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Groundwater contamination due to a coal-ash landfill - active-passive combined remediation solutions

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Abstract

The present work is based on a case study of a coal ash landfill, focusing on the evidence of groundwater contamination (acid drainage and high levels of sulfates, aluminum, manganese, zinc, iron and nickel) revealed by the periodic monitoring that has been carried on during the last years. The treatment of the available information allowed the development of the site conceptual model using the Groundwater Modelling System (GMS) software. After sampling, laboratory tests were performed to characterize, neutralize and sequentially precipitate the main metals underground using water collected from piezometers of the monitoring net,. The mathematical simulation of groundwater flow, combined with the results of laboratory tests, allowed to establish appropriated treatment alternatives for this case study.

Keywords: Acid mine drainage, coal ash landfill, groundwater modeling, remediation solutions.

1. Introduction

The rapid growth of energy consumption from fossil fuels in the last quarter of the 20th century prevented the environmental legislation response at the same pace. As a result, time gaps between the damage and the repair/protection of the environment can be observed all over the world. Statistical data from the last sixty years available in “the shift data portal”, shows that the energy production from coal combustion continues increasing, being the second largest source of energy worldwide (Baba *et al.*, 2004; Smita *et al.*, 2013; Verma *et al.*, 2016; Singh *et al.*, 2010). However, in Europe there is a trend towards the closure of thermal power plants in part because renewable sources are replacing coal combustion. For the coal ash deposits built in the past without effective environmental care, several solutions can be envisaged, depending on the topographic, climatic, geological, hydrogeological and hydrographic conditions of the region in which they are inserted. The present work focuses on a landfill of coal ashes deposited on hillside slope along approximately 50 years, having an accumulated volume of more than one million cubic meters of ash.

1.1. Assessment methodology

The evidence in groundwater of concentrations in some contaminants exceeding the maximum permissible values in the legislation triggered the establishment of a methodology for assessing a site intervention action as resumed in figure 1. Let’s start by groundwater monitoring through semi-annual piezometric net sampling: if the groundwater systematically presents contaminant concentrations higher than those legally recommended then it is necessary to analyze all the information (historical and current records) even though they may be difficult to access.

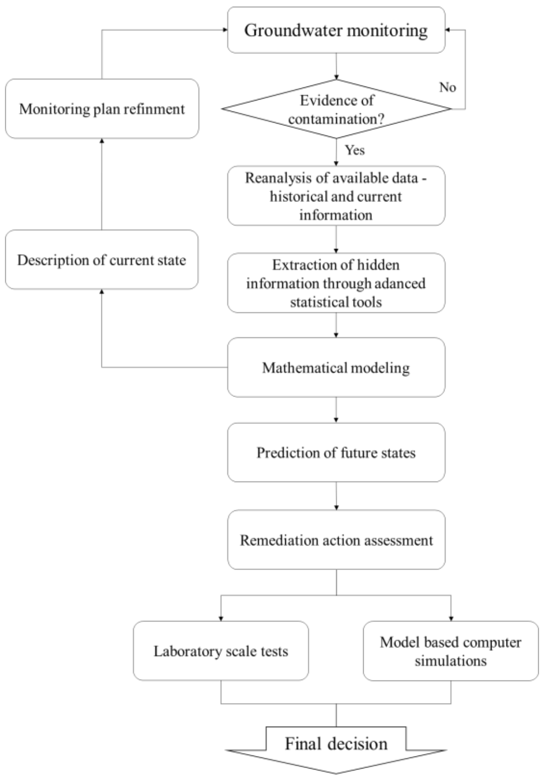

Figure 1. Conceptual decision tree for assessment of site intervention action.

The data collected should be analyzed with detail, using the advanced statistical tools such as principal components analysis among other non-linear statistical methods. Mathematical models should then be applied with two main objectives: the first is the conceptual description of the site in its main environmental compartments, namely groundwater and the porous media (soil and ashes); the second is to simulate and predict future scenarios for the site. The increase of knowledge acquired with the detailed site description allows a dynamic refinement of the monitoring plan. By another hand the simulation of future states helps on the remediation option decision and conducts to the establishment of needed laboratory tests that supported by appropriated mathematical tools will lead to the final decision.

