# **Geochemical Characterization of Acid Mine Lakes in Northwest Turkey and Their Effect on the Environment**

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Received: 5 September 2012/Accepted: 5 November 2012/Published online: 6 December 2012 © Springer Science+Business Media New York 2012

Abstract Mining activity generates a large quantity of mine waste. The potential hazard of mine waste depends on the host mineral. The tendency of mine waste to produce acid mine drainage (AMD) containing potentially toxic metals depends on the amounts of sulfide, carbonate minerals, and trace-element concentrations found in ore deposits. The acid mine process is one of the most significant environmental challenges and a major source of water pollution worldwide. AMD and its effects were studied in northwest Turkey where there are several sedimentary and hydrothermal mineral deposits that have been economically extracted. The study area is located in Can county of Canakkale province. Canakkale contains marine, lagoon, and lake sediments precipitated with volcanoclastics that occurred as a result of volcanism, which was active during various periods from the Upper Eocene to Plio-Quaternary. Can county is rich in coal with a total lignite reserve >100million tons and contains numerous mines that were operated by private companies and later abandoned without any remediation. As a result, human intervention in the natural structure and topography has resulted in large open pits and deterioration in these areas. Abandoned open pit mines typically fill with water from runoff and groundwater discharge, producing artificial lakes. Acid drainage waters from these mines have resulted in the degradation of surface-water quality around Can County. The average pH and electrical conductivity of acid mine lakes (AMLs) in

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this study were found to be 3.03 and 3831.33  $\mu$ S cm<sup>-1</sup>, respectively. Total iron (Fe) and aluminum (Al) levels were also found to be high (329.77 and 360.67 mg L<sup>-1</sup>, respectively). The results show that the concentration of most elements, such as Fe and Al in particular, exceed national and international water-quality standards.

Acid mine drainage (AMD) is generally defined as the product formed by the atmospheric oxidation (i.e., by water, oxygen, and carbon dioxide) of relatively common iron (Fe)-sulfur minerals, such as pyrite (FeS<sub>2</sub>) and pyrrhotite, in the catalyzing presence of acidithiobacillus ferrooxidans, an acidophilic bacterium, and any other products generated as a consequence of these oxidation reactions (Mills 1995; United States Environmental Protection Agency [USEPA] 1994a, b in Akcil and Koldas 2006; Ozcelik 2007). Generally AMD usually has a low pH and increased concentrations (above background) of dissolved sulfate (SO<sub>4</sub><sup>2-</sup>), Fe (Fe<sup>2+</sup>, Fe<sup>3+</sup>), manganese  $(Mn^{2+})$ , aluminum  $(Al^{3+})$ , and other metals that result from the oxidation of FeS<sub>2</sub> and the dissolution of carbonate, oxide, and aluminosilicate minerals by acidic water (Rose and Cravotta 1998).

The processes that produce AMD are natural, but they are accelerated by mining and can produce large volumes of contaminated effluents. The potential for AMD from mining activities has been known since at least 1556, and acid rock drainage was observed as early as 1698 associated with coal mining in Pennsylvania (British Columbia Acid Mine Drainage Task Force 1989). AMD is an environmental problem that has been studied extensively since the 1950s (Leathen et al. 1953). The causes and formation of AMD as a major source of water-quality degradation (McCleary and Kepler 1994), especially in coal-mining regions, have been

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well documented (Stumm and Morgan 1970; Merritt 1986; Nordstrom and Ball 1986; Erickson and Heiden 1988; Cravotta 1991; Skousen and Ziemkiewicz 1995; Younger 1995; Foos 1997; Nordstrom and Alpers 1998; Rose and Cravotta 1998; Monterroso and Macias 1998; Wood et al. 1999; Pluta and Jackowicz-Korczynski 2003; Kim and Kim 2004; Larsen and Mann 2005; Blodau 2006; Devasahayam 2006; Cravotta 2008; Doulati et al. 2010; Silva et al. 2011a).

As acidic mine water moves through the near surface environment, constituents that are soluble under acidic, oxidizing conditions—such as Fe, Al, SO<sub>4</sub>, copper (Cu), lead (Pb), zinc (Zn), cadmium Cd, and selenium—become concentrated in the effluent (Anderson and Youngstrom 1976; Davis and Boegly 1981; Brake et al. 2001). The delivery of such acid effluents to streams and rivers can lead to significant environmental impact on sediment and water quality in downstream reservoirs (Salomons 1995; Allan 1997; Paktunc 1999; Sola et al. 2004). AMD is a long-term environmental problem that requires continuous information, monitoring, and long-term performance follow-up, all of which also incur extra costs to the society (Price 2003).

AMD is a recognized problem and is covered in curricula of mining faculties in Turkey. There are very few studies related to AMD except biological ore enrichment, hydrometallurgy, and mine abatement studies (Aytekin and Akdagi 1996; Akcil and Ciftci 2003; Kesimal et al. 2003; Akcil and Koldas 2006; Ozcelik 2007). The district of Can in the Canakkale province in northwest Turkey is rich in lignite. Many small- and medium-sized mining companies have operated lignite mines within the last 32 years. Some of the activities of these enterprises have ceased over time, whereas other mines are still operational. Ceased mining operations have been abandoned without any postclosure methods or rehabilitation. Consequently, human intervention in the natural structure and topography of the surface has resulted in the large open pits and deterioration in these areas. The abandoned open pit mines now contain artificial lakes fed by both surface runoff and underground leakage. High sulfur content of lignite exists in these areas. Over time, these lakes become acidic due to acid generation from FeS<sub>2</sub> oxidation. These acid mine lakes (AMLs), near agricultural fields and villages, have low pH values and high levels of heavy metals and trace elements (Al, Fe, Mn etc.). This study focuses on the hydrogeochemistry of AML and is a detailed investigation of AMD generation within the wastes produced by mining activities. The effects of these lakes on the environment are also evaluated.

# Site Description

The study area is geographically situated immediately west of Keciagili village, approximately 17 km southwest of the

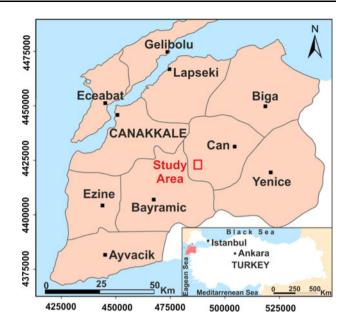


Fig. 1 Location map of the study area

county of Can of Canakkale province on Biga Peninsula in northwest Turkey (see Fig. 1). Climate effects may determine whether a mine discharge is continuous or intermittent, dilute, or highly concentrated, a characteristic that has an effect on the nature of the drainage. The hydraulic characteristics of mine waste may determine the contact time between solid and solution or the proportion of mine waste being flushed. Climatologically, the Canakkale province is situated in a climatic transition between the Mediterranean and Black Sea climate zones. The summers are hot and dry, and the winters are relatively wetter and

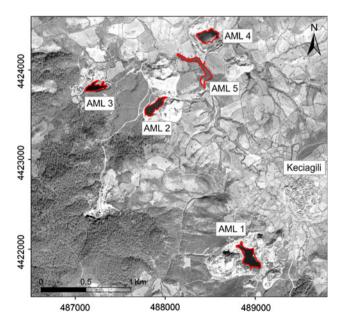


Fig. 2 Satellite imagery AMLs in study area (Worldview-1 [March 2011])

AML no.	Altitude (m)	Area (da)	Perimeter (m)	Mean depth (m)	Water volume (m <sup>3</sup> )	Reference
1	245	23.81	892	7.11	169289.1	Okumusoglu (2009)
2	172	15.55	616	7.41	115225.5	This study, July 2010
3	151	14.41	811	NM	NM	Worldview-1 satellite imagery
4	140	25.54	736	NM	NM	(March 2011)
5	128	23.83	1465	NM	NM	

Table 1 Basic characteristic of AMLs in the study area

NM not measured

colder. The long-term (1970–2010) mean annual temperature is 15.16 °C with a minimum of -12.1 °C (February) and a maximum of 39 °C (July) based on the data collected at Canakkale Meteorological Station (located approximately 45 km northeast of the study area). North winds are predominant in the region. Precipitation in the region is mostly in the form of rain, and the highest precipitation is observed in winter season. The mean annual precipitation rate was 595.7 mm between 1970 and 2011.

 $FeS_2$  oxidation and AMD generation are important processes that may take place in the wastes produced by coal-mining and coal-washing operations in Can county. The study area, one of the most polluted acid mine drainage (AMD) sites in Turkey, has many open pit coal mines that operated from 1980 to 2008. However, in 1980s the number of AMLs were increased, but they did not indicate the same increment numerically. It is clear that AMLs have risk due to their presence and areal increments (Yucel et al. 2012).

This study focused on five AMLs (AMLs no. 1 through 5) located immediately west of Keciagili village (Fig. 2). These are small lakes with surface areas not exceeding 25.54 decar (Table 1; Fig. 3a, b). Water levels in these small lakes fluctuate due variations in annual precipitation, evaporation, and drainage patterns. AML no. 1 is the oldest lake, followed by AMLs no. 2, 3, 5, and finally 4 (Yucel and Sanliyuksel Yucel 2012). AML no. 5 is situated on the

old stream bed, which was formed as a result of prevention with a huge amount of mine waste.

All of these lakes drain into Kocacay stream, which is the second largest stream in Canakkale province. The hydrology of surface drainage network in Canakkale shows a variable flow regime, which mainly depends on the seasonal precipitation pattern. The main element of the drainage network is the Kocacay stream, which flows northeast through the county of Can and eventually drains into the Sea of Marmara approximately 45–50 km north of Can.

