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ORIGINAL ARTICLE

Environmental impact of the abandoned coal mines on the surface water and the groundwater quality in the south of Bochum, Germany

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Abstract Surface water and groundwater samples were collected from 20 locations, situated in the vicinity of the abandoned coal mine fields south of Bochum. The main objective of this research is to assess the environmental impacts of these mines on the surface water and groundwater quality as well as to determine the factors controlling these impacts. The water samples were collected from stream sources, groundwater, surface water and engineered channels during April 2011. Physicochemical parameters were measured during fieldwork. Water samples were analysed for major ions and aluminium, iron, manganese, ferrous iron, zinc and hydrogen sulphide. The hydrochemistry of the surface water and the groundwater of this area is characterized by near-neutral to alkaline conditions, represented by predominance of calcium bicarbonate and sometimes calcium sulphate water types. Hence, the surface water and the groundwater quality in this region is significantly affected by abandoned coal mines. These effects resulted from oxidation of iron disulphide minerals that release iron, sulphate and hydrogen. The presence of carbonate-rich materials, which contained within the landfilling materials, has led the releasing of calcium, magnesium and bicarbonate. These materials could be the main source responsible for raising the alkalinity of the affected water. The environmental hazard of the abandoned coal mines in this area is related to the high concentration of Fe especially in the groundwater that possess the highest Fe concentration compared to other water sources: the Fe is 18 times larger than the allowed value in drinking water. Significant spatial variations of the water pollution were noted in this study. For this reason, the environmental hazards of the abandoned coal mines in Germany should be considered at closure of coal mines in the near future. Otherwise, these mines will be sources of environmental threats unless all necessary measures are taken to reduce their impact.

Keywords Environmental hazard · Abandoned coal mines · Mine hydrogeology · Acid mine drainage · Surface water and groundwater quality

Introduction

Abandoned coal mines have a negative impact on the environment. These impacts are represented by acid mine drainage (AMD), which is a common environmental problem accompanying abandoned and active coal and metallic sulphide mines (Webba and Sasowsky 1994). In abandoned coal mines, AMD results from the oxidation of iron sulphide minerals are associated with coal deposits (WCI World Coal Institute WCI 2005). This process happens by exposure of iron sulphide minerals to atmospheric oxygen and water after the closure of the mines. Water and oxygen are the major factors catalysing the oxidation processes (Hammack and Watzlaf 1990). Conversely, mine waste, resulting during the excavation process, could also contain sulphide minerals (Lottermoser 2007). These materials are normally dumped on the surface during mine construction and ore exploitation processes (Younger 2003). Thus, water infiltrating through these materials can enhance the generation of AMD.

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AMD has negative impacts on the environment due to threats to human and aquatic organisms (Singh 1987; Drever 1997; Jennings et al. 2008). These threats are related to its low pH, which could be destructive to fish and other aquatic life (Lackey 1938). In addition, the low pH value increases the concentration of total dissolved solids, heavy metals and total suspended solids of the affected groundwater and surface water (Tiwary 2001; Johnson 2003). Heavy metals also have a negative impact on human health, plants and animals by disturbing several biochemical processes (Mudgal et al. 2010). However, it should be noted that not all of the abandoned mine water is acidic. This is because mine drainage could be neutral or alkaline (Scharer et al. 2000). Therefore, the International Network for Acid Prevention INAP (2009) classified mine water into three main types: AMD by pH <6, neutral mine drainage (NMD) and saline drainage (SD) by pH >6. The distinction between NMD and SD is related to the total dissolved solids TDS (<1,000 is NMD and 10,000 > TDS > 1,000 is SD).

The generation of AMD from abandoned coal mines has been documented worldwide. Many sites have been shown significant impact such as Witbank Coalfield in South Africa (Bell et al. 2001) and the abandoned coal workings in county Durham, England (Younger 1995). On the other hand, weak impacts NMD have been observed in several other sites such as the Scottish mine waters (Younger 2001). However, conditions ranging between strongly acidic to near-neutral or alkaline were also found in some abandoned coal fields such as the coalfields of Pennsylvania, United States (Cravotta III et al. 1999) and Makum coalfield, India (Equeenuddin et al. 2010).

Germany is the world's seventh largest coal producer and has the largest coal reserves in Europa. The three main coalfields of lignite deposits, which belong to the tertiary age, are present in the Rhineland, Lusatia, and Central German basins. Significant hard coal coalfields are present in the Ruhr, the area in which the study took place, and Saar basins (Kelly 2009; Miller 2010). The rich coalfields were the basis for economic success and therefore the Ruhr area became one of the most densely populated zones in Europe. The coal deposits of this area belong to the carboniferous age (Miller 2010). The water supply depends largely on the surface waters of the Ruhr and its tributaries. This includes the industrial and drinking water for its 5 million inhabitants (Bode et al. 2003).

Following legislative action taken by the German government, all remaining black coal mines in Germany will be shut down by 2018. Thus, these mines will pose a real environmental threat, unless appropriate procedures are taken, during and after the closures.

Scientific research related to the environmental impact of abandoned coal mines on water quality is almost nonexistent in Germany. Therefore, the main aim of this research is to study examples of the groundwater and surface water affected by abandoned coal mines, discuss factors controlling the evolution of the hydrochemical composition, and evaluate the pollution and the environmental hazards caused by these mines and comparing them with other sites. Finally, this study aims to determine other factors that could affect the quality of groundwater and surface water, assess the gross risk resulting from all possible factors, and attempt to generalize the results to include larger watersheds.

Regional setting

The study area (9.6 km^2) is located in the south of Bochum (Fig. 1). This area is a part of the climate zone of northwest Germany, characterizing by marine climate of cool summers and mild winters. Continental effects sometimes dominate the climate of this zone which are represented by long periods of high atmospheric pressure and are associated with dry-hot weather in the summer and cool period in the winter (LANUV 2010). The average annual rainfall in the study area is 817.6 mm/a. The average air temperature ranges between 2.7 °C in the winter and 18.5 °C in the summer with an annual average of 10.4 °C. The study area is dominated by south and southwest winds, with mean velocities of 3.5 m s^{-1}. In addition, it has relatively high humidity with an average value of 75 %. Cloudy conditions are typical, with a sunshine duration of 1,229.5 h (Grudzielanek et al. 2011). Geographically, the study area and its surroundings are located in the transition zone separating the Bergisch-Sauerlandischen Uplands in the south and the Westphalian lowlands in the north. The topography is characterized by a layer ribs landscape that consist of small hills separated by steeply sloping v-notched valleys (Geologische Landesamt Nordrhein-Westfalen GLA-NRW 1988).

The geology of the study area is characterized by the presence of Upper Carboniferous deposits (Fig. 2), consisting mainly of the Upper Westfal A. These deposits are subdivided locally into the "Bochumer Schichten" and "Wittener Schichten" formations. Limited exposures of the Upper Namur C deposits are located in the southern part of the area (Littke et al. 1986). These deposits consist of coal seams, clay, and sandstone. In addition, the Wittener Schichten contains non-marine beds of sandy silt, sandstones, and kaolin–coal–clay stone (Hesemann 1975). The Upper Carboniferous deposits were subjected to complex tectonic processes during the Variscan Orogeny, represented by folding and associated with over thrusts as well as normal and strike–slip faults (Littke et al. 1986) as

