

Uranium in public water supplies in Migdonia Basin, Central Macedonia, Northern Greece

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Abstract: A surface and groundwater-sampling network has been set up for a period of time, to carry out accurate measurements of uranium concentration into public water supplies of the Migdonia basin in Northern Greece. About 19 samples from special wells, springs and taps that supply drinkable water were collected, in order to apply detailed uranium measurements. The collected samples were analysed using the delayed neutron activation technique. The samples were analysed at the facilities of the Greek National Centre for Scientific Research "Demokritos", using its nuclear reactor as the neutron source and a gamma-ray spectrometer system. The results of the investigation show that uranium concentrations exist in public water supplies in a significant percentage.

Key Words: Radioelements, Uranium in Drinking Water, Public Water Supply, Delayed Neutron Activation.

INTRODUCTION

Uranium is named after the planet Uranus. Uranium is a silvery white, very dense metal and it was first discovered in the mineral called pitchblende. Uranium is not a rare mineral; it is more plentiful than silver or mercury. However, it plays an important role in the nuclear age.

Most of the Uranium found in the Earth is Uranium-238, which makes it the heaviest atom found most commonly in nature. Uranium is not found in a pure form and some tons of ore have to be processed to obtain just one gram of the element. The biggest deposits of Uranium are found in Blind River, Canada, in South Africa, Australia, France, Colorado and Utah in the US.

Martin H. Klaproth, a German chemist discovered uranium in the mineral pitchblende. Antoine Henri Becquerel recognised its radioactive properties in 1896 by the action of the fluorescent salt potassium uranyl sulfate, an image on a photographic plate covered with a light absorbing substance. Radioactivity is the most unique and useful characteristic of Uranium (Fig. 1).

The element uranium is generally found in naturally occurring minerals in one of two ionic states: U^{6+} (the uranyl "oxidised" ion) and U^{4+} (the uranous "reduced" ion). Minerals containing the uranyl ion tend to be brightly coloured (red, yellow, orange and green) and occur in oxidised portions of uranium ore deposits. Common uranyl minerals include tyuyamunite ($Ca(UO_2)_2V_2O_8 \cdot 8H_2O$), autunite ($Ca(UO_2)_2(PO_4)_2 \cdot 8-12H_2O$), torbernite

($Cu(UO_2)_2(PO_4) \cdot 8-12H_2O$) and uranophane ($(H_3O)_2Ca(UO_2)_2(SiO_4) \cdot 3H_2O$) (Smith, 1984; Hutchinson and Blackwell, 1984). Minerals containing the uranous ion are more subdued in colour, typically brown or black, and occur in reducing environments. Common uranous minerals include uraninite (UO_2), pitchblende (a crystalline variant of uraninite) and coffinite (U_4SiO_{14}) (Smith, 1984; Hutchinson and Blackwell, 1984). Uranium occurs in the minerals as one of three isotopes: U-234, U-235 and the most abundant of the isotopes, U-238 (Tatsch, 1976).

Economically recoverable uranium deposits generally fit into one of four types of deposits: *stratabound*, *solution breccia pipes*, *vein*, and *phosphatic*.

Nearly any part of waste management units at active mines may be a potential source of environmental contamination. Environmental effects resulting from uranium extraction and beneficiation are chiefly derived from two sources: mining activities, and radionuclides present in the wastes. Open pit mining activities may create environmental effects typical of surface disturbances: increased run off as well as increased erosion by wind and water. Rewatering operations conducted by surface and underground mines may create groundwater depressions that may persist after mining ceases. Potential environmental effects from in situ operations are primarily groundwater-related. Since surface disturbance is not extensive, the impacts of surface operations associated with *in situ* mining (e.g. drilling wastes, ponds) are not well documented. Mill tailings, and particularly the

radionuclides contained within, appear to be a major source of environmental impact to air, soil, surface and groundwater.

Five potential exposure pathways were identified and considered for quantitative analysis:

- 1 - inhalation and ingestion of airborne radioactive particulate;
- 2 - ingestion of contaminated foods (plant and animal) produced in areas contaminated by wind-blown tailings;
- 3 - ingestion of surface water contaminated by tailings;
- 4 - inhalation of radon and radon daughters;
- 5 - direct exposure to gamma radiation emitted from the tailings.

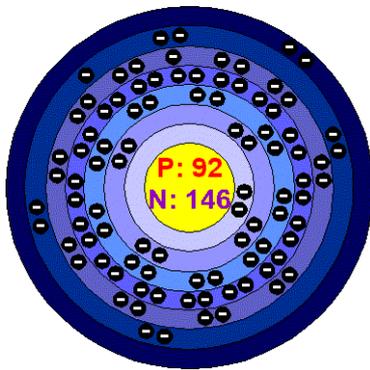


FIG. 1. Bohr model of Uranium-238 (U^{238}), Number of energy levels: 2, First energy level: 2 electrons, Second energy level: 8 electrons, Third energy level: 18 electrons, Fourth energy level: 32 electrons, Fifth energy level: 21 electrons, Sixth energy level: 9 electrons, Seventh energy level: 2 electrons.