1.2. Site description

The landfill occupies an area of about seven hectare within 20 ha belonging to the former power plant, including the inactivated coal storage park and wastewater ponds. During the operation of the thermal power plant, the wastes (bottom and fly ashes and also some slag) were deposited in a landfill that was built by taking advantage of the topography of the central neighborhood. Thus, the landfill was built on a water line, without any base lining system for protection of soils and groundwater, since at the time there was no legal constraints and the terrain did not offer structural support issues. After the power plant closure, the landfill was sealed and later classified according to the European List of Waste as 100102 coal fly ash, and a monitoring plan was then implemented. Groundwater quality has been controlled through five piezometers being two of them located inside the landfill area. Leachates are monitored through a fiber cement pipe that drains the infiltration water to the downstream of the landfill. Periodic water sampling is performed in the river where the water streamlines converge.

1.3. Geology and hydrogeology

The study area is located in an extremely metamorphized region constituted by a geological unit of schist and greywackes oriented in a cyclic *flyshes* sequence with conglomerate bankets of quartz and small ellipsoidal quartzite pebbles. In hydrogeological terms, is considered an essentially schistous homogeneous and impervious unit, constituting an anisotropic fractured medium that does not define a regional aquifer. Occurring in the studied region, the coal used in the thermal power plant was petrographically classified as an anthracite of variable composition, nevertheless with a high content on ash and sulfur (Ribeiro *et al*., 2011; Santos, 2008). A survey of the typical hydrogeochemical values for schist and greywackes formations was performed, as an attempt to distinguish groundwater contamination from background. Background values were compared with the observed values in two groundwater sampling points (PZ1 outside the landfill area and PZ3 inside the landfill area) and with the limit values for two types of groundwater usage, human consumption and agriculture irrigation according to the Portuguese legislation (Table 1). The values for PZ1 and PZ3 in table 1 show that the landfill contributes to the contamination of groundwater. This fact motivated the complementary studies to predict the extent of contamination using the GMS® software and laboratory evaluation of treatment solutions for the acidic drainage revealed by water parameters exhibited by PZ3.

2. Materials and Methods

2.1. Computational studies

The conceptual model of the region was developed using the “GMS Groundwater Modelling System” developed by U.S. Geological Survey and distributed by Aquaveo™, including the packages MODFLOW (finite difference cell-centered, saturated flow model that can perform both steady state and transient analyses with a wide variety of boundary conditions and input options) and MT3DMS a modular three-dimensional transport model for the simulation of advection, dispersion, and chemical reactions of dissolved constituents in groundwater systems (Aquaveo, 2016; Harbaugh, 2005). Input data for GMS was the site topographic map, surface and groundwater

Table 1. Typical groundwater concentrations in geological formations of schist and greywackes compared with two piezometers of the study area (PZ1 located outside the landfill area and PZ3 within the landfill area) and with the limit values for irrigation water and for drinking water.

Parameter	**Groundwater in schists /greywackes**	**PZ1** **Groundwater outside the landfill**	**PZ3** **Groundwater inside the landfill**	**Recommended limit value for irrigation**	**Recommended limit value for human consumption**
Sulfates (mg/L)	21.02 - 95.65	164.0 – 204.0	4290 – 4863	575	25
Aluminum (mg/L)	0.0 - 0.072	0.023 – 0.057	117 – 190	5	0.05
Iron (mg/L)	0.15 - 5.72	0.008 – 0.090	105 – 280	5	0.05
Manganese (mg/L)	0.11 - 1.11	1.60 – 2.60	510 – 620	0.2	0.02
Copper (μg/L)	0.8 - 31.6	< 4.0	630 – 860	200	100
Nickel μg/L)	0.4 - 153.0	4.0 – 13.0	5000 – 5600	500	50
Zinc (μg/L)	7.6 - 828.0	< 10.0 – 31.0	7000 – 10000	2000	5
Conductivity (μS/cm)	250.0 – 321.0	479.0 – 554.0	4950 – 5260	-	400
pH	-	5.1 – 5.8	3.5 – 3.8	6.5 – 8.4	6.5 – 8.5

monitoring values, some climatological data of the region, namely precipitation and evaporation. In addition to the known values it was necessary to assume, based on studies on similar formations, the values of hydrogeological parameters such as permeability, hydraulic conductivity, and dispersion-diffusion coefficients. By applying the MODFLOW package it was possible to simulate the groundwater flow as well as the seasonal variations of the piezometric levels. The application of the MT3DMS package allowed the visualization of the migration potential of the contamination plume from the ash landfill.

