

# Pilot-scale Studies of Different Covers on Unoxidised Sulphide-rich tailings, Northern Sweden: Oxygen Diffusion

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## Abstract

The efficiency of five cover systems to decrease oxygen intrusion into sulphide-rich tailings was studied in pilot-scale test cells (5x5x3 m<sup>3</sup>). The covers consisted of clayey till, sewage sludge, fine-grained apatite concentrate or Trisoplast (a mixture of a polymer, bentonite and tailings sand) as sealing layers and unspecified till as protective cover. In one reference cell, tailings were uncovered. Oxygen concentrations below the entire covers were highest below the Trisoplast and apatite layers, and lowest below the sewage sludge layer. Effective diffusion coefficients ( $D_{eff}$ ) and oxygen fluxes were estimated in the covers with non-oxygen-consuming sealing layers (clayey till and apatite). For the protective covers the for  $D_{eff}$  ranged between E-09 and E-07 m<sup>2</sup>/s, and for the sealing layers between E-10 and E-09 m<sup>2</sup>/s, and for the entire covers between E-10 and E-08 m<sup>2</sup>/s. Seasonal variations in  $D_{eff}$  were larger within the covers than between the different cover systems. Oxygen fluxes through the entire covers with clayey till and apatite ranged between 0.2 and 4 mole m<sup>2</sup>/ year, which was a reduction of more than 99% compared to uncovered dry tailings.

**Key words:** dry cover, oxygen diffusion, sulphide oxidation, mining wastes, remediation

## Introduction

In mining waste, ore-associated sulphides not extracted during concentration processes may be exposed to the atmosphere when deposited. Iron-sulphides such as pyrite (FeS<sub>2</sub>) and pyrrhotite (Fe<sub>1-x</sub>S) oxidise in the presence of oxygen and water in acid-producing reactions. The acid produced increases weathering of other minerals and mobilises metals such as Cu, Cd, Co, Pb and Zn. Remediation of sulphide mine waste and prevention of metal-rich acid drainage is, therefore, an important topic. In Sweden, as well as in other countries, remediation of sulphide-rich tailings is mainly carried out using water covers (e.g. in Sweden; Eriksson et al. 2001; Höglund et al., 2005 Öhlander et al. 1997) or soil covers (e.g. in Sweden; Lindvall et al. 1997; Lundgren 1997; Höglund et al., 2005). The aim of these covers is to reduce the oxygen diffusion and for dry cover also to decrease the water percolation into the tailings. The application of a dry cover can be simple or complex; ranging from a single layer to several layers of different materials such as soil, non-reactive mine waste, oxygen-consuming organic materials, geo-synthetic materials or ashes. There are still considerable uncertainties regarding the optimisation of dry covers with regard to long-term performance and cost-efficiency. Pilot-scale field studies have been proven useful for studies of various types of dry covers (Aubertin et al. 1997), a major advantage is that they allow studies at reasonable cost during field conditions, and also narrow the commonly observed gap between field and laboratory studies. Most field studies of the effectiveness of dry cover on tailings have been performed on tailings which have been exposed to weathering for some time. It is, however, more effective to apply dry cover on unoxidised waste to minimise sulphide oxidation. Therefore, a project was initiated to study the different dry cover on unoxidised sulphide-rich tailings in test cells under field conditions. The control of oxygen diffusion through the covers is a key requirement for the efficiency of a cover. The aim of this paper was to determine oxygen diffusion through different dry covers on sulphide-rich tailings.

The objectives were to determine the

- oxygen concentrations in the cover systems
- effective oxygen diffusion coefficients in individual layers of the different covers as well as for the entire cover systems oxygen flux through the cover systems

## Effective diffusion coefficients

Oxygen can be transported by advection and/or by diffusion in water and in air, simultaneously or separately (Cussler, 1997), into a soil. The gas phase is more important for oxygen transport, since the solubility of oxygen in water is low (c. 8.6mg l<sup>-1</sup>) compared to in air (256 mg l<sup>-1</sup>). Advection e.g., winds and barometric pressure fluctuation is usually only important in the upper few centimeters in fine

textured media, owing to the low porosity in tailings (Elberling, 1996). Diffusion is the most important transport mechanism in finer textured media such as tailings, (Harries and Ritchie, 1985; Cussler, 1997). The oxygen diffusion through a non-consuming layer can be described by Fick's first law (Eq. 1).

$$F = -D_{eff} \left( \frac{\Delta C}{\Delta z} \right) \quad (1)$$

where  $F$  is the oxygen flux ( $\text{mole m}^{-2} \text{s}^{-1}$ ), and  $D_{eff}$  ( $\text{m}^2 \text{s}^{-1}$ ) is the effective diffusion coefficient of the media. The driving force for oxygen diffusion is the oxygen concentration gradient ( $\Delta C/\Delta z$ ), while  $D_{eff}$  is an expression for the physical properties of the media such as porosity, tortuosity and water saturation of a pore space.  $D_{eff}$  can be estimated based on the physical properties of the soil and its water content and by in-situ measurements of oxygen flux.

