



Trace Metal Contamination in Soils and Waters around the Abandoned Colliery Site of Bagworth Heath, English Midlands

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Abstract

Abandoned mines and mining activities are considered one of the most significant sources of trace metal contamination worldwide. Those activities resulted in environmental contamination of particular surrounding ecosystems of abandoned ecosystems. The main aim of this study is to evaluate the trace metal contamination in the vicinity of abandoned mine located in Leicestershire English Midland. Twelve soil samples with two water samples were collected from the abandoned mine site. Test results showed a wide range of soil pH was observed from extremely acid (2.5) to slightly alkaline (7.4) as well as LOI from 8% organic matter content to 40% was found. Moreover, results demonstrated that most elements were below Soil Guideline Values (SGV) including As, Cd, pb and Ni, while the high total concentration was found for the case of (Al and Fe). In addition, a high concentration of Al and As were found in water samples in both pipe and pond water while other investigated metals were found in slightly higher than permissible limits of (10, 3, 10, 20) respectively. According to this study Al was the dominant heavy metals in the area followed by the Fe>Ti>Ba>Mn>V>Rb>Sr>Zn>Cr>Cu>Ni>As>Pb>Co>Cs>Mo>Se>U>Cd.

Keywords: soil contamination, abandoned mine, and traces metals.

تلوث المعادن في التربة والمياه حول موقع المنجم المهجور في باغورث هيث ، ميدلاندرز الإنجليزية

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الخلاصة

تعتبر أنشطة المناجم و المناجم المهجورة من أهم المصادر الملوثة للبيئة بالمعادن النزرة حول العالم. هذه الأنشطة تدخل عدد لا يحصى من الملوثات الى البيئة، و خاصة المناطق المحيطة بالمناجم المهجورة. ان الهدف من هذه الدراسة هو لتقييم الاثر الملوث للمعادن النزرة في منطقة يسترشير في ميدلاندرز الإنجليزية. تم أخذ اثني عشرة عينة من التراب و عينتان من المياه من موقع منجم مهجور. النتائج أظهرت مدى واسع لقياس الاس الهائيدروجيني (مدى الحموضة) من الحامض للغاية 2.5 الى قليل القلوية 7.4. كما و أن مستوى التكوين العضوي للمواد بعد الاشتعال ال (LOI) كان من 8% الى 40%. أظهرت النتائج ان مستوى غالبية المعادن يقع تحت مستوى قيم دلالة التربة ال (SGV)، بما فيها As, Cd, pb, Zn, Cu, Ni, Co في حين ان أعلى تركيز بين المعادن كان لنسبة الالمنيوم والحديد. اضافة الى ذلك، وجدت تركيز عالي من الالمنيوم و الارسينيك في عينات اخذت من ماء الانوب و برك المياه في حين كان مستوى المعادن الاخرى التي تمت معاينتها فوق المسموح بها بشكل طفيف. بحسب نتائج هذه الدراسة فأن الالمنيوم كان المعدن الثقيل المسيطر بين المعادن الثقيلة يتبعه الاخرين كما الاتي:

Fe>Ti>Ba>Mn>V>Rb>Sr>Zn>Cr>Cu>Ni>As>Pb>Co>Cs>Mo>Se>U>Cd.

1. Introduction

Significant amounts of chemical materials including organic and inorganic compounds are discharged into the environment each year at levels that become an environmental concern. The considerable amount of these chemicals is trace metals, which occur naturally in rocks and soils, but progressively higher quantities of metals are being released into the environment by human activities.

One of the most intractable problems exposing the world today is contamination of the environment with heavy metals which become a global phenomenon. Pekey [1] reported that the major reason for this, is that heavy metals cannot be biologically decomposed therefore, they have the ability to accumulate in living tissues, food chains and ecosystems, when they reach to the standard dosage in the living organisms, they might be directly pernicious, or can lead to serious hygienic problem. Although natural processes is one of the most significant sources of heavy metal contamination such as, erosion and weathering, volcanic eruptions, geothermal activities, natural forest fire, wind-blown dust, and sea salt spray, large quantities of heavy metals are released into the environment via anthropogenic sources, which includes many activities are created by people such as industrial activities, mining, agriculture applications, generation of energy using coal or peat, wood preservation industries, and wastes as a result of burning of preserved log. [2, 3].

