THE EFFECT OF TEMPERATURE AND PRECIPITATION ON THE LEACHING OF CONTAMINANTS FROM BALLANGEN TAILINGS DEPOSIT, NORWAY

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ABSTRACT
Leaching and transport of contaminants from tailings deposit is a highly complex interacting system affected by a suite of environmental factors, among which temperature and precipitation rates linked to climate change are two important parameters. The climate change in the Nordic region is more drastic than other regions. In this study, a laboratory batch leaching experiment was performed on the tailings deposited in Ballangen, northern Norway to investigate the impact of temperature and precipitation change on the leaching of contaminants from the tailings. The leaching was conducted at four different temperatures 5°C, 10°C, 15°C and 20°C and two precipitation rates 8mm/week and 20mm/week. In total, six leaching cycles were performed, and the leachate was collected and tested the pH and concentrations of Cr, Cu, Fe, Ni, V and Zn. The pH of leachate is lowest at 20°C under both precipitation rates, which indicate more tailings oxidation at 20°C. Highest concentrations of Cr, Cu, Fe, Ni was found in the leachate at 20°C and 10ml leaching conditions. Whilst the highest concentrations of Cr, Cu, Fe, Zn showed in the leachate at 5°C under 4ml leaching condition. The accumulated leaching amounts of Cr, Cu, Fe, V and Zn are generally higher at a precipitation rate of 20mm/week than 8mm/week. The difference between these two precipitation rates increases as temperature increases and the highest difference is generally found at a temperature of 20°C. The results indicate that a combination of high temperature and high precipitation will increase the leaching of contaminants significantly.
Keywords: temperature, precipitation, batch leaching, contaminants, tailings.

1 INTRODUCTION

The extraction of metals and minerals can affect the natural environment to a significant extent both while active operations and after the operations have ceased. Large amounts of waste rock and tailings are produced during resource extraction and it is estimated that more than 90% of the extracted materials come to waste [1]. The global production of mine wastes has been estimated at 15000–20000 million ton per year [2]. A feasibility study in Sweden demonstrated that within 10–20 years the planned mines in Norrbotten will alone produce 100 M tons of tailings per year and almost similar amount of waste rock [3]. The proper handling of these wastes is one of the biggest environmental problems associated with mining and is of great importance for the sustainable development of mining industry. One major potential long-term environmental effect of these wastes is the formation of acid rock drainage (ARD) from sulphide-bearing mine waste, which can last for hundreds or even thousands of years after deposition [1]. In an acid generating reaction, the oxidation of sulphides usually leads to the release of sulphide-associated elements into aqueous solutions [4]. The leaching of contaminants will significantly degrade the environment, and the contaminants will further transport from the environmental medium to the ecosystem and the human beings living in the area in the long-term [5]–[8].

Leaching and transport of contaminants from tailings deposits is a highly complex interacting system affected by a suite of environmental factors, among which temperature and precipitation rates linked to climate change are two important parameters. Increased temperature will affect chemical reaction kinetics. Any step along the transport and

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redistribution pathways is influenced by climate change because chemical reactivity, adsorption and accumulation are temperature-dependent. With increased flows, there will be changes in stream power and sediments loads with the potential to alter the morphology of rivers and the transfer of sediments to lakes [9]. Climate change has great impact on the process of contaminant transport and concentrating process in the environment [10]. Climate change is expected to alter environmental distribution of contaminants and their bioaccumulation due to changes in transport, partitioning, and bioaccumulation process [11].

The environmental impacts from climate change in the Nordic region are expected to be more drastic than other regions [12]. The 2007 report from the Intergovernmental Panel on Climate Change (IPCC) identified the Nordic as the region on earth where the most drastic environmental impacts from climate change are expected [12]. Changes in global climate and the associated environmental changes in the Nordic region are expected to have significant consequences for contaminant pathways. Over the past 50 years, unprecedented rates of change in both temperature and precipitation have been recorded and future predictions agree with the current trends [13]. Therefore understanding the mechanism of how climate change affects the transportation of contaminants from mine waste deposits and how climate change affects the deposition and remobilization of contaminants is of great importance for the sustainable development of the Nordic region.

In this study, a laboratory batch leaching experiment was designed and performed on the tailings from Ballangen tailings deposit in Norland county, northern Norway. The leaching was conducted at four different temperatures and two different precipitation rates based on monthly average temperature and monthly average precipitation in the region. The leachate was collected and tested the pH and concentrations of Cr, Cu, Fe, Ni, V and Zn. The effect of temperature and precipitation change on tailings oxidation rate and contaminant leaching rate from the tailings was investigated based on the leaching results.

2 STUDY AREA

The nickel mine "Nickel and Olivine AS" in Ballangen municipality in Nordland county, Norway was in operation during the period 1988–2002. The sulphidic nickel ore contains about 0.5% Ni [14]. It has an ore production of about 700,000 tons annually. The tailings from the ore dressing plant were deposited in two deposits near the Ballangen fjord, the Fornes deposit and the Ballangsleira deposit as shown in Fig. 1 [14]. The Fornes deposit and the Ballangsleira deposit were decommissioned in 1991 and 2002 respectively [14]. At Ballangsleira deposit, the tailings from the Nickel and Olivine AS covered over the old sulphide-containing tailings from the closed Bjørkåsen mine as shown in Fig. 2 [15]. In total, about 7 million tons of tailings were deposited at Ballangsleira. After the tailings deposition ceased, the tailings was covered with a thin layer cover with 10–20cm of soil. The soil was seeded and fed fertilizer afterwards in order to establish a vegetation layer on the soil to avoid erosion of soils. Deposits at Ballangsleira covers an area of about 50 hectare. However, the thin soil layer is not effective in preventing the oxidation of the underlying tailings. Therefore, this study chose Ballangsleira deposit as the study area to investigate the leaching of contaminants from tailings.