Estimation of  $D_{eff}$  based on soil physical properties has been reported by several researchers (Millington and Shearer, 1971; Reardon and Moddle, 1985; Collin and Rasmuson, 1988; Elberling et al., 1993; David and Nicholson, 1995; Stuparyk et al., 1995; Schaefer et al., 1997), and a summary is presented by Kim and Benson (2004). The following equation from Elberling et al. (1997), accounts for diffusion both in air and water.

$$D_{eff} = \tau D_a (1 - S_w)^\alpha - \tau S_w \frac{D_w}{K_H} \quad (2)$$

where  $S_w$  is the degree of saturation,  $K_H$  is Henry's constant for oxygen (25 at 10°C),  $\tau$  is the tortuosity factor ( $0.273 \pm 0.08$ ), and  $\alpha$  is an empirical coefficient ( $3.26 \pm 0.4$ ), which accounts for differences between the tortuosity factors for the liquid and gas phases. The fitting parameters,  $\alpha$  and  $\tau$ , are applicable for sandy materials with a saturation degree below 80% (Reardon and Moddle, 1985).

$D_{eff}$  was determined by a cell mass balance (Eq. 1 and 2), in a way previously used by Lundgren 2001, Carlsson 2002. The oxygen flux,  $F$  ( $\text{mole m}^{-2} \text{s}^{-1}$ ) into a lysimeter with height  $h$  can be determined with

$$F = h \frac{dC_{lm}}{dt} \quad (3)$$

where  $dC_{lm}$  is the oxygen concentration change with the time in the lysimeter beneath a layer.

Making Equation 1 equal to Equation 3 gives an expression for  $D_{eff}$ .

$$-D_{eff} = h \left( \frac{\Delta z}{\Delta C} \right) \frac{dC_{lm}}{dt} \quad (4)$$

Integrating Equation 4, using the initial condition with zero concentration in the lysimeters at time zero ( $C_{lm} = 0$ , at  $t = 0$ ) gives

$$D_{eff} = \ln \left[ \frac{C_0}{C_0 - C_{lm}(t)} \right] \frac{\Delta z}{t} h \quad (5)$$

where  $C_0$  is the oxygen concentration above a layer (assumed to be constant) and  $z$  is the distance between  $C_0$  and  $C(t)$ .

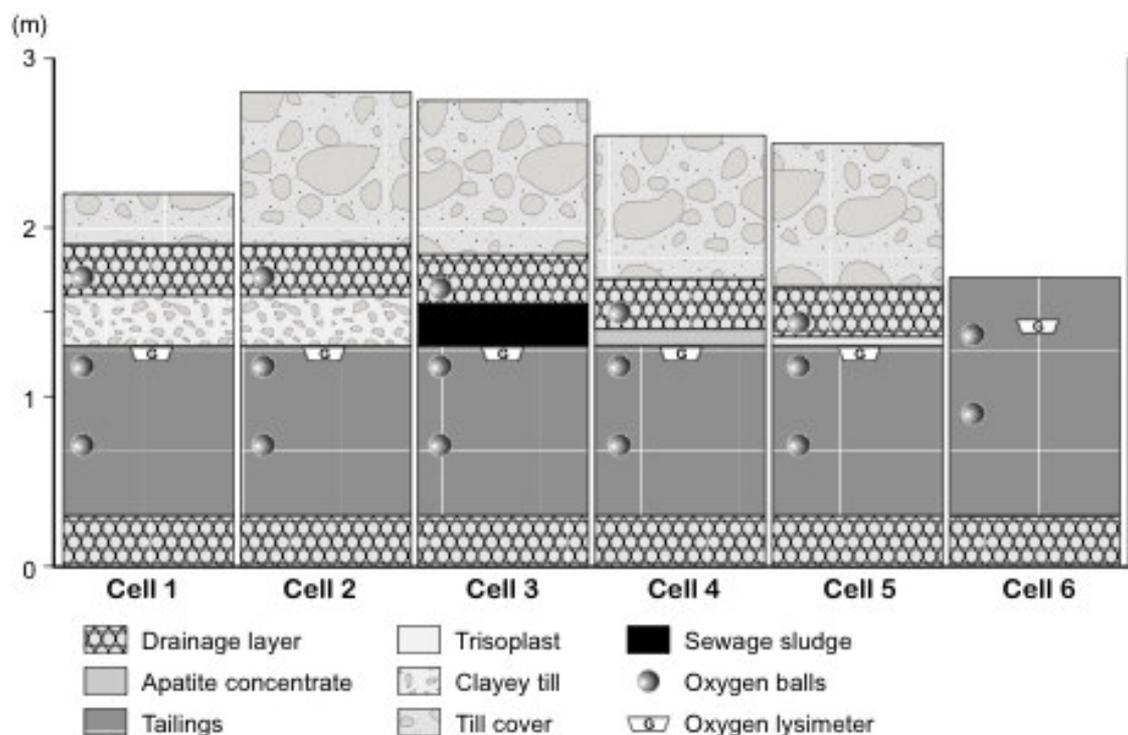
For a cover system with several layers the harmonic mean was used to estimate the equivalent effective diffusivity,  $D_{eff}^E$ , for all layers when the effective diffusivity,  $D_{eff}^i$  and thickness  $m_i$  for each layer,  $i$ , are known.

$$D_{eff}^E = \frac{\sum (m_1 + m_2 \dots m_i)}{\sum \frac{m_i}{D_{eff}^i}} \quad (6)$$

