High Arsenic Levels in Water Resources Resulting from Geogenic Resources: A Case Study from Muratlar Region, NW Turkey

Alper BABA
Izmir Institute of Technology, Department of Civil Engineering, Urla, Izmir, Turkey
alperbaba@iyte.edu.tr
Can ERTEKIN
General Directorate of Mineral Research and Exploration, Department of Energy Resources Research and Exploration, Izmir, Turkey
Deniz SANLIYUKSEL YUCEL
Canakkale Onsekiz Mart University, Department of Geological Engineering, Canakkale, Turkey

ABSTRACT
A large number of the studies on arsenic (As) in groundwater have been carried out over the last decades and a high concentration of this element has been identified in different parts of world. Muratlar Region in the northwest of Turkey is one such area having complex geology of widespread volcanic succession with active tectonics and geothermal fluids. This natural setting serves as a suitable environment for the presence of high levels of arsenic in groundwater in Muratlar Region. Arsenic was determined to be presented in water samples taken from water resources in the Muratlar Region during 2009-2010. High concentrations of arsenic were seen in the east of Muratlar Region, where densely settled argillic alteration outcrops were present. The maximum As concentration was 150 ppb. Chemical analyses revealed that arsenic was exceeding the maximum allowable limits depicted in the national and international standards for drinking water quality. The main reason for obtaining high arsenic concentrations is related to longer retention times of water resources in altered (argillic, silicification) volcanic rocks. Water-rock interaction is an important mechanism in determining the overall quality pattern of groundwater resources in this region.

1 INTRODUCTION

Arsenic (As) is an important pollutant and it has received attention recently, due to its toxic properties. A large number of studies on arsenic in groundwater have been carried out over the last decades and elevated concentration of this element have been identified especially in Bangladesh, India, Nepal, El Salvador, Ecuador, Honduras, Mexico, Chile, China, Canada, Argentina, Peru, Taiwan, United States, Bolivia and Turkey. Arsenic is a geogenic or anthropogenic contaminant in many areas and environments, which puts subsistence into risk. It is occurrence in groundwater has emerged as a major environmental catastrophe in several parts of the world. Serious pollution of groundwater by arsenic is known to occur worldwide (Ravenscroft et al. 2009). Millions of people living are exposed to high levels of arsenic from drinking water (Ahmed 2009). Approximately 125-150 millions of people are exposed to the risk of arsenic toxicity (Bhattacharya et al. 2002). International Agency for Research on Cancer (IARC) in 1987 classified inorganic arsenic as carcinogenic to humans.

Most of the aquifers that are polluted by arsenic, host anoxic groundwater and occur in alluvial setting, especially in deltaic setting. In such aquifers, pollution is caused by microbial reduction of sedimentary iron-oxyhydroxides and release to groundwater of their sorbed load of arsenic (McArthur 2010). But in some locations, arsenic is naturally found in the subsurface strata within volcanic formations, as well as in areas of geothermal systems, that are related to tectonic activity (Baba 2010; Gunduz et al. 2010). Arsenic occurs in natural waters in a variety of forms, including soluble, particulate, and organic bound species, which principally are inorganic trivalent As (III) and pentavalent As (V) of oxidation states (Lenoble et al. 2002). The trivalent bound one (arsenite H_3AsO_3) and the pentavalent bound ones (arsenate $H_2AsO_4^-$; $HAsO_4^{2-}$) are inorganic species, which tend to be more prevalent in water resources than the organic arsenic species. The toxicity of As increases greatly, when As is reduced from As (V) to As (III) in water (Chen et al. 2010). There are over 100 arsenic-containing minerals, including arsenic pyrites (FeAsS), realgar (AsS), lollingite ($FeAs_2$, Fe_2As_3, Fe_2As_5), and orpiment (As_2S_3). Arsenic retention in soils is primarily related to the contents of Fe, Al oxides, redox potential, pH as well as on the type and content of clay in soils. Clay alteration is one of many important sources of arsenic. Hydrothermal fluids carry metals in solution, coming either from a nearby igneous source, from leaching of subsurface rocks, or from both (Henley et al. 1984). These fluids then alter other rocks, changing their mineralogy and chemical composition (Nicholson 1993; Verma et al. 2005; Pandarinath et al. 2008). Complex zoned alteration patterns are well-documented to exist as a large number of important hydrothermal ore deposit types, including submarine-volcanogenic massive sulphide (Finlow-Bates and Stumpfl 1981; MacLean and Kranidiotis 1987) and epithermal Au-(Ag) deposits (Arribas 1995; Hedenquist and Arribas 1999). Many kinds of alterations exist in different parts of the northwestern Turkey, where intense zones of silicified, propylitic, and argillic alteration can be observed (Baba 2010; Baba and Gunduz 2009; Gunduz et al. 2010). These types of alteration also can be seen around the Muratlar Region.

