

Uranium in urine and hair of occupationally unexposed Finnish people

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Abstract. Uranium concentrations in the urine and hair of the occupationally unexposed Finnish population were determined using inductively coupled plasma mass spectrometry (ICP-MS). The mean concentrations of uranium in urine and hair were 0.016 µg/l and 0.216 µg/g, respectively. The mean uranium concentration in hair of the Finns was from three- to fifteen-fold higher than the values reported in the literature, while the mean uranium concentration in urine was similar to those measured elsewhere in Europe. The observed large variation in the uranium concentrations in hair and urine can be explained by the variation in the uranium concentration in drinking water. In Finland the concentrations of natural uranium in drinking water have been observed to vary markedly depending on the location. Exceptionally high concentrations have been measured in private drilled wells in the granite areas of Southern Finland. A part of the measured uranium content in hair may also be due to external contamination, which may remain in the hair after the washing procedure.

KEYWORDS: *Uranium, urine, hair, exposure*

INTRODUCTION

Uranium is a primordial radioactive element ubiquitously present in the Earth's crust. Natural uranium consists of a mixture of three radioactive isotopes: ^{238}U (99.2745% by mass), ^{235}U (0.7200%) and ^{234}U (0.0054%). All three isotopes are α emitters with half-lives of 4.468×10^9 y for ^{238}U , 7.038×10^8 y for ^{235}U and 2.455×10^5 y for ^{234}U . In Finland the concentrations of natural uranium in drinking water have been studied since 1960s and they have been observed to vary markedly depending on the location (Asikainen 1981, Kahlos 1969, Salonen 2002, Vesterbacka et al. 2004, 2005). Exceptionally high concentrations have been measured in private drilled wells in the granite areas of Southern Finland, where the arithmetic mean concentration exceeds 150 µg/l in several municipalities according to the database of Radiation and Nuclear Safety Authority. Consequently, high concentrations of natural uranium have been observed in the urine and hair samples of people using water from their own drilled wells. In an epidemiological study on the renal effects of uranium, the uranium concentration in the urine and hair of people using water from drilled wells in Southern Finland varied from 0.001 to 5.65 µg/l and from 0.0065 to 250 µg/g, respectively (Kurttio 2002). The mean uranium concentration in the urine of the study group was as high as 0.424 µg/l. Even higher uranium concentrations of 47 µg/l and 820 µg/g, respectively, in the urine and hair of Finns have been reported by Muikku et al. (2007). Although studies have been carried out on this subject in Finland, the average uranium concentrations in urine and hair of occupationally unexposed Finnish people were not known.

This paper summarizes results from a study on the levels of uranium in the urine and hair of occupationally unexposed Finns by Muikku et al. (2009).

MATERIALS AND METHODS

The Finnish Population Register Centre randomly sampled 4000 Finns from the working population (ages from 18 to 65 years) who formed two groups based on water consumption. The first sampling group comprised 2000 Finns who, according to the register, used water from the public waterworks as household water. This subgroup represented approximately 50% of the Finnish population. The second group consisted of 2000 persons using private water supplies, representing 10% of the Finnish population. The users of private water supplies represented two areas: half of them lived in Southern Finland (subgroup 2), where exceptionally high uranium concentrations have been measured in private wells, and the rest represented other parts of Finland (subgroup 3). Altogether, 1002 volunteers collected water and overnight urine samples. Fifty-one persons were excluded because the main source of their household water could not be determined. The age range of the final study group of 951 persons (443 females, 508 males) was between 18 and 66 years. The average age of the study population was 48.8 years (median 51 years). A questionnaire was prepared to collect information on the usage of household water, daily beverage consumption (water and other beverages) as well as residential history.