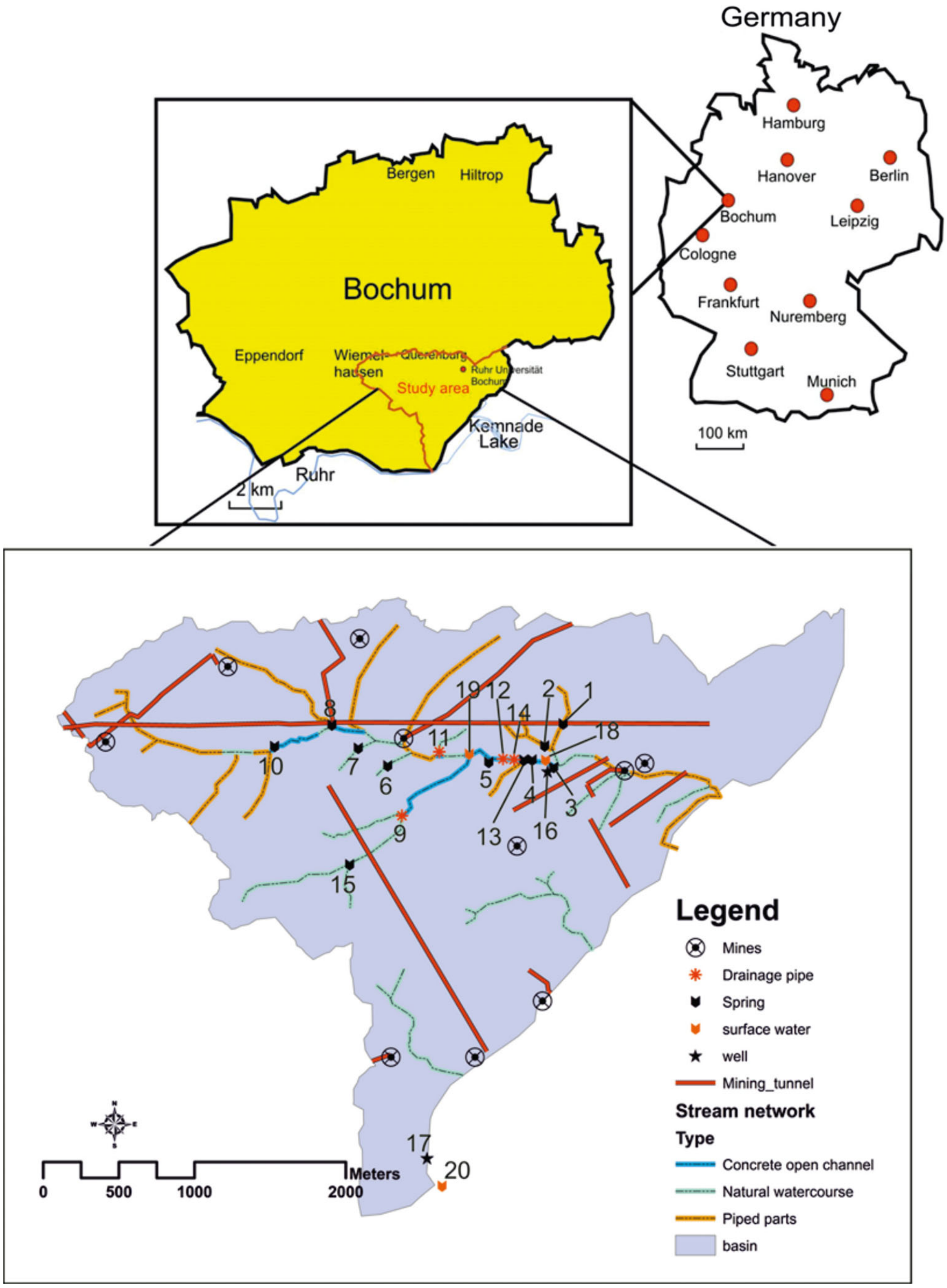

Fig. 1 Location map of the study area including sampling points, the stream network and abandoned mine sites and their facilities (location of mines and their facilities modified after Tiedt 2009)

shown in Fig. 2. The hydraulic properties of these aquifers range from moderate to very low permeability (Geologisches Landesamt Nordrhein Westfalen GLA-NRWb 1988).

With regard to the surface hydrology, the Lottenbach is the largest remaining stream system in the south of Bochum. This stream and its tributaries were subjected to significant changes, represented by reconstruction along sections of the flow path and by a large number of artificial drainage canals that connected directly to the main watercourse and its tributaries (Viebahn-Sell 2001), as a result of the mining activities and subsequent expansion of the settlement area. The eastern part of this stream was piped completely and the stream flows through this pipe into the Kemnader Lake. The Kemnader Lake reservoir was established in 1980 on the Ruhr and forms the southern boundary of the study area. This lake was developed for leisure and water sport activities and has a total storage capacity up to a maximum of 3 million m^3 (Auffermann 2010). The land use (Fig. 3) is a combination of rural activities (fields and intensive grassland) and forests. Suburban areas are situated in the western, middle and

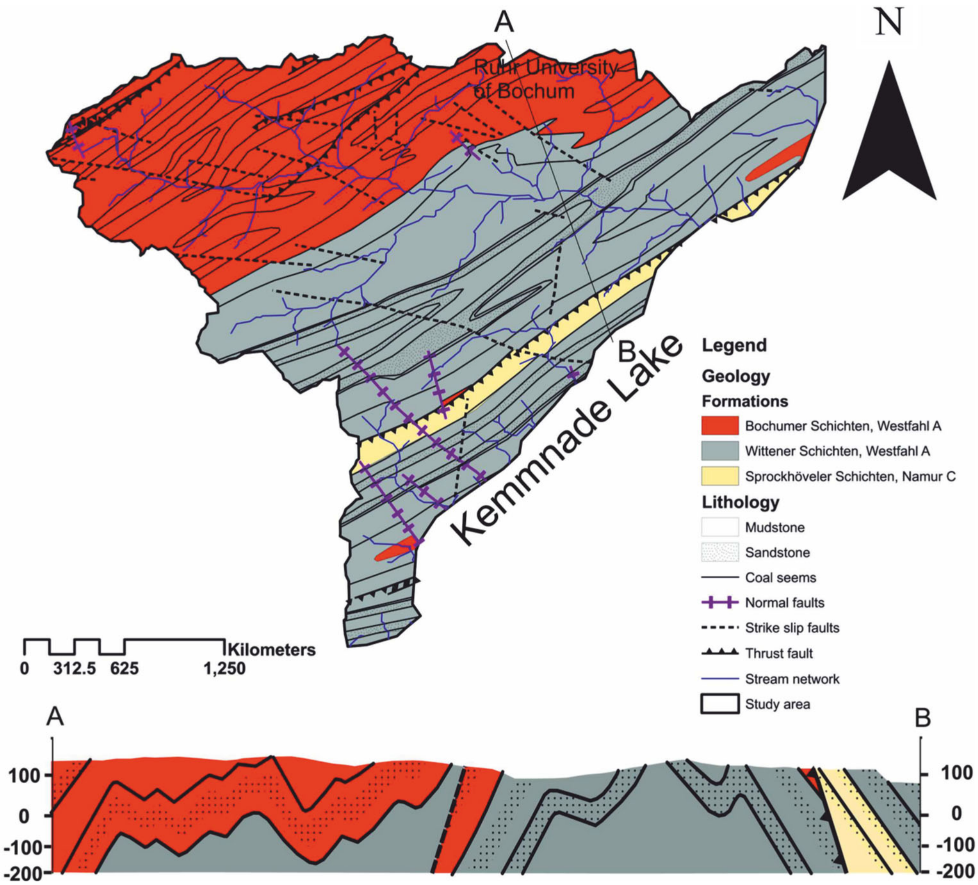

Fig. 2 Geological map and geologic cross section of the study area including the geological units, coal seams and tectonic features (modified after Geologische Landesamt Nordrhein-Westfalen GLA-NRWa 1988)

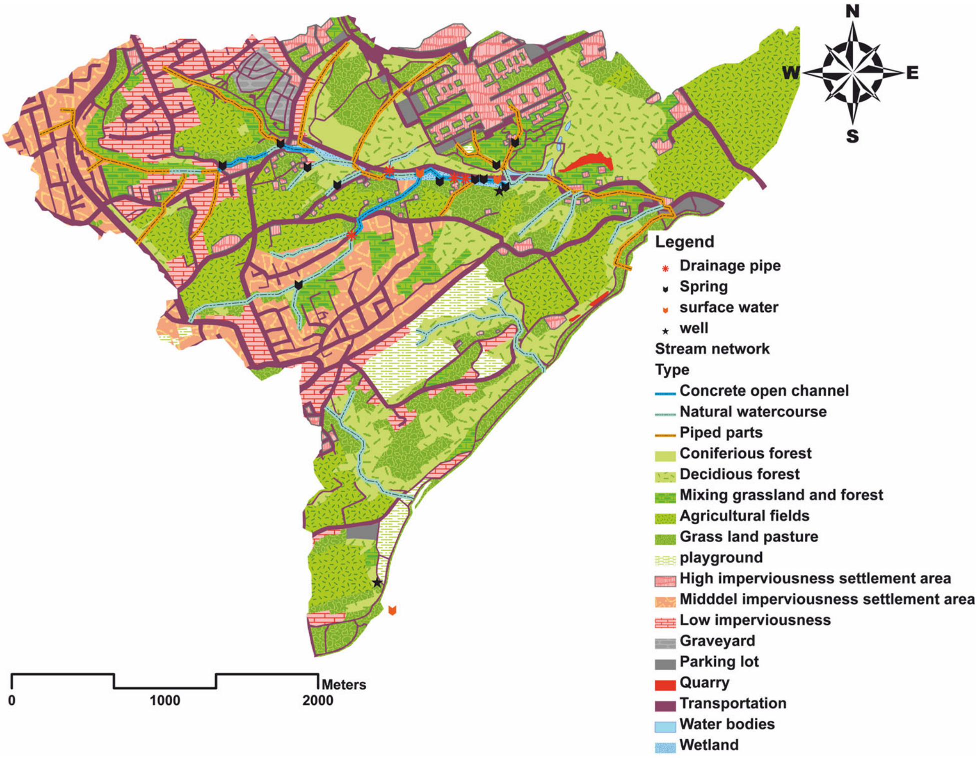

Fig. 3 Land use of the study area including the sampling points

southern part of the study area consisting of residential areas in the west and the middle and the campus of the Ruhr University of Bochum in the south (Viebahn-Sell 2006).

