were available. A few sources are in one canton but belong to and supply communes in a neighbouring canton. These are depicted on the map under the source commune but calculated statistically for the point of consumption.

Overall, there is no need to be concerned with the concentration of uranium in the vast majority Swiss drinking water, 99.7% of the communes measured have average values below the WHO guideline value. It should be noted that the samples were for the most part collected only once (areas showing a very high uranium content were later re-sampled, the values of these samples were then averaged and the mean used as the data point for the overall analysis) and so these results provide a snapshot of uranium concentration at a particular time. It is possible that due to variations in precipitation, snow melt or other such factors that uranium concentration in water could vary over time. Such factors need to be taken into account since toxicological effects result from long rather than short-term exposure. This study can therefore be used as a basis for long-term monitoring.

In general, hot spots are confined to the cantons of Fribourg, Valais, Graubünden and Ticino; the distribution in the latter three cantons corresponding to the distribution of terrestrial radiation and exhibiting an association with the crystalline rocks of the Alps (Rybach et al., 2002). All samples with  $[U] > 30 \mu\text{g L}^{-1}$  are from the canton of Valais in the communes of Martigny, Lax, Bitsch, Sion, Visp, St. Niklaus and Zermatt. Samples with  $15 < [U] < 30 \mu\text{g L}^{-1}$  are from the cantons of Valais, Graubünden, Fribourg and Ticino (see Table 4). A single high reading from a commune does not necessarily mean a high median, nor that the average is above guideline values. The high concentrations are found in mountainous regions and are most likely linked to the underlying natural rock-water interactions. There is no evidence of, nor any reason to suspect, any pollution from industrial activity in these areas.

No correlation of uranium with other elements, especially heavy metals, was found (max.  $r_s = 0.39$  for As), a phenomenon noted in previous studies and of studies in other countries (Pfammatter, 1999; Kurttio et al., 2002); presumably due to a combination of the geochemical behaviour of uranium and the unsystematic nature of uranium distribution, mineralisation and occurrence in specific lithologies (Dinelli et al., 2010).

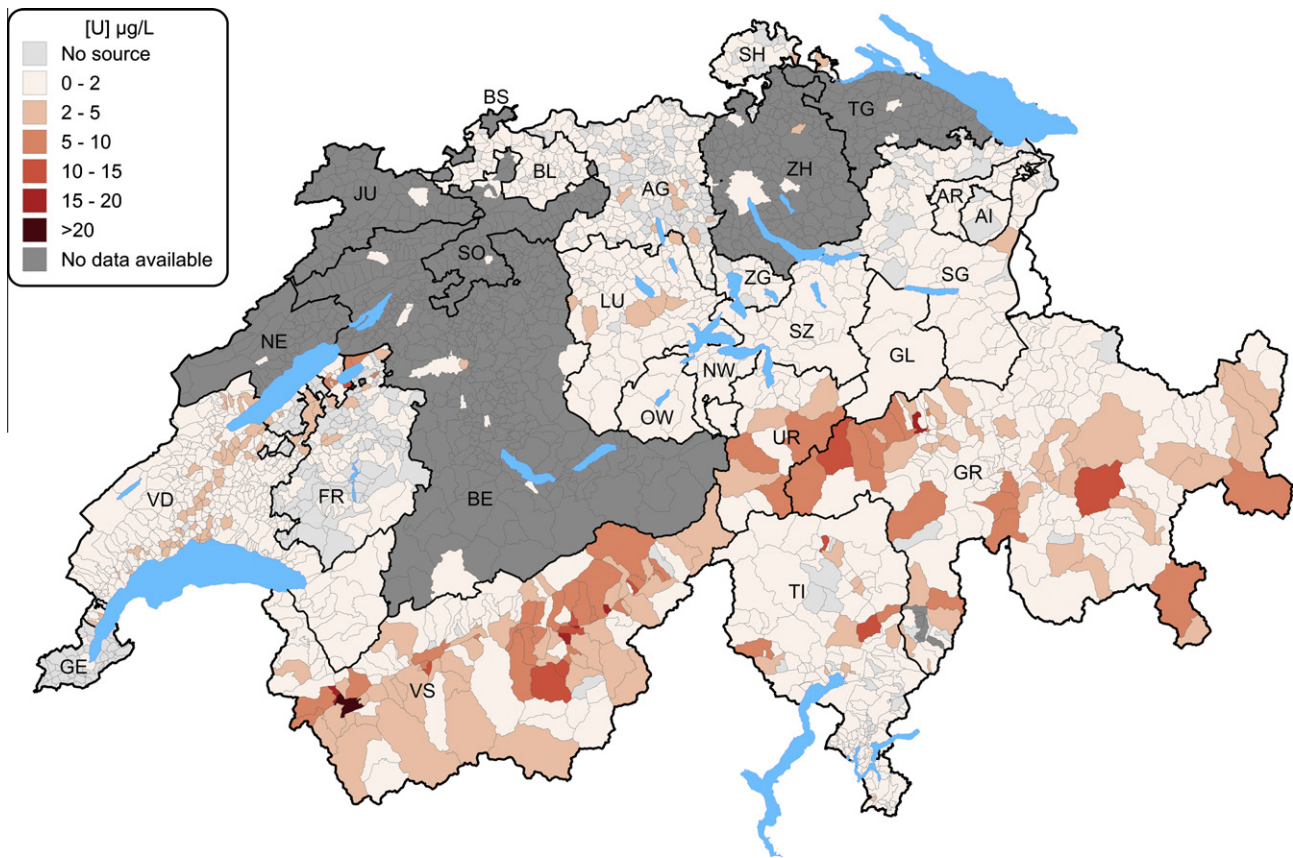
<sup>1</sup> Swiss drinking water is sourced as follows: 40% from springs, 40% from wells and 20% from surface water. Of this, 38% goes directly into the drinking water network with no treatment, 33% undergoes a single-step sterilisation process and the rest (including all surface water) a multi-step treatment process (SVGW/SSIGE, 2008).

