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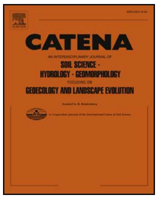

Distribution of heavy metals and metalloid in surface sediments of heavily-mined area for bauxite ore in Pengerang, Malaysia and associated risk assessment

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ABSTRACT

A detailed investigation has been conducted to evaluate the distribution of heavy metals and metalloid in the surface sediments of a bauxite mining area in association with the potential ecological and human health risk. Field sampling was carried out within the Pengerang bauxite mining areas, including mine tailings, ex-mining pond and streams. Distribution of heavy metals (Al, Cd, Co, Cr, Cu, Fe, Mn, Pb, Sr, Zn) and metalloid such as As in sediments indicated that Fe and Al constituted the greatest portion of metal elements in the sediment while Pb and Cu were found exceeding the recommended guideline values at some locations. Assessment of potential ecological risk (PERI) demonstrated low to medium ecological risk in the metal-contaminated sediments with Cd, As and Pb have generally greater risk compared to other metals, contributing the most to the total risk index (RI). The sediment enrichment factor (EF) indicated no enrichment of most metals while Pb and As at some locations were classified as having minor and moderately to severe enrichment. The geo-accumulation index (Igeo) and contamination factor (CF) indicated that the sediments were classified uncontaminated with respect to most metals. Assessment of potential human health risk revealed that the hazard index (HI) values of the carcinogenic and non-carcinogenic risks were an order of magnitude higher among children compared to adults. There were no significant non-carcinogenic risk due to metals and metalloid in the study area as HIs < 1. However, the lifetime cancer risk (LCR) for As is relatively higher than other metals and falls within tolerable LCR for regulatory purposes. Therefore, this study has highlighted the implication on potential ecological and human health risks of heavy metal occurrence in sediments of bauxite mining areas thus indicating the importance of geomorphological changes due to land exploitation for mining sector.

1. Introduction

Mineral resources are very crucial for country's mining sector development and Malaysia's national economy during the 20th century. In Malaysia, mineral resources such as iron, tin, gold, coal, silica sand, bauxite, antimony, barite, clays, copper, lead and limestone have played important roles in country's mineral production, although exploitation of some minerals had decreased significantly (Majid et al., 2013; Tse, 2015). Throughout Malaysia, minerals such as bauxite, coal, copper, gold, kaolin, silica, sand, and silver have been found in abundance in the states of Kelantan, Pahang, Sabah, and Sarawak. Specifically for bauxite, occurrence of this mineral in Peninsular Malaysia has been mostly found in Pengerang, Johor (southern Malaysia) and Kuantan, Pahang (new hot spot for bauxite mines in east-coast Malaysia) (Kusin et al., 2017). These two areas are found to be suitable for bauxite formation due to high soil temperature and annual rainfall throughout the year (Jusop, 2016). The areas are also under stable geological conditions for maximal degree of weathering of basalt and volcanic rocks, which are the major rock types in Peninsular Malaysia.

Bauxite is an ore that is mined for aluminum (Al) extraction used for production of many industrial goods, whereby the Al content could be about 12 to 25% in bauxite ore (Paramananthan, 1977). The main composition in bauxite are mineral gibbsite [$Al(OH)_3$], goethite (FeOOH), hematite (Fe_2O_3) and kaolin (Tessens and Shamshuddin, 1983; Shamshuddin and Fauziah, 2010). Bauxite is reddish brown in color which is due to the presence of mineral hematite. After the process of Al extraction, bauxite residues that consist mainly of insoluble fraction of the bauxite ore would remain. Besides aluminum, bauxite is

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also mined for Fe, Ti, Ca, Si and Na and kaolin (Jusop, 2016).

Bauxite is usually strip-mined because it is found one or two meters below the surface of the soil layer (Rajah, 1986; Paramananthan, 1977). This is typically found in open pit mining, i.e. a type of strip mining, where the material is excavated from an open pit. During every step of the process, there is always a hazard to the environment. Crushing of rock will generate large quantity of dust. During the separation process, rock slurries will produce free metals and if not properly handled, the liquids can leak into bedrocks (Saxena and Singh, 2000).

Habitat destruction, soil erosion, increase in turbidity, disturbance of hydrology are among the main impacts of mining activities (Tajam and Kamal, 2013; Demirak et al., 2013). Bauxite mining can cause caustic effluents and red-mud (Renforth et al., 2012). Harmful substances may then be released into the soil, air, and water, due to unregulated mining processes. Malaysian Ministry of Health has recently reported that increased bauxite mining activities have caused respiratory and allergy problems, and potential long-term effect of cancer (Abdullah et al., 2016). This is because the presence of metals may introduce toxicity on living organisms and also ability to be entering into the food chain (Ololade et al., 2008; Mmolawa et al., 2011; Shaari et al., 2015).

Acid mine drainage (AMD) can occur if the mined materials are exposed to the air (oxygen) and water. AMD contains high concentration of heavy metals (e.g. arsenic, cadmium, copper, lead, zinc, etc.) and sulfate. Even though the condition at the site is not acidic, toxic metals such as arsenic, cadmium and lead can leach and contaminate the ground waters and environment. The precipitates generated smothers the river bed with fine silt and hence would affect benthic organisms and the entire food chain (Penreath, 1994). This will apparently lead to huge impact on rivers and aquatic ecosystems over the long term.