Materials and methods

In situ measurements, including (pH), electrical conductivity (EC), water temperature (T), dissolved oxygen (DO) and redox potential (Eh), were performed during the field investigations. WTW and Consort portable meters were used. EC was measured by using a WTW Condi340i meter. Redox potential (Eh) was measured by CONSORT C931. (DO), (T) and (pH) were measured by CONSORT C932. Each of these instruments was calibrated using standard solutions prior to field measuring.

Water samples from groundwater, surface water and engineered channels (open channels and pipes) were collected during April 2011 from 20 sites, as shown in Fig. 1. These samples were collected intensively to include all water sources in the study area.

Samples were analysed for major ions (calcium Ca^{2+}, magnesium Mg^{2+}, sodium Na^{+}, potassium K^{+}, sulphate SO_4^{2-}, chloride Cl^{-}, bicarbonate HCO_3^{-} and nitrate NO_3^{-}), minor elements (aluminium Al_{total}, iron Fe_{total}, manganese Mn_{total}, ferrous iron Fe^{2+} and Zinc Zn_{total}) and hydrogen sulphide (H_2S). According to the diversity of the preparation procedure required by some elements, each sample set was divided into the following sub-samples; major anions sub-samples (Cl^{-}, SO_4^{2-} and NO_3^{-}) were collected by 50 ml polyethylene bottles after they passed through a 0.45-µm membrane filter.

Major and minor cation sub-samples (Ca^{2+}, Mg^{2+}, Na^{+} and K^{+}, Fe_{total}, Mn_{total}, Zn_{total} and Al_{total}) were collected by 50-ml bottles, filtered by a 0.45-µm membrane filter and preserved by concentrated nitric acid, HNO_3 1 % v/v, to prevent microbial degradation and maintain the constituents of the water samples (Nielsen and Nielsen 2006). Ferrous iron Fe^{2+} sub-samples included 25 ml of water collected using 50-ml polyethylene bottles and preserved with acetic acid.

Bicarbonate sub-samples HCO_3^{-} were collected using 250-ml glass bottles. All samples were kept in a cool box and were transported to the laboratory, where they were stored below 4 °C to limit bacterial activities and the degradation of nutrient species, including (NO_3^{-} and SO_4^{2-}) (Hiscock 2005).

Hydrochemical analyses were performed by using the instruments as follows: major dissolved ions (including K^{+}, Ca^{2+}, Mg^{2+}, Cl^{-}, SO_4^{2-} and NO_3^{-}) were analysed by the DIONEX ion chromatography system model ICS-1000 (Thermo Fisher Scientific GMBH, Dreieich, Germany) HCO_3^{-} was determined using titration assay. Fe_{total} and Mn_{total} were analysed using VARIAN flame atomic absorption spectrometry system (FAAS) model AA240F (Palo Alto, US). Zn_{total} and Al_{total} were determined by UNICAM graphite tube atomic absorption spectrometry model (q2939). All of the aforementioned analyses were performed at the hydrochemistry laboratory of the Department of Applied Geology in the Faculty of Geosciences at the Ruhr University of Bochum.

The results of field measurements and hydrochemical analyses were organized in a Microsoft Excel spreadsheet. Other software was used to perform data processing as follows: calculation of total dissolved solids TDS was processed using AquaChem software v 4.0.284 Waterloo 2003. Series plot charts of field measurements and laboratory tests were plotted using OriginLab software version 8.6. All maps used in this study were digitized using geographic information system, ArcMap version 9.3 of ESRI 2008, and reproduced using CorelDraw X6, Corel Corporation 2012.

Results

Major ions hydrochemistry

The results of field measurements and hydrochemical analyses of water samples are listed in Tables 1 and 2. Table 3 shows the correlation between the hydrochemical parameters for all water samples. Figure 4 shows a series plot of in situ measurements, while the series plot of the major and minor ions are shown in Figs. 5 and 6, respectively. Hydrogen sulphide is also included in Fig. 6a. The spatial distributions of the major ions chemistry are shown in Fig. 7 and the Piper diagram shown in Fig. 9.

The surface water and groundwater of the study area varied from weak acidic to alkaline, with pH values range between 5.95 and 8.37. The maximum value was measured at sampling point 6, while the minimum value was measured at sampling point 19. EC value ranges between 306 µs/cm, measured at sampling point 7 and 762 µs/cm, measured at sampling point 17. Table 1 and Fig. 4a show high EC values measured in the surrounding area of the Ruhr University of Bochum (samples 1, 2 and 11).

The values of dissolved oxygen measured during the field campaign show that the water in this area is generally characterized by high-to-medium oxygenated conditions, as shown in Table 1 and Fig. 4b. However, sampling points 13 and 17 show a high consumption of oxygen, especially in the deep groundwater well where the dissolved oxygen concentration had a value of 0.27 mg/l (sample 17 in Fig. 4b). These conditions are associated

Table 1 Result of physicochemical, dissolved oxygen and hydrogen sulphide measurements of surface water and groundwater

Sample No.	Sample type	*X*	*Y*	*T* (°C)	pH	EC (μS/cm)	Redox (mv)	Dissolved oxygen (O_2) (mg/l)	Hydrogen (μg/l)
1	Spring	3,781,48	56,995,03	10	6.29	739	252.50	5.17	1
2	Spring	3,792,88	57,001,36	8.6	6.46	758	238.50	4.29	0
3	Spring-pond	3,787,83.5	57,001,78	9.3	6.11	384	265.90	3.09	0
4	Spring	3,785,64.4	56,974,89	15.1	7.3	384	199.60	5.49	5
5	Spring	3,785,02.4	56,975,03	14.2	7.57	606	187.20	6.37	1
6	Spring	3,793,02.6	57,000,74	8.2	5.95	543	259.30	5.91	1
7	Spring	3,791,05.3	5,700,145	11.9	6.3	306	245.80	7.25	1
8	Spring	3,791,89.5	57,001,32	11	7.52	580	183.50	5	1
9	Spring	3,791,55.9	57,001,32	10	6.37	387	232.60	5.04	2
10	Spring	3,785,83.1	57,001,93	10.2	7.95	445	172.50	7.7	4
11	Channel	3,774,96.6	57,002,25	8.4	7.69	744	180.70	7.57	0
12	Channel	3,779,90.7	56,994,43	9.4	7.44	573	197.20	7.32	2
13	Spring	3,778,74.8	57,003,68	10.7	6.5	310	245.80	1.09	39
14	Channel	3,780,66.8	57,002,03	10.7	7.56	756	185.90	5.45	2
15	Channel	3,793,98.1	57,003,58	11.6	7.38	500	190.60	7.6	1
16	Shallow ground water	3,782,44.7	57,000,99	12	6.7	407	281.20	8	0
17	Deep ground water	3,789,12.8	57,001,22	12.3	6.4	762	220.50	0.27	–
18	Surface water (water course)	3,792,82.8	57,002,30	9.6	7.37	542	197.30	7.15	9
19	Surface water (water course)	3,793,42.7	57,001,06	13.7	8.37	554	149.00	7.79	1
20	Surface water (lake)	3,792,24	57,001,37	5.7	7.1	482	165.80	10.52	–

- not measured

with oxidation-redox potential values range between 149 and 281.2 mv and have an average of 212.57 mv. The maximum value was measured at sample 16, which represents the shallow groundwater while the minimum value was measured at sample 19, which represents the surface water of Lottenbach stream.

The results of hydrochemical analyses of water samples collected from the study area showed that Ca^{2+} is the dominant cation (Fig. 5), with a concentration ranging between 1.27 and 4.9 meq/l and an average of 2.91 meq/l. The maximum value was measured in sample 5, while the minimum value was measured in sample 7. Mg^{2+} concentrations ranged between 0.58 meq/l in lake water and 2.84 meq/l in deep groundwater, with an average value of 1.25 meq/l. Generally, samples 1, 2, 8, 11, 14 and 17 show relatively high concentrations of Mg^{2+} in comparison with the other samples. Na^{+}+K^{+} ranged between 0.38 and 2.09 meq/l. The maximum value was measured at the drainage channel of the Ruhr University of Bochum (sampling point 11), and the minimum value was measured at sampling point 5. In general, relatively high concentrations were measured in the surface water (samples 18, 19 and 20), deep groundwater (sample No. 17), springs (1, 2, 6 and 11) and channel (samples 12, 14 and 15). These samples have Na^{+}+K^{+} value ranging between 1 and 2 meq/l, while other samples contained $Na^{+} + K^{+}$ concentrations less than 1 meq/l.

