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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.

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^3 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^3) 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% $CaCO_3$ and had a $pH_{(H2O)}$ 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 $pH_{(H2O)}$ 10.2. This explains why the $pH_{(H2O)}$ 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[a]
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
$CaCO_3$ (*w/w* %)	17.1	13.7	
pH	7.99	8.4	

[a] Hungarian limit value for soil based on KvVM-EüM-FVM Joint Decree No. 6/2009.

1.2. Experimental set-up

The experiments for the potential utilization of RMSM were performed at the A.S.A. Hungary municipal landfill site at Gyál (47° 21′ 55.9476″ N, 19° 14′ 25.4868″ E).

1.2.1. Lysimeter study

The leachates of the LQS + RMSM complex were studied in lysimeters. As a first step 0.86 m^3 lysimeters were installed near the landfill. Special plastic barrels functioned as lysimeters. Four lysimeter setups were constructed: one containing only subsoil (LQS), one only RMSM and two lysimeters containing the RMSM and subsoil (LQS) mixtures (LQS + 20% RMSM and LQS + 40% RMSM). The lysimeters had a 5 cm thick gravel layer at the bottom as filter layer, followed by geo foil overlain by the 80 cm thick LQS + RMSM mixtures. Each lysimeter was irrigated with 9 L of water (from a well) three times a week, except for rainy days. During the field experiment the precipitation was mostly near average (NOAA, 2015) characterized by 699 mm total annual rainfall based on data from the Hungarian Meteorological Service (2015).

1.2.2. Field plot study

As a second step, 10 m^2 plots of land were isolated on a flat part of the landfill. The plots were randomly arranged to decrease the effect of inhomogeneity. The subsoil (LQS) was mixed with 5%, 10%, 20% and 50% RMSM by weight at 0.2 m depth. One plot contained only subsoil (LQS) and the other one only RMSM. The plots were irrigated during dry days for 1 hr/day.

1.3. Leachate and soil analysis

1.3.1. Lysimeter study

The mobility of toxic elements in the amended soil and the toxicity of the leachate from lysimeters were monitored during 10 months. Lysimeter effluent samples were collected into plastic bottles without any filtration on four occasions (after one, two, four and seven months) and the samples were stored in the fridge at 4°C prior to analysis. The metal content of the leachates was determined according to the HS EN ISO 17294-2 (2005). The pH of the leachates was measured according to the Hungarian Standard 21470/2–81 (1981). The environmental toxicity analysis of the leachate water was measured by *Aliivibrio fischeri* (ISO 11348-3, 2007) and *Sinapis alba* based on (Hungarian Standard 22902–4, 1991) test protocol.

1.3.2. Field plot study

The field plots were monitored during 10 months by an integrated methodology, combining physical–chemical–analytical methods with biological methods and ecotoxicity testing (Gruiz et al., 2005, 2009). The amount of precipitation and the temperature were recorded on a daily basis. In the field plot study the total metal content of the solid samples was determined after aqua regia digestion by ICP-AES according to the Hungarian Standard 21470-50 (2006) and the mobile metal content was determined after distilled water extraction according to the Hungarian Standard 21978-9 (1988). The pH of the solid samples was measured according to the Hungarian Standard 21470/2-81 (1981). Water holding capacity (WHC) was measured as described by Öhlinger (1995). The BIOLOG EcoPlate method was used to study the substrate utilization patterns of the treated soils' microbial community according to Nagy et al. (2013). The environmental toxicity analysis of the soil was measured by *A. fischeri* bioluminescence inhibition test based on the protocol described by Leitgib et al. (2007), *S. alba* and *Triticum aestivum* root and shoot growth inhibition test based on the Hungarian Standard 21976-17 (1993) modified to direct contact with soil as described by Leitgib et al. (2007) and *Folsomia candida* mortality test applying direct contact with soil as an international standardized method (ISO/TC 190SC4 WG2; Wiles and Krogh, 1998).

1.4. Statistical analysis

To determine whether the added RMSM amounts had any significant effect on the metal concentration, water holding capacity and microbial activity of the landfill cover, we performed analysis of variance (ANOVA) using StatSoft® Statistica 11. We established the level of significance at $p < 0.05$. We used the Fisher's least significant difference test to compare the effects of various RMSM amounts.

