Master's thesis

NTNU
Norwegian University of Science and Technology
Faculty of Natural Sciences
Department of Chemistry

Stine Steen

Treatment and monitoring of polluted leakage water from the former Killingdal concentration plant area in Trondheim.

Comparable studies of olivine and lime in a semi-scale water treatment system.

Master's thesis in Environmental Chemistry
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Abstract

Killingdal is an area in Trondheim where there was an active concentration plant from 1953 to 1986, which processed pyrite, sphalerite, and chalcopyrite. When mining activity stops, mining residue is left behind. Without proper cleaning, metals and metalloids can leach from the residue and end up in the surrounding environment. Despite clean-up efforts made in 2010/2011, the water that has filled up in the previous discharge, storage, and transport tunnel is acidic and heavily contaminated with metals and metalloids. When sulfide minerals like pyrite, sphalerite, and chalcopyrite come into contact with air and moisture, they become oxidized. This process leads to the release of metals and metalloids from the minerals, as well as acidification, and the resulting water is called acid mine drainage (AMD). The AMD inside the tunnel at Killingdal is seeping into the fjord and poses a serious threat to the aquatic ecosystems in the area.

This thesis describes the formation of AMD, as well as possible remediation strategies. It also focuses on the potential toxicity of the resulting contamination, and describes a classification system that predicts the toxic effects of the different elements in different concentrations. The system is divided into five classes from background levels (class I) to concentrations causing extensive toxic effects (class V).

The aim of this thesis was to investigate the treatment efficiency of olivine and lime on the elements Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As. Their ability to raise the pH of the AMD was also tested. The possibility of using aeration as a pre-treatment step was also explored, as well as the combination of olivine and activated carbon, an adsorbent being considered as a possible polishing step in another master thesis. In addition, the water inside the tunnel was monitored with varying regularity from October 2018 to February 2020, and the level of contamination in the water coming into the tunnel through the tunnel walls was also investigated.

Monitoring of the tunnel water showed high concentrations of particularly Cd, Cu, and Zn which were found in class V throughout the entire sampling period, and underlined the need for treatment of this water. High concentrations of Cu and Zn, and to some degree Cd, in the water entering the tunnel through the tunnel walls indicate that this water also needs to be treated before being released into the fjord.

Experiments with olivine treating the AMD from the tunnel showed between 96.9% and 100% efficiency for all of the elements except for Cr, which was released, and As, which was treated with between 63.9% and 95.6% efficiency. Olivine was found to increase the pH from 3.18 to 12.00. The high treatment efficiency of olivine resulted in Pb, As, Cd, and Ni concentrations in class II or I. Despite the high efficiency, Cu and Zn were still in class V or IV after treatment. Even though it was being released, Cr was found in class III. Olivine shows promising results as a potential treatment medium for the AMD at Killingdal, but the Cr release as well as its effect over time needs further investigation. A treatment system with olivine will probably be ameliorated if combined with other treatment steps.

Aeration was investigated as a possible pre-treatment step, but did not show any efficient metal removal, and it was concluded that aeration of the AMD at Killingdal does not have any benefits for the removal of metals through precipitation. The mixture of olivine and activated

carbon was investigated, and even though the treatment efficiencies of Cd, Pb, Fe, Ni, Cu, and Zn were high, they were not higher than olivine's treatment efficiency alone. In addition, the mixture of olivine and activated carbon increased the degree of Cr release, as well as releasing As. The mixture of olivine and activated carbon should not be considered as a treatment step for the AMD at Killingdal.

Experiments testing lime's treatment efficiency showed between 98.5% and 100% efficiency for removing Pb and Cr, and in one of the experiments 99.9% for Fe. When testing lime's efficiency over time it was found that this effect decreased over time. Lime raised the pH from 3.18 to 6.52 in one of the experiments.

Olivine was found to have high treatment efficiencies for Cd, Pb, Fe, Ni, Cu, and Zn, and in one experiment also for As, while lime was found to remove Cr and Pb, and in one experiment Fe, with high efficiencies. The treatment effect of lime on Cr in particular, should be explored and tested further in combination with olivine.

Sammendrag

Killingdal er et område i Trondheim hvor et tidligere flotasjonsverk som behandlet pyritt, sinkblende, og kobberpyritt var aktivt fra 1953 til 1986. Når gruveaktivitet stopper vil rester fra gruvedriften ligge igjen, og uten tilstrekkelig opprydding vil metaller og metalloider kunne lekke ut og ende opp i omgivelsene rundt. Til tross for oppryddingstiltak utført i 2010/2011, er vannet som har fylt seg opp i den tidligere lagringstunnelen svært forurenset med metaller og metalloider. Når sulfid mineraler som pyritt, sinkblende og kobberpyritt kommer i kontakt med luft og fuktighet vil de bli oksiderte. Denne prosessen fører til utlekking av metaller fra mineralene i tillegg til surgjøring av vannet, og resultatet kalles acid mine drainage (AMD). AMD lekker ut fra tunnelen i Killingdal til fjorden og utgjør en stor trussel for det akvatiske økosystemet i området.

Denne oppgaven beskriver dannelsen av AMD, i tillegg til mulige rensingsmetoder. Oppgaven fokuserer også på potensiell toksisitet av metall forurensningene i AMD, og beskriver et klassifikasjonssystem som sier noe om den toksiske effekten av ulike elementer i ulike konsentrasjoner. Systemet er inndelt i fem klasser fra bakgrunnsnivå (tilstrandsklasse I) til omfattende toksiske effekter (tilstandsklasse V).

Målet med denne oppgaven var å undersøke renseeffekten av olivin og kalk på elementene Cd, Pb, Cr, Fe, Ni, Cu, Zn og As. Deres evne til å øke pH i AMD ble også undersøkt. Muligheten for å bruke lufting som et for-rensetrinn ble også utforsket, i tillegg til en blanding av olivin og aktivt kull, en adsorbent som blir vurdert som et mulig poleringstrinn i en annen masteroppgave. I tillegg til dette ble vannet inne i tunnelen overvåket fra oktober 2018 til februar 2020, og metallkonsentrasjonen i vannet som kommer inn i tunnelen gjennom tunnelveggene ble undersøkt.

Overvåkingen av tunnelvannet viste høye metallkonsentrasjoner, spesielt for Cd, Cu og Zn som ble funnet i tilstandsklasse V gjennom hele overvåkningsperioden. Dette understreker behovet for rensing av vannet inne i tunnelen. Høye konsentrasjoner av Cu og Zn, og til en viss grad Cd, ble funnet i vannet som kommer inn i tunnelen gjennom tunnelveggene.

Eksperimenter med olivin viste mellom 96.9%-100% renseeffekt for alle elementene, men unntak av Cr, som ble sluppet ut, og As, som hadde en renseeffekt på mellom 63.9% og 95.6%. Olivin økte pH i vannet fra 3.18 til 12.00. Den høye renseeffekten for olivin første til konsentrasjoner av Pb, As, Cd og Ni i tilstandsklasse II eller I. Til tross for høy renseeffekt ble Cu og Zn funnet i tilstandsklasse V eller IV etter resing. Selv om Cr økte ved rensing med olivin, ble var kosentrasjonene etter rensing i tilstandsklasse III. Olivin viser lovende resultater som et mulig rensemedium i Killingdal, men Cr-utslippet og renseeffekten over tid må undersøkes ytterligere. Et rensesystem med olivin vil mest sannsynlig forbedres dersom det kombineres med andre rensetrinn.

Lufting ble undersøkt som et mulig for-rensetrinn, men viste ingen effektiv fjerning av metaller ved utfelling. En blanding av olivin og aktivt kull ble undersøkt, men til tross for høy renseeffektivitet for Cd, Pb, Fe, Ni, Cu og Zn, var ikke renseeffektiviteten høyere enn for olivin alene. Mer Cr sluppet ut fra blandingen enn fra olivin, i tillegg til at As ble sluppet ut. Blandingen bør derfor ikke vurderes som rensemedium for AMD i Killingdal.

Kalk-eksperimentene viste at kalk hadde en renseeffektivitet på mellom 98.5% og 100% for Pb og Cr, og 99.9% for Fe i ett av eksperimentene. Da kalk ble testet over tid ble det funnet at denne effektiviteten avtar over tid. Kalk økte pH fra 3.18 til 6.52 i ett av eksperimentene.

Olivin ble funnet til å ha høy renseeffektivitet for Cd, Pb, Fe, Ni, Cu og Zn, og ogå for As is ett av eksperimentene, mens kalk ble funnet til å ha høy renseeffektivitet for Pb, Cr, og Fe. Denne renseeffektiviteten til kalk, spesielt for Cr, bør utforskes videre i kombinasjon med olivin.

Foreword

This master’s project started in the fall of 2018 and was done in collaboration with Trondheim municipality. I feel so lucky to have been allowed to work with this exciting project at Killingdal. The process has taught me a lot, and it was very exciting and motivating to work on a current problem. The filed work inside the tunnel has been a really unique experience. I want to thank Kyrre Halvorsen at Trondheim municipality for welcoming me into the project and for his help and input throughout. I would also like to thank Anette Fenstad for her input, and for motivating me to speak about my master’s project at the Miljøringen conference. I would also like to thank Eirik Hindseth from TroMek for helping me with the practical setup and further development of the pipes for the experiments, and for always being willing to help.

I would like everyone who has helped me at NTNU. First and foremost I would like to thank my supervisor Øyvind Mikkelsen. He has always taken the time to help me, and has given me a lot of valuable feedback. I would also like to thank Syverin Lierhagen who did the ICP-MS analysis, for always being patient with me throughout all of my inquiries about the analysis, and for teaching me more about ICP-MS analysis. I would also like to thank Anica Simic for doing the analysis of the final samples. Finally I would like to thank the workshop at NTNU for making the pipes used in my experiments.

Two students have been working on master’s projects at Killingdal throughout this period. I have been so lucky to get to work with Hanne B. Olsen, and would like to thank her for our collaboration. We have been working on most parts of the field work, and I would like to thank her for her patience with me, her determination, knowledge, and skills. This project would not have been the same without her, and I am so grateful for our collaboration and, most of all, for our friendship.

I would also like to thank my parents for their support, and their help proof-reading this thesis. Finally I would like to thank my husband for always believing in me, supporting me through the stressful times, and for taking such good care of me. This has been essential for me, especially in the last stressful week.

Stine Steen
Trondheim 15.05.20

Index

Abbreviations

Abbreviation	Meaning
AA-EQS	Annual Average Environmental Quality Standard
AF	Assessment Factor
AMD	Acid Mine Drainage
As	Arsenic
Cd	Cadmium
Cr	Chromium
Cu	Copper
Fe	Iron
H	Hydrogen
HR-ICP-MS	High Resolution Inductively Coupled Plasma Mass Spectrometry
ISO	International Organization for Standardization
IUPAC	International Union of Applied Chemistry
MAC-EQS	Maximum Admissible Concentration Environmental Quality Standard
Mg	Magnesium
Ni	Nickel
NTU	Nephelometric Turbidity Unit
Pb	Lead
PNEC	Predicted No Effect Concentration
ppb	Parts per billion
RSD	Relative Standard Deviation
S	Sulphur
TM	Trondheim Municipality
XRD	X-ray Diffraction
Zn	Zinc

Figures

Tables

Background

Killingdal is an area in Ilsvika in Trondheim where Killingdal Grubeselskap had an active concentration plant from 1953 to 1986. Ore was transported from mines at Killingdal in Holtålen to this location in Trondheim, which from now on will be referred to as Killingdal. Here sphalerite (ZnS), pyrite (FeS_2), and chalcopyrite ($CuFeS_2$) were extracted, stored and shipped.[1,2] Killingdal Gruber AS went bankrupt in 1986 leaving the plant abandoned until Trondheim municipality, the Norwegian National Rail Administration, and Rom Eiendom demolished the old buildings, removed the most contaminated residue, and created a park and bike path in 2010-2011. A deposit was created under the bike path in an attempt to store the less contaminated mass without leakage. The deposits were covered in a bentonite membrane and topped with a layer of non-contaminated mass. Below the deposits lies the discharge-, storage- and transport-tunnel.[3] Water coming in through the tunnel wall has filled the tunnel and is visibly contaminated with residue from the activity at the old concentration plant; it has a distinct, opaque orange color. The water seeps out from the tunnel and into the sea along the shore.

Before the clean-up in 2010/2011 approximately one ton of metals leaked out into the fjord every year.[1] After surveillance of the area from 2011 to 2015 it became clear that the initial cleanup was not sufficient, and a more extensive cleanup was needed. Some emergency measures were executed in 2016, among them attempts to decrease the amount of water entering the tunnel. A cost-benefit analysis was performed, and it was concluded that cleaning the tunnel water would be the best alternative to prevent future contamination of the fjord in a cost-effective manner. To find the best treatment medium(s) and setup the municipality was granted a four-year trial period (2018-2021) by the Norwegian Environment Agency, with some conditions regarding the release of metals to the fjord during this period.[4,5]

Two master students have been involved in the project, and the project has been split in two; one student will focus on the first treatment step and one will focus on a polishing step in the treatment process. In my master project I will be looking at the first treatment step, and I will investigate lime and olivine as possible treatment mediums, through their effect on pH and their adsorption abilities. In addition I will look into using olivine in combination with activated carbon, which is one of the treatment mediums being studied as a possible polishing step. I will also consider the removal efficiency of metals through precipitation by aeration of the water, but the main focus will be on removing the aqueous metals by the aforementioned treatment mediums. I will also monitor the contamination level inside the tunnel over time.

The contaminated water is seeping into the fjord polluting it with several elements in high concentrations. Because of their abundance as well as possible adverse effects on marine life, these elements have been chosen as the most important ones to study, and will be the focus of this thesis: Copper (Cu), zinc (Zn), cadmium (Cd), chromium (Cr), arsenic (As), nickel (Ni), lead (Pb), and iron (Fe). These are also the elements mainly focused on by Trondheim Municipality.

1 Theory

1.1 Trace elements

Trace elements have several definitions, but in short they are elements that are only found or needed in trace amounts. IUPAC defines trace elements as “any element having an average concentration of less than about 100 parts per million atoms (ppma) or less than 100 μg/g.”.[6] In this thesis the elements in focus are copper (Cu), zinc (Zn), chromium (Cr), cadmium (Cd), lead (Pb), nickel (Ni), arsenic (As), and iron (Fe). Throughout this thesis the expressions trace metal and trace element will be used interchangeably. The elements of focus are all metals except for As which is a metalloid, and their position in the periodic table is shown in figure 1.1.

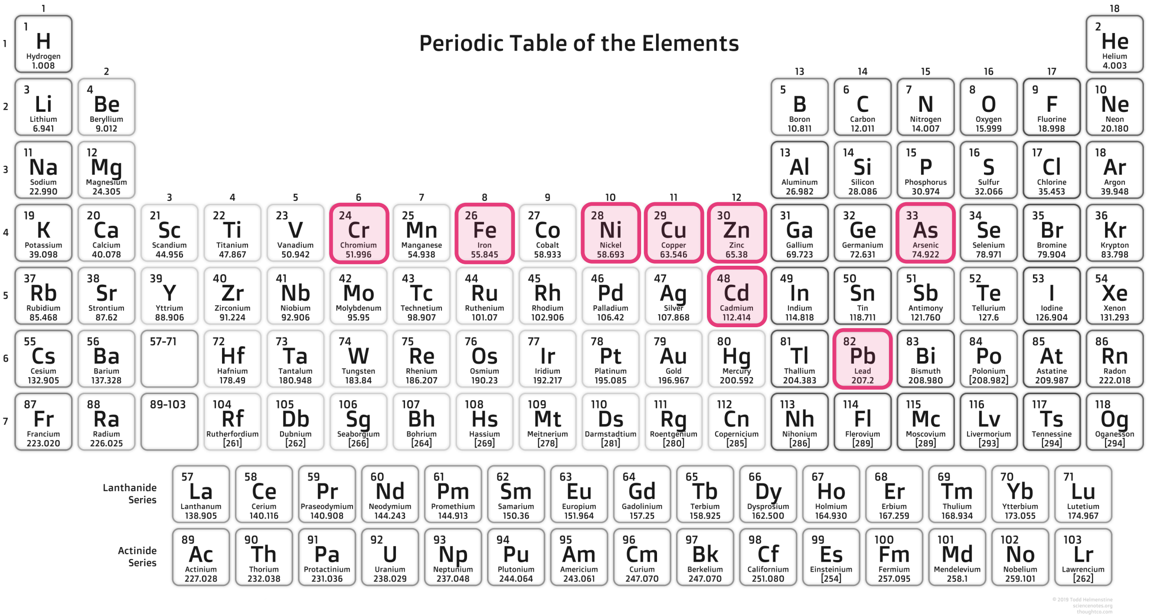

Figure 1.1 The Periodic Table of the Elements[7] with the eight elements of focus in this thesis marked in pink.

Metals and metalloids can exist in several different species, meaning different chemical forms. These include free ions with different charge, complexed, colloidal, and organic. It is not the total concentration of an element that decides its toxicity, mobility, or other behaviors, but each species has its own geochemical and biological properties and behaviors. In both water and soil, metals may exist as dissolved, solid, or colloidal, and the surrounding environment variables, like pH and redox potential, control what species each metal is found as.[8,9]

In metal contaminated soil several factors impact the mobility of the metals. Due to various functional groups and a high surface area, organic matter, Fe oxides, and other colloidal minerals in the soil have the ability to adsorb metals. When metals are adsorbed to these surfaces their mobility and bioavailability will be limited. There are several surface-related mechanisms leading to this control of solubility of the metals. Some examples are specific adsorption to hydroxyl groups on surfaces, surface complex formation, cation exchange, precipitation, and co-precipitation. Variables in the soil like pH, redox potential, and temperature will impact these mechanisms.[8,9]

The behavior of trace elements in water is largely influenced by adsorption. Adsorption is an interfacial reaction which can result from both chemical and electrostatic interactions.[10] These reactions occur between the surface of a solid and solute ions, and the resulting association differs in strength depending on the type of bond. If the ion becomes covalently bonded to the surface of the solid it is called chemisorption, or chemical adsorption.[6] This is stronger than physisorption, or physical adsorption, which is the association between the ions and the surface resulting from the weaker Van der Waals forces (intermolecular forces) alone.[6]

The different chemical properties of solid surfaces control the adsorption of different trace metals, because in aquatic environments solids have a surface charge. This charge attracts counter ions to balance it out, leading to a layer of water molecules coordinated on the surface. These may react and form a hydroxylated surface due to dissociative chemisorption.[10] The hydroxyl groups may be deprotonated, which leads to a negatively charged surface. Each functional group has a specific dissociation constant, pK_a, and if a surface has more than one functional group the net charge will be a function of all of these. A group will be deprotonated at pH above pK_a and stay protonated at pH below. Deprotonated groups can act as binding sites for positively charged metal ions. This adsorption property of minerals is used to remove metals from polluted water. Mineral surfaces may also become positively charged through the association of protons on the surface functional groups. The mineral is then working as a buffer, buffering the pH of the surrounding solution. Mineral surfaces are often amphoteric, meaning that the surface can become both positively and negatively charged, therefore having the ability to bind both anions and cations, depending on the pH of the surrounding environment.[10,11]

The master variable controlling metals in the aquatic environment is generally said to be pH. In interfacial reactions between surface solids and solute ions it is the controlling factor because it controls the surface charge of solids, as well as the speciation of metals. Changes in pH will affect reactivity, mobility, bioavailability, and toxicity. An example of pH controlling solubility is the effect it has on metal hydroxides. Most metals form insoluble complexes with the ligand hydroxide, and the solubility varies with pH.[11] Different metals precipitate at different pH ranges, and have different optimal precipitation pH. Table 1.1 shows the different ranges and optimal pH for the precipitation of different metals.[12] It is important to note that the precipitation of the different ions is also dependent on the other ions and ligands present in the water, as their presence may impact the efficiency of the hydroxide precipitation. This complication causes variations in numbers in literature.[12]

Table 1.1 The different precipitation ranges for different metal ions as well as their optimal pH for precipitation. From the book Lime and Limestone.[12]

Metal ion	Precipitation pH range	Optimum pH
Fe(III)	> 4.0	7-8.5
Cr(III)	> 5.3	9.5
Cu	> 6	7.5
Pb	> 6.0	10.0-10.3
Ni	> 6.7	9.8-10.2
Cd	> 6.7	10.5
Zn	8.0-11	10.5-11.0

Some metals exist in several oxidation states and the oxidation states may precipitate at different pH ranges. One example is Fe which becomes much more readily removed as a solid hydroxide at lower pH when oxidized from Fe(II) to Fe(III). Another example is the reduction of Cr(VI), which is soluble in water, to Cr(III), which is readily precipitated.[12] The degree of precipitation of these metal hydroxides may therefore vary in oxic and anoxic waters.