Figure 5b shows that HCO_3^{-} dominates the anionic composition of samples 3, 4, 5, 10, 13, 16 and 17. The maximum value of 5.4 meq/l was measured in the groundwater (sample 17), while the minimum value of 0.65 was measured at sampling point 6. The average value of HCO_3^{-} is 2.69 meq/l. $SO4^{2-}$ dominates in the surroundings of the settlement areas including samples 1, 2, 9 and 11, as well as sample 8. This sample is located downstream of the adjacent mine sites as shown in Fig. 1. The maximum value is 4.58 meq/l measured at sampling point 1, whereas the minimum value is 0.26 meq/l measured at sampling point 13. The average value of SO_4^{2-} in this area is 1.72 meq/l. Cl^{-} ranges between 0.52 and 0.96 meq/l, with a maximum concentration in sample 11 and a minimum concentration in sample 8. In general, Cl^{-} concentrations of more than 1 meq/l were measured in channel water, surface water (samples 2 and 6). NO_3^{-} was also detected in this area and its concentration range between 0.02 and 0.71 meq/l and has an average of 0.26 meq/l.

Figure 5b shows that HCO_3^{-} dominates the anionic composition of samples 3, 4, 5, 10, 13, 16 and 17. The maximum value of 5.4 meq/l was measured in the groundwater (sample 17), while the minimum value of 0.65

Table 2 Result of hydrochemical data of the water samples collected from surface water and groundwater

Sample No.	Ca^{2+} (meq/l)	Mg^{2+} (meq/l)	Na^+ (meq/l)	K^+ (meq/l)	HCO_3^- (meq/l)	SO_4^{2-} (meq/l)	Cl^- (meq/l)	NO_3^- (meq/l)	Mn_{total} (mg/l)	Fe_{total} (mg/l)	Fe^{2+} (mg/l)	Al_{total} (µg/l)	Zn_{total} (µg/l)	TDS (mg/l)	Ion balance error (%)
1	4.31	2.01	0.92	0.26	2.74	4.58	0.84	0.23	<0.1	0.1	0.03	10	6	572.35	5.24
2	4.61	2.01	1	0.07	3.33	3.52	1.52	0.09	<0.1	<0.1	0.04	12	7	578.91	4.68
3	1.94	0.72	0.47	0.03	1.64	1.27	0.67	0.19	<0.1	<0.1	0.07	15	6	256.05	8.26
4	2.11	0.65	0.54	0.03	4.6	0.7	0.73	0.06	<0.1	0.9	0.53	98	23	273.22	7.27
5	4.85	1.08	0.38	0	4.93	0.94	0.56	0.38	<0.1	<0.1	0.7	11	6	508.13	2.72
6	1.87	1.19	1.2	0.12	0.65	1.4	2.23	0.71	<0.1	0.3	0.08	43	8	314.6	4.82
7	1.27	0.68	0.57	0.09	0.75	1.01	0.96	0.44	<0.1	0.1	0.16	8	8	205.9	7.75
8	3.57	1.85	0.63	0.09	3.75	1.4	0.52	0.02	<0.1	0.9	0.38	87	21	403.25	–
9	1.86	0.77	0.64	0.04	1.24	1.43	0.74	0.22	<0.1	<0.1	0.09	12	8	247.3	3.88
10	1.86	0.78	0.69	0.16	1.94	1.03	1.02	0.19	<0.1	0.2	0.23	11	6	284.52	8.45
11	3.01	1.68	1.91	0.18	2.54	2.98	2.96	0.06	<0.1	<0.1	0.08	38	6	538.6	11.33
12	3.36	0.9	1.2	0.12	2.64	1.41	1.43	0.58	<0.1	0.4	0.27	34	22	426.13	2.96
13	2.11	0.71	0.6	0.06	2.54	0.26	0.71	0.1	<0.1	0.5	3.17	29	15	269.1	0.07
14	4.13	2.18	1.28	0.08	4.53	2.67	1.43	0.02	<0.1	0.5	0.24	17	19	598.36	5.73
15	2.26	0.99	0.88	0.12	1.34	1.25	1.49	0.6	<0.1	<0.1	0.03	11	8	314.83	3.55
16	2.13	0.89	0.51	0.04	2.2	1.44	0.61	0.03	–	–	–	–	–	297.54	7.16
17	4.95	2.84	0.81	0.16	5.4	3.04	0.6	0.14	<0.1	4.9	3.51	175	52	668.92	0.98
18	3.04	1.11	0.94	0.09	2.84	1.42	1.27	0	<0.1	0.7	–	32	31	404.27	5.14
19	2.81	1.32	0.84	0.09	2.14	1.54	1.29	0.25	<0.1	<0.1	0.14	27	6	361	0.97
20	2.12	0.58	1.3	0.09	2	1.11	1.3	0.27	<0.1	0.2	0.35	215	67	321.76	5.83

- not measured

Table 3 Statistical correlations of the hydrochemical parameters for all water samples

Element	Parameter	*T*	pH	EC	Eh	DO	H_2S	Ca	Mg	Na	K	HCO_3	SO_4	CI	NO_3	Fe	Al	Zn
T	Pearson Corr.	1.00																
pH	Pearson Corr.	0.28	1.00															
EC	Pearson Corr.	−0.15	0.18	1.00														
Eh	Pearson Corr.	−0.09	−0.89	−0.25	1.00													
DO	Pearson Corr.	−0.20	0.49	−0.11	−0.44	1.00												
H_2S	Pearson Corr.	0.01	−0.12	−0.39	0.12	−0.60	1.00											
Ca	Pearson Corr.	0.14	0.13	0.85	−0.18	−0.33	−0.19	1.00										
Mg	Pearson Corr.	0.02	−0.02	0.88	−0.05	−0.44	−0.29	0.80	1.00									
Na	Pearson Corr.	−0.61	0.20	0.57	−0.33	0.32	−0.17	0.15	0.32	1.00								
K	Pearson Corr.	−0.31	0.01	0.50	−0.09	0.04	−0.14	0.21	0.47	0.52	1.00							
HCO_3	Pearson Corr.	0.44	0.27	0.53	−0.28	−0.39	0.04	0.78	0.57	−0.08	−0.10	1.00						
SO_4	Pearson Corr.	−0.23	−0.18	0.83	0.13	−0.19	−0.39	0.65	0.80	0.45	0.64	0.26	1.00					
CI	Pearson Corr.	−0.49	0.18	0.39	−0.23	0.37	−0.20	−0.06	0.12	0.89	0.38	−0.30	0.26	1.00				
NO_3	Pearson Corr.	−0.10	−0.17	−0.14	0.06	0.25	−0.21	−0.23	−0.29	0.07	0.12	−0.49	−0.21	0.25	1.00			
Fe	Pearson Corr.	0.36	−0.18	0.43	0.02	−0.66	0.14	0.58	0.66	−0.15	0.12	0.66	0.29	−0.38	−0.24	1.00		
Al	Pearson Corr.	−0.17	−0.02	0.09	−0.26	0.01	0.06	0.08	0.15	0.17	0.05	0.32	−0.04	−0.11	−0.15	0.53	1.00	
Zn	Pearson Corr.	−0.25	0.00	0.09	−0.27	0.04	0.27	0.13	0.12	0.18	0.02	0.33	−0.05	−0.15	−0.18	0.49	0.92	1.00