Some trace metals are essential for living organisms in a small amount, but others become a major problem. The most serious damage to human health has demonstrated in Bangladesh and West Bengal, India. In the 1970s and 1980s, long term exposure or overdoses of heavy metals can cause acute toxic effects including gastrointestinal symptoms such as, loss of appetite, vomiting and diarrhea and dysfunction of cardiovascular and nervous systems, for instance, muscle cramps, heart complains finally death [4]. Worldwide, a massive amount of people are exposed to heavy metals at concentrations greater than permissible limits. Recent studies indicate that some inorganic heavy metals including arsenic, cadmium and chromium are carcinogenic, mutagenic, and teratogenic if used in high dosage [5].

The soil is a key component of natural and agricultural environments, in both ecosystems, it is essential for flourishing, decomposition and recycling of all biological communities. It has contained a myriad of organic and inorganic materials in different physical phases, solid, aqueous and gaseous [6]. Trace metals are considered as the significant potential sources of soil and water contamination. They can be passed to, dispersed to and accumulated in the biological organism and then may be transported through the food web to the human population as an end user [7]. Heavy metals accumulate in soils and may contaminate them through emissions from unexpected development of industrialization and urbanization, mine tailings, agricultural application particularly, fertilizer and pesticide, wastewater irrigation, coal, and fossil fuel combustion, dispose of high level contained waste and precipitation from the atmosphere [8]. Although the total concentration of metals in soils remain for a long time after their release, the change of their chemical forms might be possible through the process of speciation and bioavailability [9].

Heavy metal contamination is considered one of the most serious problems in the surrounding of abandoned mine sites (AMS). These heavy metals have the potential to contaminate environmental compartments including soil and water. They can be dispersed and accumulated in the food chain, and taken in by human beings as a final consumer. Trace metals can be distributed to the soil in the surrounding of an (AMSs) by many ways after their disposals such as winds in the form of dust particles and water through surface run off, seasonal ground water discharges and surface water overspill. All these pathways are affected by hydrogeological conditions and climatic situation of the area [10]. Coal mine spoils represent one kind of high metals industrial solid waste, via coal mine application a copious amount of this kind of wastes has been excavated from the underground to the surface of the earth. Natural erosion and weathering conditions may lead to these exposed coal mine spoils to break into fine clay size particles. Through these processes, huge quantities of tiny particles can be discharged to the environmental compartments within a particular time [11].

The common parts of all mine coals are metal sulfides particularly, pyrites, they can be easily oxidized during the natural actions such as erosion and weathering of coal mine spoils. Wiggering [12] reported that these processes are partly controlled by microorganisms, especially *Bacillus ferroxidans*. It occurs when the pH value drops to below 4, in this case, the hydrogen ions can accelerate the weathering of coal mine spoils and expedite the release of toxic trace metals. The main aim of this study is to evaluate the trace metal contamination in the abandoned coal mine area.

2. Materials and method

2.1 Description of sampling site

Soils and water samples were collected from the reclamation coal mine site at Bagworth Heath, about 10 miles west of Leicester, East Midlands, England. Bagworth Heath is located less than a mile south of Bagworth village towards Merry Lees and Desford. All sampling points were geo referenced by GPS. Figure-1 shows the sampling points within the site. Coal mining took place in this area from 1825 to February 1994, it had been in the peak of success and produced more coal per man-shift than any other pit in Europe. Bagworth colliery is entered in the Guinness Book of Records for its exceptional production per man-shift. Bagworth finally ceased in February 1991 after 166 years of production. Upon closing, a massive quantity of mine solid wastes was left behind without proper protection and monitoring.

Bagworth heath contains the greatest variety of plant species, including trees, grasses and herbaceous vegetation, as well as there are many bare land spots sparsely covered with grass and herbaceous vegetation or even completely excavated because of exposed solid waste of coal mines. Bagworth Heath is moderate elevation lying between 121-134 m above sea level the local geology includes mostly Mudstones.

