Radioprotection, vol. **46**, n° 6 (2011) S255–S263

DOI: 10.1051/radiopro/20116566s

Uranium and ^{226}Ra in drinking water supplied by Finnish waterworks

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Abstract. In a survey on uranium and ^{226}Ra concentrations in water distributed by Finnish waterworks, 548 samples were analysed that covered drinking water networks servicing 84% of all consumers. Among all consumers, the mean and median uranium concentrations at the point of use were 0.58 and 0.15 μg L^{-1}. The mean uranium concentration in drinking water produced from groundwater was 0.88 μg L^{-1}, and when surface water was used in production the mean concentration was only 0.15 μg L^{-1}. ^{226}Ra concentrations were generally below our detection limit of 0.01 Bq L^{-1}. The annual effective doses to the public arising from these radionuclides were only a few microsieverts, and the 0.1 mSv limit laid down in the EU Drinking Water Directive was not exceeded in any of the studied water distribution networks.

1. INTRODUCTION

Continuous access to clean, potable water sources is vital for humans. Water conducts several functions in the body such as the transport of nutrients, maintenance of tissue structures, regulation of body temperature and waste discharge [1]. Drinking water supplies are, however, susceptible to both chemical and microbiological contamination. Contamination can originate from human settlement discharges, industrial activities, accidental releases or water treatment processes. It can also be attributable to natural sources in the soil, as is the case with fluoride and arsenic, each affecting millions of people [2]. Naturally occurring radionuclides also belong to the latter category.

Naturally occurring radionuclides have existed in the Earth's crust since its formation about 4.6 billion years ago. Uranium and thorium have half-lives that are of the same order as the Earth's age. As these isotopes decay, they turn into new radioactive elements. The decay and formation of new isotopes continue until a stable isotope is reached. Hence, we refer to the uranium and thorium decay series. Geological processes during the history of the Earth have caused the migration of naturally occurring radionuclides from minerals to organic soil grains, sediments, water bodies and groundwater. Naturally occurring radionuclides are also found in air due to the resuspension of soil or, as in the case of radon, due to the emanation of gas. Plants take up these radionuclides, and naturally occurring radionuclides are thus also present in all food chains.

Natural sources of radiation exposure (inhalation of radon gas, external radiation from the ground and building materials, ingestion of naturally occurring radionuclides and cosmic radiation) contribute the largest share, about 80%, to the average effective doses worldwide. The ingestion of uranium and thorium series radionuclides has been assessed to account for about 5% of this. Drinking water is an important part of the human diet and hence it should be included in dietary intake estimates, especially since the concentrations of naturally occurring radionuclides in individual drinking water sources (wells, springs, ground waters) have varied by several orders of magnitude worldwide and high concentrations are occasionally encountered [3]. Uranium exhibits chemical toxicity along with radiotoxicity, which is why its concentration is often expressed as the mass concentration instead of the activity concentration. The mean annual intake of uranium (^{238}U) and ^{226}Ra from public drinking water supplies in different countries has ranged from 0.3–50 Bq (0.02–4 mg) and 0.2–20 Bq, respectively (by employing an annual water ingestion rate of 500 litres) [3]. The mean annual intake of uranium and ^{226}Ra in the diet,

including drinking water, has ranged in different countries from 2.9–57 Bq (0.2–5 mg) and 9–40 Bq, respectively [4].

The radioactivity of Finnish well waters has been comprehensively studied since the 1960s, and elevated concentrations of naturally occurring radionuclides have been observed [5–10]. However, systematic surveys aiming at assessing exposure from radioactivity in water distributed by waterworks are from the 1970s [11]. Nevertheless, naturally occurring radionuclides in all new groundwater sources commissioned by waterworks have been statutorily screened since 1993 [12]. The present exposure assessment in Finland is based on these data collected during the past four decades. The mean ^{238}U and ^{226}Ra concentration in water supplied by Finnish waterworks has been estimated as 0.015 Bq L^{-1} (1.2 μg L^{-1}) and 0.003 Bq L^{-1}, respectively [13]. In the oldest samples, the activity concentrations have been determined at the point of use (i.e. water from the tap), whereas the activity concentrations of the samples since 1993 have been measured from samples retrieved from the waterworks (and may thus represent concentrations prior to purification processes in which radionuclides may be partly removed). Furthermore, the uranium concentration has usually been estimated from the results of gross-alpha screening, and true uranium exposure to the public has therefore been difficult to assess.