3 METHODS

3.1 Tailings sampling and analysis

Both un-oxidized and oxidized tailings from Ballangsleira deposit were collected into several polyethylene plastic bags with plastic spade in July 2016. Both tailings were stored at 4°C in

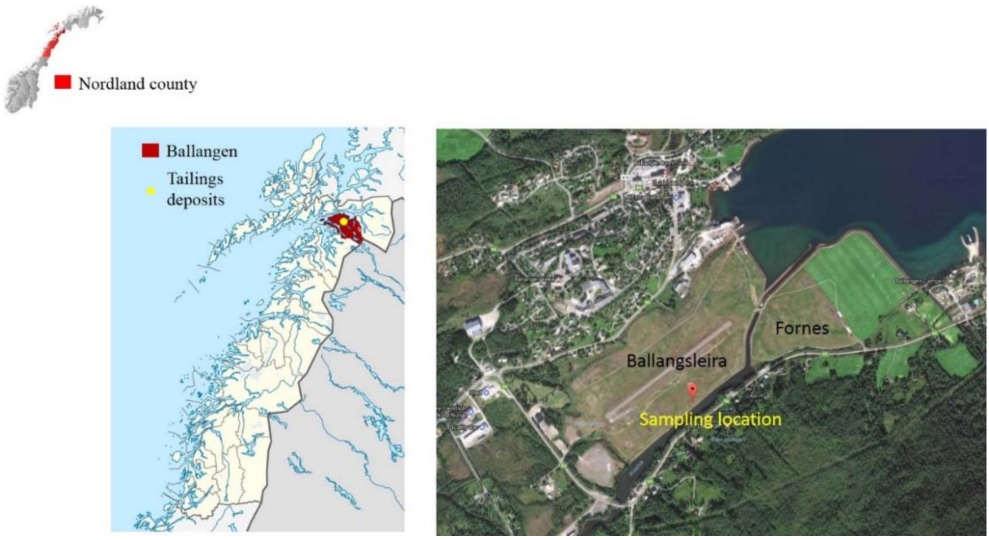

Figure 1: Tailings deposits and sampling location.

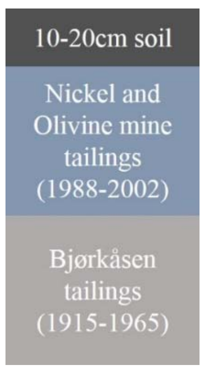

Figure 2: Tailings deposit at Ballangsleira.

the refrigerator until analysis. The samples were sent to ALS Laboratory Group AS for elemental analysis. The EPA modified methods 200.7 and 200.8 were used for elemental analysis. The total solid content was determined by drying the tailings at 105°C according to the Swedish standard SS 028113. The dried samples were dissolved by smelting with $LiBO_2$ and dissolved in 1:1 HNO_3 (1 ultrapure nitric acid and 1 water) according to the ASTM 3682. The digestion solution was analyzed the concentration of elements with ICP-AES and ICP-MS.

3.2 Grass sampling and analysis

To investigate the leaching of metals to the surrounding environment, two grass samples were collected into polyethylene plastic bags. One is from the Ballangsleira deposit and one is from a reference area without tailings deposition. The samples were sent to ALS Laboratory

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Group AS for elemental analysis. The samples were digested with nitric acid and H_2O_2 in the microwave oven first. The digestion solution was analyzed the concentrations of elements with ICP-SFMS according to the modified EPA methods 200.7 and 200.8.

3.3 Laboratory batch leaching experiment

The column experiment method is complex and expensive. In recent years, some researchers have indicated that the low-cost and simple batch experiment method can substitute the column experiment method in studying the release of heavy metals from tailings [16]. To investigate the leaching of contaminants from tailings at different precipitation rates and different temperatures, a small-scale laboratory batch leaching experiment was designed and performed on the oxidized tailings. 10g tailings was weighed and added to a 50ml centrifuge tube. 4 and 10ml of deionized water was added to the tubes and capped respectively. The addition of water was based on a precipitation rate of 8mm/week and 20mm/week. The leaching after every second times are equal to that after one week in reality. The tubes were then put into four different incubators. The incubators were set at temperature of 5, 10, 15 and 20°C respectively. There are 8 tubes in total in the experiment. The tubes were let mixing completely inside the incubator with a shaking speed of 150 rpm for about 48 hours. They were then taken out from the incubator and centrifuged in a centrifuge. The leaching solution were collected from the top into a 20ml tube and stored at 4°C in the storage room until analysis. A new identical amount of deionized water was added to the tube, capped and put into the incubator again. The same process was repeated. For the tube added 4 ml water each time, the water was collected every second time due to limited amount of water. In total, six leaching cycles were performed. However, for the tubes with 4ml water addition, only 3 leachates were collected since the leachate is collected every second time. The collected leachates were tested pH and sent to ALS Laboratory Group AS for analysis of Cr, Cu, Fe, Ni, V and Zn. The samples were acidified first with 1ml nitric acid per 100ml water samples and then analyzed with ICP-AES according to the EPA methods 200.7.