The samples were collected separately, from the base of the heap to the top and grassland Around the heap (Table-1) and two samples of surface water were taken from the piped and pond water in the vicinity of the spoil heap.

Table 1-Description of sampling sites at Bagworth Heath Colliery

Sample no	depth (cm)	GPS	Position and description
1	0-20	52°39'27.43"N 1°19'07.94" W. elev 124m	Base of spoil heap, non-vegetated area (drainage ditch)
2	0-20	52°39'27.38"N 1°19'07.04" W. elev 125m	Above drainage ditch, non-vegetated (bare area)
3	0-20	52°39'27.39"N 1°19'06.98" W. elev 125m	Mid slope on spoil heap (bare area)
4	0-20	52°39'27.84"N 1°18'59.07" W. elev 132m	Top of the spoil heap, bare area
5	0-20	52°30'32.43"N 1°19'03.96" W. elev 132m	Top spoil heal, soil cover above spoil heap (vegetated area)
6a	0-20	52°39'32.83"N 1°19'09.07" W. elev 129m	Bottom of slope, fenced area (vegetated and wooded)
6b	20-40	52°39'32.83"N 1°19'09.07" W. elev 129m	- - -
6c	40-60	52°39'32.83"N 1°19'09.07" W. elev 129m	- - -
7a	0-20	52°39'33.50"N 1°19'10.62" W. elev 126m	Grass slope
7b	20-35	52°39'33.50"N 1°19'10.62" W. elev 126m	-
7c	40-60	52°39'33.50"N 1°19'21.43" W. elev 124m	-
8	0-20	52°39'31.57"N 1°19'07.94" W. elev 124m	Soil cover near the path
PipeW		52°39'30.05"N 1°19'18.44" W. elev 121m	Bottom of the spoil heap
PondW		52°39'28.23"N 1°19'26.52" W. elev 119m	Main fishing lake

2.2 Sample preparation and analysis

Soil samples were collected using a clean stainless steel trowel or auger and sealed in plastic bags for transport. Soils were air dried in aluminum trays, gently disaggregated using a pestle and sieved to

obtain a <2 mm fraction. A portion of each sample was finely ground using an agate ball mill (Retsch, Model PM400), and then stored in polyethylene bags in preparation for elemental analysis. Water samples were immediately filtered through 0.45 μm cellulose acetate syringe filters into acidified polyethylene bottles and stored at ambient temperature until laboratory analysis.



Figure 1-satellite image of the study area showing sampling sites

2.2.1 Soil properties

Soil properties, such as pH, organic matter (OM%, measured by loss on ignition), were determined based on the sample fraction <2mm to obtain basic information about the soil conditions. To determine pH, 5 g of soil (< 2mm) was suspended in 12.5 mL of MilliQ water (18.3 M Ω) and shaken on a reciprocal shaker for 30 minutes. The pH of the soil suspension was measured using a Hanna pH-209 pH meter and a combined glass electrode (Ag/AgCl; PHE 1004), calibrated with pH 7 and pH 4.01 buffers, allowing 5 minutes for the reading to stabilize. Water pH was determined in the field using a portable pH meter. The standard method for loss of ignition was used to measure the percentage of organic carbon in the samples. A known weight of <2mm dried soils in a weighted silica crucible was placed in muffle furnace for overnight at 550 $^{\circ}$ C, to ignite organic matter. The weight loss is expressed as a percentage of soil organic matter.

2.2.2 Total elemental concentrations

Approximately 200 mg of the finely ground soil was digested using 70% hydrofluoric acid, nitric acid and perchloric acid (Trace Element Grade (TEG); Fisher Scientific, UK) in a Teflon-coated graphite Block Digestor (Analysco, UK) containing places for 48 PFA digestion vessels. The digested samples were diluted to 50 mL using Milli-Q water (18.3 M Ω cm) and stored un-refrigerated in universal sample bottles (5% HNO $_3$) pending elemental analysis. All digests were diluted to 1 in 10 with Milli-Q water using a compudil – D auto diluter (Hook and Tucker Instruments) immediately prior to analysis.

