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The effect of temperature and digested sewage sludge cover over tailings on the leaching of contaminants from Ballangen tailings deposit

Jinmei Lu[a,*], Tiina Leiviskä[b] and Ingar Walder[c,d]

[a] Department of Technology and Safety, UiT The Arctic University of Norway, N-9037 Tromsø, Norway
[b] Chemical Process Engineering, University of Oulu, P.O. BOX 4300, FI-90014 Oulu, Finland
[c] Kjeøy Research and Education Ctr, Vestbygd, Norway
[d] New Mexico Institute of Mining and Technology, Socorro, NM, USA
*Corresponding author. E-mail: jinmei.lu@uit.no

ABSTRACT

Dry covers can be applied above tailings to reduce and prevent formation of acid mine drainage and leaching of contaminants. Efficiency of covers is affected by different parameters, of which temperature change under climate change context is one. Here, a laboratory column leaching experiment was performed under four temperatures, 5, 10, 14, and 18 °C on unoxidized tailings from Ballangen, Norway. 600 mL of water was added to each column every second week and leachate was collected and analyzed for pH, salinity, alkalinity, concentrations of sulfate, Co, Fe, Mn, Ni, and Zn. A thin layer of digested sewage sludge was added to columns after the 16th leaching cycle. In total, 21 leaching cycles were performed. Results showed low oxidation of tailings and therefore high pH and low salinity, SO_4^{2-}, Fe, Ni, and Co in the leachates at leaching temperature of 5 °C. Addition of sludge cover slowed down oxidation of underlying tailings and decreased leaching of SO_4^{2-}, Fe, Mn, Co, Ni, and Zn from the tailings deposit, especially at relatively high temperatures. 10 °C is a threshold temperature, below which leaching is not affected by the cover addition so much. At a leaching temperature higher than 10 °C, the sludge cover addition can reduce the leaching of elements significantly.

Key words: column, dry cover, sewage sludge, tailings, temperature

HIGHLIGHTS

- Temperature as an important impact on the efficiency of dry covers in preventing leaching of contaminants from waste deposit.
- 10 °C is a threshold temperature, below which the cover addition is not very efficient.
- At a leaching temperature higher than 10 °C, the sludge cover addition can reduce the leaching of elements significantly.

INTRODUCTION

The tailings or mine waste disposal facility has proven to be the most contentious component of mining activities and represents the source of significant environmental and economic impacts due to poor management in many cases. Dry covers can be applied to reduce the environmental impacts of mining wastes (Hallberg *et al.* 2005). The use of different cover materials over the tailings to prevent the oxidation of tailings has been proposed in various studies, such as till, clay, or some mixed artificial material such as fly ash and sludge or cement-stabilized fly ash (Hallberg *et al.* 2005). A mixture of fly ash and biosludge produced in a paper mill was applied as a dry cover over the sulfide mine tailings in Falun, Sweden and the efficiency of the dry cover was evaluated (Hallberg *et al.* 2005). The efficiency of fly ash cover on the mobilization and accumulation of metals in the underlying tailings was investigated (Lu *et al.* 2014). Organic covers can be used to prevent the diffusion of oxygen into the reactive sulfidic tailings and therefore eliminate the oxidation of sulfide minerals in the tailings and generation of acidic leachate afterwards (Peppas *et al.* 2000). The efficiency of organic cover over reactive sulfidic tailings was investigated by a column experiment and the results showed that the cover inhibited the oxidation of the tailings and therefore the generation of acidic drainage to a great extent (Peppas *et al.* 2000). Oxygen diffusion coefficient through the candidate cover materials was investigated in another study (Yanful 1993). Jia *et al.* (2013) investigated the use of green liquor dregs (GLD) and fly ash from paper mills as a paste additive to tailings as a cover to seal the mine waste and found that a proportion of 7:2:1 for tailings:GLD:FA proved to be a satisfactory geotechnical condition for the sealing of mine waste. Column tests were performed to investigate the effectiveness of covers made of low-sulfide tailings in prevention of acid mine drainage generation and the results showed that the application of the cover significantly reduced the amount of contaminants in the percolated water, with the reduction greater than 99% for Zn, Cu, and Fe (Bussiere *et al.* 2004). The use of sewage sludge as a sealing layer over sulfidic mine tailings at Kristineberg mine, northern Sweden was investigated in a pilot experiment (Nason *et al.* 2013). The results showed that the sludge was an effective barrier to oxygen influx as it formed both a physical obstruction and an organic reactive barrier to prevent oxygen to the underlying tailings (Nason *et al.* 2013). The long-term efficiency of paper mill sludge as cover over the sulfidic mine wastes from historic Au mines in Northern Ontario, Canada was studied and the results showed that the paper sludge covers are suitable for use in medium- to long-term mining reclamation strategies (Cousins *et al.* 2009).