4 RESULTS

4.1 Elemental composition of tailings

The elemental compositions of the un-oxidized tailings and oxidized tailings were shown in Table 1. From Table 1, we can see that the concentrations of Co, Cr and Ni are much higher in the oxidized tailings than in the un-oxidized tailings. However the concentrations of Al, Ca, K, Na, Ba are higher in the unoxidized tailings. This showed that the oxidation of tailings will lead to the leaching of Co, Cr and Ni to the surrounding environment. As the tailings oxidize, the buffering minerals such as Al_2O_3, CaO, K_2O, Na_2O, and Ba oxides will be consumed by the generated acid, which leads to the lower concentration of these elements in the oxidized tailings.

4.2 Elemental composition of grass samples

The elemental composition of grass samples was shown in Table 2. The concentrations of As, Co, Cu, Ni were higher in the grass sample from Ballangen than that from the reference area without tailings deposition. The concentration of these elements in the grass from Ballangen is between one to five times of that in the grass from the reference area. This indicates that the deposition of tailings has led to the leaching of these elements from tailings to the surrounding environmental mediums.

Table 1: Elemental compositions of the tailings [17].

Element	Unit	Unoxidized tailings	Oxidized tailings
Total solid	%	81.5	88.2
SiO_2	% TS	39.5	40.1
Al_2O_3	% TS	4.47	3.95
CaO	% TS	3.18	2.22
Fe_2O_3	% TS	17.3	14.2
K_2O	% TS	0.417	0.211
MgO	% TS	27	31.3
MnO	% TS	0.165	0.149
Na_2O	% TS	0.619	0.481
Ba	mg/kg TS	65.8	43.9
Co	mg/kg TS	38	83.1
Cr	mg/kg TS	820	1410
Ni	mg/kg TS	77.8	476
Cu	mg/kg TS	144	81.3
Zn	mg/kg TS	144	60.1

Table 2: Elemental compositions of grass samples.

Element	Unit	Grass from Ballangen	Grass from reference area
As	mg/kg	0.111	<0.08
Cd	mg/kg	0.0248	0.0261
Co	mg/kg	0.421	0.147
Cr	mg/kg	0.0446	0.0785
Cu	mg/kg	19.9	7.14
Mn	mg/kg	106	152
Ni	mg/kg	7.43	1.53
Zn	mg/kg	38.6	43.5

4.3 Batch leaching results

4.3.1 pH in the leachate

The pH evolvement in the leachate at different temperatures were shown in Fig. 3. From this figure, we can see that the pH of the leachate is lowest at a temperature of 20°C under both 4ml and 10ml leaching conditions. The pH of the leachate at 10ml leaching is slightly over 5.8 in the first leaching cycle and continuously decrease afterwards and has slight increase again in the 6 leaching cycle. The pH of the leachate at 4ml leaching is around 5.8 in all the leaching solutions at 20°C. In general, the pH is under 6 in all the six leaching cycles under 20°C leaching condition. This indicates that more oxidation of tailings is ongoing in the tube at 20°C leaching condition compared with the leaching at other temperatures.

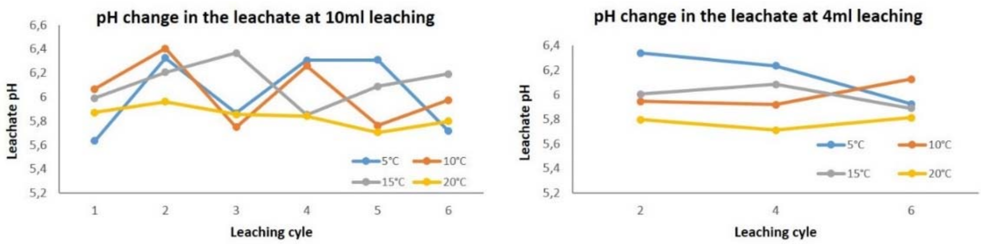

Figure 3: Changes in leachate pH at 4ml and 10ml leaching.

4.3.2 Concentrations of Cr, Cu, Fe, Ni, Zn in the leachate under 10ml leaching

Concentrations of Cr, Cu, Fe, Ni, Zn in the leachate under 10ml leaching were shown in Fig. 4. From Fig. 4, we can see that concentrations of Cr, Cu, Fe, Ni were highest under 20°C leaching conditions at 10ml leaching condition, whereas concentration of Zn was highest under 10°C leaching conditions. Concentrations of Cr, Cu, Fe were highest in the leachate of the fourth leaching cycle. Concentrations of Ni and Zn were highest in the leachate of the second leaching cycle. This indicate that a temperature increase from 5°C to 20°C will increase the leaching of most tested metals Cr, Cu, Fe, Ni. However, the highest leaching of Zn under 10°C indicates that Zn may have a different leaching mechanism than the other elements.

4.3.3 Concentrations of Cr, Cu, Fe, Ni, Zn in the leachate under 4ml leaching

Concentrations of Cr, Cu, Fe, Ni, Zn in the leachate under 4ml leaching were shown in Fig. 5. From Fig. 5, we can see that concentrations of Cr, Cu, Fe, Zn were highest under 5°C and 4ml leaching conditions, whereas concentration of Ni was highest under 20°C leaching condition. Concentrations of Cr, Cu, Fe were highest in the leachate of the fourth leaching cycle and concentrations of Ni and Zn were highest in the leachate of the second leaching cycle, which is the same as that under 10ml leaching. It showed different results from that under 10ml leaching.