Muratlar Region in the northwestern Turkey is one such area of complex geology having widespread volcanic succession with active tectonics. This geological setting serves as a suitable environment for the presence of high levels of arsenic in subsurface waters. Based on these fundamentals, this study presents a general overview of arsenic presence in northwestern of Turkey. The geochemical and hydrogeological processes related to As mobilization in alteration rock aquifers are investigated in the Muratlar Region, NW Turkey. This research aims to gain understanding of the extent and spatial scale that bedrock geology influences As distribution and what geochemical reactions mobilize As in the Muratlar Region.

2 GENERAL CHARACTERISTICS OF THE STUDY AREA

Canakkale province is geographically located in the Northwestern Turkey, between the coordinates of 39.4^o - 40.8^o N latitudes and 25.6^o - 27.6^o E longitudes (Figure 1A). The study area is located at about 60 km southeast of Canakkale City (Figure 1B). The Muratlar Region is also located in Canakkale province, between the coordinates of 39.9^o – 40.0^o (4420000 – 4425000 in UTM coordinates) N latitudes, 26.8^o – 26.9^o (480000-487000 in UTM coordinates) E longitudes and includes Muratlar Town, Yenikoy District and Tepekoy District (Figure 1C). Many private companies have been conducting exploration activities in the region for gold prospecting. Also, many private and governmental companies have been extracting coal in this region.

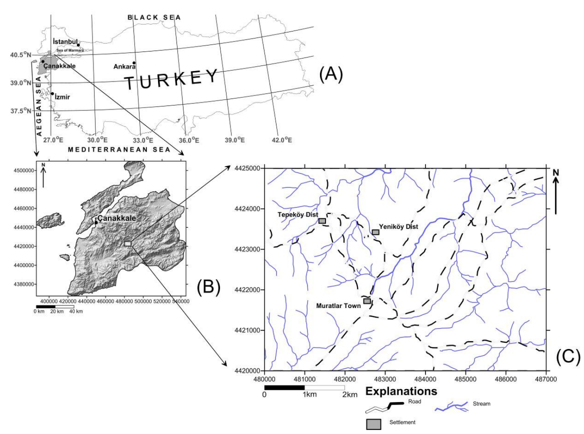

Figure 1. (A) Location map of the Çanakkale region. (B) Location map of the Çanakkale City and the study area. (C) Location map of the study area. Abbreviation: Dist, district

Climatologically, the study area is situated within a transitional climate region between Black Sea Climate zone and Mediterranean Climate Zone. North winds are predominant in the area and the mean annual precipitation rates range between 600-850 mm. Precipitation is typically in the form of rain and is mostly observed in autumn, winter and spring months. The maximum and minimum temperatures are observed to be 38.7°C and 11.5°C, respectively (Baba et al. 2009).