Pearson Corr Pearson correlation

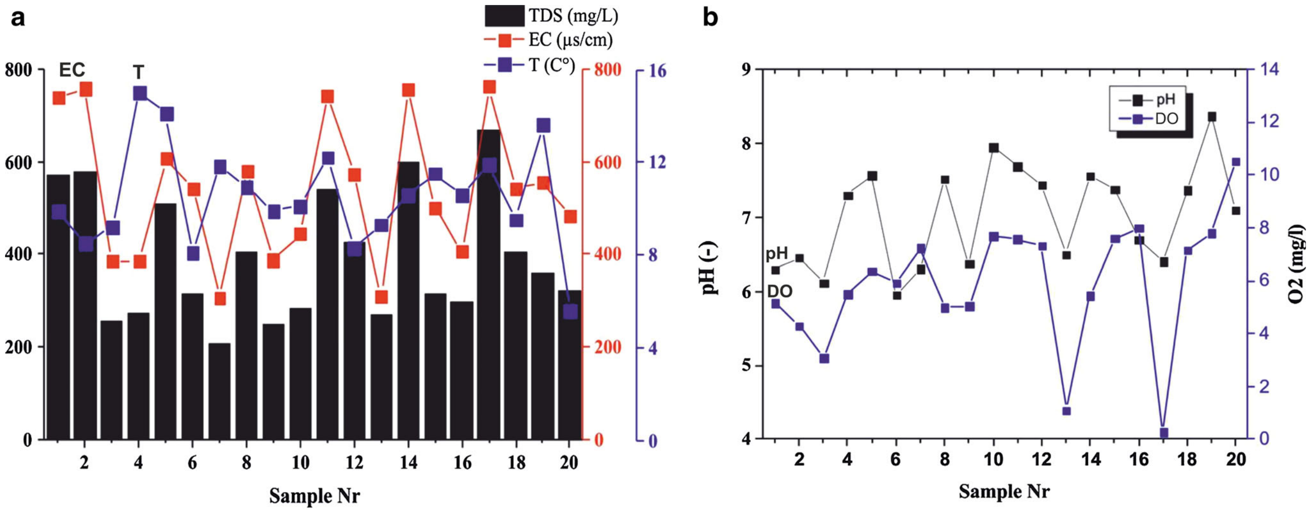

Fig. 4 Series plot of the in situ parameters of the surface water and groundwater (**a** EC, T, TDS; **b** DO, pH)

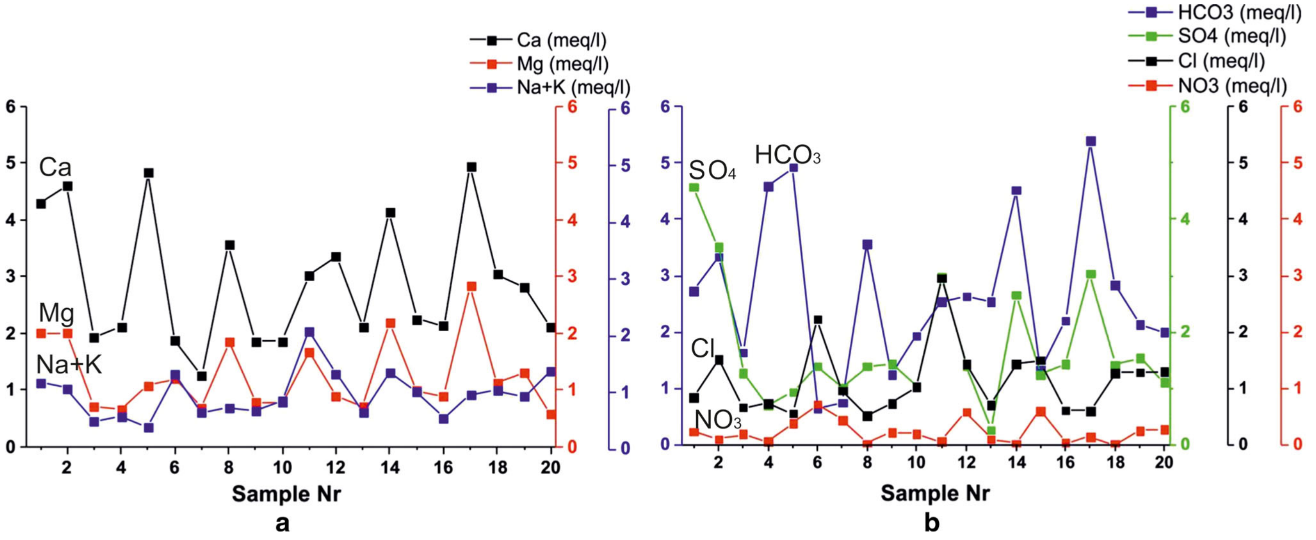

Fig. 5 Series plot of major elements of the surface water and groundwater

was measured at sampling point 6. The average value of HCO_3^- is 2.69 meq/l. $SO4^{2-}$ dominates in the surroundings of the settlement areas including samples 1, 2, 9 and 11, as well as sample 8. This sample is located in a downstream area of the adjacent mine sites as shown in Fig. 1. The maximum value is 4.58 meq/l measured at sampling point 1, whereas the minimum value is 0.26 meq/l measured at sampling point 13. The average value of SO_4^{2-} in this area is 1.72 meq/l. Cl^- ranges between 0.52 and 0.96 meq/l, with a maximum concentration in sample 11 and a minimum concentration in sample 8. In general, Cl^- concentrations of more than 1 meq/l were measured in channel water, surface water (samples 2 and 6). NO_3^- was also detected in this area and its concentration range between 0.02 and 0.71 meq/l and has an average of 0.26 meq/l.

The calculated TDS of the water samples show that the study area is characterized by fresh water, with TDS values <1,000 mg/l (Cherry and Freeze 1979). The TDS values range between 658.9 and 178 mg/l. The maximum value was calculated for sample 17, collected from deep groundwater wells, while the minimum value was calculated for sample 7. The average TDS value in this area is 392 mg/l. All samples had an ion balance error <10 %. An exception was found for sample 11, which has an error close to 11 %. The ion balance error could be the result systematic laboratory error including salt standards and dilutions, and could be either positively or negatively charged (Fritz 1994).

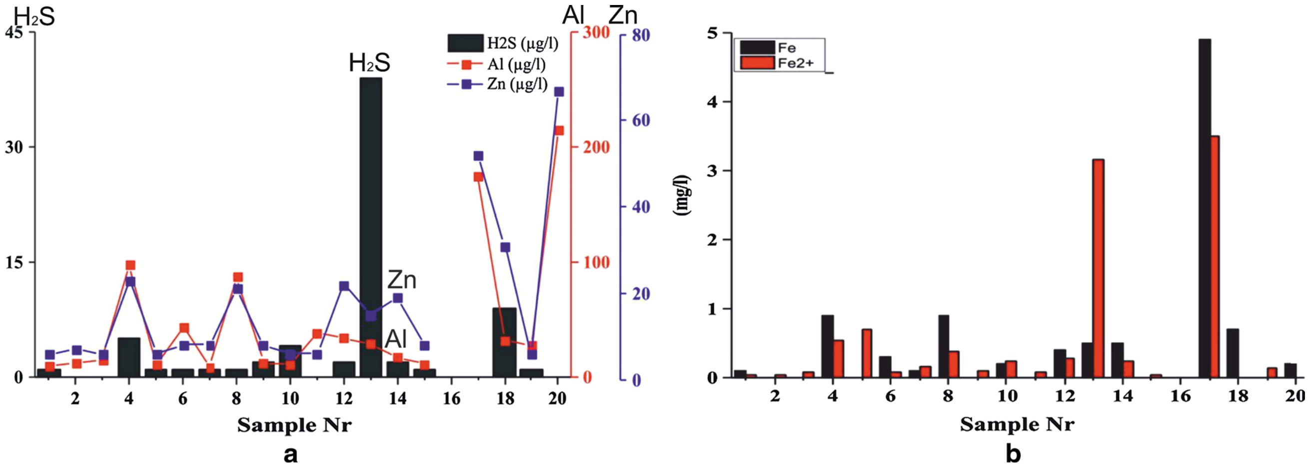

Fig. 6 Series plot of minor elements and hydrogen sulphide of the surface water and groundwater (**a** H_2S, Al, Zn; **b** Fe, Fe^{2+})