Currently, there are many studies on the use of different candidate cover materials to prevent the oxidation of tailings. These covers can reduce the infiltration of oxygen and water to reach the underlying tailings to different extents, and thus reduce the production of acidic leachate and leaching of metals to the surrounding environment. Few studies have focused on the factors that will influence the efficiency of different covers. The oxidation of tailings is controlled by many factors, including pH, acid neutralizing capacity, temperature, concentration and reactivity of redox species, oxygen availability, concentration of carbon dioxide, nutrient requirements, sulfide mineralogy and particle size, galvanic interactions, and hydrology (Hallberg *et al.* 2005). Of the different factors, temperature change linked to climate change can be one important factor for the efficiency of covers, and more research is needed to determine how temperature affects leaching rates.

In this study, a laboratory column leaching experiment on the unoxidized tailings was performed under four different temperatures, 5, 10, 14, and 18 °C. The leachate was collected periodically and analyzed for pH, salinity, alkalinity and concentrations of sulfate, Co, Fe, Mn, Ni, and Zn. A thin cover of digested sewage sludge from a local wastewater treatment plant was applied on the tailings after several leaching cycles. The aim was to investigate the effect of temperature change on the efficiency of the digested sewage sludge covers in reducing the leaching of metals from the tailings deposit.

STUDY AREA

In this study, Ballangen tailings deposit, located in Ballangen municipality, Nordland County, northern Norway was selected as the study area (Figure 1). The tailings in the deposit are from the nickel mine 'Nickel and Olivine AS' and were deposited at the two deposits of Ballangsleira and Fornes (Figure 2) in tidal flat areas. In total, about 7 million tons of tailings were deposited at Ballangsleira deposit. The tailings are mainly composed of mafic mineral (forsterite, orthopyroxene, amphibole, clinochlore) plagioclase, with minor to trace amounts of pyrrhotite and traces of chalcopyrite (Embile *et al.* 2018, 2019). The tailings were covered with a thin soil layer of 10–20 cm to prevent the weathering and oxidation of the underlying tailings. However, the thin soil layer is not efficient in preventing the oxidation of the underlying tailings (Segalstad *et al.* 2006; Embile *et al.* 2018). Leaching of metals such as Ni from the oxidation of tailings to the surrounding environment has been reported in a previous study (Embile *et al.* 2018). Therefore, Ballangsleira deposit was selected as the study area to investigate the leaching of contaminants from tailings and the efficiency of a cover made with digested sewage sludge from a local wastewater treatment plant in reducing the leaching of contaminants from tailings deposit.

MATERIAL AND METHODS

Tailings sampling and analysis

Tailings samples were collected from below the oxidation front at Ballangsleira deposit in June 2016 (Figure 2). The tailings were collected into several buckets with a plastic spade and were covered with a lid after the sampling. The collected tailings were transported to Kjeøy Research & Education Center (KREC), Norway and stored at 4 °C for analysis. The experiments were set up and run at KREC.

Probe measurements of temperature, pH, Eh, TDS, and salinity were performed immediately. Alkalinity was measured by titration using Metrohm titrando on samples showing pH above 4.5. Sulfate was measured using IC Metrohm 870.

