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Uranium natural levels in water and soils: assessment of the Italian situation in relation to quality standards for drinking water

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Abstract In view of the establishment of quality standards for uranium in drinking water by the European and Italian authorities, we report a comprehensive overview of the Italian situation regarding uranium concentrations in natural waters. More than 3100 data on waters all over Italy are reviewed in addition to new data for Latium Region (Central Italy) where relatively high concentrations in rocks are long been known. Our study evidences that uranium in bedrock is not the main factor ruling uranium presence in fresh and groundwater, indicating that redox conditions as well as bicarbonates and silicates control solution equilibria and accordingly the uranium content in waters. At present, and considering the provisional guideline value of 30 μg/L established by [WHO (2012) Guidelines for Drinking-Water Quality. World Health Organization, Geneva], no critical situation is envisaged in Italy, but this can be overruled if lower limits will be actually imposed. There is also a need of further studies on uranium speciation in water and of wider epidemiological studies to assess its real toxicity to overcome the uncertainties that have so far hampered the definition of reliable water quality standards.

Keywords Uranium · Natural background · Drinking water · Regulatory limits · Italy

1 Introduction

In recent years, epidemiological studies and further investigations have focused the attention of the scientific community on some potentially toxic and/or carcinogenic elements that in some regions show natural high concentration due to "geochemical anomalies". In fact, the nature of the rocks and their alteration (during the soil formation and weathering) cause the redistribution of the elements, producing a great variability in their concentrations. In some cases enrichments of potentially harmful elements may create concern for human health. Particular attention is paid, of course, to water intended for human consumption. A well-known case is that of arsenic, for which in 2001 the European drinking water standard was lowered from 50 to 10 μg/L. As a result, in Italy many municipalities had to face the problem of drinking water that had suddenly become "illegal" and were forced to find alternative sources or to adopt treatment systems, but the ultimate solution is still far to come.

Recently, attention has been put also on uranium due to its known toxicity. Even if at the moment standard limits for drinking water have not yet been set in Europe, uranium toxicity could cause the same difficulties depending on the natural background levels of different areas.

Uranium occurs naturally in soils and waters with three isotopes: ^{238}U (abundance 99.285 %), ^{235}U (0.71 %) and ^{234}U (0.005 %); it is mainly associated with the suite of "granitic rocks" and during magmatic differentiation shows a tendency to increase with the silica content. It is also found in various mineral deposits as phosphate rocks,

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pegmatites, felsic rocks and into a variety of minerals. Leaching is the main cause of natural uranium mobility while anthropogenic uranium is released from mine mill tailing and nuclear industry emission. Uranium is also present in the combustion products of coal as far as other fuels and it is also present in fertilisers, mainly in phosphates. (Allard et al. 1999; Giammar and Hering 2001; WHO 2004). Natural organic matter can significantly influence uranium speciation in environmental settings, as carboxyl groups of naturally occurring humic and fulvic acids can strongly bind uranium (Gascoyne 1992; Lenhart et al. 2000).

The concentration of uranium in groundwater is usually in the range 0.1–50 μg/L and about 0.9–1.7 mg/kg in the Earth's crust (Kabata-Pendias and Mukherjee 2007). Uranium solubility in aqueous systems is predominantly controlled by three factors: oxidation–reduction potential, pH and dissolved carbonate (Murphy and Shock 1999). When dissolved carbonate concentrations are high, uranium is highly mobile due to the formation of uranyl-carbonate $UO_2(CO_3)$ species. In aqueous solution under environmental oxidising conditions uranium can exist in oxidation states of +IV and +VI; U(VI) is considerably more soluble than U(IV). Under reducing conditions, the precipitation of U(IV) is the dominant process leading to naturally enriched zones of uranium in the subsurface (Osmond and Cowart 1992). The sorption of uranium to aquifer rocks is frequently dominated by the association with iron oxyhydroxide minerals.

While its natural occurrence is not correlated to a significant risk in terms of radioactivity, this element, like other "heavy metals", may have a non-negligible chemotoxicity. Because of the long physical half-lives of the three isotopes found in the environment, the specific activity (the amount of radioactivity per unit weight) of uranium is low; thus, chemical toxicity may often be of greater concern than radiotoxicity. Uranium from the environment enters the human body by ingestion with food and drink and by inhalation of airborne particles. Daily intake of uranium in food and water varies from ~1 to ~5 μg/day in uncontaminated regions to 13–18 μg/day or more in uranium mining areas (Taylor and Taylor 1997).

The main organ affected by the toxicity of uranium is the kidney; nephritis is the primary chemically induced effect of uranium in humans (Hursh and Spoor 1973), and the principal route of elimination is the urinary excretion: consequently, the uranium content in the urine can be used to assess the exposure levels. Intake through drinking water is normally low; however, in circumstances in which uranium is present in a drinking water source, the majority of intake can be through drinking-water (WHO 2012).

