

Environmental pollutants enrichment in Soil, surface sediments and water resources by Mining activities, a case study; Zarshuran mine, NW Iran

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Abstract

Contamination of soil and water environment by heavy metals in a mining area and their effect on the soil and water characteristics were determined. Despite the ever-increasing development in mining activities in Iran, a little attention has been given to the environmental impacts in some cases. In this study, distribution of pollutants (As, Sb, Ag, and Zn) in natural resources such as surface soil, sediments and water, in addition to adjacent rock of river bed has been investigated. Correlations between these elements, sizes of sedimentary grains, and the reasons of solution and precipitation of minerals have also been studied and compared. Additionally the effects of some of the natural geochemical barriers on transferring elements have been introduced. Distribution of pollutant elements from distances far from sources of pollution can be traced in the regional plants. Finally, while presenting environmental and health problems, some practical solutions for the prevention of pollution distribution is cited.

Keywords: Zarshuran, Pollution Arsenic, sediment and geochemistry.

Introduction:

Indiscrete disposal of industrial waste leads to several geo-environmental disasters on land and it has huge socio-economic effects on the population residing in the impacted area especially in terms of livelihood and health. Once this happens, it becomes a news item and comes into focus for geo-environmental evaluation. Mining and processing of heavy and rare earth minerals inevitably involves distress of the land environment, the magnitude and intensity of which depends on the type of chemicals and processes used, the efforts taken in the management of waste as well as on environmental fragility of location. It also involves extreme disturbance to local residents by increased diseases, water pollution and destruction of farmland thereby violating the rights of local communities. Some of the contaminants are even classified as human carcinogens by international and federal health agencies (Adler & Rascher, ۲۰۰۷; APHA, ۲۰۰۵; Bell, ۱۹۹۸).

Numerous anthropogenic sources of pollutants can contaminate the soil and water environment, including inputs from waste waters flowing from mines and waste storage, runoff of pesticides from agricultural land or atmospheric deposition (Ackman & Klienmann, ۱۹۸۸; Song et al. ۲۰۱۰). A common problem in vicinity of mines is distribution of hazardous elements in the surrounding environment. In most cases, several factors control their migration. The factors underlying pollution and recipient parameters of these toxic elements are diverse. The elements studied in this investigation are considered as toxic pollutants. They are many products of mining at the Zarshuran ore deposit. Generally, bed sediments and suspended materials in the river water are major places of receipt and storage of pollutants in the aqueous system and for economic, hygienic and ecologic reasons; these are highly important (Bell, ۱۹۹۸). Soil As concentration abruptly decreased with an increase in the distance from the polluting source. Arsenic accumulated in significantly different levels between leafy vegetables and non-leafy vegetables (Liao et al, ۲۰۰۵).

In this area, it has been a major notice on black shale deposits, because they are major host of Arsenic ore deposits. Arsenic (As) naturally occurs in the form of sulfides in association with sulfides of ores of silver, lead, copper, nickel, etc (Liao et al, ۲۰۰۴; Virkutyte & Sillanpaa, ۲۰۰۶). Arsenic mining in Zarshuran introduced this element in surrounding environment. Erosion factors are able to transport and distribute these elements in various ways easily. From pollutant distribution mechanisms, the following can be mentioned: weathering, flood leaching and wind that all of them are active in Zarshuran area. Analysis of water, river sediments, soil, adjacent rock of riverbed and plant samples, reveal very high degrees of these pollutant elements, which are present far from sources of pollution. Soil As was reported in high quantities through atmosphere deposition and As containing water around some industrial areas due to mining and smelting activity (Gidhagen et al., ۲۰۰۲). However, the dramatic increase in population resulted in an enormous consumption of the world's water reserves (Ho et al., ۲۰۰۳). Natural contamination of water resources mainly results from normal geological phenomena such as ore formation (Al Fraij et al., ۱۹۹۹; Williamson et al., ۱۹۹۲). High concentrations of arsenic in groundwater in addition this area have been reported from several countries, including Argentina, Bangladesh, Chile, China, India, Japan, Mexico, Mongolia, Nepal, Poland, Taiwan, Vietnam, and some parts of the United States (Das et al, ۲۰۰۴).

Geological setting:

Takab area is located in Northwest of Iran in the west Azerbaijan province and in terms of morphology; it belongs to cold and mountainous areas. On the basis of annually isohyets contours, the rate of rainfall in this area varies between ۲۰۰ to ۴۵۰ mm.