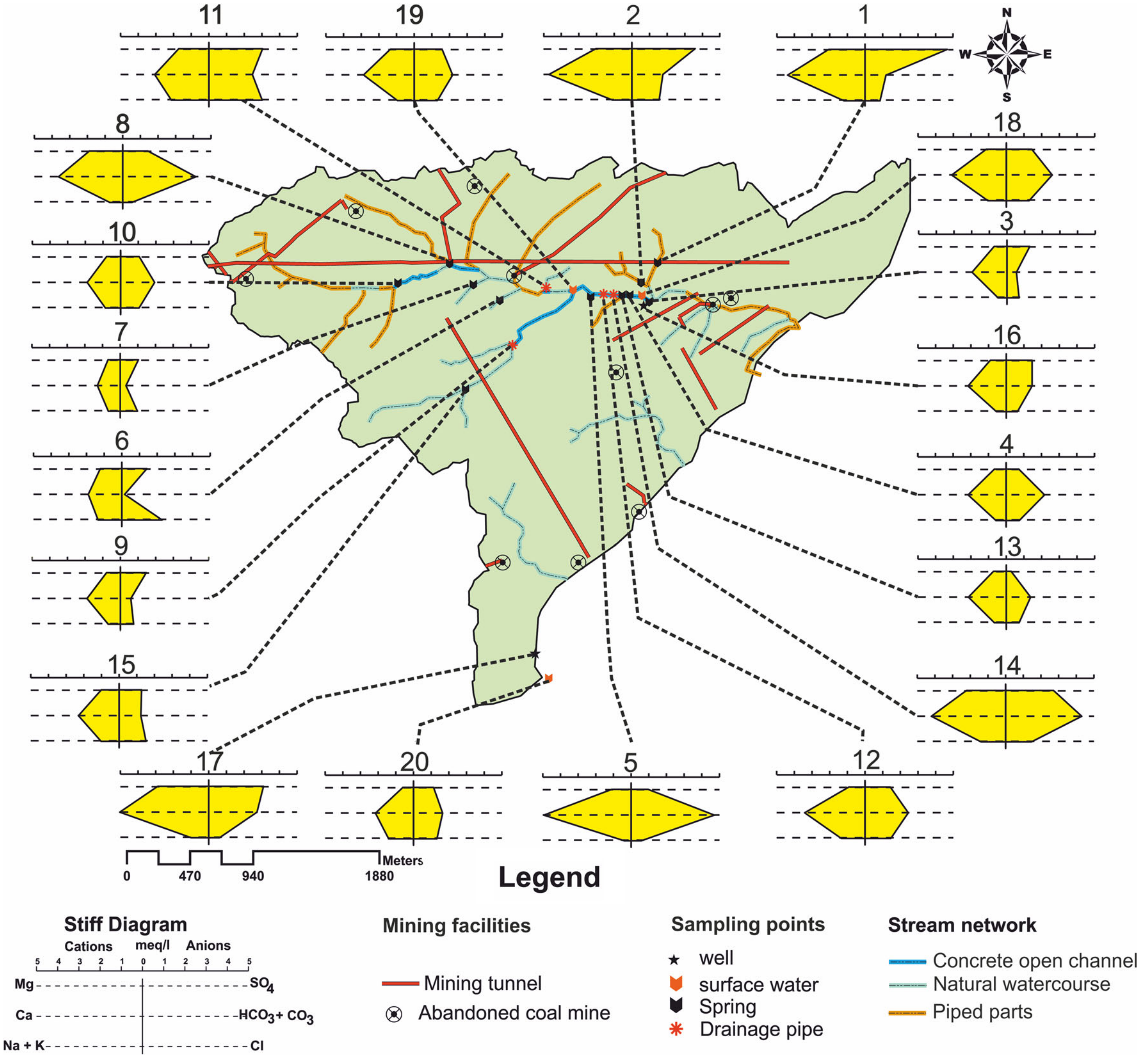

Fig. 7 Spatial distribution of the major ion chemistry in the south of Bochum, Germany

Heavy and trace elements

The Zn_{total} concentration ranged between 6 and 67 µg/l, the minimum value measured in the samples 1, 3, 5, 10, 11 and 19 and the maximum value measured in the lake water (sample 20).

Figure 6a shows that Al_{total} has a high concentration of 215 µg/l in the lake water (sample 20). This value is the maximum value measured in the study, while the minimum value is 8 µg/l measured in sample 7, and the average concentration is 46.58 µg/l.

Fe_{total} concentration ranged from undetectable (<0.1 mg/l), measured in samples 2, 3, 5, 9, 11, 15 and 19, and 4.9 mg/l, measured in sample 17 (deep groundwater), with an average of 0.81 mg/l. Fe^{2+} showed a measurement error resulting from the chosen reagent and is not included in these results. Mn_{total} was undetectable (<0.1 mg/l) for all samples in this study and was also not included herein. H_2S showed a relatively high value of 39 µg/l in sample 13. This value is associated with a high-depleted concentration of ${SO_4}^{2-}$, while all other samples showed low H_2S concentrations that ranged between 0 and 9 µg/l.

Discussion

The results of the hydrochemical analyses show that the surface water and groundwater samples can be divided into two groups: the first group includes samples 4, 6, 8, 12, 13, 14, 17, 18 and 20, while the second group includes samples 1, 2, 3, 5, 7, 9, 10, 11, 15, and 19. This classification is based on the concentration of the minor elements especially Fe_{total}. Consequently, the water samples that had Fe_{total} concentration greater than 0.2 mg/l were included in the first group, while the other water samples, which had Fe_{total} concentration equal to or smaller than 0.2 mg/l, were included in the second group. Samples of the first group were also characterized by relatively high concentrations of Zn_{total} and Al_{total} compared to the sample of the second group. Sample 16 contained no data about heavy and trace elements. Therefore, this sample can only be used to compare the major ion composition with the other samples.

The relatively high concentrations of minor elements of the first group were accompanied by the presence of ${SO_4}^{2-}$. In addition, the sampling sites of these samples located in the vicinity or the downhill of the abandoned coal mines and their facilities as shown in Fig. 1. These conditions give a strong indication of the impact of the abandoned mines on these samples. This impact may be represented by the oxidation of sulphide minerals (pyrite, marcasite and sphalerite), associated with coal deposits (Skousen et al. 2000), which lead to the release of Fe^{2+}, Zn^{2+}, ${SO_4}^{2-}$ and H^+ (Evangelou and Zhang 1995; Banks et al. 1997; Costello 2003; Lottermoser 2007). Oxidation of iron disulphide (pyrite and marcasite) can be described in the following reactions (Wolkersdorfer 2008):

$$2FeS_{2(s)} + 7O_{2\,(aq)} + 2H_2O \leftrightarrow 2Fe^{2+} + 4SO_4^{2-} + 4H^+ \tag{1}$$

This reaction causes an increase in the acidity of the mine water and consumption DO. Microbiological activities catalyse and enhance this reaction (Kleinmann 1998). Water and oxygen are major factors controlling the oxidation reaction (Hammack and Watzlaf 1990). Thus, removal of one or both of these factors from the oxidation zone will slow down or cease the process (Skousen et al. 1998). In contrast, if sufficient oxygen is present in the oxidation medium, the equilibrium condition of ferrous iron Fe^{2+} resulting from reaction 1 will change, and the iron will be oxidized to ferric iron Fe^{3+} as described in the following reaction (Banks 2003):

$$2Fe^{2+} + 1/2O_2 + 2H^+ \leftrightarrow 2Fe^{3+} + H_2O \tag{2}$$

This process is associated with the consumption of hydrogen, which reduces the acidity of mine water. As the reaction continues, the released ferric iron Fe^{3+} is precipitated as iron hydroxide $Fe(OH)_3$. This process is associated with the release of hydrogen ions, which again increases the acidity of mine water as illustrated in the following reaction (Rose and Cravotta 1998):

$$2Fe^{3+} + 6H_2O \leftrightarrow 2Fe(OH)_{3(s)} + 6H^+ \tag{3}$$

Sphalerite is often associated with pyrite in coal deposits, this mineral is also oxidizes by exposure to oxygen and water as described by the following reaction (Younger et al. 2002).

$$ZnS_{(s)} + 2O_{2(aq)} \leftrightarrow Zn^{2+} + SO_4^{2-} \tag{4}$$

This reaction only increases the Zn^{2+} and ${SO_4}^{2+}$ concentration. Therefore, no direct acidity will arise from these processes (Banks 2003).

In contrast, samples of the second group are characterized by low contents of minor elements compared to the first group, despite the fact that most of them (such as 1, 2, 3, 5, 7, 9, 10, 11 and 19) have been taken from the surrounding areas of the mines or mine facilities. These conditions may be related to the presence of these points within zones of low sulphide-content, whereas other points, such as 7 and 10, may represent the surface water drained from the settled areas as shown in Fig. 3.