2.2. Experimental work

The laboratory tests aimed to verify if the combination of passive treatment neutralization systems (anaerobic lagoons) with active treatment systems could promote the gradual precipitation of the different contaminants. The contaminated groundwater sample used was collected in piezometer PZ3. Physical parameters, such as pH, electrical conductivity, dissolved oxygen and temperature were measured both in the field and in the laboratory, where the redox potential as well as the concentrations in aluminum, manganese, zinc and total iron were also determined. Neutralizing/passive treatment tests were performed in an acrylic column with 50 cm height and 3.7 cm in diameter. After a few exploratory tests the following configuration was selected: the column was filled with a limestone neutralizing layer to a height of 10 cm, followed by a 10 cm thick organic compound cover layer. After filling the column with the contaminated water, the system was closed and kept protected from light at laboratory temperature. The duration of the test was 72 hours, and the physical parameters were measured for different pre-established residence times of 0.5, 1, 2, 4, 6, 8, 10, 12, 24, 36, 48 and 72 hours. From the final liquid phase a small sample was withdrawn for chemical analysis and the remainder was used in the following precipitation assays. The remaining solid phase was separated, dried and the chemical composition was analyzed using a portable XRF equipment (Innov-X System). The precipitation/active treatment tests were phased due to the simultaneous presence of high concentrations of aluminum and manganese in the contaminated water. It is known that aluminum precipitates at a pH near 5.5, but it re-dissolves at pH’s above 9 that is the minimum value to precipitate manganese (Skousen, 2002). To the effluent from the neutralization assay it was added NaOH 1M until it reached pH 7. The obtained solution (A) was then oxidized with the addition of 3% hydrogen peroxide, adding further 1M NaOH until pH 10.5 resulting in solution B (treated water) plus a solid phase that follows for drying and weighing. Solutions A and B were subjected to chemical analysis by atomic absorption spectrometry.

3. Results

3.1. Simulated scenarios

In the 3D resulting conceptual model it is possible to distinguish between the landfill and the bedrock. It can still be observed how much the slope of the terrain is accentuated (figure 2).

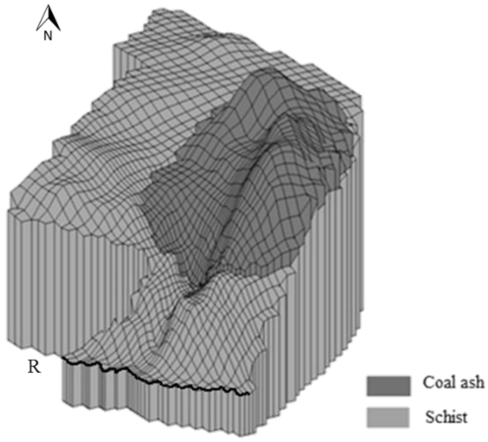

Figure 2. Conceptual model for the terrain.

The simulated average for piezometric heads in a summer situation is presented in Figure 3. It varies from over about 10 m near the river line (R) until about 60 m following the topography. In a time horizon of 100 years several simulations were performed concerning the contamination plume migration: winter and summer scenarios for the main contaminant species (Al, Mn, Zn, Ni Cu, Fe and SO_4^{2-}), as well different actions as natural attenuation and the landfill cover. Figure 4 resumes the simulations for manganese, as an example of prediction results.

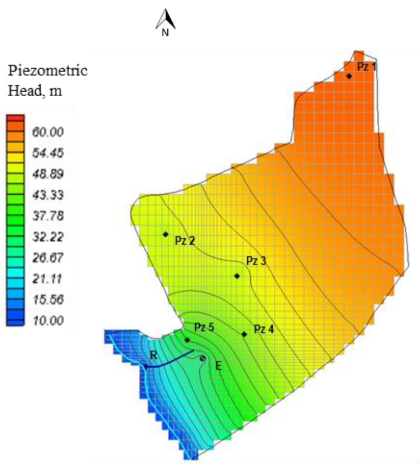

Figure 3. Simulation results for piezometric heads in a summer situation.