3 MATERIALS AND METHODS

All water samples obtained either from the wells dug up various depths or from springs and reservoirs, during their discharging process. Polyethylene bottles of 500 mL and 50 mL capacity were obtained and were then kept in portable coolers. The 500 mL samples were used in anion and cation analyses and the 50 mL samples were used in the trace element and heavy metal analysis. All samples were filtered (0.45 µm) in the field. The samples for the trace elements and heavy metal analyses were acidified with nitric acid solution to achieve a pH value of below 2. The samples were then refrigerated and kept at 4 ºC, until analyses finished. In the meantime, physical field parameters (i.e., temperature, pH, oxidation reduction potential and electrical conductivity) were instantly measured in-situ (i.e., inside the field) at all sampling points (wells, springs and reservoir) with a multi-parameter probe (WTW 340i). Major anions and major cations were analyzed with ion chromatography (Dionex ICS-3000) at the Izmir Institute of Technology laboratories. Trace elements and heavy metals in water samples were analyzed with inductively coupled plasma-optical emission spectrometry (Agilent 7500ce Octopole Reaction System (ORS) ICP-MS) at the Izmir Institute of Technology and Hıfzıssıhha laboratories. Rock samples were obtained from eight different locations near the Muratlar Region. A total of 1 kg rock samples were collected. Major and minor element contents of these samples were analyzed at the ACME Laboratories in Canada, via the inductively coupled plasma mass spectrometry (ICP-MS) method.

Aquachem 3.70 computer program was used to conduct primary computation of water chemistry. Parameter distribution map are generated in the GIS platform (ArcGIS 9.2) used in this study.

4 GEOLOGICAL AND HYDROGEOLOGICAL SETTING OF THE STUDY AREA

Turkey is one of the most seismically active regions in the world. Its geological and tectonic evolution has been dominated by the repeated opening and closing of the Paleozoic and Mesozoic oceans (Dewey and Sengor 1979; Jackson and McKenzie 1984; Bozkurt 2001). It is located within the Mediterranean Earthquake Belt, whose complex deformation results from the continental collision between the African and Eurasian plates (Bozkurt 2001). The border area of these plates constitutes seismic belts marked by young volcanics and active faults, the latter allowing circulation of water, as well as heat. The distribution of hot springs in Turkey roughly parallels the distribution of the fault systems, young volcanism and hydrothermally altered areas (Simsek et al. 2002). There are a total of about 1000 thermal and mineral water spring groups in the country (MTA 1980; Simsek et al. 2002) (Figure 2). The activity of Western Anatolia is believed to be a result of tensional forces that resulted from rigid behavior during the Neogene and Quaternary and the development of extended near-coastal graben areas (Baba and Ármannsson 2006). Another potential arsenic source near the study area is the geothermal fluid that has 290 ppb mg/l arsenic levels (Yilmaz et al. 2009).