4.3.4 Effect of precipitation on the leaching of elements

The accumulated mass of elements that are leached out from tailings under a precipitation of 8mm/week and 20mm/week was calculated and compared. The mass of elements that are leached out after 3 weeks' precipitation for Cr, Cu, Fe, Ni, V, Zn was calculated, which is the number of elements that is leached out after six leaching cycles.

4.3.4.1 Effects of precipitation on the leaching of Fe

The accumulated mass of Fe that are leached out from the tailings at different temperatures after 3 weeks under a precipitation of 8mm/week and 20mm/week was calculated and showed in Fig. 6. The accumulated mass of Fe leached out are generally higher at 20mm/week's precipitation at 10°C, 15°C and 20°C. The difference between 8mm/week and 20mm/week is the most significant at 20°C. This indicates that a combination of high precipitation and high temperature can significantly increase the total amount of Fe that are leached out. However, the accumulated mass of Fe at 5°C seems higher at 4mm/week's precipitation than 10mm/week's precipitation. This may indicate that at 5°C low temperature, the reaction rate is low. At high precipitation, the high flushing of water does not have enough time to dissolve the Fe in the tailings, which lead to low concentration of element.

4.3.4.2 Effects of precipitation on the leaching of Cr

The accumulated mass of Cr leached out are higher at 20mm/week's precipitation than 8mm/week's precipitation under all leaching temperatures. The difference increases as the leaching temperature increases from 5°C to 10°C, 15°C and 20°C. The difference is the most significant at 20°C. The same as Fe, it indicates that a combination of high temperature and high precipitation can significantly increase the mass of Cr that are leached out from tailings.

4.3.4.3 Effects of precipitation on the leaching of Cu

In general, the accumulated mass of Cu that are leached out are higher at a precipitation of 20mm/week than 8mm/week (Fig. 8). The difference between the two precipitation rates increases as temperature increases, with the highest difference found at 20°C. However, the difference is not as significant as that of Fe and Cr (Figs 6–8).

4.3.4.4 Effects of precipitation on the leaching of Ni

The accumulated mass of Ni that are leached out from the tailings at different temperatures after 3 weeks was calculated and showed in Fig. 9. In contrary to other elements, the effect of precipitation on the leaching of Ni are not significant at all the leaching temperatures (Fig. 8). The accumulated mass of Ni leached out at 5°C and 10°C is slightly higher at a

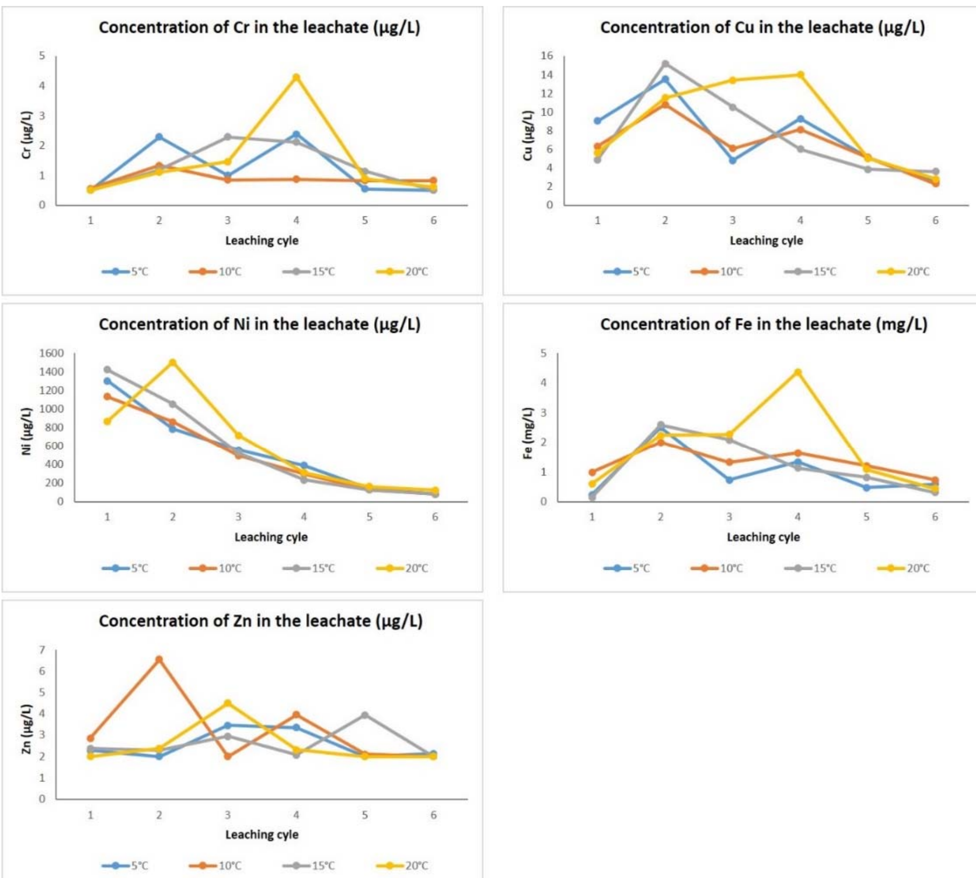

Figure 4: Concentrations of Cr, Cu, Fe, Ni, Zn in the leachate under 10ml leaching.