In this survey, we investigated the uranium and ^{226}Ra concentration in drinking water samples collected from households that were connected to public water distribution networks. In addition, the daily water ingestion rates were determined via a questionnaire. The primary aim of this survey was to investigate the relationship between the uranium concentration in drinking water and the concentration in hair and urine. The results relating to the main aim have already been presented elsewhere [14]. This paper reports our effort in assessing uranium and ^{226}Ra exposure to the public from public water supplies by weighting the measured concentrations by the number of consumers connected to these waterworks and taking the varying water consumption rates into consideration. Uranium and ^{226}Ra are the most interesting radionuclides in the present EU legislation, since radon and its progeny were excluded from the Drinking Water Directive which was laid down in 1998.

1.1 Uranium and radium in groundwater

Groundwater is formed as melt water from snow and rainwater percolate through the soil grains until they reach the zone where all soil pores are saturated with water. Chemical and biochemical reactions take place during this process and change the water composition. Depending on factors such as rainwater chemistry, soil chemistry, soil composition and climate, varying amounts of ions and soluble organic carbon are dissolved in groundwater from the soil. The biological decomposition of plant debris produces carbon dioxide, which dissolves in groundwater and forms bicarbonate ions. A positive correlation between bicarbonate and uranium concentration in groundwater has been observed [15]. Oxidation-reduction reactions involving organic matter and metal oxides also increase the solubility of uranium in groundwater [16]. The solubility of radium increases as a function of increasing water salinity [17].

Natural uranium is a mixture of three isotopes, ^{238}U, ^{235}U and ^{234}U, which all decay by alpha emission. More than 99% of the mass of natural uranium consists of the ^{238}U isotope, and only 0.72% and 0.0055% of ^{235}U and ^{234}U isotopes, respectively. The ^{234}U isotope is part of the uranium (^{238}U) decays series, and since the decay series is in radioactive equilibrium, its activity in the global inventory is the same as that of ^{238}U. However, in natural waters there is generally excess activity of ^{234}U [18]. The mechanism resulting in excess ^{234}U has been explained by the alpha-recoil ^{234}Th atoms that are ejected into the groundwater [19]. Another mechanism resulting in the observed excess is chemical: recoil nuclides ejected from mineral grains are embedded into adjacent grains and are hence more prone to chemical leaching than the parent isotopes [20].

In Finnish groundwaters, the activity ratio ^{234}U/^{238}U has varied from 0.76 to 4.67 [6]. Nowadays, the uranium concentration in drinking water samples is usually determined by common mass spectrometers

and only the mass concentration of uranium is recorded. Therefore, information on the ^{234}U activity concentration is often lacking as was the case in this study.

1.2 Health effects of uranium

The toxicity of uranium has been well established for almost two centuries. In 1885, Chittenden and Hart referred to experiments carried out as early as in 1824 in which the administration of uranyl salts caused nausea and death in laboratory animals. In their own dose-response experiments, continuous daily doses of uranium (3–4 mg kg^{-1} body weight) administered orally to a dog showed little effect on protein levels in the dog's urine. As the dosage was increased to 8–24 mg kg^{-1}, a cumulative effect was noted and both the protein and sugar levels in the urine significantly increased. In a set of experiments performed on rabbits (body weight not given), uranium administered both orally and subcutaneously caused effects on several endpoints, including nephropathy and death [21]. Based on these findings, uranium nephritis was used as an experimental condition in studying renal functions in the early 1900s e.g. [22]. At that time, more detailed histological studies on uranium poisoning also became available [23–25].

In its 1950 recommendations, the International Commission on Radiological Protection (ICRP) stated that "as the specific activity of natural uranium is so low, it is considered that the hazards arising from its use are mainly chemical" [26]. Some studies on the toxicity of uranium were conducted during the following decades, and the mechanisms by which uranium reduces glomerular filtration rates were explained e.g. [27]. The ICRP also developed an age-specific biokinetic model for uranium [28].