Drinking water was sampled directly from the tap, urine samples were collected over night and hair samples were collected by cutting the hair close to the scalp near the ears from an area of about 1 cm². For the uranium analysis, 12 cm of the hair sample from nearest the scalp was used. As the average growth rate of hair is approximately 12-18 cm per year (Ho 2001), the hair samples represented an exposure of one year or a shorter period. The hair samples were cut into 1 cm pieces and washed according to the IAEA standard procedure (Chatt 1985) before the analysis using ICP-MS (Elan-6000, Perkin-Elmer-Sciex, Thornhill, Canada). The creatinine concentrations of urine samples determined by United Laboratories Ltd in Helsinki were used to normalize the urine results. According to ICRP Publication 89 (ICRP 2002), the average daily creatinine excretion is 1.7 g d⁻¹ for males and 1.0 g d⁻¹ for females. The measurement uncertainty and detection limit for uranium in water and urine samples were 15% and 0.010 µg/l, respectively. A more detailed description of the methods is found in the publication by Muikku et al. (2009).

RESULTS

The population weighted mean concentration of ²³⁸U in drinking water estimated in this study, 1.25 µg/l, is over ten times higher than the worldwide average ²³⁸U concentration of 1 mBq/kg (0.081 µg/l) in drinking water reported by UNSCEAR (2000). The uranium concentration in water from the public water supplies varied from < 0.01 µg/l to 25 µg/l. The arithmetic mean and median were 0.720 and 0.148 µg/l, respectively. The minimum uranium concentration in the household water from private water sources was below the detection limit of 0.01 µg/l. The maximum concentration, 1770 µg/l, was found in a drilled well in Southern Finland. The arithmetic mean and median for the water samples of the private water users in Southern Finland (subgroup2) were 18.1 and 1.13 µg/l, respectively. In subgroup 3 (the private water users in other parts of Finland) the corresponding values were 2.54 and 0.158 µg/l, respectively.

The mean ²³⁸U concentration in the urine of the Finnish occupationally unexposed working population (0.016 µg/l) determined in the present study is similar to values measured elsewhere (e.g. Beyer et al. 1993, Byrne and Benedik 1991, Dang et al. 1992, Galletti et al. 2003, Ting et al. 1999, Tolmachev et al. 2006). Over 70% of the urine results were below the detection limit, 0.010 µg/l (Table 1). The variation in the concentrations was high, ranging from <0.01 µg/l to 3.7 µg/l. At low uranium concentrations in household water there was a poor correlation between the concentration of uranium in water and in urine, whereas when the uranium concentration in water was high, the correlation was rather good.

Food is on average more important source of uranium than drinking water and may affect the uranium concentration in urine at low levels.

Table 1. The number of study subjects in the groups formed according to the ^{238}U concentration in urine.

Sampling group	^{238}U in urine ($\mu\text{g}/\text{l}$)				^{238}U in urine (% of all)			
	< 0.01	0.01-0.10	0.10-1.0	≥ 1	< 0.01	0.01-0.10	0.10-1.0	≥ 1
Communal	429	113	5	1	45	12	1	0
Private (South)	125	74	14	2	13	8	1	0
Private (other)	132	48	8	-	14	5	1	-
All	686	235	27	3	72	25	3	0

The minimum ^{238}U concentration in hair of the study population was below $0.0005 \mu\text{g}/\text{g}$. The maximum value of $140 \mu\text{g}/\text{g}$ was determined from a hair sample taken from the person using drinking water with the highest recorded uranium concentration. The mean ^{238}U concentration in the hair samples, $0.216 \mu\text{g}/\text{g}$, was from three- to fifteen-fold higher than the values reported in the literature (e.g. Akamine et al. 2007, Byrne and Benedik 1991, Gonnén et al. 2000, Imahori et al. 1979, Rodushkin and Axelsson 2000). This can be partly explained by the higher than average uranium concentration in drinking water. A good correlation between the concentrations in water and in hair was observed. Although the IAEA standard washing procedure was used, a part of the measured uranium content in hair may be due to external contamination. No tendency for higher uranium concentrations in older participants was observed, as seen in the study by Akamine et al. (2007).