## Material and Methods

Six concrete cells (surface 5x5 m<sup>2</sup>, depth 3 m) (Figure1) were constructed during the summer of 2001 at the Kristineberg mine site, owned by the mining company Boliden. An inert HDPE liner (high density polyethylene) covers the concrete inner walls and floor to prevent attack of possible acid produced by the pyrite oxidation. The cells were insulated from the inside and outside in an attempt to prevent horizontal freezing. Windows were installed in the walls in cells 2, 4 and 6, where the different layers could be viewed. At the bottom of the cells a 0.3-m thick drainage layer was applied followed by 1.0 m sulphide-rich tailings (Figure1). Different sealing layers were used on the tailings, and above these a drainage layers (0.3 m) and finally a protective cover of unclassified till was applied at the top. The protective cover was 0.6 m thick in cell 1, whereas the other cells had a thickness of 1.2 m. The sealing layers and the sewage sludge layer consisted of 0.3 m clayey till in cells 1 and 2; 0.25 m sewage sludge in cell 3; 0.1 m of apatite concentrate in cell 4 and 0.05 m of Trisoplast in cell 5. The upper drainage layer was intended to simulate run-off. In cell 6, tailings were left uncovered as a reference.

**Figure 1** Schematic picture over the layers in the test-cells 1-6 at Kristineberg, Northern Sweden



The grain size of the tailings was classified as sandy-silt and the dominating sulphides were pyrite (FeS<sub>2</sub>), approximately 48% and pyrrhotite (Fe<sub>1-x</sub>S), approximately 4.8%. Gangue minerals were quartz [SiO<sub>2</sub>], muscovite [KAl<sub>2</sub>(Si<sub>3</sub>AlO<sub>10</sub>)(OH)<sub>2</sub>], cordierit [Mg<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub>], clorite [(Mg,Fe)<sub>6</sub>(SiAl<sub>4</sub>O<sub>10</sub>)(OH)<sub>8</sub>], talc [Mg<sub>3</sub>Si<sub>4</sub>O<sub>10</sub>(OH)<sub>2</sub>], calcite [CaCO<sub>3</sub>], dolomite [Mg,Ca(CO<sub>3</sub>)<sub>2</sub>], microcline [KAlSi<sub>3</sub>O<sub>8</sub>], diopside [Ca(Mg,Al)(Si,Al)<sub>2</sub>O<sub>6</sub>] and albite [NaAlSi<sub>3</sub>O<sub>8</sub>]. The quartz content was 20% and the content of other silicates was lower. The calcite and dolomite content was approximately 2.5%, respectively.

The **clayey till** in cells 1 and 2 was a local glacial soil, with a clay content of 9%. The **sewage sludge** in cell 3, a municipal waste from a nearby community, constituted a non-compacted layer that did not allow geotechnical measurements.

The **apatite concentrate** in cell 4 was a waste from the Kiruna iron mine in northernmost Sweden. The grain size was dominated by silt. The concentrate consisted of approximately 83.5% apatite (Ca<sub>5</sub>(PO<sub>4</sub>)<sub>3</sub>(OH)), 5% of other calcium phosphate minerals, and 11.5% of calcite. The **Trisoplast** in cell 5 was a mixture of 88.0% tailings, 11.8% bentonite and 0.2% polymer. The physical properties such as porosity, saturated hydraulic conductivity, dry density and bulk density determined in laboratory and in field for the sealing layers are summarised in Table 1. Trisoplast had the lowest saturated hydraulic conductivity according to the laboratory measurements.

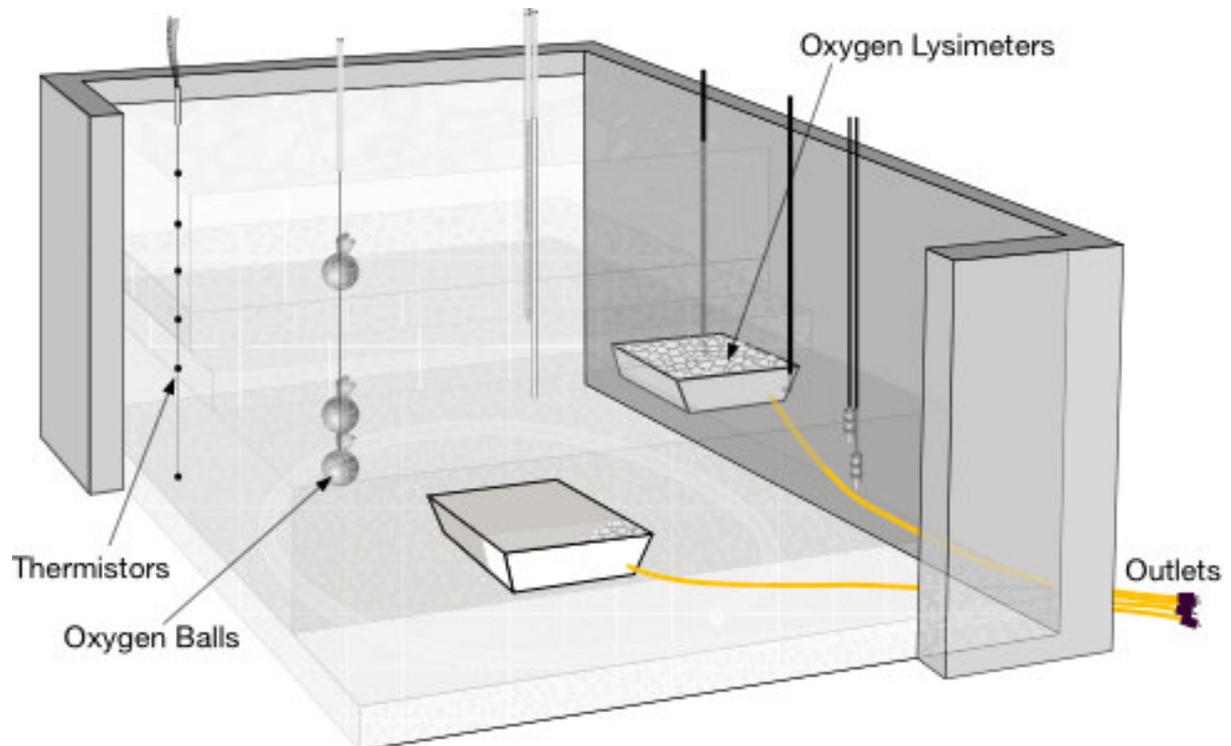
**Table 1** Physical properties of the different materials used in the cover systems in the test-cells determined by laboratory measurements, SLU (2005)