It has been noted that metal contamination and land degradation from mining can give harmful effects to communities living in nearby areas (Yelpaala, 2004; Pan and Li, 2016). Therefore, it is important to understand properly the characteristics of trace metals contamination in mine-impacted areas particularly in water and sediments and their potential impact. Viers et al. (2009) stated that people are confronting with the increments of threat with regard to water security because of the sediments in many rivers and lakes that have been polluted by trace metals. Therefore, this study was conducted to determine the composition of metals and the extent of contamination in the surface sediments from bauxite mining activities, and to determine the impact in terms of the potential ecological risk and also impact on human health.

2. Materials and methods

2.1. Site description

The study was undertaken in the vicinity of bauxite mining areas located in the Pengerang, Johor (southern Malaysia) (Fig. 1). The Pengerang was once known as the only bauxite mining area in Malaysia. A new area was recently found in the Kuantan, Pahang (east-coast Malaysia). The Pengerang bauxite mining area is located on the south-eastern of Johor state and approximately 50 km from east of Singapore, specifically within the area of the Teluk Ramunia, Johor. Historically, since the early 50's, Teluk Ramunia has been well known as progressive bauxite mining town.

The Pengerang bauxite mines have been said to have the best grade of bauxite but are mostly exported. In the Pengerang, the valuable bauxite occurs in alluvial and residual deposits. In these deposits, bauxite occurs in concretions, pisolites, nodules and in shaly structure (Patterson et al., 1986). Geologically, the bauxites in the vicinity of Pengerang are formed from the acid volcanic rock coupled with stable geological conditions that has favored sufficient weathering of the rocks (Raj, 2009; Jusop, 2016). The area mined for bauxite in Pengerang is about 1800 ha which is on formerly agricultural land prior to bauxite discovery in the 50–60's (Paramananthan, 2000; Jusop, 2016).

2.2. Field sediment sampling and analysis

Sampling was carried out at several locations of the bauxite mines (Fig. 1). Triplicate sampling was done by stainless steel trowel, whereby (0 – 20) cm surface sediments were collected into a zip locked bag. The sampling was performed in May 2016 at seven sampling points. There were four samples that were collected from the mine tailings (S3, S4, S5 and S6), and the rest were collected at the nearby streams (S1 and S2) and an ex-mining pond (S7). The samples were kept in an icebox (< 4 °C) and transported to laboratory for further analysis. Sediments were maintained at room temperature, air-dried and homogenized by using mortars and pestles. Then, they were sieved through 2 mm sieve for physicochemical analyses. The sample preparation for analyses was carried out according to the EPA method 3050B (US Environmental Protection Agency (USEPA), 1996).

The sediments had to be completely decomposed before analyzing it using ICP-MS. Acid digestion method was applied for the extraction of heavy metals using hydrochloric and nitric acids (3:1 ratio). Approximately, 1 g of sediment sample was weighed and placed in digestion tubes, then, were allowed to pre-digestion overnight. The samples were digested at 40 °C in first hour, 80 °C in second hour, 120 °C in third hour and 140 °C in the fourth hour. The obtained suspensions were then filtered using 0.45 μm membrane filter. Afterwards the suspensions were placed into centrifuge tube and ready for heavy metals analyses using an ICP-MS (Perkin Elmer ELAN DRC-e). For quality assurance/quality check (QA/QC) procedure, the analytical instrument was checked with standard reference material NIST, SRM 1646a (estuarine sediment). The percentage of recoveries for the metals studied ranged between 82 and 117% (Table 1).

2.3. Potential ecological risk index (PERI)

Potential ecological risk index (PERI) can be used to determine the potential risk due to exposure to ecological sensitivity, concentration and toxicity of heavy metals (Nabholz, 1991; Douay et al., 2013). It is also regarded as a comprehensive potential ecological risk assessment, $\mathrm{E_r^i}$, which is the sum of the potential risk of individual metal element. It represents the sensitivity of the biological community to the toxic substance and illustrates the potential ecological risk caused by the overall contamination. The estimation of total risk index (RI) of PERI was calculated using Eq. (1) as proposed by Guo et al. (2010) and was first introduced by Hakanson (1980):

$$\mathrm{RI} = \sum E_r^i = \sum \mathrm{T_r^i(C_s^i/C_n^1)} \tag{1}$$

where, RI is the sum of all the potential risk factors for all heavy metal, $E_r{}^i$ is the potential ecological risk index for single heavy metal pollution that can be calculated as:

$$E_r^i = C_r^i \times T_r^i \tag{2}$$

$T_r{}^i$ is the toxic-response factor for a single heavy metal contamination. $C_r{}^i$ is the pollution index for a given heavy metal and can be defined as:

$$C_r^i = C_s^i/C_n^i \tag{3}$$

where $C_s{}^i$ is the present concentration of heavy metal in the sediment and $C_n{}^i$ is the reference value of heavy metal in the sediment. The toxic-response factors were taken as 5 for Cu, Pb and Co, 1 for Zn, 10 for As and 30 for Cd (Guo et al., 2012; Islam et al., 2015). The classification of the potential ecological hazard and the risk criteria of heavy metals in surface sediments are given in Table 2.