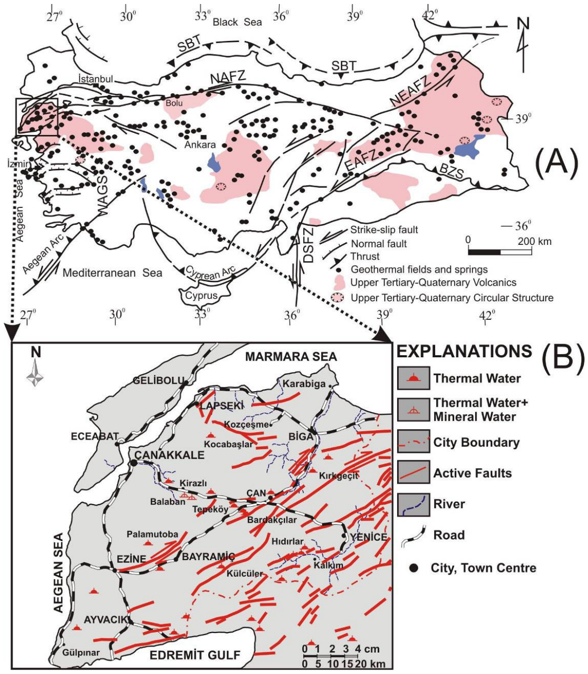

Figure 2. (A) Tectonic map showing structures developed during the Miocene to Holocene time and distribution of major hot water supplies around Turkey (compiled from Simsek 1997 and Yigitbas et al. 2004). Abbreviations: WAGS, Western Anatolia Graben System; NAFZ, North Anatolian Fault Zone; NEAFZ, Northeast Anatolian Fault Zone; EAFZ, Eastern Anatolian Fault Zone; BZS, Bitlis-Zagros Suture; DSFZ, Dead Sea Fault Zone; SBT, Southeastern Black Sea Thrust. (B) Tectonic features and geothermal areas around Canakkale Region (modified from Saroglu et al. 1992)

This study focuses on studying high arsenic levels in water resources coming from geogenic resources in the Muratlar Region, located in Northwestern Anatolia-Turkey, which is one such area of complex geology of volcanic succession with active tectonics (Figure 3). The basement rocks, which form Oligocene aged Can volcanic rocks, are composed of andesitic, dacitic, basaltic lava flows, agglomerate, tuff and the alteration materials of these, containing silicified tuffs and kaolin. The trace element geochemistry of different Can volcanics indicates that they are volcanic in nature and derived from the similar origin magma (Bozcu et al. 2008). Most of these rocks are altered and fractured, due to the effects of active faults. Widespread and intense zones of alteration in volcanic rocks are found to be present in this region (Figure 4). Since the majority of alteration was observed in volcanic rocks of the study area, it was concluded that this formation is the primary source of high As concentration in water sources (Figure 5). The volcanic structure that is dominant in the geological formation of Turkey in general and particularly in northwestern Turkey, in particular, is the primary mechanism for presence of numerous trace elements in earth's crust, including, but not limited to arsenic, antimony, boron, lead and zinc. Andesitic unit is overlain by feldspar quartz porphyry that mostly holds the gold mineralization in this region. Miocene aged Çan formation is composed of conglomerate, claystone, lignite, laminated organic claystone, sandstone-siltstone with intercalated tuff. The lignite accumulated in lacustrine environment indicates inundated swamp and limno-telmatic conditions (Bozcu et al. 2008). They are also found as impurities in the ores of other minerals including lignite levels (Karayigit et al. 2000; Baba et al. 2009) reaching as high as 6413 ppm, as in the case of arsenic. These lignite deposits just outcrop around the Muratlar Region. Quaternary alluvium overlies all the units unconformably, located in the north part of the study area.

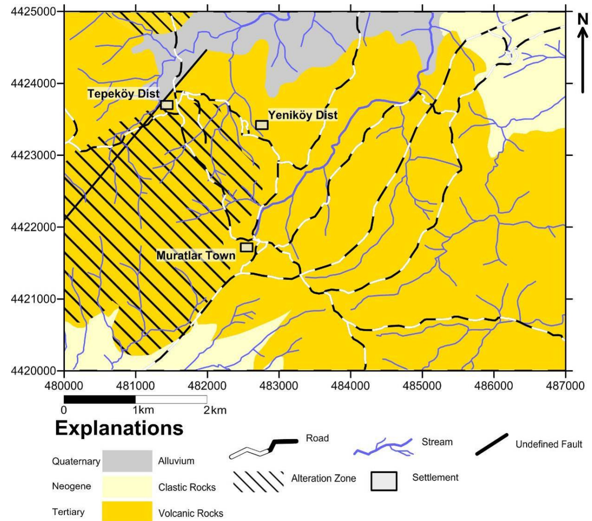

Figure 3. Geology of the study area (modified from MTA 2007)