Taking into account the water consumption data collected via the questionnaires and the analysed uranium concentrations in water, the population weighted mean daily intake of uranium via water in food and fluids has been estimated to be $2.49 \mu\text{g}/\text{d}$ (median $0.29 \mu\text{g}/\text{d}$) for the occupationally unexposed Finnish working population. The mean committed effective dose from ^{238}U via water, $0.51 \mu\text{Sv}/\text{y}$ (median $0.06 \mu\text{Sv}/\text{y}$), is double the dose caused by the average intake of ^{238}U in the whole diet according to UNSCEAR (2000). The dose conversion coefficient of $4.5 \times 10^{-8} \text{ Sv}/\text{Bq}$ (ICRP 1995) has been used in the calculations. Only the dose caused by the isotope ^{238}U (abundance 99.2745% by mass) has been estimated because the activity ratio $^{234}\text{U}/^{238}\text{U}$ has been observed to vary in the water of Finnish wells, the average ratio being between 1 and 2 (Salonen 2002). Thus, the total uranium exposure is twice the exposure of ^{238}U at the minimum.

The annual committed effective doses ($\mu\text{Sv}/\text{y}$) for the different study groups formed according to the water source in use are presented in Table 2. The mean annual committed effective dose from ^{238}U in water is 0.68 and $10 \mu\text{Sv}/\text{y}$ for the users of dug wells (or springs) and drilled wells, respectively. The median values for the groups above are accordingly 0.08 and $0.86 \mu\text{Sv}/\text{y}$. The highest estimated dose is about $880 \mu\text{Sv}/\text{y}$. The mean annual doses are $0.08 \mu\text{Sv}/\text{y}$ (median $0.04 \mu\text{Sv}/\text{y}$) and $0.36 \mu\text{Sv}/\text{y}$ (median $0.08 \mu\text{Sv}/\text{y}$) for those groups using water from communal water supplies that have surface water and groundwater as the raw water sources, respectively. The significant difference between the mean and median doses reflects the wide range of annual doses, which can be explained by the large variation in the concentration of ^{238}U in drinking water. In previous studies the mean annual effective doses from

^{238}U in drinking water in Finland have been estimated to be 0.5, 0.5 and 8 $\mu\text{Sv}/\text{y}$ for people using water from the public waterworks, private dug wells and private drilled wells, respectively (Mäkeläinen 2001, Vesterbacka 2005). The median doses for the users of private dug and drilled wells were 0.1 and 0.7 $\mu\text{Sv}/\text{y}$ (Vesterbacka 2005). The findings of this report support the previous estimations for users of private water sources. The dose estimations presented in this study for the users of different water sources are not population weighted values, but are doses estimated for particular subgroups. More profound estimation of uranium and ^{226}Ra exposure to the public from public water supplies has been presented in Turtiainen et al. (2011).

Table 2. The statistical parameters describing the ^{238}U concentrations in drinking water ($\mu\text{g}/\text{l}$) and hair ($\mu\text{g}/\text{g}$), the water consumption rate (l/d), the intake of ^{238}U via water ($\mu\text{g}/\text{d}$) and the annual committed effective dose ($\mu\text{Sv}/\text{y}$) in the different study groups formed according to the water source in use. The number of study subjects in each group is provided in brackets.