Multi-element analysis was undertaken by ICP-MS (Model X-Series I, Thermo-Fisher Scientific, Bremen, Germany) in ‘collision cell mode’ (7% hydrogen in helium) to reduce polyatomic interferences. Samples were introduced from an autosampler (Cetac ASX-520 with 4 x 60-place sample racks) through a concentric glass venturi nebulizer (Thermo-Fisher Scientific; 1 mL min $^{-1}$). Internal standards were introduced to the sample stream via a T-piece and included Sc (100 ng mL $^{-1}$), Rh (20 ng mL $^{-1}$) and Ir (10 ng mL $^{-1}$) in 2% TEG HNO $_3$. External multi-element calibration standards (Claritas-PPT grade CLMS-2, Certiprep/Fisher) included Al, As, Ba, Bi, Ca, Cd, Co, Cr, Cs, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, Pb, Rb, Se, Sr, U, V, and Zn, all in the preferred range of 0-100 $\mu\text{g L}^{-1}$.

Sample processing was undertaken using Plasma lab software (version 2.5.4; Thermo-Fisher Scientific) set to employ separate calibration blocks and internal cross-calibration where required.

3. Results and discussion

3.1 Soil properties

Values for pH and loss on ignition (LOI) values are given in Table-2. In general most of the samples on or around the spoil heap have an acidic pH ranged from 2.5 to 7.4 with a mean value of 3.82. The lowest values might be due to the weathering of sulfides minerals (pyrite (FeS₂) from the mine dump. Sometimes acidic pH may be related to oxidation of metal sulfides and iron sulfide such as pyrite because metal sulfides, particularly pyrite can easily be oxidized under runoff situation. and the highest values of the pH refers to presence considerable quantity of carbonate which neutralizes the acid water generated in the coal mines [13]. LOI is an estimation of the percentage of organic carbon in soil. The organic carbon of the soil samples ranges from 8% to 40% with a mean of 21%. The presence of vegetation in most parts of the mining area could explain the high values of organic matter in some samples of [14]. Catherine [15] reported that both properties are a significant variable in modeling the behavior of the biological availability of metals and solubility in soil.

3.2 Total metal concentrations in soil samples

When comparing total concentrations, Al was the highest Potentially Toxic Element (PTE) the order observed for other studied metals are Fe>Ti>Ba>Mn>V>Rb>Sr>Zn>Cr>Cu> Ni> As>Pb> Co>Cs> Mo>Se>U>Cd (Table-2). Although remediated mines considered one of the most important sources of studied PTEs, the total content of metals were below the Soil Guideline value SGV except Al and Fe content were high if compare to other investigated PTEs. The distribution of metals in sampling sites is given in Figure-2. It can be seen that the most abundant PTEs[16] in all sampling sites was Al followed by Fe. Interestingly, As and Cu concentrations were significantly higher in the spoil (S1, S2, S3 and S4) then the soil. In contrast to Cd, Ni, Co and Zn, which are higher in soil (S5, S6a, S7a and S8) compared to colliery waste

Table 2-Soil properties and Concentration of some potentially toxic elements in Bagworth Heath samples (mg/kg) with mean, SD and SGV

Samples	pH	LOI%	As	Cd	Cr	Co	Ni	Cu	Zn	Mn	Fe	Al	Ti	V	Ba	U	Se	Cs	Mo	Rb	Sr	Pb
1	2.83	40	15.68	0.071	26.6	3.518	11.69	24.67	15.9	42.38	13875	27240	3137	48.01	287	1.527	3.321	2.099	29.49	55.84	2.832	
2	2.61	26	24.27	0.032	28.58	2.866	8.805	16.88	15.25	29.28	13080	32261	3604	57.1	311.8	1.452	4.639	2.343	38.87	50.2	48.06	
3	2.5	22	13	0.042	28.84	2.695	12.29	17.67	21.31	34.42	13896	28016	3806	53.5	293.1	1.449	4.15	1.567	38.3	46.19	3.214	
4	2.78	30	26.69	0.036	25.85	2.372	7.904	17.95	16.34	26.11	13082	25827	3275	45.67	282.1	1.231	4.594	2.519	35.56	50.22	27.5	
5	7.4	24	8.111	0.109	22.35	5.726	16.1	13.88	41.82	177.9	11352	21426	2597	37	190.8	1.161	2.847	1.13	33.54	37.07	6.402	