Metals can precipitate as oxides and hydroxides, and the solubility of some of the different metals are presented in figure 1.2.

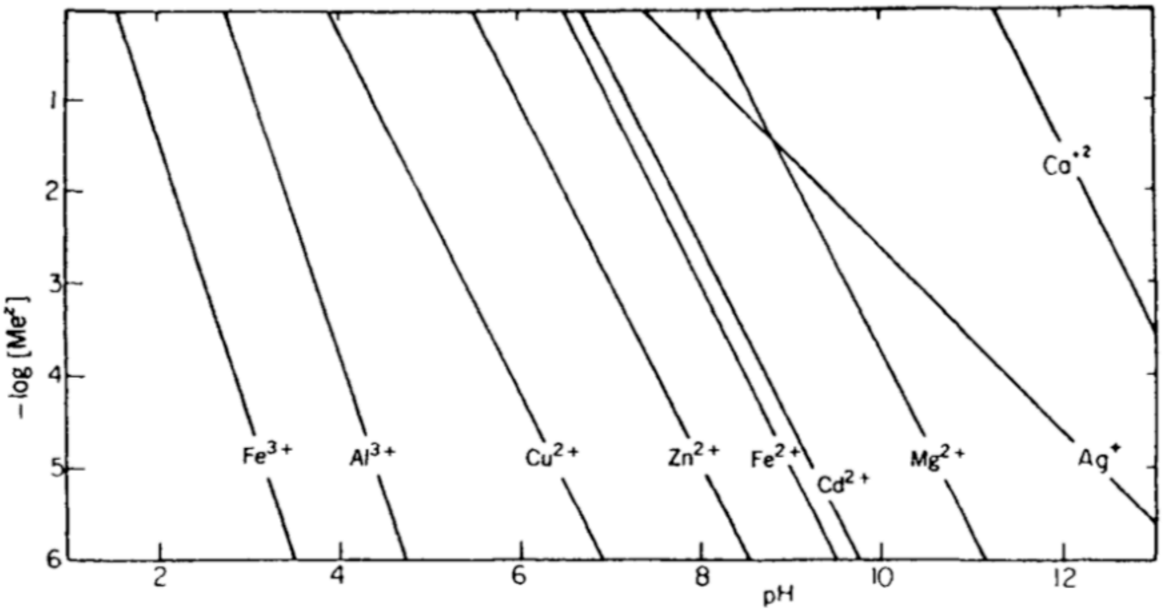

Figure 1.2. The solubility of different metal oxides and hydroxides, where the lines indicate equilibrium between free metal ions and solid (hydr)oxides. The (hydr)oxide in question will be solid to the right of the equilibrium line.[13]

Metal cations in water will be hydrated, i.e. surrounded by a number of H_2O molecules, also called aquo complexes. The number of H_2O molecules per metal ion varies, but most metal ions coordinate four or six. The radius, and oxidation state, of the central ion decides the acidity of the coordinated H_2O molecules. When the oxidation state becomes more and more positive, they become more and more acidic. For this reason the different metal ions exist as different species, and a simplified scheme of these is presented in figure 1.3.[13]

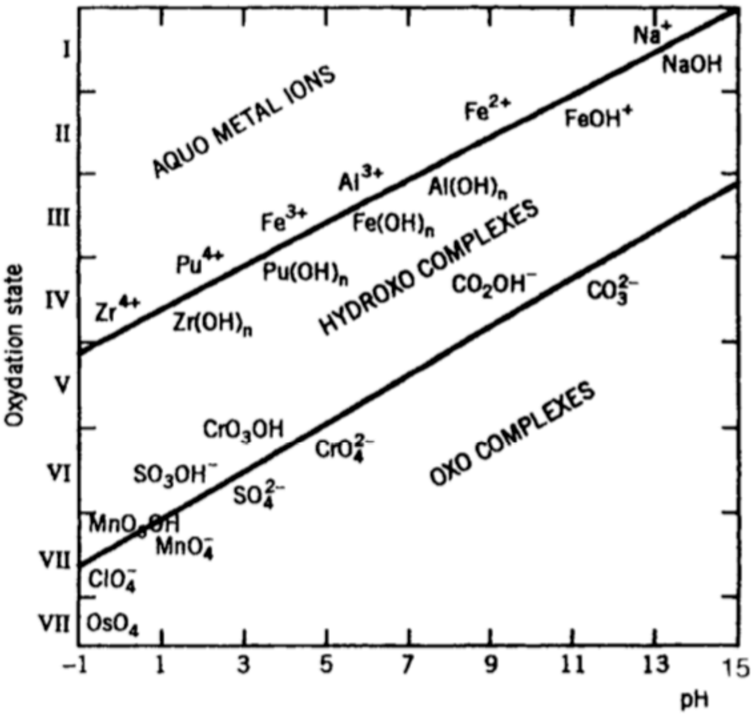

Figure 1.3. The predominant species (aquo, hydroxo, hydroxo-oxo, and oxo complexes) for different metals in aqueous solutions depending on oxidation state and pH. [13]

One example of different species can be found when looking at $Cr(OH)_3$. When dissolved we have the species Cr(III) and OH^-. Different reactions will occur between these two species, depending on the pH, and we will also have different species like $CrOH^{2+}$, $Cr(OH)_2^+$, $Cr(OH)_4^-$, and $Cr_3(OH)_4^{5+}$ present in the solution. There will be a similar situation for the other metal ions as well. Considering OH^- as a ligand, it becomes clear that the presence of ligands enhances the solubility of the solid phase.[13] This is the case with other ligands as well, not just OH^-. Other ligands in the AMD will be ligands such as Cl^-, SO_4^{2-}, and organic ligands, which are typical ligands in aquatic systems. In addition ligands such as S^{2-} and HS^- may be present. In this thesis the focus will not be on speciation, but on solid vs. dissolved. It is still of importance to consider that the picture is not as straight forward as it seems, and that the presence of other species than free metal ions exists in the dissolved phase, and precipitations other than hydroxides exist, and may complicate the understanding of precipitation in the water.

1.2 Trace element toxicity

Trace elements can be categorized as essential or non-essential. Essential elements are elements that are essential for life to an organism.[14] Each essential element has an optimal range of concentration, which can be regarded as a window of essentiality. An intake of less than this will lead to deficiency of the element, and an intake of more than the maximum of the essential window is toxic.[14] This effect can be reflected in a dose-response curve, of which an example is presented in figure 1.4.

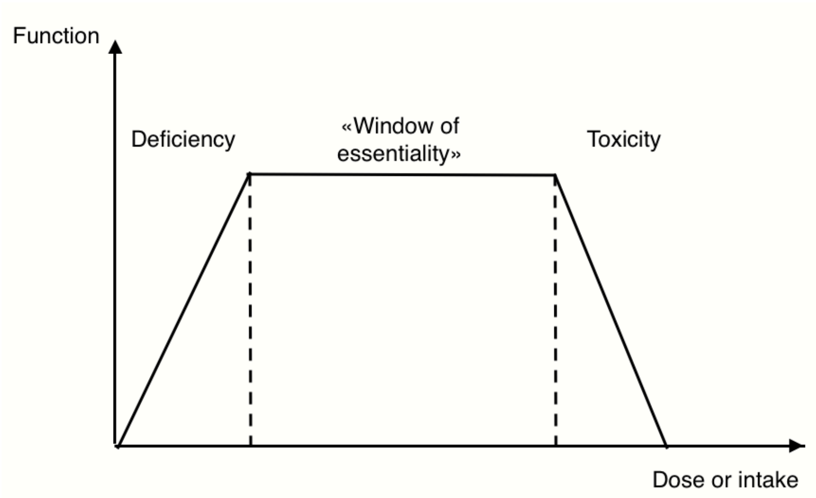

Figure 1.4 Conceptional illustration of a dose-response curve for an essential element. Intake within the "window of essentiality" is safe and adequate for normal function. Illustration inspired by Mason[15], Das et al.[16] and Walker et al.[14]

Fe, Ni, Cu, Zn, and Cd are all essential to aquatic life. They all play parts in enzymes, for example as co-factors, which is a common role for trace elements, and some are parts of important proteins or play roles in photosynthesis or redox regulation as well.[15]

Non-essential elements are not needed for normal function. This means that a low dose will not do any good, but it is not necessarily toxic either. A higher dose will cause toxicity. Figure 1.5 illustrates this effect.

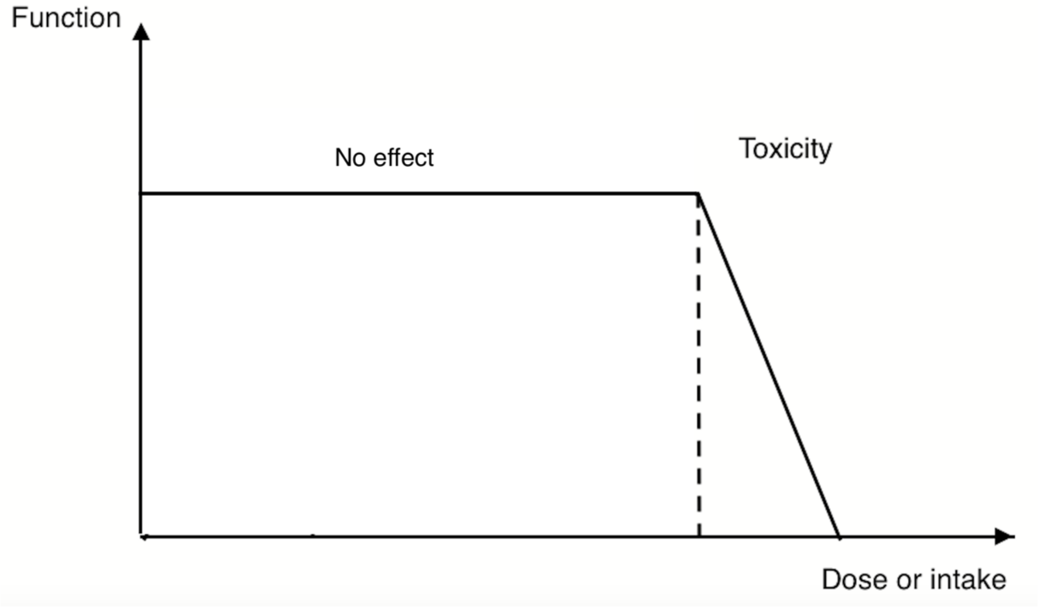

Figure 1.5 Conceptional illustration of a dose-response curve for a non-essential element. Illustration inspired by Walker et al.[14]

In addition to their toxicity at high doses, elements may act as inhibitors to each other, by competing for active sites. This competition can lead to one element causing the deficiency of an essential element. Both essential and non-essential elements can have this effect.[14]

Whether an element is toxic or not, and to what degree it is toxic, depends on several factors. The concentration, or dose, is an important factor for the toxicity of both essential and non-essential elements, since concentrations above the threshold of toxicity, as shown in figure 1.4 and 1.5, will be toxic, while concentrations below will not. In addition to the concentration, other factors also impact the degree of toxicity of elements. Different organisms have different requirements, so an element that is essential to one species is not necessarily essential to another. An example of this is boron (B) which is only essential to plants.[14] Different organisms also have different mechanisms to deal with trace elements, both essential and non-essential, e.g. mechanisms for uptake, excretion, regulation, and detoxification.[8] Therefore the toxicity of an element will vary with the species. In addition to being species-dependent, the degree of toxicity also varies with the speciation of the element.[17] Some forms of elements are more easily taken up than others, i.e. more bioavailable. Which form is the most bioavailable depends on the element, as well as the organism. The oxidation state of the element is also of importance. An example of this is Cr where Cr(III) is essential and Cr(VI) is toxic.[18]

Metals are non-biodegradable, meaning that they cannot be broken down to less harmful components. Therefore detoxification of metals that have entered the aquatic environment must be done by removing or immobilizing the metals either before they enter the organisms, e.g. by adsorption or precipitation of insoluble species, or by mechanisms like protein binding or long-term intracellular storage inside the organism.[14] Aquatic organisms will accumulate metals and metalloids that are released to the aquatic environment either directly from the water, or indirectly through the food chain. In high concentrations, above the threshold for toxicity, metals may cause severe oxidative stress, disturb growth, reproduction, and metabolism, and ultimately death.[19,20] Short-term exposure to high concentrations, or acute exposure, may cause death if the concentrations are high enough. Long-term exposure to lower concentrations, or chronic exposure, may have such outcomes as effects on growth and reproduction, deformities, and mortality.[21,22]

This thesis focuses on the elements of interest at Killingdal, and their effect on aquatic organisms, because the contaminated water is leaking into the fjord along the shore, possibly affecting the aquatic ecosystem there. Therefore, examples of the toxicity of some of the elements to aquatic organisms will be looked into in further detail.

1.2.1 The toxicity of selected elements

Cu is an essential element to vertebrates, including fish, playing vital roles in several enzymes and being important for the function of the nervous system. Cu is also considered one of the most toxic elements to fish and other aquatic organisms, causing irreversible harm to some species at levels just above the threshold for toxicity. Some known effects of Cu to fish is oxidative stress, behavior alteration (e.g. migration), effects on respiration, and reduction of resistance to disease.[23]

Cr(III) is an essential trace element that is a component of several enzymes. Cr(VI), the other stable form of Cr, is toxic to aquatic life. Cr is found to have acute toxic effects on fish behavior (e.g. swimming and balance) and effects on mortality, as well as biochemical and physiological alterations. Chronic exposure to Cr may cause reduced immune function, damage to DNA, effects on growth and survival, as well as other effects.[24]

Cd was earlier thought to be a toxic element, but in recent years it has been discovered that it may replace Zn in the enzyme carbonic anhydrase, therefore having a functional biological role.[15] Chronic exposure may cause developmental effects, effects on growth and reproduction, malformations, behavior alterations, and reduced immune response, among others.[25]

Pb is a non-essential element to aquatic life. Pb will bioaccumulate in fish and affect biochemical and physiological functions. The degree of bioaccumulation will vary with different species and environments. Pb affects various biological systems and examples of its effects are oxidative stress, behavioral and cognitive dysfunction, and reduced immune function.[26]

1.3 Mining consequences

Mining can be a significant anthropogenic source to metal pollution, even after the mining activity itself has ceased. When a mining company is closed down, the area might be left behind without proper cleaning. In the case of the Killingdal area in Trondheim, the discharge-, storage- and transport-tunnel as well as contaminated residues were abandoned at the site. Cleaning attempts were made in 2010/2011, but the surrounding grounds to the tunnel system are still contaminated, and water that has filled the tunnel by leaching through the foundation walls is contaminated with metals, as well as acidic with a pH of around 3.5 before treatment. This type of water created by mining residue is called acid mine drainage (AMD).[27] Hanrahan defines AMD as "acidic water laden with iron, sulfate, and other metals that forms under natural conditions when strata containing pyrite are exposed to the atmosphere or oxidizing environments."[11].

AMD is characterized by a pH below 5.5,[27] and while most AMD has a pH greater than 1,[27] some rare cases with drainage pH below one have been found, and pH as low as -3.6 has been reported.[28] The low pH of AMD is derived from the oxidation of sulfide minerals.[27] The minerals pyrite, sphalerite, and chalcopyrite are all sulfide minerals. Sphalerite is a sulfidic mineral mainly consisting of ZnS. It is the main ore for Zn, as well as the most common Zn mineral. In addition to Zn, it commonly contains Fe, sometimes up to as much as 26%.[29] It can therefore also be written (Zn, Fe)S. Chalcopyrite is a copper mineral often found together with pyrite. Its chemical formula is $CuFeS_2$.[29] The most common sulfide mineral is pyrite (FeS_2),[30] which is also the most common reason for acidification of natural waters due to mining activity.[31] In reducing conditions, where there is virtually no oxygen, pyrite, and other sulfide minerals, will remain in their natural form, but when exposed to oxygen, oxidation occurs. When these minerals are mined, the surface will come into contact with the oxygen in air, which in turn will lead to the oxidation of the sulfides, a process which acidifies the water, and releases metals and metalloids into it.

Pyrite oxidation can occur through both biotic and abiotic oxidation, i.e. with or without the presence of microorganisms. In addition the oxidation may be caused by oxygen alone or by the combination of oxygen and iron, i.e. direct or indirect oxidation. Direct oxidation will have the same outcome both through biotic and abiotic oxidation[30]:

$$\mathrm{FeS_2(s) + {}^{7}/_{2}\,O_2(g) + H_2O(l) \rightarrow Fe^{2+}(aq) + 2SO_4^{2-}(aq) + 2H^+(aq)} \qquad (1.1)$$

Indirect oxidation, meaning oxidation in the presence of both oxygen and Fe(III), happens in several steps. They can be summarized in three main steps; pyrite oxidation by oxygen (eq. 1.1), oxidation of Fe(II) to Fe(III) (eq. 1.2), and finally the oxidation of pyrite aided by Fe(III) (eq. 1.3).[30]

$$\mathrm{Fe^{2+}(aq) + {}^{1}/_{4}\,O_2(g) + H^+(aq) \rightarrow Fe^{3+}(aq) + {}^{1}/_{2}\,H_2O(l)} \qquad (1.2)$$

$$\mathrm{FeS_2(s) + 14Fe^{3+}(aq) + 8H_2O(l) \rightarrow 15Fe^{2+}(aq) + 2SO_4^{2-}(aq) + 16H^+(aq)} \qquad (1.3)$$

In the final reaction (eq. 1.3) Fe(III) acts as the oxidizing agent and is reduced to Fe(II). This Fe(II) can then react with oxygen (eq. 1.2), and will generate more Fe(III), which in turn will oxidize more pyrite. This cycle will continue until the supply of pyrite or Fe(III) runs out. The second reaction (eq. 1.2) only happens in low pH, because the solubility of Fe(III) is low in alkaline and neutral water. If the pH is raised above approximately 3.5, Fe(III) will precipitate as Fe hydroxides, giving the water its characteristic reddish-yellow color.[30] Then again, the precipitation reaction of Fe(III) will supply the solution with hydrogen ions, lowering the pH, as shown in eq. 1.4, which in turn allows more Fe(III) to stay in the solution.

$$\mathrm{Fe^{3+}(aq) + 3H_2O(l) \rightarrow Fe(OH)_3(s) + 3H^+\,(aq)} \qquad (1.4)$$

The Fe(III) in solution can oxidize more pyrite, following eq. 1.3, and this will again lower the pH of the solution.[30] What becomes evident is that this is a complex set of reactions that are all linked together and drive each other. The reactions described above are simplified, and in reality there will be several other factors influencing the generation of AMD, such as the presence of neutralizing agents, trace element substitution, complex formation, adsorption and similar reactions, in addition to the structure, porosity, particle size, surface area, and other characteristics of the pyrite.[30]

Even though the minerals processed at Killingdal were Fe, Zn, and Cu sulfides, several other trace elements are found in high concentrations in the AMD. In addition to pyrite, sphalerite, and chalcopyrite, several other minerals existed in the ore where the pyrite, sphalerite, and chalcopyrite were extracted from. These include bournonite ($PbCuSbS_3$), arsenopyrite (FeAsS), and galena (PbS).[32] Sulfides can contain other elements through cation substitution, as mineral inclusions, or crystal lattice impurities.[30] Chalcopyrite, pyrite, and sphalerite may all contain several other trace elements, presented in table 1.2.