During the Persian Gulf War in 1991, a vast number of people were wounded by fragments of ammunition containing depleted uranium or were exposed to dust generated by the explosion of this ammunition (in depleted uranium the ^{235}U content has been reduced to about 0.2%). In the following years, many of them developed a multi-symptom illness now known as the "Gulf War Syndrome" e.g. [29]. Consequently, considerable resources were allocated to studying the effects of depleted uranium in the human body, and numerous research papers on the health effects of (depleted) uranium are now available. Research on solid uranium dust and its transfer in the environment also led scientists to investigate the relationship between uranium speciation and toxicity e.g. [30]. The World Health Organization (WHO) recently provided a review of biokinetic, toxicological and epidemiological studies on uranium in drinking water and confirmed the long-ago observation that renal dysfunction is the primary health effect of uranium [31].

1.3 Maximum concentrations permissible in drinking water

In the EU, there is presently no common maximum permissible concentration of uranium or ^{226}Ra in drinking water. Instead, the EU Drinking Water Directive sets a reference dose (total indicative dose) of 0.1 mSv per year, which corresponds to the annual dose received from all the natural and artificial radionuclides in water except for tritium, ^{40}K and radon and its progeny [32]. The monitoring methods and frequencies of the total indicative dose have not been specified and hence its compliance is not enforced. Nevertheless, assuming a 500 litre annual water ingestion rate, the 0.1 mSv limit corresponds to 4.5 Bq L^{-1} of ^{238}U, 4.1 Bq L^{-1} of ^{234}U or 0.71 Bq L^{-1} of ^{226}Ra if these radionuclides were to individually occur in water (which never occurs in practise). By using the typical activity ratio of ^{234}U/^{238}U for groundwater (1–1.5), a ^{238}U mass concentration of about 150 μg L^{-1} would cause the 0.1 mSv effective dose if only uranium isotopes were present in the water.

Nevertheless, Germany has set a 2 μg L^{-1} limit for uranium in water used for preparing infant formulae. The German Federal Environmental Agency (UBA), which is responsible for assessing German drinking water quality, recommends compliance with a guideline value of 10 μg L^{-1} for uranium in drinking water [33]. In Canada, a guideline value of 20 μg L^{-1} is given for uranium and 0.5 Bq L^{-1} for ^{226}Ra [34]. The US Environmental Protection Agency has issued a maximum contaminant level of 30 μg L^{-1} for uranium and 0.2 Bq L^{-1} for ^{226}Ra [35]. In Australia, the National

Health and Medical Research Council has set a guideline value of 20 μg L^{-1} for uranium, and the overall radioactivity of drinking water should be limited to less than 1 mSv per year assuming a 730 litre annual water ingestion rate [36]. However, these guideline values are now under revision, and a 15 μg L^{-1} limit has been proposed for uranium.

The World Health Organisation (WHO) has given a provisional guideline value for uranium in drinking water which is 15 μg L^{-1} [37]. WHO will continue to review uranium under a programme of rolling revision of the Guidelines for Drinking Water Quality. In the latest revision, a new provisional guideline value of 30 μg L^{-1} has been presented [31].

2. EXPOSURE ASSESSMENT

2.1 Activity concentration of uranium and ^{226}Ra in water supplied by waterworks

Water samples for this survey were collected in 2005–2006 during a research project gathering information on public exposure to uranium that included sampling of water, urine and hair [14]. This paper is a reanalysis of the data, which have been supplemented with information on the ^{226}Ra concentration in the samples and with a more detailed number of customers using water from waterworks. In the survey, uranium and ^{226}Ra were determined from 548 samples of tap water collected at the point of use (i.e. from household taps). The sampling covered 204 municipalities out of the 431 in 2006.

Uranium concentrations were determined by ICP-MS (Elan-6000, Perkin-Elmer-Sciex, Thornhill, Canada) at Ramboll Analytics Ltd (earlier Paavo Ristola Consulting Engineers Ltd). ^{226}Ra concentrations were determined by liquid scintillation spectrometry [38]. The uncertainty of the uranium concentrations was 15% and the minimum detectable concentration (MDC) 0.010 μg L^{-1}. The MDC of ^{226}Ra was 0.01 Bq L^{-1}.