**Table 2**Statistical summary of uranium data for water samples from this study and literature; *n* = number of samples, *N* = total number of samples.

	Cantons of GR, TI, VS [U] ( $\mu\text{g L}^{-1}$ )		All cantons excluding GR, TI, VS [U] ( $\mu\text{g L}^{-1}$ )		All cantons [U] ( $\mu\text{g L}^{-1}$ )	
N	2539		3009		5548	
Minimum	0.025		0.025		0.025	
Maximum	92.02		27.90		92.02	
Median	1.12		0.69		0.77	
Arithmetic mean	3.27		1.00		2.04	
Geometric mean	0.96		0.66		0.78	
Standard deviation	5.72		1.36		4.16	
95th percentile	13.7		2.70		8.84	

[U] ( $\mu\text{g L}^{-1}$ )	<i>n</i>	%	<i>n</i>	%	<i>n</i>	%
<1	1199	47	2097	70	3296	59
<2	1607	63	2735	91	4342	78
<10	2325	92	2997	99.6	5322	96
>15	107	4.2	3	0.10	110	2.0
>20	59	2.3	2	0.07	61	1.1
>30	18	0.7	0	0	18	0.3

**Fig. 2.** Overview of uranium concentration in Swiss water. Majority of data for Graubünden taken from literature (Deflorin, 2004).

### 3.1. Valais (VS)

In the west, Martigny and Vernayaz lie on the Mont-Blanc massif and the Aiguilles Rouges massif respectively. Here the underlying geology is mainly granitic and gneiss rock. In this region, localised but rich deposits of uranium have been discovered (Gilliéron, 1986).

Central Valais is situated on the St. Bernard nappe and here it also known that there are localised deposits of uranium minerals, notably in the areas of St. Niklaus–Zeneggen–Turtmann and of the Great St. Bernard Pass (Gilliéron, 1986).

From north eastern Valais, through the south east of canton Bern, southern Uri to western Graubünden, disappearing at Ilanz

(and thus encompassing Andermatt, Disentis and Rueun) is the region of the Aaremassif and the Gotthard Massif. In Valais, the communes of Bitsch, Lax, Visp and Visperterminen all fall within this region. The presence of rich, but very localised uranium deposits is already documented (Gilliéron, 1986) and it is known that the granites from this area are mineralised with readily soluble uranium (Labhart and Rybach, 1974).

### 3.2. Graubünden (GR)

The geology in the region of Disentis is dominated by crystalline rocks of the Aaremassif (rhyolite, granite, metagranitoid and gneiss), Tavetscher massif (metagranitoid, granodiorite, gneiss)

**Table 3**

Statistical summary of uranium concentrations ( $\mu\text{g L}^{-1}$ ) by canton (AM: arithmetic mean, GM: geometric mean).