Table 1.2 Possible minor and trace element substitutions in chalcopyrite, pyrite and sphalerite (after B. G. Lottermoser)[30]. The elements of focus in this thesis are marked in blue.

Mineral name	Chemical formula	Minor and trace element substitution
Chalcopyrite	$CuFeS_2$	Ag, As, Bi, Cd, Co, Cr, In, Mn, Mo, Ni, Pb, Sb, Sn, Ti, V, Zn
Pyrite	FeS_2	Ag, As, Au, Bi, Cd, Co, Ga, Ge, Hg, In, Mo, Ni, Pb, Sb, Se, Sn, Ti, Tl, V
Sphalerite	ZnS	Ag, As, Ba, Cu, Cd, Co, Cr, Fe, Ga, Ge, Hg, In, Mn, Mo, Ni, Sb, Se, Sn, Tl, V

When the minerals are mined and processed, these elements will also become exposed and enter the water. Hence AMD will not only affect the surrounding environment because of its low pH and high Fe, Zn, and Cu content, but it will also contain concentrations of the minor and trace elements in table 1.2. Yue et al. found that the leaching of Ni, Zn and Cd to AMD is likely controlled by the rate of oxidation of pyrite. They also suggest that the concentration of Pb, Cr, and As in AMD is controlled by their solubility, which in turn is governed by the pH of the solution.[33]

The presence of other sulfides, such as sphalerite may affect the oxidation of pyrite. Cruz et al. found that the presence of other sulfides causes galvanic protection.[34] In the case of sphalerite, this is caused by the fact that its rest potential is lower than that of pyrite. Consequently sphalerite is corroded before pyrite. This was seen by the high concentrations of zinc, and lower concentrations of iron and sulfate. Cruz et al. also found that chalcopyrite could cause the same effect.[34]

Similar to pyrite, sphalerite can be oxidized by oxygen or Fe(III). Oxidation by Fe(III) results in the release of H⁺ and therefore also acidification of the water, while oxidation by oxygen does not. The oxidation reactions of sphalerite are shown in eq. 1.5 and 1.6.[35]

$$\mathrm{ZnS(s)} + 2\mathrm{O_2(g)} \rightarrow \mathrm{Zn^{2+}(aq)} + \mathrm{SO_4^{2-}(aq)} \tag{1.5}$$

$$\mathrm{ZnS(s)} + 8\mathrm{Fe^{3+}(aq)} + 4\mathrm{H_2O(l)} \longrightarrow \mathrm{Zn^{2+}(aq)} + 8\mathrm{Fe^{2+}(aq)} + \mathrm{SO_4^{2-}(aq)} + \mathrm{H^+(aq)} \tag{1.6}$$

The Fe(III) in eq. 1.6 comes from the Fe(II) being released from the sphalerite, due to oxidation, and oxidized to Fe(III).

Chalcopyrite can also be oxidized either in the presence of oxygen or Fe(III), but neither of the oxidation reactions directly acidify the water, as none of them release H⁺. The oxidation reactions of chalcopyrite are shown in eq. 1.7 and 1.8.[21]

$$\mathrm{CuFeS_2(s)} + 4\mathrm{O_2(g)} \rightarrow \mathrm{Cu^{2+}(aq)} + \mathrm{Fe^{2+}(aq)} + 2\mathrm{SO_4^{2-}(aq)} \tag{1.7}$$

$$\mathrm{CuFeS_2(s)} + 4\mathrm{Fe^{3+}(aq)} \rightarrow \mathrm{Cu^{2+}(aq)} + 5\mathrm{Fe^{2+}(aq)} + 2\mathrm{S^0(aq)} \tag{1.8}$$

1.4 Treatment of metal contaminated waters

The best way to prevent pollution due to AMD would be to prevent the formation of AMD. This may not always be possible, or attempts may fail, and remediation becomes the best solution to the pollution problem. AMD formation happens in places with different surrounding conditions, and various options of systems to combat the pollution exist. These remediation strategies are divided into abiotic and biological systems, i.e. if they use chemicals or biological mechanisms to remove metals and neutralize the AMD.[36] These two categories include both active and passive systems, where active systems are those who require continuous input of resources, and passive systems require almost no input. Active abiotic systems could be for example aeration or addition of lime, while an example of a passive abiotic system is anoxic limestone drains.[36] Bioreactors can be an example of both passive and active biological systems, depending on the type. Figure 1.6 shows examples of each of the categories of remediation strategies for AMD.

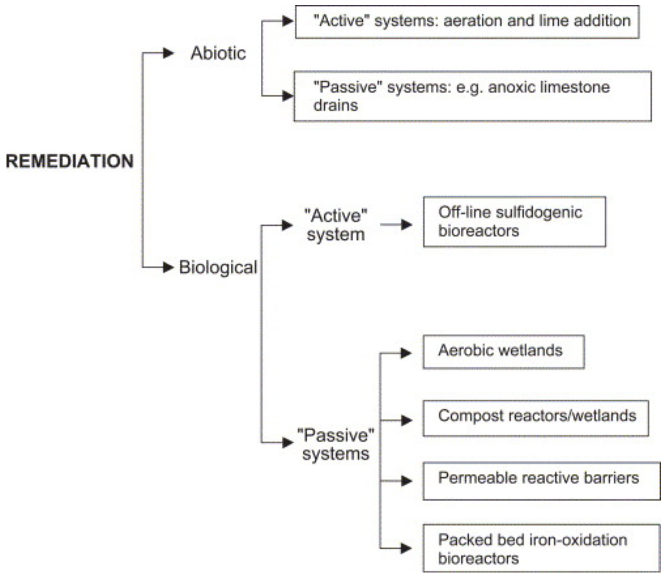

Figure 1.6 Different options of remediation systems for AMD in their respective categories. From Johnson et al.[36]

1.4.1 Abiotic remediation

Abiotic remediation, also called chemical remediation, involves the use of chemical processes to neutralize AMD and remove metals. The most widespread method is to use active technologies.[36] The added chemicals are alkaline and will therefore raise the pH of the AMD. Raising the pH is desired because this will cause Fe(II) to oxidize at a higher rate and cause the precipitation of metals as hydroxides.[36] One of the problems with this method is that it leaves behind a sludge that is rich in Fe and other metals. This sludge usually has a high water content, making the texture fluffy and therefore difficult and possibly expensive to handle. The density of the sludge may be improved by adding other steps to the precipitation process, like partial recycling of the sludge and dewatering, making the removal simpler and less expensive. Active abiotic techniques are usually viewed as a low cost technology, in its basic concept.[36] To increase efficiency and deal with sludge-removal difficulties, different refinement steps can be added to the process. This may of course make it more expensive, but a cost-benefit analysis has to be done to see if it is more expensive overall, seeing as efficiency and simple sludge removal may lead to lower total costs in the end.

There is also the possibility of using passive abiotic remediation systems. This involves the polluted water going through an anoxic limestone drain, which will raise the pH while maintaining Fe in its reduced form. When Fe is in its reduced form it will not precipitate as $Fe(OH)_3$. This is optimal for the lime, because the precipitation of $Fe(OH)_3$ would reduce the effectiveness of its neutralizing abilities.[36] The drain is constructed in a way that makes it impermeable to air and water from the outside, creating an anoxic environment. This environment accelerates the dissolution of the limestone because of the heightened CO_2 partial pressure, and therefore raises the pH faster, compared to an open system.[36] Anoxic limestone drains are considered passive because they require minimal maintenance after being built and activated. They are efficient, but may not be suitable for treating all types of AMD. For example, if the AMD contains high levels of Fe(III) or aluminum, precipitation can build up over time, and the drain may fail.[36] In addition, if the water is too aerated, it may need to pass through an anoxic pond before entering the anoxic lime drain, to lower the O_2 levels and prevent Fe(II) oxidation.[36] Anoxic limestone drains are often used as a component in a treatment system, for example in a system with wetlands, a biological remediation system.

1.4.2 Biological remediation

Some organisms have the ability to increase pH of AMD through sulfate reduction and Fe reduction. This ability is utilized in biological remediation processes of AMD. Both active and passive systems exist, and they all take advantage of microbial processes by different bacteria. In addition to raising the pH, metals are also immobilized through these processes. Sulfate reduction removes metals because several metals form insoluble sulfides.[36]

Passive biological systems are easily maintained, but may be expensive to set up, and require a large area of land. Examples of passive biological systems are anoxic ponds, modified wetland systems, and infiltration beds, where wetlands are the most common. Even though they require minimal maintenance, some monitoring may be needed to make sure that changes in abiotic factors like pH and temperature are observed, as changes in these may affect the efficiency of the treatment.[37]

Active biological treatment is performed through bioreactors. There are several types, but in general it is a system which is not in contact with the surrounding environment, such as is the case with passive systems. It needs robust pumps and precise piping networks of tough material, so the cost of installing such a system can be high. In addition the operating costs are high. Because of this, passive biological systems have been preferred and focused on, rather than active biological systems.[37]

1.4.3 Other remediation solutions

A combination of abiotic and biological systems may be preferable in some situations. For example, in some cases it has been observed that poorly performing constructed wetlands seem to improve their performance with the addition of anoxic lime drains.[36] The current remediation practices create significant amounts of sludge and are of varying efficiency. A new method of reuse and resource recovery has been suggested to replace the current approach of remediation. This is a sustainable practice and can reduce sludge generation, enhance efficiency, and recover valuable metals.[38]

1.5 Olivine, lime, activated carbon, and aeration

Both acidity and high concentrations of metals and metalloids may arise in AMD formation, and it is important to tackle this problem. Metals may be removed through precipitation, but with high concentrations, additional measures have to be taken. One possibility is to adsorb metal and metalloid ions onto the surface of a mineral. Adsorption of metal ions onto solid surfaces is an important mechanism for controlling the fate and behavior of trace metals.[39] In this thesis experiments were performed using mainly olivine and lime, and two minor experiments with activated carbon and aeration were also carried out. These treatment options will therefore be looked into in closer detail.

1.5.1 Olivine

Olivine is a mineral with the general formula (Mg, Fe)$_2$SiO$_4$, and it is a combination of the minerals forsterite (Mg$_2$SiO$_4$) and fayalite (FeSiO$_4$) in varying composition.[40] It can be found in nature as a green, sometimes yellow-ish or brown-ish, mineral often covered in a brown surface layer due to oxidation of the Fe content.[40] The Olivine used for the treatment of metal polluted water is often in the form of granules, created by crushing olivine into a fine powder, adding a binding medium, and packing it into a granule shape.[41] The olivine used in the experiments performed in this thesis was Blueguard G1-3 produced by Sibelco Nordic. They are granules produced for the purpose of "adsorbing heavy metals and organic environmental toxins" and for "purification of polluted water".[42] When the granules are produced in this specific way they allow water to flow through the olivine more easily than if using olivine powder, but it also allows for an internal flow through the granules, which leads to high contact surface between the contaminated water and the olivine.

Olivine has the ability to increase the pH of a solution by adsorbing H^+ ions. The most commonly studied form of olivine in this regard is magnesium rich olivine, and the studies show that pH is increased by substituting Mg^{2+} with H^+.[43] This process happens during the dissolution of the mineral. The dissolution rate of olivine is slow at conditions with a pH above 4, but becomes higher, and of practical importance, at pH < 3.[44] This property makes olivine a possible first treatment step for neutralizing AMD with pH < 3. The most commonly used passive abiotic treatment system is anoxic limestone drains, but problems with precipitation clogging the system can often occur. Using olivine as a first neutralization step may help prevent this, by increasing the pH and thereby reducing the amount of Ca ions that could potentially precipitate and clog the anoxic limestone systems.[44] The dissolution of olivine can be described by the following equation (eq. 1.9):

$$\mathrm{(Mg,Fe)_2SiO_4(s) + 4H^+(aq) \rightarrow 2(Mg^{2+},Fe^{2+})(aq) + H_4SiO_4(aq)} \qquad (1.9)$$

As this reaction occurs, four moles of H^+ will be neutralized per mole of olivine. The concentration of Fe(II) in the solution will increase, and if this happens in an anoxic environment there will be no precipitation, but in oxic conditions Fe precipitation will occur, first by the oxidation of Fe(II) to Fe (III) (eq. 1.2) and then by the hydroxide precipitation of Fe (III) (eq. 1.4). However, the precipitation of Fe(III) is pH dependent, and if the pH is below 4 the reaction will become so slow that it is negligible.[44]

Olivine is also an adsorbent and has been found to be a good copper adsorbent,[45] in addition to other trace metals.[46] The adsorption of divalent metal ions onto a silicate surface, such as olivine, has a tendency to increase with increasing pH.[43] This effect is due to the competition of the adsorption seats between the metal ions and H^+. Kleiv et al. found that the adsorption of copper to olivine increased rapidly when the pH increased from 4 to 6.[47] In addition, it has been shown that the solid/solution ratio is positively correlated to the adsorption of Cu.[43] A higher solid/solution ratio means more surface area, and therefore more active binding sites for the metal ions to react to.

The presence of Fe in a solution can affect the adsorption of metals to olivine. This is due to the change in pH that occurs. When olivine is introduced to a solution, it will raise the pH, and Fe(II) will oxidize to Fe(III) (eq. 1.2), which will precipitate as $Fe(OH)_3$ (eq. 1.4). The precipitation of $Fe(OH)_3$ will result in 3 moles of H^+ released per mole of Fe(III) precipitating. This will lead to a decrease in pH, which again leads to a desorption of metals from the olivine surface.[43]

1.5.2 Lime and limestone

Lime is a term which includes CaO and $Ca(OH)_2$, which are called quicklime and slacked lime respectively.[12] The usage of lime to treat AMD is the most commonly applied method because of its low cost and ability to remove metals through neutralization.[48] Treatment with lime leads to an increase of pH in AMD, and as a consequence metals are removed from the water through precipitation.

The principle of AMD treatment with lime is based on lime dissolution, which increases the pH of the solution. If CaO is used, it is added to the solution and reacts with water to form $Ca(OH)_2$, which then dissolves. The dry form of $Ca(OH)_2$ can also be used directly. The dissolution is presented in eq. 1.10 and 1.11.[48]

$$\mathrm{CaO(s) + H_2O(l) \rightarrow Ca(OH)_2(s)} \tag{1.10}$$

$$\mathrm{Ca(OH)_2(s) \rightarrow Ca^{2+}(aq) + 2OH^-(aq)} \tag{1.11}$$

After the dissolution of $Ca(OH)_2$ the pH will have increased. The free hydroxide ions are then available for the precipitation of metals as metal hydroxides. An example of this precipitation is shown in eq. 1.12, using Zn as an example.[48]

$$\mathrm{Zn^{2+}(aq) + 2OH^-(aq) \rightarrow Zn(OH)_2(s)} \tag{1.12}$$

This precipitation occurs at different pH ranges for each element, as presented in table 1.1. Within this range the metals will most likely exist as metal hydroxides, but both at pH lower and higher the metals will most exist as dissolved species. When the pH is lower than the optimal pH for hydroxide precipitation, we will find the metals as hydrated ions. If the pH increases above the range for hydroxide precipitation, i.e. the hydroxide concentration increases, it is likely that the metal hydroxides will react with another hydroxide and form a soluble metal complex. This leads to an increase in solubility of the metals.[27] In the presence of Fe(III), which precipitates at lower pH than the other metals, the metals may be removed at a lower pH due to co-precipitation and adsorption to $Fe(OH)_3$.[49]

Crushed $CaCO_3$ can also be used in the treatment of AMD. Using $CaCO_3$ is inexpensive and produces a less voluminous sludge, but this treatment method has several disadvantages and is not widely used. When using $CaCO_3$ it is difficult to raise the pH above 6 because of the buffering effect CO_2 will have on the reaction. In addition it is more challenging to use in water with a high Fe(II) content, because the $CaCO_3$ will become coated with $Fe(OH)_3$ precipitation and rendered ineffective. Another disadvantage is the slow dissolution, and consequently slow reaction time.[49,50]

The dissolution of $CaCO_3$ is shown in eq. 1.13, where it can be observed that the dissolution uses one mole H^+ per mole $CaCO_3$ dissolved.[27] This will increase the pH of the solution.

$$\mathrm{CaCO_3(s) + H^+(aq) \rightarrow Ca^{2+}(aq) + HCO_3^-(aq)} \tag{1.13}$$

This dissolution will also introduce a buffering capacity to the solution.[27] The buffering capacity is a result of the carbonate system, shown in eq. 1.14 and 1.15.[11] The HCO_3^- gives the water its buffering capacity through the ability to incorporate H^+. HCO_3^- in water can take up one H^+ from the acidic solution and become CO_2, which can become a gas and exit the water to the air.

$$\mathrm{CO_2(g) \rightleftharpoons CO_2(aq) + H_2O(l) \rightleftharpoons H_2CO_3(aq)} \tag{1.14}$$

$$\mathrm{H_2CO_3(aq) \rightleftharpoons HCO_3^-(aq) + H^+(aq) \rightleftharpoons CO_3^{2-}(aq) + 2H^+(aq)} \tag{1.15}$$

1.5.3 Aeration

Aeration adds oxygen to the water making it a more oxidizing environment. This will oxidize some of the metals in the solution. The main goal of aeration of AMD is often to oxidize Fe(II) to Fe(III) so that it will precipitate as Fe hydroxides.[48] If aeration is used as a pretreatment of AMD, before treatment with for example olivine or lime, there might be less problems with precipitation clogging the system, and the following treatment may become more effective.

