DETERMINATION OF URANIUM-BEARING SAMPLES IN TERMS OF POSSIBLE CONTAMINATION, ARIKLI URANIUM REGION, ÇANAKKALE, TURKEY

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1. INTRODUCTION

Radioactive mineralization sites and related exploration activities threaten the ecosystems of surrounding areas. Nowadays, this is an issue of concern to many countries [1, 2]. Natural and artificial factors cause the dispersion of radioactive isotopes from the sites and increase the likelihood of contamination in the surrounding area [3, 4]. It is observed that the isotopes may be liberated, transported and precipitated under certain oxidation conditions and can accumulate in different regions and this process may even lead to secondary mineralization under appropriate conditions. Over time, these accumulations and transport environments may act as secondary sources and damage human and environmental health [5–7].

The radioisotope distribution varies continuously among landscape components (e.g. rock, soil, groundwater and surface water) [3, 8, 9]. As an example, groundwater interacts with rocks, leaches radioisotopes and then influences the chemical composition of the surface water and soils [10].

Sampling strategy is one of the most important issues in contamination research. Methods which are suitable for one environment may be quite inappropriate for another. For example, the mechanisms of formation of uranium deposits vary widely and hence the geochemical signature of the deposits also vary [3]. Therefore, careful measurements and analyses are needed to understand the geological and physical structure of the area before cost-effective analyses.

Generally, for the determination of the radioactive element distribution originating from an exploration site, the easiest and cheapest step is to carry out outdoor absorbed gamma dose rate measurements (OAGDR). The distance between the measurement points should be adjusted according to the detector range and the integration time. As a result of the gamma measurement values, the target area can be reduced. For example, if erosion is dominant in the area, drawing the borders along the hilltops, gives the advantage of understanding the flow of elements. The catchments of the exploration sites should be investigated first, but measurements should also be taken in neighbouring catchments to effect comparison of the results and assess the risk of contamination.

Then, the next step is geochemical analysis of the samples. Geochemical parameters such as pH, electrical conductivity, organic matter, carbonate and clay contents, and oxidation state help to evaluate the migration patterns.

Proper GIS operations and the use of maps such as lithology, topography, soil type, land cover, erosion, hydrology, flow accumulation, run-off, etc., can significantly reduce the number of samples required. Statistical analyses such as homogeneity test of univariate distributions, bivariate scatter plots, multivariate cluster analysis and principal component analysis are used to separate populations in the dataset which may indicate various geochemical processes.
For the case study reported in the present work, which was conducted in the Arikli uranium region, a methodology was developed which included geochemical, radiometric and GIS based landscape analysis for the determination of uranium-bearing samples to assess the possible uranium-related contamination.

2. DESCRIPTION

The restricted study area, after the OAGDR orientation measurements, covers the Arikli site (Ayvaçık–Çanakkale) where uranium mineralization is present in the surrounding area. In 1959, the Turkey General Directorate of Mineral Research and Exploration Office discovered radioactive anomalies in the area during an airborne survey. In 1967, the field was surveyed by exploration ditches and from 1968 to 1982 a total 56 uranium exploration drill holes were completed [11]. Although it had been reported that the area contained favourable resources of uranium, the last drilling programme (1982) was terminated as the Government considered the resources were uneconomic [12].

According to the geochemical analyses performed on samples taken from rock dumps and exploration ditches, the phosphate–uranium association, and in some samples thorium enrichment, were detected. Although, based on these analyses, the area was considered to be favourable as regards phosphate, it was abandoned due to insufficient economic reserves. Later, as stated in the report of Gök [13], magnesite lenses and radioactive minerals were also found in some parts of volcanic tuff layers in samples taken from the vicinity of Arikli and in 1980 the Mineral Research and Exploration Office took two samples from the region for analysis and values of 1050 mg/kg and 1300 mg/kg uranium were detected by the Turkish Atomic Energy Authority laboratories [13, 14].

Using results from the phosphate related studies on Arikli tuff (ignimbrites) performed by Günaydın [11], Çelik et al. [15], and Günaydın and Çolak [16], it was concluded that uranium and phosphate enrichments were the result of hydrothermal fluids and that they accumulated in brecciated fault zones [11, 15, 16]. Bayleyite [Mg$_2$(UO$_2$)(CO$_3$)$_3$.18H$_2$O] and ningyoite [(U,Ca,Ce)$_2$(PO$_4$)$_2$.1–2H$_2$O] were defined as uranium minerals in the area. During the studies, 1:5000 scale geological maps were prepared [11]. It was noted that significant faults were developed in NE–SW and NW–SE directions.

Using results from the outdoor gamma measurements, the final study area was reduced to approximately 12 km$^2$ at the south-east of Ayvacik (in the Ayvalık İ17-d4 pedestal). In 2000, about 256 people were living in Arikli village, where cattle and chicken farming, beekeeping and a very important olive production are the main activities in the village. The region also has its own private drinking water source originating in the Kaz Mountains, but groundwater is also used as drinking water. Additionally, groundwater is also used for agriculture and animal farming in the region. Streams flow into the Edremit Gulf, but the water flows are intermittent during the year. The area contains deep valleys with narrow plains. Kocakaya Hill, the highest peak in the region, is 742 m high. Olive trees grow on the southern coastline, pine forests to the northern side and some small trees and bushes can also be found there.