Studies done in the past decade by the World Health Organization (WHO) and the US Environmental Protection Agency (EPA) have, over time, suggested safety levels for drinking water with limits ranging between 10 and 30 μg/L, the latter being the current value recommended by the WHO (2012). The US EPA fixes a maximum contaminant level (MCL) of 30 μg/L but states that their MCL 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 the European Food Safety Authority (EFSA) endorsing the total daily intake of 0.6 μg/kg b.w./day as described by the WHO (EFSA 2009). Germany has recently introduced a limit of 10 μg/L (Bundesministerium für Gesundheit 2011). No other country in Europe has fixed limits, but UK Food Standards Agency advices to avoid using natural mineral water to prepare infant food (COT 2006), and in Switzerland the uranium maximum permissible concentrations of uranium as radionuclide is 10 Bq/kg in liquid foodstuffs (EDI 2010).

Starting from the numerous data available for many Italian regions acquired by studies done since late 50s in our laboratory (formerly "Geomining Laboratory"), we provide an overview of the distribution of uranium in groundwater, freshwater and soils of various geographical areas in Italy, to assess the average concentrations and highlight any values differing significantly from the average. In addition, new data are reported for a particular area, the Latium region, where high content of uranium and other elements of toxicological interest in rocks has long been known. This characterization of uranium distribution in natural matrices in Italy aims to be a preliminary reference in case of setting a regulatory limit value for drinking water as well as giving indications on the uranium background values on the Italian territory.

2 Geological setting of Latium volcanic districts

The uraniferous mineralization in Latium is related, directly or indirectly, to the Pliocene and Quaternary volcanism. The volcanism occurring in the uraniferous volcano-sedimentary basin belongs mostly to the potassic 'Mediterranean' magmatic suite. The cause of this volcanism, which led to the formation of the major volcanic districts along a NW–SE direction, is in turn linked to the different phases of tectonics that have affected the area from the Late Miocene.

The study areas (Fig. 1) are characterized by the presence of volcanic formations belonging to the so-called "Roman Comagmatic Province" outcropping extensively in Latium and developed in relation to the final stages of the Apennine orogeny and to the opening of the Tyrrhenian back-arc basin (Wezel 1982, 1985; Serri et al. 1991; Barberi et al. 1994). As a result of these phenomena, from the