The precipitation of Fe hydroxides will cause the co-precipitation of other trace metals present in the water. This will remove them from the water column, decreasing their concentration. Co-precipitation involves the incorporation of the trace metal into the hydroxide lattice structure.[51] In addition the newly formed precipitate can adsorb metals to its surface. Adsorption results in a weaker binding of the metals than inclusion in the precipitate.[52] $Fe(OH)_3$ has been found to be a good adsorbent for As, Pb, and Cu, as well as Zn and Cd to some extent.[9]

Through aeration, As is removed by co-precipitation and adsorption of As with $Fe(OH)_3$, or by precipitation of As as an arsenate (AsO_4^{3-}) form.[27] Arsenate forms can be both stable and unstable, depending on the other element in the precipitation, where for example $FeAsO_3$ is considered stable, but $Ca_3(AsO_3)_2$ is unstable. Aeration may therefore only remove As temporarily if it dissolves as an unstable form.[27] As has a high affinity to Fe, and adsorption and co-precipitation with $Fe(OH)_3$ has been found to be one of the main controls of As.[9]

1.5.4 Activated carbon

Activated carbon is defined as porosity enclosed by carbon atoms.[53] It can be made from several sources including coals, hard wood, coconut shells, and fruit stones. It is activated by thermal decomposition and physical modification in a furnace under controlled environments.[54] Its surface has a high degree of microporosity and therefore a large surface area, which makes it ideal as an adsorbent. It is widely used in the removal of metal contaminants in polluted waters.[55] The adsorption onto activated carbon can be both physisorption and chemisorption.[56] Because the surface area is so large, and because of different functional groups present (e.g. hydroxyl, carbonyl, etc.), it has a high adsorption capacity for many different types of contaminants, both organic and inorganic, and it can be used to remove most metal contaminants.[37,57] It has been used in permeable reactive barriers to treat AMD. In this method water passes through a barrier of activated carbon by natural flow and metals become adsorbed to the activated carbon surface.[37] Because of the depletion of commercial coal-based activated carbon, the price is increasing.[55] One solution to this problem could be to use activated carbon mixed with other adsorbents, for example olivine.

1.6 Classification of condition

The Norwegian Water Regulation was updated in 2015 and now includes limit values for 45 prioritized environmental pollutants, including seven of the elements of focus in this thesis.[58] The prioritized pollutants have properties like low degradability, bioaccumulatory properties, and toxicity, causing them to pose a risk on the environment and on the health of the organisms living there.[59] The classification system is based on an expected degree of harm on the organisms, and the effects range from background to very poor, and these are presented in table 1.3. Biologically the classes are divided into background, no toxic effect, chronic toxic effect by long-term exposure, acute toxic effect by short-term exposure, and extensive toxic effects. These classes are derived from results from laboratory tests, risk assessments, and information on the acute and chronic toxicity to organisms.[58,59]

Table 1.3 The different classifications of conditions of water and their definitions. From the Norwegian Environment Agency's guide *Quality standards for water, sediment and biota*.[58]

I Background	II Good	III Moderate	IV Poor	V Very Poor
Background level	No toxic effect	Chronic toxic effect by long-term exposure	Acute toxic effect by short-term exposure	Extensive toxic effects
Upper limit: Background	Upper limit: AA-EQS, PNEC	Upper limit: MAC-EQS, $PNEC_{acute}$	Upper limit: $PNEC_{acute}$*AF	

To quantify the limits for each class, ecotoxicological parameters are used. For class II, the upper limit is defined by the AA-EQS, which is the Annual Average Environmental Quality Standard, and is the limit for chronic effect by long-term exposure. For class III the upper limit is defined by the MAC-EQS, which is the maximum EQS value, and is the limit for acute effect by short-term exposure. Class IV is based on the same value as class III, but a safety factor (AF) is taken into account, when needed. This is a factor that is used to ensure protection of prospective organisms that are more vulnerable than those who have been used for testing in the lab, and is used when there is not enough information from lab testing. The values for each class are given in [µg/l] for water, and are specific to each pollutant.[58]

Different limits are defined for classification of condition used for fresh water, coastal water, sediments, and biota. This thesis considers the classes for coastal waters only. This is because the water at Killingdal eventually ends up in the sea, and it is therefore important to use the classification system that has taken the aquatic ecosystem into account, as opposed to the fresh water classes, which are based on terrestrial ecosystems. Water samples should in general be unfiltered when being classified with this system, but metals are an exception.[59] In this thesis all samples are filtered. When the classification system is used for defining the condition of a body of water, samples should not be taken in proximity to the source of emission of pollution. If the classification system is used for monitoring of an area to decide what measures to potentially introduce, the samples may be taken close to where the pollution occurs. In this case, the results will not say anything about the whole body of water, but rather give an indication of the urgency to remediate this specific area of pollution.[59]

The concentrations for each class for the elements of focus in this thesis are given in table 1.4. They are all given in [µg/l], and this is the classification of condition for coastal water. Class III, IV, and V for Cd are different depending on the hardness of the water.

Table 1.4 The limits for each class of condition for each of the elements Cd, Pb, Ni, Cu, Zn, As, and Cr. All of the values are given in [µg/l]. From the Norwegian Environment Agency's guide *Quality standards for water, sediment and biota.*[58]

Element	I Background	II Good	III Moderate	IV Poor	V Very Poor
Cd	0.03	0.2	*	◆	◆
Pb	0.02	1.3	14	57	>57
Ni	0.5	8.6	34	67	>67
Cu	0.3	2.6	2.6	5.2	>5.2
Zn	1.5	3.4	6	60	>60
As	0.15	0.6	8.5	85	>85
Cr	0.1	3.4	36	358	>358

* Class III for Cd depends on the hardness of the water:
≤ 0.45 for hardness < 40 mg $CaCO_3$/L
0.45 for hardness 40-50 mg $CaCO_3$/L
0.60 for hardness 50-100 mg $CaCO_3$/L
0.9 for hardness 100-<200 mg $CaCO_3$/L
1.5 for hardness ≥ 200 mg $CaCO_3$/L

◆ Class IV for Cd depends on the hardness of the water:
≤ 4.5 for hardness < 40 mg $CaCO_3$/L
4.5 for hardness 40-50 mg $CaCO_3$/L
6.0 for hardness 50-100 mg $CaCO_3$/L
9 for hardness 100-<200 mg $CaCO_3$/L
15 for hardness ≥ 200 mg $CaCO_3$/L
Values over the given value above belongs to class V

1.7 Sampling for analytical use

The sampling procedure is important in order to produce reliable results, especially when the concentrations are in the [µg/L] range. Samples may become contaminated by the sampling equipment, cross-contamination between samples, and by incorrect sample preservation and storage. There are international standards in place to ensure the quality of the sampling, as well as to make experiments reproducible. For this thesis the different standards in the ISO 5667 series are the most relevant, as they refer to water quality. NS-EN ISO 5667-1:2006 gives a guideline for designing the sampling program and techniques.[60] NS-EN ISO 5667-3:2018 describes the different requirements for sampling, preservation, handling, transport, and storage of water samples in general. For example it specifies the container that should be used for specific analytes and analytical methods, as well as how to preserve samples with these analytes.[61] A guidance of quality assurance and quality control is provided in NS-EN ISO 5667-14:2016.[62] In addition to these there are specific standards for the sampling of different matrices, e.g. freshwater and marine waters, as well as standards for measurement of water properties like pH (NS-EN ISO 10523:2008)[63] and turbidity (NS-EN ISO 7027-1:2016)[64]. Following these standards ensures quality of sampling and measurements, and allows for universal methods being used internationally, facilitating understanding and reproducibility of experiments.

1.8 Inductively coupled plasma mass spectrometry

Inductively coupled plasma mass spectrometry (ICP-MS) is an analytical method based on ionizing a sample and then separating and detecting each isotope separately. It gives you the total amount of an element as well as specific isotope information in a sample. It does not specify the species or type of chemical bonds.[65]

The ICP in ICP-MS serves as both an atomizer and ionizer. ICP is the plasma source in the ICP-MS. Plasma is a gas that has been partially ionized and maintains high temperature. Its ions, including argon ions and electrons, give it a conducting property. Argon flows through a radio-frequency generator, and the use of this generator has a great advantage over other generators such as micro-wave or a dc arc source, in terms of liberation from interferences and sensitivity.[65]

The sample is usually nebulized before being introduced to the argon flow. This is done by transporting the liquid sample into the nebulizer, where it is broken up into fine droplets by a high-velocity gas. The drops are then carried into the argon plasma. Because the temperature of the plasma is extremely high (6000-8000 K), the atomization efficiency is high, which gives fewer chemical interferences, compared to combustion flames. Another advantage is the high concentration of electrons from the ionization of argon in the plasma. It leads to a more or less constant electron concentration in the plasma, which in turn leads to ionization interferences being small to non-existent. In addition, the chemically inert environment where the atomization happens is favorable to the reactive environment that occurs in flames and other plasma sources. This, along with the relatively uniform temperature of the plasma, results in calibration curves that are linear over several orders of magnitude of concentration. ICP also has a tendency to lead to ionization of the analytes, not just atomization, and this characteristic is exploited in ICP-MS, where ICP is the ionizations source.[65]

Mass spectrometry (MS) separates ions based on their mass-to-charge ratio, and converts the abundance of ions into an electrical signal. The mass-to-charge ratio (m/z) is the ratio between the exact mass of the ion/isotope, m, and the number of fundamental charges, z. [65] ICP-MS combines ICP and MS, where ICP ionizes the sample, making the analytes gaseous ions and accelerating them into a mass analyzer, where the ions are separated based on their m/z ratio, which is then turned into an electrical signal. A data handling system processes the results and prints them as mass spectrums. Elaborate vacuum systems are required for the mass spectrometer, because this will lead to a low collision frequency, which, in turn, maintains the free ions and electrons. This poses a problem in ICP-MS because the ICP operates in atmospheric pressure. The interface between ICP and MS therefore has to connect an area of atmospheric pressure to an area of high vacuum. This is done with two cones; the sampler and the skimmer.[65] Ions from the plasma, and the plasma itself, pass through the sampling cone due to pressure differences, where the pressure in the area after the sampling cone is of moderate pressure. The skimmer (second cone) is placed close to the sampling cone and the gas then flows through this cone which typically has a smaller opening than the first cone. The ions then pass through the skimmer to ion optics, and the ion optics aids in taking the ions from atmospheric pressure to the low pressure of the mass analyzer, while minimizing background noise. Ion optics focuses the ions as well as prevents particulates, neutral species and photons from getting through to the mass analyzer. The ion beam is guided further into the instrument to the mass analyzer.[66]

The mass analyzer is the component in the MS that separates ions based on m/z ratio. In ICP-MS the mass analyzer is one or more quadrupoles. It works as a mass filter that only lets through ions of a certain m/z ratio. It consists of four rods that are connected to dc and radio-frequency (time-dependent ac[66]) voltages.[65] Adjusting the voltages creates a stable path for the ions of a certain m/z to pass through to the detector, while the other ions become unstable and are ejected from the quadrupole. The voltages are adjusted again to let through a different m/z ratio, and this process is repeated until all of the elements have been analyzed.[66]

Finally, the ions exit the mass analyzer and reach the detector. It converts the ions into electrical pulses to quantify them. The pulses' magnitude corresponds to the number of analyte ions in the sample, and is used for quantification by comparing the ion signal to known reference or calibration standards,[66] which usually is a calibration standard with the ratio of the ion signal of an internal standard and the signal of the ions in the analyte plotted against concentration.[65] The most common detector used is the continuous dynode electron multiplier, but other detectors may also be used. Dynode detectors amplify the signals efficiently.[66]

There are two types of interferences which may occur in ICP-MS; matrix and spectroscopic interferences. When the concentration of the species in the matrix is high, a matrix effect may become notable, reducing the signal of the analyte ions. This problem can be fixed by diluting the sample. When another ionic species has the same m/z ratio as the analyte ion, spectroscopic interferences occur. This problem usually occurs with polyatomic ions, isotopes with the same mass, or ions that have been doubly charged, which all result in overlapping ions with the analyte ions. Today we have High-resolution (HR-)ICP-MS which reduces and in some cases eliminates this problem.[65] With HR-ICP-MS the analyte is separated from the interferences by taking advantage of small differences in m/z, which is possible due to the high mass resolution[67].

ICP-MS is a technique with a wide dynamic range. The detection limit varies for each element, but most elements can be detected down to below ppb, and even lower for HR-ICP-MS (even down to sub-ppt). Internal standards are often used to compensate for drift, instabilities, and matrix effects. Calibration curves using external standards are usually used for quantitative analysis.[65]

There are standards for the use of ICP-MS analysis for determination of elements in water. NS-EN ISO 17294-1:2006 describes ICP-MS in general, including information about the principle and apparatus, common interferences by contaminant elements, apparatus adjustments, preparation steps, and the procedure.[68] NS-EN ISO 17294-2:2016 specifies a method for the determination of selected elements, including the ones that are in focus in this thesis. It includes information about the interferences, reagents, sampling, sample preparation, and the procedure.[69] These standards should be followed as much as possible to ensure quality of the analysis and results. Following these allows for a universal method being used internationally, facilitating understanding of the results.

Statistical methods used

To check if some data is significantly different from other data it needs to be known whether the data is normally distributed. A Shapiro-Wilk test can be used to check if a sample fits the normal distribution model in a population. The data is entered in the test, and the test compares the observed distribution to the normal distribution and computes the probability of finding the observed distribution assuming that the population is normally distributed. The test's biggest limitation is its bias to sample size, where the larger the sample size, the more likely the model is not correct.[70,71]

If data is normally distributed a t-test can be used to indicate if there is a statistically significant difference between the data in two treatment conditions. It is used when looking at a numerical value, e.g. concentration of a metal in water, and it compares the mean between two different sets of data, e.g. concentration of a metal in water before and after some treatment. The test should only be used when the sample is < 30.[72,73]

If data is not normally distributed, a Mann-Whitney U test can be used. It checks for a significant difference between two independent groups, but unlike the t-test it compares the whole distribution, not just the mean. This test is more robust to outliers than the T-test, but is slightly weaker than the T-test.[74]

2 Materials and methods

In this section a general description of the sampling period, sampling methods, and materials used will first be described. Then a description of the sampling inside the tunnel, including a description of the sampling sites, will be provided. Following this the methods for the different experiments will be specified. After the experiments, the ICP-MS analysis and sample preparation will be described, followed by the statistical methods.

The sampling period for this thesis was from October 2018 to February 2020. During this period Trondheim municipality had a treatment system inside the tunnel, which was used by them to test different cleaning mediums. A visualization of the tunnel is presented in figure 2.1, and a picture of the tunnel with Trondheim municipality's system is shown in figure 2.2. Samples were taken at different sampling points inside the tunnel, and will be described in further detail below. The sampling frequency varied, in part, due to Trondheim municipality's system being changed during the sampling period. To see all the changes that have been done to Trondheim municipality's treatment system throughout the sampling period, see appendix A.

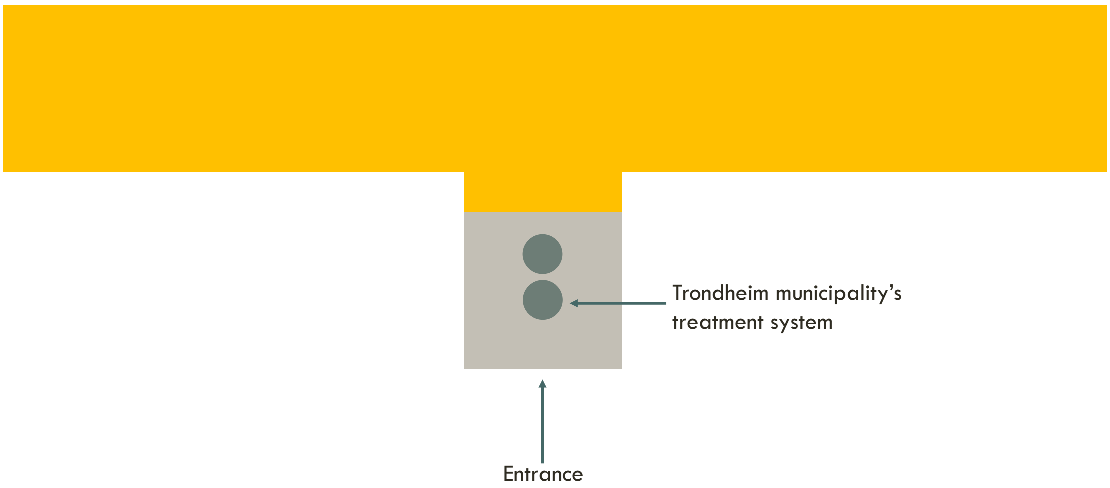

Figure 2.1 The tunnel seen from above. The orange color represents the water inside the tunnel, and the gray area is where Trondheim municipality's system is placed, and where experiments for this thesis have been performed.

Figure 2.2 The tunnel with Trondheim municipality's system in January 2019. The system changed over time. For more information about the changes made in Trondheim municipality's system, see appendix A.

All samples in this thesis were taken and preserved according to ISO standard NS-EN ISO 5667-6 and NS-EN ISO 5667-3.[61,75] Measurements of pH were performed according to ISO standard NS-EN ISO 10523.[63] Turbidity measurements were performed according to ISO standard NS-EN ISO 7027-1.[64]

The olivine used in the experiments and in Trondheim Municipality's system was Blueguard G1-3 produced by Sibelco Nordic.[42,76] The lime used was $Ca(OH)_2$ of the type Miljø Hydratkalk VK. The activated carbon was of the type SUPER KULL Chemviron 0.4-1.4mm.

All samples were taken according to the following method, unless otherwise specified:
Samples were taken with a polypropylene syringe and water was drawn into and discharged from the syringe three times before the sample was taken. The sample was filtered through a 0.45 μm filter. A 15 mL polypropylene tube (MetalFree for ICP-MS use) was washed with the filtered sample three times. 10 mL of the final filtered sample was then added to the tube. 3 drops of ultrapure HNO_3 was added for preservation.

2.1 Samples taken in Trondheim municipality's system

Trondheim municipality's system changed over time, but there was always one point in the system representing the water inside the tunnel; either an open tub where untreated water passed through before entering the treatment medium, or a hose taking water in from the tunnel. The height of the intake of water may have changed, where it is assumed that the hose took in surface water while the intake supplying water to the open tub was deeper, but this is uncertain. For more information about the changes made in Trondheim municipality's system, see appendix A. Samples were taken of the untreated tunnel water, as well as of the treated water, when possible, in Trondheim municipality's system.

2.2 Samples taken inside the tunnel

Samples were taken at different sampling points inside the tunnel, presented in figure 2.3. Samples were taken according to the general sampling method described above. At sampling points where drawing in water with the syringe was not possible, the syringe was filled from the top (open syringe without the plunger). This was also repeated three times before the sample was taken.

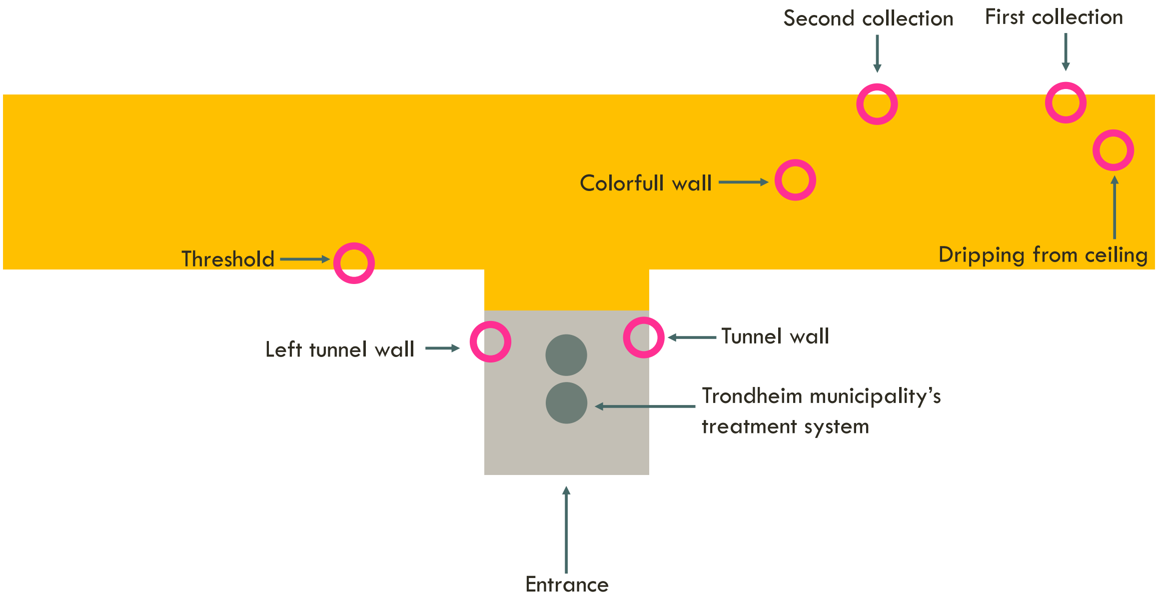

Figure 2.3 The tunnel seen from above with the different sampling points marked by pink circles. The orange color represents the water inside the tunnel, and the gray area is where Trondheim municipality's system is placed, and where experiments for this thesis have been performed.