2. Results and discussion

2.1. Lysimeter study

To assess the effect of the LQS + RMSM landfill surface cover on the subsurface waters the pH and the toxic metal content of the leachate from lysimeters were analyzed. The average results of the 10 months monitoring are presented in Table 2.

The pH of the leachate from the mixtures did not differ significantly from subsoil (LQS) and RMSM. The metal content of the leachates was below the Hungarian limit value for subsurface water (Hungarian 6/2009 (IV.14.) KvVM-EüM-FVM decree) except for the following seven elements: B, Mo, Na, Ni, Se, and Zn shown in Table 2. Since the subsoil (LQS) has already had high B, Mo, Na, Ni, Se, and Zn content without being mixed with RMSM, the addition of the RMSM did not influence significantly the metal content of the leachates, except for B and Mo. The As content of the subsoil (LQS) did not exceeded the Hungarian limit value but it was significantly higher in the subsoil (LQS) than in the RMSM or in the different mixtures. In our experiment the Cr was below the detection limit (0.5 μg/L) in both the subsoil (LQS) and the RMSM in contrast to other studies (Rékási et al., 2013; Burke et al., 2012). Anton et al. (2012) modeled the effects of the Hungarian red mud disaster in a soil column experiment focusing on element solubility concluding that the red mud affected the total Mo and Na concentrations of the topsoil layer due to leaching of the red mud particles. Rékási et al. (2013) investigated the conditions after the Hungarian red mud disaster in a soil column experiment, which showed that leaching from the red mud layer increased the total, plant-available, exchangeable and water-soluble fractions of Na, Mo, Cu and Cr, and resulted in a rise of the pH and DOC (dissolved organic carbon) concentration. Therefore, the elevated Mo content of the LQS + RMSM landfill surface cover was attributed to the RMSM although the subsoil (LQS) has already had high Mo content. Mo in soils is associated with wet conditions, alkaline reactions and high concentrations of organic matter (Fleming, 1980; Gupta and Lipsett, 1981). In terms of Mo,

Table 2 – Observed element concentrations in leachates from the lysimeters (average of 4 samplings).

RMSM amount in LQS/Elements	LQS	LQS + 20% RMSM	LQS + 40% RMSM	RMSM	HLV*
As (μg/L)	2.15 a	1.03 b	1.75 c	1.90 c	10
B (μg/L)	1610 a	2300 b	2050 c	1810 d	**
Mo (μg/L)	29.6 a	22.7 b	31.3 a	23.3 b	20
Na (mg/L)	1170 a	1223 a	1287 a	1203 a	–
Ni (μg/L)	21.3 a	20.3 a	27.0 a	26.1 a	20
Se (μg/L)	128 a	13.0 b	84.0 a	129 a	10
Zn (μg/L)	2260 a	2130 a	2130 a	94.8 b	200
pH	7.71 a	7.62 a	7.82 a	7.65 a	

Values followed by the same letters, indicate no significant differences at the level of $p < 0.05$, Decree No. 6/2009.

* Hungarian limit value for subsurface water based on KvVM-EüM-FVM Joint.

** Essential to plant growth, but even tolerant plants may be damaged when boron exceeds 2.000 μg/L (US EPA, 1986).

there is a potential risk of metal transfer through soil down to ground water, aquifer or via plant – root uptake (bio available). When the leachate is flowing through the soil, the heavy metals in leachate may be adsorbed or complexed to the soil particles (McBride et al., 1997; Sewwandi et al., 2011). On the other hand, usually in a landfill, the leachates are collected by a leachate collection system, thus the risk of groundwater contamination by any leachate is decreased.

Addition of RMSM did not change the toxicity of the LQS + RMSM leachates as compared to subsoil (LQS) (Table 3). Rékási et al. (2013) found that the leachate from the column experiment with red mud contributed to a rise in the *S. alba* root and shoot growth in an ecotoxicity test. Based on the above findings the RMSM material may be applied as landfill surface cover at landfill sites, especially when considering that most modern landfills' key component is a leachate collection system to avoid seepage of the leachate into the underlying soil.