Figure 4. Scenes of argillic alteration in the study area

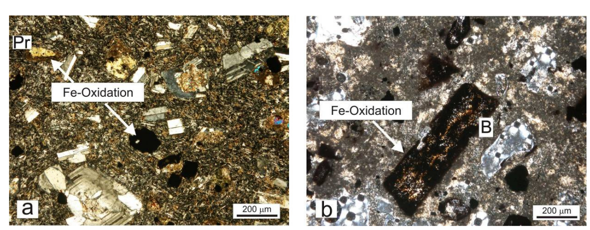

Figure 5. Fe oxidation in volcanic rocks a) basalt b) trachyandesite (Pr: pyroxene, B: biotite)

The Tertiary volcanic units are widely distributed across this part of the region. The silicified volcanics are extensively jointed and fractured and therefore have secondary (fractured) hydrogeological characteristics. Although these rocks have some water bearing potential they tend to be very limited in extent, invariably from spring sources, and yields are neither large nor sustained (0.01 l/s to 2 l/s). The silicified volcanics do contain localized alterations where there is mineralization in the study area and in the neighboring properties of the Muratlar Region. These altered units, commonly contain impervious clay layers that act as a hydrogeological barrier. Likewise, much of the high ground in the areas of mineralization are capped by silica layers that tend to be impervious to recharge, resulting in presence of perched water tables and high level springs. Although the volcanic rocks have some water bearing potential, they do not form good aquifers of regional extent. Their cooling fractures extend several meters in depth, providing a good avenue for deep penetration and circulation of groundwater. Because of the fact that the property area is on a ridge, underlain by these rocks, they form a recharge area for the groundwater system. Limited information on the water table depths obtained from some exploration holes indicates that the depth to water table varies between 0.5 m to 12.40 m, in autumn seasons. General groundwater flow is from SW to NE. Several springs emerge from these rocks in the Muratlar Region. Most of these spring water have been used by local people (Baba et al. 2009).

5 RESULTS AND DISCUSSION

Arsenic was determined on eleven water samples taken from water resources from Muratlar region during 2009-2010 periods (Figure 6). The pH values ranged from 3.46 to 7.34, electrical conductivity values (E.C.) were between 684 and 2170 µS/cm (Figure 7 and 8). The electric conductivity and pH showed significantly different values through the transition of the alteration zone and the fresh zone of volcanic rocks. Major ions constituents of the samples are evaluated by Piper and Schoeller diagrams.