Samples 1, 2, 7, 9 and 11 are also located in the vicinity of the settlements. Some of these sampling sites present near a drainage adit, such as 1, 2, 7 and 11, and show relatively high concentrations of several major ions, such as Ca^{2+}, Na^+, Cl^-as shown in Fig. 7. Hence, the chemical

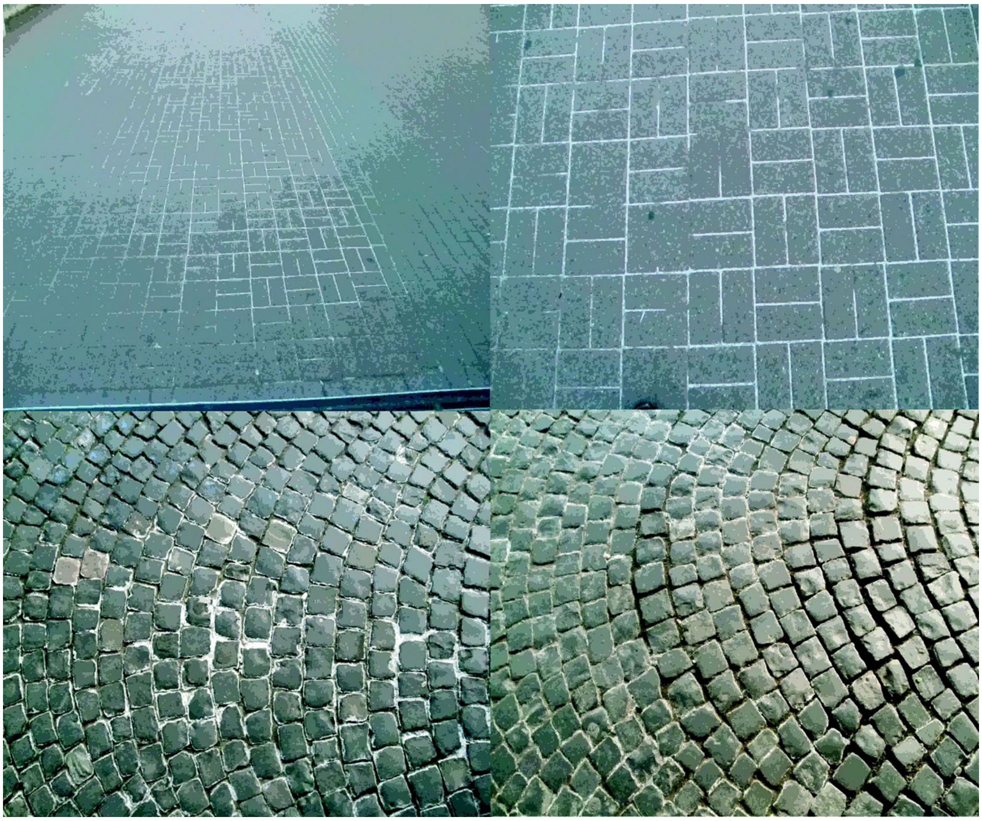

Fig. 8 Traces of the halite (*white colour* between the blocks) in the facilities of the Ruhr University of Bochum

composition of this water could be affected largely by the urbanization. In particular, the relatively high concentrations of Na^+ and Cl^- that measured in samples 1, 2 and 11. These samples were taken from the surroundings of the Ruhr University of Bochum. Thus, the urbanization activities can be considered the main source of these elements. Samples 15 and 19, representing surface water, show similar conditions. The most likely source of these two elements is the road salt, used heavily during snowy and icy conditions. The highly correlated relationship between Na^+ and Cl^-, shown in Table 3, reinforces this hypothesis.

Traces of the halite in the facilities of the Ruhr University of Bochum, shown in Fig. 8, are a further indication of anthropogenic source the Na^+ and Cl^-.

Additional anthropogenic influences on the quality of surface water and groundwater in this area could arise from diversity in land use, which is characterized by pastures and fields (Fig. 3). These influences are represented by the additional load of several nutrients, such as NO_3^-, SO_4^{2-} and other elements (Na^+, K^+, $Ca^{2+}$and Mg^{2+}) contained in the fertilizer widely used in Germany (BLFU 2004; LNW 2012).

A significant match was found between the average of the major ion chemistry of the first group and the second group as shown in Table 4, especially Ca^{2+} and HCO_3^- dominating the ions compositions of the most samples of these groups.

The high correlation between calcium-carbonate and magnesium-carbonate, shown in Table 3, gives an indication of the dissolution of carbonates. These reactions consume H^+ ions, released by oxidation of iron sulphide minerals or by precipitation of metal hydroxides. The dissolution of carbonates can be described by (Younger 2001; Wolkersdorfer 2008);

$$CaCO_{3(s)} + H^+ \leftrightarrow Ca^{2+} + HCO_3^- \quad (5)$$

$$Ca\,Mg(CO_3)_{2(s)} + 2H^+ \leftrightarrow Ca^{2+} + Mg^{2+} + 2HCO_3^- \quad (6)$$

However, carbonate minerals are not common in the study area. Therefore, anthropogenic sources may be the main source of carbonates control the neutralization of the abandoned mine water in this area. The anthropogenic sources can be related to the materials used in sealing of the coal mines. These materials consist mainly of mining waste and to a lesser extent of ash, garbage, slags, sludge, construction waste, industrial residue and household waste (GLA-NRW 1988). Mine waste was also used in road construction (Schulz 2002a). These materials often have low amounts of calcite (Strömberg and Banwart 1999). Dissolution of these carbonates in some cases could be

Table 4 Comparison between the mean chemical composition of the samples affected by abandoned coal mines (group 1) and the samples not affected (group 2)

Element	Units	Group	Statistical parameter			
			Min	Max	Average	Std
Ca^{2+}	meq/l	Group 1	1.87	4.95	3.03	1.01
		Group 2	1.27	4.85	2.81	1.18
Mg^{2+}	meq/l	Group 1	0.58	2.84	1.33	0.74
		Group 2	0.68	2.01	1.18	0.48
Na^{+}	meq/l	Group 1	0.54	1.30	0.94	0.29
		Group 2	0.38	1.91	0.80	0.40
K^{+}	meq/l	Group 1	0.03	0.16	0.09	0.04
		Group 2	0.00	0.26	0.10	0.07
HCO_3^{-}	meq/l	Group 1	0.65	5.40	3.22	1.40
		Group 2	0.75	4.93	2.25	1.10
SO_4^{2-}	meq/l	Group 1	0.26	3.04	1.49	0.82
		Group 2	0.94	4.58	1.91	1.16
Cl^{-}	meq/l	Group 1	0.52	2.23	1.14	0.52
		Group 2	0.56	2.96	1.15	0.66
NO_3^{-}	meq/l	Group 1	0.00	0.71	0.21	0.25
		Group 2	0.03	0.60	0.24	0.16
Fe_{total}	mg/l	Group 1	0.20	4.90	1.03	1.39
		Group 2	0.10	0.20	0.13	0.05
Al_{total}	µg/l	Group 1	17.00	215.00	81.11	66.64
		Group 2	8.00	38.00	15.50	9.00
Zn_{total}	µg/l	Group 1	8.00	67.00	28.67	17.83
		Group 2	6.00	8.00	6.70	0.90
TDS	µg/l	Group 1	269.10	668.92	408.85	132.62
		Group 2	205.90	578.91	378.65	135.49

adequate to change the mine water from acidic to neutral conditions (Watzlaf et al. 2004). In addition, these materials could also include sulphide minerals (Schulz 2002b). Thus, the dissolution of carbonates may be responsible for reducing the oxidation of sulphides in these materials and in the abandoned coal mines of this area. Construction waste and slags could also contain significant amounts of calcium and magnesium carbonate (Hime 2001; Melzer 2011). Dissolution of the carbonate contents of these materials can participate in the increasing of the Mg^{2+}, Ca^{2+}, HCO_3^{-} as well as the pH value of the water flow through the mines. Conversely, coal beds and the hosted rocks of this area also contain carbonates (Kukuk and Hahne 1962). These carbonates could also be dissolved in water flowing into mines and contribute in elevation of the HCO_3^{-} and the other associating cations.

In addition, Table 5 shows a convergence between averages of the major ion chemistry of all samples collected south of Bochum and the sample taken from Kemnader Lake that receives its water from a relatively larger catchment area. These conditions could be an indication of the similarity of the conditions controlling the hydrochemical evolution of the surface water and ground water in the south of Bochum and the surface water and groundwater feeding the Kemnader Lake, which represents the surface water of the Ruhr. On the other hand, the lake water shows a low concentration of Fe_{total} and very high concentration of Al_{total} and Zn_{total}. Nevertheless, deep groundwater has a very high concentration of Fe_{total} compared to the surface water and the shallow groundwater, while the content of Al_{total} and Zn_{total} is fairly similar to lake water. Stream water has relatively low concentrations of these elements compared to the ground water and lake water. Most of the major elements of the lake water, such as Ca^{2+}, Mg^{2+}, K^{+}, HCO_3^{-} and SO_4^{2-}, have concentrations much lower than the concentration of groundwater and slightly lower compared with stream water. These situations may be the result of a higher impact of the abandoned coal mines and post-mining activities on the groundwater and the surface water (stream water) in comparison with other sources feeding into the lake or it may be caused by dilution of lake water by precipitation.