The most recent information available on the number of connections (and consumers) to waterworks is from 1999, and this was employed in weighting of the data [39]. If more than one water sample was collected from a waterworks, the median concentration was calculated and used in the statistical analysis. In 30 samples, the uranium concentration was below the MDC, in which case a value of 0.5 $\times$ MDC was used in the statistical analysis. At large waterworks, there may be more than one water abstraction plant and both groundwater and surface water are processed. In these cases, the water was classified according to the main type of water used. The statistical analyses were carried out with Stata/IC 10.1 (Stata Corp, TX, USA).

The mean and median uranium concentration in tap water produced from surface water was 0.15 and 0.099 μg L^{-1}, respectively (Table 1). If (artificial) groundwater was used for water production, the uranium concentration was higher, with mean and median values of 0.88 and 0.15 μg L^{-1}, respectively. The standard deviation and range of values were also significantly larger when (artificial) groundwater was used.

The data on the number of consumers connected to waterworks were collected in 1999. In that year, the total number of people using water from waterworks was 4.6 million and the proportions of surface water and (artificial) ground water used were 41 and 59%, respectively [39]. Our water samples thus covered waterworks providing water for 3.9 million consumers, or 84% of the consumers in 1999. The respective proportions in 2006 when this survey was carried out were 40 and 60%, and the total number of consumers was 4.8 million [40]. In our data set, 39% of the determined concentrations represented ground water sources. Therefore, weighting was performed on the data accordingly in order to determine a representative uranium concentration for all consumers.

The highest uranium concentration in a single sample, 25 μg L^{-1}, was found in a water sample from Riihimäki waterworks. The two other samples from the municipality contained 0.89 and 2.6 μg L^{-1} of uranium (Table 2). At Riihimäki, two separate groundwater abstraction plants were in operation and both plants were feeding water into the water mains. The clear difference in the three measured

Table 1. Summary statistics for the uranium concentration in drinking water from waterworks. The mean, median, SD and centile values have been weighted by the number of consumers.

	All consumers	(Artificial) groundwater	Surface water
N samples	548	396	152
N consumers	4.80 million	2.38 million	1.50 million
Arithmetic mean [μg L^{-1}]	0.58	0.88	0.15
Median [μg L^{-1}]	0.15	0.15	0.099
SD [μg L^{-1}]	1.5	1.5	0.13
10% centile [μg L^{-1}]	0.021	0.020	0.033
90% centile [μg L^{-1}]	1.7	2.4	0.16
Min [μg L^{-1}]	<0.010	<0.010	<0.010
Max [μg L^{-1}]	10	10	0.92

Table 2. Summary statistics for the samples collected at the largest waterworks and at Riihimäki, where the highest uranium concentration was recorded. The proportion of groundwater produced is based on the 2006 figures supplied by the waterworks.

Municipality	N cons. [$\times 10^3$]	N samples	N abstr. plants	Ground-water use [%]	Mean [μg L^{-1}]	Median [μg L^{-1}]	SD [μg L^{-1}]	Range [μg L^{-1}]
Helsinki	550	45	1	0	0.23	0.16	0.32	0.05–2.0
Espoo	203	36	5	2.5	0.12	0.10	0.09	0.02–0.3
Tampere	186	11	10	30	0.14	0.17	0.12	< 0.01–0.4
Vantaa	166	19	3	6	0.20	0.15	0.13	0.05–0.5
Turku	165	16	3	3.5	0.15	0.09	0.14	0.06–0.6
Oulu	117	8	6	5.2	<0.01	<0.01		<0.01–0.02
Lahti	95	8	7	100	2.7	2.4	1.0	1.3–4.0
Riihimäki	25	3	2	100	9.5	2.6	13	0.9–25

concentrations may be due to variation between the plants in the water mixing ratios. However, a more likely explanation is the structure of water mains and pipes. The inner surfaces of the pipes are always covered with varying amounts of deposits and biofilms. Ferric deposits, in particular, may adsorb a range of metallic species that in some circumstances might be released into the water. Variation between samples from the same water network was also observed in several large municipalities such as Helsinki, Espoo, Tampere, Vantaa and Turku.

The summary statistics presented in Table 1 were calculated based on median concentrations at individual each waterworks. If arithmetic mean concentrations had been used, the mean (and median) concentration for all consumers, (artificial) groundwater consumers and surface water consumers would have been 0.66 (0.20), 0.97 (0.20) and 0.19 (0.15) μg L^{-1}, respectively.