### **Geological Setting**

Canakkale is located on the westernmost part of the Biga Peninsula. Biga Peninsula is one of the focal points of current mineral exploration in Turkey (Yigit 2012). Mineral deposits of the Biga peninsula are dominated by volcanic and intrusion-related hydrothermal systems, which is a natural corollary of the preeminent geology (Yigit 2012). Prevalent magmatic and associated volcanic rocks dominate in Biga peninsula. Apart from these plutonic rocks, Biga peninsula is generally made up of a metamorphic and granitic basement (Sengun et al. 2011) and the Permo– Triassic Karakaya complex (Okay et al. 1990; Okay and Goncuoglu 2004). Early Eocene–Pliocene–Quaternary



Fig. 3 a Photographs of AMLs no. 1 and b no. 2

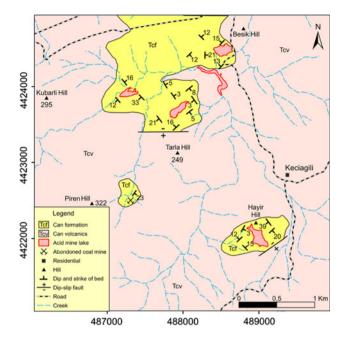


Fig. 4 Geological map of the study area

volcanic and sedimentary rocks form the youngest cover units (Siyako et al. 1989). Cenozoic volcano-plutonic rocks, covering extensive areas, dominate the geology of the Biga peninsula (Yigit 2012). Moreover, Late Eocene-Miocene volcanic activity is accompanied by Late Oligocene-Miocene granitic intrusions (Ercan et al. 1995; Delaloye and Bingol 2000; Okay and Satir 2000).

The study area mainly consists of acidic and agglomeratic, andesite, trachyandesite, and andesitic tuff which are named "Can volcanics" by Ercan et al. in 1995). During the Early-Middle Miocene, terrestrial units were deposited along with the calkalkaline volcanic activity (Siyako et al. 1989). These terrestrial units consist of bituminous shale, siltstone, claystone, sandstone, tuff, and coals, named the "Can formation," which is  $\leq 250$  m thick and unconformably overlies the Miocene-aged andesitic volcanic rocks (Siyako et al. 1989) (Fig. 4). According to the palynological studies, the coal and claystone of the Can formation were deposited during the late Early Miocene to early Middle Miocene (Middle Orleanian-Early Astaracian [Bozcu et al. 2008]). Early-Middle Miocene times are characterized by coeval volcanism and sedimentation. Lacustrine sediments, such as shale, siltstone, and tuffs, were deposited in small basins, including economic coal deposits, e.g., Can lignite (Yigit 2012). In the Can Basin, the total coal reserves are > 100 Mt (Bozcu et al. 2008) and are exploited mainly by opencast mining in the study area. The Can formation is rich in lignite (also known as brown coal) that is 17 m thick (Bozcu et al. 2008) and characterized by its low calorific value and high sulfur content. The Can coals are lignite to subbituminous coal (Ro = 0.32-0.44 %) and have an intermediate to high moisture content ranging from 14.67 to 28.42 %. The ash, volatile matter, and sulphur (S) content (on original basis) vary between 3.48 and 29.83 %, 26.43 and 35.81 %, and 0.64 and 8.1 %, respectively (Gurdal 2008). The major oxides in Can coals are dominated by SiO<sub>2</sub> (0.65–20.64 %), Al<sub>2</sub>O<sub>3</sub> (0.4–6.09 %), and Fe<sub>2</sub>O<sub>3</sub> (0.29–4.09 %) with lesser amounts (<1.0 %) of MgO, CaO, NaO<sub>2</sub>, TiO<sub>2</sub>, K<sub>2</sub>O, P<sub>2</sub>O<sub>5</sub>, MnO, and Cr<sub>2</sub>O<sub>3</sub> (Baba et al. 2007).

Widespread and intense zones of silicified, argillic, and propylitic alterations in the study area can be observed in Can volcanics. These alteration zones give rise to distinct mineral forms. In general,  $Al^+$  and  $K^+$  as well as  $Mg^+$ , Ca<sup>+</sup>, and Fe<sup>++</sup> are enriched in argillic and propylitic alteration, respectively (Baba and Gunduz 2010). Ca<sup>+</sup>, Mg<sup>+</sup>, and Fe<sup>++</sup> were leached during argillic alteration, whereas Na<sup>+</sup> leaching is evident in all alteration types (Sanliyuksel Yucel et al. 2012). The alteration system in the study area displays all porphyry-related alterations, including epithermal and skarn systems, containing porphyry-related high-sulphidation style gold (Au) and Cu-Au mineralization (Yigit 2012). The alteration preserves the texture with feldspars altering to kaolinites and smectites, with the groundmass being replaced by varying degrees of silica. A surficial argillic alteration zone has developed on weathered outcrops due to the oxidation of  $FeS_2$ .  $FeS_2$  is the most abundant primary sulfide mineral related to Au in the prospect (Yigit 2012).  $FeS_2$  is a common mineral in abandoned coal mines, especially around Keciagili village. FeS<sub>2</sub> oxidation and AMD generation are important processes that may take place within the wastes produced by coal mining operations in study area.

# **Materials and Methods**

Geochemistry and Mineralogy of the Study Area

Mine wastes and sedimentary and volcanic rocks were analyzed for major element content and chemical characteristics. The mineralogical characteristics of mine waste and soils are the most important factor of AMD quality. It is therefore critical to evaluate the mineralogical characteristics of a mine waste. Mine wastes having much greater surface area than undisturbed geologic material due to their smaller grain size and are more prone to generating AMD. Because large masses of sulfide minerals are exposed quickly during the mining and milling processes, the surrounding environment can often not attenuate the resulting low pH conditions (Jennings et al. 2008). Therefore, mine wastes were collected around Keciagili village to evaluate the mineralogical characteristics of mine wastes. All samples were analyzed by inductively coupled plasma-mass spectrometry (ICP-MS) at ACME Labs (Canada).

The morphology of the mine waste and bottom sediment at the macro and micro levels were obtained by microscopy. Scanning electron microscopy (SEM) (FEI Philips XL30 sFEG, OR) coupled with energy-dispersive X-ray spectrometry (EDX) were performed using secondary electrons (SE) and back-scattered electrons (BSE) detectors to determine surface features and local chemical contents. X-ray diffraction (XRD) analyses of the samples were performed on a Philips X'Pert diffractometer and in a Debye-Scherrer camera. XRD was performed on samples pulverized in alcohol for 3 min in a McCrone micronizer equipped with agate grinding pellets. The powder patterns were collected with a Scintag X1 automated powder diffractometer equipped with a Peltier detector having CuKa radiation. The XRD patterns were analyzed with the use of Material Data Inc.'s JADE software and standard reference patterns. Waste and sediment samples were analyzed at the Center for Material Science at the Izmir Institute of Technology, Turkey.

### Hydrogeochemistry of AMLs

The five AMLs, from which water samples were taken, were monitored from 2009 to 2012 in the study area. In all AMLs, water was sampled. In addition, a 9 m-deep vertical-depth profile was sampled and measured at AML no. 2. Water samples were filtered through 0.45-µm filter paper, transferred to 50-mL polyethylene bottles. and stored at 4 °C without acidification. All water samples were collected in duplicate. The samples were analyzed for major cations, anions, heavy metals, and trace elements. Major and trace elements were analyzed with ICP-MS, and SO<sub>4</sub> was analyzed by ion chromatography at ACME Labs (Canada). Physical parameters (pH, redox potential, temperature, salinity, and electrical conductivity) were also measured in situ using a multiparameter probe (WTW 340i).

Based on the results of the bathymetry study and the characteristics of the general lake morphology of AML no. 2, sampling locations were identified on the lake surface. The locations of these sampling points always coincided with some of the bathymetry points. In general, the aim was to achieve a relatively homogenous distribution of sampling points within the lake's surface area. Waterquality samples were then collected from all stations with a Niskin sampling bottle (HydroBios) with a capacity of 5 L. The bottle was made from hard plastic materials to avoid cross-contamination from metal ions. Although surface samples were collected directly into polyethylene sampling bottles, all other samples were obtained with this equipment. All of collected data was transferred to a geographic information system (GIS) platform (ArcGIS v9.3) for data visualization and interpretation.

### **Results and Discussion**

### Geochemical Data

The geochemistry of the mineral-water interaction is dependent on the geologic setting. The potential sources for the generation of AMD are reactive sulphide minerals and their oxidation products. The production and persistence of acidity largely depends on the nature of the sulphide mineral being oxidized, the reaction mechanism (*i.e.*, oxygen *vs.* ferric Fe as the oxidant), and the presence of acid-consuming minerals, such as carbonates and aluminosilicates.

A coal-rock sampling campaign was also conducted in the vicinity of the AMLs sampled. A total of 10 samples were collected from volcanic and sedimentary rocks, coal, and mine wastes and analysed for major element constituents (Table 2). SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and Fe<sub>2</sub>O<sub>3</sub> constitute the other important parts of the rocks, with maximum values reaching as high as 61.25, 19.97, and 10.46 %, respectively.

Mineralogy and petrography data were also used to examine sulphide mineralogy in the study area. The most common sulphide mineral is FeS<sub>2</sub>, which is the mineral of most relevance from an acid-generation perspective. The Greek philosopher Theophrastus (approximately 325 BC) recognized the oxidation of FeS<sub>2</sub>, the formation of metal salts, and the production of acid (Lottermoser 2010). FeS<sub>2</sub> produces AMD, and, generally, crystals are <3 mm in size, yellowish brown in color, and highly lustrous in the study area.

FeS<sub>2</sub> oxidation is directly controlled by the mineral and crystal type (Malmstrom et al. 2000) and by particle size, i.e., mass normalized surface area (Stromberg and Banwart 1999). Morrison (1988) defined nine classes of FeS<sub>2</sub> morphology, with end members having framboidal and euhedral crystal structures. Framboidal FeS2 consists of aggregates of very small FeS<sub>2</sub> crystals (< 1  $\mu$ m), whereas euhedral crystals are generally greater (tens to thousands of microns). Among a variety of morphologies, such as framboids, octahedral, and cubic crystals with irregular surfaces, the small, polycrystalline framboids are particularly susceptible to oxidation owing to their porosity and large surface area (Evangelou and Zhang 1995). FeS<sub>2</sub> crystals have a cubic crystal form in coals in the study area (Fig. 5) containing 69.60 % sulfur and 30.40 % Fe (Fig. 6).