2.2. Field study

Table 4 shows the effect of RMSM on the pH, the total (aqua regia extract) and water soluble metal contents of the subsoil (LQS) in the treated plots during the 10-month experiment. The 5%, 10% and 20% RMSM increased the slightly alkaline pH (7.99) of the subsoil (LQS) by 0.1 unit; 50% RMSM increased pH by 0.2 unit, but the effect was not significant. This negligible increase differs from the results of previous studies applying more alkaline red mud directly into slightly acidic (pH 5.5 and 6.4) soil. Lombi et al. (2002) and Ujaczki et al. (2015) described an increase of the pH by 1.4 and 1.7 units after the addition of 2% and 5% red mud to soil. The RMSM addition resulted in a much slighter pH increase than the red mud in the above studies, because the RMSM had a pH of only 8.4 compared to pH 10.2 of the carbonated red mud covering the agricultural soil. In addition, at the time of the spill the pH of the red mud suspension was very caustic (pH 13) (Szépvölgyi, 2010). The carbonation of the Ajka red mud occurred as a result of atmospheric contact (Lockwood et al., 2014). According to previous studies an important issue is the greater mobility of oxyanionic trace elements such as As, Cr, Mo, and V at elevated pH (Langmuir, 1997; Klebercz et al., 2012).

The total metal content of the subsoil (LQS) was below the Hungarian limit value for soil (Hungarian 6/2009 (IV.14.) KvVM-EüM-FVM decree) as shown in Table 4. The metal content of RMSM was also below the Hungarian limit value for soil except for two metalloids: As and Ni. The total concentration of As was 30% higher than the limit value (15 mg/kg), however the As was insoluble in water (Table 4). The concentration of Ni was 9% higher than the limit value, which is negligible, but there was 5.84 mg/kg water soluble Ni in the RMSM. Nevertheless the Na content was three times higher in the aqua regia extract than the risk based site specific screening value at Ajka based on the soil properties of the area (900 mg/kg) (Kádár, 2009; Gruiz et al., 2013), which indicates increased sodification potential, but might be too strict limit in case of a landfill site. Based on previous microcosm experiments conducted by Ujaczki et al. (2015) the As, Ni and Na concentration of RMSM was higher than the limit value (15 mg/kg, 40 mg/kg, 900 mg/kg, respectively) when the amount of red mud was 10% and more in the soil. Other authors also observed an increase of As, Ni, and Na in the aqua regia extract when mixing red mud with soil (Gruiz et al., 2013; Rékási et al., 2013; Ruyters et al., 2011). According to Ujaczki et al. (2015) the soils became slightly saline at 5%, 10% and 20% red mud dose, therefore the growth of only some very salt-sensitive crops would be inhibited. The metal analysis of the LQS + RMSM mixtures indicated that the RMSM is applicable as landfill surface cover component at up to 20% RMSM dose because the concentration of the examined toxic metals and metalloids did not exceed the Hungarian limit value for soil.

Table 3 – Summary of environmental toxicity test results of the leachates from the lysimeters (inhibition % calculated compared to leachate from subsoil (LQS)).

		Aliivibrio fischeri bioluminescence inhibition (%)	*Sinapis alba* root growth inhibition (%)	*Sinapis alba* shoot growth inhibition (%)
LQS + 20% RMSM	10th month	<5 (non toxic)	<5	7
LQS + 40% RMSM	10th month	<5 (non toxic)	<5	7
RMSM	10th month	<5 (non toxic)	<5	<5

Table 4 – Total (aqua regia extract) and distilled water-soluble element concentrations in the different subsoil (LQS) and RMSM in field plots (during the experiment) ($n = 3$).