^{226}Ra concentrations were generally below the MDC (0.01 Bq L^{-1}). Only in twelve water samples from nine separate waterworks could the concentration be measured, and the values ranged from 0.010 to 0.035 Bq L^{-1}. When employing 0.5 $\times$ MDC for samples that were below the limit of detection, a median concentration could only be calculated for four water works servicing water to about 47 000 consumers. The median values in these networks ranged from 0.016–0.021 Bq L^{-1}.

2.2 Water ingestion rates

Tap water is directly ingested as drinking water, coffee, tea or cordial, but also indirectly in foods such as soups, porridges and purées. Packaged waters, soft drinks and beers are also normally prepared from water distributed by waterworks, although often located in another municipality or even abroad. Most foodstuffs additionally contain various amounts of biological water, most notably fruits, vegetables and milk. In the questionnaire sent during this survey, the respondents were asked to estimate their daily

Table 3. Summary statistics for daily tap water ingestion rates among Finns in the age group 18–65 years. The values reported for both genders have been weighted according to gender among the age group.

	All	Men	Women
N replies	548	235	313
Arithmetic mean [L d^{-1}]	1.95	1.90	2.00
Median [L d^{-1}]	1.90	1.85	1.95
SD [L d^{-1}]	0.81	0.82	0.80
10% centile [L d^{-1}]	1.0	0.90	1.1
90% centile [L d^{-1}]	3.1	2.9	3.1
Min [L d^{-1}]	0	0	0
Max [L d^{-1}]	5.6	5.3	5.6

beverage consumption (water and other beverages) and the amount of water ingested indirectly in food. The ingestion of biological water was not taken into account. According to the replies, the daily mean tap water ingestion rates among adult men and women were 1.90 and 2.00 litres, respectively (Table 3). The response rate among women was higher, and hence gender weighting was applied for all consumers based on the data from the population census of 2006.

The daily water need covering the majority of men and women has been assessed as 3.7 and 2.7 litres, respectively. Physical exercise and heat stress may increase the need up to 6 litres [1]. This could partly explain why water ingestion rates varied substantially within the studied group. Data are also available from the National FINDIET 2007 survey carried out in Finland [41]. Daily water ingestion rates from drinking water, coffee, tea and cordial among men and women in the age group 25–64 years were 1.36 and 1.47 litres, respectively. Since the indirect use of water was not included in these figures, the 1.9–2.0 litres quantified by our survey seems valid.

The International Commission for Radiological Protection (ICRP) has adopted reference values for water intake among adult men and women of 2.6 L day^{-1} and 1.96 L day^{-1}, respectively [42]. However, these values include biological water, and such a difference between genders was not obtained from our data. The daily mean ingestion rate of "community water" among the population of the US has been comprehensively studied and has been estimated at 0.926 litres per capita, including those who do not consume community water. The daily water ingestion rate was found to be slightly higher among adult men ($\sim$1.16 litres) than among women ($\sim$1.04 litres), except for lactating women, among whom the rate was the highest, 1.38 litres [43]. These values are significantly lower than those obtained in our study. Hence, it is evident that dietary intake assessments require data on country-specific ingestion rates.

2.3 Magnitude of exposure

The annual intake of uranium from drinking water supplied by waterworks was first assessed by dividing daily ingestion rates into 22 categories (each covering a 0.25 L d^{-1} range). The number of consumers belonging to these categories was then calculated for each of the 204 waterworks. Hence, we obtained 4488 groups, whose annual uranium exposure was calculated by multiplying the uranium concentration in the water by the respective water ingestion rate. Weighting according to the proportion of the water source was carried out (Figure 1).

The source-weighted median value for the annual intake of uranium from water was 87 μg among all consumers. The median values among consumers of surface water and (artificial) ground water were very similar, being 80 and 100 μg, respectively. However, the mean annual intake values ranged from 100 μg among consumers of surface water to 620 μg among consumers of (artificial) ground water, being 410 μg among all consumers. Again, if arithmetic mean concentrations had been used to represent the water of an individual network, the median intake among all consumers would have been 110 μg. The difference in the distribution of concentrations for surface water and (artificial) groundwater was evident when examining the most highly exposed groups. Among consumers of surface water, the most highly