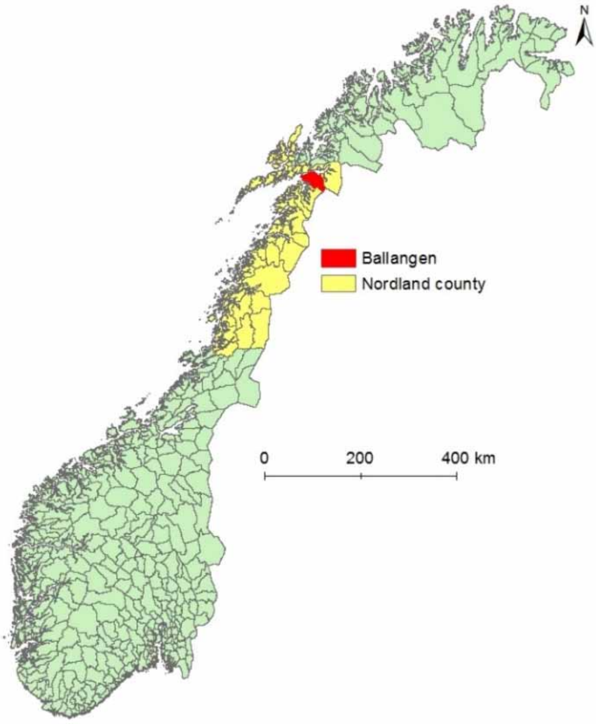

Figure 1 | Location of Old Ballangen municipality in Norway.

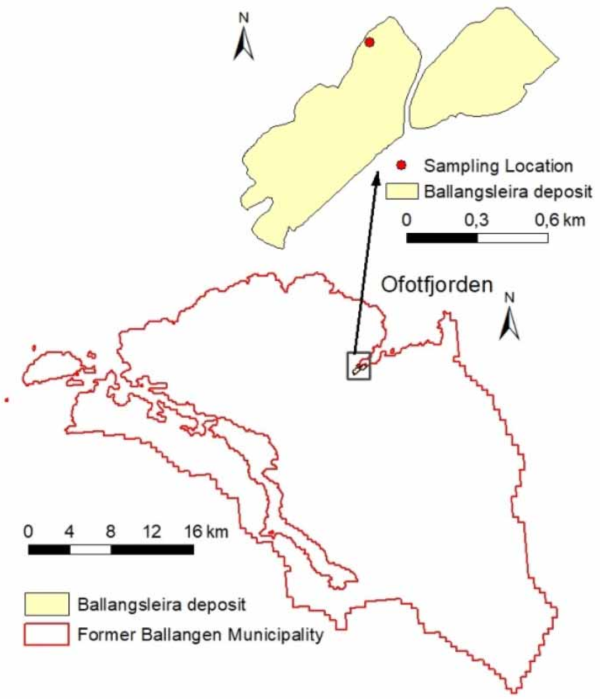

Figure 2 | Tailings deposits and sampling location.

Chemical analysis of sludge

The sewage sludge was collected from a local wastewater treatment plant at Narvik municipality. The sewage sludge was transported to the laboratory and dried under room temperature to decrease the water content first. Chemical composition was determined by X-ray fluorescence (XRF) analysis (Panalytical Axios Max).

Prior to the analysis, samples were dried at 60 °C for 24 hours, ground to a fine powder, mixed with a binder (Hoechst wax C, 6%), and pressed into a pellet.

Column experiment

Wet unoxidized tailings were fed into columns with 10 cm diameter and 40 cm length. A 10 cm gap from the top of the column was set. The columns were put into four wine fridges that were set at a temperature of 5, 10, 14, and 18 °C. There were four columns in total. 600 mL of deionized water (pre-cooled to the specific column temperature) was added to the columns from the top with a shower every second week. The addition of water volume was calculated based on the diameter of the column and a monthly precipitation of 80 mm. Leachates were collected at the bottom after 24 hours of water addition. The collected leachates were analyzed for the pH, salinity, alkalinity and concentrations of sulfate, Co, Fe, Mn, Ni, and Zn. After the leachate collection, the same amount of deionized water was added to the column again after 2 weeks. The leachate was collected at the bottom until at least 24 hours after the water addition. The same process was repeated.

In order to investigate the effect of digested sewage sludge on the reduction of leaching of contaminants from the tailings, a thin cover of 3 cm of digested sewage sludge from a wastewater treatment plant at Narvik community was added to the columns above the tailings after 16 leaching cycles. The same leaching procedure was continued afterwards. In total, 21 leaching cycles were performed.