Neutralization reactions play a key role in determining the compositional characteristics of drainage originating from sulphide oxidation. Most carbonate minerals are capable of dissolving rapidly, thus making them an effective acid neutralizer. Calcite is more easily dissolved than other carbonate minerals. Dissolved calcite neutralizes acid

Rock type	SiO <sub>2</sub>	$Al_2O_3$	Fe <sub>2</sub> O <sub>3</sub>	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	TiO <sub>2</sub>	$P_2O_5$	MnO	Cr <sub>2</sub> O <sub>3</sub>	LOI	Sum
Coal	24.18	8.28	10.19	0.18	9.28	0.85	0.44	0.12	0.1	< 0.01	< 0.002	45.6	99.19
Coal	40.4	19.97	10.46	0.49	0.37	0.21	0.38	0.72	0.18	0.66	0.003	26	99.88
Claystone	51.35	19.81	2.96	0.93	3.75	0.35	1.76	0.64	0.41	< 0.01	< 0.002	17.8	99.79
Claystone	61.25	14.8	2.34	0.71	0.31	0.36	0.36	0.56	0.31	0.01	0.003	18.8	99.81
Mine waste	44.61	14.13	5.57	2.09	0.22	1.14	0.93	0.64	0.14	< 0.002	< 0.004	29.17	98.64
Mine waste	35.51	13.6	3.93	1.79	0.37	1.25	1.29	0.39	0.09	0.01	< 0.004	40.85	99.08
Andesite	58.9	17.77	6.63	0.98	3.65	2.77	3.22	0.79	19	0.04	0.003	4.8	99.76
Andesite	56.99	19.49	7.39	0.44	0.67	0.91	4.23	0.71	0.1	0.09	0.004	8.7	99.74
Andesite	56.89	19.85	3.1	0.75	1.55	1.59	3.56	0.84	0.12	< 0.01	0.005	11.5	99.76
Trachyandesite	58.31	17.64	7.37	0.5	4.97	3.18	3.18	0.7	0.21	0.06	0.002	3.7	99.8

Table 2 Major element oxides of rocks in study area (%)

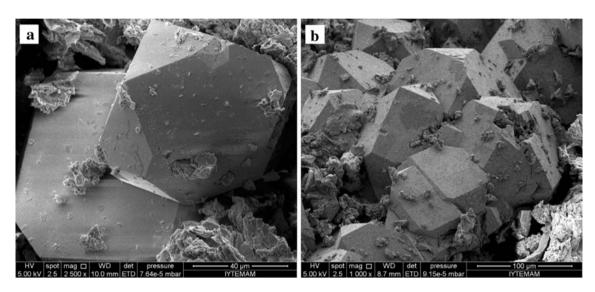


Fig. 5 SEM photomicrographs showing cubic  $FeS_2$  crystals in coal (crystal structures are. a pentagondodekaeder and b pyritoeder)

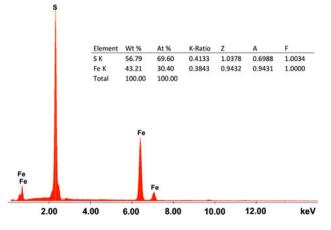


Fig. 6 EDX results of cubic FeS<sub>2</sub> crystals

by complexing with hydrogen ions to form bicarbonate and carbonic acid (Stumm and Morgan 1995). Calcareous minerals are the dominant components of fractures in claystone, shale, and associated strata of coal-bearing rocks in Keciagili village.

The major reservoir for buffering capacity in the environment is silicate minerals, which make up the majority of the minerals found in the Earth's crust (Lottermoser 2010). Chemical weathering of silicate minerals consumes hydrogen ions and occurs by way of congruent or incongruent weathering (Lottermoser 2010). Dissolution of silicates, such as plagioclase-feldspars and olivine, can also neutralize acid (Lapakko 2002). However, their rates of dissolution and consequent acid neutralization are slow relative to the carbonate minerals (Nesbitt and Jambor 1998). Feldspar, labradorite, quartz, orthoclase, and mica are silicate minerals and can be detected by XRD analysis in mine waste. The clay minerals also occurred as in association with FeS<sub>2</sub> and calcite. The XRD analyses performed detected illite, smectite, were also observed in the SEM analyses (Fig. 7).

Secondary minerals are important in water-mineral reaction and result from weathering of primary sulphide

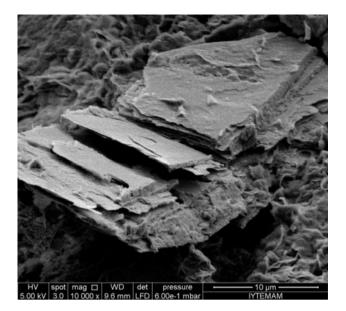


Fig. 7 SEM photomicrographs showing clay minerals in claystone

minerals. Secondary minerals are younger than the rock inn which they form (Neuendorf et al. 2005). Some of these secondary minerals may have deleterious effects on water quality because of the release of additional acidity during their formation (*e.g.*, metal (hydro)oxides) or release of stored acidity, sulphate, or metals (or both) during their dissolution (*e.g.*, Fe and Al hydroxysulphates). The formation of secondary minerals is the most common form of element fixation in porewaters of sulfidic wastes (Lottermoser 2010). A significant fraction of the metals released by sulfide oxidation is retained in the wastes as the secondary mineral precipitates (Lin 1997; Lin and Herbert 1997; Lottermoser 2010).

Mineralogical studies of AMD sites indicate that gypsum is the most common sulfate phase controlling sulfate concentrations in such environments (Davis et al. 1991; Jambor and Blowes 1994). Gypsum was found in tailings in Keciagili village (Fig. 8a–c). The gypsum formed during the earlier stages of AMD generation and dissolved as a

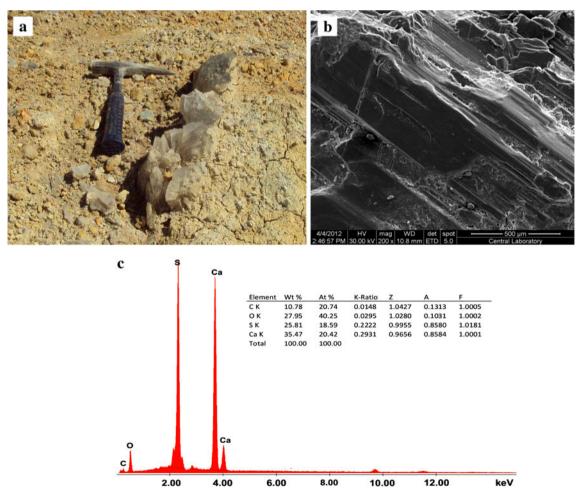
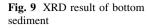
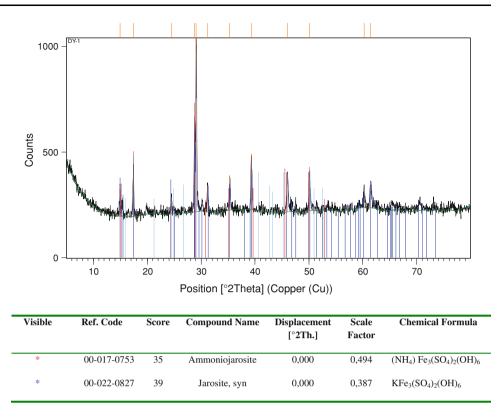


Fig. 8 a Field view and b SEM image showing gypsum and c its EDX analysis





consequence of low calcium (Ca) concentration in porewater. Sulfate was flushed out of the tailings pile.

Jarosite and amoniojarosite are the main secondary minerals in bottom sediment (Fig. 9). Jarosite is a strawyellow mineral that is common in the weathered zone of sulfide ore deposits and in acid sulfate soils. Jarosite is less soluble than the other acid-producing sulfate minerals. Jarosite commonly occurs intimately mixed with clays in seams and in thick clay beds (Dutrizac and Jambor 2000). The general assumption is that the Fe and sulfate originate from the oxidation of FeS<sub>2</sub>, whereas acid leaching of the clay minerals provides the alkalies (Warshaw 1956). The  $FeS_2$  may occur within the clay (Krazewski 1972; Goldbery 1978), thereby generating the ferric sulfate solutions in situ. Alternatively, the jarosite could have precipitated directly, such as from ponded solutions (Gryaznov 1957; Alpers et al. 1992; Long et al. 1992), or the solutions could have been transported into the sedimentary unit from distal sources (Khlybov 1976; Dutrizac and Jambor 2000). Jarosite can assume a cubic morphology, suggesting that it forms as a pseudomorph after  $FeS_2$  (Silva et al. 2011b).