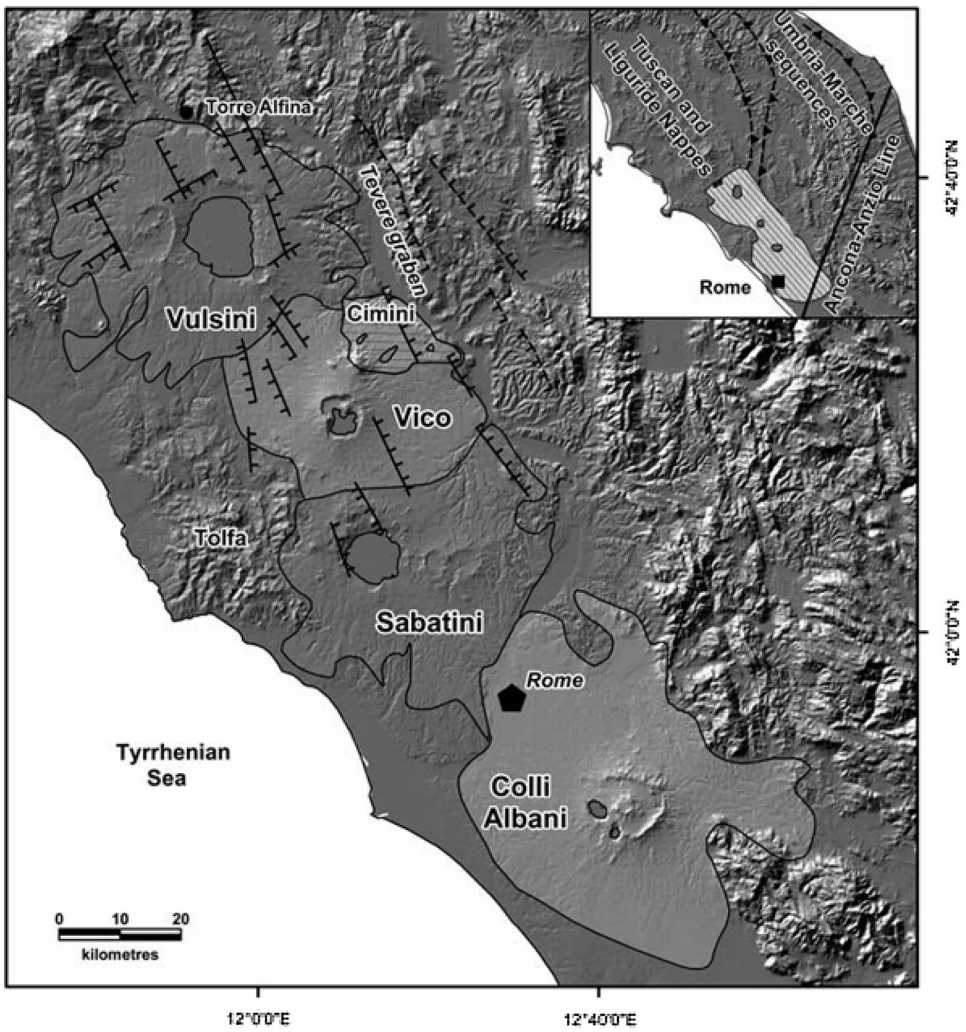

Fig. 1 Location map of volcanoes of the Roman Province. (From: Peccerillo 2005). With kind permission from Springer Science and Business Media

Pliocene the region was affected by large volcanic events, whose final stages, represented by widespread degassing processes, are still present causing the enrichment in some elements characterising this area.

The Cimino volcano, active between 1.35 and 0.95 Ma, showed an uncommon magmatic evolution from acidic anatectic products to final potassic olivine-latitic melts, which are very similar to the 'Mediterranean' suite. This volcano is believed to has been derived from partial melting of the crust due to the upward migration of the geotherms accompanying the uplift of the mantle-derived magmas (Locardi 1985). Several domes are radially arranged around the top of Mt. Cimino, which developed along a NW–SE trending fracture and included pyroclastic deposits.

The Vico complex consists of a stratovolcano with the centre of the caldera housing the eponymous lake. The volcano of Vico was active between 419 and 95 ka from present and developed through alternating explosive and effusive phases which have led to several pyroclastic deposits and lava flows of phonolitic, tephritic and trachytic composition (Locardi 1965; Mattias and Ventriglia 1970; Bertagnini and Sbrana 1986). The rocks and soils of the area, in addition to the known high concentrations of arsenic (Armiento et al. 2015), show a high average content in other elements including uranium.

The Sabatini volcanic district extends over an area of about 1800 km^2 and its volcanic products interfinger towards south with the 560–350 ka-old pyroclastic deposits of the Tuscolano-Artemisio phase of the Alban Hills volcanic district, and in the northern part they are buried by the 0.15 Ma-old Vico C ignimbrite of the Vico volcanic district (Sottili et al. 2004). The oldest products, dated by Cioni et al. (1993) to 600 ka from present, are represented by trachytic lava. Major eruptive events are represented by the pyroclastic succession of the Tufo Giallo della Via Tiberina (561–548 ka) and Tufo Rosso a Scorie Nere (449 ka). The final phases (dated to 300–200 ka) of the eruptive activity occurred over the areas of the Sacrofano Caldera and the Bracciano

volcano-tectonic depression. The typical lava rock-types of the Sabatini volcanic district have a prevailing potassic composition ranging from trachybasalts to trachytes to phonolites.

The volcanic complex of Alban Hills is located SE of Rome and was characterized by an intense volcanism, whose potassium-alkaline products overlay the Mesozoic carbonate sequence of the Central Apennines. The Alban Hills were built up by several effusive and explosive events, which occurred during two main phases of activity, separated by a huge explosive eruption responsible for the emplacement of the Villa Senni Eruption Unit and the subsequent collapse of a central caldera (Peccerillo et al. 1984). The earliest period of this volcanic activity (called "Tuscolano-Artemisio" phase) occurred between 700 and 300 ka and is characterized by several large explosive eruptions, which have put in place pyroclastic products in an area of over 1500 km^2. The Villa Senni Eruption Unit (338 ka) is the last major explosive event of this phase. After the caldera collapse the activity resumed in a smaller area, with the emission of lava flows and scoriae, and terminated within the calderic depression with small phreatomagmatic explosions (Peccerillo et al. 1984). Alban Hills lavas are basic or ultrabasic in composition, and typically enriched in potassium.

3 Materials and methods

3.1 Data and samples

Several data from previously published studies, mainly on water (groundwater, freshwater, tap and mineral bottled) were collected and systematised, obtaining an overview of the Italian situation. In addition, new data were produced during this study, concerning a particular area where uranium is long known to be present in deposits with relatively high concentrations.

In particular, a detailed characterization has been made in the area of the Vico-Cimino volcanic complex, where uranium shows the highest content of the investigated region, principally in rocks and soils, in addition to high concentrations of other potentially harmful elements (PHEs), such as arsenic, beryllium, vanadium, antimony, copper, lead and thorium. In this area 75 samples of soils and rocks were sampled, as well as 32 water samples were collected from drinking troughs in all municipalities of the Viterbo province.

Samples belonging to the Sabatini area are represented by 6 drilled core, for a total of 24 rocks and 6 topsoils, in addition to 28 water samples collected from rivers, lakes, springs and wells.

In the area of Alban Hills 43 samples of soils (*n*. 36) and rocks (*n*. 7), belonging to the widespread Villa Senni Unit, were collected, while 19 water samples from wells and springs were sampled in a broad area comprising the entire Alban Hills volcanic district.