The average altitude of Zarshuran ore deposit from sea level is about ۲۳۵۰m and the difference between maximum and minimum altitude of basin is ۵۶۹m. The Zarshuran River is a relatively small river, which passes toward the south, joins to Saroque River and finally forms Zarrineh rood, which enters the Orummieh Lake. The Zarshuran, Alochelo, Yar Aziz and Shirmard villages are in direct contact with river (figure.۱). The history of mining in this area is long-term and the evidences can be traced back to ۱۴۰۰ years ago.

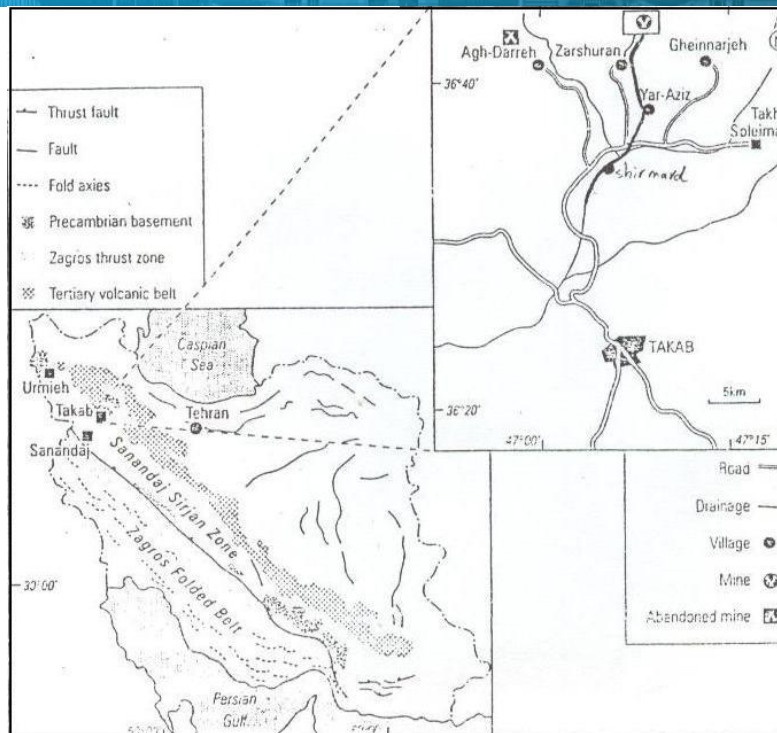


Fig ۱: The location of studied area.

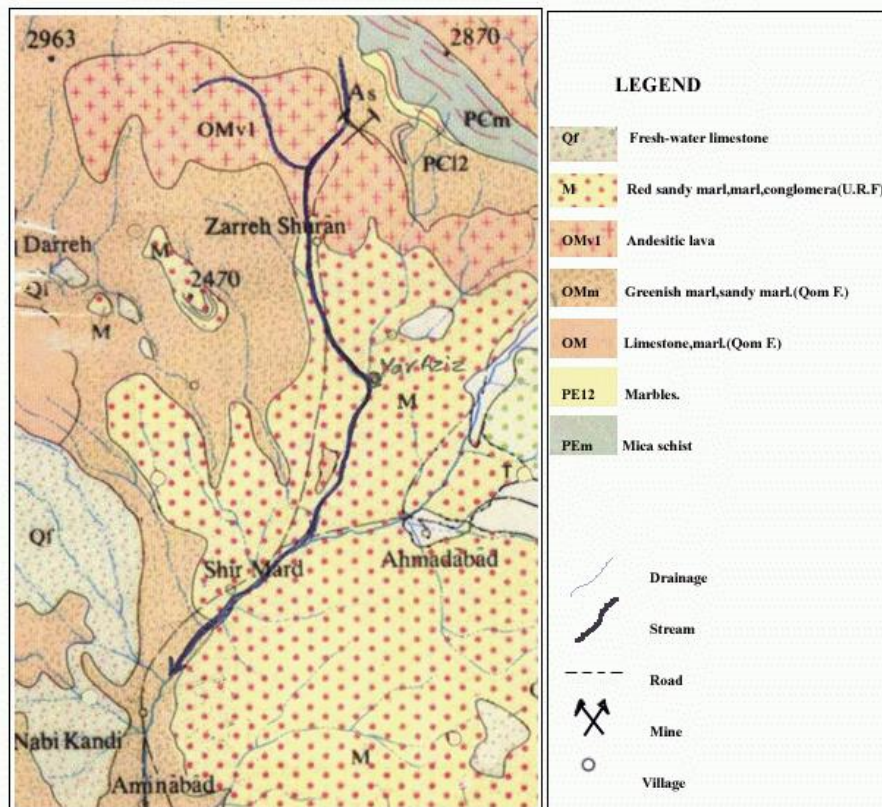


Fig ۲: geological map of this area.