	-1.42	3.679	5.67	0.892	-1.16	-1.41	-5.30	7.412	15.21	450
	17.2	47.68	50.28	19.17	18.33	33.23	17.72	36.92	15.15	
	37.45	36.22	34.95	38.49	39.54	36.97	46.43	37.15	4.022	
	0.349	1.557	2.573	0.413	0.408	1.175	0.274	1.367	0.879	
	2.173	4.053	4.099	2.271	2.271	3.526	2.986	3.41	0.904	
	0.716	1.445	1.837	0.682	0.772	1.133	0.702	1.172	0.404	350
	0.730	1.520	1.736	0.710	0.708	1.209	0.794	1.185	0.368	
	156.4	269.7	264	166.2	166.1	213.6	173.5	231.2	58.71	
	26.03	56.28	58.29	27.93	27.53	44.7	33.08	42.93	12.22	
	2582	3667	3948	2678	3119	3924	3627	3330	508.4	
	18835	27604	29286	18684	17929	25239	19599	24329	4835	
	10589	11678	11920	11672	10994	9797	12482	12035	1284	
	290.5	44.44	38.06	760.7	270	56.68	285	171.3	214.9	
	39	16.66	10.85	56.16	48.18	14.01	34.46	27.5	15.55	
	9.032	18.3	17.59	6.632	7.356	16.81	6.081	14.4	5.833	
	15.22	11.58	11.87	26.44	21.48	10.99	17.45	14.32	5.386	130
	7.243	3.216	3.544	9.793	6.181	3.896	5.743	4.732	2.238	
	21.22	29.48	28.87	22.25	22.35	27.41	27.88	25.97	3.081	130
	0.149	0.034	0.04	0.29	0.262	0.037	0.07	0.098	0.09	10
	4.785	11.04	12.3	5.504	5.099	8.558	4.871	11.66	7.387	32
	8	26	30	8	8	22	10	21.2	10.5	
	4.54	2.83	2.61	4.25	4.24	2.76	6.5	3.82	1.64	
6a								Mean	SD	SGV

