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## The potential application of red mud and soil mixture as additive to the surface layer of a landfill cover system

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### ABSTRACT

Red mud, the by-product of aluminum production, has been regarded as a problematic residue all over the world. Its storage involves risks as evidenced by the Ajka red mud spill, an accident in Hungary where the slurry broke free, flooding the surrounding areas. As an immediate remediation measure more than 5 cm thick red mud layer was removed from the flooded soil surface. The removed red mud and soil mixture (RMSM) was transferred into the reservoirs for storage. In this paper the application of RMSM is evaluated in a field study aiming at re-utilizing waste, decreasing cost of waste disposal and providing a value-added product. The purpose was to investigate the applicability of RMSM as surface layer component of landfill cover systems. The field study was carried out in two steps: in lysimeters and in field plots. The RMSM was mixed at ratios ranging between 0 and 50% w/w with low quality subsoil (LQS) originally used as surface layer of an interim landfill cover. The characteristics of the LQS + RMSM mixtures compared to the subsoil (LQS) and the RMSM were determined by physical–chemical, biological and ecotoxicological methods. The addition of RMSM to the subsoil (LQS) at up to 20% did not result any ecotoxic effect, but it increased the water holding capacity. In addition, the microbial substrate utilization became about triple of subsoil (LQS) after 10 months. According to our results the RMSM mixed into subsoil (LQS) at 20% w/w dose may be applied as surface layer of landfill cover systems.

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### Introduction

On 4 October 2010, the wall of a red mud storage facility at the MAL Hungarian Aluminum Production Company (MAL Co. Ltd.) in Ajka, western Hungary broke and more than 800,000 m<sup>3</sup> of toxic (highly alkaline, pH = 13) red mud slurry flooded the environment (Szépvölgyi, 2011) covering 1017 ha

of agricultural land (Uzinger et al., 2015). Red mud is a by-product derived from the treatment of bauxite with concentrated NaOH under elevated temperature and pressure (Gräfe and Klauber, 2011). At the time of the Ajka spill the highly caustic (pH = 13) red mud suspension engulfed the downstream villages of Kolontár, Devecser and Somlóvásárhely in Western Hungary and contaminated the Torna Creek and the

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Marcal and Rába rivers, prior to the outlet of the Rába system to the Danube (Gruiz et al., 2013; Mayes et al., 2011). The immediate emergency management measures focused on the removal of red mud from residential areas where the average thickness of the red mud layer on soil surfaces was 5–10 cm (min. 3 cm; max. 45 cm) (Anton et al., 2012). The removal of the red mud layer from the soil surfaces in inhabited areas begun after the spill, but in the agricultural areas the red mud had covered the soil for more than 3 months before removal (Uzinger et al., 2015). The removed red mud and soil mixture (RMSM) (estimated 530,000 m<sup>3</sup>) was collected and disposed of in the dams at MAL Co. Ltd.

In recent years, extensive work has been done by researchers to develop various economically favorable ways for the utilization of red mud (Sutar et al., 2014). Many applications have been investigated including its re-use as stabilization material (Kalkan, 2006), as adsorbent (Pradhan et al., 1999), for the recovery of trace metals (Kumar et al., 2006) and production of building material (Kalkan, 2006; Thakur and Sant, 1983). Some attempts have been made to use red mud for soil improvement. It has been used in agriculture to increase the phosphorus retention of sandy soil (Summers et al., 1993; Summers and Pech, 1997) and to increase the low pH (Summers et al., 2001; Snars et al., 2004). Due to the combined presence of ferric, aluminum, and tectosilicate like compounds in red mud, it is capable of immobilizing toxic metals from polluted soils (Gadepalle et al., 2007) or removing toxic metals from wastewaters (Castaldi et al., 2010a, 2010b; Garau et al., 2011; Santona et al., 2006) or to reduce the leaching of soil nutrients (Phillips, 1998).