^{238}U	Mean	Median	25th percentile	75th percentile	Minimum	Maximum
Drilled wells						
Drinking water ($\mu\text{g}/\text{l}$) (167)	23.8	2.00	0.46	7.60	< 0.01	1770
Consumption rate (l/d)	2.02	1.90	1.40	2.45	0.40	5.30
Intake ($\mu\text{g}/\text{d}$)	51.1	4.24	0.86	14.0	< 0.01	4340
Dose ($\mu\text{Sv}/\text{y}$)	10.4	0.86	0.18	2.86	< 0.01	884
Hair (153)	2.14	0.215	0.045	0.850	0.001	140
Dug wells and springs						
Drinking water ($\mu\text{g}/\text{l}$) (226)	1.68	0.20	0.09	0.82	< 0.01	102
Consumption rate (l/d)	2.04	1.89	1.50	2.50	0.30	7.95
Intake ($\mu\text{g}/\text{d}$)	3.33	0.40	0.16	1.44	< 0.01	240
Dose ($\mu\text{Sv}/\text{y}$)	0.68	0.08	0.03	0.29	< 0.01	48.8
Hair (204)	0.133	0.034	0.014	0.096	< 0.001	3.44
Communal water, surface						
Drinking water ($\mu\text{g}/\text{l}$) (152)	0.22	0.11	0.06	0.19	< 0.01	2.81
Consumption rate (l/d)	1.83	1.73	1.24	2.34	0.35	5.60
Intake ($\mu\text{g}/\text{d}$)	0.41	0.19	0.09	0.39	< 0.01	7.82
Dose ($\mu\text{Sv}/\text{y}$)	0.08	0.04	0.02	0.08	< 0.01	1.6
Hair (138)	0.108	0.040	0.017	0.079	0.002	5.21
Communal water, ground						
Drinking water ($\mu\text{g}/\text{l}$) (394)	0.91	0.20	0.05	1.16	< 0.01	25.0
Consumption rate (l/d)	2.02	1.91	1.48	2.45	0.1	5.0
Intake ($\mu\text{g}/\text{d}$)	1.75	0.40	0.09	1.87	< 0.01	39.5
Dose ($\mu\text{Sv}/\text{y}$)	0.36	0.08	0.02	0.38	< 0.01	8.1
Hair (345)	0.203	0.051	0.013	0.216	< 0.001	3.02

DISCUSSION

The mean concentrations of uranium in the drinking water, urine and hair of occupationally unexposed Finns were presented in this paper. The large variation observed in the uranium concentrations in hair and urine can mostly be explained by the large variation in the uranium concentration in drinking water. The uranium concentration in private wells as well as water consumption rates may vary considerably over time, and therefore an overnight urine sample is not necessarily a good indicator of long-term uranium exposure. The uranium concentration in hair reflects the natural uranium contamination caused by continuous use of drinking water better than the uranium concentration in a spot urine sample. However, a part of the measured uranium content in hair may be due to external contamination, which may remain in the hair after the washing procedure. Occupational exposure can be reliably distinguished from the natural background only by determining the isotope ratio in urine samples.

The present WHO guideline for uranium concentration in drinking water, 30 µg/l (WHO 2011) was exceeded in 2.4% of the cases (23 samples). All of them were from private water sources.

REFERENCES

- Akamine, A.U., Duchen Silva, M.A., Saiki, M., Vasconcellos, M.B.A., de Andrade, S.L., Fulfaro, R. (2007). Determination of uranium in human head hair of a Brazilian population group by epithermal neutron activation analysis, *J Radioanal Nucl Chem*, 271, 607-609
- Asikainen, M., (1981). State of disequilibrium between ^{238}U , ^{234}U , ^{226}Ra and ^{222}Rn in groundwater from bedrock. *Geochimica et Cosmochimica Acta*, 45(2), 201-206.
- Beyer, D., Buehl, R., Pilwat, G. (1993). Normal concentration of uranium in urine. *Health Phys*, 64, 321
- Byrne, A.R., Benedik, L. (1991). Uranium content of blood, urine and hair of exposed and non-exposed persons determined by radiochemical neutron activation analysis, with emphasis on quality control. *Sci Total Environ*, 107, 143-157
- Chatt, A., Sajjad, M., DeSilva, K.N., Secord, C.A. (1985). Human scalp hair as an epidemiologic monitor of environmental exposure to elemental pollutants, IAEA-TECDOC-330, International Atomic Energy Agency, Vienna, 33-49
- Dang, H.S., Pullat, V.R., Pillai, K.C. (1992). Determining the normal concentration of uranium in urine and application of uranium in urine and application of the data to its biokinetics, *Health Phys*, 62, 562-566
- Galletti, M., D'Annibale, L., Pinto, V., Cremisini, C. (2003). Uranium daily intake and urinary excretion: A preliminary study in Italy, *Health Phys*, 85, 228-235
- Gonnen, R., Kol, R., Laichter, Y., Marcus, P., Halicz, L., Lorber, A., Karpas, Z. (2000). Determination of uranium in human hair by acid ingestion and FIAS-ICPMS, *J Radioanal Nucl Chem*, 243(2), 559-562
- Ho, J., Boenheim, F., Pangborn, J., Spitz, J., Smith, B. (2001) Hair Element Testing: Clinical Utility and Reliability. A referenced white paper by Great Smokies Diagnostic Laboratory, Asheville