Considering the risks associated with the ingestion of uranium, using current risk factors for the radiocarcinogenicity of uranium, and the chemical toxicity of uranium, Environmental Protection Agency (EPA) has concluded that the level proposed, 30 pCi/liter, provides an adequate margin of safety against both carcinogenic and toxic effects of uranium, and that the level should be expressed in terms of the concentration of radioactivity. Because it is related to the principal health risk, and can accommodate different levels of radioactive disequilibrium between uranium-234 and uranium-238. EPA's Office of Groundwater and Drinking Water has also examined these factors, and, on July 18, 1991, proposed that the Maximum Concentration Level (MCL) for uranium in drinking water should be set at a chemical concentration comparable to the limit on radioactivity promulgated in this regulation. Should the MCL for drinking water, as finally promulgated, provide a level of health protection different from that provided by the limit in this regulation, Environmental Protection Agency (EPA) will reconsider the limit at that time.

On the basis of the above considerations, the limit for uranium has been established as being 30 pCi/liter for this regulation.

AREA OF STUDY

The water quality problem of the Migdonia basin in Northern Greece is well known (Fig. 2). The water is supplied from boreholes in the basin sediments as well as in the metamorphic rocks and granites. The rural population that is estimated in 15000-18000 inhabitants uses this water for drinking and irrigation purposes.

The regional and local tectonics of the complex has greatly influenced the development of geomorphology of the study area. The tectonic depression of the Migdonia Basin, which holds the ecosystems of the lakes Koronia and Volvi, is controlled by at least one active fault with recent seismic activity, the catastrophic earthquake of 1978.

The stratigraphic sequence of the study area (Fig. 2) consists of meta-alpine sediments, shaly cleavage granites, Vertiskos formation gneiss, meta-ophiolithic complex of Vertiskos formation that overlies to the Kerdilios formation.

Granite and/or igneous granitoid bodies that hold rather high concentrations of natural radioelements (uranium, radon, radium, thoron, etc.) are in existence around the Migdonia basin. Through the process of natural corrosion they enrich the surface and subsurface water horizons with the above mentioned radioelements. There are also thermal waters in the basin. The area under investigation is presented on Figure 4 and its extent is about 1100 km².



FIG. 2. Geological elements of the study area (Papanikolaou 1983). 1: Meta-alpine, 2: Paionia formation, 3: Periropiki formation, 4: Mesozoic formations, 5: shaly cleavage granites, 6: Vertiskos formation gneiss, 7: meta-ophiolithic complex of Vertiskos formation, 8: Kerdilios formation limestone, 9: gneiss, amphibolites, migmatites of Kerdilios formation, 10: Rodopiki formation of Pagaios.

APPLIED METHODOLOGY

Nineteen (19) samples were totally collected from twenty (20) sampling points on two sampling dates in October and December of 1999. A more detailed sample collection took place in respect to the intense tectonic features of the Migdonia basin. A map of the area showing the sampling locations combined with topographic elements is presented in Fig. 4. Water from wells, bore-holes as well as from taps was analyzed for the determination of uranium concentration. The water samples were collected by the procedure of slowly filling each vial directly from the water source. The result of the analyses as well as the sampling points and dates are presented in Table 1.

The samples are analysed at the facilities of the Greek National Centre for Scientific Research “Demokritos”, using its nuclear reactor as the neutron source and a gamma-ray spectrometer system. The U238 is determined using the delayed neutron counting technique, which is based on the proportionality of delayed neutrons, released by fission of U²³⁸ after neutron activation in the sample. The method guarantees high accuracy (measurement error less than 1%), and high sensitivity with detection limits of less than 0,5 ppm.

RESULTS AND DISCUSSION

The results of the investigation (Table I) show that uranium concentrations in public water supplies are in a significantly high percentage. The level of 20 pCi/l is not exceeded. A percentage of 89% (17 out of 19 samples) fluctuates from 0,01 pCi/l to 3,0351 pCi/l and the rest

11% (2 out of 19 samples) varies from 5 pCi/l to 16,38 pCi/l. The two highest concentrations were detected in samples at Vagiochori (16.38 pCi/l, U6) and Gerakarou (5.092 pCi/l, U9) from within two different systems of faults with NE/SW and NW/SE direction (Fig. 3).