RMSM amount in LQS/elements		LQS	LQS + 5% RMSM	LQS + 10% RMSM	LQS + 20% RMSM	LQS + 50% RMSM	RMSM	HLV *
As (mg/kg)	Total	11.3 a	10.5 a	14.7 b	14.5 b	16.5 b	19.5 c	15
	Water soluble	<DL	<DL	<DL	<DL	<DL	<DL	
B (mg/kg)	Total	31.4 a	20.1 b	20.5 b	18.2 b	20.0 b	9.55 c	–
	Water soluble	1.08 a	0.84 a	1.01 a	0.87 a	0.89 a	0.61 b	
Cr (mg/kg)	Total	45.4 a	44.0 a	47.6 a	47.2 a	56.5 b	69.6 c	75
	Water soluble	<DL a	0.03 a	0.04 a	0.03 a	0.05 a	0.04 a	
Cu (mg/kg)	Total	22.5 ac	19.1 bdf	21.5 ce	19.0 df	20.9 e	18.0 f	75
	Water soluble	0.06 a	0.16 a	0.13 a	0.11 a	0.20 a	0.24 a	
Mo (mg/kg)	Total	0.29 a	0.71 b	0.94 bc	1.19 c	1.92 d	2.31 e	7
	Water soluble	0.04 a	0.07 a	0.09 a	0.13 a	0.13 a	0.24 b	
Na (mg/kg)	Total	509 a	603 a	956 ab	1143 ab	1782 b	3070 c	900 **
	Water soluble	0.14 ab	0.30 b	0.38 bc	0.50 bc	0.54 bc	0.73 bc	
Ni (mg/kg)	Total	32.0 a	33.4 a	35.5 ab	33.9 a	38.1 b	43.5 c	40
	Water soluble	<DL a	<DL a	<DL a	<DL a	5.26 b	5.84 b	
Pb (mg/kg)	Total	18.3 a	18.2 a	23.7 ab	22.7 a	29.1 b	38.5 c	100
	Water soluble	<DL	<DL	<DL	<DL	<DL	<DL	
Se (mg/kg)	Total	0.91 a	1.07 a	<DL b	0.74 a	1.41 c	2.20 d	30
	Water soluble	<DL	<DL	<DL	<DL	<DL	<DL	
Zn (mg/kg)	Total	75.5 a	77.5 a	72.4 a	63.3 ab	66.9 ab	54.9 b	200
	Water soluble	<DL a	<DL a	<DL a	<DL a	0.26 b	0.24 b	
pH		7.99 a	8.11 a	8.14 a	8.13 a	8.22 a	8.40 a	

Values followed by the same letters, indicate no significant differences at the level of $p < 0.05$;
DL (As): 0.08 mg/kg; DL (Cr): 0.02 mg/kg; DL (Ni): 0.04 mg/kg; DL (Se): 0.12 mg/kg; DL (Zn): 0.06 mg/kg.
* Hungarian limit value for soil based on KvVM-EüM-FVM Joint Decree No. 6/2009;
** Risk based site specific screening value at Ajka, indicates increased sodification potential.

Fig. 1 presents the WHC change during the experiment, where the WHC is the total amount of water a soil can hold at field capacity (Öhlinger, 1995). Due to the RMSM content, the planned landfill surface cover is able to store significantly more water (by 5% at 10% RMSM) than the subsoil (LQS) itself, but the availability of this water will be determined by infiltration patterns and rooting depth (Van Gool et al., 2005). These changes in the water holding characteristics could be explained by changes in the porosity and the pore size distribution (Buchanan et al., 2010).

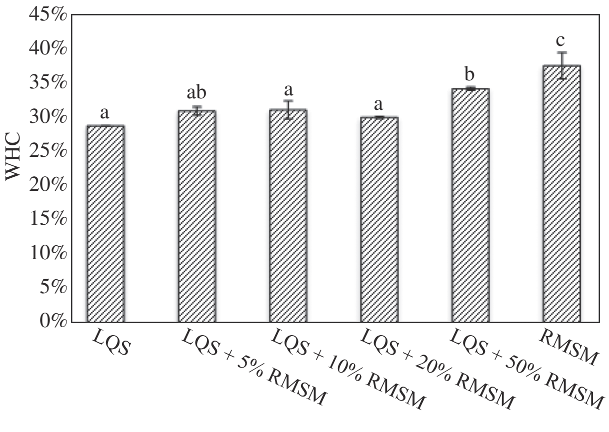

Fig. 1 – Water holding capacity (WHC) in field plots (during the experiment) ($n = 3$), (LQS: low quality subsoil; RMSM: red mud and soil mixture). Values followed by the same letter indicate no significant differences at the level of $p = 0.05$ at each sampling.