Canton	n	Low	High	Median	AM	GM	95th percentile
AG Aargau	310	0.025	4.54	0.90	1.04	0.87	2.40
AR Appenzell Ausserrhoden	150	0.15	2.72	0.72	0.83	0.73	1.83
AI Appenzell Innerrhoden	15	0.025	1.05	0.29	0.38	–	1.00
BL Basel-Landschaft	365	0.13	2.72	0.53	0.60	0.51	1.27
BS Basel-Stadt	–	–	–	–	–	–	–
BE Bern	17	0.025	5.42	0.38	1.02	–	5.11
FR Fribourg	201	0.08	27.90	0.84	1.76	0.91	6.23
GE Genève	6	1.02	2.21	1.46	1.61	–	2.21
GL Glarus	61	0.025	3.20	0.40	0.66	–	2.09
GR Graubünden	379	0.025	28.80	1.40	2.55	1.20	9.96
JU Jura	3	0.025	0.33	0.33	0.23	–	0.33
LU Luzern	246	0.10	6.25	0.71	0.95	0.72	2.74
NE Neuchâtel	2	0.025	0.33	0.18	0.18	–	0.33
NW Nidwalden	27	0.18	0.59	0.30	0.34	–	0.57
OW Obwalden	21	0.10	0.80	0.22	0.31	–	0.78
SH Schaffhausen	71	0.20	4.86	0.68	1.04	–	2.58
SZ Schwyz	83	0.07	1.76	0.53	0.62	0.48	1.57
SO Solothurn	4	0.22	1.10	0.49	0.57	–	1.10
SG St. Gallen	631	0.025	6.65	0.54	0.70	0.48	0.93
TG Thurgau	8	0.73	1.47	0.88	1.00	–	1.47
TI Ticino	687	0.025	26.12	0.32	0.89	0.29	3.41
UR Uri	78	0.03	14.47	2.28	3.73	–	11.99
VS Valais	1473	0.025	92.02	1.94	4.57	1.59	17.27
VD Vaud	586	0.025	14.21	0.89	1.16	0.84	2.95
ZG Zug	109	0.22	5.12	0.80	0.83	0.74	1.36
ZH Zürich	15	0.27	2.57	1.03	1.20	–	2.53

and the Gotthard massif (Swisstopo, 2005). In this region, the St. Placidus Source is noted for its red colour, a result of iron precipitate present in the water. Analysis of this precipitate shows a composition of 75.4%  $\text{Fe}_2\text{O}_3$  and 22%  $\text{SiO}_2(\text{H}_2\text{O})_x$  with 145 ppm of uranium (Bärtschi and Keil, 1992). The water itself contains a much lower concentration than this, unsurprising as uranium is known to co-precipitate with  $\text{Fe}_2\text{O}_3$  as it comes into contact with the air.

At the eastern end of the Aarmassif and Tavetscher massif is the commune of Rueun. The geological formation of this region is known as Verrucano comprising sedimentary rocks, slates, sandstones and conglomerates. It has previously been shown to contain low concentrations of uranium containing minerals of sporadic distribution (Gilliéron, 1986).

Other points of note in this canton are the neighbouring communes of Davos and Bergün. Samples with a relatively high maximum value were measured in both these communes. These communes are situated on the crystalline bedrock of the Upper Austroalpine Silvretta nappe. Rocks found in this region include rhyolite, metagranitoid, gneiss, schist and granite (Swisstopo, 2005). It is known that soil in the region the Dischma Valley shows anomalously high uranium content, supposed to originate from dissolution of the bedrock by groundwater (Regenspurg et al., 2010).

In Val Poschiavo, the crystalline Bernina nappe (predominantly ortho- and paragneiss) of the Eastern Alps meets with the Margna-Sella nappe (mainly paragneiss and slate) of the Penninic Alps (Deflorin, 2004). Elevated concentrations are also found in mineral waters sourced in the neighbouring regions of Italy (Cicchella et al., 2010; Dinelli et al., 2010).

A more detailed discussion of the underlying geology and interaction with the groundwater of these sources in Graubünden can be found in (Deflorin, 2004) but no correlation can be drawn between the underlying geology of the area and the sources which have high uranium concentrations indicating that these waters

**Table 4**

Statistical summary of uranium data for 'hotspots' with a highest measured concentration above  $15 \mu\text{g L}^{-1}$ . All concentrations are given in  $\mu\text{g L}^{-1}$  (AM: arithmetic mean).