3.2. Active-passive remediation testing results

Measured physical parameters for the main liquid samples are presented in table 2. A slight difference between the values measured in the field and those recorded in the laboratory is noticeable. The reducing conditions created by the compost layer in neutralizing/passive treatment test allowed a decrease in dissolved oxygen in sample N, increasing this parameter in the precipitation/ active treatment tests occurred in oxidizing conditions (samples A and B). The concentrations on Al, Fe, Mn and Zn for the liquid samples (PZ3, N, A and B) are presented in figure 5.

Table 2. Physical parameters measured in the liquid samples.

Sample / parameter	PZ3 (field)	PZ3 (lab)	N	A	B
pH	3.57	3.26	5.66	7.03	10.54
Conductivity (□S/cm)	4300	4230	5250	5690	4820
O_2 dissolved (mg/L)	1.41	2.43	1.08	4.28	3.92
Temperature (ºC)	17.9	25.6	27.2	25.0	25.0

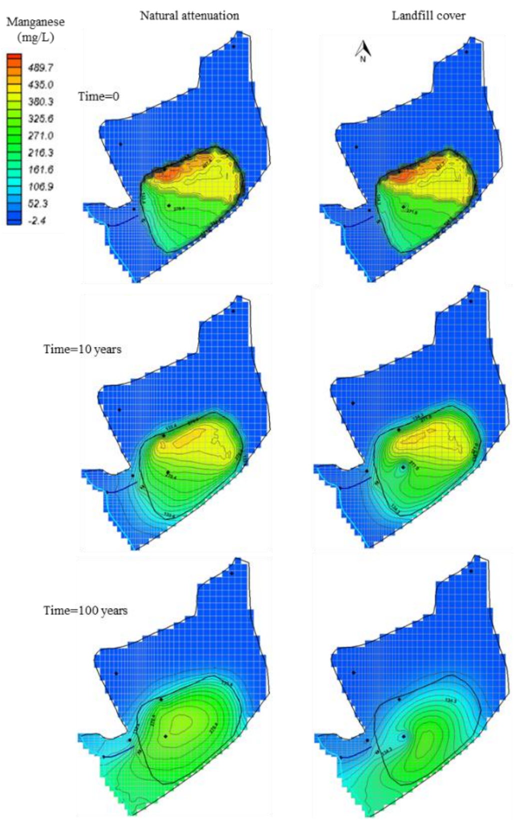

Figure 4. Simulation results for manganese concentrations in groundwater in a summer situation.

The removal of aluminum under reducing conditions allowed the further removal of manganese at a pH above 10 without the aluminum re-solubilizing. The treated water (solution B) presented values in all the studied contaminants below the maximum recommended values by the legislation. The obtained metals removal are in accordance with the respective Eh-pH diagrams, as can showed in figure 6.

4. Conclusions

In the face of a real problem of suspected contamination of groundwater by drainage from a coal ash landfill, a holistic methodology for environmental diagnosis, as well as the envisagement of feasible corrective actions were established. The use of robust computational tools, as GMS, in groundwater flow modeling and in the prediction of the transport and fate of contaminants proved to be very useful in supporting the decision of remediation techniques to test at lab scale. Combined passive-active treatment

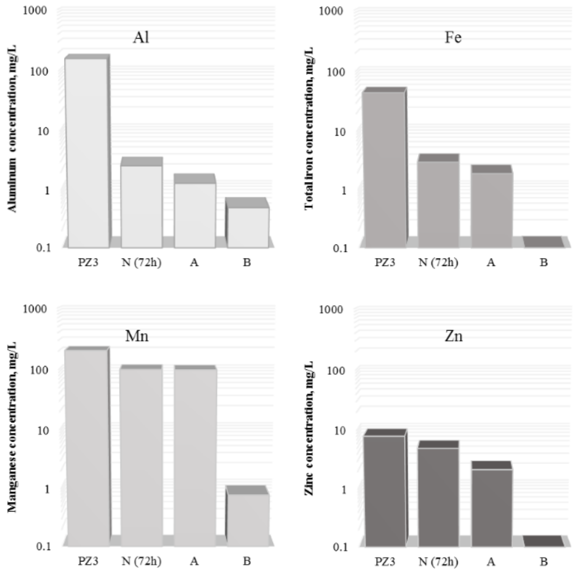

Figure 5. Determined contaminants concentration in the liquid samples under study.