Material	Porosity (%)	Saturated hydraulic conductivity (m/s)	Dry density (g/cm <sup>3</sup> )	Bulk density (g/cm <sup>3</sup> )
Clayey till	40.5	3.5E-08	1.62	2.72
Apatite	35	3.2E-08	1.77	2.72
Trisoplast		4E-11		2.45
Tailings	55.4	1.31E-05	1.7	3.82

In order to determine  $D_{\text{eff}}$  and oxygen flux for the entire cover systems and for the sealing layers, oxygen concentration in quartzite filled lysimeters (1x1x0.2 m<sup>3</sup>) and oxygen balls were measured (Figure 2). This technique has earlier been used by Lundgren (2001) and Carlsson (2002). The quartzite filled lysimeters had a porosity of 54% and were located directly below the sealing layers and the balls above and beneath the sealing layers. They were covered with geotextile on top to prevent material from the sealing layer from falling into the lysimeters, and the sides were sealed with bentonite to prevent oxygen consumption by tailings through the sides. Three pipes were attached to the lysimeters; one water outlet at the bottom and two pipes up to the surface for oxygen sampling.

The geotextile oxygen balls were filled with quartzite and installed 0.1 m above and 0.1 m and 0.5 m beneath the sealing layers (Figures 1 and 2). A tube was inserted into the balls and extended to the surface for sampling (Figure 2).

**Figure 2** Location of installed equipment in the cells.



To determine the  $D_{\text{eff}}$ , (Equation 5) for the entire cover and for the sealing layers, the lysimeters were flushed with nitrogen to achieve initial zero oxygen concentration. Thereafter, the oxygen concentrations were measured after certain times. Atmospheric oxygen concentration was used as  $C_0$  for the protective cover. The oxygen concentrations in the oxygen balls above the sealing layer were used as  $C_0$  for the sealing layers. Oxygen may be consumed within the covers by oxidation of organic matter and by minerals, but this consumption was probably minor and has been neglected. The effective diffusion

coefficients ( $D_{eff}$ ) were estimated in field for the cover systems without oxygen-consuming layers (clayey till and apatite). Trisoplast, tailings and sewage sludge in cells 3, 5 and 6 are highly oxygen-consuming soils;  $D_{eff}$  was therefore not determined for these soils.

The estimated  $D_{eff}$  for the entire cover system and for the sealing layers were used for determination of  $D_{eff}$  for the protective cover by using Equation 6, and the thickness of the layers.

Oxygen fluxes were calculated with Equation 1 and the oxygen gradients determined with oxygen balls above and beneath the sealing layers.

### ***Analysis of gases***

Gas in the lysimeters and in the oxygen balls was sampled every second week during spring, summer and autumn in 2004 and once a month during 2005. The samples were analysed for oxygen, methane and carbon dioxide using Maihak S710 (Hallberg, 2005). Methane and carbon dioxide were calibrated with specific gas concentrations. The precision of the instrument was better than 2% of the analysed value, according to the manufacturer.

### **Results and discussion**

Problems with frozen probe connections during the winter and high water content in the balls and in the lysimeters during the spring reduced the quantity of data. In the uncovered tailings (cell 6), oxygen concentrations decreased with depth. At depths of 0.5 m and 0.9 m the concentrations ranged from zero to almost atmospheric concentration (Figure 3). In general, lower concentrations at 0.9 m, indicating that sulphide oxidation occurred. The change in oxygen concentration with depth could be attributed to variations in the moisture content. In the autumn the concentrations were lower than during the summer, probably as a result of higher moisture content in the autumn, decreasing the oxygen diffusion.

In the cover systems the oxygenated zone extended from the surface to the tailings below the cover (Figure 3). The concentrations in the lower part of the protective cover (above the sealing layer) varied between 14 and 21 vol% in cells 1, 2, 4 and 5, and was less than 14 vol% above the sewage sludge layer (cell 3). On a few occasions, in April and during autumn, even lower concentrations were observed in the protective cover, probably due to high moisture content resulting from infiltrated water from snow melt or heavy rainfalls.

The concentrations were, in general, lowest below within the sulphide tailings throughout the sampling period. Layers with low saturated hydraulic conductivity such as the sealing layer in cells 1, 2, 4 and 5, are expected to reduce the oxygen concentration to near zero below the layers. Such low values were only found throughout the sampling period beneath the sewage sludge layer. Beneath the other sealing layers, the oxygen concentrations were, in general, below 10 vol%, but sometimes during early summer (June) almost as high as in the atmosphere.