For pictures of the different sampling sites, see appendix A.

2.3 Experiments

In addition to monitoring the metal concentration in the water inside the tunnel, a series of experiments were performed to assess the treatment efficiencies of olivine, lime, aeration, and the combination of olivine and activated carbon. To carry out the experiments, three plexiglass pipes with nozzles at the bottom were made. These are shown in figure 2.4. One of the pipes was connected to a pump taking water in from the tunnel and through the pipe, while the other two pipes remained open. The pipes will hereon be referred to as either open or connected to pump.

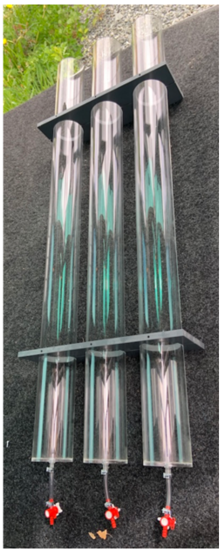

Figure 2.4 The three plexiglass pipes used for experiments.

2.3.1 Olivine

Gravel was added to the bottom of one of the open pipes to prevent the treatment medium clogging the nozzle. The gravel was rinsed three times with 7 L of water from the tunnel taken from an open tub in Trondheim municipality's system. A sample was taken of the water from the third rinse before adding it to the pipe, and a sample was taken of last water that passed through the gravel. A plastic ladle was used to add 10*220 g of olivine to the pipe on top of the gravel, before the pipe was filled up with approximately 6 L of tunnel water (taken from Trondheim municipality's system). The nozzle was opened to the point where water was running slowly through the olivine. A sample was taken after approximately 0.6 L of water had passed through, after approximately 3 L had passed through and after approximately all 6 L of the water had passed through. The pipe was refilled with 6 L more of tunnel water and a sample was taken of the last of this water to pass through the olivine.

Samples that were taken of water that had passed through the olivine were first collected in a polyethylene bottle. Samples were then taken from the collection bottle according to the general sampling method described above.

Gravel was added and rinsed with the same method as above, and new 10*220 g olivine was added to the pipe connected to the pump. This pump brought water from inside the tunnel into the pipe. The pump was turned on and the nozzle opened slightly, and the balance between water in and out was found. This was left for two days.

2.3.2 Lime

Gravel was added and rinsed with the same method as described in the method for the olivine experiment, and 10*220 g of lime was added to the pipe connected to the pump. The pump was turned on and the nozzle opened slightly, and the balance between water in and out was found. A sample was taken as soon as the water passing through the lime lost the milky white color that came from lime dust. Samples were also taken after approximately 75 and 110 minutes. The system was left running over night and a sample was taken the next day after approximately 20 hours.

Rinsed gravel and 10*220 g of lime was added to one of the open pipes and the pipe was filled with approximately 6 L tunnel water (from Trondheim municipality's system). The nozzle was opened slightly and a sample was taken as soon as the water lost the initial milky white color. Another sample was taken of the last water that went through the pipe.

Samples that were taken of water that had passed through the lime were first collected in a polyethylene bottle. Samples were then taken from the collection bottle according to the general sampling method described above.

2.3.3 Olivine and lime experiments with pH measurement

The experimental setups with olivine and lime in the open pipes described above were replicated in the two open pipes. Samples were taken of the water before going through the treatment medium, after 6 L had passed through and after 12 L had passed through. The pH was measured at the time of each sample with a benchtop pH meter, which was first calibrated with standard buffers of pH 4 and 7.

Samples that were taken of water that had passed through olivine or lime were first collected in a polyethylene bottle. Samples were then taken from the collection bottle according to the general sampling method described above.

2.3.4 Turbidity

Untreated water was taken from a hose taking water in from inside the tunnel, added to a vial, and put in the accompanying nephelometer. The turbidity was checked after 0, 10, 20, 30, 40, 50, 60, 120, and 300 seconds. The turbidity of the water at the water's edge inside the tunnel was checked in the same way.

2.3.5 Olivine + activated carbon

Gravel was added and rinsed with the same method as described in the method for the olivine experiment (in section 2.3.1), and added to an open pipe. 8*220 g olivine and 2*56 g activated carbon granules were mixed and added on top. The pipe was filled with approximately 6 L of untreated tunnel water (taken from a tap on Trondheim municipality's system) and the nozzle was slightly opened. A sample was taken of the water before it went through the cleaning medium, and samples were taken of the treated water after 3 L had passed through and after 6 L had passed through. The pipe was then refilled with 6 L more water, and a sample was taken of the last water that passed through.

Samples that were taken of water that had passed through olivine and activated carbon mixture were first collected in a polyethylene bottle. Samples were then taken from the collection bottle according to the general sampling method described above.

2.3.6 Aeration

Approximately 7 L of tunnel water was collected with a bucket a few meters into the tunnel; it had a distinct orange, but still translucent, color. The water was aerated using a portable aquarium pump (Marina battery air pump) with the aeration stone approximately half way down into the water. A filtered and an unfiltered sample was taken of the water before aeration, after 10 minutes and after 40 minutes.

Filtered samples were taken according to the general sampling method described above, and the unfiltered samples were taken in the same way with the exception of the filtration.

2.4 ICP-MS analysis and sample preparation

All water samples were preserved with three drops of ultrapure HNO_3 and analyzed with ICP-MS by Syverin Lierhagen or Anica Simic at NTNU. For more information about the ICP-MS analysis, see appendix C. Blanks were made when a new batch of acid was used for sample preservation. For the first batch of acid, two blanks were made following the same procedure as the sampling, but using MilliQ water instead of sample. The average concentrations in the blanks were used to correct for samples preserved with this batch of acid (1-61 and 1-35, see appendix C). For the second batch of acid, five blanks following the same procedure as the sampling, but using MilliQ water instead of sample, and the average was used to correct for the samples preserved with this batch of acid (36-81 and 82-162, see appendix C).

The first samples (1-61, 1-35, and 36-81, see appendix C) were analyzed with HR-ICP-MS in the instrument ELEMENT 2, while the last samples (82-162) were analyzed in the instrument Agilent 8800.

2.8 Statistics

The following procedure was repeated for each element (Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As) separately. A Shapiro-Wilk test was used to check for normality of the samples taken before and after the gravel was rinsed; three samples before entering gravel, and five samples after being passed through the gravel. The significance level (α) was set to 0.05.

When the distribution was found to be normal, a T-test was used to check for significant differences between the before and after samples. The significance level (α) was set to 0.05.

When the distribution was not found to be normal, a Mann Whitney U test was performed. The significance level (α) was set to 0.05.

3 Results and discussion

In this sections the results will be presented and discussed. After general results and discussion from the experiments are presented, the olivine experiment results will be presented and discussed, followed by the results from samples taken in Trondheim municipality's system when they tested olivine. The results from the experiment with the combination of olivine and activated carbon will then be presented, followed by the aeration experiment. The next section will be the results from the lime experiments. Finally the experiments where the pH was measured will be presented and discussed, followed by the turbidity measurements. Each experiment will be presented with concentrations and treatment efficiencies, and at the end a table summarizing the treatment efficiency in each experiment will be presented.

After the experiments follows a section with results from monitoring, starting with the concentrations of each element of interest in the untreated tunnel water over time. These are presented in graphs, and the lower limit for the different classes in the classification of condition of coastal water are included in the graphs as colored lines. The color of the line corresponds to a class, where green is the lower limit for class II, yellow is class III, orange is class IV, and red is class V. When the concentration of a sample is above a certain line it is in that class.

All presented concentrations are corrected with blank values. For the detailed ICP-MS results with corresponding RSDs, as well as the blanks, see appendix C.

3.1 Experiments

3.1.1 Gravel

To prevent obstruction of the nozzle by the small-grained olivine, lime, and activated carbon, gravel was added before the treatment medium. The gravel was washed with the untreated water from the tunnel. Statistical analyses indicate that there was no significant difference in metal concentration in the water before and after going through gravel. This means that the use of gravel did not affect the results of the experiments, and that any change in metal concentration before and after going through the treatment mediums is due to the medium itself and not the gravel. Results from the gravel washing as well as statistics are presented in appendix D.

3.1.2 Before treatment

Before being treated by any medium the concentrations of the different elements in the untreated water were found. Experiments were carried out in June, September, and December 2019, and February 2020, and Trondheim municipality did large scale treatment attempts continuously throughout this time period. During their treatment attempts they tried different methods and therefore made several changes in their setup. This led to different outlets where untreated tunnel water was collected. In addition the depth of the pipe bringing in tunnel water changed. As a consequence of different changes in Trondheim municipality's system, the condition of the water could vary between experiments. The concentrations of each element before each experiment is presented in table 3.1. The concentrations of the elements in the tunnel water before Trondheim municipality added olivine to their system are also presented in table 3.1. These samples are the before samples later referred to in the results. The results are color coordinated in accordance with table 1.4.

Table 3.1 Concentrations in µg/L of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As of the water in the tunnel before the addition of olivine to Trondheim municipality's system (18.01.19) and before each experiment was performed. The table is color coordinated according to the classification of condition for these elements in coastal water, where blue is class I, green is class II, yellow is class III, orange is class IV, red is class V, and gray is no classification.

	Cd [µg/L]	Pb [µg/L]	Cr [µg/L]	Fe [µg/L]	Ni [µg/L]	Cu [µg/L]	Zn [µg/L]	As [µg/L]
18.01.19	25.1	29.0	2.40	10 000	26.3	8 360	7 220	0.746
06.06.19	28.4	22.9	3.08	10 400	36.9	11 700	8 120	0.624
12.09.19	35.0	33.2	6.22	29 100	33.2	18 100	9 500	10.3
18.12.19	50.0	3.35	0.492	5 700	53.2	20 500	13 200	0.258
24.02.20	12.1	21.6	2.60	9 530	18.2	7 060	3 230	0.692

The classification system for condition can be used to determine the severity of the contamination. It gives an indication of the toxicity of the contaminated water to the aquatic ecosystem near the release. Class I and II are the only classes to not cause any toxicity. Therefore it would be ideal to treat the water so that all concentrations were in class I and II. Because the water is being released into the fjord, there will be some dilution. It might therefore be sufficient to reduce concentrations to class III, assuming the dilution will result in the aquatic environment being exposed to lower concentrations.

The limits for class III, IV, and V for Cd are dependent on the hardness of the recipient water. This was calculated to be 841 mg $CaCO_3$/L, which is >200 mg $CaCO_3$/L, and the upper limit for class II is therefore 1.5 µg/L and for class IV 15 µg/L. Further information and calculations are presented in appendix B.

3.1.3 Olivine

The intention of the olivine experiments was to investigate the capacity and efficiency of olivine on removing Cu, Zn, Cd, Cr, As, Ni, Pb and Fe from the polluted water at Killingdal. The aim was to do experiments using both gravity where water is running through the treatment medium only due to gravity, and a pump. The experiment using the pump led to precipitation clogging the pipe. Therefore only two successful experiments were realized; both with olivine using gravity. The results of the first experiment will be presented in this section. In the second experiment pH was measured, and these results will be presented further below in section 3.1.8.

Four samples were taken during treatment with olivine in this experiment; one after approximately 0.6 L had passed through the olivine (hereby referred to as 1st), one after 3 L had passed through (2nd), one after 6 L had passed through (3rd), and one when 12 L had passed through (4th). The results are presented in figure 3.1-3.3 with elements in similar concentration ranges presented together.

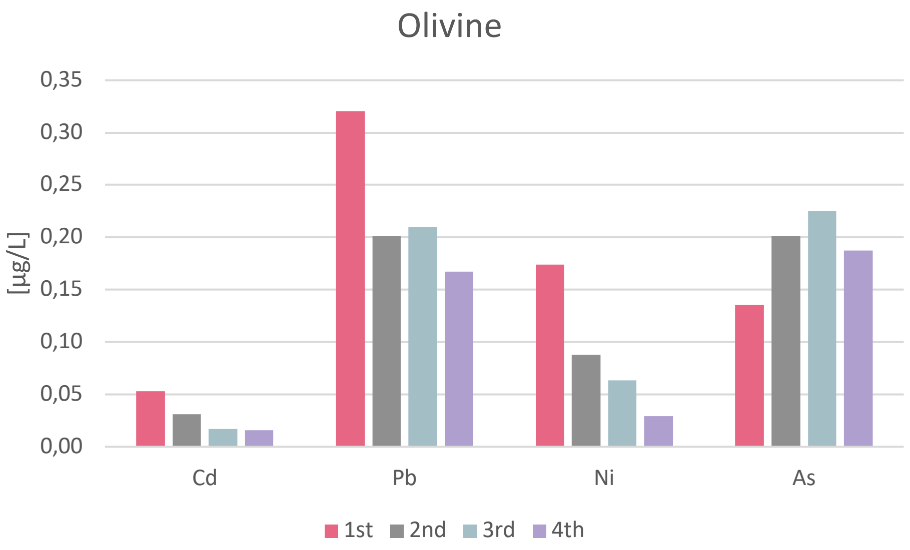

Fig. 3.1 Concentrations of Cd, Pb, Ni, and As in the water after being treated with olivine. Samples were taken after 0.6 L (1st), 3 L (2nd), 6 L (3rd), and 12 L (4th) of water had passed through the olivine.

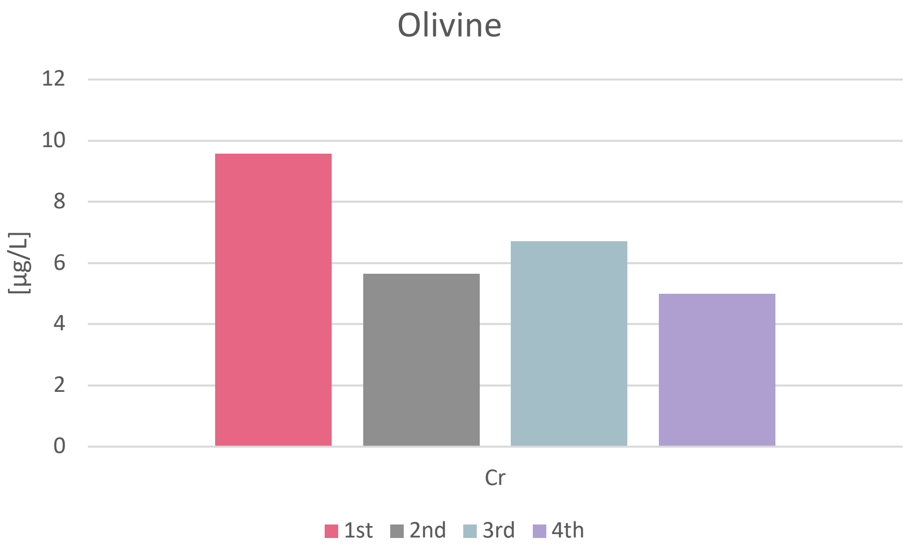

Fig 3.2 Concentration of Cr in the water after being treated with olivine. Samples were taken after 0.6 L (1st), 3 L (2nd), 6 L (3rd), and 12 L (4th) of water had passed through the olivine.

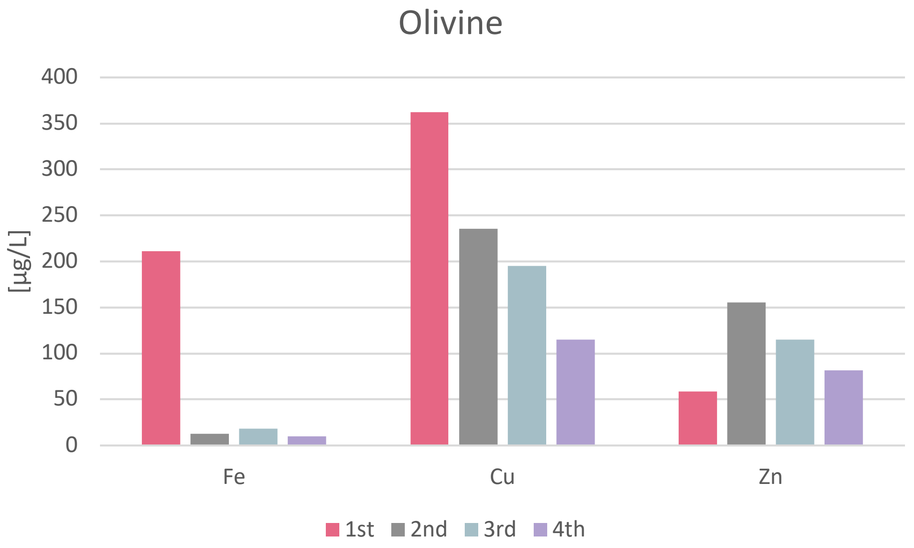

Fig 3.3 Concentrations of Fe, Cu, and Zn in the water after being treated with olivine. Samples were taken after 0.6 L (1st), 3 L (2nd), 6 L (3rd), and 12 L (4th) of water had passed through the olivine.

The first observation to be noted about these results is the magnitude of contamination. Fe, Cu and Zn levels are up to 38 times higher than the highest Cr concentration, which is the highest of the other elements. Another observation is that the trends of the concentrations (i.e. if they are declining, rising or other) are different for the different elements. This shows that the elements react differently to the olivine. This may be due to different affinities to the olivine surface, differences in solubility at the pH the olivine increases the water to, or it may also just be variations in the samples taken, due to spatial variation of the ions.

This experiment looks at olivine's capacity to remove the selected metals from the water. New water is continuously passing through the medium, but we can still say something about olivine's capacity over time. In figure 3.3 it can be seen that Fe is about 15 times lower in the 2nd to 4th samples than in the 1st sample, even though they are all samples of treated water. Fe(III) is expected to precipitate at pH above approximately 3.5, and the olivine is expected to raise the pH of the solution. One explanation in the difference in Fe concentration between the 1st sample and the others could be that the Fe has not precipitated as much in the 1st sample as in the others. When olivine is exposed to acid it will dissolve, releasing Mg(II) and Fe(II) to the solution, and capturing H^+ as H_4SiO_4, consequently raising the pH. This results in an increase in Fe(II) concentration. If the Fe(II) is oxidized to Fe(III) it can precipitate as hydroxides. The oxidation of Fe(II) to Fe(III) has a half-life of 30 minutes at neutral pH, but becomes slow at pH below 4.[44] Because of the high concentrations in the other metals, there might have been competition between the H^+ ions and the metals. This will lead to a slower increase of pH, and if the pH is lower in the 1st sample than in the rest, then the Fe(II) might not have been oxidized to Fe(III) to the same degree. This could explain the difference in Fe concentration between the 1st sample and the others.