For all the three areas, water samples were collected according to the principle that they would represent a natural water, i.e. sampled from wells, springs, streams and drinking troughs, but avoiding household tap water, that in some cases could have been treated or supplied from aqueducts whose water originate far from the areas of interest.

3.2 Sample treatment and analyses

Soil and rock samples were oven dried at 40 °C, soil samples were then sieved and the fraction with a size <2 mm was selected. Sieved soils and bulk rocks were successively ground and uranium concentrations were measured by ICP-MS (Perkin Elmer ELAN 6100) after acid dissolution of the powdered samples and performed according to the EPA procedure 3052 using 69 % HNO_3, 40 % HF and 30 % H_2O_2 as reagent mixture.

The water samples were collected in polyethylene tubes, after being filtered through 0.45 μm nitrocellulose membrane filters. They were then acidified to pH <2 with HNO_3 and immediately stored at 4 °C until being analysed for uranium by ICP-MS.

4 Results and discussion

4.1 Northern and Southern Italy: literature data review

A collection and systematisation of the available literature on uranium content in surface and groundwater in Italy has been carried out, starting from the numerous data available from previous studies performed at our laboratory since the early 1960. At that time, the former CNEN (Italian National Council for Nuclear Energy), now ENEA, conducted several surveys and sampling campaigns all over Italy with the aim of finding evidence of uranium economically valuable deposits and starting a comprehensive geochemical characterization of the Italian territory. This record represents a particularly valuable dataset because, dating to some decades ago, it gives information on natural background concentrations without the addition of diffuse anthropogenic input that eventually occurred lately. Data reported in Table 1 represent the results of 2249 water samples (plus data on rocks and alluvial sediments) analyses performed at CNEN/ENEA in the timespan from 1960s to 2009. In addition, data derived from other

Table 1 Uranium content in water and rock samples collected in various regions of Italy

Region	Locality	Sample type	N. samples	*U*	References
Italy	–	Rivers	17	0.15–4.40	Dall'Aglio et al. (1983), Brondi et al. (1986)
Lombardy	Milan area	Well	7	2.6–4.8	Brondi et al. (1986)
		Groundwater	15	0.05–4.82	Dall'Aglio and Gragnani (1983)
	Novazza/ Valvedello (BG)	Surface water	60	0.04–1.2	Clemente et al. (1982)
		Groundwater		0.05–1.9	
		River sediment, soil	n.r.	0.5–29 mg/kg	
Piedmont	Alessandria area	Well	7	<0.4–4.4	Brondi et al. (1986)
	Bosco Marengo (AL)	Various	51	0.719 ± 0.426	Dall'Aglio et al. (1976)
Veneto	Vicenza	River, groundwater	20	0.07–1.9	Dall'Aglio et al. (1983)
	(Alpi Dolomitiche, BL)	River, spring water	28	0.025–1.8	Brondi et al. (1989)
Trentino AA	Val Venosta	Water	241	2.53 (st. dev. 6.7)	Dall'Aglio (1966, 1971)
	Martello Valley	Spring water	12	6.2 (st. dev. 3.4)	
		Surface water	17	7.6 (st. dev. 3.1)	
Latium	Latium region	Lake	6	0.6–4.9	Brondi et al. (1986)
		River	4	0.7–5.2	
		Spring	5	0.4–3.8	
	Mts Cimini	Surface water	126	1.9	Dall'Aglio (1968)
		Cold spring	39	0.6	
	Northern Latium	Water in equilibrium with carnotite	1	1.3	Dall'Aglio et al. (1974)
	Vico-Cimino complex	Cold water	60	7.2 (0.04–49)	Angelone et al. (2009)
		Thermal water	5	0.07 (0.06-0-09)	
Tuscany	Tuscany region	Surface water	934	0.71 (st. dev. 0.73)	Dall'Aglio (1968)
		Alluvial sample	952	0.67 (st. dev. 0.62) mg/kg	
Campania	Phlegrean fields	Spring	8	0.3–18	Brondi et al. (1986)
		Well, thermal spring	11	0.5–96	
Apulia	Apulia region	Spring	8	0.7–4.6	Brondi et al. (1986)
		Groundwater	21	0.74–13	Brondi et al. (1983)
	Taranto	River water	6	0.72–2.3	Brondi et al. (1985)
Basilicata	Rotondella (MT)	River water	12	0.32–2.7	Boeri et al. (1979)
Calabria	Sila Plateau	Spring water in equilibrium with autunite	1	0.55	Dall'Aglio et al. (1974)
		Spring water leaching granite rock and emerging near autunite	1	0.64	Dall'Aglio (1973)
Sicily	Western Sicily	Stream water	299	1.09	Dall'Aglio (1970)
	Mts Peloritani	Surface water	228	2.5	Dall'Aglio et al. (1975)
		Fluvial deposit	296	3.27	
Sardinia	Sardinia region	Surface water	91	1.53	Carrara et al. (1974)
		Rock	88	0.19–5.04 mg/kg as U_3O_8	

