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Review

# The use of red mud as an immobiliser for metal/metalloid-contaminated soil: A review

Yumei Hua[a], Kate V. Heal[b,*], Wolfgang Friesl-Hanl[c]

[a] *College of Resources and Environment, Huazhong Agricultural University, Wuhan 430070, PR China*
[b] *School of GeoSciences, University of Edinburgh, Edinburgh EH9 3FF, UK*
[c] *Energy Department, AIT Austrian Institute of Technology GmbH, 3400 Tulln, Austria*

## HIGHLIGHTS

- The concentrations and mobility of metals/metalloids in red mud were assessed.
- The response of soil extractable metals/metalloids to red mud addition was compared.
- Key controls on metal/metalloid fixation due to red mud application were identified.
- Effects of red mud amendment of contaminated soil on plant growth were described.
- Bacterial activity was intensified in red mud-amended contaminated soil.

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## ABSTRACT

This review focuses on the applicability of red mud as an amendment for metal/metalloid-contaminated soil. The varying properties of red muds from different sources are presented as they influence the potentially toxic element (PTE) concentration in amended soil. Experiments conducted worldwide from the laboratory to the field scale are screened and the influencing parameters and processes in soils are highlighted. Overall red mud amendment is likely to contribute to lowering the PTE availability in contaminated soil. This is attributed to the high pH, Fe and Al oxide/oxyhydroxide content of red mud, especially hematite, boehmite, gibbsite and cancrinite phases involved in immobilising metals/metalloids. In most cases red mud amendment resulted in a lowering of metal concentrations in plants. Bacterial activity was intensified in red mud-amended contaminated soil, suggesting the toxicity from PTEs was reduced by red mud, as well as indirect effects due to changes in soil properties. Besides positive effects of red mud amendment, negative effects may also appear (e.g. increased mobility of As, Cu) which require site-specific risk assessments. Red mud remediation of metal/metalloid contaminated sites has the potential benefit of reducing red mud storage and associated problems.

## Contents

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* Corresponding author.
*E-mail address:* k.heal@ed.ac.uk (K.V. Heal).

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## 1. Introduction

Red mud (also called bauxite residue) is a by-product of alumina production from bauxite by the Bayer process [1]. The Bayer process involves washing bauxite in a hot solution of sodium hydroxide (NaOH) which leaches the aluminium from the bauxite. Development of the process was largely responsible for the major expansion of the industry during the 20th century and it currently underpins over 95% of alumina production globally [2]. The basic chemistry and operational steps of a contemporary Bayer plant are fundamentally the same as described in Bayer's patents in 1887. The chemical and physical properties of red mud depend primarily on the bauxite mineralogy and, to a lesser extent, the effect of the Bayer process on it [3]. The main processes in a Bayer plant which affect red mud production and composition are bauxite milling, pre-desilication (to react silica-containing minerals so as to reduce the time of the digestion stage), digestion with NaOH, clarification, and washing [4]. Red mud comprises the solids remaining after separation from the green liquor after digestion [4].

Red mud is produced in enormous quantities that require safe disposal [5]. Power et al. [4] estimated the annual production of red mud as 120 million t and a global inventory of over 2.6 billion t in 2007. Based on data for annual alumina production (Fig. 1a) it can be inferred that production of red mud has generally increased from 1998 to 2015. Since 2–3 t of bauxite are used in the Bayer process to produce 1 t of alumina, the amount of red mud produced can be estimated by applying a mean ratio of 1.5 to alumina production data [4], yielding approximately 170 million t of red mud produced worldwide in 2015. Red mud production is expected to increase further in the future since, due to initial exploitation of the highest quality bauxite reserves, the grade of bauxite ore is decreasing, resulting in an increased ratio of red mud production to bauxite [6]. Recently global alumina production has been driven by production in China (Fig. 1a). In 2015 China, Oceania and South America accounted for 50.4%, 18.6% and 11.6% of the global production of alumina, respectively, with the rest of the world accounting for about 20% of production (Fig. 1b). In 2011 annual red mud production in China was 42.6 million t of which 5.2% was reused [7], and in 2013 it reached 73 million t of which 2.9 million t was utilised (4% of the production amount) [6]. The total inventory of red mud deposited in China over time was estimated to be more than 300 million t by 2013 [6].