Imahori, A., Fukushima, I., Shiobara, S., Yanagida, Y., Tomura, K. (1979) Multielement neutron activation analysis of human scalp hair: A local population survey in the Tokyo metropolitan area, *J Radioanal Chem*, 52(1), 167-180

International Commission on Radiological Protection. (1995). Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 3 Ingestion Dose Coefficients. Oxford: Pergamon Press; ICRP Publication 69

International Commission on Radiological Protection.(2002). Basic Anatomical and Physiological Data for Use in Radiological Protection: Reference Values. Oxford: Pergamon Press; ICRP Publication 89

Kahlos, H. (1969). Radon-222 concentration of some water samples determined with gamma-spectrometric measurements. Report SFL-A14 (Institute of Radiation Physics, Helsinki)

Kurttio, P., Auvinen, A., Salonen, L., Saha, H., Pekkanen, J., Mäkeläinen, I., Väisänen, S.B., Penttilä, I.M., Komulainen, H. (2002). Renal effects of uranium in drinking water, *Environ health perspectives*, Vol 110, 337-342

Muikku, M., Heikkinen, T., Puhakainen, M., Rahola, T., Salonen, L. (2007). Assessment of occupational exposure to uranium by indirect methods need information on natural background variations, *Radiat Prot Dosimetry*, 125, 492 - 495

Muikku, M., Puhakainen, M., Heikkinen, T., Ilus, T. (2009). The mean concentration of uranium in drinking water, urine, and hair of the occupationally unexposed Finnish working population. *Health Physics*, 96(6), 646-654.

Mäkeläinen, I., Huikuri, P., Salonen, L., Markkanen, M., Arvela, H. (2001) Radioactivity of drinking water in Finland - basis for quality requirements (in Finnish). STUK-A182, Edita Oyj, Helsinki

Rodushkin, I. and Axelsson, M. (2000) Application of double focusing sector field ICP-MS for multielemental characterization of human hair and nails. Part II. Study of inhabitants of northern Sweden, *Science Total Environ* 262, 21-36

Salonen, L. and Huikuri, P. (2002) Elevated Levels of Uranium Series Radionuclides in Private Water Supplies in Finland. BfS Schriften. Strahlenhygiene. High Levels of Natural Radiation and Radon Areas: Radiation Dose and Health Effects. Volume II: Poster Presentations. *General Exposure Assessment*, 24, 87-91

Ting, B.G., Paschal, D.C., Jarrett, J.M., Pirkle, J.L., Jackson, R.J., Sampson, E.J., Miller, D.T., Caudill, S.P. (1999) Uranium and thorium in urine of United States residents: Reference range concentrations, *Environ Research Section A*, 81, 45-51

Tolmachev, S., Kuwabara, J., Noguchi, H. (2006) Concentration and daily excretion of uranium in urine of Japanese, *Health Phys*, 91, 144-153

Turtiainen, T., Muikku, M., Vesterbacka, P., Heikkinen, T. (2011) Uranium and ^{226}Ra in drinking watersupplied by Finnish waterworks. Presented in The International Conference on Radioecology and Environmental Radioactivity (ICRER-2011), Hamilton, Canada, June 18-24, 2011.

UNCEAR 2000. Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation. UNSCEAR 2000 Report to the General Assembly, with scientific annexes, Vol 1: Sources; New York 2000

Vesterbacka, P., Mäkeläinen, I., Tarvainen, T., Hatakka, T., Arvela, H. (2004) Kaivoveden luonnollinen radioaktiivisuus - Otantatutkimus 2001 (in Finnish), STUK-A199

Vesterbacka, P., Mäkeläinen, I., Arvela, H. (2005) Natural radioactivity in drinking water in private wells in Finland. *Radiation Protection Dosimetry*, 113, 223-232

WHO Guidelines for Drinking-water Quality. 4th ed. Geneve: World Health Organization, 2011