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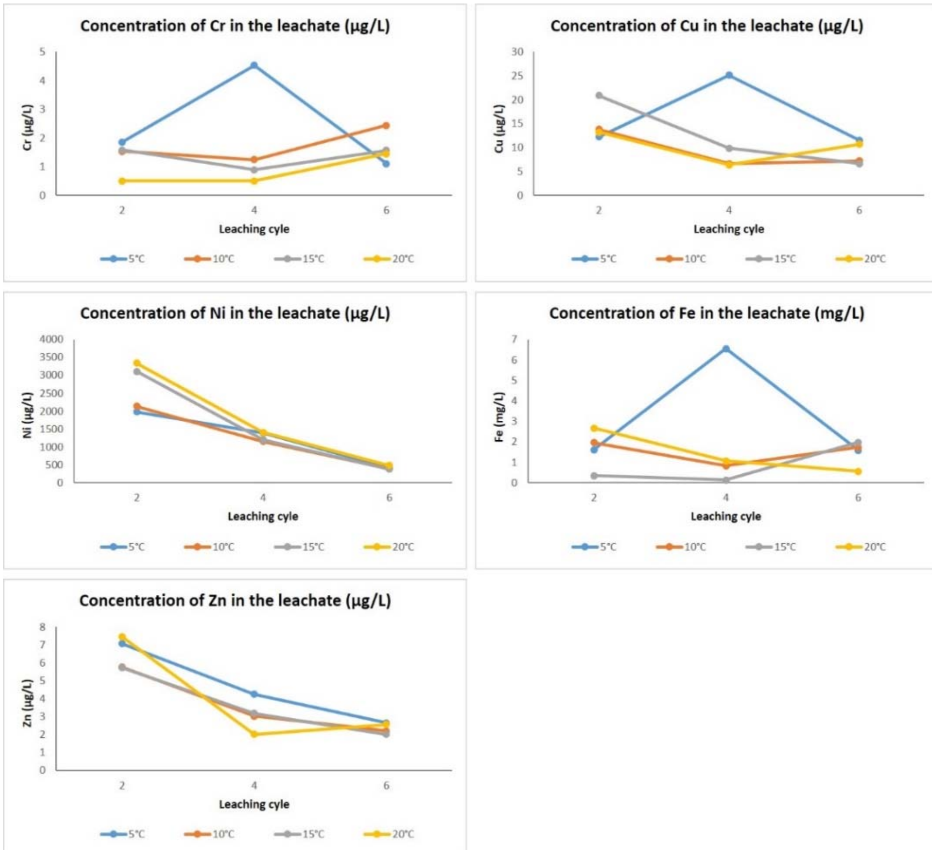

Figure 5: Concentrations of Cr, Cu, Fe, Ni, Zn in the leachate under 4 ml leaching.

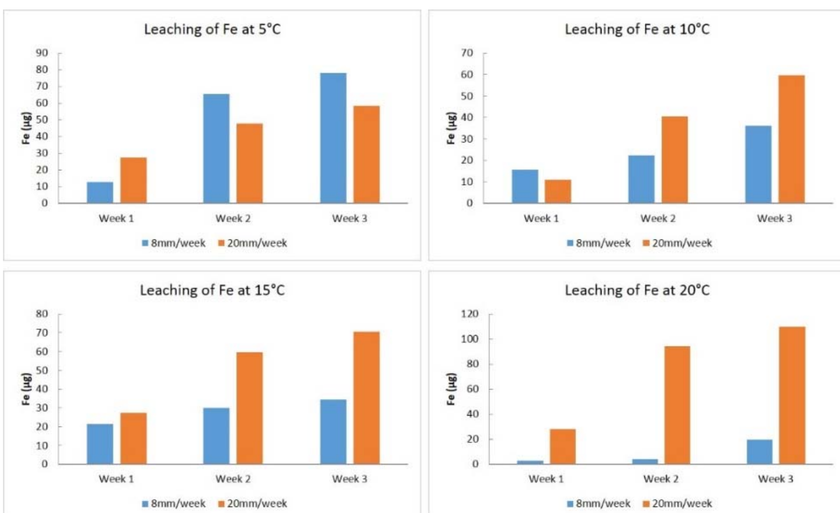

Figure 6: Effects of precipitation on the leaching of Fe.

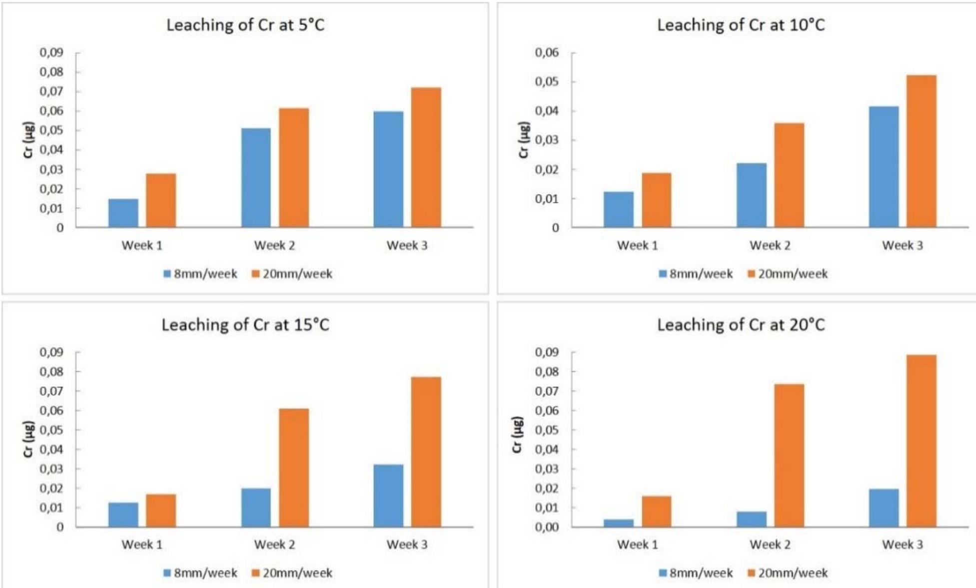

Figure 7: Effects of precipitation on the leaching of Cr.