High concentrations indicate that the layers were rather permeable, which may be a result of insufficient compaction of the layers during installation. For the Trisoplast layer, which consists of tailings, bentonite and polymers, it can be concluded by the high oxygen concentrations below this layer that it was not efficient as a barrier against oxygen. Laboratory studies of Trisoplast showed very low saturated hydraulic conductivity (Lars Eriksson, MRM, personal communication, 2005). Probably, there was a failure in the structure of the barrier. The combination of the Trisoplast and tailings instead of common sand has never been used in field before.

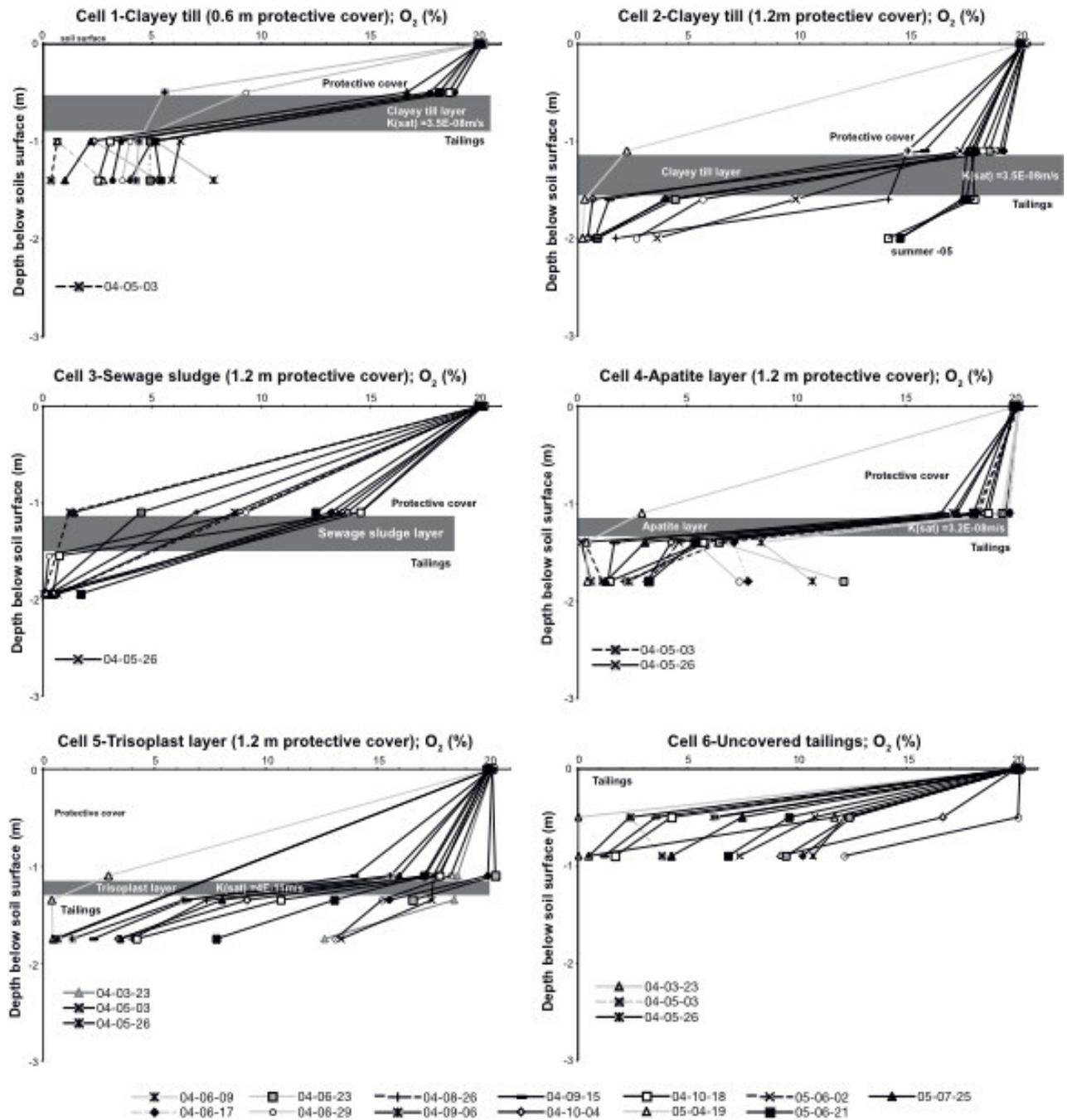
The oxygen concentrations were lower beneath the other barriers (clayey till and apatite), but not so low to consider the layers as ideal barriers against oxygen. The only layer that reduced the oxygen concentration to near zero was sewage sludge

Because the sealing layers in the cells were frozen during the winter it might be possible that the layers were damaged. Depending on the climate, the required thickness of a protective cover varies. In northern Sweden, where the frost depth can be deep, the protective cover ought to be at least 1 m thick to prevent freezing of the sealing layer. The construction of the test-cells might have increased the frost depth.

Calculations of the effects of drought on the saturation of a sealing layer of clayey till during long dry periods showed that the water content was reduced marginally, and that it took a very long time to dry (Moreno and Neretnieks, 2004). Modelling of six consecutive dry years in northern Sweden showed that the water content in the sealing layer remained close to saturation, and that the protective cover had a saturation of 80% at a depth of 1 m, and of 10% in the upper part (Moreno and Neretnieks, 2004). The lack of moisture measurements in the test-cells complicated the evaluation of the capacity of the sealing

layers to maintain saturation throughout the year, but high oxygen concentrations during the summer indicate that the layers were only partially saturated.