Significant differences were found by comparing the chemical composition of the water samples affected by the mines in the south of Bochum and samples taken from the abandoned Upper Bavarian coal mining (tertiary age) (Wolkersdorfer 2009) as shown in Table 6.

Table 5 Comparison between the mean chemical compositions of water samples collected south of Bochum (surface water, groundwater and engineered channel), the stream water (Lottenbach) and lake water (Kemnader)

Sample No.	T (°C)	pH	EC (µs/cm)	Eh (mv)	DO (mg/l)	H_2S (µg/l)	Ca^{2+} (meq/l)	Mg^{2+}	Na^+ (meq/l)	K^+ (meq/l)	HCO_3^- (meq/l)	SO_4^{2-} (meq/l)	Cl^- (meq/l)	NO_3^- (meq/l)	Fe_{total} (mg/l)	Al_{total} (µg/l)	Zn_{total} (µg/l)	TDS (mg/l)
A	10.89	7.01	541.05	215.03	5.66	3.89	2.95	1.28	0.84	0.10	2.72	1.75	1.14	0.23	0.86	37.22	14.33	395.95
18	9.60	7.37	542.00	197.30	7.15	9.00	3.04	1.11	0.94	0.09	2.84	1.42	1.27	0.00	0.70	32.00	31.00	404.27
20	5.70	7.10	482.00	165.80	10.52	–	2.12	0.58	1.30	0.09	2.00	1.11	1.30	0.27	0.20	215.00	67.00	321.76

A average value of all samples collected from the south of Bochum

The Upper Bavarian abandoned coal mines are represented by wider ranges and higher maximum values of all the elements compared with south of Bochum. These conditions could be an indication of the largest vulnerability by abandoned mines, especially the very high concentrations of SO_4^{2-} and Fe. Conversely, the very high concentrations of the other elements can be most likely caused by geogenic factors related to the lithology and the geographical position.

Nevertheless, the possibility of generating mine drainage with higher concentrations of sulphate and iron south of Bochum remains possible, due to the fact that the rain drainage system was connected directly to the shafts and adits so that the storm water or surface runoff generated on the urban areas enters the mine facilities cause dilution of the mine drainage. This situation can be responsible for the non-predominant anion of the most of sampling points shown in the Piper diagram (Fig. 9). In addition, the presence most of the samples affected by mines in the dilution-mixing zone on the Piper diagram could be another indication of the dilution processes. The classification of water samples collected south of Bochum into three groups on the Piper diagram was created based on the well-known samples, so that the strong abandoned coal mines-affected zone was chosen based on samples 8 and 13. These samples represent water flowing directly from the mines as shown in Fig. 1.

Table 6 Comparison between the mean chemical compositions of the samples affected by abandoned coal mines south of Bochum and the samples of the abandoned coal mining in Upper Bavaria

Element	Unit	Location	Min	Max	Average	Std
Ca^{2+}	meq/l	South Bochum	1.87	4.95	3.03	1.01
		South Bavaria	3.24	20.26	2.32	1.94
Mg^{2+}	meq/l	South Bochum	0.58	2.84	1.33	0.74
		South Bavaria	1.14	9.66	3.82	3.19
Na^+	meq/l	South Bochum	0.54	1.30	0.94	0.29
		South Bavaria	0.04	26.97	4.56	7.41
K^+	meq/l	South Bochum	0.03	0.16	0.09	0.04
		South Bavaria	0.02	4.99	0.51	1.22
HCO_3^-	meq/l	South Bochum	0.65	5.40	3.22	1.40
		South Bavaria	2.54	13.93	7.34	3.16
SO_4^{2-}	meq/l	South Bochum	0.26	3.04	1.49	0.82
		South Bavaria	0.25	50.67	11.43	16.97
Cl^-	meq/l	South Bochum	0.52	2.23	1.14	0.52
		South Bavaria	1.20	97.80	14.40	22.60
NO_3^-	meq/l	South Bochum	0.00	0.71	0.21	0.25
		South Bavaria	0.00	0.47	0.10	0.13
Fe_{total}	mg/l	South Bochum	0.20	4.90	1.03	1.39
		South Bavaria	0.04	12.6	1.7	3.72

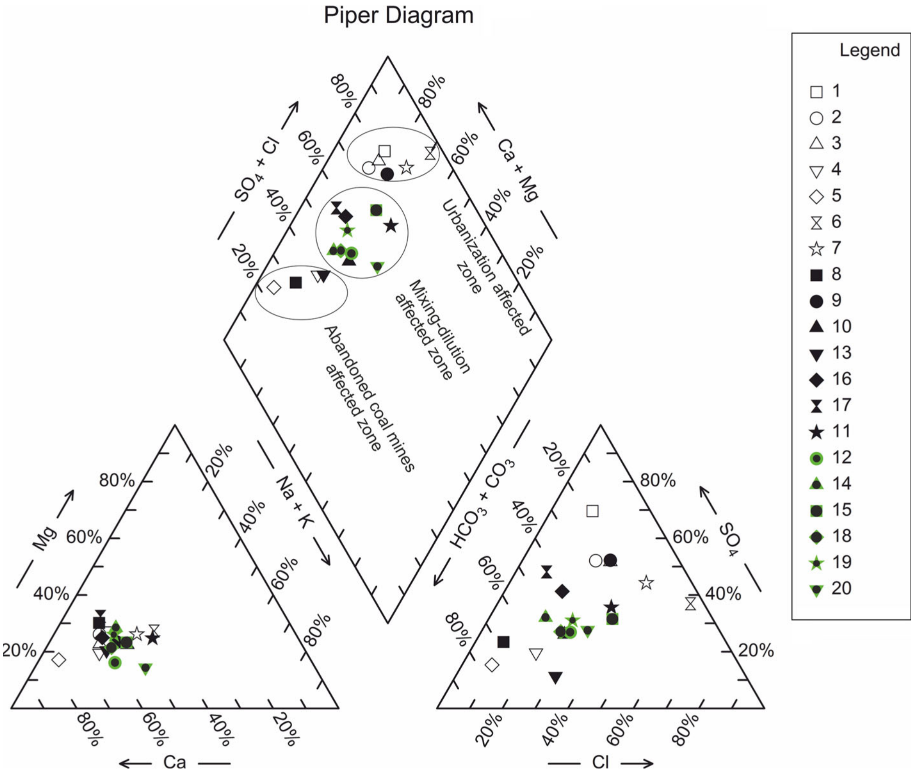

Fig. 9 Piper diagram showing the hydrochemical characteristics of the water samples listed in Table 2

The location of the strong urbanization-affected zone was based on samples 1, 2 and 9, which were collected directly from the surrounding of the settled areas as shown in Fig. 3. The dilution-mixing zone was determined by the surface water samples of the Lottenbach stream and Kemnader Lake, which normally consists of different water sources of the Lottental and the other watersheds, respectively.

Other sulphides, such as galena, greenockite, covellite, chalcopyrite, millerite and others, are often associated with pyrite. These minerals are also subject to the same oxidation conditions of iron disulphide minerals when exposed to oxygen and water (Wolkersdorfer 2008). Thus, oxidation of these minerals causes an additional increase of sulphate and undesirable ions, such as Pb, Ni, Cd and Cu, in mine water (Younger et al. 2002). Most of these metals exist in very low concentrations in abandoned mine-affected water (Wolkersdorfer 2009). Thus, no direct environmental hazard could exist from these elements. Investigation of these elements was intended to be a focus in this study, but unfortunately, these tests were not possible due to financial and technical issues.

Conclusions

The abandoned coal mines of the Upper Carboniferous, located south of Bochum, have a negative impact on surface water and ground water. This impact is represented by the pollution of groundwater and surface water by Fe, as a result of the oxidation of pyrite and marcasite, as well as the generation of AMD. Materials used in the sealing and settlement expansion include relatively high contents of carbonates. The dissolution of these minerals increases pH, consumes H^+ and releases Ca^{2+}, Mg^{2+} and HCO_3^- to the affected water. This process mitigates the generated AMD. Thus, the discharge from abandoned mines in this area is mainly characterized by near-neutral to alkaline conditions. However, the likelihood of generating acid drainage, with high concentrations of heavy metals, is still possible especially during low flow conditions from mines or adits. The deep groundwater is characterized by weak acidic conditions and very high concentration of Fe. Therefore, the use of surface water and groundwater, affected by abandoned coal mines, locally for water supply requires detailed studies to determine the temporal and spatial variations that could occur to the concentrations. Additionally, it allows for the determination of the concentrations of other harmful elements that could be associated with iron. All harmful elements must be removed from the water before delivering it to inhabitants.

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