The concentrations from the treatment experiment were compared to the concentrations before treatment, and the treatment efficiency was calculated in percent. Calculations are shown in Appendix B. The results are presented in % change in metal concentration after treatment. Positive numbers indicate a decrease in concentration and negative numbers indicate an increase. All values are related to the concentrations in the sample taken before any treatment. The treatment efficiency of olivine in this experiment is shown in table 3.2.

Table 3.2 Treatment efficiency of olivine on the elements Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As as % at four different times in the treatment process; after 0.6 L (1st), 3 L (2nd), 6 L (3rd), and 12 L (4th) of water had passed through the olivine. The results are presented in % change in metal concentration after treatment, compared to before, and are based on concentration (µg/L). All values are related to the concentrations in the sample taken before any treatment. The results are color coordinated where green means that the treatment efficiency of olivine on this particular element was >95%, yellow means <95%, but still positive, and red means negative.

	Cd	Pb	Cr	Fe	Ni	Cu	Zn	As
1st	99.8	98.6	-211	98.0	99.5	96.9	99.3	78.3
2nd	99.9	99.1	-83.5	99.9	99.8	98.0	98.1	67.7
3rd	99.9	99.1	-118	99.8	99.8	98.3	98.6	63.9
4th	99.9	99.3	-62.3	99.9	99.9	99.0	99.0	69.9

From table 3.2 it can be observed that olivine seems to have a relatively good capacity to remove several elements. An increase in Cr is however observed. The binding material in this olivine is Cr-free,[77] but previous studies have shown that the olivine granules lead to an increase in Cr.[41,46] According to Weim-Andersen et al., X-ray diffraction (XRD) determined that the Cr-free granules may contain approximately 3% cement. This cement is used as a binder and contains Cr.[41] It may therefore seem that the granules may still include some chromiferous cement. Former olivine products have used chromiferous cement as a binder, but the granules used in this thesis use a Cr-free binder.[77] If these olivine granules contain Cr-free binder as they should, then the increase in Cr must come from other sources. Cr can exist in the olivine mineral structure as substitutions, but the levels are small.[78] It is therefore unlikely that mineral inclusions can explain the increase in Cr. A possible explanation is that Cr in particulate form dissolves because of the changes in water chemistry. Cr(III) will precipitate as hydroxide when the pH increases, but if the pH increases above 12, the occurrence of Cr(III) hydroxide is negligible.[79] In this case it will likely be complexed by four hydroxides, due to the high presence of hydroxides at high pH, and be $Cr(OH)_4^-$. Therefore it is likely that the Cr precipitates became dissolved, if the olivine raised the pH too high. This would insinuate that the Cr was on particulate form before treatment. This is unlikely considering the low pH of AMD, however the pH of the water was not tested on this day. The water inside the tunnel is a complex solution with many different elements in high concentrations. This makes it more difficult to predict and understand the reactions that occur. Maybe, because of the high concentrations, there is a competition between Cr and other elements over ligands, like hydroxide, and the precipitated Cr dissolves because of it. Maybe Cr(III) becomes oxidized to Cr(VI), which is more soluble in water. If olivine is to be used as a treatment medium for this water it is important to monitor Cr particularly, to make sure that the concentration does not exceed unacceptable levels. It would be preferable to combine olivine with another treatment step that removes Cr, or a pre-treatment step to eliminate this problem if possible.

Looking at the treatment efficiency, As may also need extra attention. A possible solution that may improve the removal of As is pre-treatment with aeration. This can remove As by co-precipitation, as well as adsorption onto precipitated $Fe(OH)_3$.[27] Aeration was later investigated and the results are presented below in section 3.1.6.

Even though olivine has promising results based on its treatment efficiency it does not treat the water well enough on its own. To define the environmental status of water we use the classification of condition provided by the Norwegian Environment Agency.[58] The classes are based on concentration, and in this case the classification of coastal water will be used since the final recipient of the treated water will be coastal water. The classification of the water before and during treatment with olivine is shown in table 3.3.

Table 3.3 The concentrations, in µg/L, of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As before treatment with olivine and during four steps of the treatment process; after 0.6 L (1st), 3 L (2nd), 6 L (3rd), and 12 L (4th) of water had passed through the olivine. The table is color coordinated according to the classification of condition for these elements in coastal water, where blue is class I, green is class II, yellow is class III, orange is class IV, red is class V, and gray is no classification.

	Cd [µg/L]	Pb [µg/L]	Cr [µg/L]	Fe [µg/L]	Ni [µg/L]	Cu [µg/L]	Zn [µg/L]	As [µg/L]
Before	28.4	22.9	3.08	10400	36.9	11700	8120	0.624
1st	0.0528	0.321	9.57	211	0.174	362	58.8	0.135
2nd	0.0312	0.202	5.65	12.4	0.0878	236	155	0.201
3rd	0.0169	0.210	6.71	18.3	0.0633	195	115	0.225
4th	0.0159	0.168	5.00	10.3	0.0290	115	81.9	0.188

Comparing table 3.2 and 3.3, the severity of the degree of pollution becomes evident, especially for Cu and Zn. These elements are in class V both before and after treatment with olivine, but looking at table 3.2 we can see that olivine removed at least 96.9% Cu and 98.1% Zn in this experiment. These results emphasize the high levels of Cu and Zn and the fact that removing most of the contamination is still not enough to reach an acceptable level to release into the fjord. These results accentuate again the need for additional treatment steps.

Olivine seems to be a promising choice for a system to clean this water adequately so that it can be released back into the fjord, but other steps, either before, after or in combination with olivine, need to be included. To my knowledge no one has looked into the efficiency of trace metal removal from AMD using olivine in combination with other steps. This should be researched in the future to see if even better results can be achieved.

The treatment efficiency of As was lower than the other elements (except for Cr), but looking at table 3.3 it can be observed that As concentrations are in class II and I after treatment with olivine. This indicates that olivine removes enough As, despite the lower efficiency. Olivine's treatment efficiency of Ni causes a decrease in concentration from class IV to class I. Class I is background levels, and being able to remove Ni from concentrations potentially causing acute toxic effect by short-term to background levels is very promising. Cd is also found in class I in the 3rd and 4th samples, lowering the concentrations from potentially causing extensive toxic effects, to background levels. Concentrations of Pb, and Cd concentrations in the 1st and 2nd samples, are decreased to class II, which will cause no toxic effect. These results are very promising regarding olivine as a potential treatment medium for this water.

This experiment was conducted with gravity, and an experiment attempting to treat water with olivine over time using a pump, failed because of precipitation clogging the system. This precipitation was a combination of orange and white solids, which may be Fe hydroxides and Mg hydroxide. The precipitation problem that arose might have been prevented if water was being pumped up through the olivine and not down. This would allow for mixing of the medium, which is beneficial for two reasons; it would allow the water to come into contact with more of the surface area of the olivine, and a layer of precipitation might not be formed on top. If a precipitation layer still was to form on top of the olivine, the water would break through it more easily upwards than when the water is pushing down and hitting the precipitation layer before the olivine. The precipitation would also most likely form inside the olivine layer, not on top.

Looking at the results presented in figure 3.1, 3.2, and 3.3 it seems like there is some variation in the four samples (1st, 2nd, 3rd, and 4th), but no general trends. An experiment testing olivine over time, with samples taken regularly will give a better view into how its treatment efficiency holds up over time. This should be tested in the future before making any final conclusion about olivine's potential use as a treatment step in treating the water at Killingdal. A previous study testing olivine's ability to adsorb Cu was run for more than 300 hours, and it was found that the treatment efficiency remained 99% for the first 300 hours, but started to decrease dramatically after this.[77] A suggested reason for this decrease is that the internal permeability of the granules becomes lower over time, and therefore the contaminated water is only in contact with the outer surface of the olivine granules. This means less surface and fewer sites for the Cu to bind to. It may also suggest that olivine became saturated in regard to adsorption sites after 300 hours.

3.1.4 Olivine in Trondheim municipality's treatment system

In the period from 29.01. to 22.03. olivine was tested by Trondheim Municipality in their system. The day the olivine was added, there was no way to sample the tunnel water coming into the system, therefore a sample of the tunnel water from 18.01.19 has been used as the before sample. On the day olivine was added, two samples were taken; one after approximately 5 minutes and one after 20 minutes. Between these two samples someone walked inside the tunnel disturbing the sedimented precipitations. This caused a difference in the water coming into the system, visible by the bright orange colored water entering the system after the disturbance, and the sample taken after 20 minutes has therefore not been included in the graphs. Figure 3.4 to 3.11 represent the concentrations of the different elements in Trondheim municipality's system when olivine was used as the treatment medium. The purple line is the day when the olivine was added to the system. The concentrations in the samples taken on this day (approximately 5 minutes after the addition of olivine) are sometimes too small to be visible, but are all presented in the figures right next to the purple line (to the left, even though the sample was taken after addition of olivine).

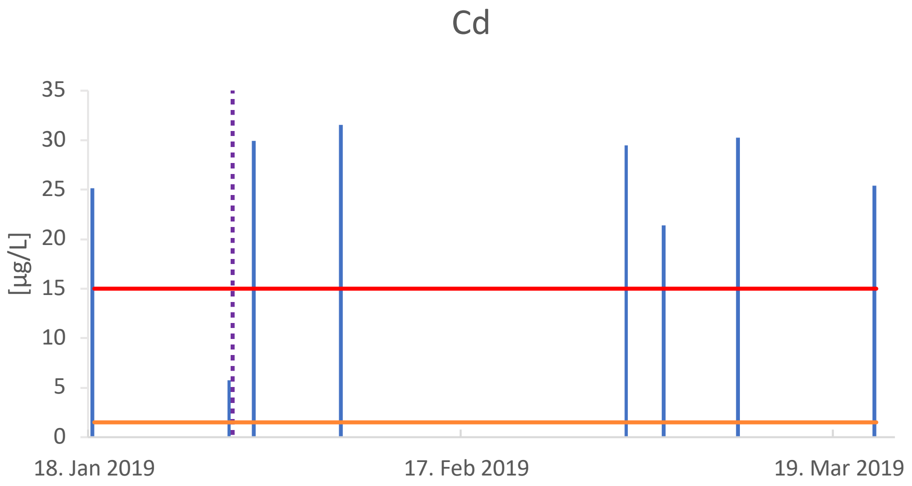

Figure 3.4 The concentration of Cd in Trondheim municipality's system after the addition of olivine. The purple line represents the day olivine was added to the system, and the sampling point before this represents the concentration of Cd in the untreated tunnel water. Colored lines define the lower limit for a class in the classification of condition for coastal water, where orange is class IV and red is class V.

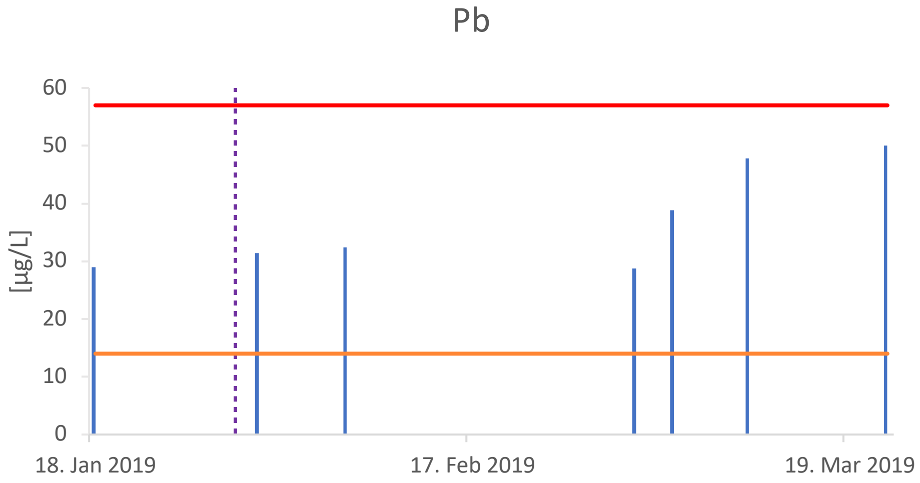

Figure 3.5 The concentration of Pb in Trondheim municipality's system after the addition of olivine. The purple line represents the day olivine was added to the system, and the sampling point before this represents the concentration of Pb in the untreated tunnel water. Colored lines define the lower limit for a class in the classification of condition for coastal water, where orange is class IV and red is class V.

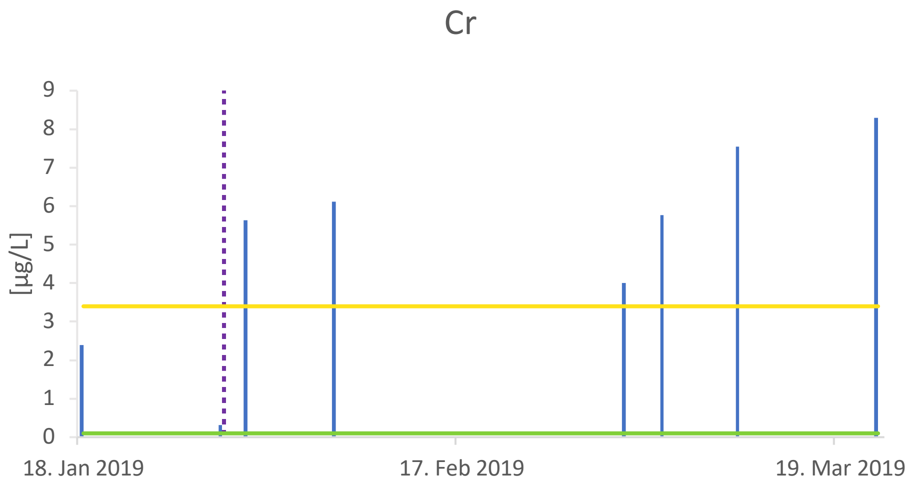

Figure 3.6 The concentration of Cr in Trondheim municipality's system after the addition of olivine. The purple line represents the day olivine was added to the system, and the sampling point before this represents the concentration of Cr in the untreated tunnel water. Colored lines define the lower limit for a class in the classification of condition for coastal water, where green is class II and yellow is class III.

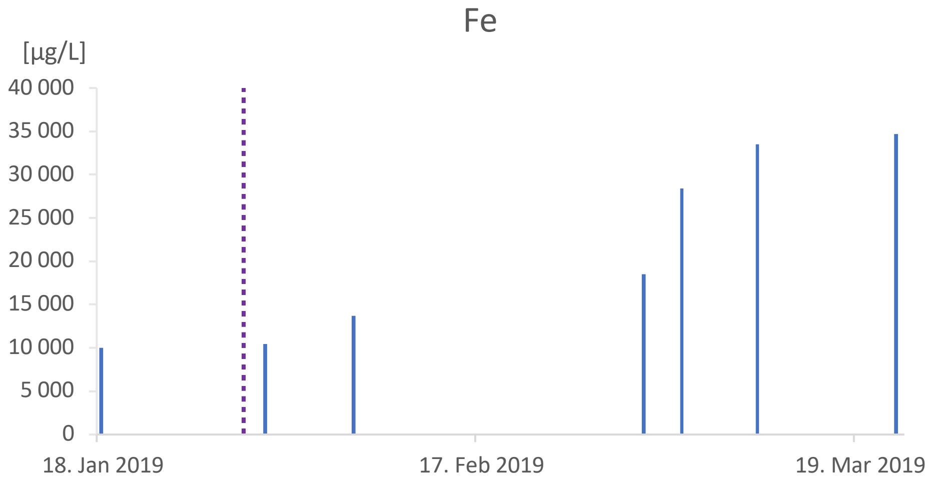

Figure 3.7 The concentration of Fe in Trondheim municipality's system after the addition of olivine. The purple line represents the day olivine was added to the system, and the sampling point before this represents the concentration of Fe in the untreated tunnel water.

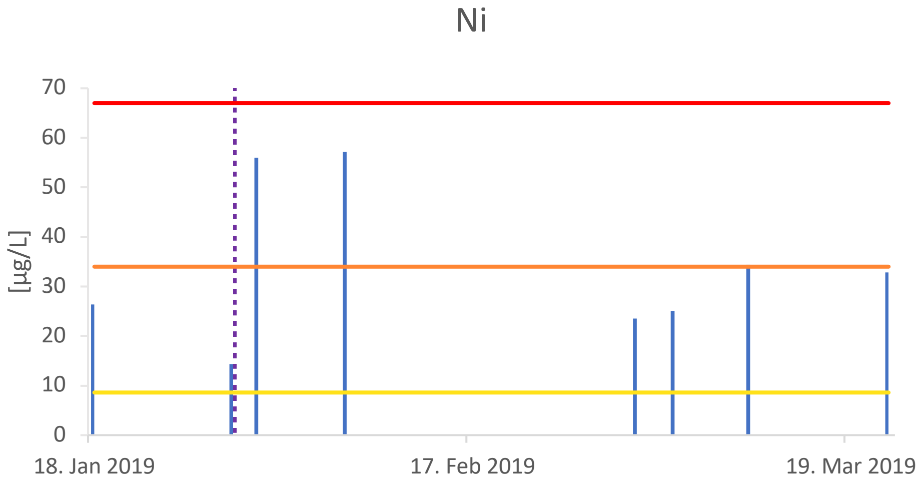

Figure 3.8 The concentration of Ni in Trondheim municipality's system after the addition of olivine. The purple line represents the day olivine was added to the system, and the sampling point before this represents the concentration of Ni in the untreated tunnel water. Colored lines define the lower limit for a class in the classification of condition for coastal water, where yellow is class III, orange is class IV, and red is class V.

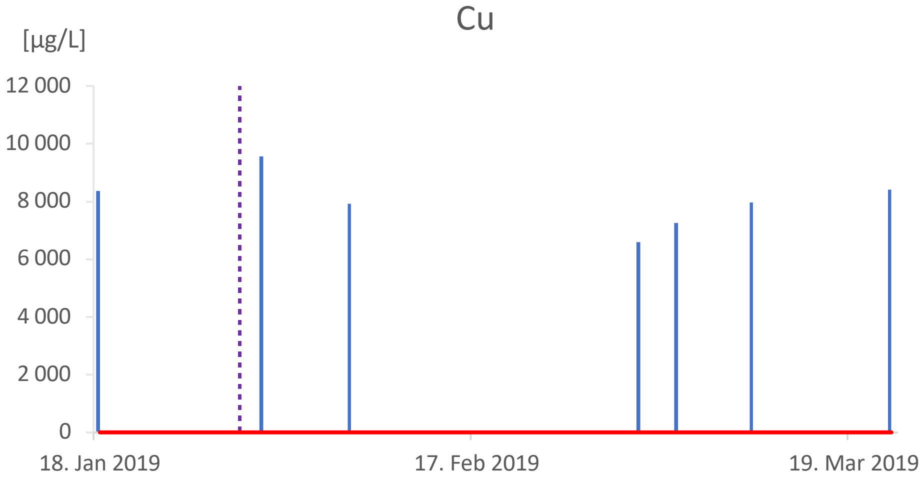

Figure 3.9 The concentration of Cu in Trondheim municipality's system after the addition of olivine. The purple line represents the day olivine was added to the system, and the sampling point before this represents the concentration of Cu in the untreated tunnel water. The red line defines the lower limit for class V in the classification of condition for coastal water.