Canton	Commune	n	Low	High	Median	AM	Population
VS Martigny		10	0.43	92.02	8.33	22.64	15778
VS Lax		15	0.11	49.45	1.53	14.25	306
VS Bitsch		13	0.35	47.93	13.91	19.30	844
VS Visp		33	4.67	37.72	7.68	12.82	6842
VS Sion		37	0.31	35.61	0.61	5.79	29718
VS St. Niklaus		23	0.45	32.12	13.75	12.89	2283
VS Visperterminen		36	0.04	28.17	1.72	4.97	1390
VS Lalden		6	11.21	27.71	19.19	19.59	665
VS Stalden (VS)		12	0.42	26.86	1.22	5.44	1129
VS Conthey		24	0.05	26.74	0.32	4.84	7445
VS Zermatt		34	0.025	25.91	0.58	2.28	5828
VS Les Agettes		18	1.97	25.65	11.14	12.52	346
VS Grächen		21	1.10	25.12	6.73	10.56	1365
VS Steg-Hohtenn		19	0.97	24.42	4.22	5.65	1514
VS Fully		21	0.02	24.04	4.70	7.30	7411
VS Baltschieder		12	2.57	23.75	4.72	7.37	1200
VS Vernayaz		3	8.24	21.61	16.94	15.60	1799
VS Fiesch		9	2.75	21.19	3.97	9.24	972
VS Turtmann		18	0.32	20.43	8.02	7.78	969
VS Agarn		5	5.93	20.00	6.26	8.94	771
VS Vex		67	0.06	19.26	2.03	4.60	1631
VS Naters		24	1.04	18.81	2.98	5.73	8096
VS Eggerberg		4	9.69	18.53	14.21	14.16	343
VS Evolène		21	0.03	17.79	0.72	3.20	1636
VS Gampel-Bratsch		6	2.15	17.38	3.39	7.20	1858
VS Zeneggen		3	10.84	17.36	16.98	15.06	245
VS Ernen		23	0.025	17.08	1.03	3.55	527
VS Blitzingen		13	3.33	17.01	7.97	8.28	79
VS Simplon		21	0.04	15.73	0.88	3.31	349
VS Unterbäch		6	0.50	15.62	3.24	4.63	409
VS Brig-Glis		23	0.53	15.35	2.49	3.91	12254
TI Lodrino		2	2.02	26.12	14.07	14.07	1641
GR Poschiavo		14	1.30	28.80	7.10	9.19	3521
GR Bergün/Bravuogn		4	2.20	28.40	10.90	13.10	474
GR Davos		21	0.00	23.39	1.30	3.21	11248
GR Disentis/Mustér		6	2.00	23.10	9.30	11.12	2121
GR Rueun		1	15.30	15.30	15.30	–	416
FR Haut-Vully		8	0.55	27.90	6.30	8.36	1335
FR Bas-Vully		16	2.74	24.37	5.56	6.74	1924

do not originate locally but are transported from other areas along fault zones before reaching the source.

### 3.3. Ticino (TI)

The highest concentrations in Ticino are found in Lodrino, situated on the Leventina nappe where deposits of brannerite ( $\text{UTi}_2\text{O}_6$ ) are documented (Gilliéron, 1988). The mineral is found in alkali-feldspar-pegmatites, veins of quartz and at joints in Leventina granite gneiss.

### 3.4. Fribourg (FR)

The two communes in Fribourg where higher concentrations of uranium were measured are Haut- and Bas-Vully. These both neighbour Mont Vully, a molasse outcrop of sandstones, shales and conglomerates. Uranium deposition most likely took place in early diagenetic times when waters containing soluble U(VI) species flowed through the sand and U(IV) was precipitated. A detailed analysis of the geology and its effects on the groundwater was carried out (Schott and Wiegand, 2003) and shows areas of organic deposits of high uranium concentration, for example bone fragments with concentrations up to four orders of magnitude higher than found in vivo resulting from a low but persistent supply of uranium in the groundwater. Of the sources measured in the study by Schott et al., those which were spring water showed a higher

concentration than those collected from the groundwater via drainage pipes. This implies the spring water originates from highly permeable regions such as faults or jointed sandstones. The rocks of this area have been shown to contain carbonate which, in the presence of CO<sub>2</sub>, can dissolve into the water. Such conditions are favourable for the dissolution of uranium from the deposits (Ramseyer, 1952). Indeed, the high content of uranium in waters of this region can be traced back to the uranium rich rocks with a high content of organic matter in the lower parts of the aquifer (Schott and Wiegand, 2003).