Data from studies conducted at the ENEA (formerly CNEN) laboratories. (U μg/L, unless otherwise noted)

n.r. not reported

Table 2 Uranium content in natural waters, from various published studies

Region	Locality	Sample type	N. samples	*U* (μg/L)	References
Latium	Vico-Cimino complex	Cold volcanic water	223	6.0 (st. dev. 8.2)	Cinti et al. (2015)
		Cold sedimentary water	57	2.5 (st. dev. 3.9)	
		Thermal water	33	0.91 (st. dev. 2.4)	
	Viterbo province	Spring, well	231	2.7–13.8	Sappa et al. (2014)
Sardinia	Osilo epithermal gold deposit	Surface and groundwater	48	1–24	Biddau and Cidu (2005)
		River (winter)	34	<0.04–4.2	Cidu and Biddau (2007)
		River (summer)	16	0.48–3.7	
		River, stream	141	0.78 (<0.01–6.4)	Cidu and Frau (2009)
		Surface waters draining abandoned mines	27	1.5 (0.25–15.5)	
Basilicata	Mt Vulture	Groundwater	34	1.6 (0.1–5.6)	Parisi et al. (2011)
Campania	Vesuvius	Groundwater	54	33.3 (0.3–137)	Aiuppa et al. (2005)
Sicily	Vulcano	Groundwater	21	11.6 (0.1–57)	Aiuppa et al. (2000a)
	Etna	Well, spring, galleries	53	3.3 (0.07–11.5)	Aiuppa et al. (2000b)
	Etna	Well, spring, galleries	13	0,16–5.1*	Kozlowska et al. (2009)

*As radioactivity, conversion assuming ^{238}U specific activity of 1 Bq/L = 80.37 μg/L (EFSA 2009)

published studies are reported in Table 2 and consist of 863 analyses, bringing the total data collected to 3112. Besides, other new analyses of 47 waters and 148 rocks and soils performed for this study have been added. This is quite an impressive number of data, also compared to the samples analysed, e.g. for geochemical surveys conducted at European continental scale for the Geochemical Atlas of Europe (Salminen et al. 2005).

In Italy the geological and environmental settings vary over a wide range of conditions, influencing the mobility and distribution of uranium in various media. The Alpine range has high altitudes, heavy rainfalls and low average temperatures. The outcropping formations are principally silicic crystalline (granite, gneiss, metamorphic rocks) in composition (Dall'Aglio 1968). Known uranium anomalies along the Alpine arc are in Lombardia, near Novazza, where uranium deposits occur (Tedesco 1984; Nash 2010). In the Novazza/Valvedello uraniferous district uranium shows low concentrations in water, apparently in contrast to the high geochemical background of this area (Table 1). This behaviour should be related to the high silicization of the uranium-enriched orebodies that makes them very resistant to leaching, in addition to a poor water–rock interaction due the hydrodynamic conditions of the mountain environment (Clemente et al. 1982).

Anomalous uranium contents were recognised in the alluvia of Val Venosta (Trentino Alto Adige) (Dall'Aglio 1966, 1971), with concentrations up to 90 mg/kg. Nevertheless, the reported values for surface and groundwater are on average 7.6 μg/L in the sub-area of Martello valley, never approaching the guideline value of 30 μg/L recommended by WHO (2012).

The outcropping rocks belong to the crystalline basement ("Cristallino Antico") consisting of metamorphic formations such as micaceous schists, gneiss, amphibolite, all rocks characterized by low solubility. This feature, combined with the circulation of surface waters through preferential paths is the explanation for a poor leaching and, therefore, for low uranium concentrations into the waters (Dall'Aglio 1966).

In Southern and Insular Italy the environmental conditions are widely different. Rainfalls are generally scarce (<600 mm/year) and temperatures average 20 °C, even if a climate similar to the Alpine one is present in some areas where mountains reach 1000 m of altitude. The outcropping geolithologic formations and the weathering conditions differ widely from area to area, ranging from the Apenninic carbonaceous range to volcanic and metamorphic genetic environments. Two particular settings in Southern Italy are relevant as far as uranium is concerned. One is the Sila Plateau in Calabria, where the leaching of crystalline rocks, even with not high uranium content, under particular erosion and weathering conditions in the wet temperate climatic zone causes the precipitation of uranium minerals, namely autunite (Dall'Aglio et al. 1974). The uranium content in surface and spring water (Table 1) of this area is moderate and differentiated in two different zones, according to the tectonic setting that influences the bicarbonate content and consequently the activity of uranyl ion on uranium solubility (Dall'Aglio et al. 1974).

