Assessing the Impact of Sewage Sludge on a Formally Remediated Tailings Impoundment in Relation to the Transport of Metals in Groundwater

Peter Nason, Lena Alakangas, Björn Öhlander

Luleå University of Technology, Department of Chemical Engineering and Geosciences, SE-97187, Luleå, Sweden, peter.nason@ltu.se

Abstract In 2009 sewage sludge was applied to an already successfully remediated saturated and dry covered sulphide tailings impoundment in northern Sweden as a vegetation substrate. The aim of this study was to identify the dispersion, magnitude and duration of sludge-borne constituents released. Readily leachable sludge-borne constituents (DOC, Fe, Co, Cr, Ni, Pb and Zn) created a contamination plume which affected the impoundment groundwater within the first year of application. The majority of the plume originated from the water-saturated area of the impoundment that was only covered with a 10% surface area of sludge. The dry-composite covered area of the impoundment, which contained a 70% sludge cover, did not contribute to the contamination plume due to the efficiency of the cover at reducing water infiltration and contamination dispersal. The constituents were transported along the dominant groundwater flow direction, underneath the dry covered area of the impoundment, to the effluent of the impoundment groundwater system due to inflowing uncontaminated groundwater from an adjacent till slope. All metal and DOC concentrations in the impoundment groundwater returned to pre-sludge application concentrations within two years, due to successful plant establishment on the sludge.

Key Words Sewage Sludge, Sulphidic Tailings, DOC, Groundwater

Introduction

Preventative remedial measures against acid rock drainage (ARD) formation in tailings repositories have been widely studied globally (Lottermoser, 2003). Sewage sludge, which is the solid material generated during the treatment of domestic waste-water, is often utilised as a final reclamation cover above pre-existing remedial systems, and may act as a suitable vegetation substrate. Approximately 210 000 tonnes of sludge are produced annually in Sweden and it has been widely used as a vegetation substrate on the surface of mine wastes (Forsberg, 2008). However, sewage sludge contains high concentrations of readily leachable trace elements (Eriksson, 2001) that may migrate and accumulate into underlying soil profiles or contaminate groundwater systems (Ahlberg *et al.*, 2006). It contains elevated concentrations of organic matter which in its dissolved form may enhance metal mobility, forming organo-metallic complexes in solution (Garcia-Gil *et al.*, 2007). The aim of this study was to identify the dispersion controls, magnitude and duration of sludge-borne metals released from a surface application of sewage sludge on a successfully remediated sulphidic-tailings impoundment.

Site Description

The study site is located in the western part of the Skellefte ore district in northern Sweden at the Kristineberg Zn-Cu mine operated by New Boliden AB. The impoundment tailings storage facility comprises tailings deposited from 1946 to 1996, and has an area of 0.11 km^2 with a tailings thickness varying from 2 to 10m deep (Alakangas, 2006) (Figure 1). The impoundment was successfully remediated in 1996—97 with a combination of water-saturated and dry-composite cover systems (Figure 1). Groundwater wells were installed on the impoundment with the groundwater geochemistry being thoroughly investigated from 1998 to 2006 (Alakangas, 2006; Alakangas and Nason, 2010; Carlsson, 2002). From July-August 2009, the impoundment was subject to further remediation by using sewage sludge as a final vegetation substrate. Approximately 12 000 tonnes of sludge were applied to a depth of 0.2m on 70% of the dry cover and 10% of the water-saturated remediated areas of the impoundment (Figure 1). The sludge was seeded with grass in summer 2010 and plant establishment began in 2011. The geochemical composition of the original sludge material (Table 1) shows elevated concentrations of organic matter (LOI), Ca, P_2O_5, Fe_2O_3, Ba, Co and Ni, when compared to the unoxidised and oxidized tailings compositions derived in 2001 (Holmström *et al.*, 2001).