### 3.5. What does this mean for Switzerland?

The WHO guideline value is calculated using an assumed water consumption of 2 L. However, dietary reference values for water determined both by EFSA (EFSA, 2010) and a committee consisting of the German, Austrian and Swiss Societies for Nutrition (DGE et al., 2011) determine a total adequate intake of around 2.5 L; subtracting the water intake due to food (20–30%) and oxidation water due to metabolism of macronutrients (typically 250–350 mL) a total of 1.5 L is left to be consumed from water and beverages. Calculating, on the basis of a 60 kg adult drinking 1.5 L of water, the current TDI will be reached with a concentration of 40 µg L<sup>-1</sup> and the 2004 TDI will be reached with a water concentration of 20 µg L<sup>-1</sup>.

If Switzerland were to adopt a value of 40 µg L<sup>-1</sup> as its maximum level, three communes would need to implement measures to reduce uranium concentrations in parts of their water supply; if the current WHO guideline of 30 µg L<sup>-1</sup>, six communes would be affected (0.7% of the total population); if a guideline of 20 µg L<sup>-1</sup>, 20 communes; if the 2004 WHO guideline of 15 µg L<sup>-1</sup>, 39 communes (1.8% of the total population) (BFS, 2011). If Switzerland were to follow suit with Germany, 65 communes would be affected.

A special case should be noted, regarding children. Proportionally, infants drink a much greater volume of water with respect to their body weight than adults. There is reason to suspect that absorption of uranium in infants is higher than in adults (Chen et al., 2011). Taking the infant scenario used by the EFSA (EFSA, 2009), of a 3 month old child weighing 6.1 kg drinking on average 675 mL and a 95th percentile of 917 mL the exposure values in µg kg<sup>-1</sup> b.w. d<sup>-1</sup> for a formula fed child in Switzerland are shown in Table 5. Infants exposed to concentrations at the 95th percentile and consuming at the 95th percentile will consume 1.33 times excess the WHO TDI. For such an infant, the TDI will be exceeded with concentrations higher than 6 µg L<sup>-1</sup>.

In Switzerland, 78% of the water samples measured were below 2 µg L<sup>-1</sup> and 92% are below 6 µg L<sup>-1</sup>. Looking at the geographical distribution of uranium, it is clear that the vast majority is found in the alpine regions in the cantons of GR, TI and VS. Descriptive statistics for this region are compared to that of the rest of the country and the overall results (Table 2). For all cantons excluding GR, TI and VS the proportions rise to 91% of samples having [U] < 2 µg L<sup>-1</sup> and 99% having [U] < 6 µg L<sup>-1</sup> whilst the data for these cantons alone return 63% and 83% respectively.

**Table 5**  
Uranium consumption (µg kg<sup>-1</sup> b.w. d<sup>-1</sup>) for a 3 month old formula fed infant at mean, median and 95 percentile concentrations in Switzerland.

U concentration (µg L <sup>-1</sup> )	Water volume consumed (mL)	
	675 (Mean)	917 (95th Percentile)
2.04 (Mean)	0.23	0.31
0.77 (Median)	0.09	0.12
8.84 (95th Percentile)	0.98	1.33

A reduction of uranium concentration in drinking water can be achieved in several ways. The simplest, most practical solution would be via dilution, simply mixing water from a source with higher concentration with that of a lower. This has the advantage that there is no residue requiring disposal. If this is either not possible or does not result in a low enough concentration then it may be possible to remove uranium from the water. Several methods have been postulated, such as coagulation with ferric or aluminium sulphate at an optimal pH (80–95% removal), filtration, lime softening, anion exchange or reverse osmosis (99% removal) although not all methods have been tested on an industrial scale (Giddings, 2005; ATSDR, 2009) and disposal of the residue must also be considered.

## 4. Conclusions

This survey has provided an insight into the uranium levels in the drinking water of some cantons of Switzerland. A few sources have been identified which produce water with elevated uranium concentrations and so indicate to the cantonal authorities areas requiring monitoring and further action. The data depicted here is for each commune and so dependent on the number of samples measured in a particular area. In some regions a “problem” area was identified and re-sampled with more samples being collected along the network to ascertain the breadth of the problem, potentially leading to a higher than otherwise expected mean. It should be noted that the most extensive surveys were carried out in the alpine cantons of Valais, Graubünden and Ticino and are not representative of the rest of the country. Concentrations depend on the underlying geology of the region, although it is difficult to specify a direct relationship owing to the fact that the water flows underground and so does not necessarily originate in the same rock as the source of uranium. These sources in which high concentrations were measured should be monitored and if possible determined how many people are affected and whether or not they are permanent residents or merely seasonal. Another area worth further investigation is the Bernese Alps for which no data is available. As these are situated on the same massif as parts of Valais, Uri and Graubünden, it seems highly likely that elevated uranium concentrations would be found in this area also.

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