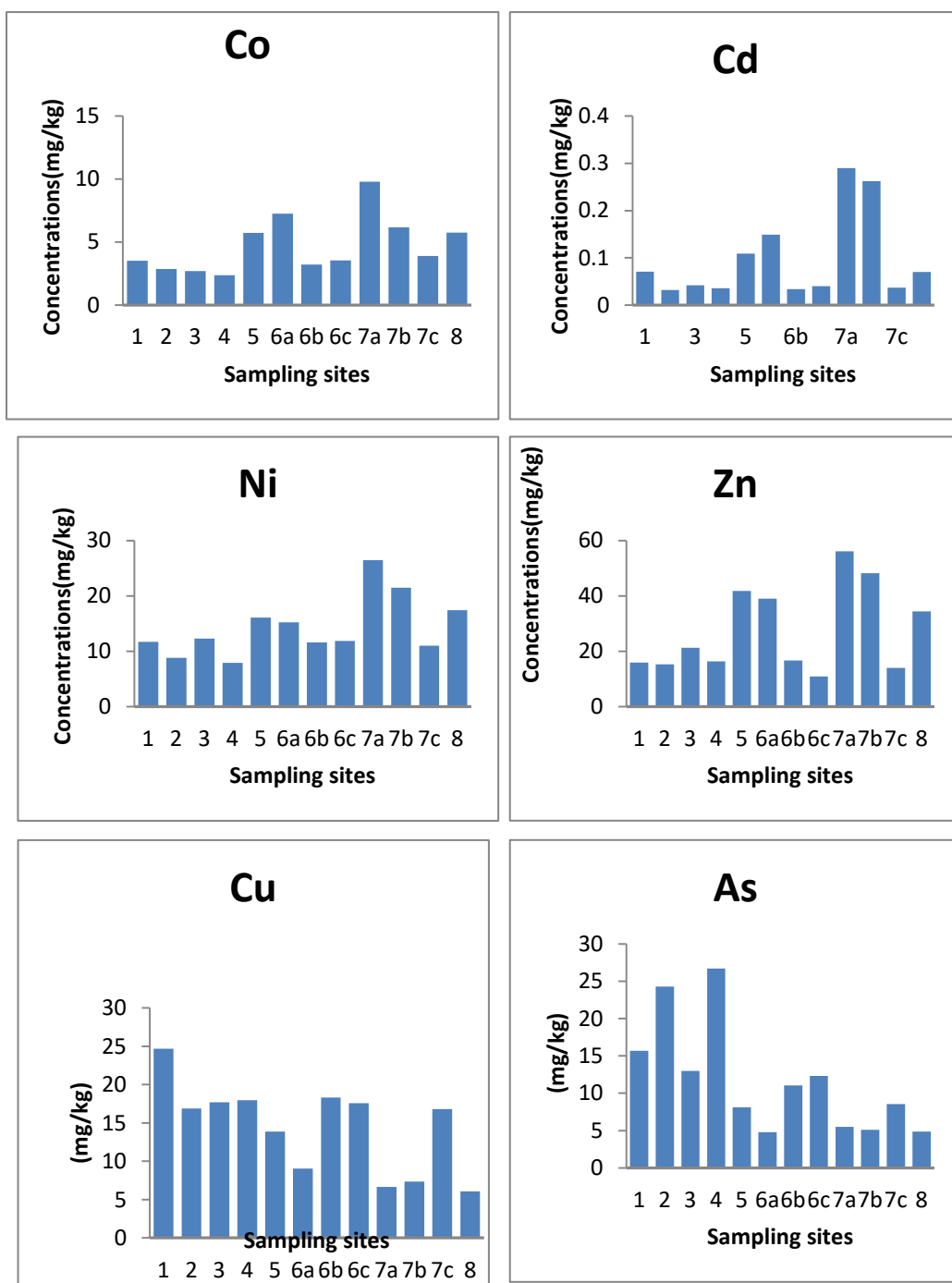


Figure 2-Distribution of Potentially Toxic metals in Bagworth Heath

Table 3- Concentration of some major elements (mg/kg) in Bagworth heap samples with mean, range and SD

Samples	Na	Mg	K	Ca
1	400.5	1173	5567	649.2
2	483.7	1319	6819	604.9
3	440.1	1358	7065	928.1
4	416.4	1348	6905	348.1
5	530.5	5736	7744	9006
6a	685.8	4239	11484	644.1
6b	438.2	1811	7582	462.1
6c	601.6	1334	6536	672.5
7a	698.4	4408	12306	690.8
7b	736.4	4442	12537	649.1
7c	437.8	1664	7462	1571
8	600.8	6173	15008	1150
mean	539.2	2917	8918	1448
Range	400.5-698.4	1172-6173	6536-15008	348-9005
SD	121	1922	3050	2402

Concentrations of major elements in soil at sampling points are shown in Table-3. In general, order of occurrence of major elements in soil samples was $K > Mg > Ca > Na$. The concentrations of these major elements in the studied area are higher if compare to the other trace elements in the soil.

Possible correlations were observed between metal-metal, metal-pH and metal-LOI throughout the study, using the Pearson correlation coefficient, r , $p < 0.05$ and 0.01 as shown in Table-4. The statistical relationship between metals is related to the same common sources. For example, the correlation between on one hand Zn and Cd and on the other Cu and Pb are related to their sources, Zn and Cd from sphalerite Pb and Cu from galena and As from pyrite [13].

The negative correlation between pH and metals, particularly Cu, As and Fe and Al indicate that, these metals may have a common source in pyrite oxidation. The amount of some metals, for instance, Al in the present study, increases dramatically, once the soil pH decrease to below 5.0 [13].

LOI is the paramount important feature which controls the binding of trace elements in soil [15]. In the present investigation, the concentration of some metals, including Al, Cu, As, Mo increase as the percentage of organic carbon in the samples increase, Table-4, and vice versa for Mn, Co, Ni, Zn, and Cd.