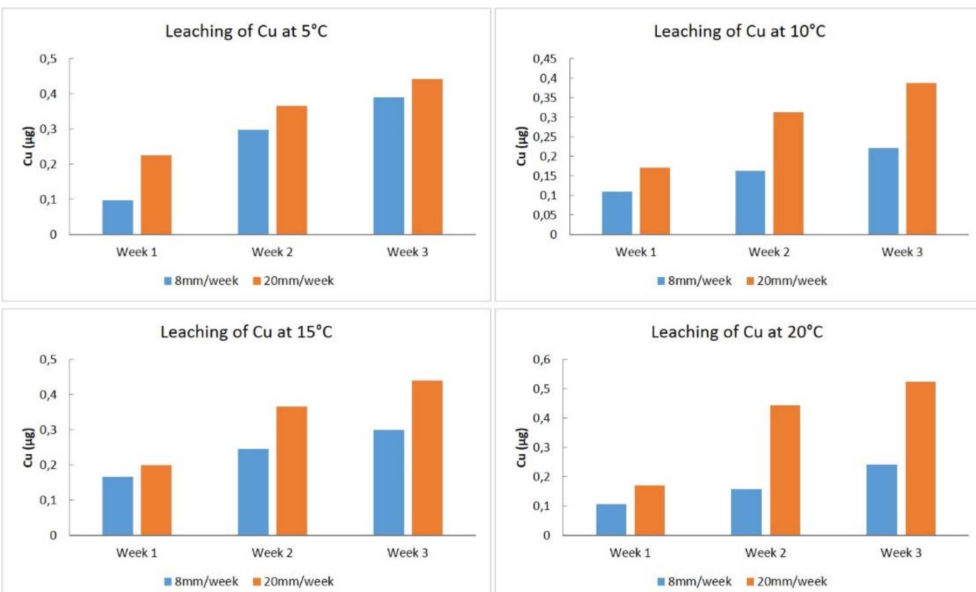

Figure 8: Effects of precipitation on the leaching of Cr.

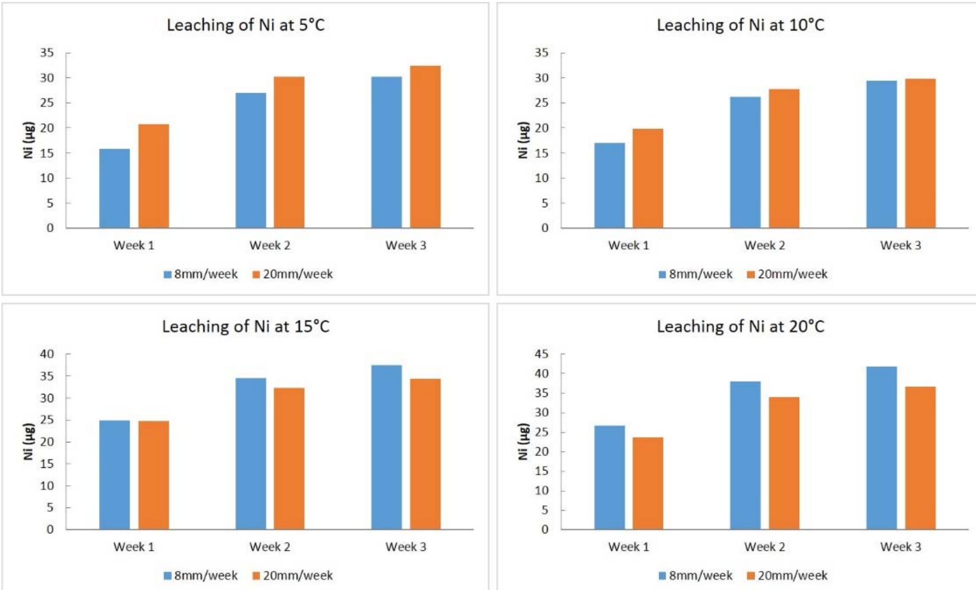

Figure 9: Effects of precipitation on the leaching of Ni.