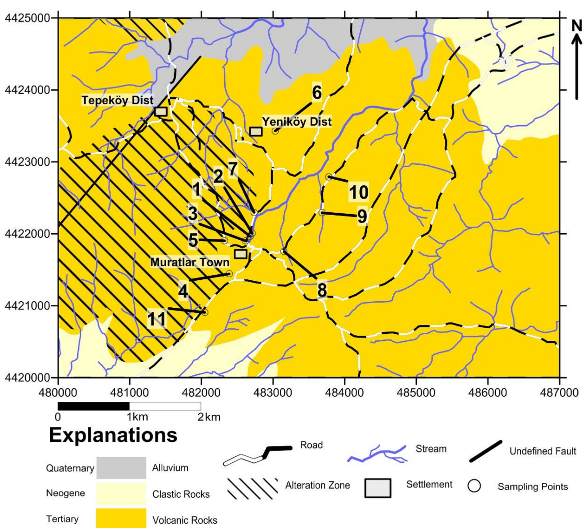

Figure 6. The map of sampling locations

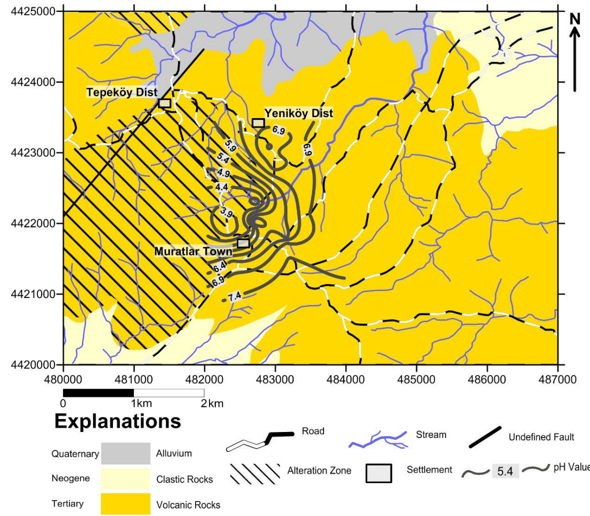

Figure 7. Distribution of pH

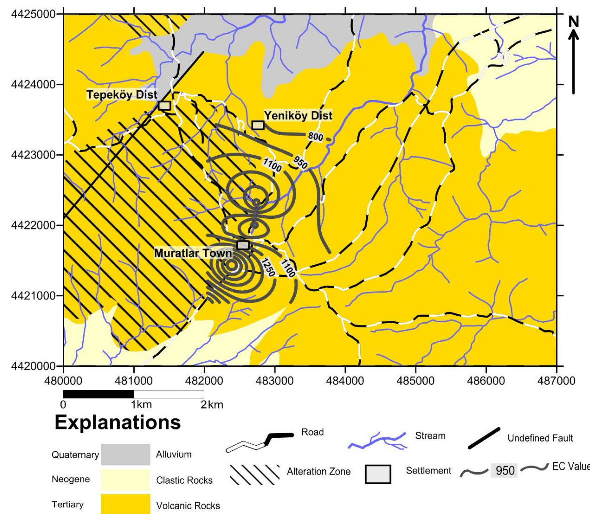

Figure 8. Distribution of EC (in µS/cm)

According to Piper diagram, the water in the Muratlar Region can be classified as being of Ca-Na-Cl or Ca-Cl types in the alteration zones (1, 2, 4, 7) and as either Na-Ca-Na-HCO_3 or Ca-HCO_3 types (6, 8, 9, 10, 11) in the fresh zone of volcanic rocks (Figure 9). To Schoeller diagram, the water can be classified into four patterns (Figure 10). As seen in Fig. 10, Samples from the alteration zone (1, 2, 4) have a different pattern, in accordance with the sample of 4 results. This can be explained by difference of shallow wells and spring water chemistry resulting from different retention times of water for the both causes. This explanation is also matched to the samples from the fresh zone of volcanics rocks. That is; the sample of 8 has a different pattern than the others because of taking from spring, but the others are well samples. Therefore different retention times acts on the chemistry of the samples of the fresh rock zones.