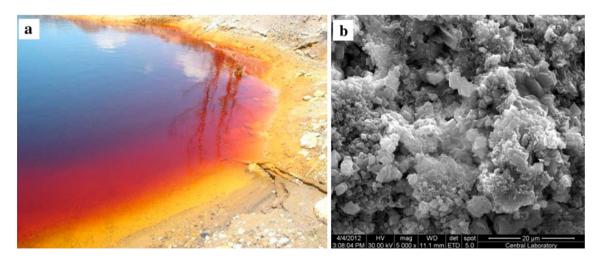


Fig. 10 a Field view and b SEM photomicrographs showing pseudocubic jarosite crystals

Date $\overline{\rm PH}$ $\overline{\rm AML}$ $\overline{\rm O7.11.2009}$ $2.72$ $2.85$ $NM$ $NM$ $NM$ $NM$ $0.8.12.2009$ $2.91$ $3.18$ $NM$ $NM$ $NM$ $NM$ $0.5.01.2010$ $2.86$ $2.93$ $NM$ $NM$ $NM$ $NM$ $0.5.01.2010$ $2.66$ $2.58$ $2.74$ $NM$ $NM$ $10.05.2010$ $2.69$ $2.63$ $3.06$ $3.06$ $3.22$ $17.07.2010$ $3.05$ $3.14$ $3.53$ $3.79$ $17.07.2010$ $3.06$ $2.00$ $2.12$ $3.60$ $3.76$ $17.07.2010$ $3.06$ $2.00$ $2.12$ $3.60$ $3.76$ $17.07.2010$ $3.06$ $2.200$ $2.12$ $3.73$ $3.79$ $17.04.2011$ $2.99$ $3.14$ $3.53$ $3.36$ $3.76$ $17.04.2011$ $2.90$ $3.12$ $3.20$ $3.31$ $3.76$ $17.04.2011$ $2.90$ $3.12$ $3.20$ $3.31$ $3.76$ $17.04.2011$ $2.99$ $3.269$ $3.31$ $3.32$ $3.76$ $17.04.2011$ $2.99$ $3.269$ $3.316$ $3.76$ $17.04.2011$ $2.99$ $3.269$ $3.316$ $3.76$ $17.06.2011$ $2.99$ $2.61$ $2.60$ $3.00$ $25.02.2012$ $2.99$ $2.61$ $2.77$ $2.76$ $26.02.2012$										Ī
AML I     AML 2     AML 3     AML 4       2.72     2.85     NM     NM     NM       2.91     3.18     NM     NM     NM       2.66     2.93     NM     NM     NM       2.69     2.63     2.74     NM     NM       3.05     3.06     3.06     NM     3.50     3.60       3.05     3.09     3.12     2.94     3.53     3.51     3.53       3.09     3.12     3.26     3.14     3.53     3.61     3.71       3.17     3.22     3.20     3.14     3.53     3.61     3.71       3.17     3.22     3.23     3.26     3.71     3.76     3.71       2.99     3.12     3.26     3.71     3.72     3.66     3.16       2.91     2.91	рН					Temperature (°C)	(0°C)			
2.72   2.85   NM   N	AML 1	AML 2	AML 3	AML 4	AML 5	AML 1	AML 2	AML 3	AML 4	AML 5
291   3.18   NM   NN   N   Substance   2.66   2.58   2.82   NM   N	2.72	2.85	NM	NM	MM	16.8	17.2	NM	MN	NM
286   2.93   NM   NM   NM   N     2.66   2.58   2.82   NM   N   N     2.69   2.63   2.63   2.74   NM   N   N     2.69   2.63   2.63   2.74   NM   N   N     2.91   2.96   3.05   3.12   3.66   NM   N   N     3.05   3.05   3.05   3.14   3.53   3.60   N   N   N     3.75   3.24   3.12   3.14   3.53   3.71   3.53   3.71     3.75   3.24   3.26   3.14   3.53   3.71   3.53   3.71     3.17   3.22   3.26   3.12   3.26   3.71   3.73     3.17   3.22   3.26   3.26   3.71   3.76   3.71     2.99   3.17   3.22   3.26   3.71   3.76   3.76     3.17   3.22   3.26   3.70   3.71   3.76   3.76     2.99   2.71   2.73   3.00   3.76 <t< td=""><td>2.91</td><td>3.18</td><td>NM</td><td>NM</td><td>MN</td><td>11.7</td><td>10.6</td><td>NM</td><td>NM</td><td>NM</td></t<>	2.91	3.18	NM	NM	MN	11.7	10.6	NM	NM	NM
2.66   2.8   2.82   NM   N     2.69   2.63   2.63   2.74   NM   N     2.91   2.96   3.05   3.06   NM   N     3.05   3.09   3.12   3.60   N   N   N     3.06   3.05   3.09   3.14   3.53   3.60   N   N   N     3.05   3.05   3.24   3.12   3.53   3.71   3.53   3.71     3.75   3.24   3.26   3.14   3.53   3.71   3.53   3.71     3.17   3.24   3.26   2.12   2.71   3.73   3.34     3.17   3.22   3.20   3.20   3.61   3.71     3.17   3.22   3.20   3.61   3.71     2.97   3.00   3.26   3.36   3.16     2.98   2.61   2.09   3.16   3.16     2.99   3.10   2.69   3.16   3.16     2.59   2.61   2.69   3.16   3.05     2.51   2.52   3.2	2.86	2.93	NM	NM	NM	7.5	7.9	NM	NM	NM
2.69   2.63   2.74   NM   NI     2.91   2.96   3.06   3.06   NM   NI     3.05   3.05   3.12   3.60   NM   NI     3.06   3.05   3.14   3.53   3.60   NM   NI     3.06   3.05   3.14   3.53   3.60   3.60   3.60   3.60   3.60   3.60   3.61   NI   3.60   3.60   3.60   3.71   3.23   3.71   3.23   3.71   3.23   3.71   3.23   3.71   3.23   3.71   3.72   3.71   3.72   3.71   3.74   3.71   3.74   3	2.66	2.58	2.82	NM	NM	17.9	17.3	17.9	NM	NM
2912963.06NMNI $3.05$ $3.09$ $3.12$ $3.60$ $3.60$ $3.06$ $3.05$ $3.14$ $3.53$ $3.06$ $3.05$ $3.14$ $3.53$ $1.99$ $2.00$ $2.12$ $2.71$ $3.75$ $3.24$ $3.26$ $3.71$ $3.75$ $3.24$ $3.26$ $3.71$ $3.75$ $3.24$ $3.26$ $3.71$ $2.9$ $2.00$ $2.12$ $2.71$ $2.9$ $3.12$ $3.26$ $3.71$ $2.97$ $3.00$ $2.93$ $3.39$ $3.17$ $3.22$ $3.26$ $3.16$ $2.97$ $3.00$ $3.09$ $3.46$ $2.97$ $3.00$ $3.09$ $3.46$ $2.97$ $3.00$ $3.09$ $3.46$ $2.97$ $3.00$ $3.09$ $3.16$ $2.97$ $2.01$ $2.03$ $3.16$ $2.59$ $2.61$ $2.69$ $3.16$ $2.51$ $2.61$ $2.69$ $3.16$ $2.73$ $2.61$ $2.73$ $3.01$ $2.74$ $2.92$ $2.73$ $3.01$ $2.74$ $2.92$ $2.73$ $3.01$ $2.71$ $2.79$ $2.73$ $3.01$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.74$ $2.90$ $2.77$ $2.74$ $2.90$ $2.12$ <t< td=""><td>2.69</td><td>2.63</td><td>2.74</td><td>NM</td><td>MN</td><td>21.1</td><td>26.2</td><td>25.1</td><td>NM</td><td>NM</td></t<>	2.69	2.63	2.74	NM	MN	21.1	26.2	25.1	NM	NM
3.05 $3.09$ $3.12$ $3.60$ $3.06$ $3.05$ $3.14$ $3.53$ $1.99$ $2.00$ $2.12$ $2.71$ $3.75$ $3.24$ $3.26$ $3.71$ $3.75$ $3.24$ $3.26$ $3.71$ $2.9$ $2.80$ $2.93$ $3.39$ $3.09$ $3.12$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $2.97$ $3.00$ $3.20$ $3.61$ $2.95$ $2.91$ $3.09$ $3.16$ $2.59$ $2.61$ $2.69$ $3.16$ $2.59$ $2.61$ $2.69$ $3.16$ $2.59$ $2.61$ $2.69$ $3.16$ $2.51$ $2.61$ $2.69$ $3.16$ $2.51$ $2.61$ $2.69$ $3.16$ $2.71$ $2.61$ $2.73$ $3.01$ $2.74$ $2.92$ $2.73$ $3.02$ $2.71$ $2.88$ $3.32$ $2.81$ $2.82$ $2.75$ $3.22$ $2.81$ $2.82$ $2.73$ $3.32$ $2.71$ $2.90$ $2.72$ $3.33$ $2.71$ $2.91$ $2.92$ $3.33$ $2.72$ $2.88$ $3.33$ $2.71$ $2.90$ $2.12$ $2.71$	2.91	2.96	3.06	NM	NM	25.9	31.3	28.1	NM	NM
3.06 $3.05$ $3.14$ $3.53$ $1.99$ $2.00$ $2.12$ $2.71$ $3.75$ $3.24$ $3.26$ $3.71$ $3.75$ $3.24$ $3.26$ $3.71$ $2.9$ $2.80$ $2.93$ $3.39$ $3.09$ $3.12$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $2.97$ $3.00$ $3.26$ $3.61$ $2.97$ $3.00$ $3.09$ $3.46$ $2.95$ $2.91$ $3.09$ $3.16$ $2.59$ $2.61$ $2.69$ $3.16$ $2.59$ $2.61$ $2.69$ $3.16$ $2.51$ $2.61$ $2.69$ $3.16$ $2.73$ $2.61$ $2.73$ $3.01$ $2.51$ $2.61$ $2.73$ $3.02$ $2.74$ $2.92$ $2.73$ $3.02$ $2.74$ $2.92$ $2.75$ $3.25$ $2.74$ $2.88$ $3.33$ $2.81$ $2.82$ $2.75$ $3.22$ $2.71$ $2.90$ $2.12$ $2.71$ $2.91$ $2.00$ $2.12$ $2.71$	3.05	3.09	3.12	3.60	3.22	25.6	26.7	28.7	30.6	30.5