BIOLOG EcoPlates were used to study the substrate utilization patterns of the microbial community in the LQS + RMSM mixtures, where the average well color development (AWCD) value was the indicator of the general microbial activity (Garland et al., 1991). At the beginning (1st month) the 5%–20% RMSM containing landfill surface cover had the lowest AWCD values, but by the end of the experiment the microbial activity of these mixtures indicated the highest values (Fig. 2). The AWCD values of 5%, 10% and 20% RMSM mixed with subsoil (LQS) increased largely during the monitored time period,

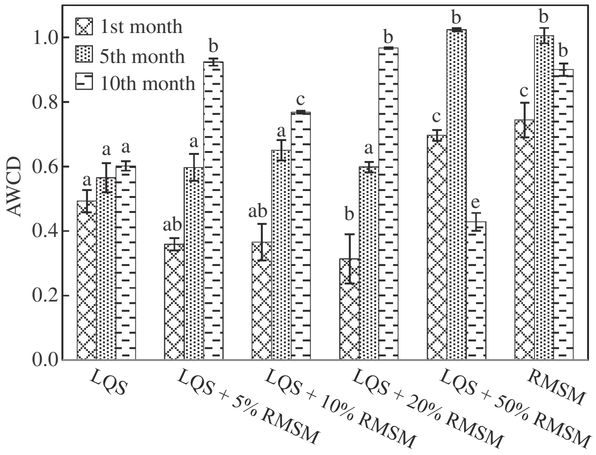

Fig. 2 – Average well color development (AWCD) at 72 hr in the samples from field plots. Values followed by the same letter indicate no significant differences at the level of $p = 0.05$ at each sampling.

reaching a higher microbial activity than the subsoil (LQS) itself. The results for the RMSM demonstrate clearly the existence of an active microflora in the red mud–soil mixture as indicated by the highest AWCD values even after one month. Based on the AWCD values in the 10th month we do not recommend mixing more than 20% RMSM into the subsoil (LQS).

Our previous experiments with red mud mixed into agricultural soil showed that the number of aerobic heterotrophic living cells did not increase at 5% red mud dose compared to the untreated soil (Ujaczki et al., 2015). Castaldi et al. (2009) found positive effect on microbial cell numbers when an acidic soil was treated with red mud. Lombi et al. (2002) also observed an increase in the microbial biomass volume in metal contaminated soil after red mud treatment. Garau et al. (2007) detected that the red mud caused a significant ($p < 0.05$) increase of fast-growing heterotrophic bacteria, but the addition of red mud did not change the amount of heterotrophic bacteria.

The effects of the RMSM mixed into subsoil (LQS) on testorganisms were measured and compared to the subsoil (LQS) as control. Table 5 summarizes the toxicity results of the bioassays. The various bioassays showed different sensitivity to the mixed RMSM due to their sensibility to the contaminants (Leitgib et al., 2007). Direct contact tests with microbial testorganism (*A. fischeri*) and plant testorganisms (*S. alba*, *T. aestivum*) were the most sensitive to all mixtures (Table 5).

Based on the $\sum$Cu20 values (Cu-equivalent values) of the *A. fischeri* bioluminescence inhibition the LQS + 20% RMSM mixture was the most toxic of all, but still in the slightly toxic category. However, the inhibiting effect of the RMSM shows a decreasing trend towards the non-toxic category during the 10 months of the experiment, which is in accordance with the results of previous microcosm experiments when different red mud doses were mixed with soil (Ujaczki et al., 2015). Contrary to these results, Klebercz et al. (2012) reported that the red mud contaminated sediment after the accident in Ajka was very toxic to *A. fischeri* according to the bioluminescence inhibition test. The toxic effect suggested that other background sources of pollution in the catchment may have contributed to the response (Klebercz et al., 2012). The highest inhibition percentage compared to the subsoil (LQS) was observed in case of the *S. alba* root growth in plots containing only RMSM. In this case the initial 34% inhibition decreased to 15% nine months later. The root and shoot growth inhibition in case of *T. aestivum* after 10 months was 37% and 39%, respectively in the above plots. The toxic effect of RMSM might originate from its heavy metal content (Table 4). Ujaczki et al. (2015) found that 5% red mud application directly to soil did not have any significant harmful effect on the testorganisms, a fact confirmed also by Lombi et al. (2002), who reported positive effect of red mud on crops. Klebercz et al. (2012) also found that the inhibition of *S. alba* shoot and root growth by the contaminated sediment was relatively low and there were significant positive correlations between root inhibition percentage and exchangeable trace element concentrations. None of the RMSM and subsoil (LQS) mixtures showed inhibition in case of the *F. candida* (Collembola) compared to the subsoil (LQS) in contrast to previous microcosm experiments, where the high red mud concentration (>30%) in soil was toxic to the test animals (Ujaczki et al., 2015). This finding was similar to Winkler (2013) who analyzed the Collembolan community structure and species abundance at the red mud polluted areas in Western Hungary and observed no adverse effect in the Collembola abundance.