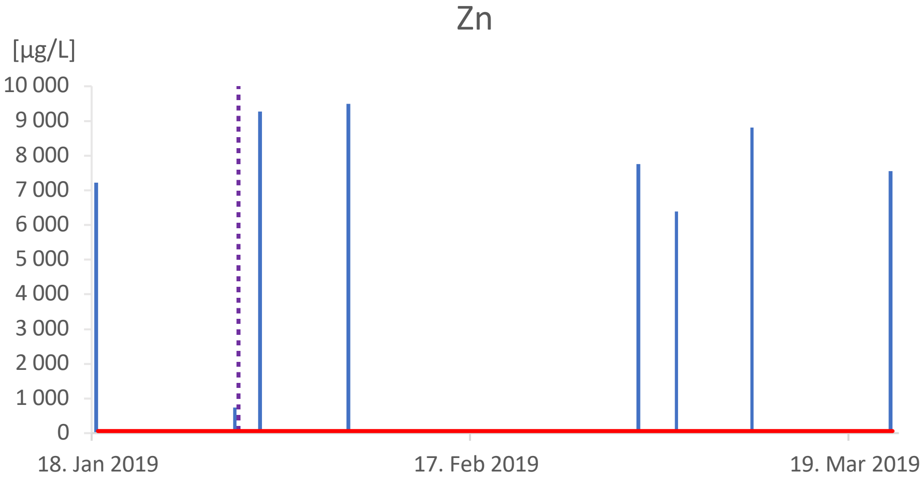

Figure 3.10 The concentration of Zn in Trondheim municipality's system after the addition of olivine. The purple line represents the day olivine was added to the system, and the sampling point before this represents the concentration of Zn in the untreated tunnel water. The red line defines the lower limit for class V in the classification of condition for coastal water.

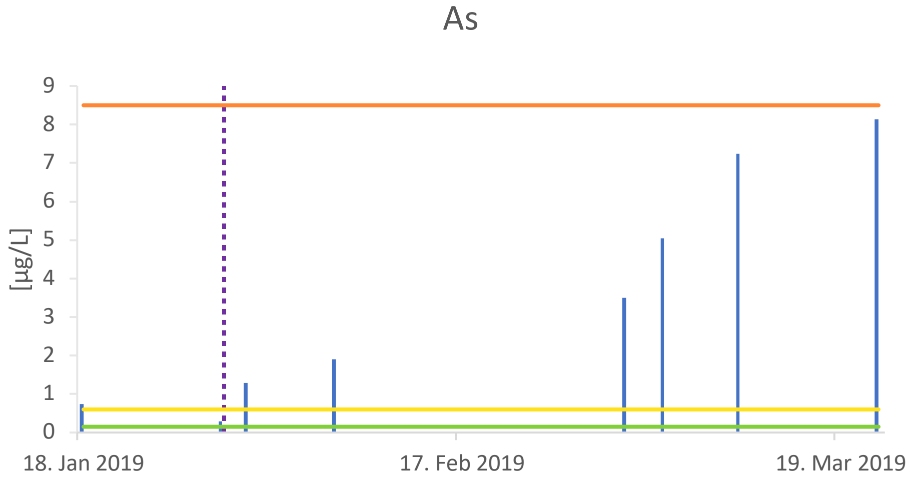

Figure 3.11 The concentration of As in Trondheim municipality's system after the addition of olivine. The purple line represents the day olivine was added to the system, and the sampling point before this represents the concentration of As in the untreated tunnel water. Colored lines define the lower limit for a class in the classification of condition for coastal water, where green is class II, yellow is class III, and orange is class IV.

The concentration of all of the elements drops with the addition of olivine. The sample taken the same day that olivine is added to the system is taken only five minutes after the olivine has been added, and therefore all of the active sites are available for adsorption. In addition there will be some olivine dust among the granules, and this provides an even greater surface area. This effect will wear off as the dust is washed out from the system. Looking at the concentration of the elements on the next sampling date, 31.01., it seems that olivine's effect on the metal concentration has worn off. In all of the samples taken after 29.01., all of the elements are found in concentrations that are approximately the same as before treatment with olivine, and most are found in higher concentrations. This indicates that using olivine over time did not work on the levels of contaminations in water at that time. The concentrations of all of the elements in the untreated tunnel water 18.01., before the addition of olivine, can be found in table 3.1. The concentrations before treatment in the olivine experiment can also be found in table 3.1., under the date 06.06.19, which is the date the experiment was performed. Comparing the concentrations of the elements 18.01.19 and 06.06.19 it can be observed that they are similar. Since the composition of the water before olivine was added to Trondheim municipality's system 18.01.19 and before treatment with olivine in the experiment 06.06.19 were similar, it could be assumed that the efficiency of olivine would be approximately the same. One observed exception is Cr, which increases in the experiment 06.06.19, but decreases 29.01. The dimensions of the experiment and of Trondheim municipality's system were not measured, and were therefore not the same. The municipality passed larger amounts of water through the olivine. The effect of olivine over time using more olivine than was added 29.01.19 should be investigated to see if more olivine will make the effect last longer. It is also important to keep in mind that the before sample used in figure 3.4-3.11 is not taken on the same day that the olivine was added to the system. There were no major changes in the system between these two dates, but it cannot be said with certainty that the composition of the metals in the water was the same 18.01. and 29.01 before olivine.

The effect of the olivine has most likely been used up some time between 29.01. and 31.01. This may be due to the extent of pollution in the water and that olivine's active sites where adsorption occurs are saturated. A possible explanation could be that initially the metal ions were adsorbed by the olivine, but over time, since this water is AMD and is acidic, they were replaced by H^+ ions. The amount of olivine used was not enough to raise the pH of the whole tunnel system, so when the active sites were filled with H^+ there was no metal retention, but also no further increase in pH. The adsorption of metal ions onto the surface of olivine increases with increasing pH. A suggestion could therefore be that the pH should be raised before using olivine as a treatment medium, and that even though olivine has the ability to raise the pH in a solution, it did not have the capacity to do so on a large scale, and therefore the efficiency of the olivine wore off relatively fast. The capacity for olivine to increase pH was investigated in another experiment, and the results are presented in figure 3.18-3.20.

Looking at figure 3.7 and 3.11 it can be seen that Fe and As follow the same trend. This is due to the affinity of As to Fe. It is known that dissolved As is highly controlled by Fe hydroxides, both through co-precipitation and adsorption. If the pH decreases Fe hydroxide will dissolve and As will desorb, or the co-precipitated As will be released. From the figures it becomes clear that as olivine's effect on the water wears off, the dissolved Fe concentration increases, and the As concentration follows the same pattern of increase. This indicates that Fe and As were removed from the solution by olivine due to an increase in pH and subsequent Fe hydroxide precipitation.

Because olivine seems like a promising treatment medium, but other steps are needed, two additional experiments were conducted. The combination of olivine and activated carbon was investigated, and aeration was tested because of its potential use as a pre-treatment step. The results from these experiments are presented below.

3.1.5 Combination of olivine and activated carbon

Looking at the treatment efficiency there is a lot of potential for olivine, but resulting classes of condition suggest that another step needs to be added in the treatment process to end up with concentrations that can be released into the fjord. In addition, the negative impact of added Cr needs to be counteracted. One possible solution can be to have a polishing step after treatment with olivine. Previous studies have found that activated carbon can adsorb metals from aqueous solutions.[56,80,81] Using activated carbon as a polishing step in a potential treatment system for the AMD at Killingdal is being investigated by another master student.[82] To explore the possibility of removing one step, the mixture of olivine and activated carbon was looked into. The experiment was performed in the same way as the olivine experiment, using one of the open pipes, but with a mixture of olivine and activated carbon as the treatment medium. In theory, the activated carbon in the mixture should adsorb the Cr being released by the olivine. In addition the treatment efficiency of As was hoped to increase.

The concentrations from the olivine and activated carbon treatment experiment were compared to the concentrations before treatment, and the treatment efficiency was calculated in percent. The results are presented in % change in metal concentrations after treatment. Positive numbers indicate a decrease in concentration and negative numbers indicate an increase. All values are related to the concentrations in the sample taken before any treatment. The treatment efficiency of the olivine and activated carbon mixture on each element is presented in table 3.4.

Table 3.4 Treatment efficiency of the mixture of olivine and activated carbon on the elements Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As as % at three different times in the treatment process; after 3 L (1st), 6 L (2nd), and 12 L (3rd) of water had passed through the olivine and activated carbon mixture. The results are presented in % change in metal concentration after treatment, compared to before, and are based on concentration (µg/L). All values are related to the concentrations in the sample taken before any treatment. The results are color coordinated where green means that the treatment efficiency of olivine on this particular element was >95%, yellow means <95%, but still positive, and red means negative.

	Cd	Pb	Cr	Fe	Ni	Cu	Zn	As
1st	99.9	97.5	-1900	99.7	99.1	99.2	98.9	-686
2nd	99.9	98.2	-1860	99.8	99.7	99.3	98.8	-568
3rd	99.9	98.3	-702	99.9	99.7	99.8	99.4	-123

This experiment shows that a mixture of olivine and activated carbon does not improve the treatment efficiency on Cr and As, but rather worsens it. This is an unfortunate effect and it seems that the combination of olivine and activated carbon should not be considered as a treatment step for this water.

It should be noted that the olivine experiment presented in section 3.1.3 and the mixture experiment were carried out on different dates; the first one in June and the second one in December. The concentrations of the different elements on these dates are presented in table 3.1. It is clear to see that there is a difference. This difference may be due to altered physical and chemical properties in the water, and may come from Trondheim municipality's treatment attempts carried out between these dates. Changes in the water may both increase and decrease the concentration of elements. If the pH rises, more will precipitate. Each metal has a different optimal precipitation pH, so they may not all have precipitated. During precipitation co-precipitation and adsorption on the surface of precipitates occurs. This may lead to elements being removed from the water in a different degree of stability. Adsorption onto surfaces may be unstable, and therefore adsorbed metals may desorb if the precipitation is disturbed.

In addition to the changes that may have happened to the water between the two experiments, there was a disturbance of the sedimented precipitates on the day of the experiment with the olivine and activated carbon mixture in December. People were walking inside the tunnel right before the experiment was performed. This will disturb the settled precipitation, and because the texture of the precipitation is light and fluffy, the particles will not have had time to settle before water is sampled from the tunnel and used in the experiment. Metals that are weakly adsorbed to the surface of the precipitations may become desorbed and released back into the water.

The resulting concentrations after treatment with the olivine and activated carbon mixture are shown in table 3.5.

Table 3.5 The concentrations, in µg/L, of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As before treatment with a mixture of olivine and activated carbon, and during three steps of the treatment process; after 3 L (1st), 6 L (2nd), and 12 L (3rd) of water had passed through the olivine. The table is color coordinated according to the classification of condition for these elements in coastal water, where blue is class I, green is class II, yellow is class III, orange is class IV, red is class V, and gray is no classification.

	Cd [µg/L]	Pb [µg/L]	Cr [µg/L]	Fe [µg/L]	Ni [µg/L]	Cu [µg/L]	Zn [µg/L]	As [µg/L]
Before	50.0	3.35	0.492	5700	53.2	20500	13200	0.258
1st	0.0571	0.0824	9.83	16.3	0.500	165	140	2.03
2nd	0.0369	0.0589	9.64	9.27	0.136	153	159	1.72
3rd	0.0390	0.0573	3.94	6.28	0.156	45.3	78.1	0.576

Looking at table 3.5 it can be noted that the increase in Cr and As caused them to be found with concentrations in class III. This is a class higher than the concentrations before treatment, but it is still not as dramatic as the treatment efficiency suggests. Since olivine had a higher efficiency for Cr and As, and similar efficiencies for the other elements, the use of olivine mixed with activated carbon shows no benefits over using olivine alone. Even though the mixture did not fix the issue with Cr release or ameliorate the treatment efficiency of As, it does not mean that activated carbon cannot be used as a polishing step. This experiment was conducted to investigate the possibility of eliminating an extra treatment step by combining two steps. The results indicate that this is not a preferable option. The use of activated carbon should be investigated as a possible adsorbent in a polishing step following treatment with olivine.

3.1.6 Aeration

An alternative, or addition, to a polishing step is to implement a pre-treatment step. One option is aerating the water. By adding oxygen into the water, the reduction potential is changed. More oxygen present makes the conditions more oxidizing, and several elements may be oxidized and precipitate as hydroxides. An aeration step may in particular remove some of the Fe in the solution, as Fe hydroxides. Precipitation of Fe is beneficial in several different ways. By precipitating before going through the cleaning medium, clogging of the cleaning medium may be prevented. In addition, the precipitation of Fe hydroxides may cause the co-precipitation of other elements. If the co-precipitation is stable, those elements will also be removed from the water column. Kleiv et al. studied the adsorption of Cu on olivine in synthetic mine water, and found that when excluding Fe they reached higher pH and better retention values than when including Fe in the synthetic mine water. They concluded that if olivine is to be used to remediate mine water, in their case at Løkken mines, olivine loaded with Cu needs to be separated from the Fe rich solution to avoid desorption.[43] Pre-treatment with aeration removing Fe from the water could increase olivine's treatment efficiency even further.

Another benefit of precipitation Fe is the adsorptive qualities of the precipitated Fe. Other elements may be removed from the solution if the adsorption onto Fe particles is strong and stable. In an environment as polluted as the one at Killingdal, the removal of some contaminants will benefit the following treatment with a treatment medium. Pre-treatment with aeration will improve a following treatment step with an adsorbent, due to the resulting less stable forms of metals. In the case of olivine, its treatment efficiency was high, but the complication was the amount of metals present. If some of these can be removed, or made less stable, prior to the treatment with olivine, and the efficiency is upheld, olivine is an even more promising treatment medium. The results from the experiment with aeration are presented in table 3.6 and 3.7 as treatment efficiencies and concentrations, respectively.

Table 3.6 Treatment efficiency of aeration of the water for the removal of the elements Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As as % after 10 and 40 minutes of aeration. The efficiency is based on concentration (µg/L) and on filtered samples. All values are related to the concentrations in the sample taken before any treatment. All values are related to the concentrations in the sample taken before any treatment. The results are color coordinated where green means that the treatment efficiency of aeration on this particular element was >95%, yellow means <95%, but still positive, and red means negative.

	Cd	Pb	Cr	Fe	Ni	Cu	Zn	As
After 10 minutes	1.94	2.47	-4.97	1.75	4.50	2.21	0.932	-13.2
After 40 minutes	-0.321	0.243	-1.35	1.07	-0.690	-0.259	-0.647	-18.5

Table 3.7 The concentrations, in µg/L, of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As in filtered samples before aeration and after 10 and 40 minutes. The table is color coordinated according to the classification of condition for these elements in coastal water, where blue is class I, green is class II, yellow is class III, orange is class IV, red is class V, and gray is no classification.

	Cd [µg/L]	Pb [µg/L]	Cr [µg/L]	Fe [µg/L]	Ni [µg/L]	Cu [µg/L]	Zn [µg/L]	As [µg/L]
Before	12.1	21.6	2.60	9530	18.2	7060	3230	0.692
After 10 minutes	11.8	21.0	2.73	9370	17.4	6910	3200	0.784
After 40 minutes	12.1	21.5	2.64	9430	18.3	7080	3250	0.820

These results indicate that there was virtually no change in concentration before and after aeration. As increased by up to 18% which was the most negative result. The most positive result is for Ni which decreased by 4.50% after 10 minutes, but this effect was lost after 40 minutes where Ni had increased by 0.690% from the before concentration. Theoretically it was expected that Fe would precipitate and co-precipitate or adsorb some of the other elements. In addition it was expected that As would be removed, by co-precipitation and adsorption onto Fe hydroxides or as an arsenate phase (AsO_4^{3-}), not increase in concentration.

In a study of the removal of As from AMD Yuan et al. found that the removal efficiency was highly dependent on pH.[83] After aerating the water for 40 minutes the removal efficiency of As was under 10% at pH 2, but at pH 7 it was found to be over 95%. The experiment reported by Yuan et al. was performed in a laboratory, and with higher temperatures, so it is not directly comparable to the aeration experiment presented in this thesis. However the pH dependence of As removal is still relevant. In this study aeration caused the release of As. This may not be explained by the pH, but the absence of efficient As removal may suggest that the pH was too low. The pH was not measured on the day of the aeration experiment. A future experiment with aeration in combination with pH increasing mediums like olivine and lime should be conducted, and the removal efficiency, particularly of Fe and As should be investigated. In theory aeration could be used as a pre-treatment of metal removal of AMD, but it might need to be combined with a neutralizing agent that increases the pH.

Another possible reason for the inefficiency of the aeration in this experiment is that the conditions of the water may already have been oxic enough to cause the precipitation of Fe hydroxide. Increasing the oxygen makes the environment more oxidizing, but if the water is already oxic, aeration will not have any effect.

Unfiltered samples were also taken before and during aeration, but due to sample loss of the before sample during sample preparation, the unfiltered samples are not presented here. The results of these samples can be found in appendix C.

3.1.7 Lime

In addition to olivine, lime was also investigated as a possible treatment medium. Its abilities to raise the pH and consequently remove metal contamination by precipitation is well known.[38,48] Two experiments were performed with lime, one with one of the open pipes and one with the pipe connected to the pump, and the experiment over time using a pump was successful as opposed to the equivalent experiment with olivine, where precipitation caused the system to clog. Another lime experiment in an open pipe was performed where pH was measured, and these results will be presented further below in section 3.1.8.

The first experiment is comparable to the successful olivine experiment; it is only gravity dependent. A sample was taken of the first water to pass through the lime that did not have a white milky color, and another sample was taken after 6 L had passed through the lime. Concentrations of the different elements during treatment of lime are presented in figure 3.12-3.14 with elements in similar concentration ranges presented together.

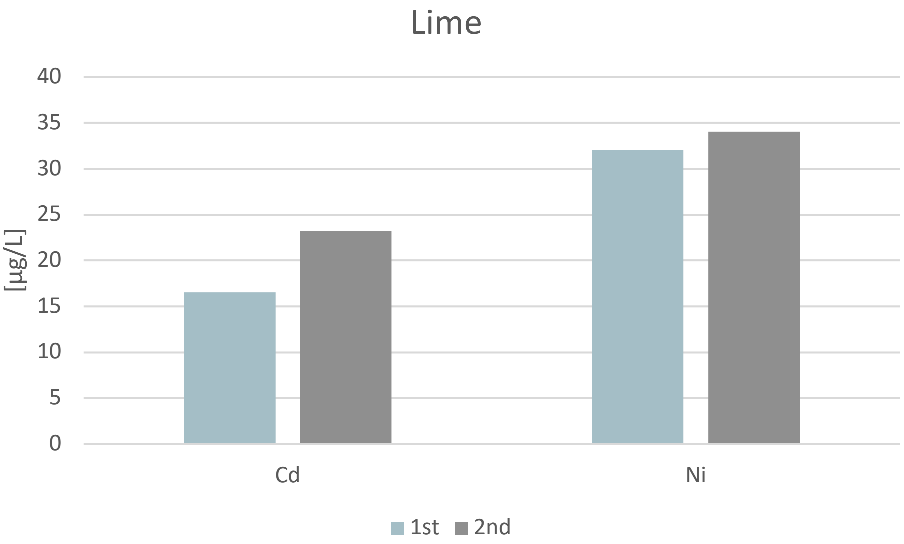

Figure 3.12 The concentrations of Cd and Ni in the first water to pass through the lime that did not have a white milky color (1st) and after 6 L had passed through the lime (2nd).