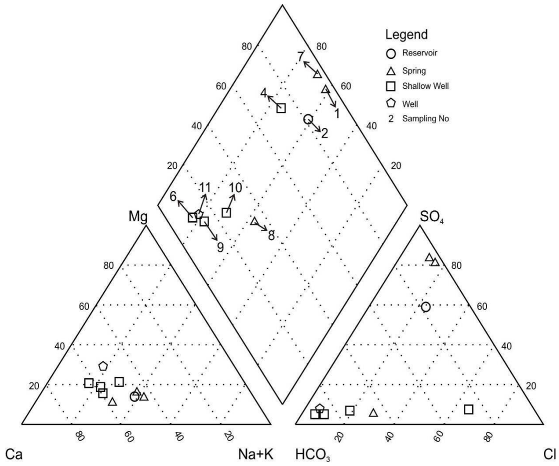

Figure 9. Piper Diagram

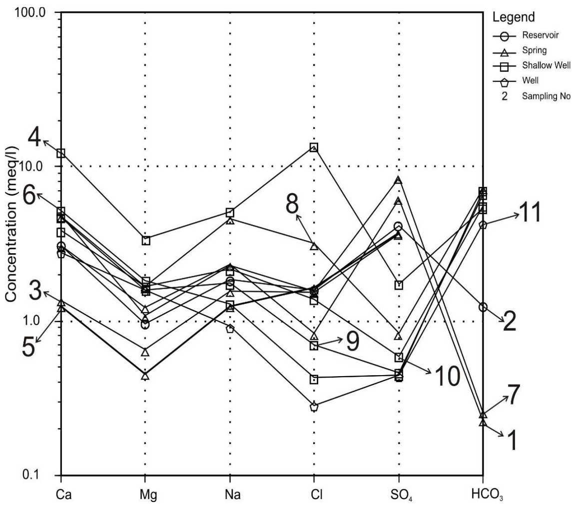

Figure 10. Schoeller Diagram

The chemical analyses revealed that arsenic was exceeding the maximum allowable limits depicted in the national and international standards (10 ppb) for drinking water quality (Table 1). High concentrations of arsenic were seen in the west and southwest of the Muratlar Region, where densely populated argillic alteration outcrops are present. The maximum As concentration was 150 ppb. Low arsenic concentrations were seen in the eastern part of the Muratlar Region, where the fresh zones of volcanic rocks were also seen. The magnitude of As contamination in groundwater samples from the Muratlar Region decreased gradually from southwest to northeast, due to being near the alteration zones (Figure 11). The main reason for obtaining high arsenic concentrations is related to having longer retention times of water resources in the altered geological formations (Table 2). The average As content in rocks around the Muratlar Region changed from 57.2 ppm to 735.7 ppm. The maximum content of As in rocks was 735.7 ppm and was found mostly in silicified rock. Arsenic occurred as an impurity in sulfide minerals, such as; realgar (AsS), orpiment (As_2S_3) and arsenopyrite (AsFeS) in the Muratlar Region.

Table 1: Average physical and chemical properties of water samples.

Sample No	X	Y	pH	T (°C)	E.C. (µS/cm)	E.h. (mV)	As (ppb)
1	482681	4421965	4.8	14.7	873	122	7.15
2	482707	4422009	6.59	12.2	709	18	25
3	482615	4421902	4.34	15.2	795	143	10.89
4	482388	4421443	6.56	16.8	2170	20	0.32
5	482324	4421903	3.46	14.3	1070	189	1.41
6	483030	4423429	6.77	15.3	726	7	0.15
7	482736	4422298	3.64	15.7	1608	180	1.71
8	483153	4421754	6.73	14	986	10	1.77
9	483683	4422292	7.13	14	928	-11	0.43
10	483778	4422792	6.96	15.5	877	-3	2.19
11	482046	4420907	7.34	14.1	684	-24	150

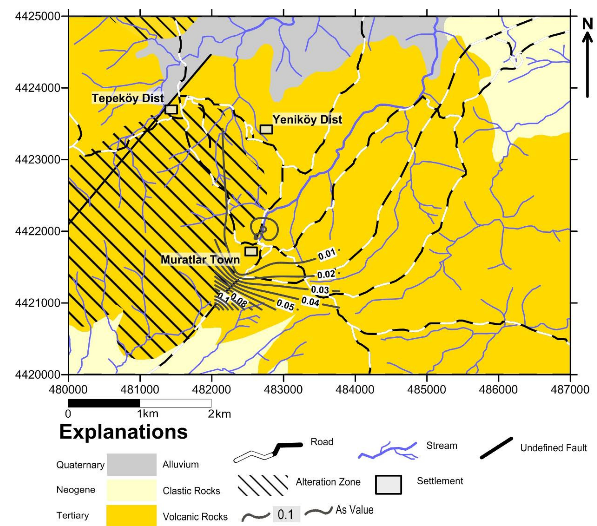

Figure 11. Distribution of As (in ppm)

Table 2. Minor element distributions in rock samples.