RESULTS AND DISCUSSION

After the end of the experiments, the tailings columns were taken apart. The internal part showed iron oxides/hydroxides in part of the tailings, and other areas were dark gray as the tailings were before the start of the experiments. Figure 3 shows the column run at 5 °C a few weeks after the leaching experiment started and at the end of the leaching experiment. This indicates a preferential flow through the tailings.

Sludge composition

The chemical composition of the sludge is shown in Table 1. The sludge has high organic content. Therefore, it can be a good organic barrier to consume the oxygen and prevent the oxygen from reaching the tailings underneath. The sludge has a high concentration of nutrients, such as Na, Mg, Al, P, K, and Ca, which indicates that in addition to preventing the penetration of oxygen to underlying tailings, the sludge also works as a substrate to support the growth of vegetation afterwards. The concentrations of metals in the sludge only account for a small percentage of the mass, that is, the concentrations of Cr, Cu, Mn, Ni, and Zn are all lower than 0.05%. This indicates that the addition of the sludge cover will not add extra metals to the underlying tailings. However, the sludge had a relatively high Fe content, which might affect the concentration of Fe in the leachate.

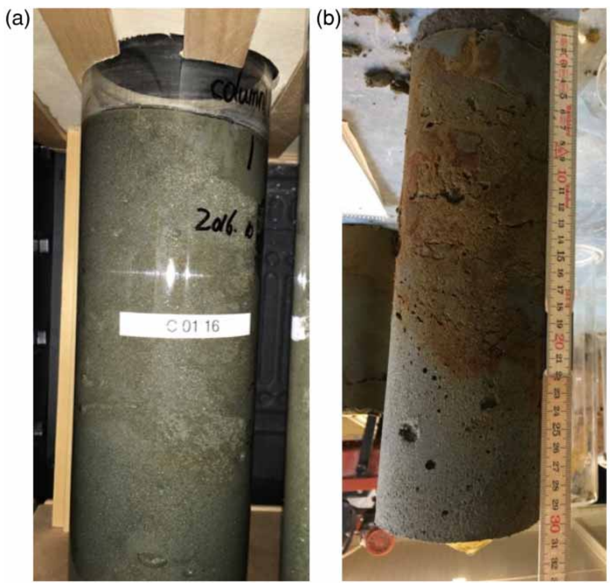

Figure 3 | (a) The column run at 5 °C a few weeks after the leaching experiment had started and (b) the same column at the end of the leaching experiment (taken out of the acryl-plastic column). The column has a 2 cm layer of sludge material on top.

Table 1 | Chemical composition of the sludge

Element	Concentration (%)	Element	Concentration (%)
CO_2	66.89	Cr_2O_3	0.01
Na_2O	0.45	MnO	0.06
MgO	2.02	Fe_2O_3	7.26
Al_2O_3	3.09	NiO	0.006
SiO_2	9.12	CuO	0.016
P_2O_5	2.99	ZnO	0.039
SO_3	1.63	Ga_2O_3	0.001
Cl	0.08	Br	0.005
K_2O	0.63	Rb_2O	0.002
CaO	5.41	SrO	0.012
TiO_2	0.26	Y_2O_3	0.001
V_2O_5	0.01	BaO	0.016

The evolvement of different parameters before sludge addition

The leaching results before the sludge cover addition are shown in Figures 4–6. The salinity showed a strong decrease during the first four leaching cycles. After the fourth leaching cycle, the salinity maintains a relatively stable level until the sludge cover addition. The strong leaching during the first cycles indicates leaching of easily soluble secondary minerals. The salinity is lowest at leaching temperature of 5 °C compared with that at other leaching temperatures. This indicates less oxidation of the tailings at low temperatures, which leads to low release of salts to the leaching solutions.

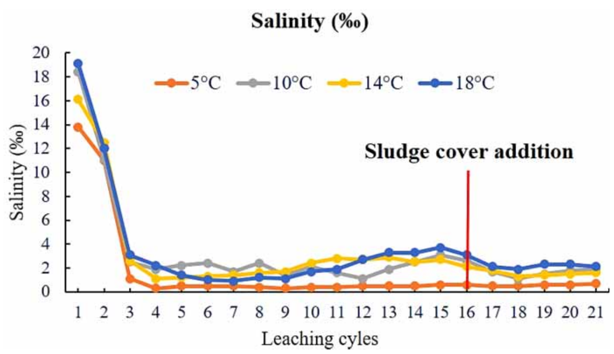

Figure 4 | Changes of salinity before and after the addition of the sludge cover (the 17th cycle and onwards had the sludge cover).