The high concentrations of U²³⁸ at the sites U6 and U9 can be interpreted in terms of the active tectonism in combination with the geology of the surrounding area. The granitoid bodies of the area present high values of radioelements’ concentration (Papakonstandinou *et al.*, 1996). Through the hydrologic cycle, amounts of the radioactive elements, in solution, are concentrated in deeper geological formations. Due to the active tectonism of the area, the geological formations with rather high concentration of radioactive elements come to the surface at U6 and U9 sites.

It is important to pinpoint that the site U6, where the highest concentration of U238 was detected is very close to the epicentre of several seismic events (Fig. 3).



FIG. 3. Geological and tectonic elements of the study area.

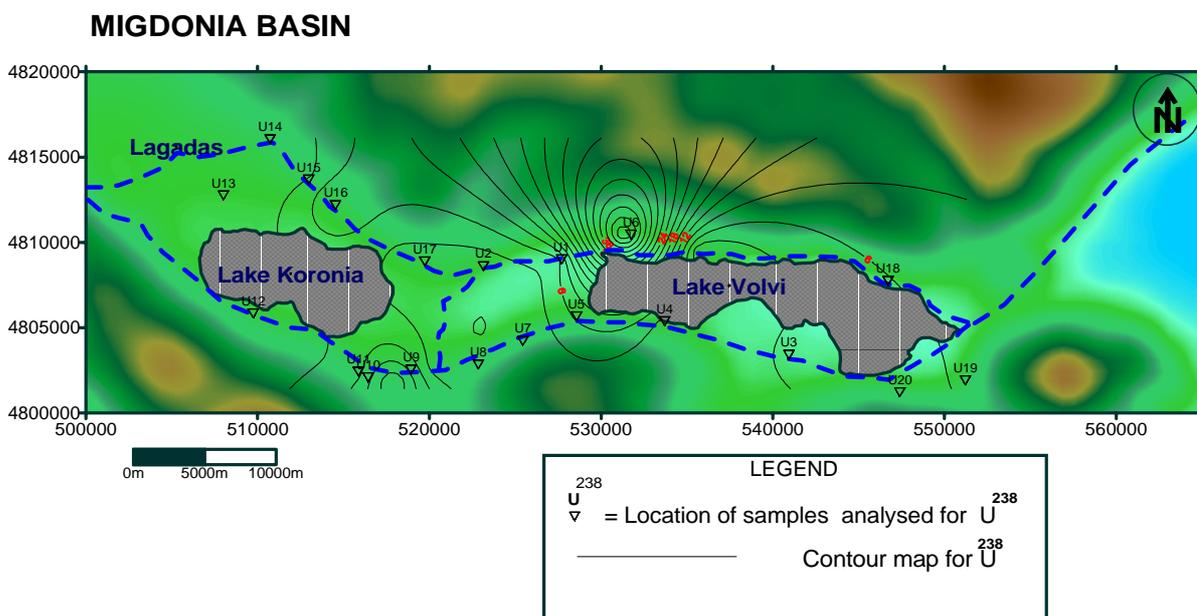


FIG. 4. U²³⁸ contour map of the area showing the sampling locations and topographic details.

TABLE 1. U²³⁸ concentrations in water samples from Migdonia basin, sampling period October - December of 1999

Village	SYMBOL	Type of water source	X	Y	URANIUM (pCi/l)
Nymfopetra	U1	Community Tap	527452	4808980	1,62
Profitis	U2	Aqueduct Tank	523033	4808610	0,365
Apolonia	U3	Church Tap	541015	4803470	1,39
Loutra Apolonia	U4	House Tap	533626	4805220	0,54
Peristeronas	U5	Community Tap	528298	4805320	1,85
Vagiochori	U6	Church Tap	531340	4810430	16,38
Stivos	U7	Square Tap	525193	4804200	0,224
Lagadikia	U8	House Tap	520387	4802740	0,315
Gerakarou	U9	House Tap	517903	4801820	5,092
Vasiloudi	U10	House Tap	515590	4802410	1,306
Ayios Vasilios	U11	Community Tap	509542	4806080	0,234
Loutra Lagada	U12	Community Tap	506896	4813800	0,171
Kolxiko	U13	Community Tap	510685	4816110	0,01
Drakontio	U14	Church Tap	512971	4814160	0,713
Analipshy	U15	Community Tap	514348	4812350	3,0351
Evagelismos	U16	Community Tap	518200	4809340	0,754
Megali Volvi	U17	Community Tap	535687	4809500	0,104
Modi	U18	Community Tap	551276	4801780	2,271
Nea Maditos	U19	Community Tap	547154	4801420	1,176

CONCLUSIONS

The results of the rather detailed investigation of public water supplies in the Migdonia basin region show that uranium concentrations are not above the accepted by the EU safety level of 30 pCi/l. Furthermore due to the intense tectonism and the geology of the area, there have been detected two high concentrations of U²³⁸ from within two different systems of faults with NE/SW and NW/SE direction that surround the Lagadas lake basin.

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