Table 4- Pearson correlation coefficients for pH, LOI and trace elements

	pH	LOI	Al	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Se	U	Pb	Cd	Mo
pH	1															
LOI	-0.483	1														
Al	0.610*	0.727**	1													
Cr	-0.489	0.437	0.802**	1												
Mn	0.439	0.730**	0.73**	0.633*	1											
Fe	-0.287	0.433	0.541**	0.507*	-0.286	1										

3.3 Chemical properties and metal concentrations in water samples

Hydrogen ion and total trace metal concentrations are demonstrated in Fig 5. The pH values were observed being in alkaline ranges, were 7.6 for piped water and 8.12 for pond water in the vicinity of the old mine area. The slightly alkaline pH consequences from the dissolution of calcite and dolomite and as a result of carbonate buffering ($\text{HCO}_3^-/\text{H}_2\text{CO}_3^*$; $\text{HCO}_3^-/\text{CaCO}_3$; $\text{HCO}_3^-/\text{CaMg}(\text{CO}_3)_2$). As long as carbonate equilibrium is maintained or estimated, slightly alkaline pH can be maintained despite continued amounts of pyrite oxidation. Trace metal results of this investigation reveal that the surface water (piped and pond) is severely contaminated with some metals such as As, Al, Fe, Ni and Zn and slightly, with Cd, Pb, Cr and Co, Table-5. This contamination may due to the discharge of trace metals from the oxidation of metal-bearing sulfides in the mine dumps [17].