The composition of red mud varies depending upon the quality of the ore and processing methods [10–13]. On average, red mud dry mass contains large amounts of $Fe_2O_3$ (41%) and $Al_2O_3$ (17%), smaller amounts of $SiO_2$ (10%), $TiO_2$ (9%), CaO (9%) and $Na_2O$ (5%), trace elements (e.g. Cr, Cu, Pb, V and Zn) and naturally occurring radionuclides (U, Th isotopes and their daughters, and $^{40}K$) [2,13–16]. Due to washing with NaOH red mud has a high pH (9–13) and electrical conductivity (EC) ranging from 0.67 to 60.8 mS $cm^{-1}$ [5,11,17–23].

Currently, red mud is treated as a waste and is typically stored in large lagoons or in land-based disposal pits, depending on the method of bauxite processing [14]. The original water content of red mud is more than 80% [24]. Additional thickening or filtration of the slurry are applied in order to store red mud as dry stacks (45–52% water content) or dry cake (<35% water content) to minimise the land area required for storage [4]. Storage of large volumes of red mud in lagoons can lead to environmental disaster caused by high rainfall and/or failure of the containment structure [4,14]. The most serious accident [25] involving red mud was on 4 October 2010, when the western dam of the red mud reservoir at an alumina plant in Ajka (Hungary) collapsed. The red mud release killed 10 people and injured more than 100. In addition, rivers and land were contaminated across a large area, with an estimated 800 ha of agricultural land covered by a red mud layer 5–10 cm deep [25]. Other accidents involving red mud have been reported in other countries [26,27]. Li [28] identified 51 red mud pollution incidents distributed across 20 countries on 5 continents from 1900 to 2012, with the majority occurring since 1950. These numbers are expected to be an underestimate as not all red mud pollution incidents are reported.

Although the safe disposal and storage of red mud is an international issue and has been extensively researched, better solutions to the storage of red mud and the risks it poses are to reuse red mud. Red mud should be considered a valuable material instead of a waste since it has many potential reuse applications. The abundance of red mud has led to extensive research into possible uses [12]. These include: recovery of Al, Fe, and rare earth metals [29,30]; a sorbent for treatment of contaminated water [31,32]; sequestration of $CO_2$ [33]; an additive to ceramics and building materials [34,35]; embankment construction [36]; and soil amendment [2,12,14,17,37–42]. All of these uses may play a role in reducing the storage of red mud.

More recently, various industrial residues have been considered for their potential as amendments for the remediation of contaminated soils characterised by high concentrations of metals/metalloids and low pH [43]. The remediation of metal-contaminated soil is often difficult and expensive, and one cost-effective and feasible option is reduction of risk by metal fixation [44,45]. In situ fixation of metals is based on the reduction of metal mobility and availability which can be achieved by applying soil amendments [38,46]. Consequently, the detrimental effects of metals to environmental receptors such as microorganisms, plants, animals, water bodies and humans are reduced [43]. Whilst red mud represents a disposal problem worldwide due to its high concentrations of potentially toxic elements (PTEs), it has a large specific surface area suitable for remediation of metal-contaminated substrates [37]. In recent years red mud has been added to contaminated soils, mainly resulting from mining activities, to neutralise low pH and reduce metal mobility through different physicochemical mechanisms [44,47]. These include immobilisation of metals through adsorption and surface complexation with Fe oxides and formation of inner sphere complexes with Fe and Al oxides and hydroxides [47,48].

A number of studies have reported on the application of red mud from different sources for amending contaminated soil, and the mobility of different PTEs, mainly metals and metalloids, has been investigated globally [37,38,46,49–52]. Furthermore, increasing numbers of field experiments have been conducted utilising red mud to amend contaminated soils, in which a range of plants have