Impoundment 1 Groundwater Hydrology

The groundwater within the impoundment is highly dependant upon hydrological doming caused by inflowing uncontaminated groundwater from the western till slope, producing flow gradients from the western till slope to the eastern end of the impoundment (Figure 2) (Corrège *et al.*, 2001). The resultant hydraulic head in the west

***Table 1** Average Composition of unoxidised and oxidised tailings from Impoundment 1 in 2001 and average composition of sewage sludge applied in 2009. *modified from (Holmström et al., 2001).*

	Unoxidised Tailings* (73 Samples)	Oxidised Tailings* (12 Samples)	Sewage Sludge (3 Samples)
Major Elements (wt. %)			
SiO_2	42.8	63.1	10.5
Al_2O_3	9.35	11.4	3.11
CaO	1.01	1.24	2.62
Fe_2O_3	24.0	8.45	12.38
K_2O	0.81	1.88	0.62
MgO	7.73	6.65	0.53
MnO	0.12	0.11	0.03
Na_2O	0.46	1.46	0.40
P_2O_5	0.07	0.08	6.65
S	14.4	1.81	1.31
LOI	12.4	5.03	53.4
Minor Elements (ppm)			
As	183	36.2	5.56
Ba	281	481	265
Cd	21.5	1.06	0.91
Co	56.4	7.77	9.02
Cr	46.2	60.7	42.5
Cu	956	159	422
Hg	2.42	0.94	0.77
Ni	5.95	4.52	26.7
Pb	463	454	29.5
Zn	8861	559	626
Zr	117	205	48.8

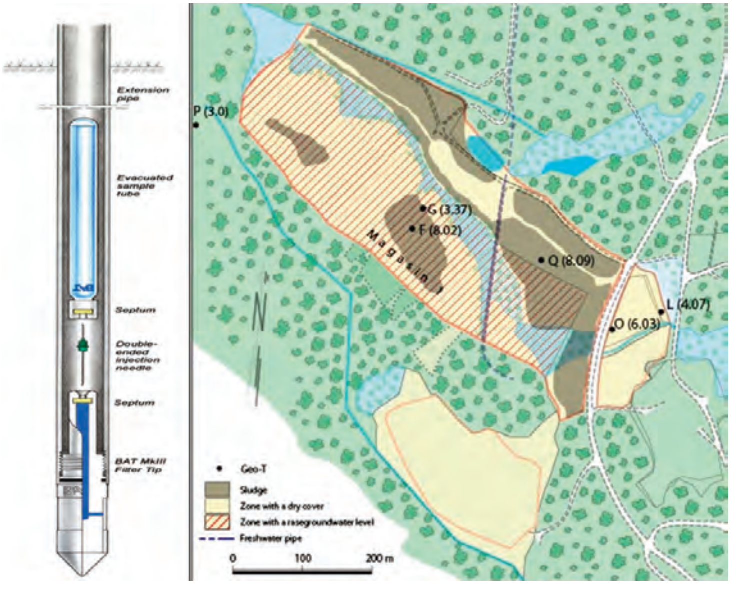

***Figure 1** Left Illustration of the BAT® Groundwater well and Envirosampler®. Right: Map illustrating the location of the BAT® Groundwater wells at Impoundment 1, Kristineberg Mine.*

may cause artesian water to break above groundlevel in the water-saturated cover during snowmelt. This hydrological flow regime controls both flow direction and dilution of the groundwater within the impoundment.