**Figure.3** Oxygen concentration in the cover systems applied on sulphide-rich tailings and in uncovered tailings in the test-cells.



Carbon dioxide was observed in the uncovered tailings as well as above and below the tailings with covers. The concentrations were up to 10 vol% below the sewage sludge layer and up to 6 vol% below the other layers. Upward diffusion of carbon dioxide and large amounts of infiltrated water into the lysimeters sometimes affected the oxygen concentration in the lysimeters, and these occasions were therefore omitted when estimating the effective diffusion coefficients ( $D_{eff}$ ). The oxygen-consuming character of the Trisoplast, the uncovered tailings and the sewage sludge made it impossible to calculate  $D_{eff}$  of these covers with this method, since the oxygen concentration in the lysimeters was affected. The protective covers in all cells consisted of similar material, and cells 2-4 have the same thickness (1.2 m), while in cell 1 it is only 0.6 m thick. The  $D_{eff}$  values were higher for the entire covers and

protective covers than for the sealing layer alone, which was expected, due to the larger differences in physical properties such as porosity and hydraulic conductivity. The approximate  $D_{eff}$  for the protective covers in the three cells varied between  $10^{-09}$  and  $10^{-07} \text{ m}^2 \text{ s}^{-1}$  (Figure 4). Higher  $D_{eff}$  values were, in general, found during dry periods such as the summer and during snow covered periods.

The major aim of the sealing layers and the sewage sludge layer was to decrease the oxygen entry, either by oxygen consumption by the decomposition of organic matter in the sewage sludge or by reduction of the oxygen diffusion by maintaining a high degree of saturation in the other sealing layers. The reduction of oxygen diffusion through the sealing layers is expected to be much greater than through protective covers, because of the lower saturated hydraulic conductivity. This means that the oxygen flux into the tailings is controlled by diffusion in the sealing layers and the oxygen consumption below the sealing layers. The  $D_{eff}$  values for the sealing layers ranged from  $10^{-10}$  to  $10^{-08} \text{ m}^2 \text{ s}^{-1}$  in cells 1, 2 and 4. The sealing layers differ in thickness, porosity and saturated hydraulic conductivity but the observed differences in  $D_{eff}$  were more related to seasonal variations than to the type of sealing layer. The limited number of estimated  $D_{eff}$  values made it difficult to prove higher diffusion through one of the sealing layer than through another, and the average  $D_{eff}$  values were rather similar between the sealing layers, on average  $2.8\text{E-}09 \text{ m}^2 \text{ s}^{-1}$  for the clayey till in cell 1, and  $2.6\text{E-}09 \text{ m}^2 \text{ s}^{-1}$  in cell 2, and for the apatite layer  $1.2\text{E-}09 \text{ m}^2 \text{ s}^{-1}$ . The thickness of the apatite layer was only one third of the thickness of the clayey till, which may have reduced the effectiveness of the layer. Thicker snow cover was measured in cell 1, since the soil surface was deeper down. The higher cell walls shadowed the soil surface and thereby probably blocked the solar radiation, which led to later and slower snow melting than for the other cells. For the other cells, thinner snow cover caused some soil surfaces to dry earlier (cells 3-5) than others in the spring. The rapid melting in these cells may have resulted in percolation of water along the cell walls, as the cover was partially frozen. This probably resulted in that the protective covers drying out faster and consequently an increase in oxygen diffusion, which may explain the high  $D_{eff}$ . Large amounts of percolated water created oxidised channels (yellow-brown precipitates) along the cell walls in which oxygen could be transported into the tailings. These  $D_{eff}$  values were of the same order of magnitude as those modelled and observed for other covers (Table 2).