In this area, clastic and carbonate sedimentary rocks are widespread. The lithology of the adjacent riverbed are; Precambrian Micaschiste and marbles, sandstone and green sandy marl of the Qom formation (Oligo-Miocene), conglomerate and sandstone of Upper Red formation, porphyritic

Andesite and dolerites of Oligo-Miocene, travertine and recent alluvium (Mehrabi, ۱۹۹۹) (figure. ۲). The presence of thermal spring, sedimentation of travertine and formation of oxides such as Pyrolosite, limonite and goethite, are evidences for presence of an active geothermal system (۲). The major sulfide ores in this mine are Realgar (As₂S₃) and Orpiment (As₂S₅).

Pyrite (FeS₂), the major mineral in the formation of acidic mine waters, is also present in this area. Gold storage extension in Zarshuran mine is nearly similar to arsenic. The Arsenic ore reserve of the Zarshuran deposit (۱ gr/ton) is assessed about ۲۳ M.T. (Mehrabi, ۱۹۹۹). The study of topographic map in the area shows that the ground water level in the mine area is higher than the river surface water. As a result, ground water recharges surface water and gradually dilutes it.

The mechanical transportation of elements also occurs as coarse grains. While in lowland areas due to low and smooth topography, ground water in downstream area is polluted by rivers causing a spread of pollutant elements in the subsurface areas. The transportation of elements is also occurs as chemical dissolution, which has dissolves materials in the water.

Methodology:

To accomplish this study, at the outset the topographic and geological maps of area were used as base maps and, then, the river tributary was traveled. Sampling from water and river sediments was done in ۱۰ sampling stations at the same time. In the locations that river bedrocks have well outcrops ۱۲ soil and water samples were taken. Soil (۶ samples) and grassy plants samples (۱۲ samples) in ۱-۲ km around the mining site were taken according to a systematical sampling procedure (Hasanipak, ۱۹۹۴).

After granulometry (sieving and Hydrometry) and calcimetry laboratory experiments, the soil and river sediment samples were analyzed by atomic absorption spectrophotometer (A.A.S.). The soils were digested by using HNO₃-H₂O₂. The plants and hairs were digested by using HNO₃-HClO₄ (Liao et al, ۲۰۰۴). Also, Arsenic and heavy metal levels were determined using Inductively Couple Plasma (ICP) emission spectroscopy following EPA procedures (Rios-Arana et al, ۲۰۰۳; Naicker et al., ۲۰۰۳; Reimann and de Caritat, ۲۰۰۵).

Rock samples were studied using a polarizing microscope and prepared thin sections. Also, after samples preparation, all elements were detected by A.A.S. The soil and river sediment samples were also analyzed by X.R.D. and their constitution minerals were recognized.

Table ۱ and figures. ۳ show the concentration of elements for water samples, river sediments, soils and rock samples. In compared to World Health Organization (WHO) standards concentration of Arsenic in water should be ۰.۰۵ mg/l. The normal range for As in soils of various countries was ۰.۱ to ۴۰ mg/kg (mean of ۶ mg/kg) (Chatterjee et al., ۱۹۹۳).

The concentration of this element (As) in the Zarshuran River water is estimated between ۱۰-۲۶۰۰ ppm and in the soil samples between ۹۲۰-۱۰۰۰۰ ppm. Therefore serious pollution appears can be observed in this area. The pH of water samples in the stations no. ۱-۴ was measured. The pH ranges (۴.۵-۶.۲) showed acidic and slightly acidic water and in other stations were in neutral range (۷.۰).

The study of heavy minerals in the river and tributaries in ۱۹۸۹ (for geochemical explorations) and recent studies show the presence of the heavy minerals in the local alluviums. The minerals included such as: realgar (As₂S₃), orpiment (As₂S₅), stibnite (Sb₂S₃), pyrite (FeS₂), cinnabar (HgS), marcasite (FeS₂), fluorite (CaF₂), barite (BaSO₄), sphalerite (ZnS), galena (PbS), and cerostie (PbCO₃).