Methodology

Sampling occurred within two intensive monitoring periods in which samples were collected during the summer months of May to September. The first sampling period was initiated after full-scale remediation from 1998 to 2006. The second occurred just prior to, and for two years after a surface application of sewage sludge was deposited in July 2009. The BAT® groundwater wells were designed to contain an impermeable membrane to allow sample collection with minimal exposure to atmospheric oxygen in compliance with the BAT® Envirosampler (Figure 1), allowing sample collection and analysis without contact with atmospheric oxygen. Six wells were sampled in this study (Figure 1). They were chosen because of their relative position in the impoundment groundwater system (Figure 2). Well P represents the background inflowing groundwater from the western till slope. Wells G and F are located in the water saturated remediated part of the impoundment at different depths respectively. Wells Q and O are situated in the dry covered part of the impoundment. Well L represents the groundwater outflow in the east. The BATper®per groundwater tubes were acid-washed with 5% HNO_3 for three days and rinsed with Milli-Qper®per water before sampling. They were purged with argon and vacuumed. Water for major and minor elemental analysis was filtrated in the field using 0.22µm Millipore nitrocellulose membrane filters, acid-washed with 5% acetate acid and with syringes that had been acid-washed using HNO_3, then rinsed with Milli-Qper®per water. The dissolved fraction was transferred into 125ml acid-rinsed bottles. Separate samples were collected unfiltered into non-acid washed bottles for dissolved organic carbon (DOC). Determinations of pH were conducted with a Metrohm® combined pH electrode from 2003 to 2006 and with a Mettler-Toledo MP125 pH electrode from 2009—2011. Determination of dissolved fractions of major (Ca, Fe, K, Mg, Na, S, Si, Al) and minor (As, Ba, Cd, Co, Cr, Cu, Mn, Mo, Ni, P, Pb, Zn) elements in the leachate were performed using ICP-AES (Inductively Coupled Plasma – Atomic Emission Spectrometry) and ICP-SFMS (Inductively Coupled Plasma – Sector Field Mass Spectrometry) respectively. Dissolved organic carbon (DOC) samples in the leachate water were calculated by filtration through 25mm diameter glass micro fibre filters (0.47 mm) mounted in stainless steel filter holders and DOC analysis was performed using a Shimadzu TOC-5000 high-temperature combustion instrument.

Results and Discussion

Water Saturated Groundwater Geochemistry

The geochemistry was identified from the data from wells F and G. DOC peaked in 2010 at a magnitude of approximately 8 mg/L from the pre-sludge background of 3—4 mg/L (Figure 3). The deeper groundwater received peak concentrations of Fe, Ni and Zn immediately after the application in 2009, whereas the shallower groundwater received a lower magnitude concentration pulse in 2010 except for Ni, which was elevated (365µg/L) in 2009 (Figure 4). Due to artesian pressure from the dominant hydraulic regime in the impoundment groundwater system (Corrège *et al.*, 2001) a raised groundwater level above the ground surface occurred during May to June, mobilising sewage sludge constituents directly from the material and contributing to the removal of elevated trace element concentrations in the form of colloidal matter.

Ahlberg (2006) reported that after this period, with the accelerated degradation of organic matter in the sludge, trace metals such as Ni are dominantly released in the form of dissolved organo-metallic complexes. This may explain why peak concentrations of metals in 2009 spiked prior to the peak DOC release in 2010 and that Ni may have continued to persist until 2010 due to possible binding as organic-metallic complexes with peak DOC. However, concentrations of metals and DOC subsided after one year respectively, probably due to dilution from uncontaminated groundwater from the western till slope (Werner *et al.*, 2001), resulting in 2011 metal concentrations that were similar to pre-sludge application conditions (Figure 4).

Dry Covered Groundwater Geochemistry

The geochemistry was identified from the data from wells Q and O, though no samples were retrieved from well O during 2010—2011. Well Q displayed peak concentrations of metals (Fe, Co, Cr,

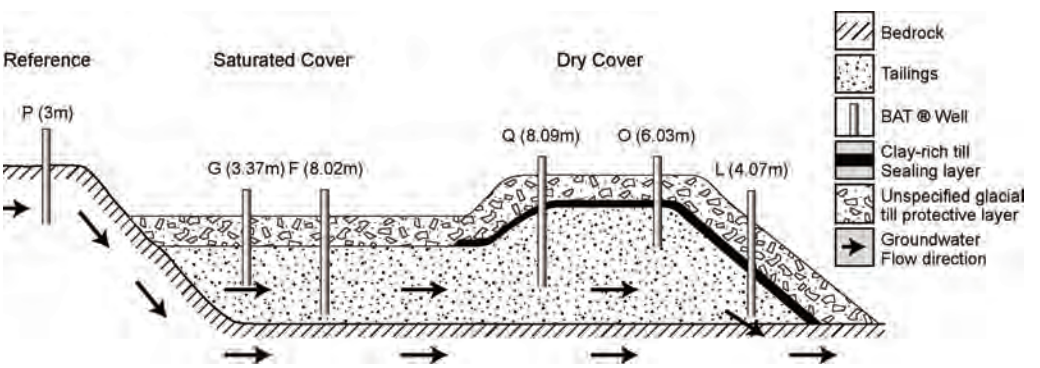

Figure 2 *Schematic cross-section of the placement of the six groundwater wells used in this study in relation to their location in the impoundment cover systems, depth and generalised groundwater flow.*