The Campania volcanic area represents another relevant case in Southern Italy. Data concerning the Vesuvius groundwater show very high uranium concentrations with values up to 137 μg/L (Table 2, Aiuppa et al. 2005). This active volcanic area, characterised by a potassic magmatic suite, is affected by extensive water–rock interactions that are promoted by magmatic and hydrothermal gas dissolution into groundwater. The elevated uranium concentrations are explained by the high total carbon content (bicarbonate is the predominant anionic species into solution) of the groundwater and to the high stability of metal-carbonate complexes. These species dominate to a large degree the equilibrium speciation of uranium, due to the strong affinity for carbon of the uranyl ion (UO_2^{2+}), the main hydrolysis product of U(VI) (Langmuir 1978). Farther, extreme ^{238}U enrichment in Vesuvius lavas is a known occurrence, whose origin is not completely clear, and has been tentatively attributed to metasomatism and the addition of subduction-related carbonate fluids (Avanzinelli et al. 2008).

Data on uranium concentrations in thermal spring water collected in the area of Phlegrean Fields (near Naples) (Brondi et al. 1986) and Vulcano (Sicily) (Aiuppa et al. 2000a) are reported in Tables 1 and 2, respectively. The high uranium content, up to 57 μg/L, originates from an extensive hydrothermal circulation, with the widespread occurrence of surface manifestations including fumarole discharges and boiling to hypothermal waters (Aiuppa et al. 2006). The complex setting of these two volcanic areas, characterized by the mixing of meteoric recharge, usually rich in bicarbonates, with volcanic fluids with a large compositional range spanning from chloride to sulphate-chloride to bicarbonate, generates highly variable U content and can cause favourable conditions for U solubilisation into circulating waters (Aiuppa et al. 2000a, 2006; Boschetti et al. 2003).

Data regarding the other active volcanic area of Southern Italy, namely Mt. Etna, show lower uranium levels in groundwater, although with a high variability in a range between 0.07 and 11.5 μg/L (Table 2, Aiuppa et al. 2000b; Kozlowska et al. 2009). This variability is linked to different Eh–pH conditions that in some cases include the occurrence of U scavenging by Fe oxides/hydroxides (Aiuppa et al. 2000b). The basaltic rocks of Mt. Etna show relatively low uranium content (mean 3, range 2.1–4.1 mg/kg) and a low leachability (Kozlowska et al. 2009).

4.2 Central Italy: literature review and new data

In Central Italy the geological setting is represented by the calcareous Apennines overlaid by volcanic rocks on the Tyrrhenian border in the Tuscany and Latium territory. Latium quaternary alkali-potassic volcanites are characterised by a peculiar chemical and mineralogical composition, with exceptionally high contents of many trace elements, such as U, F, B, Be, Co, Cs, Ba, Pb and Th. These elevated concentrations are attributed to the uplift of mantle enriched in incompatible elements by deep fluids and metasomatic products (Locardi 1982). This highly potassic province presents a range of uranium content increasing in the more alkaline suites, corresponding to different degrees of partial melting in the mantle. An additional mechanism for uranium enrichments has been proposed, i.e. the transport by a very mobile fluid phase, to explain the uranium variable content in similar products of different volcanic groups in the region (Locardi 1982).

Starting from the evaluation of the data collected about the Italian situation (Tables 1, 2), we carried out further studies in the areas where the main volcanic complexes occur in Latium. Thus, in addition to the area of Vico-Cimino complex, already extensively investigated for uranium occurrence, also the area of Sabatini volcanic complex and of the Alban Hills district were addressed, mainly with the aim of gathering information on uranium content in drinking water, starting from the assessment of uranium content in the aquifer rocks.

Data concerning uranium concentration in 75 rocks of the Vico-Cimino district (Table 3) show an average of 24 mg/kg, with values ranging from 2.4 to 85 mg/kg. The high concentrations of uranium in the area are associated with high content of other elements, e.g. arsenic (Armiento et al. 2015) and beryllium (Armiento et al. 2012), and are also documented by the presence of abandoned uranium mining sites. During the exploration activities for uranium research carried out starting from the late 50s, the territory comprising the volcanic complexes of the Northern Latium and Southern Tuscany was classified as "uranium area" and the uranium-related deposits were estimated as of sub-economic importance. Uranium mineralizations occur always in association with CO_2 and H_2S exhalations that, alongside travertinization, are widespread all over the region (Locardi 1985). Where these gases percolated the groundwater in the upper oxygenated part of the aquifer,

Table 3 Uranium content in rocks and soils of the volcanic district of Latium analysed for this study

Volcanic district	N. samples	*U* (mg/kg)			
		Mean	Min	Max	Median
Vico-Cimino (rock)	75	24	2.4	85	21
Sabatini (topsoil)	6	4.3	0.41	14	0.73
Sabatini (rock)	24	6.1	0.25	32	1.2
Alban Hills[a] (topsoil)	36	5.2	2.4	8.4	5.1
Alban Hills[a] (rock)	7	7.5	5.1	9.9	7.2