1.99 $2.00$ $2.12$ $2.71$ $3.75$ $3.24$ $3.26$ $3.71$ $3.75$ $3.24$ $3.26$ $3.71$ $2.9$ $2.80$ $2.93$ $3.39$ $3.09$ $3.12$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.09$ $3.12$ $3.22$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $2.97$ $3.00$ $3.09$ $3.46$ $2.98$ $2.61$ $3.09$ $3.46$ $2.59$ $2.61$ $2.69$ $3.16$ $2.53$ $2.61$ $2.69$ $3.10$ $2.51$ $2.61$ $2.69$ $3.10$ $2.73$ $2.61$ $2.73$ $3.01$ $2.74$ $2.61$ $2.73$ $3.01$ $2.51$ $2.66$ $2.75$ $3.25$ $2.74$ $2.92$ $2.75$ $3.25$ $2.74$ $2.88$ $3.33$ $2.81$ $2.82$ $2.75$ $3.25$ $2.74$ $2.00$ $2.12$ $2.71$	3.06	3.05	3.14	3.53	3.15	23.6	22.9	22.0	23.1	22.4
3.75 $3.24$ $3.26$ $3.71$ $2.9$ $2.80$ $2.93$ $3.39$ $3.09$ $3.12$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $2.97$ $3.00$ $3.09$ $3.46$ $2.95$ $2.91$ $3.00$ $3.46$ $2.95$ $2.91$ $3.00$ $3.46$ $2.59$ $2.61$ $2.69$ $3.16$ $2.53$ $2.61$ $2.69$ $3.16$ $2.71$ $2.61$ $2.69$ $3.10$ $2.73$ $2.61$ $2.73$ $3.01$ $2.74$ $2.61$ $2.73$ $3.01$ $2.74$ $2.92$ $3.32$ $2.74$ $2.92$ $3.32$ $2.74$ $2.92$ $3.32$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.74$ $2.88$ $3.33$ $2.90$ $2.00$ $2.12$ $2.91$ $2.00$ $2.71$	1.99	2.00	2.12	2.71	2.65	15.1	15.6	15.9	14.6	15.3
2.9 $2.80$ $2.93$ $3.39$ $3.09$ $3.12$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $3.17$ $3.22$ $3.20$ $3.61$ $2.95$ $2.91$ $3.09$ $3.34$ $2.95$ $2.91$ $3.09$ $3.46$ $2.68$ $2.63$ $2.69$ $3.16$ $2.59$ $2.61$ $2.69$ $3.16$ $2.59$ $2.61$ $2.69$ $3.16$ $2.51$ $2.61$ $2.66$ $3.05$ $2.71$ $2.61$ $2.73$ $3.01$ $2.51$ $2.66$ $2.73$ $3.01$ $2.51$ $2.66$ $2.73$ $3.01$ $2.74$ $2.92$ $3.25$ $3.25$ $2.71$ $2.82$ $2.73$ $3.33$ $2.91$ $2.82$ $2.75$ $3.25$ $2.74$ $2.92$ $2.75$ $3.25$ $2.74$ $2.88$ $3.33$ $2.91$ $2.88$ $3.33$ $2.91$ $2.88$ $3.33$ $2.91$ $2.00$ $2.12$ $2.71$	3.75	3.24	3.26	3.71	3.79	12.5	9.5	11.1	11.8	11.7
3.09   3.12   3.20   3.61     3.17   3.22   3.20   3.61     3.17   3.22   3.22   3.86     2.97   3.00   3.09   3.46     2.95   2.91   3.00   3.46     2.95   2.91   3.00   3.46     2.68   2.63   2.69   3.16     2.59   2.61   2.69   3.16     2.53   2.61   2.69   3.10     2.53   2.61   2.69   3.10     2.51   2.61   2.69   3.10     2.53   2.61   2.69   3.10     2.51   2.61   2.69   3.05     2.71   2.61   2.73   3.01     2.59   2.74   2.92   3.32     2.74   2.92   3.25   3.25     2.74   2.88   3.32     2.81   2.82   3.33     2.91   2.88   3.33     2.91   2.00   2.12   2.71	2.9	2.80	2.93	3.39	3.88	10.9	11.5	12.6	12.0	12.1
3.17   3.22   3.22   3.86     2.97   3.00   3.09   3.46     2.95   2.91   3.09   3.46     2.95   2.91   3.00   3.46     2.95   2.91   3.00   3.46     2.59   2.61   3.00   3.46     2.53   2.61   2.69   3.16     2.53   2.61   2.66   3.05     2.71   2.61   2.66   3.01     2.59   2.61   2.73   3.01     2.71   2.61   2.73   3.01     2.51   2.61   2.73   3.01     2.51   2.66   2.75   3.25     2.74   2.92   3.25   3.25     2.74   2.92   3.25   3.25     2.74   2.88   3.33   3.25     2.81   2.82   2.88   3.33     2.91   2.90   2.12   2.71	3.09	3.12	3.20	3.61	4.35	13.4	11.9	11.8	12.2	12.0
2.97   3.00   3.09   3.34     2.95   2.91   3.00   3.46     2.68   2.63   2.69   3.16     2.59   2.61   2.69   3.10     2.53   2.61   2.66   3.01     2.53   2.61   2.66   3.01     2.71   2.61   2.66   3.01     2.73   2.61   2.73   3.01     2.71   2.61   2.73   3.01     2.73   2.61   2.73   3.01     2.74   2.61   2.73   3.01     2.51   2.66   2.75   3.25     2.74   2.92   3.25   3.25     2.74   2.86   2.75   3.25     2.74   2.82   2.75   3.25     2.81   2.82   2.75   3.32     2.99   2.00   2.12   2.71	3.17	3.22	3.22	3.86	3.96	18.7	19.3	21.0	21.0	20.1
2.95   2.91   3.00   3.46     2.68   2.63   2.69   3.16     2.59   2.61   2.69   3.10     2.53   2.61   2.66   3.05     2.71   2.61   2.65   3.01     2.73   2.61   2.73   3.01     2.71   2.61   2.73   3.01     2.74   2.61   2.73   3.01     2.51   2.66   2.73   3.01     2.51   2.66   2.75   3.25     2.74   2.92   3.32   3.25     2.74   2.66   2.75   3.25     2.74   2.82   2.75   3.25     2.81   2.82   2.75   3.32     2.99   2.00   2.12   2.71	2.97	3.00	3.09	3.34	3.45	23.7	24.8	25.0	28.5	27.7
2.68   2.63   2.69   3.16     2.59   2.61   2.69   3.10     2.53   2.61   2.69   3.10     2.53   2.61   2.66   3.05     2.71   2.61   2.65   3.05     2.71   2.61   2.73   3.01     2.59   2.74   2.92   3.32     2.51   2.66   2.75   3.25     2.74   2.92   3.32     2.51   2.66   2.75   3.25     2.74   2.88   3.33     2.74   2.88   3.33     2.74   2.88   3.33     2.91   2.82   2.88   3.33     2.91   2.00   2.12   2.71	2.95	2.91	3.00	3.46	3.20	27.3	28.9	28.2	26.7	25.4
2.59   2.61   2.69   3.10     2.53   2.61   2.66   3.05     2.71   2.61   2.66   3.05     2.71   2.61   2.73   3.01     2.59   2.74   2.92   3.32     2.51   2.66   2.75   3.25     2.74   2.66   2.75   3.25     2.74   2.66   2.75   3.25     2.74   2.66   2.75   3.25     2.74   2.66   2.75   3.25     2.74   2.66   2.75   3.25     2.74   2.60   2.75   3.23     1.99   2.00   2.12   2.71	2.68	2.63	2.69	3.16	2.77	26.6	24.9	24.1	25.3	25.1
2.53   2.61   2.66   3.05     2.71   2.61   2.73   3.01     2.71   2.61   2.73   3.01     2.59   2.74   2.92   3.32     2.51   2.66   2.75   3.25     2.74   2.66   2.75   3.25     2.74   2.66   2.75   3.25     2.81   2.82   2.88   3.33     2.81   2.82   2.88   3.33     1.99   2.00   2.12   2.71	2.59	2.61	2.69	3.10	2.85	9.3	9.3	8.0	12.0	9.6
2.71 2.61 2.73 3.01   2.59 2.74 2.92 3.32   2.51 2.66 2.75 3.25   2.74 2.66 2.75 3.25   2.74 2.66 2.75 3.25   2.81 2.82 2.88 3.33   1.99 2.00 2.12 2.71	2.53	2.61	2.66	3.05	2.77	7.6	7.6	7.0	8.9	7.5
2.59 2.74 2.92 3.32   2.51 2.66 2.75 3.25   2.74 2.66 2.75 3.25   2.74 2.66 2.75 3.22   2.81 2.82 2.88 3.33   1.99 2.00 2.12 2.71	2.71	2.61	2.73	3.01	3.20	9.4	11.5	11.9	10.8	10.9
2012 2.51 2.66 2.75 3.25   2012 2.74 2.66 2.75 3.22   2012 2.81 2.82 2.88 3.33   101 1.99 2.00 2.12 2.71	2.59	2.74	2.92	3.32	3.70	9.1	10.0	7.6	8.2	8.5
2012     2.74     2.66     2.75     3.22       2.81     2.82     2.88     3.33       uun     1.99     2.00     2.12     2.71	2.51	2.66	2.75	3.25	3.06	22.1	22.6	21.9	21.3	21.4
2.81 2.82 2.88 3.33 um 1.99 2.00 2.12 2.71	2.74	2.66	2.75	3.22	2.86	26.9	26.7	26.8	27.3	27.1
1.99 2.00 2.12 2.71	2.81	2.82	2.88	3.33	3.30	17.39	17.91	18.66	18.39	17.95
	1.99	2.00	2.12	2.71	2.65	7.5	7.6	7.0	8.2	7.5
Maximum 3.75 3.24 3.26 3.86 4.35	3.75	3.24	3.26	3.86	4.35	27.3	31.3	28.7	30.6	30.5