Sa. No	As (ppm)	Sb (ppm)	Hg (ppm)	Zn (ppm)	Ba (ppm)	Sr (ppm)	Rb (ppm)	Zr (ppm)	Th (ppm)	Cu (ppm)	Pb (ppm)	Ni (ppm)
1	57.2	0.5	0.08	3	160	7.9	0.5	69.9	0.3	9.4	1.2	2.1
2	143.1	2.8	0.01	2	245	101.1	0.7	49.7	0.4	6.2	4.2	2.3
3	112.5	0.2	0.03	3	437	12.4	0.3	55.2	0.2	5.2	0.8	2.5
4	282.3	0.4	0.01	2	915	1603	1.5	327	57.3	8.5	25.7	1.1
5	735.7	1.7	0.01	17	345	818.1	12.9	148.9	29.6	133.1	30.9	1.8
6	462.8	3.5	0.01	10	442	737.5	2.1	141.2	28.6	69.8	41.6	4.4
7	113.8	4.6	0.39	5	251	88.4	1.1	79.3	1.5	8.8	2.4	1.7
8	207.6	10.7	0.64	6	481	52.4	1	66.3	1	7.8	3.1	2.5

Rock-water interaction is an important mechanism in determining the overall quality pattern of groundwater resources. Volcanic rocks generally consist of andesites, basalts, tuffs and agglomerates present in the Muratlar Region. Rock samples were collected from the immediate vicinity of region with particular reference to the outcropping units. The results of rock sampling and some basic statistics are given in Tables 2 and 3 for major constituents and minor elements, respectively. Generally, results show that all rock samples consist of SiO_2, Al_2O_3, and Fe_2O_3 (Table 3). Particularly, SiO_2 contents reached to levels 97.58% (at no:1), which is a clear indication of the silicified zone exists. When minor elements are considered, arsenic is the most significant minor element. Generally, arsenic, strontium and zircon levels were also high and showed a parallel trend to each other in the study area.

Table 3: Major element distribution in rock samples

Sa. No	SiO_2 (%)	Al_2O_3 (%)	Fe_2O_3 (%)	MgO (%)	CaO (%)	Na_2O (%)	K_2O (%)	TiO_2 (%)	P_2O_5 (%)	MnO (%)	Cr_2O_3 (%)
1	97.58	0.22	0.83	0.02	0.03	0.01	0.03	0.67	0.01	0.01	0.005
2	96.38	0.76	1.01	0.01	0.01	0.02	0.06	0.82	0.03	0.01	0.005
3	97.18	0.29	1.37	0.01	0.01	0.01	0.01	0.54	0.01	0.01	0.003
4	47.47	33.81	2.33	0.07	0.22	0.1	0.09	1.3	0.45	0.01	0.004
5	53.53	15.52	18.51	0.14	0.18	0.23	0.06	0.61	0.23	0.01	0.005
6	58.27	14.72	17.17	0.01	0.08	0.03	0.02	0.5	0.19	0.01	0.004
7	95.33	0.7	1.69	0.01	0.01	0.01	0.04	0.7	0.07	0.01	0.003
8	95.35	0.77	1.91	0.01	0.02	0.01	0.04	0.68	0.07	0.01	0.008

Although the volcanic rocks have some water bearing potential they do not form good aquifers of regional extent. Their cooling fractures extend several meters in depth, providing a good avenue for deep penetration and circulation of groundwater. Because the property area is on a ridge, underlain by these rocks, they form a recharge area for the groundwater system. Several springs emerged from these rocks in this region. Some of these springs are used as a source of drinking water for the towns. The field water quality parameters were measured at some of these springs and drill wells, which are used as a source of water supply by local population. As groundwater is currently the source of drinking water in the area, it is extremely vital to find an urgent sustainable, reliable, affordable and acceptable solution to the problem of As toxicity in water supplies.

6 CONCLUSION

Based on this paper, it is evident that arsenic deserves more attention in Turkey. The health risk of arsenic in drinking water may not be neglected. Groundwater of a large area in the Muratlar Region, where densely altered rock outcrop, is contaminated with As and health of the thousands of people residing in the area is at risk. The increasing trend of contamination with time is also a matter of great concern. Therefore; well-planned water management and mitigation strategy, supported with specialized adequate medical facilities should be implemented without any delay.

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