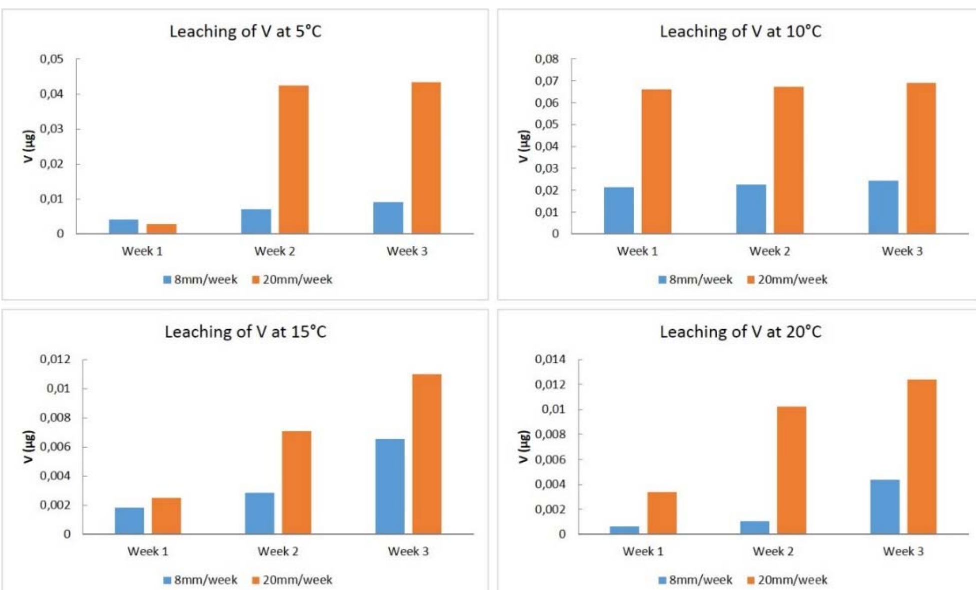

Figure 10: Effects of precipitation on the leaching of V.

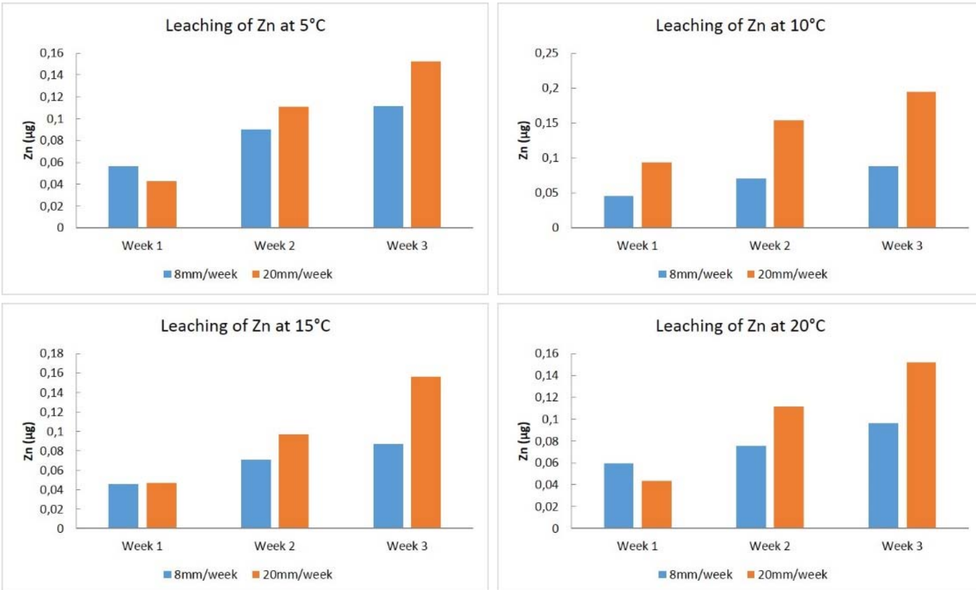

Figure 11: Effects of precipitation on the leaching of Zn.

precipitation of 20mm/week. However, the accumulated mass of Ni leached out at 15°C and 20°C is generally slightly lower at a precipitation of 20mm/week.

4.3.4.5 Effects of precipitation on the leaching of V
The accumulated mass of V that are leached out from the tailings at different temperatures under a precipitation of 8mm/week and 20mm/week was shown in Fig. 10. In general, the accumulated mass of V that are leached out are higher at a precipitation of 20mm/week than 8mm/week (Fig. 10), which showed similar trend as that of Cu (Fig. 8).

4.3.4.6 Effects of precipitation on the leaching of Zn
The accumulated mass of Zn that are leached out from the tailings at different temperatures after 3 weeks was calculated and showed in Fig. 11. In general, the accumulated mass of Zn that are leached out are higher at a precipitation of 20mm/week than 8mm/week (Fig. 11), which showed similar trend as that of Cu and V (Figs 8 and 10).

5 DISCUSSIONS

5.1 The effect of temperature on the weathering of tailings

The pH of leaching solution is lowest at 20°C under both 4ml and 10ml leaching conditions, which indicates that an increase in temperature will lead to more oxidation of the tailings and thus lower pH of the leaching solution. The effect of temperature on the oxidation rates of sulphides in tailings has been discussed in many studies [18]. The overall oxidation of pyrite is temperature-dependent and both chemical and biological pyrite oxidation rates decrease with decreasing temperatures as described by the Arrhenius equation [18]. The temperature coefficient, or Q_{10}, is widely used to describe the proportional change in rate, given a 10° change in temperature, and it is reported to be between 2 and 3 for pyrite oxidation for

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temperatures between 3 and 25°C [18]. The rate of acid production is controlled by the availability of oxygen at the sulphide surface [19]. A steady oxygen supply to the surface of pyrite grains is a prerequisite for oxidation in both frozen and unfrozen mine tailings [18]. High temperature will increase the free movement of oxygen through the tailings, thus increase the availability of oxygen for pyrite oxidation. The rate of both chemical and biological oxidation of pyrite are affected by temperature [20]. The temperature dependence of the oxidation rate follows the Arrhenius equation [20]. From least square fit of experimental data, Nicholson calculated an activation energy E_a of 88 kJ mol^{-1} which implies that the rate of oxidation approximately triples for a 10°C rise in temperature [20].