365

Date	Redox po	Redox potential (mV)				Electrical	Electrical conductivity (µS/cm)	(µS/cm)			Salinity (%)	(%)			
	AML 1	AML 2	AML 3	AML 4	AML 5	AML 1	AML 2	AML 3	AML 4	AML 5	AML 1	AML 2	AML 3	AML 4	AML 5
07.11.2009	243	243	NM	MN	NM	5110	7310	MN	MN	MN	2.7	4.0	NM	MN	MN
08.12.2009	237	221	MN	MN	MN	5130	7280	MN	MN	MN	2.7	3.9	MN	MN	MN
05.01.2010	234	229	MN	MN	MN	4860	6410	MN	MN	MN	2.5	3.4	MN	MN	MN
02.05.2010	245	251	233	MN	MN	3970	5430	2530	MN	MN	2.0	2.8	1.2	MN	MN
10.05.2010	249	253	246	MN	MN	3970	5350	2510	MN	MN	2.0	2.9	1.2	MN	NM
04.06.2010	258	261	252	MN	MN	4120	5590	2180	MN	MN	2.1	3.0	1.3	MN	NM
17.07.2010	256	255	257	230	254	4960	6434	2990	3180	3070	2.4	3.5	1.5	1.6	1.5
15.09.2010	256	257	251	229	251	5160	7070	4050	3660	3810	2.7	3.9	2.1	1.8	1.9
27.11.2010	242	242	237	206	210	4460	5450	3430	2732	3490	2.3	2.9	1.7	0.7	1.7
01.04.2011	201	228	227	201	198	3040	5550	3080	3240	1345	1.4	2.9	1.5	1.6	0.5
17.04.2011	233	235	228	205	203	2985	4820	2770	3160	1085	1.3	2.5	1.3	1.4	0.3
22.04.2011	237	234	230	207	176	4120	5340	2870	3040	674	2.1	2.8	1.3	1.4	0.1
19.05.2011	235	237	238	201	194	4130	5350	2920	3120	1384	2.2	2.8	1.4	1.5	0.5
17.06.2011	253	251	246	217	210	4120	5340	2960	3110	1736	2.3	2.9	1.4	1.6	0.7
04.07.2011	256	260	254	227	242	4180	5440	2990	3070	2050	2.2	2.9	1.5	1.5	0.9
23.08.2011	253	254	250	224	247	4730	6530	3840	3370	2880	2.5	3.6	1.9	1.7	1.4
19.11.2011	228	227	222	201	214	5210	7010	4280	3550	2940	2.7	3.8	2.1	1.7	1.4
26.11.2011	226	221	218	199	214	5260	7030	4360	3620	3030	2.7	3.7	2.2	1.8	1.4
06.12.2011	232	224	217	200	211	4370	7030	4340	3600	2990	2.2	3.8	2.2	1.7	1.3
25.02.2012	235	233	221	198	177	4320	5820	3380	3240	1040	2.2	1.4	1.6	1.5	0.4
19.05.2012	239	245	238	217	220	4235	5640	3050	3100	1915	2.7	3.0	1.5	1.4	0.8
03.07.2012	242	248	243	215	236	4280	5870	3230	3260	2870	2.2	3.2	1.6	1.6	1.4
Mean	240.45	241.31	237.26	211.06	215.43	4396.36	6049.72	3250.52	3190.75	2269.31	2.27	3.16	1.60	1.53	1.01
Minimum	201	221	217	198	176	2985	4820	2180	2732	674	1.3	1.4	1.2	0.7	0.1
Maximum	258	261	257	230	254	5260	7310	4360	3660	3810	2.7	4.0	2.2	1.8	1.9
NM not measured	Isured														

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Table 3 continued

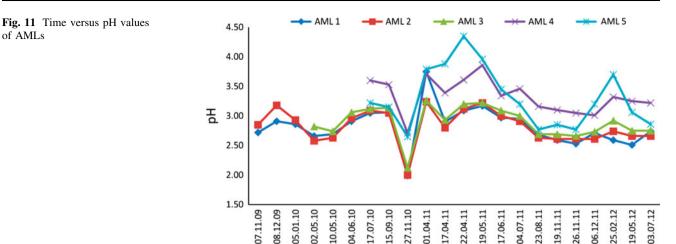


Figure 10 shows a field view and SEM image of jarosite pseudomorph after  $FeS_2$ .

The formation of ammoniojarosite is rare in nature because of the scarcity of concentrated ammonium-bearing solutions (Dutrizac and Jambor 2000). The ammonium originates from the decomposition of organic substances, whereas the Fe and sulfate are derived from the oxidation of associated FeS<sub>2</sub>. Aside from the scarcity of NH<sub>4</sub>-rich solutions, the K<sup>+</sup>:NH<sub>4</sub><sup>+</sup> partition coefficient strongly favors the formation of jarosite rather than ammoniojarosite (Dutrizac and Jambor 2000).

### Hydrogeochemical Data

The oxidation of FeS<sub>2</sub>, a common form of sulfur bearing Fe mineral associated with coal, often results in the formation of AMD, which typically forms extremely acidic lakes in the depressions of abandoned open-pit mines (Gunduz et al. 2007). The process of FeS<sub>2</sub> oxidation is complex and involves both chemical and biological mechanisms as well as a number of controls (Blodau 2006). The reaction of FeS<sub>2</sub> with oxygen and water produces a solution of ferrous

sulfate and sulfuric acid. Ferrous Fe can further be oxidized producing additional acidity.

AMLs are commonly observed in Germany (Blodau et al. 1998; Abel et al. 2000; Weber 2000; Lessmann et al. 2003), Greece (Triantafyllidis and Skarpelis 2006), and the United States (Shevenell et al. 1999; Nordstorm et al. 2000). A number of such lakes were also recently discovered in the Can coal basin located in Canakkale province in northwestern Turkey where sulfur-rich lignite is extracted by way of surface mining operations (Gunduz et al. 2007; Baba et al. 2009).

## Field Parameters

The field parameter results of AML measurements are listed in Table 3. These results show that the pH of AMLs ranged from 1.99 to 4.35 between 2009 and 2012 (Fig. 11). The generation of low pH is attributed to sulfide oxidation found primarily in FeS<sub>2</sub> and, to a certain extent, in other sulfide minerals (Blodau 2006). The pH of AMD can be unstable because of a general tendency for the exsolution of dissolved CO<sub>2</sub> and H<sub>2</sub>S, the dissolution of O<sub>2</sub>, and the

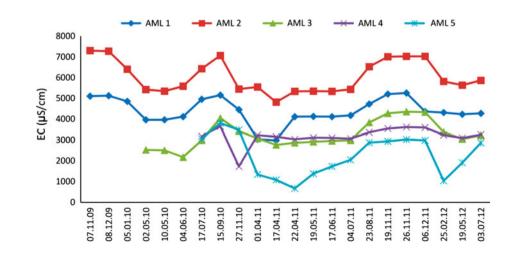


Fig. 12 Time versus EC values of AMLs

Major cations	Na <sup>+</sup>					$K^+$				
Date	AML 1	AML 2	AML 3	AML 4	AML 5	AML 1	AML 2	AML 3	AML 4	AML 5
08.12.2009	125.51	279.69	NM	NM	NM	0.99	2.93	NM	NM	NM
17.07.2010	104.8	164.27	53.02	NM	NM	0.99	0.99	3.36	NM	NM
01.04.2011	115.92	176.93	76.93	216.15	44.39	1.90	1.90	2.09	9.94	9.10
19.05.2011	96.3	181.65	74.92	207.73	52.14	1.40	0.87	2.30	10.07	6.21
04.07.2011	108.8	168.30	98.60	265.70	81.21	1.60	2.00	4.80	15.80	12.41
06.12.2011	105.38	164.17	87.49	217.03	48.11	1.80	2.94	4.30	8.82	16.57
25.02.2012	77.01	157.97	108.52	243.13	43.97	0.95	2.00	2.00	12.00	4.78
03.07.2012	81.43	146.60	137.55	197.72	50.41	1.25	2.89	2.70	7.14	5.88
Mean	101.89	179.94	91.00	224.57	53.37	1.36	2.06	3.07	10.62	9.15
Minimum	77.01	146.60	53.02	197.72	43.97	0.95	0.87	2.00	7.14	4.78
Maximum	125.51	279.69	137.55	265.70	81.21	1.90	2.94	4.80	15.80	16.57
08.12.2009	232.53	261.55	NM	NM	NM	444.25	513.84	NM	NM	NM
17.07.2010	230.53	230.53	63.25	NM	NM	377.65	477.65	98.51	NM	NM
01.04.2011	178.10	278.10	98.53	125.43	49.67	350.70	450.70	147.28	386.49	122.06
19.05.2011	198.40	238.17	64.69	101.00	34.40	392.80	470.20	177.89	315.80	107.70
04.07.2011	228.90	357.80	80.60	119.40	62.61	478.10	520.10	229.00	411.30	172.10
06.12.2011	174.21	306.63	69.08	101.88	73.95	304.34	451.29	212.02	306.87	145.53
25.02.2012	182.00	306.00	92.00	118.00	28.74	356.10	461.90	245.10	368.80	85.47
03.07.2012	134.40	271.20	68.48	94.97	53.61	290.20	445.30	177.55	302.30	141.30
Mean	194.88	281.24	76.66	110.11	50.49	374.20	473.87	183.90	348.59	129.02
Minimum	134.40	230.53	63.25	94.97	28.74	290.20	445.30	98.51	302.30	85.47
Maximum	232.53	357.80	98.53	125.43	73.95	478.10	520.10	245.10	411.30	172.10

Table 4 Major cation values of AMLs (mg  $L^{-1}$ )