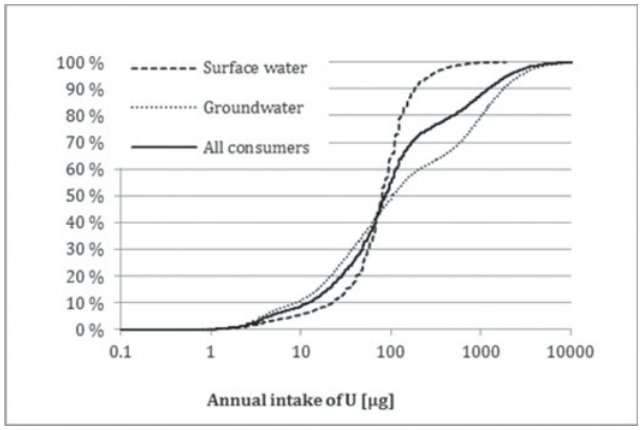

Figure 1. Cumulative distribution of annual intake of uranium from water provided by waterworks among the age group 18–65 years.

exposed one per cent (19 000 consumers) was determined to ingest 0.6–1.8 mg of uranium annually. This range among consumers of groundwater (29000 people) was 5.7–21 mg.

When assessing effective doses to consumers, we must also consider the ^{234}U isotope, which was not investigated in this survey. However, data on 1530 alpha spectrometric determinations of uranium isotopes are available in the database of the Radiation and Nuclear Safety Authority (STUK). Most of these measurements were performed on water from drilled wells, i.e. from bedrock aquifers. Bedrock aquifers are abundant in Finland, but their storage capacities and yields are typically very low and hence they are not suitable as water sources for waterworks. The number of measurements performed on public water supplies in which the uranium concentration ranged from 0.016 to 22 μg L^{-1} was 223. The mean and median activity ratios of ^{234}U/^{238}U among these samples were 1.3 and 1.1 and the range 0.8–3.4. By using the median activity ratio and dose conversion coefficients from ICRP publication 72, the median annual effective dose among all users was determined to be only 0.1 μSv [44]. According to this assessment, one per cent of all consumers (48000) receive an effective dose exceeding 5.5 μSv from uranium isotopes in water.

Only at four waterworks servicing a total of 47 000 customers could the median value of ^{226}Ra be calculated. Among this group, the annual median ^{226}Ra exposure was 11 Bq, or 3 μSv expressed as the annual median effective dose. The exposure among the most highly exposed group (4700 customers) was assessed as 19–43 Bq, or 5.2–12 μSv. Our assessment, however, covered only one per cent of all consumers, and hence the previously estimated mean ^{226}Ra concentration of 0.003 Bq L^{-1} remains our best available estimate on the general levels.

3. CONCLUSIONS

According to this survey, the mean uranium concentration in water supplied by Finnish waterworks was 0.58 μg L^{-1}, or expressed as an activity concentration for ^{238}U, 7.2 mBq L^{-1}. This value is only half of the previously assessed concentration of 15 mBq L^{-1}, and hence a better insight was gained into the uranium exposure of Finns. It is also noteworthy that although the uranium concentrations in Finnish wells drilled in bedrock are among the highest in the world, the uranium concentration in water distributed by the waterworks is generally low, even if groundwater is used in the production. One water sample contained 25 μg L^{-1} of uranium, which is close to the 30 μg L^{-1} maximum level currently proposed by WHO. However, the other two samples retrieved from this waterworks did not support a generally elevated uranium concentration in water. There is an ongoing project at STUK in

which the adsorption/desorption of radionuclides onto/from water pipe precipitates and biofilms is being investigated. This will hopefully explain the varying concentrations measured at several waterworks. In addition, ^{226}Ra concentrations were very low and generally below our MDC of 0.01 Bq L^{-1}. Therefore, the intake assessment from 2001 remains valid.

The European Commission decided against revising the Drinking Water Directive in early 2011 [46]. Hence, it is now the decision of the member countries whether to include uranium in the list of monitored parameters, and what the maximum permissible concentration should be. A positive finding of this survey was that the measured uranium and ^{226}Ra concentrations all comply with the total indicative dose of 0.1 mSv presently stated in the directive. Furthermore, if a new maximum permissible uranium concentration of 15–30 μg L^{-1} were to come into effect in Finland, all waterworks studied in the survey would also comply with this.

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