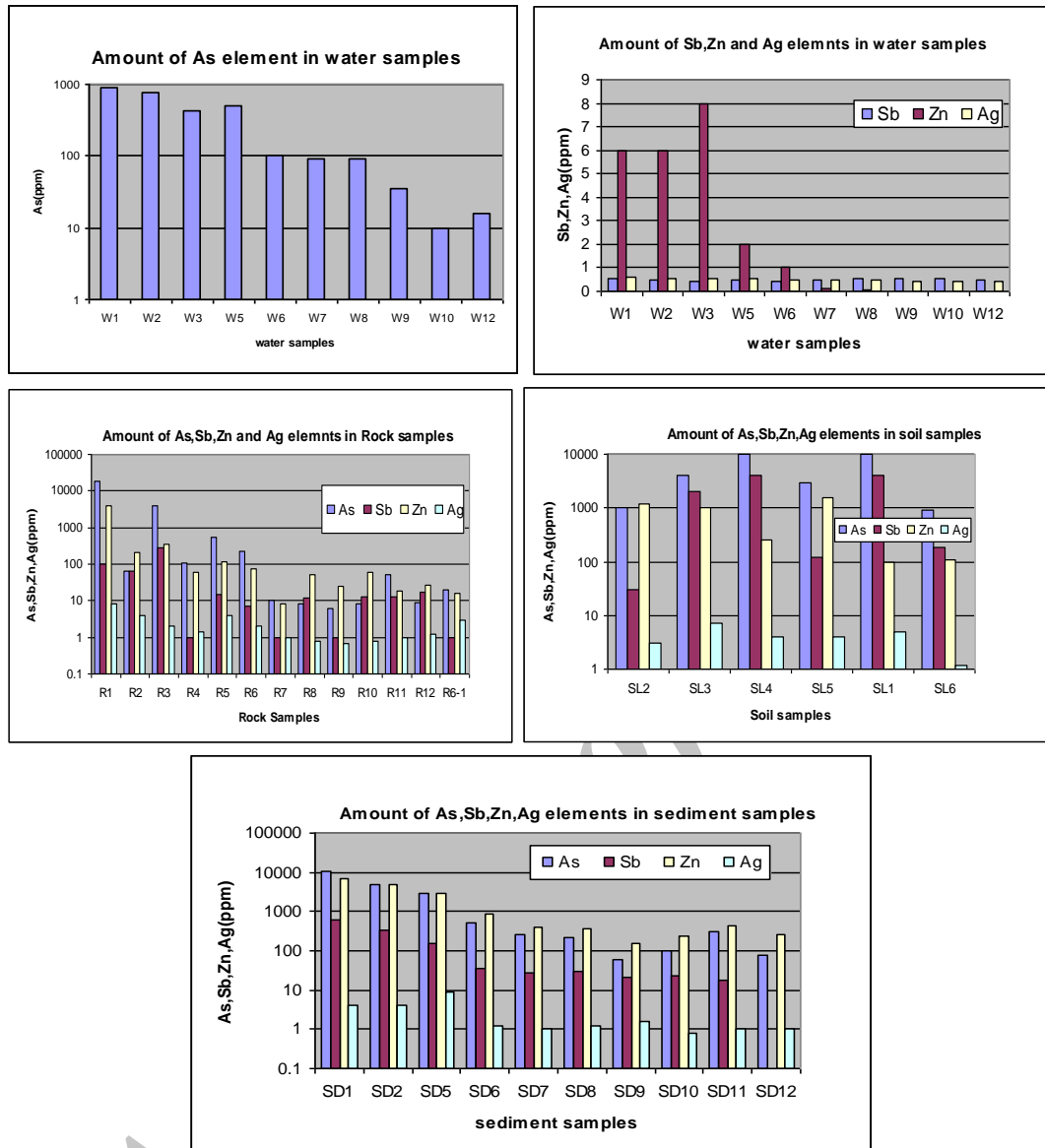
Figure ۴ also indicate the concentration of elements in wastes that have been dumped around the Mine (Mehrabi, ۱۹۹۹). The enrichment factor (EF) of a vegetable was calculated using the following equation: $EF = a/b$ that $a=As$ concentration in edible part grown on contaminated soil and $b=As$ concentration in edible part grown on control soil (Liao et al, ۲۰۰۵). X.R.D. results of river sediments and soil samples are shown these minerals: Quartz, Nordstrandite, Calcite, Clay minerals, Dolomite, Goethite, Feldspar, Hematite, Amphibole. All these concentrations indicate the increment and enrichment of toxic elements in the selected samples, which obviously show high concentrations, compared to the standards and acceptable limits.