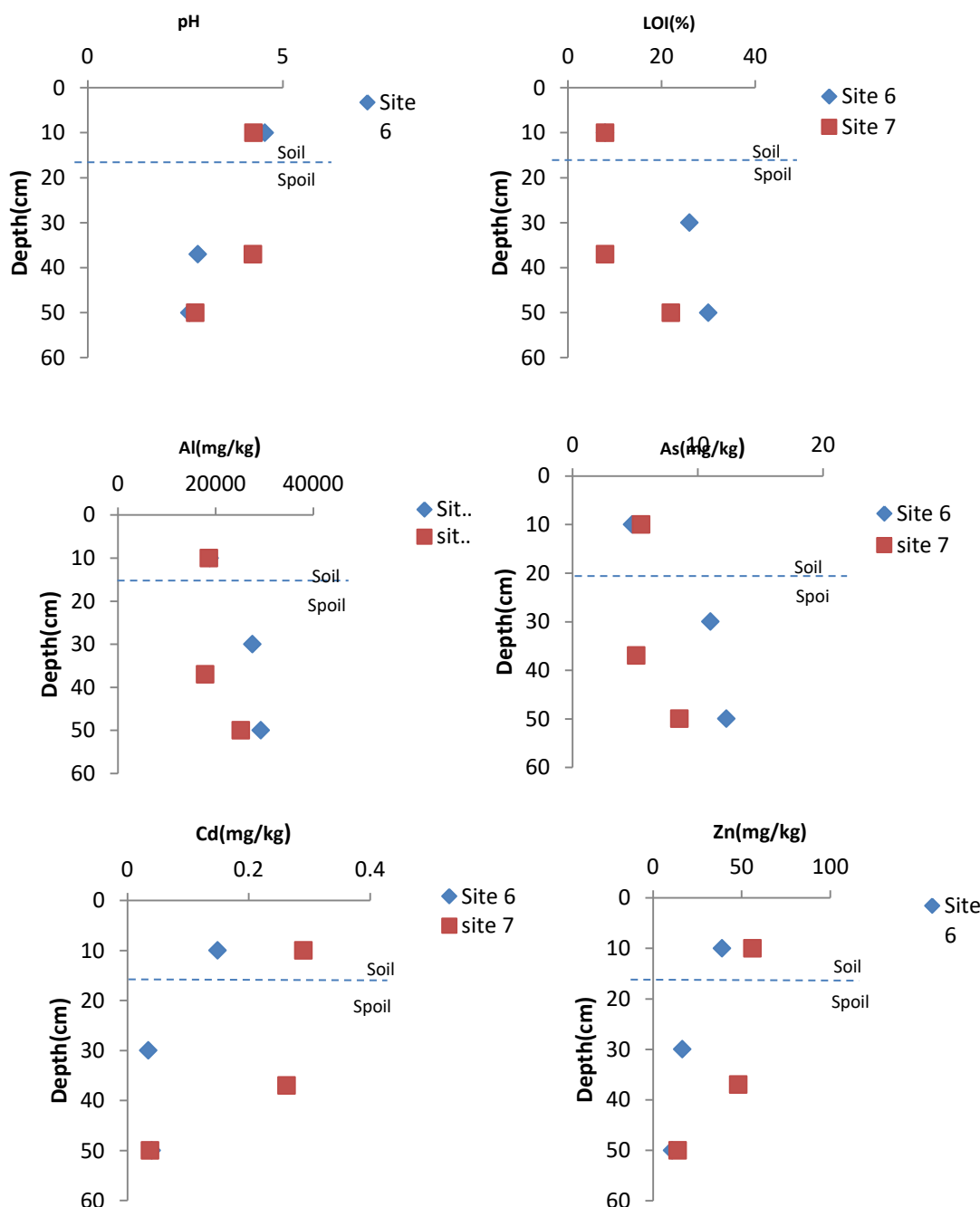


Figure 3-Relationship between soil depths with some studied soil properties and PTEs in sites 6 and 7.

Table 5-Concentrations of some PTEs (mg/l) in water samples around Bagworth Heath with mean, range and SD

Elements	Piped W	Pond W	Mean	SD	WHO GV
pH	7.6	8.12	7.86	0.367	7
Na	15.9	15.46	15.68	0.311	
Mg	26.16	24.51	25.335	1.166	
K	6.702	6.974	6.838	0.192	
Ca	102.7	99.77	101.23	2.07182	
Al	22.96	16.07	19.51	4.871	
Cr	0.06	0.045	0.0525	0.01	0.05
Mn	14.03	2.226	8.128	8.346	0.05
Fe	31.21	21.89	26.55	6.59	0.05
Co	0.148	0.123	0.135	0.017	0.05
Ni	3.041	1.815	2.428	0.866	0.02
Zn	3.729	0.09	1.909	2.573	3
As	0.819	0.775	0.797	0.031	0.01
Mo	0.645	0.623	0.634	0.015	
Cd	0.017	0.011	0.014	0.004	0.002
pb	0.039	0.027	0.033	0.008	0.01
Ti	2.613	1.283	1.948	0.940	
V	0.749	0.739	0.744	0.007	
Cu	-5.118	-5.368	-5.243	0.176	
Se	0.534	0.558	0.546	0.016	
Rb	3.463	3.477	3.47	0.009	
Sr	166.4	158.6	162.5	5.515	
Cs	0.011	0.009	0.01	0.001	
Ba	96.76	96.61	96.68	0.106	
U	0.732	0.745	0.738	0.009	

Although the high concentration of Al rarely occurs in natural water, a noticeable amount of it was found in the studied water samples. This concentration in the study area may be as a result of drainage from coal mines, acid precipitation, and breakdown of clays and aluminosilicates in shale [18]. According to his investigation dissolved Al > 0.5 mg/L may generally lead to destroy all fish and many micro invertebrates.

The concentration of major elements in the water sample is shown in Table-5. Unlike soil samples the order of presentation of major elements in soil samples was Ca > Mg > Na > K. These concentrations are relatively high with regard to other trace elements in the same samples this may be due to mineral hydrolysis in the water bodies [13]. In addition, the results indicate that there is no significant difference between pipe and pond water for the case of major elements Figure-4.

4. Conclusions

It can be concluded from the results that, the total concentration of most metals has been below the relevant soil guideline value (Environmental agency, 2009) whereas Bagworth spoil heap has been contaminated by some other heavy metals including Al and Fe due to past mining activities. Furthermore, surface water in the vicinity of abandoned mines has been polluted by most PTEs due to coal mine drainage.

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