[a] Tufo di Villa Senni

the sulphur oxidisation caused pH and Eh variations that induced uranium to precipitate as oxide. As a consequence, the waters flowing in the areas where mineralizations occur show usually very low uranium concentrations, being the redox conditions favourable to its removal from solutions. This geochemical behaviour explains also the difference in uranium content between cold and thermal waters in the Latium region (Tables 1, 2, Angelone et al. 2009; Cinti et al. 2015). Cold waters are mostly characterized by nearly neutral and oxidising conditions, favouring the uranium mobility and, consistently, uranium concentrations in water are higher. Conversely, the prevailing anoxic conditions of most thermal waters decrease the uranium mobility, causing its precipitation and relatively low contents in waters. The highest uranium concentrations in groundwater were found in the area southwest the Vico Lake, which is characterized by oxidising conditions and elevated HCO_3 concentrations (Angelone et al. 2009), all these factors favouring uranium mobility through the formation of the soluble uranyl-carbonate complex.

This occurrence has been confirmed by the analyses that we carried out on 32 water samples collected from drinking troughs all over the Viterbo province (Table 4). The highest values (range 10-15.1 μg/L) were found in 4 municipalities southwest the Vico Lake, while in other 9 municipalities the values ranged from 5 to 10 μg/L and in the remaining 19 the values were lower than 5 μg/L.

The Sabatini Volcanic district belongs to the same magmatic province of the Vico-Cimino and is also characterized by relatively high content of incompatible elements in rocks. In an area about 15 km^2 south of Bracciano Lake 30 soil and rock samples were collected from 6 cores drilled through the volcanic formations down to the sedimentary substrate. The results (Table 3) show mean uranium content of 5.7 mg/kg that is quite lower than that measured in the Vico-Cimino rocks, but still significant. The geochemical behaviour of uranium explains the slight depletion of the element in topsoils (mean 4.3 mg/kg) observed with respect to the underlying rock formations (mean 6.1 mg/kg), due to the enhanced element mobility in oxidising environments. On the contrary, its tendency to precipitate in reducing conditions is evidenced by higher uranium concentrations in strata formed by clayey lacustrine deposits (13.1 mg/kg) and in a core drilled in proximity of a hydrothermal fluids vent (mean 15.5 mg/kg, range 7.2–2 mg/kg).

In Table 4 the results of the analyses of different types of waters collected in the entire area of the Sabatini volcanic district are reported, including the Bracciano Lake water that is used as a source for supplying drinking water to the city of Rome. Data show relatively low mean uranium concentrations, correlating with HCO_3 concentrations. The uranium content of Martignano Lake water (Table 4), lower than that of Bracciano Lake, can be explained by the different origin of the two lakes. The latter is hosted in a volcano-tectonic depression and its waters are fed by numerous springs flowing at the contact between basic lava flows or tuffs and the underlying sedimentary series. On the contrary, the Martignano lake is a close basin fed by rainfall and with surface water circulation; consequently, a lower water–rock interaction prevents uranium leaching from rocks.

In the area of the Alban Hills volcanic district 36 topsoil and 7 rock samples were collected and analysed. Also in this case we found slightly higher uranium content in rocks than in soils, matching the same geochemical behaviour explained for the Sabatini district soils. The mean uranium content in rocks and soils is similar to that of the Sabatini, but much lower than the Vico-Cimino, confirming that the latter represents an area with significant uranium enrichment in rocks even compared to the other volcanic districts of Latium.

Table 4 Uranium content in waters from the volcanic districts of Latium analysed for this study

Volcanic district	Type	Name/subtype	N. samples	*U* (μg/L)		
				Mean	Min	Max
Vico-Cimino	Groundwater	Drinking trough	32	5.1	0.25	15.1
Sabatini	Freshwater	Mignone River	6	3.3	2.8	3.7
		Arrone River	6	3.1	1.1	4.4
		Treja River	1	3.5	–	–
		Bracciano Lake	2	3.2	2.8	3.6
		Martignano Lake	2	0.82	0.80	0.84
	Groundwater	Spring	7	2.9	0.4	6.0
		Well	6	2.4	0.3	3.5
Alban Hills	Groundwater	Spring	14	4.4	0.07	17
		Well	5	9.5	1.4	21

The 19 water samples collected from springs and wells located all over the Alban Hills volcanic district showed uranium concentrations slightly higher than those of the Sabatini district and somehow similar to those of the Vico-Cimino district, already measured in our laboratory and reported in Angelone et al. (2009). Uranium enrichments in Vico rocks are due to hydrochemical conditions favouring uranium secondary deposition, clearly involving the removal from water solutions (Locardi 1985) and resulting in concentration similar to those of areas with lower uranium content in rocks. In particular, about 80 % of the analysed waters presents uranium concentrations lower than 15 μg/L, while the remaining 20 % shows uranium contents between 15 and 21 μg/L. As evidenced for other water samples, uranium content in waters shows a correlation with bicarbonate and calcium ions. This behaviour, common to all the analysed samples and frequently described in the literature, is definitely the dominant factor affecting uranium mobility and distribution in waters.