Table ۱: The concentration of the measured elements in water, River sediments, soil and rocks samples (ppm).

N.S	As	Sb	Zn	Ag	N.S.	As	Sb	Zn	Ag
W۱	۹۲۰	۰,۵۲	۶	۰,۵۹	R۱	۱۸۰۰۰	۱۰۳	۴۰۰۰	۸
W۲	۷۶۰	۰,۵۱	۶	۰,۵۷	R۲	۶۶	۶۵	۲۱۵	۴
W۳	۴۲۰	۰,۴۵	۸	۰,۵۵	R۳	۳۹۰۰	۲۷۰	۳۶۱	۲
W۵	۵۰۰	۰,۴۶	۲	۰,۵۳	R۴	۱۰۴	۱	۶۰	۱,۴
W۶	۱۰۰	۰,۴۵	۱	۰,۵۱	R۵	۵۳۶	۱۵	۱۱۹	۴
W۷	۹۲	۰,۵۱	۰,۱	۰,۴۹	R۶	۲۲۴	۷	۷۴	۲
W۸	۹۲	۰,۵۲	۰,۰۵	۰,۴۷	R۷	۱۰	۱	۸	۱
W۹	۳۶	۰,۵۳	۰,۰۲	۰,۴۵	R۸	۸	۱۲	۵۱	۰,۸
W۱۰	۱۰	۰,۵۷	۰,۰۲	۰,۴۲	R۹	۶	۱	۲۵	۰,۷
W۱۲	۱۶	۰,۵۱	۰,۰۲	۰,۴۱	R۱۰	۸	۱۳	۶۰	۰,۸
SD۱	۱۱۰۰۰	۶۳۳	۷۰۰۰	۴	R۱۱	۵۰	۱۳	۱۸	۱
SD۲	۵۰۰۰	۳۳۰	۵۰۰۰	۴	R۱۲	۹	۱۷	۲۶	۱,۲
SD۵	۳۰۰۰	۱۵۵	۳۰۰۰	۹	R۱۰-۱	۲۰	۱	۱۶	۳
SD۶	۵۰۰	۳۴	۸۴۵	۱,۲	SL۲	۱۰۰۰	۳۰	۱۱۸۰	۳
SD۷	۲۶۰	۲۷	۴۱۵	۱	SL۳	۴۰۰۰	۲۰۰۰	۹۹۰	۷
SD۸	۲۲۰	۲۹	۳۶۵	۱,۲	SL۴	۱۰۰۰۰	۴۰۰۰	۲۶۰	۴
SD۹	۶۰	۲۲	۱۶۰	۱,۶	SL۵	۳۰۰۰	۱۲۰	۱۵۲۰	۴
SD۱۰	۱۰۰	۲۳	۲۴۰	۰,۸	SL۱	۱۰۰۰۰	۴۰۰۰	۱۰۰	۵
SD۱۱	۳۲۰	۱۸	۴۳۰	۱	SL۶	۹۲۰	۱۸۳	۱۱۰	۱,۲
SD۱۲	۸۰		۲۶۰	۱					
W=water	R=Rock	N.S= Sample Number			SD=Sediment			SL=Soil	

Table ۲: Name of plant and concentration of Sb and As measured In the plant samples around Zarshuran mining site

No.	Name	Weight(g)	Sb(ppm)	As(ppm)
۱	Fabeaceae onions Spinoza	۵,۰۶	۹,۹	۱۷
۲	Apicea umbrelliferae Erynium	۱۰,۰۵	۱۰	۲,۲
۳	Fabeaceae Astragalus L.	۷,۱۳	۱۱	۲,۵
۴	Rosaceae Rosa L.	۱۰,۵۴	۱۲,۳	۱۵,۵
۵	Fabeaceae lotus L.	۳,۰۳	۹,۶	۱۳
۶	Scrophulariaceae scrophularial L.	۱۹,۵۲	۱۱,۳	۱۵,۶
۷	Labiatae lamaceae menthe	۳,۸۷	۱۳,۵	۴,۱
۸	Lamiaceae menthe logifoin	۲,۸۷	۱۴,۴	۱۸,۶
۹	Composita picris strigosa M.B.	۹,۵۹	۱۴,۷	۹,۶
۱۰	Composita Inula Helenium L.	۶,۵۱	۱۰,۶	۰
۱۱	Labiatae	۱,۲۲	۵,۷	۲,۱
۱۲	Composita Inula Acuheriana	۱,۰۴	۱۳,۸	۰



Figures. 7: The concentration of the measured elements in the water, River sediments, soil and rocks samples (ppm).