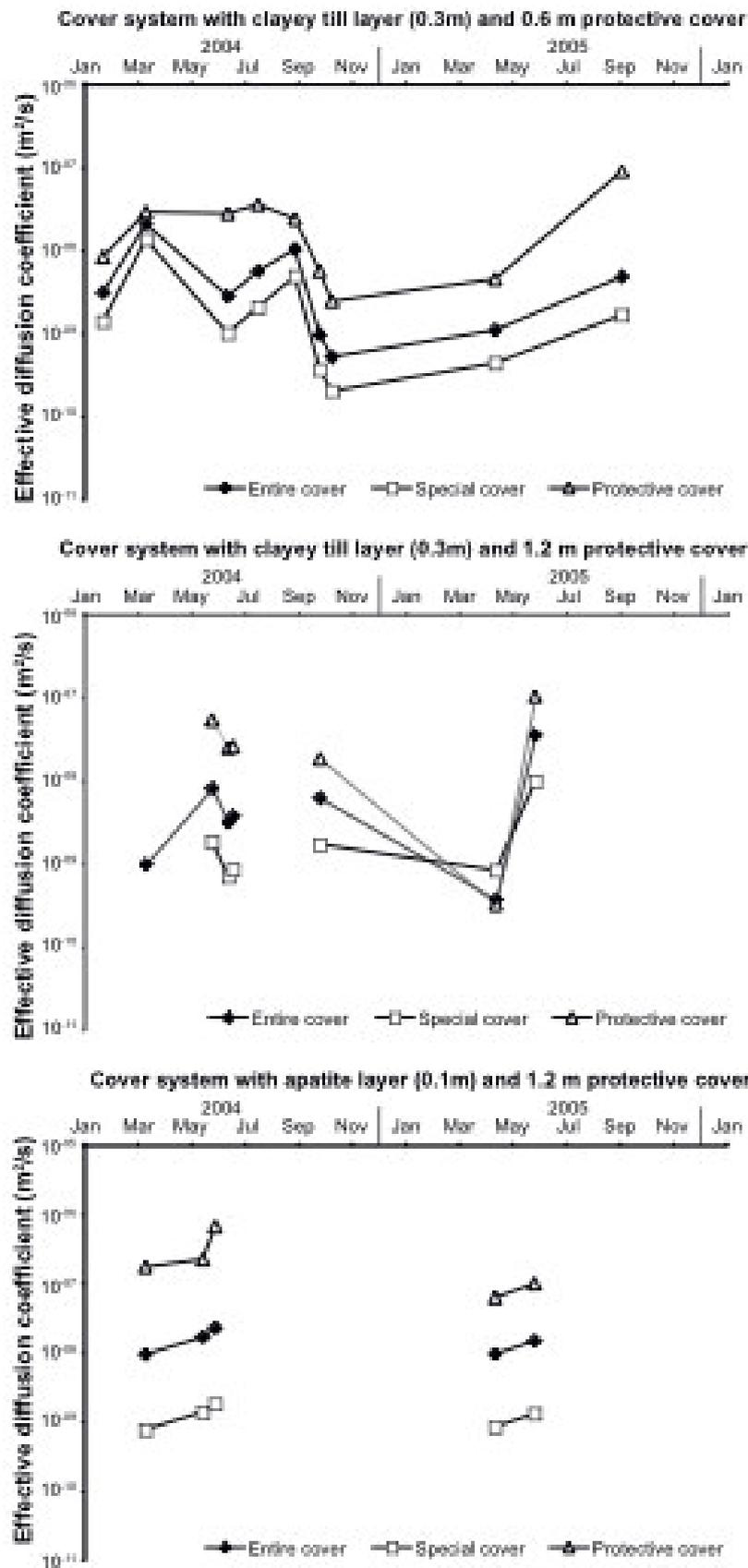
$D_{eff}$  values are considered to be higher in field than in laboratory (Yanful, 1993). Various investigators (Reardon and Moddle, 1985; Elberling et al., 1993; Schaefer et al., 1997; Aubertin et al., 2000) have demonstrated that the degree of saturation in a soil media is the factor that has the greatest influence in the  $D_{eff}$ .

The variation in  $D_{eff}$  for fine sandy to silty media at low water saturation (0 to 0.6) was estimated to be less than one order of magnitude, while at saturation between 0.6 and 1.0 the  $D_{eff}$  varied in four orders in magnitude (Elberling et al., 1993). Several studies have shown that a water saturation of more than 0.85 would result in  $D_{eff}$  values lower than  $1.0\text{E-}08$  for; fine sand (Reardon and Moddle, 1985; Elberling et al., 1993) and for clayey till similar to the one used in cells 1 and 2 (Moreno and Neretnieks, 2004). Sealing layers of clayey till should have approximately 95% saturation to be effective as a barrier against oxygen, which corresponds to a  $D_{eff} < 1.0\text{E-}09 \text{ m}^2 \text{ s}^{-1}$  (Moreno and Neretnieks, 2004). At dry periods, the sealing layer saturation may decrease to 70%, which could increase  $D_{eff}$  three orders of magnitude (Moreno and Neretnieks, 2004). Because of the lack of moisture measurements the relation between the diffusion coefficients and moisture is uncertain in the test-cells.

Upward diffusion of carbon dioxide may induce an oxygen gradient that is not related to oxygen consumption in the tailings, this effect was neglected in the calculations, but could be significant due to the relatively high carbon dioxide concentrations (up to 10%).

The estimated  $D_{eff}$  values (Equation 1) in saturated and dry uncovered tailings were  $2.9\text{E-}09$  and  $3\text{E-}06 \text{ m}^2 \text{ s}^{-1}$ , respectively. The iron-sulphide content was very high; almost 50% and the tailings were fresh, which may increase the sulphide oxidation rate (Nicholson et al., 1997). However, fresh tailings may be wet at the time of deposition and therefore show low sulphide oxidation rate initially (Elberling et al., 1993; Woyshner and Swarbrick, 1997). This implies that new tailings may have low initial sulphide oxidation rate just after deposition, but this rate increases as water content decreases by drainage and evaporation. The oxygen gradient as measured in the uncovered tailings and a  $D_{eff}$  of  $3\text{E-}06 \text{ m}^2 \text{ s}^{-1}$  gives an oxygen flux of approximately  $940 \text{ mole year}^{-1} \text{ m}^2$ . In wet tailings with an oxygen concentration gradient close to zero, the pyrite oxidation rate would be less than  $1 \text{ mole year}^{-1} \text{ m}^2$ .