NM not measured

**Table 5** Major anion values of AMLs (mg  $L^{-1}$ )

Major	$F^{-}$					$Cl^{-}$					$SO_4^{2-}$				
cations Date	AML 1	AML 2	AML 3	AML 4	AML 5	AML 1	AML 2	AML 3	AML 4	AML 5	AML 1	AML 2	AML 3	AML 4	AML 5
08.12.2009	2.94	2.45	NM	NM	NM	44.63	100.84	NM	NM	NM	3857.26	5296.00	NM	NM	NM
17.07.2010	1.40	1.40	3.10	NM	NM	78.30	78.30	39.80	NM	NM	3512.40	5012.40	903.80	NM	NM
01.04.2011	1.12	1.12	2.65	1.20	1.30	73.20	73.20	38.75	99.41	104.56	3017.26	5017.26	988.63	1018.45	895.00
19.05.2011	1.89	1.30	1.29	1.55	1.58	53.60	182.42	48.67	83.54	65.87	3192.50	5273.24	1455.12	1823.67	772.24
04.07.2011	2.98	2.77	2.91	1.80	1.70	63.00	143.00	69.00	106.00	72.00	3000.00	4600.00	1500.00	1700.00	1000.00
06.12.2011	4.12	3.68	4.80	2.27	3.24	65.33	191.45	73.90	89.25	85.27	3122.16	4413.20	1656.70	1036.76	964.55
25.02.2012	3.05	3.02	3.09	2.10	3.12	46.00	97.00	67.00	100.00	54.00	3260.00	5370.00	2520.00	1760.00	594.00
03.07.2012	1.71	2.76	1.48	3.76	1.93	42.78	83.01	44.28	76.16	53.84	3181.56	4774.08	2330.27	1605.84	1091.64
Mean	2.40	2.31	2.76	2.11	2.14	58.35	118.65	54.48	92.39	72.59	3267.89	4969.52	1622.07	1490.78	886.23
Minimum	1.12	1.12	1.29	1.20	1.30	42.78	73.20	38.75	76.16	53.84	3000.00	4413.20	903.80	1018.45	594.00
Maximum	4.12	3.68	4.80	3.76	3.24	78.30	191.45	73.90	106.00	104.56	3857.26	5370.00	2520.00	1823.67	1091.64

NM not measured

consequent oxidation and hydrolysis of dissolved Fe and Mn (Rose and Cravotta 1998; Cravotta et al. 1999). The electrical conductivity (EC) values of AMLs ranged between 674 and 7310  $\mu$ S/cm (Fig. 12). These values were consistent with EC values of numerous AMLs published in the literature (Lessmann et al. 1999; Espana et al. 2008).

Table 6 Heavy-metal and trace-element values of AMLs  $(\mu g L^{-1})$ 

	Al	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	S	Zn
AML no. 1: Date												
08.12.2009	278245	3.9	14.16	2030	26	340.6	88093	74630	1009	1.67	1350000	4666
17.07.2010	247950	4.1	20	1270	19	220	90315	67760	520	1.6	1289000	2820
01.04.2011	252683	5.2	21.3	1938	33.75	312.84	83739	72413	1071	1.95	1511000	4471
04.07.2011	286613	6	15	2150	20	365	91903	83947	1075	2	1275000	4881
06.12.2011	204677	4.5	16	1784	25	197.89	90108	64201	515	1.8	1434000	3569
25.02.2012	219618	2.9	13.8	1524	23	196.4	54198.9	66489	579	1.3	1206000	2758
03.07.2012	201450	3.1	11.2	1100	17	190	75580	67610	493	1.1	1318000	2990
Mean	241605.14	4.24	15.92	1685.14	23.39	260.39	81990.98	71007.14	751.71	1.63	1340428.6	3736.42
Minimum	201450	2.9	11.2	1100	17	190	54198.9	64201	493	1.1	1206000	2758
Maximum	286613	6	21.3	2150	33.75	365	91903	83947	1075	2	1511000	4881
EPA (2003)	200	10	5	_	100	1000	300	50	_	15	_	5000
WHO (2004)	200	10	5	_	50	2000	200	400	20	10	_	5000
ITASHY (2005)	200	10	5	_	50	2000	200	50	20	10	_	5000
AML no. 2: Date	200	10	5	-	50	2000	200	50	20	10	-	5000
08.12.2009	311980	14.9	29.1	4781	40	645	284540	206430	2150	0.4	1796000	16270
17.07.2010												
	345800	16.2	21.8	4290	43	610	328296	217935	2436	0.5	1991400	15481
01.04.2011	327510	12.9	19.3	4954	39	486	227210	225650	1800	0.25	1851000	15730
19.05.2011	346624	15.3	20.9	3820	41	590	270414	208360	2396	0.4	1898000	13117
04.07.2011	360675	12.1	27.9	4245	51	952	329770	187079	2367	0.5	1932000	12037
06.12.2011	352699	17.7	26.2	4087	57	457	277169	222585	2149	0.3	1839000	15013
25.02.2012	308835	11.4	25.9	3918	45	420	154650	182203	2022	0.44	2084000	10281
03.07.2012	356310	14.9	18.7	3460	34	511	284400	216580	1840	0.1	2125000	9610
Mean	338804.12	14.42	23.73	4194.37	43.75	583.87	269556.13	208352.75	2145	0.36	1939550	13442.37
Minimum	308835	11.4	18.7	3460	34	420	154650	182203	1800	0.1	1796000	9610
Maximum	360675	17.7	29.1	4954	57	952	329770	225650	2436	0.5	2125000	16270
EPA (2003)	200	10	5	-	100	1000	300	50	-	15	-	5000
WHO (2004)	200	10	5	-	50	2000	200	400	20	10	-	5000
ITASHY (2005)	200	10	5	-	50	2000	200	50	20	10	-	5000
AML no. 3: Date												
17.07.2010	118745	6.1	20.6	835	94	143	27780	24010	510	2.1	577000	2370
01.04.2011	124792	5.3	19	1220	150	190	28721	25650	578	1.9	612380	1896
19.05.2011	123639	5.9	11.43	1317	87	103	31790	20213	614	1.5	657300	2015
04.07.2011	129498	5	12.5	1168	96	155	34370	27574	580	1	682000	1238
06.12.2011	142765	6.2	15.78	1289	146	152	31951	24322	502	1.8	786400	2211
25.02.2012	155878	7.1	17.2	1316	133	79	21550	33820	563	2	914000	3923
03.07.2012	166730	6.3	11.63	780	78	101	35962	21370	437	1.3	1143800	3120
Mean	137435.28	5.98	15.44	1132.14	112	131.86	30303.43	25279.86	540.57	1.65	764413.33	2396.14
Minimum	118745	5	11.43	780	78	79	21550	20213	437	1	577000	1238
Maximum	166730	7.1	20.6	1317	150	190	35962	33820	614	2.1	1143800	3923
EPA (2003)	200	10	5	_	100	1000	300	50	_	15	_	5000
WHO (2004)	200	10	5	_	50	2000	200	400	20	10	_	5000
ITASHY (2005)	200	10	5	_	50	2000	200	50	20	10	_	5000
AML no. 4: Date	200	10	5		50	2000	200	50	20	10		2000
01.04.2011	20600	2.37	6.9	930	4	210	9670	35480	410	0.7	585600	1720
19.05.2011	20000 19687	2.97	0.9 4.7	930 836	4 1.7	174	9670 9645	33480 38603	429.6	0.7	585000 597490	1720
04.07.2011	19687		4.7 6.2	830 819.3	1.7 4	174		38003 32737	429.6 354	0.9 3	613700	1549 1607
04.07.2011	10884	4	6.2 5.5	819.5 713.4		114	7142	32737 31546	334 321		653860	1982
00.12.2011	19703	3.2	5.5	/13.4	1.4	152	7588	31340	321	0.4	00000	1902

Table 6 continued

	Al	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	S	Zn
25.02.2012	18262	2.8	8.1	719	2.3	76	9856	32509	229	0.3	726000	611
03.07.2012	20531	2.7	4.2	669	1.1	69	8563	30850	210	0.1	734900	790
Mean	19291.8	3	5.93	781.11	2.41	129.16	8744	33620.83	325.6	0.9	651925	1376.5
Minimum	16884	2.37	4.2	669	1.1	69	7142	30850	210	0.1	585600	611
Maximum	20600	4	8.1	930	4	210	9856	38603	429.6	3	734900	1982
EPA (2003)	200	10	5	-	100	1000	300	50	-	15	-	5000
WHO (2004)	200	10	5	-	50	2000	200	400	20	10	-	5000
ITASHY (2005)	200	10	5	-	50	2000	200	50	20	10	-	5000
AML no. 5: Date												
01.04.2011	36810	3.05	3	210	1.9	18	15680	12240	110	2.2	286000	960
19.05.2011	38940	3.3	2.4	275	1.73	16.48	17004	11876	148	1.5	295400	764
04.07.2011	62880	2.1	2.39	468.2	2	27.7	25335	19978	183	1	393000	1238
06.12.2011	41339	3.1	2.03	328	1.76	13.28	18188	18200	139	0.8	201500	930
25.02.2012	18192	2.9	1.05	150.3	0.5	12.9	12330	9880	77	0.4	173000	438
03.07.2012	51494	2.67	0.9	350	0.3	11.7	14590	18290	112	0.1	387500	1210
Mean	41609.16	2.85	1.96	296.91	1.36	16.67	17187.83	15077.33	128.17	1	289400	923.33
Minimum	18192	2.1	0.9	150.3	0.3	11.7	12330	9880	77	0.1	173000	438
Maximum	62880	3.3	3	468.2	2	27.7	25335	19978	183	2.2	393000	1238
EPA (2003)	200	10	5	-	100	1000	300	50	-	15	-	5000
WHO (2004)	200	10	5	-	50	2000	200	400	20	10	-	5000
ITASHY (2005)	200	10	5	-	50	2000	200	50	20	10	-	5000

Eh values are likely to be meaningful in high-Fe and lowpH waters (Nordstrom et al. 1979; Sracek et al. 2004). Eh values of AMLs are between 176 and 261 mV. The Eh values probably reflect the variations in the dominant  $Fe^{2\pm}$  $Fe^{3+}$  redox couple because concentrations of Fe were generally high. In general, temperature of surface waters change with geographical location, altitude, season, time of day, and depth of water body (Sracek et al. 2004).