Discussion:

In terms of geochemistry, river environment belongs to secondary or supergene environments. The formation and accumulation of sedimentary materials are accomplished at low pressure and temperature. Therefore, the role of thermodynamic parameters is nearly insignificant. The extremity fate of elements depends on affinity of them in the water. The mobility of elements relate to their solubility. In addition to the nature of elements and chemical properties, pH and Eh conditions, the presence of ions such as Fe and Mn, electro negativity, ionic potential, colloidal surface absorption and other geochemical barriers also affect the mobility of elements. The major element in this ore deposit is arsenic; Chalcophile element and marker soluble complex anions. This element can deposit as hydration or co-precipitation with other elements. In high pH and low Eh conditions, the relative mobility of elements decrease and in low pH and high Eh condition, mobility of elements increases intensely (Bando et al, ۲۰۰۰). In term of oxidation of sulfide minerals, it results in acidic waters, which in turn cause on increase in the dissolution and transportation of elements (Leblanc, ۱۹۹۶; Kendeler et al., ۲۰۱۲).

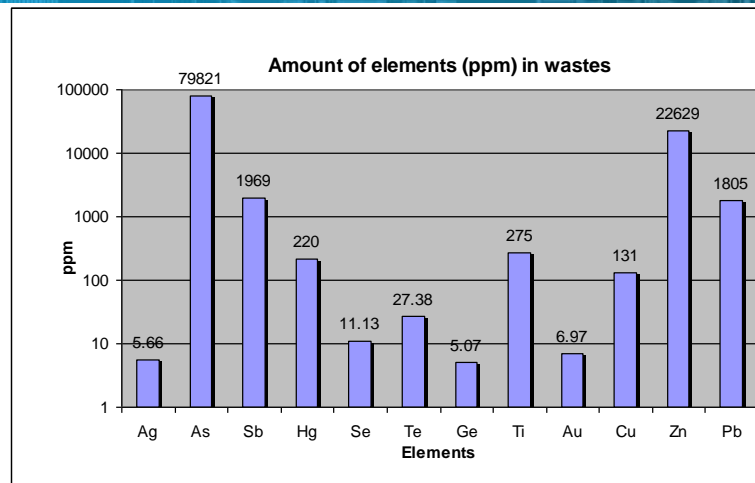


Figure 4: The concentration of elements in wastes that have been dumped around the Mine.

In Zarshuran mine, rainfall penetrates into the mining tunnels easily and it is in direct contact with wastes (figures 4 and 5). The rainfall season in this area is long and the rate of precipitation is also high. Before the mining, the oxidation of sulfide minerals and formation of sulfuric acid, during the weathering processes were accomplished slowly. Their natural entry into the water system was very insignificant. The mining activities increase the rate of some chemical reactions. In these reactions, at beginnings, from combination of sulfide minerals with oxygen and water, produces the sulfates and (H^+) ions and then in the transportation channel, produces sulfuric acid (Cravotta et al, ۱۹۹۰).

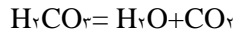
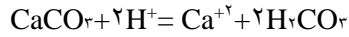
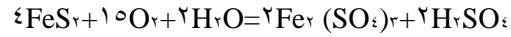


Figures 4 and 5: In Zarshuran mine, rainfall penetrates into the mining tunnels easily and it is in direct contact with wastes

The decrease of the water pH also causes the release of toxic cations. In flood time and seasonal inundations, the increasing of pH causes the possibility of production of acid and metal rich fluids. The catalysis actions of autotrophic bacteria such as Thiobacillus Ferroxidans increase the amount of oxidation and production of acidic waters strongly. This bacterium is microaerophile and needs little oxygen for life. The poisons are used to take away microbes in these processes more especially; anionic surfactants cause to decrease the production of acidic waters (Sasowsky et al, ۱۹۹۶; Bhattacharya et al., ۲۰۰۶; Jaya and Narayanan, ۲۰۱۳).