The pH generally showed a decreasing trend throughout the leaching, except for an increase in the first three leaching cycles. The pH of the leachate was highest at leaching temperature of 5 °C before the sludge cover addition, which corresponds to the lowest salinity at this leaching temperature. This indicates low oxidation of the tailings at this leaching temperature. As leaching goes on, the oxidation of tailings without any remediation measures may occur even at a low temperature. The pH of the leachate at 18 °C was generally lowest in the first 12 leaching cycles. After the 12th leaching cycle, the pH of the leachate started to increase. The three columns operated at 10, 14, and 18 °C showed pH levels similar to the column study of Embile *et al.* (2018, 2019) that was done at around 20 °C.

The concentration of SO_4^{2-} in the leachate was lowest at a leaching temperature of 5 °C, which corresponds to the high pH at this temperature. The highest concentration was observed in the first three leaching cycles. Afterwards, it decreased significantly to low levels and remained at relatively low levels thereafter. The same phenomenon was observed at 10, 14, and 18 °C. The highest level was observed at the first two leaching

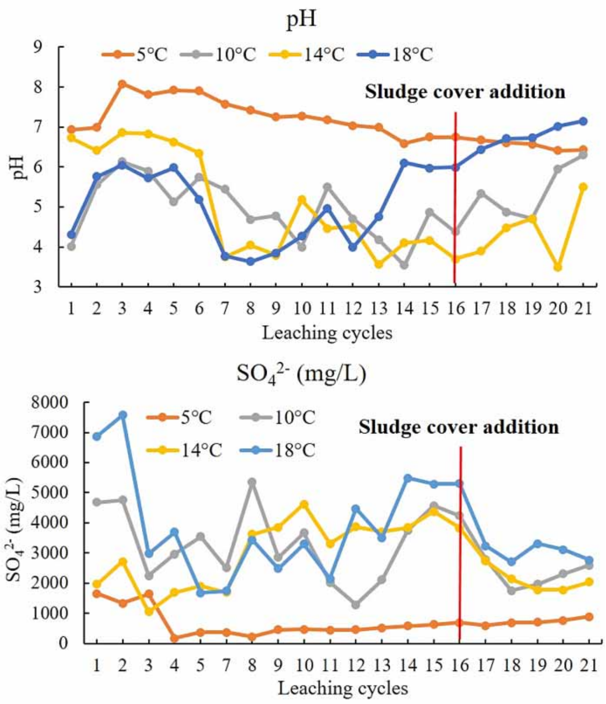

Figure 5 | Changes of pH and SO_4^{2-} before and after the addition of the sludge cover (the 17th cycle and onwards had the sludge cover).

cycles, which decreased significantly afterwards. After the third leaching cycle, the concentration of SO_4^{2-} in the leachate fluctuated. This fluctuation may be due to the variable flow path through the tailings.

The concentrations of Fe and Mn in the leachate were the lowest at the leaching temperature of 5 °C compared with those at other leaching temperatures. This corresponds to the high pH and low sulfate concentration in leachates at this leaching temperature. Their concentrations showed an obvious decrease in the first several leaching cycles and a constant increase afterwards, which was more obvious for Fe. This corresponds to the slight increase of the pH in the first several leaching cycles and decrease afterwards.

For the highest concentrations, there is considerable variation between different temperatures. For Fe, the highest concentration was observed at 18 °C in the first five leaching cycles. The concentration of Fe decreased significantly afterwards. The highest concentration was observed at 10 °C from the 6th to 16th leaching cycle before the sludge cover addition and, to a large extent, corresponds with the lower pH. This may indicate that 10 °C is an important threshold temperature for the leaching of Fe. However, flow paths' variations between the columns may also be an important factor in the oxidation rate.