This field study used a complex methodology for monitoring the potential effects of RMSM as additive to the landfill surface cover (LQS) and found conformity between the results of the various test methods. Both the analytical, biological and the toxicity measurements showed that up to 20% RMSM mixed into the low quality subsoil (LQS) had no adverse effects on the soil ecosystem.

3. Conclusion

The effect of different red mud-soil mixture (RMSM) doses to the borrow soil material (LQS) to produce a landfill surface cover at a landfill site in Hungary was modeled in a 10 month long lysimeter and field plot study. RMSM is a soil and red mud mixture originated from the red mud disaster affected area at Ajka (Hungary) in 2010. The aim of the study was to assess the RMSM dose at which the natural habitat of the landfill surface cover is not adversely affected. Mixing 20% *w/w*

Table 5 – Summary of environmental toxicity test results for the RMSM and subsoil (LQS) mixtures from the field plots (inhibition % calculated compared to subsoil (LQS)).

		Aliivibrio fischeri inhibition in Cu equivalent	*Sinapis alba* root growth inhibition	*Sinapis alba* shoot growth inhibition	*Triticum aestivum* root growth inhibition	*Triticum aestivum* shoot growth inhibition	*Folsomia candida* mortality
		ED20 (μg Cu/g soil)	Inhibition (%)	Inhibition (%)	Inhibition (%)	Inhibition (%)	Inhibition (%)
LQS + 5% RMSM	1st month	<5 (non toxic)	<5	<5	<5	<5	<5
	10th month	1 (non toxic)	11	13	7	15	<5
LQS + 10%RMSM	1st month	<5 (non toxic)	32	14	6	<5	<5
	10th month	9 (non toxic)	11	<5	<5	<5	<5
LQS + 20% RMSM	1st month	204 (slightly toxic)	21	<5	12	12	<5
	10th month	5 (non toxic)	18	<5	<5	<5	<5
LQS + 50% RMSM	1st month	81 (slightly toxic)	3	1	<5	<5	<5
	10th month	10 (non toxic)	7	6	2	<5	<5
RMSM	1st month	108 (slightly toxic)	34	22	8	<5	<5
	10th month	6 (non toxic)	15	11	37	39	<5

RMSM into subsoil (LQS) to create a landfill surface cover caused an increase in the total amount of Na, and in the water soluble fractions of Na. In the leaching experiments the Mo, Na, Ni, Zn, B and Se concentrations exceeded the Hungarian limit value for subsurface water, however due to the leachate collection system at landfill sites its harmful effect may be limited. RMSM addition did not change the pH during the experiment. The result of the substrate utilization test demonstrated that there is an active microflora in the RMSM, as indicated by the AWCD values which increased during the monitored time period at 5%, 10% and 20% RMSM mixing. The ecotoxicity test results of leachate and soil showed that 20% *w/w* RMSM had no adverse effect on the following testorganisms: *S. alba*, *T. aestivum*, *A. fischeri* and *F. candida* during the experiments (10 months). Assessing all the environmental toxicity test results we concluded that 20% RMSM application did not have any harmful effect on the subsoil (LQS). Based on this study we may recommend the use of the removed RMSM as additive to the surface layer of landfill cover systems at landfill sites.

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