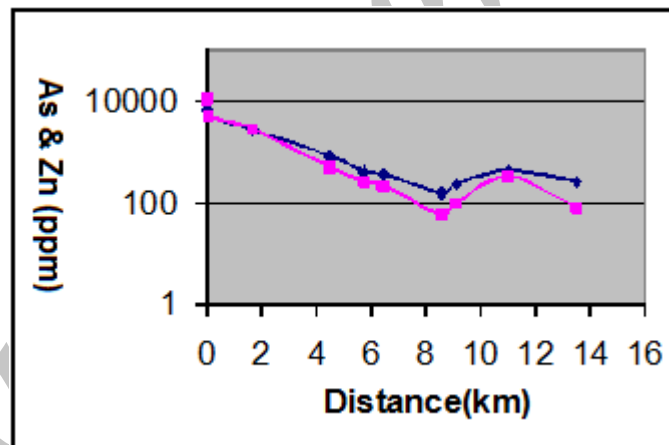
Residual sulphide minerals, especially pyrite, in the tailings dumps are unstable when exposed to atmospheric oxygen and undergo oxidation resulting in the generation of acid mine drainage (AMD) and the subsequent release of heavy metals and metalloids (Tutu et al., ۲۰۰۸). The problem of sulphide oxidation and the associated AMD, or more generally acid rock drainage (ARD), as well as the solution and precipitation processes of metals and minerals, has been a major focus of

investigation for over half a century. In Zarshuran, the limestone rocks in vicinity of mine decreases the capacity of elemental transportation of acidic waters because of neutralizing properties and increases the pH of water excessively (Tutu et al., ۲۰۰۸; Cravotta et al, ۱۹۹۰; Stumm & Morgan, ۱۹۸۱).



The composition of waters and sediment resources of Zarshuran River show the distribution of pollution and environmental destructive elements that are spread to ۱۲ km far from the mine (Shirmard village). Also, these elements seep into the ground waters resources. It has been detected that, where the minor branches of river connect to the main river, the water dilutes and decreases the concentration of toxic elements. The diagram of concentration of elements in water and sediments versus the distance from the source of pollution (mine) shows an exponential decreasing manner (see figure. ۶).

During sampling, a pH of γ , λ was recorded for the tailings. This slightly elevated pH in the Slimes is attributed to the addition of lime during gold extraction. This is done to keep the cyanide in solution as it escapes at low pH posing a toxicity problem. The results for elemental concentrations in tailings following total digestion in the microwave point to tailings as potential sources of leachable metals to the environment. The results for leaching at pH γ showed an initial increase in pH to about λ (roughly the pH of the tailings) and then a drop. This could be attributed to the effects of buffering by the tailings material. This effect was also apparent when pH γ was used for leaching. The leachates gave an initial pH above ε , \circ which dropped gradually to γ as leaching progressed up to two pore volumes. This trend of decreasing pH is likely to occur beyond two pore volumes.



Fig^۶: The diagram of concentration of elements in water and sediments versus the distance from the source of pollution (mine) shows an exponential decreasing manner.

The biological treatment of groundwater is used primarily to remove electron donors from water sources, providing (biologically) stable drinking water, which preclude bacterial re-growth during subsequent water distribution. To the electron donors belong also the dissolved metal cations of ferrous iron and manganese, which are common contaminants found in most (anaerobic) ground waters. The removal of iron and manganese is usually accomplished by the application of chemical oxidation and filtration (Zoubouli & Katsoyiannis, ۲۰۰۵; Bálintová et al., ۲۰۱۲; Friedlová, ۲۰۱۰).

The oxides and hydroxides sediments exist between the sampling stations of study area which (figure ۷) shows the sudden change of concentrations of soluble pollutant elements (λ , γ), in a short distance. So, it is interesting that the concentration of elements (As, Sb, Ag and Zn) decrease excessively in short distance as results of a natural geochemical barrier. Arsenic exists mainly in three valences states (i.e., -۳ , $+۳$, $+۵$). The trivalent arsenic ($\text{As}^{۳+}$) and the pentavalent arsenic ($\text{As}^{۵+}$) are widely present in

natural waters and are soluble over a wide range of pH and Eh conditions (Bell, ۱۹۹۸, Duker et al, ۲۰۰۵; Oelofse et al., ۲۰۰۷; McCarthy, ۲۰۰۸).