The study area consists of four main geological formations, Upper Cretaceous ophiolitic rocks at the base, volcanic units, lacustrine sediments and alluvial sediments. The Çetmi ophiolitic melange is a complex unit with volcanic lavas, pyroclastic rocks, limestone blocks, shale and greywacke and these form the basement of the study area. They are covered by volcanic and Neogene lacustrine sediments known as the Küçükkuş Formation, which contains volcanic intercalations with conglomerate, sandstone, siltstone, claystone and marl [17–20]. In the study area, the main unit is represented by the Arikli tuff which consists of andesite/andesitic tuffs and andesitic agglomerates. The SiO$_2$ content indicates that the unit has a moderate acidic character and the chemical composition corresponds to rhyolites and rhyodacites [11]. Siliceous and altered nodules of 2–7 cm radius, some exhibiting iron oxide banding, are present within the tuffs. Uranium enrichment developed via Ca$^{2+}$–U$^{4+}$ ion exchange in the phosphate nodules found in the Arikli volcanics [20].
3. MATERIALS AND METHODS

Taking into consideration the studies made by the Mineral Research and Exploration Office, OAGDR orientation measurements were first undertaken in the current survey and included the villages of Arıklı, Nusratlı, Ahmetçe, Hüseyinfakı, Demirciköy and Kayalar, representing an area of approximately 50 km². For the measurements, a portable ESP-2 Na(I) probed Eberlin gamma detector was used at 1 m above the ground level using a 100-s interval for each measurement [21]. The measurements were planned according to the lithological units. The map of the OAGDR data was prepared by the kriging geostatistical interpolation method using Arc-GIS software. As a result, the study area was restricted to a 2.63 km × 4.25 km rectangle (~11 km²) which included the catchment area of the Arıklı mineralization site and Arıklı village.

The restricted area was first split into a 500 m × 500 m grid inside the catchment and the grid was then reduced to 250 m × 250 m. OAGDR measurements were taken at the corners of each square. Open exploration ditches were identified and their OAGDR measurements were also taken.

Soil samples were collected from the measurement points. Before collecting soil samples, the sampling points were cleared of vegetation, roots and rocks, and then 250 g of material from the first 10 cm of the topsoil was packaged. The samples were dried under sunlight to remove the moisture, transported to the laboratory and then they were sieved in 1 mm mesh, packaged and labelled.

For pH measurements, a 6-g sample was mixed with 15 mL of distilled water and after 12 h, measurements were taken. For calibration of the pH meter (Multi 340i and pH/Cond 340i Handheld Multimeter), pH4.00, 7.00 and 9.00 buffer solutions were used. The same solution was also used at the same time and with the same device, but with a different probe, for electrical conductivity measurements [22].

For carbonate analysis, 3–10 g of soil was mixed with 0.1M HCL solution. The probe of the Scheibler calcimeter was filled with the HCL solution and once fixed into the polyethylene wide-mouth 250 mL tube, the solution was mixed into the samples which were agitated by magnetic stirrer. The CO₂ pressure was measured by the Scheibler calcimeter chamber filled with 0.1M sodium hydroxide solution. The results of the gas volume were converted to per cent CaCO₃ by calculation [23].

The barium chloride method was used for cation exchange capacity analysis. A 4-g sample was mixed in 0.1M BaCl₂ solution and buffered up to pH8.0 with tetraethyl ammonium. After 2 hours of agitation by shaker at 480 rpm, the solution was filtered and centrifuged. Finally, the Ba²⁺ content was measured by ICP-OES [24].

To take account of the topography, run-off, slope break and watershed, models were derived from the digital elevation model for 5 m × 5 m grid cells [25]. A drainage map and a land cover map were derived from the topographic map using Arc-GIS software. All the multisource prediction maps were superimposed by GIS operations using Surfer Software Homogeneity tests of univariate distributions, bivariate scatter plots, multivariate cluster analysis and principal component analysis being used for statistical analysis.

4. RESULTS

The highest outdoor gamma levels were detected in the Karakisla region. In the study area, other anomalous measurement levels were found in the exploration ditches of Feyzullah Hill. At both sites, the ditches were still open and the maximum depth reached 5 m. It was advised that the ditches be filled in and the radon emission at the sites checked.

Since the area was under the effect of slope driven soil erosion, the OAGDR measurements, there was more correlation with topographical units than with the lithological units. The gamma levels at the alluvial
accumulating flat bottoms of valleys were also higher than the background level owing to the erosion effect and the hills acting as physical barriers to prevent the dispersal of the radioactive contaminants from the catchment.

According to the results of the soil chemical analyses, it is concluded that the higher values of electrical conductivity and cation exchange capacity measurements were driven by topographical and hydrological barriers. For example, in the alluvial accumulations at the bottoms of valleys and along the meanders of stream branches, where water flow is slower, deposition took place which resulted in higher values for electrical conductivity and cation exchange capacity. As regards pH and carbonate measurements, their results correlated with each other and recorded the highest values in the sediment samples.

5. CONCLUSION

This interdisciplinary developed methodology helps to analyse the characteristics of the uranium distribution originating from the Arıklı mineralized site. The methodology characterizes the geochemical and topographic units and provides an insight into the mechanisms controlling the distribution of elements.

REFERENCES


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