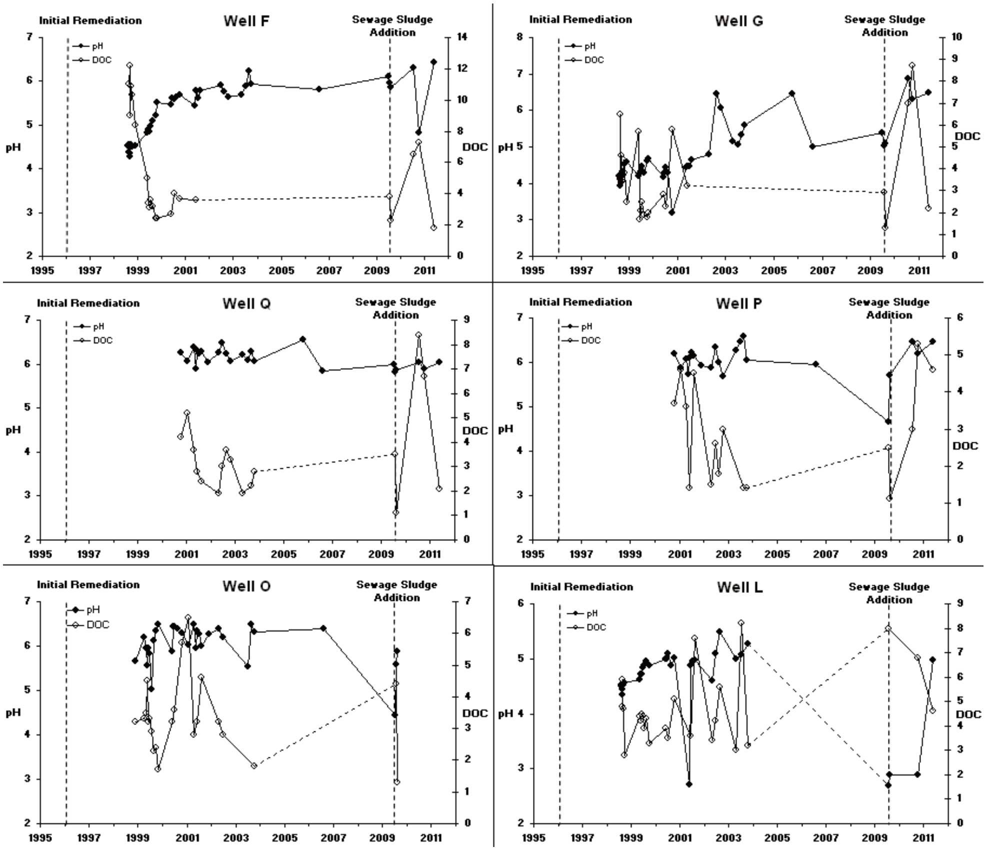

Figure 3 *pH and dissolved organic carbon (DOC) concentrations of the six groundwater wells from 1998—2006, and from after sewage sludge application from 2009—2011.*

Ni, Pb and Zn) during 2010 (Figure 4), together with elevated concentrations of DOC (Figure 3). However the peak groundwater concentrations were much lower than the groundwater concentrations exhibited underneath the water-saturated covered area of the impoundment. The pulse subsided in 2011 to pre-sludge application concentrations. Werner, *et al.* (2001) indicated that water infiltration through the dry composite cover on the impoundment was largely within 0—5% of the mean annual precipitation (400—800mm/year) as most of it was stored within the protective layer above the low permeable sealing layer. This has hence both limited and delayed the transport of contaminants from the sludge downwards into the underlying groundwater. Therefore the contamination in the underlying groundwater is likely caused by the horizontal migration of the contamination plume derived from the adjacent water-saturated cover.