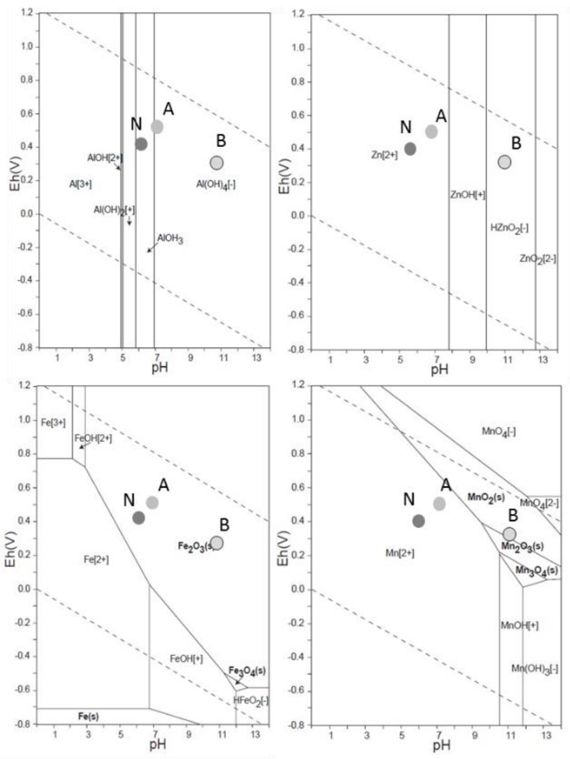

Figure 6. Laboratory results projection in Eh-pH diagrams.

solutions were tested at laboratory scale leading to promising contaminants removal efficiencies.

References

Aquaveo LLC (2016), MODFLOW - Conceptual Model Approach II. 2016.

Baba, A. and Kaya, A. (2004), Leaching characteristics of solid wastes from thermal power plants of western Turkey and comparison of toxicity methodologies. *Journal of Environmental Management*, **73**(3), 199–207.

Singh, R., Singh, R. K., Gupta, N. and Guha, B. (2010), Assessment of heavy metals in fly ash and groundwater - A case study of NTPC Badarpur thermal power plant, Delhi, India. *Pollution Research*, **29**(4), 685–689.

Verma, C., Madan, S. and Hussain, A. (2016), Heavy metal contamination of groundwater due to fly ash disposal of coal-fired thermal power plant, Parichha, Jhansi, India. *Cogent Engineering*, **3**(1), Available at: http://cogentoa.tandfonline.com/doi/full/10.1080/23311916.2016.1179243.

Cheremisinoff P.N. (1995), Handbook of Water and Wastewater Treatment Technology, Marcel Dekker Inc., New York.

Ribeiro, J, Silva, E. F., Jesus, A. P., Flores, D. (2011), Petrographic and geochemical characterization of coal waste piles from Douro Coalfield (NW Portugal). *International Journal of Coal Geology*, **87**, 226–236. http://dx.doi.org/10.1016/j.coal.2011.06.014.

Santos, R. (2008), Caracterização Hidrogeológica e Hidrogeoquímica da Área Mineira de Germunde (Pejão), PhD thesis, Instituto Superior Técnico da Universidade Técnica de Lisboa.

Smita. H.V., Panaskar, D.B.F. and Pawar, R.S. (2013), Impact of Indiscriminate Disposal of Waste from Thermal Power Plant on Groundwater Resources *Universal Journal of Environmental Research and Technology*, **3**(6), 630–640.

Skousen, J.G. (2002). A Brief overview of control and treatment technologies for acid mine drainage. *National Meeting of the American Society of Mining and Reclamation*, Lexington, 6108, 879–899.

Harbaugh, A.W., (2005). MODFLOW-2005, The U. S. Geological Survey Modular Ground-Water Model — the Ground-Water Flow Process. U.S. Geological Survey Techniques and Methods.

http://www.tsp-data-portal.org/