Covers placed over landfills are typically multicomponent systems constructed directly on top of the waste shortly after a specific unit has been filled to capacity. Nowadays, many operators are looking towards alternative materials to use as landfill cover instead of topsoil. In an effort to recycle and reclaim industrial and municipal wastes, wastes generated by agriculture, livestock farming, forestry are being successfully used as landfill cover constituent for plant production (Abad et al., 2001; Benito et al., 2005; Grigatti et al., 2007). Several studies have investigated different inorganic materials, as partial substitutes for soil, like sewage sludge (Ingelmo et al., 1997), drinking water treatment residuals (Dayton and Basta, 2001) and incinerator bottom ash (Rivard-Lentz et al., 1997). Application of RMSM as additive to the surface layer of a landfill cover system may contribute to the reduction of the stored red mud inventory.

Since the disposal of the removed RMSM at Ajka involves considerable costs and also potential risks, the aim of our research was to develop a technology for the re-utilization of the stored RMSM. So far other studies have not dealt with the re-use of RMSM. The overall objective of the present study was to characterize and evaluate the applicability of the RMSM as additive to the surface layer of the landfill cover system at the municipal solid waste deposit in Gyál (A.S.A. Hungary). Ujaczki et al. (2015) in previous microcosm studies on the potential utilization of the Ajka red mud (main components: Fe, Ti and Al oxides and hydroxides, pH = 10–12) as soil ameliorant found that the Ajka red mud may be mixed into soil at up to 5% w/w without any mid-term adverse effects on the natural habitat of the soil. Based on this finding, we assumed that the RMSM might be applicable also as landfill surface cover.

To select the best RMSM and low quality subsoil (LQS, originally used as surface layer of an interim landfill cover) combination the LQS + RMSM mixtures were applied at various ratios in a two-step field study. In the first step a leaching experiment was performed in lysimeters. The toxicity of the leachate and the average leaching of metals from the LQS + RMSM lysimeters were monitored and compared. In the second step we applied RMSM in field plots. The experiments have been monitored for over 10 months by an integrated methodology combining physical, chemical, biological and ecotoxicological methods.

## 1. Materials and methods

### 1.1. Materials

The landfill surface cover system in the field study was made up of two components. The first component was a borrow LQS material excavated from the underground construction of the metro line No. 4 in Budapest, hereinafter referred to as LQS, originally used as surface layer of the interim landfill cover system in the study area. This LQS had clay loam texture (USDA Textural Soil Classification), contained 17.1% CaCO<sub>3</sub> and had a pH<sub>(H<sub>2</sub>O)</sub> of 7.99. The second component of the landfill surface cover system was RMSM obtained from the temporary storage facility at Ajka 2 years (2012) after the red mud spill. The red mud contained in the RMSM mixture is carbonated (i.e., neutralized with atmospheric contact), for this reasons it has pH<sub>(H<sub>2</sub>O)</sub> 10.2. This explains why the pH<sub>(H<sub>2</sub>O)</sub> of the RMSM is only 8.40. The metal concentrations of subsoil (LQS) and RMSM were analyzed on site with a portable X-ray fluorescence (XRF) (Niton® XL3t 600, Thermo Scientific, USA) analyzer before mixing of the amendments (Table 1). The Co and Ni levels in the subsoil (LQS) were above the Hungarian limit value for soil (Hungarian 6/2009 (IV.14.) KvVM-EüM-FVM decree). The As, Co and Ni concentrations in the RMSM were above the Hungarian limit value for soil (Hungarian 6/2009 (IV.14.) KvVM-EüM-FVM decree).

**Table 1 – Metal composition (XRF) of low quality subsoil (LQS) and red mud and soil mixture (RMSM) on site before mixing of the amendments.**

RMSM amount in LQS/elements	LQS	RMSM	HLV <sup>a</sup>
As (mg/kg)	8.59	19	15
Ca (g/kg)	52.8	62.8	–
Co (mg/kg)	115	181	30
Cu (mg/kg)	24.8	30	75
Fe (g/kg)	21.3	33.2	–
K (g/kg)	14.4	10.6	–
Mo (mg/kg)	2.42	2.44	7
Ni (mg/kg)	93.7	98.4	40
Ti (g/kg)	3.02	27.4	–
Zn (mg/kg)	64	57.7	200
CaCO <sub>3</sub> (w/w %)	17.1	13.7	
pH	7.99	8.4	

<sup>a</sup> Hungarian limit value for soil based on KvVM-EüM-FVM Joint Decree No. 6/2009.

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