Impoundment Groundwater Effluent Geochemistry

Concentrations of metals and DOC from the effluent groundwater (Well L) were similar to the deeper water-saturated groundwater (Figure 4). Concentrations of DOC and the metals Cr, Ni, Pb and Zn peaked in 2009 and were the highest of any of the wells (Figure 4), with 8 mg/L, 4.1µg/L, 619µg/L, 33µg/L and 20 400µg/L respectively. This confirms that readily leachable sludge-borne constituents derived from sewage sludge on the water-saturated area of the impoundment leached down into the deep groundwater and were transported along the dominant groundwater flow direction (Figure 2) due to inflowing uncontaminated groundwater from the western till slope. The pH of well L was the only one which drastically changed after the sludge application (Figure 3). It reduced during 2009 to pH 3 but recovered during 2011 to pre-sludge conditions as did the metal and DOC concentrations. This illustrates that any inflow of sludge-borne con-

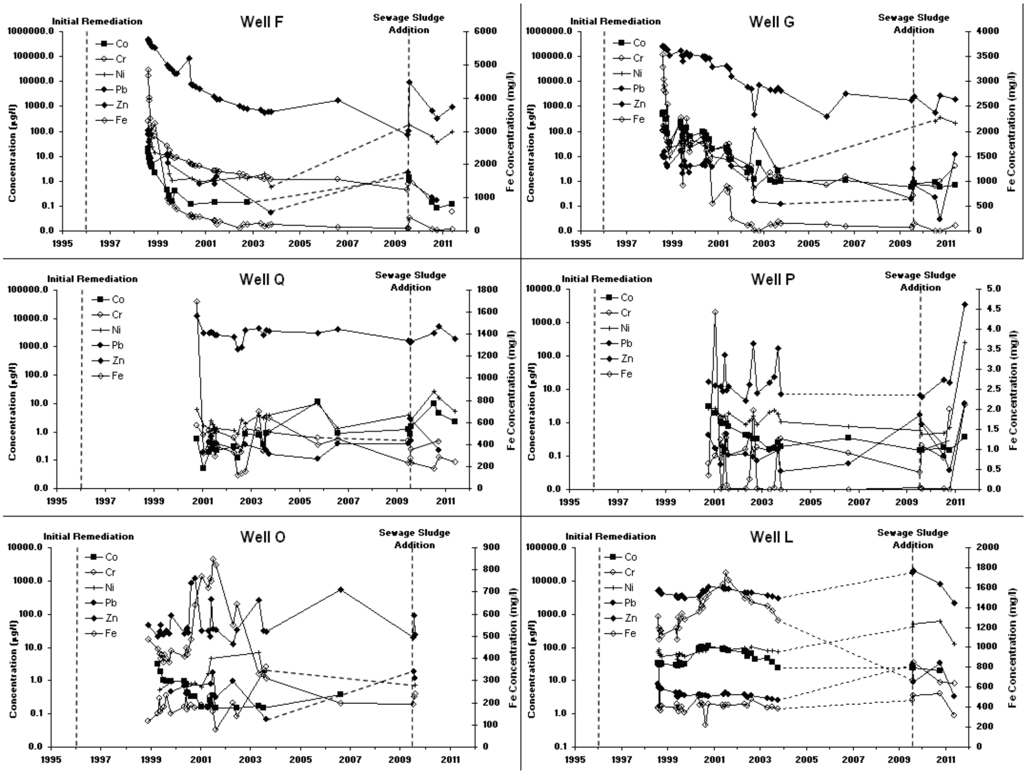

Figure 4 Dissolved metal concentrations of the six groundwater wells from 1998—2006, and from after sewage sludge application from 2009—2011(doted lines indicate sample absence of minimum of four years).

stituents from the dry-covered area of the impoundment did not reach the effluent groundwater before 2011.

