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# Leaching of copper and nickel in soil-water systems contaminated by

# bauxite residue (red mud) from Ajka, Hungary: The importance of organic

### matter

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This section consists of 7 pages and 2 Tables and 4 Figures

**Table A. Full XRF analysis of red mud and soils** Concentrations of selected elements present in the red mud sample and soil samples prior to amendment (Lehoux et al. 2013)

Major Elements	Red Mud	Organic-rich	Wetland soil	Sandy soil
(Weight %Oxides)		soil		
SiO <sub>2</sub>	12.9	72.9	80.6	89.8
TiO <sub>2</sub>	5.24	0.48	0.33	0.37
$Al_2O_3$	16.0	8.9	6.4	4.2
$Fe_2O_3$	38.5	3.0	1.8	1.7
MnO	0.31	0.04	0.03	0.06
MgO	0.61	0.84	0.98	0.06
CaO	8.0	1.0	2.27	0.60
Na <sub>2</sub> O	8.0	1.0	0.90	0.71
K <sub>2</sub> O	0.10	1.6	1.3	0.87
$P_2O_5$	0.19	0.11	0.07	0.08
$SO_3$	0.36	0.02	0.02	0.006
LOI	8.0	10.2	5.1	1.8
Minor Elements				
$(mg kg^{-1})$				
As	196	11	8	2
Ba	66	396	267	148
Ce	607	47	34	17
Co	59	11	5	3
Cr	864	68	62	50
Cu	104	12	6	2
Ga	26	10	6	4
La	283	26	18	10
Мо	15	1	1	1
Ni	361	23	14	5
Pb	215	25	12	9
Sb	22	1	2	1
Sr	318	78	94	47
Th	98	6	4	2
U	21	3	2	1
V	1132	72	51	30
W	17	<1	<1	<1
Zn	162	52	26	21
Zr	1223	122	102	88

### Table B. Sequential extraction data for Cu and Ni.

Amount of Cu and Ni Leached from triplicate Ajka red mud samples collected from location K1 in Mayes et al. (2011) during sequential extractions (Rauret et al. 1989). Values in parenthesis are percentages of total Cu or Ni leached.

Leaching Step	Cu (mg kg <sup>-1</sup> )	Ni (mg kg <sup>-1</sup> )
1 mol $L^{-1}$ Mg(Cl) <sub>2</sub> at pH 7	$0.1 \pm 0.0 (0.2 \pm 0.0\%)$	0.1 ±0.0 (0.2 ±0.0%)
1 mol L <sup>-1</sup> NaOAc / HOAc at pH 5	24.9 ±1.8 (38 ±3%)	2.7 ±0.1 (0.7 ±0.0%)
$0.5 \text{ mol } \text{L}^{-1} \text{ NH}_2 \text{OH.HCl} / \text{HCl} \text{ at pH } 1.5$	18.5 ±1.0 (28 ±2%)	54.4 ±3.2 (14 ±1%)
$30\% H_2O_2 / HNO_3$ at pH 2	8.8 ±3.0 (13 ±5%)	43.3 ±0.6 (11 ±0.2%)
Concentrated HF / HCl / HNO <sub>3</sub>	13.4 ±1.1 (20 ±2%)	280.7 ±2.7 (74 ±0.7%)

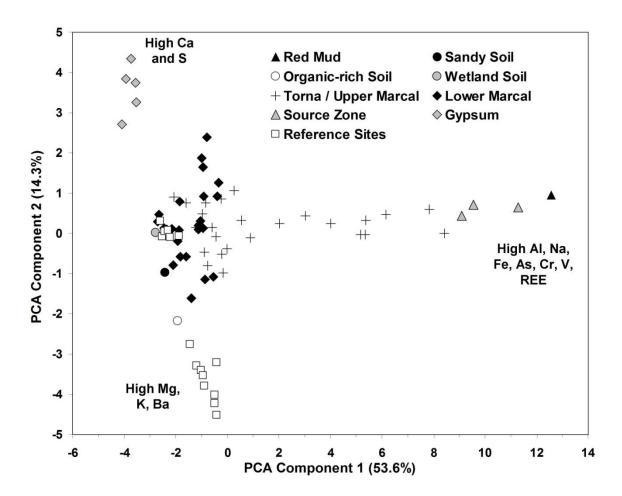
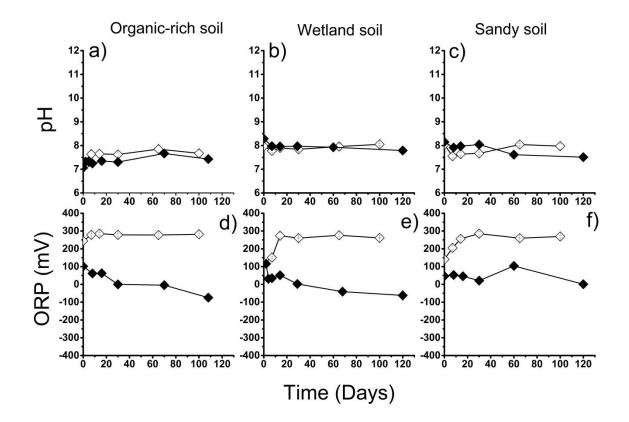
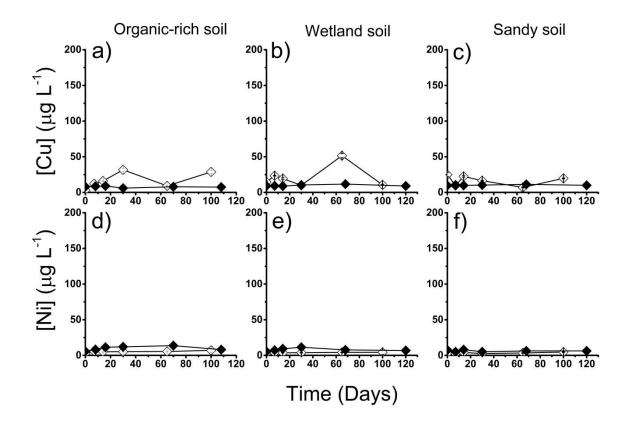