FigV: The oxides and hydroxides sediments exist between the sampling stations of study area show the sudden change of concentrations of soluble pollutant elements in a short distance.

The surface absorption of trace element ions can be used to remove the pollution and file away the poisoning elements. The soluble hydroxyl sulfates, which from in the alluvium are a temporary storage source for metal pollutants and in the course of intense rainfall and floods, dissolve and enter the aquatic system (Bowell, ۱۹۹۴, Salminen et al, ۱۹۹۶).

The high amount of fine sedimentary grains (clay size fraction) increases the amount of absorption of pollutant elements from the water resources (Thorton, ۱۹۹۶). Other geochemical barriers (intensive changes in physiochemical conditions in the migration of elements which cause to settling them from solution) that are present in this area can indicate the presence of sulfates and carbonate, evaporate rocks and mechanical barriers (changes in velocity and other hydrological factors of water and deposition of heavy minerals) (Evagelu, ۱۹۹۸, Thorton, ۱۹۹۶, Sasowsky et al, ۱۹۹۶).

The fine and soft sediments and, naturally pollutant elements, often deposit in tails of sedimentary bars (current cells are close to returned movement) and towards them (Ackman&Klienmann, ۱۹۹۸). The presence of calcite in sediments causes the decrease of mobility of metals and increases their buffer capacity.

The statistical studies on soil and sediment samples of the area (cluster analysis), indicates the presence of pollutant elements, while this correlation is very low for sandy grains and the coarser ones. Also, from all of studied elements, the high correlation appears between As, Sb and Zn and these three elements nearly show a concordant behavior while silver (Ag) has a negative correlation and unique behavior. Dolomite ($\text{CaMg}(\text{CO}_3)_2$) also shows a good correlation with As, Sb and Zn. The water sample N.۷ used for irrigation of farms and agriculture. The water sample N.۱۰ is a Thermal spring in vicinity of Yar Aziz village that consummated by inhabitants and the amounts of elements in its sediments is higher than normal.

The river water and also grassy plant in vicinity of river is available to the retinues and animals of area easily. The presence of arsenic in drinking water causes the skin diseases, cancers, and disorder in blood circulation system, bloody flux (dysentery), respiratory inflammation and black foot disease. Systemic and chronic exposure to arsenic (As) is known to lead to serious disorders, such as vascular diseases (Blackfoot disease (BFD) and hypertension) and irritations of the skin and mucous membranes as well as dermatitis, keratosis, and melanosis (Chou et al, ۲۰۰۶).

Conclusion:

In comparisons to world health organization (WHO) standards, the pollution is heavily observed in this area. The pH of water samples were acidic and slightly acidic [۴,۵-۶,۵] and in some stations were neutral [۷]. All resources (Water, Sediments, Soil, and Plants) indicate enrichment of toxic

elements in the tested samples, compared to standards and acceptable limits. In this mine, in addition to conditions and factors, which are necessary for production of acidic, mine waters, the prevention factors and neutralizer of them also are present.

In this mine, rainfall penetrates to tunnel of mine easily and it is in direct contact with wastes. The formation of the low pH (acidic) water also causes the release of toxic metals cations. In this area, limestone exists in vicinity of ore deposit, which cause neutralization and increases the pH of acidic waters excessively and decreases the capacity of their elemental transportation.

The study of water and sediments of this river demonstrates that the distribution of pollution and environmental destructive elements is spread to Shirmard village (۱۶km far from the mine) and the penetration of these elements occur in to ground waters of area. The oxides and hydroxides sediments cause the absorption of pollutant elements as a natural geochemical barrier

In the river branches, everywhere that the amount of fine sedimentary grains (clay fragments) is high, the amount of absorption of pollutant elements from the water also increases.

The statistic studies on soil and sediment samples of the area (in cluster analysis), indicated the presence of pollutant elements, while this correlation is very low for sandy grains and the coarser ones. Among studied elements, the high correlation appears between As, Sb and Zn.

Based on our investigation, we proposed that, with construction some sediment-trap barriers at the upstream of Zarshuran River and in vicinity of the mining area and using from depositional materials (see geochemical barriers) for pollutant elements will prevent the distribution of pollutants. Also it is necessary that diseases, which are related to these elements, be studied in area carefully and informing the risk factor to inhabitants, in order to decrease the destructive environmental impacts of the mining activities in Zarshuran area.

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