The concentration of Mn in the leachate was highest at a leaching temperature of 18 °C in the first three leaching cycles. From the 4th to 7th leaching cycle, the highest concentrations were observed at 10 °C. From the 8th to 12th leaching cycle, the highest concentrations shifted to a leaching temperature of 14 °C. Then, in the last four leaching cycles (13th to 16th) before the sludge addition, the highest leaching concentrations were observed at 18 °C again. The big variation throughout the leaching indicates that the leaching of Mn is a complex process among several factors. Both pH and redox conditions can play a part in mobilizing and stabilizing Mn that may be interacting results of many different factors.

For Zn, the highest concentration was observed at 10 °C in the first ten leaching cycles. From the 11th to 15th leaching cycle, the highest leaching concentration was observed at 14 °C. The 5 °C and 14 °C columns had a higher Zn leaching rate after the 15th leaching cycle than in the beginning, while 18 °C and 10 °C both showed the opposite. This indicates that 10 °C might be an important temperature for the leaching of Zn.

Ni and Co showed a similar leaching phenomenon throughout the leaching. Their concentrations were highest at a leaching temperature of 10 °C in the first eight leaching cycles. From the 9th to 13th leaching cycle, their highest concentrations shifted to 14 °C. In the last three leaching cycles (14th to 16th) before the sludge cover addition, the highest concentration shifted back to 10 °C again.