**Figure 4** Time series for estimated effective diffusion coefficients for the protective covers, sealing layers and the entire cover in the test cells



**Table 2** Modelled and measured effective diffusion coefficients ( $D_{eff}$ ) and oxygen fluxes for tailings and cover

Study	Sulphide	Method	Saturation	$D_{eff}$ ( $m^2s^{-1}$ )	Oxygen flux ( $mole\ m^{-2}\ year^{-1}$ )
Eberling et al., 1993	40Fine grained	F	Dry	1.2E-05 **	1560 <sup>b</sup> , at surface
		F	Modest	2E-06**	581b
		F	Wet	4.2E-10**	0.25b
Kim and and Benson, 2004	Silty loam	M	Wet	1.5E-08*	
		M	Dry	6.2E-06*	
	Silty clay loam	M	Wet	1.9E-08*	
		M	Dry	7.7E-06*	
Tibble and Nicholson, 1997	Fresh	F	Wet		33b
	Old (5-6 years) and (10 years)	F			192 and 81b
Woysner et al., 1997	Non-reactive	F	28%	1.7E-06	
Yanful, 1993	Pyrite and pyrrhotite	L+ M	27.5-46.9%	1.0-6.1E-08	66.5
Alakangas and Öhlander, 2005	Old sandy (60 years)	F		5.5E-08	9.9
This study	Fresh silty	F	Wet 90%	2.9E-09b**	
		F	Dry 17%	3E-06b**	0.8 at <sup>0.5m</sup> depth
<b>Covered tailings</b>					
Kim and and Benson, 2004	Bentonite	M	D+W	1.1E-08b*	94l <sup>at 0.5m</sup> depth
	Compacted till	M	D+W	1.7E-07*	
Woysner et al., 1997	clay	F	60%	2.5E-07	
Tibble and Nicholson, 1997	Sand (0.5m) and non-sulphide tailings (0.8m) and Sandy gravel (0.3m)	F			7b
Cabral et al. 2000	Pulp and paper residues	L+M		8.3E-09-9.7E-07	6.2
Yanful, 1993	Fine sand (0.3m)+ clay	L+M	25%	3.9E-09	0.13
Carlsson, 2002	(0.6m)+coarse sand (0.3m)				
	Clayey till (0.3m)	F		1.6E-09	0.008
	+ protective till cover (1m)				
	Clayey till, 0.3m	F		3.9E-09	0.4
	+ protective cover (1.3 m)				
Moreno and Neretnieks, 2004	Clayey till (0.3m)	F		4.9E-10	0.34
	+ protective cover (1.5m)				
	Clayey till (0.3m)	M	Wet 95%	1.0E-09	1.0
	Clayey till (0.3m)	M	Dry 0%	2.0E-06	1974
	Clayey till (0.3m)	F		2.8E-09	1.1
This study	protective cover (0.6 m)				
	Clayey till (0.3m)	F		2.6E-09	1.1
	+ protective cover (1.2 m)				
	Apatite concentrate (0.1m)	F		1.2E-09	2.0
	+ protective cover (1.2m)				

L= Laboratory, M= Modeling, F = Field , <sup>b</sup> Oxygen consumption method \* diffusion in air \*\* diffusion in water and air

The maximum oxygen intrusion was estimated to 592 mole year<sup>-1</sup> m<sup>-2</sup> in a dry cover system with a sealing layer of clayey till and a protective cover, and if the oxygen was instantly consumed at the bottom of the cover (Moreno and Neretnieks, 2004). An oxygen flux of 1 mol m<sup>2</sup> year<sup>-1</sup> was estimated by modelling, when assuming a high degree of saturation in a sealing layer of clayey till similar to the one in this study, and with an oxygen concentration close to zero below the cover (Moreno and Neretnieks, 2004). A saturation of only 85% in such a cover may increase the oxygen flux by almost three orders of magnitude compared to a water-saturated sealing layer, (Moreno and Neretnieks, 2004). The estimated oxygen fluxes (Figure 5) for each layer in the entire cover system in each cell were similar, while the  $D_{eff}$  values and the oxygen gradients varied. In general, low oxygen fluxes were observed in the entire cover system in the cells (Figure 5) when both the diffusion coefficients and oxygen consumption are considered to be low (during cool or wet seasons). The oxygen fluxes ranged from 0.2 to 4 mole year<sup>-1</sup> m<sup>-2</sup> in the cells with clayey till and apatite (Figure 5), which is rather similar to

fluxes estimated for other covers (Table 2). High degree of saturation in the tailings or in a cover seems to reduce the oxygen flux significantly (Table 2). The variation of  $D_{\text{eff}}$  values and oxygen fluxes between the cover systems was not as large as might be expected, considering the variations of oxygen concentrations below the covers. No obvious difference between the ability of the cover systems to limit the oxygen intrusion could be confirmed from this study, possibly due to the limited number of  $D_{\text{eff}}$  values. The cover systems in the test-cells reduced the oxygen flux into the tailings by more than 99% compared with dry, uncovered tailings. Analysis by modelling showed that transient oxygen transport in a complex cover system with clay layers occurs for about 10 years, and thereafter the transport is steady (Kim and Benson, 2004). This implies that the cover layers may improve in the future.

The decomposition of organic matter in the sewage sludge layer probably consumed most oxygen below and above the layer, and consequently little oxygen diffused into the tailings. The release of sulphate and metals in the tailings below the sewage sludge was lower than for the other cover systems, and decreased significantly during 2005 (Alakangas, 2006). This suggests that the sewage sludge was most effective as a barrier for oxygen in the short term. The long-term efficiency is probably limited, since the organic matter will be consumed (Hallberg et al., 2005).

The relatively high oxygen concentrations below the Trisoplast suggest that this layer failed as a barrier against oxygen. High amounts of infiltrated water through this layer also indicate its failure as barrier against water. This may be a result of the mixing properties of the Trisoplast, since tailings never have been used as matrix before.

The large advantage with the field technique used here compared to other techniques is the ability for long-term studies without interrupting the cover systems. The method is however also afflicted with some shortcoming.

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