Conclusions

The sewage sludge application drastically altered the metal concentrations in the tailings groundwater system. An initial plume of readily leachable sludge-borne constituents (DOC, Fe, Co, Cr, Ni, Pb and Zn) leached down into the deep groundwater below within the first year of application. The majority of the plume originated from the water-saturated area of the impoundment that was only covered with a 10% surface area of sludge. The dry-composite covered area of the impoundment, which contained a 70% sludge cover, did not contribute to the contamination plume due to the efficiency of the cover at reducing water infiltration and contamination dispersal. The constituents were transported along the dominant groundwater flow direction, underneath the dry covered area of the impoundment, to the effluent of the impoundment groundwater system due to inflowing uncontaminated groundwater from an adjacent till slope. Organic complexation modelling using the chemical equilibrium Visual MINTEQ modelling software (Version 3.0., Beta) will further enable an interpretation of the interaction between DOC concentrations and dissolved metal concentrations in the groundwater. All metal and DOC concentrations in the impoundment groundwater recovered to pre-sludge application concentrations within two years due to successful plant establishment on the sludge.

References

Ahlberg G, Gustafsson O, Wedel P. Leaching of metals from sewage sludge during one year and their relationship to particle size Environmental Pollution 2006; 144: 545—553.

Alakangas L. Sulphide Oxidation, Oxygen Diffusion and Metal Mobility in Sulphide-bearing Mine Tailings in Northern Sweden. Department of Chemical Engineering and Geosciences. Doctoral Thesis. Luleå University of Technology, Luleå, 2006, pp. 27.

Alakangas L, Nason PA. Declining element concentrations in groundwater after remediation in sulphide-rich tailings at Kristineberg, northen Sweden. In: Wolkersdorfer C, Freund A, editors. International Mine Water Association Symposium 2010: Mine Water and Innovative Thinking. Cape Breton University Press, Cape Breton University, Sydney, Nova Scotia, Canada, 2010, pp. 323—326.

Carlsson E. Sulphide-Rich Tailings Remediated by Soil Cover - Evaluation of cover efficiency and tailings geochemistry, Kristineberg, northern Sweden. Department of Environmental Engineering. Doctoral Thesis. Luleå University of Technology, Luleå, 2002, pp. 44.

Corrège O, Carlsson E, Öhlander B. Geochemical investigations of the groundwater in sulphide-bearing tailings remediated by applying till cover. Securing the Future. International Conference on Mining and the Environment. I, Skellefteå, 2001, pp. 97—114.

Eriksson J. Concentrations of 61 trace elements in sewage sludge, farmyard manure, mineral fertiliser, precipitation and in soil and crops. Swedish University of Agricultural Sciences, Uppsala, 2001, pp. 69.

Forsberg SL. Reclamation of Copper Mine Tailings Using Sewage Sludge. Department of Soil and Environment. Doctoral Thesis. Swedish University of Agricultural Sciences, Uppsala, 2008, pp. 88.

Garcia-Gil JC, Plaza C, Senesl N, Brunetti G, Polo A. Effects of long-term sewage sludge amendment on the composition, structure and proton binding activity of soil fulvic acids. Clean-Soil Air Water 2007; 35: 480—487.

Holmström H, Salmon UJ, Carlsson E, Petrov P, Öhlander B. Geochemical investigations of sulfide-bearing tailings at Kristineberg, northern Sweden, a few years after remediation. Science of the Total Environment 2001; 273: 111—133.

Lottermoser B. Mine Wastes, Characterisation, Treatment and Environmental Impacts. Germany: Springer, Berlin Heidelberg, 2003.

Neuschütz C, Isaksson K-E, Lundmark L, Gregor M. Evaluation of a dry-cover treatment consisting of vegetation, sewage sludge and fly ash. 8th International Conference on Acid Rock Drainage, Skellefteå, Sweden, 2009, pp. 9.

Werner K, Carlsson E, Berglund S. Oxygen and water fluxes into a soil-cover remediated mill tailings deposit: Evaluation of field data from the Kristineberg mine site, Northern Sweden. Securing the Future. International Conference on MIning and the Environment. Volume II, Skellefteå, 2001, pp. 896—905.