4.3 Tap and bottled mineral water: review from the literature data

Table 5 shows the results of a major study carried out by Cicchella et al. (2010) and Dinelli et al. (2010) relatively to the geochemical characterization of bottled mineral water. In particular, the results shown refer to the concentrations of uranium analysed in more than 150 samples. The measured concentrations range from <0.0005 to 31 μg/L, with median and mean values of 0.47 and 1.29 μg/L, respectively. The highest concentration was observed in samples from Sardinia. The authors highlight the influence of the geological context, as in that region the high concentrations of uranium may be due to leaching from Hercynian granites. Similarly, high values (11 μg/L) are reported by Cicchella et al. (2010) for water samples coming from western Alps, due to the interaction of waters with the weathered graphitic shales characterized by an abundance of U-bearing minerals.

Table 5 reports also the uranium data from Dinelli et al. (2012) resulting from an extensive study regarding major and trace elements in tap water from Italy. The database consists of 157 tap waters sampled in private or public distributing systems all over Italy between 2009 and 2010. The uranium concentrations in Italian tap water range from 0.003 to 18.1 μg/L, and the median is 0.62 μg/L. Although relatively high concentrations are found in correspondence of the Roman Comagmatic Province regions (Central Italy), only in Trentino Alto Adige a sample with concentration (18.1 μg/L) exceeding the FAO guideline value of 15 μg/L (Dinelli et al. 2012) has been detected.

A comparison between inorganic components in bottled water and Italian tap water was performed by Cidu et al. (2011). In this study 37 bottled water and 15 tap water samples supplied by the municipalities of some large Italian cities were considered, all samples collected during 2005. The results (Table 5) showed that uranium in bottled waters is found at median and maximum concentration significantly higher than the corresponding median and maximum concentration in tap waters (0.83 and 20 vs 0.39 and 1.6 μg/L, respectively). According to Cidu et al. (2011), this result is not surprising because the studied bottled waters are mineral waters that can be naturally enriched in these elements as a result of water–rock interaction processes.

5 Conclusions

This extensive review of literature data in Italy and the addition of new analyses of samples from a region where high uranium content is a well-known occurrence provided an insight into the uranium concentrations in waters with the aim of comparing them to the current standards for drinking water.

Although rocks and soils of some Italian areas, as the Vico-Cimino volcanic district and the Novazza uraniferous district, contain relatively high amounts of uranium, almost

Table 5 Uranium content in bottled mineral and tap waters, from various published studies

Region	N. samples	*U* (μg/L)						References
		Sample type	Min	Mean	Median	Max	St. dev.	
Italian territory	186	Bottled	<0.0005	1.29	–	31	2.88	Cicchella et al. (2010)
Italian territory	158	Bottled	<0.0005	–	0.47	31	–	Dinelli et al. (2010)
Italian territory	157	Tap water	0.003	1.1	0.62	18.1	1.7	Dinelli et al. (2012)
Italian territory	37	Bottled	–	–	0.83	20	–	Cidu et al. (2011)
Italian territory	15	Tap water	–	–	0.39	1.6	–	Cidu et al. (2011)

all waters considered have comparably low uranium concentrations. Uranium water content is not directly correlated to that of host rocks, but is regulated by solution equilibria controlled by the presence of bicarbonate ions and by redox conditions (pH, Eh). In some environments (e.g. where uranium is hosted in secondary minerals), the contents in rocks and in waters seem to have an inverse correlation, due to the fact that the redox conditions that favour uranium precipitation and enrichment in rocks are the same that subtract the element from aqueous solutions causing its decrease in waters. Uranium is highly soluble and even little changes in redox conditions can cause its dissolution, transport or precipitations with formation of new mineral species.

Comparing uranium concentration in Italian natural waters with the limit values set for drinking water standards, there is evidence that, at the moment, no particular concern is related to the presence of this element. The same conclusion can be drawn for tap and most bottled mineral waters, although for the latter relatively high concentrations can occur, sometimes exceeding the recommended values. This will also largely depend on the limit value that Europe, and consequently Italy, will establish in the future. If the current provisional limit suggested by WHO (30 μg/L) will be adopted, there will be no need to implement measures to reduce uranium concentrations in water supplies, as it has been in the case of arsenic. At the moment the only situation needing attention is that of Vesuvius groundwater, that, according to literature data, presents concentrations not suitable for supplying drinking water. But if stricter standards will be established, also other areas in Italy may face problems in the management of drinking water supplies. More generally, also chemical speciation in water should be tackled to assess which are the species into solution and their toxicity as far as human health is concerned. These factors, together with extensive epidemiological studies, could help overcoming the current lack of knowledge that causes uncertainties in establishing a definite limit value for uranium in drinking water and clear rules for health protection.

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