The influence of temperature on biological ferrous iron oxidation by Thiobacillus ferrooxidans is similar to that of chemical oxidation. In the temperature range of 3 to 24°C, the biological rate is approximately doubled for every 10°C rise in temperature [20].

5.2 The effect of temperature on the leaching of contaminants from tailings

Cr, Cu, Fe, Ni showed highest concentrations in the leachate at 20°C and 10ml leaching conditions. There are other studies, which also showed the increased leaching of elements with temperatures. The released amounts of Cu, Fe, Mn, Ni and Zn from the pyrite tailings at Sichuan province, China, at a temperature range of 15–45°C were investigated in a laboratory batch experiment, and showed the released amount of each metal had an increasing trend as temperature increases [16]. Temperature had a significant effect on the release of Fe, Cu, Mn and Ni from the investigated pyrite tailings and high temperature can significantly promote the release of Fe, Cu, Mn and Ni from the pyrite tailings [16]. The release characteristic caused by temperature is related to the solubility product constant K_{sp}, which is positively correlated with temperature [16]. The effect of temperature on the leaching of copper from a sulphide tailings were investigated, and highest degree of copper leaching was achieved at the highest experiment temperature of 80°C [21]. However their experiment temperatures are much higher than the experiment in this study [21]. The sulphide oxidation rate consists of a biological and an abiological component [22]. In our study, the reaction should be more dominated by abiological process since the sulphide concentration of the tailings is low and the pH of the leachate is around 6 generally. Increase of temperature will increase the abiological surficial reaction rate [22].

5.3 The effect of precipitation on the leaching of contaminants from tailings

The accumulated leaching amounts of Cr, Cu, Fe, V and Zn are generally higher at a precipitation rate of 20mm/week than 8mm/week. The difference between the two precipitation rates increases as temperature increases and the highest difference is generally found at a temperature of 20°C. There are also previous studies that showed the increased leaching of contaminants as precipitation changes. An investigate on the pyrite tailings in Sichuan, China found that high LS ratio significantly promoted Fe, Cu, Mn, Ni and Zn release from the pyrite tailings [16]. The leaching of Zn from refractory zinc silicates in alkaline conditions was studied and the results showed that L/S ratio had a significant effect on the leaching of Zn and the dissolution of Zn increases with increasing L/S ratio when the L/S ratio below 10 [23]. The L/S ratio in this study was much lower than 10, and thus the results are comparable. The leaching behavior of metals from mineral processing waste was investigated in both batch and column leaching experiments [24]. The results of both batch and column experiment showed that the release of As and Se increased with the L/S ratio, while the release of Cu and Zn increased initially and tapered towards equilibrium values at high L/S ratio [24]. While the L/S ratio used in this study fell well below the equilibrium values of that study. Therefore, the results in this study are comparable with the results from that study.

5.4 A combination of temperature and precipitation effect on the leaching of contaminants from tailings

Cr, Cu, Fe, Ni showed highest concentrations in the leachate at 20°C and 10ml leaching conditions. Whilst the concentrations of Cr, Cu, Fe, Zn in the leachate were highest in the leachate at 5°C under 4ml leaching condition. This showed that leaching of contaminants was affected by the combination of temperature and precipitation. At too low temperature such as 5°C, the reaction rate is low. If the precipitation rate is high, the water doesn't have enough time to dissolve the metals in the tailings. Therefore, under 4ml leaching condition, the highest concentrations of Cr, Cu, Fe, Zn were shown in the leachate at 5°C. At high temperature of 20°C in this study, the reaction rate is high. It needs more water to dissolve the released elements. Therefore, a combination of high temperature and high precipitation can lead to high dissolution of metals from tailings.

6 CONCLUSIONS

In this study, a laboratory batch leaching experiment was performed on the tailings from Ballangen deposit in northern Norway. The leaching was conducted at four different temperatures 5, 10, 15 and 20°C and 2 precipitation rates of 8mm/week and 20mm/week. The leachate was collected for six leaching cycles and tested the pH and concentrations of Cr, Cu, Fe, Ni, V and Zn. The pH of leaching solution is lowest at 20°C under both precipitation rates, which indicates more oxidation and weathering of tailings at 20°C. Concentrations of Cr, Cu, Fe, Ni showed highest concentrations in the leachate at 20°C and 20mm/week leaching conditions. Whilst the concentrations of Cr, Cu, Fe, Zn in the leachate showed highest concentrations in the leachate at 5°C and 8mm/week leaching condition. The accumulated leaching amounts of Cr, Cu, Fe, V and Zn are generally higher at a precipitation rate of 20mm/week than 8mm/week. The difference between the two precipitation rates increases as temperature increases and the highest difference is generally found at a temperature of 20°C. The results indicate that a combination of high temperature and high precipitation will significantly increase the leaching of contaminants. If the global climate change continues in the near future, the temperature and precipitation will definitely increase especially in the Nordic region. This will have a significant effect on the contaminant leaching and transport from different environmental medium, not only from the mine waste deposits. Therefore, more study is needed to investigate the mechanism of temperature and precipitation change on the contaminant leaching and transport in the Nordic region.

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