# Water Chemistry

Sulfate is the predominant anion in all of AMLs in the study area. Sulfate levels were observed to range between 594 and 5370 mg L<sup>-1</sup> (mean 2447.30), which exceeds the water-quality standard of 250 mg L<sup>-1</sup> by a factor of 10 (USEPA 2003; World Health Organization [2004]; ITA-SHY 2005) (Table 4). Similar values have been measured in other AMLs worldwide (Bachmann et al. 2001; Trian-tafyllidis and Skarpelis 2006; Espana et al. 2008). Mg and Ca values ranged between 28.74 and 357.8 and between 85.47 and 520.1 mg L<sup>-1</sup>, respectively (Table 5).

Most metals have increasing ionic solubility under acidic and oxidizing conditions. A typical attenuation sequence for AMD in surface water is described in Plumlee (1999). When AMD enters a stream, it is progressively diluted, which causes an increase in pH. This leads to precipitation of the typical orange ferric (hydr)oxide colloids and coatings commonly observed in acidic mine effluents. As the pH of the water continues to increase, Al and Mg precipitates form because sorption onto the suspended Fe, Mn, and Al particulates becomes more effective.

Fe and Al are the most predominant metal species in AMLs in the study area. Similar values have been measured in other AMLs worldwide (Silva et. 2011a; Brake et al. 2001; Bladau 2006; Espana et al. 2008). High Fe levels are typically characteristic of FeS<sub>2</sub> oxidation. Fe concentrations ranged between 7.14 and 329.77 mg  $L^{-1}$ (Table 6). The concentration of Al was between 16.88 and  $360.67 \text{ mg L}^{-1}$ . The observed high aluminum concentrations are also a clue for the high clay content of the coals and regional rocks. High S concentrations were found in all five lakes, the maximum of which reached a level as high as 2125 mg  $L^{-1}$ . The high S concentrations are no surprise because it has been well known that Can coals contain high S contents. These lakes also have extremely high concentrations of manganese, which were observed to range between 9.88 and 225.65 mg L<sup>-1</sup>. Manganese contents are closely related to the lithology and the abundance of clay minerals. All of these values exceed the ITASHY (2005), United States Environmental Protection Agency (2003) and WHO (2004) water-quality standards value.



Fig. 13 Snapshots from AML no. 2 during field work

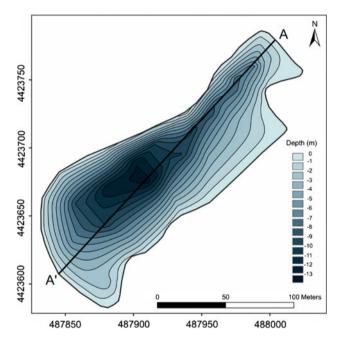


Fig. 14 Bathymetry of AML no. 2 (July 2010)

Maximum and minimum nickel concentration in AMLs were measured to be 2436 and 77  $\mu$ g/L, respectively (Table 6). The mean value was 778.21  $\mu$ g/L, which is

Fig. 15 The vertical profile on  $A-A^1$  cross-section

approximately 40 times greater than the ITASHY and WHO standard value. Cobalt is a rare element in the earth's crust and thus not typically found in high concentrations in surface waters. Thus, no standard value has been set for this chemical and for its compounds despite the fact that they are extremely toxic for humans (Okumusoglu 2009). Zn values ranged between 438 and 16270  $\mu$ g/L (mean 4374.95). Total Cd levels ranged between 0.9 and 29.1  $\mu$ g/L (mean 12.59), which exceeds all national and international standard values of 5  $\mu$ g/L. The Pb levels in AMLs ranged between 0.1 and 3  $\mu$ g/L (mean 1.1).

### Limnology of AML 2

Morphological and some chemical properties of AML no. 2 were studied during July 2010 (Fig. 13). The bathymetry of the lake was measured using a 30-m marine rope marked at 1-m intervals that was tied to a 1-kg weight. Lake depth was measured at 15-m intervals along a 235-m long transect (A-A') across the longest part of the lake for a total of 15 depth measurements (Fig. 14). The deepest point in the lake was 13.45 m deep, and the average depth of the lake was 7.41 m (Fig. 15). The perimeter and surface area of the lake were 616 and 15.55 m<sup>2</sup>, respectively. pH concentrations increased as depth increased; the average pH was

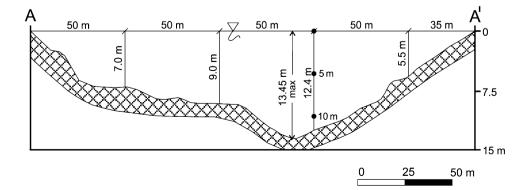




Fig. 16 Environmental impacts of AMD in study area

3.32. Iron concentrations ranged between 328.29 and 461.83 mg  $L^{-1}$ , and concentrations increased with depth.

Environmental Effects of AMLs

Water contaminated by AMD, often containing increased concentrations of metals, can be toxic to aquatic organisms, leaving receiving streams devoid of most living creatures (Kimmel 1983). In addition, some metals bioaccumulate in the aquatic food chain.

The AMLs are located at the upstream areas of the Kocacay Stream, which passes through Etili village, Can and Biga counties, and eventually discharges into the Sea of Marmara to the north. Several residential areas along this stream system, including Keciagili, Etili, and Can, obtain drinking water from wells within the flood plain (Fig. 16). Thus, the water quality in Kocacay Stream and its tributaries have great importance for providing safe drinking water to the local residents.

pH values of the AMLs and surrounding creeks were between 2.85 and 5.75 (Fig. 17). The surrounding creeks drain into Katran Creek to the northeast, which is a tributary of Kocacay Stream. Coal-mining activity has caused the flow direction of some creeks to change in the study area.

One of the AMLs (AML no. 2) was discharged to Kocacay Stream in 2008, resulting in serious environmental problems. It is also worthwhile to mention that in Can Basin, some fish deaths have been observed because of the seepage and discharge of the AML waters into surface waters (Sanliyuksel and Baba 2011). However, this lake reoccurs from time to time over very short periods.

The majority of this type of pollution results from old mining operations that have simply been abandoned after coal was extracted in the study area. The open-pit coal mines, where waste rocks are placed around the mining area, are exposed to weathering and leaching by rain. The tailings and altered volcanic rocks around mining area are composed of various kinds of minerals containing high amounts of heavy metals. Metals in the altered rocks and tailings can be released by the low-pH AMD and transported by rainwater, surface water, and groundwater away from the mine. Residual-waste crushed rock from mining activities represents a major source for metal-rich AMD water. The tailing minerals can release metal ions over long time periods due to oxidation of sulfide minerals leading to the production of sulfuric acid. In addition, groundwater moves from the topographically high tributary–watershed areas (Piren Hill) toward the topographically lower areas (coal mine sites) (Fig. 18).

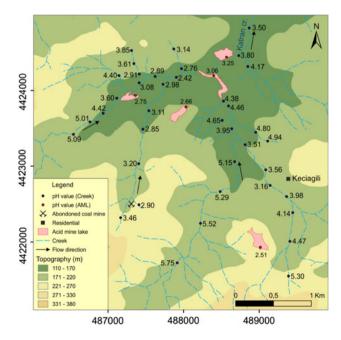


Fig. 17 pH distribution of creeks and AMLs (May 2012)

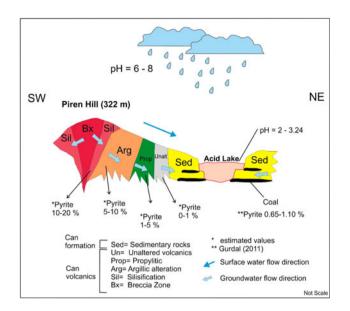


Fig. 18 A simplified conceptual model of AML

# Conclusion

This study focused on characterizing the geochemistry and mineralogy of AMD from the Can region, northwest Turkey. FeS<sub>2</sub> is the main cause of water pollution originating from abandoned coal mine deposits. Oxidation of FeS<sub>2</sub> gives rise to the generation of AMD, which is one of the most significant environmental consequences of mining operations, especially tailing dumps. These acidic waters can typically dissolve metals, metalloids, and other toxic materials and pose many environmental problems. Therefore, the study of the geochemistry of FeS<sub>2</sub> and its oxidation regarding AMD generation can help in devising minemanagement plans to help minimize the environmental impact associated with AMD. Based on the results of this study, the lakes exhibited characteristics typical of AMLs, including reddish-brown color, low pH, and high heavymetal and trace-element concentrations. The concentrations of Al, Fe, and Mn were found to exceed national and international water-quality standards.

Management of AMD aims to decrease the impact of the effluent to levels that can be tolerated by the environment without significant damage. To minimize this pollution, precautions must be taken to ensure that rainwater does not come into contact with FeS<sub>2</sub>. Abandoned waste and lack of rehabilitation measures on closure could be considered as the consequence of poor mining operations. Particularly, in mining areas with high FeS<sub>2</sub> content, special precautionary measures should always be implemented on completion of the operational phase of mining to prevent degradation of water quality and the environment.

Both the AMLs and Can county suffer from the same hydrochemical problem: the oxidation of sulfide minerals in mine waste and consequent acidification of mining lakes and leaching of metals from the sediment and groundwater. Therefore, it is very important to monitor the effect of this wastewater on water resources and soil.

Acknowledgments The authors are grateful to Mehmet ali Yucel and Firat Sengun for their support for field studies and suggestions and Mehmet Karadeniz for helpful comments. Herb Maier is acknowledged for proofreading and suggestions. This research was funded by the Teck Mining Company and partially supported by the Canakkale Onsekiz Mart University, Scientific Research Project No. 2011/082.

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