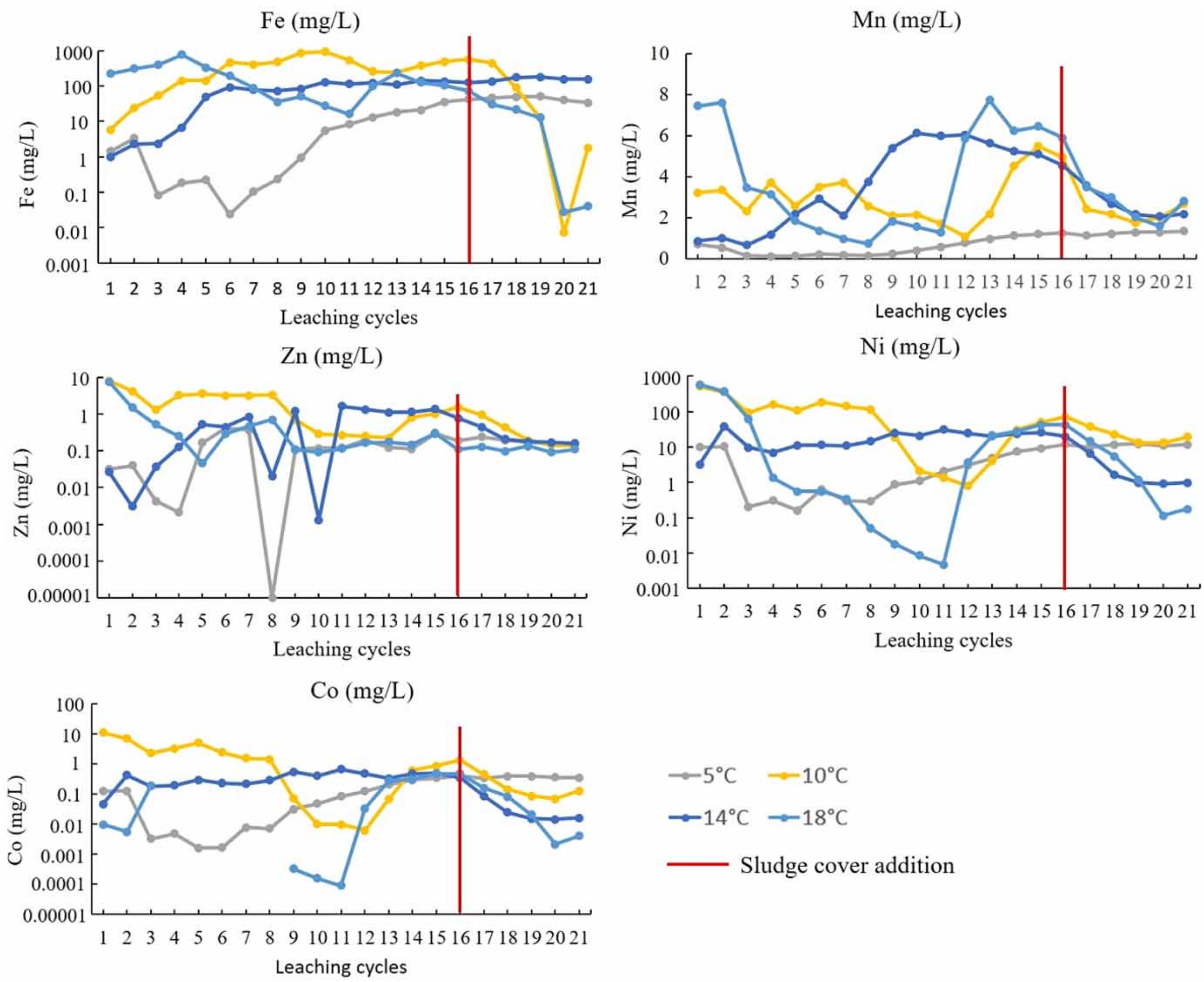

Figure 6 | Changes of concentrations of Fe, Mn, Ni, Zn, and Co in the leachate before and after the sludge cover addition (the 17th cycle and onwards had the sludge cover).

The efficiency of sludge cover on the leaching

The leaching results were compared before and after the sludge cover addition and are shown in Figures 4–6.

The effectiveness of sludge cover on the pH and salinity of the leachate

The salinity of the leachate showed a slight decreasing trend at 10, 14, and 18 °C leaching temperatures, but remained at a constant level at 5 °C.

The pH of the leachate at 5 °C already showed a decreasing trend before the sludge cover addition. However, after the addition of the sludge cover, the decreasing speed in pH slowed down. However, the pH of the leachate under leaching temperatures of 10, 14, and 18 °C showed an increasing trend after the sludge cover addition. This indicates that the addition of the sludge cover probably slowed down the weathering and oxidation process of the underlying tailings.

The effectiveness of sludge cover on the concentration of SO_4^{2-} in the leachate

The concentration of SO_4^{2-} in the leachate remained relatively constant after the sludge cover addition at a leaching temperature of 5 °C. However, an obvious decrease in sulfate concentration was observed at leaching temperatures of 10, 14, and 18 °C. The results correspond to the pH change, which indicated the sludge cover addition slowed down the weathering process of the underlying tailings, and therefore lowered the concentration of SO_4^{2-} in the leachate.

The effectiveness of sludge cover on the concentrations of Co, Fe, Mn, Ni, and Zn in the leachate

The concentrations of Co, Mn, and Ni in the leachates showed a decreasing trend after the sludge cover addition at leaching temperatures of 10, 14, and 18 °C, while the column operated at 5 °C showed relatively constant concentrations for these elements. The concentration of Fe in the leachate decreased significantly at leaching temperatures of 10 and 18 °C. The concentration of Zn in the leachate decreased slightly at 10 and 14 °C after the sludge cover addition. This indicates that the addition of sludge cover slowed down the leaching of contaminants from the tailings deposit, especially at a relatively high temperature. It seems that 10 °C is a threshold temperature, below which the leaching is not affected by the cover addition so much. At a leaching temperature higher than 10 °C, the sludge cover addition can reduce the leaching of elements significantly. Water quality data also confirm that the used sewage sludge did not contain leachable metals (Mn, Zn, Ni, Co). Several other studies have reached similar results, which show that sludge is an efficient cover that can reduce the leaching of metals from tailings and will not add extra metals to the leachates (Lu *et al.* 2013; Nason 2013; Nason *et al.* 2013). Sludge quality is an important factor and sometimes can result in leaching of metals (Andrés & Francisco 2008). Therefore, before use of the sludge as a cover over tailings, a thorough investigation of the sludge is needed and necessary.

CONCLUSIONS

The addition of sludge cover slowed down the oxidation of the underlying tailings and leaching of SO_4^{2-}, Fe, Mn, Co, Ni, and Zn from the tailings, especially at relatively high temperatures. 10 °C is a threshold temperature, below which the leaching is not affected by the cover addition so much. At a leaching temperature higher than 10 °C, the sludge cover addition can reduce the leaching of elements significantly. When a dry cover technique is used as a mitigation measure for mine waste management, local temperature change should be taken into account.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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