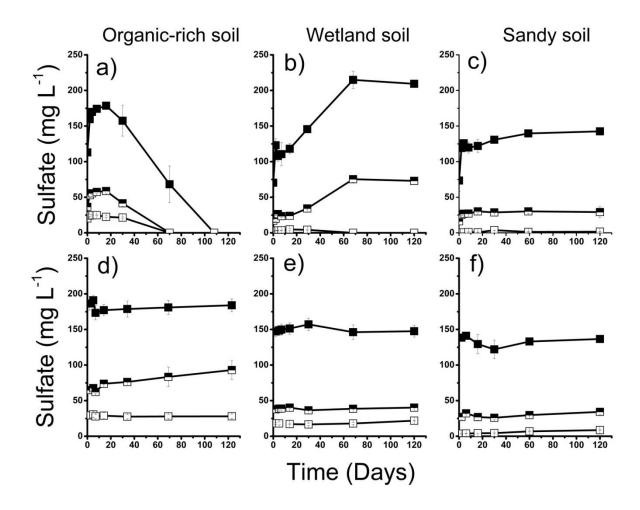
Figure A. Taken from Lehoux et al. (2013) - Principle Component Analysis - shows the samples used in this study in the broader context of large scale soil and sediment sampling efforts across a 100km reach of the Torna-Marcal-Raba River system in the immediate aftermath of the spill (Mayes et al. 2011). Each point represents a single sample station and three end members are apparent in the PCA. Samples of red mud from the Ajka repository plot at the extreme right hand side of the figure, being characterised by enriched Fe, Al, Ti, Na, Cr, V and various other trace elements. The red mud samples used in this study (labelled 'Red Mud' in the legend) plot alongside those red mud taken from previous sampling efforts at the same site (labelled 'Source', sample K1 from Mayes et al. (2011). To the upper left of Figure A, samples enriched with Ca and S are indicative of river sediments in reaches subject to extensive gypsum dosing (to neutralise the high pH) shortly after the spill. To the lower left, unaffected reference sites are apparent as a final end member and are relatively enriched in K, Ba and Mg, consistent with bedrock lithology (Mayes et al. 2011). The samples plotting between these end members represent mixing of these different extremes, for example the samples that plot in a roughly straight horizontal line from the red mud to the left hand side of Figure A represent dilution of red mud through mixing with soils and sediments with distance downstream from the source area. The soil samples used in this study all plot in a group on the left hand side with unaffected sites from the lower Marcal River and reference samples showing them to be representative of the broad geochemical constituent of unaffected sediments in the Marcal catchment. The pH of the waters in this area are neutral (pH 7 - 8) as this area is dominated by dolomite and limestone bedrock (Mayes et al. 2011).



**Figure B** – pH and ORP (as an indicator for Eh) in unamended control experiments over time. Full symbols = anaerobic experiments, empty symbols = aerobic experiments. Error bars are 1  $\sigma$  of triplicate results (where not shown, errors are within the symbol size).



**Figure C** – Evolution of aqueous Cu and Ni over time in unamended control experiments. Full symbols = anaerobic, empty symbols = aerobic. Error bars are 1  $\sigma$  of triplicate results (where not shown, errors are within the symbol size).



**Figure D** – Evolution of sulfate over time in anaerobic red mud amended soil experiments (a-c), and the associated controls (d-f). Full symbols = 33% RM, half squares = 9% RM and empty squares = no RM. Error bars are 1  $\sigma$  of triplicate results (where not shown, errors are within the symbol size). Sulfate was not recorded for aerobic experiments as sulfate reducing conditions were not anticipated. See Lockwood et al. (2014).

References

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