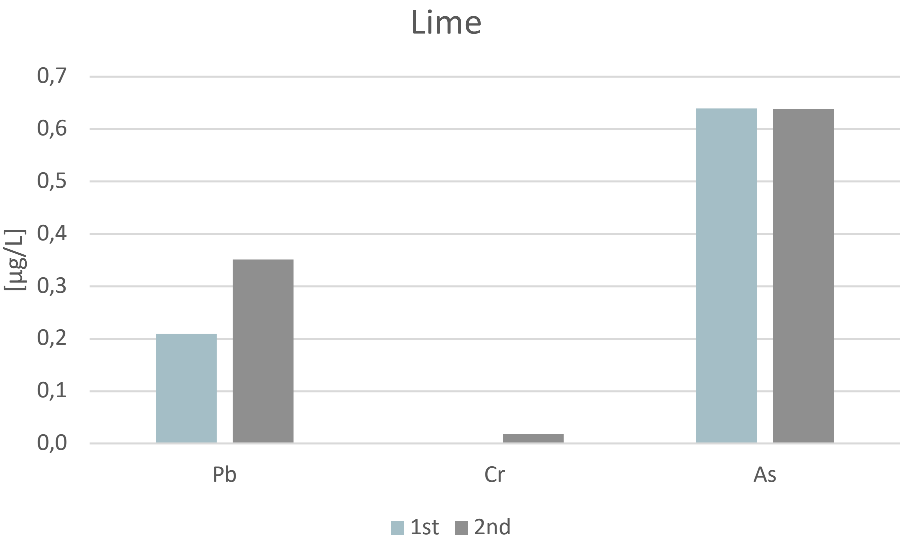

Figure 3.13 The concentrations of Pb, Cr and As in the first water to pass through the lime that did not have a white milky color (1st) and after 6 L had passed through the lime (2nd).

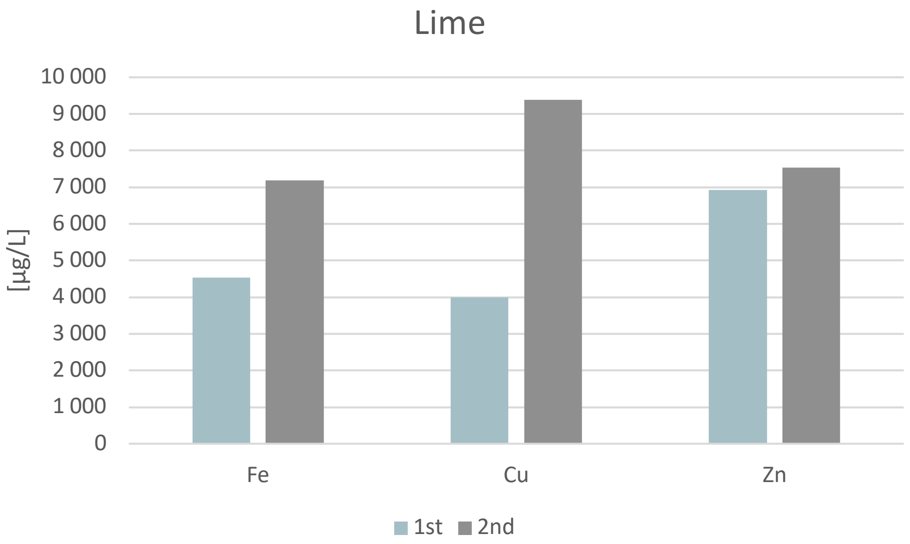

Figure 3.14 The concentrations of Fe, Cu and Zn in the first water to pass through the lime that did not have a white milky color (1st) and after 6 L had passed through the lime (2nd).

Looking at figure 3.12-3.14 one trend can be observed; Cd, Fe, and Cu, and to some degree Ni, Pb, and Zn have lower concentrations in the first sample than in the second one. The first sample was taken shortly after the experiment started. When the first water passes through the lime there will be some lime dust among the grains. The small size of the dust particles might provide rapid dissolution and therefore a higher initial pH increase. This effect will wear off as the dust is washed out from the system.

The concentrations from the lime treatment experiment were compared to the concentrations before treatment, and the treatment efficiency was calculated in percent. The results are presented in % change in metal concentrations after treatment. Positive numbers indicate a decrease in concentration and negative numbers indicate an increase. All values are related to the concentrations in the sample taken before any treatment. The treatment efficiency of lime on each element is presented in table 3.8.

Table 3.8 Treatment efficiency of lime of the elements Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As as % after the first water to pass through the lime that did not have a white milky color (1st) and after 6 L had passed through the lime (2nd). The results are presented in % change in metal concentration after treatment, compared to before, and are based on concentration (µg/L). All values are related to the concentrations in the sample taken before any treatment. The results are color coordinated where green means that the treatment efficiency of olivine on this particular element was >95%, yellow means <95%, but still positive, and red means negative.

	Cd	Pb	Cr	Fe	Ni	Cu	Zn	As
1st	41.8	99.1	100	56.3	13.2	65.8	14.8	-2.61
2nd	18.2	98.5	99.4	30.6	7.74	19.8	7.28	-2.31

Results indicate that lime does not have the same capacity to remove metals as olivine, when comparing these results to table 3.2. However, for Pb and Cr the results are very promising. This may be of some importance, especially for Cr, since olivine released Cr. These results suggest that pre-treatment with lime before treating it with olivine may be an option. If pre-treatment with lime was performed, most of the Cr present before olivine treatment would be removed, meaning that if Cr is released from olivine it might not have such a negative impact on the quality of the water. This will depend on the amount of Cr released by treatment with olivine. The upper limit for class II of Cr in coastal water is 3.4 µg/L, so a concentration up to 3.4 µg/L would keep Cr within class II. This might not be an optimal solution, but it should be investigated. Another option would be to pass the water that was treated with olivine through lime after olivine treatment, to remove the released Cr. Future experiments should look into both the combination of lime and olivine as a mixture and as two consecutive treatment steps.

The resulting classification of condition after treatment with lime, shown in table 3.9, combined with the resulting treatment efficiencies in table 3.8, indicate that lime is not a treatment medium that will be adequate on its own.

Table 3.9 The concentrations, in µg/L, of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As before treatment with lime and of the first water to pass through the lime that did not have a white milky color (1st) and after 6 L had passed through the lime (2nd). The table is color coordinated according to the classification of condition for these elements in coastal water, where blue is class I, green is class II, yellow is class III, orange is class IV, red is class V, and gray is no classification.

	Cd [µg/L]	Pb [µg/L]	Cr [µg/L]	Fe [µg/L]	Ni [µg/L]	Cu [µg/L]	Zn [µg/L]	As [µg/L]
Before	28.4	22.9	3.08	10400	36.9	11700	8120	0.624
1st	16.5	0.209	-0.00131	4530	32.0	4000	6920	0.640
2nd	23.2	0.351	0.0180	7190	34.0	9390	7530	0.638

It can be observed from table 3.9 that not only does lime remove Cr efficiently, but the resulting concentrations are in class I; background level. This ability to remove Cr, as well as Pb, should be investigated over time if lime is to be considered as a treatment medium for this water.

A lime treatment experiment using the pipe connected to the pump was performed with continuous water flow over approximately 20 hours. Resulting concentrations from the lime experiment using the pump are presented in figure 3.15-3.17.

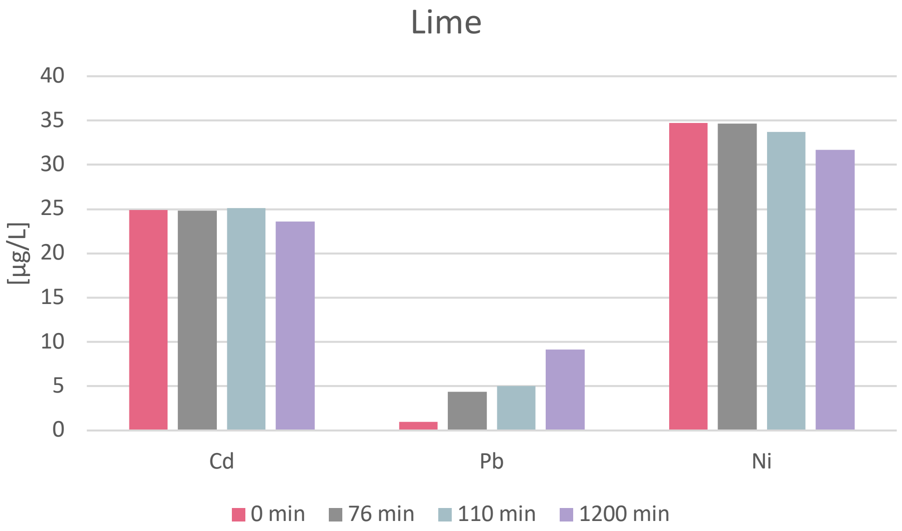

Figure 3.15 The concentration of Cd, Pb, and Ni after 0, 76, 110, and 1200 minutes of water continuously passing through lime.

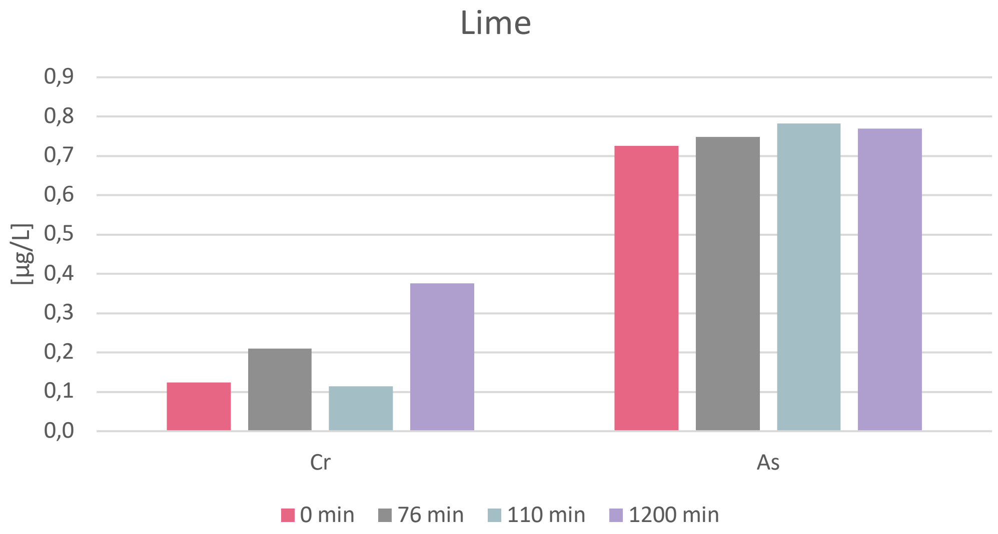

Figure 3.16 The concentration of Cr, and As after 0, 76, 110, and 1200 minutes of water continuously passing through lime.

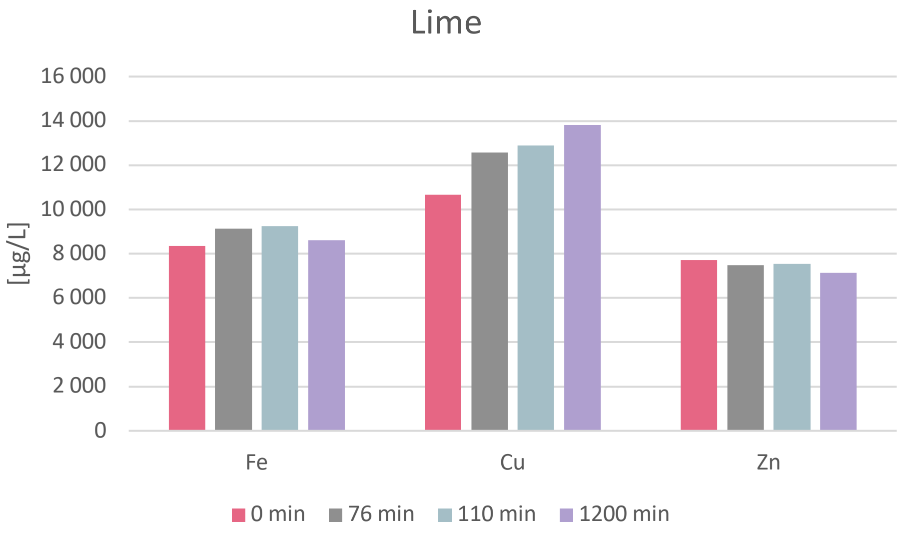

Figure 3.17 The concentration of Fe, Cu, and Zn after 0, 76, 110, and 1200 minutes of water continuously passing through lime.

The water being treated was taken continuously from the tunnel and passed through the lime using a pump. This facilitated the treatment of water over time, and samples were taken four times. The reason samples were taken at different times was to see if the treatment capacity of lime would change over time. Lime will raise the pH of the water, and this causes the precipitation of metals. This effect could wear off due to precipitation covering the lime, and it would therefore be of interest to see if the efficiency was the same after 20 hours as it was at the start of the experiment.

Looking at the trends of the concentrations in figure 3.15-3.17 it can be observed that lime's treatment efficiency seems to decrease over time for Pb, Cr, and Cu. There are some small variations for the other elements, but the overall treatment efficiency does not change much. The concentrations from the lime treatment experiment were compared to the concentrations before treatment, and the treatment efficiency was calculated in percent. The results are presented in % change in metal concentrations after treatment. Positive numbers indicate a decrease in concentration and negative numbers indicate an increase. All values are related to the concentrations in the sample taken before any treatment. The treatment efficiency of lime over time is presented in table 3.10.

Table 3.10 Treatment efficiency of lime for the elements Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As as % after 0, 76, 110, and 1200 minutes of water continuously passing through lime. The results are presented in % change in metal concentration after treatment, compared to before, and are based on concentration (µg/L). All values are related to the concentrations in the sample taken before any treatment. The results are color coordinated where green means that the treatment efficiency of olivine on this particular element was >95%, yellow means <95%, but still positive, and red means negative.

	Cd	Pb	Cr	Fe	Ni	Cu	Zn	As
After 0 min	12.1	95.8	96.0	19.5	5.82	8.87	4.95	-16.4
After 76 min	12.6	80.8	93.2	12.0	6.02	-7.35	7.72	-20.0
After 110 min	11.4	78.0	96.3	10.8	8.50	-10.1	7.01	-25.6
After 1200 min	16.8	60.0	87.8	16.9	14.2	-18.0	12.3	-23.4

The concentration of both As and Cu is increasing over time, compared to before treatment, in this experiment. As increased in the lime experiment in the open pipe as well (table 3.8). The increase in Cu was not expected. Cu is expected to precipitate at pH > 6, which can be seen in table 1.1. Lime is expected to raise the pH of the solution, and one possible explanation to the lack of Cu removal could be that the pH was not raised sufficiently. This would still not explain the increase. Looking at table 3.8 it can be observed that Cu was removed in the experiment with lime in the open pipe.

The removal of Pb and Cr was found to be 95% in the beginning and decreases slightly over time to 60% for Pb and 88% for Cr, indicating that the effect wears off over time. Further experimentation is needed to investigate if the possible trends in figure 3.15-3.17 and table 3.10 really are trends. In general, it appears that the treatment efficiency is neither increasing nor decreasing much over time, and the results indicate that using lime over time does not have the same capacity to remove metals from the water as olivine. The efficiency of lime over time is not directly comparable to the olivine experiment in the open pipe, and the olivine experiment over time failed.

To what degree the treatment with lime over time ameliorated the contaminated water is presented in table 3.11.

Table 3.11 The concentrations, in µg/L, of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As before treatment with lime and after 0, 76, 110, and 1200 minutes of water continuously passing through lime. The table is color coordinated according to the classification of condition for these elements in coastal water, where blue is class I, green is class II, yellow is class III, orange is class IV, red is class V, and gray is no classification.

	Cd [µg/L]	Pb [µg/L]	Cr [µg/L]	Fe [µg/L]	Ni [µg/L]	Cu [µg/L]	Zn [µg/L]	As [µg/L]
Before	28.4	22.9	3.08	10400	36.9	11700	8120	0.624
After 0 min	24.9	0.969	0.124	8340	34.7	10700	7720	0.726
After 76 min	24.8	4.39	0.210	9120	34.7	12600	7490	0.748
After 110 min	25.1	5.03	0.114	9240	33.7	12900	7550	0.783
After 1200 min	23.6	9.15	0.377	8610	31.7	13800	7120	0.769

After treatment with lime over time, only Cr and Pb are in class II. The other elements are in class III, IV, and V, and this indicates that treatment with lime alone is not enough. One of the biggest concerns is that Cu and Zn remain in extremely high levels. This is of concern because of their toxicity to aquatic life. Cu is regarded to have extensive toxic effects, i.e. class V, at concentrations above 5.2 µg/L, and Zn at concentrations above 60 µg/L. After being treated with lime the Cu concentration in the water increased. After 20 hours the Cu concentration was 2654 times higher than the limit for class V. Zn was 119 times higher than its limit for class V. Cd also remains in class V after treatment and is almost twice as high as the lower limit for class V after 20 hours. It becomes clear that lime is not sufficient as a treatment medium on its own to treat this water.

3.1.8 pH

The ability to increase the pH of a solution is a property olivine and lime have in common. To assess the effect of olivine and lime on pH, the aforementioned experiments with olivine and lime in the open pipes using gravity were recreated. Samples were taken before treatment and after 6 L and 12 L of water had passed through the treatment medium. The results from the recreated olivine experiment in combination with the corresponding pH of each sample is presented in figure 3.18-3.20.

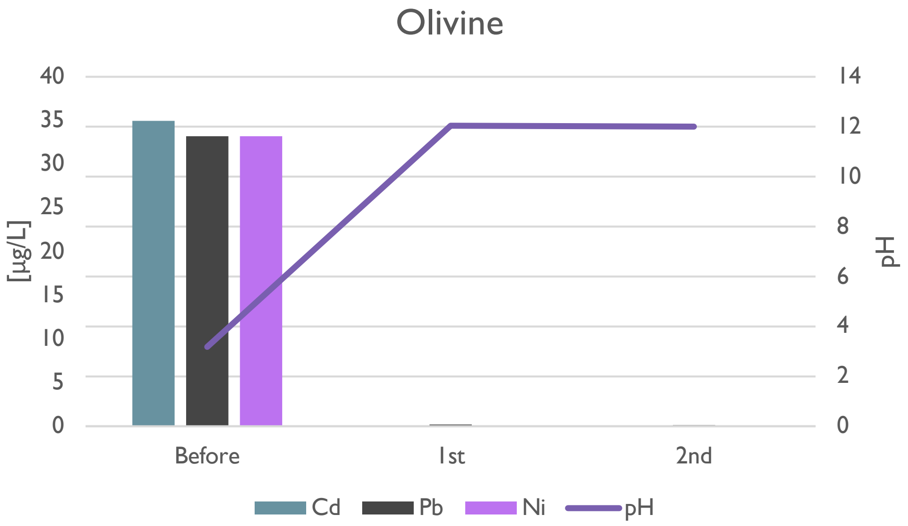

Figure 3.18 Concentrations of Cd, Pb, and Ni, and the corresponding pH, before and after treatment with olivine. Two samples were taken during treatment; one after 6 L had passed through the olivine (1st) and one after 12 L had passed through (2nd). The left y axis is concentration [µg/L] and corresponds to the bars, and the right y axis is pH and corresponds to the line.

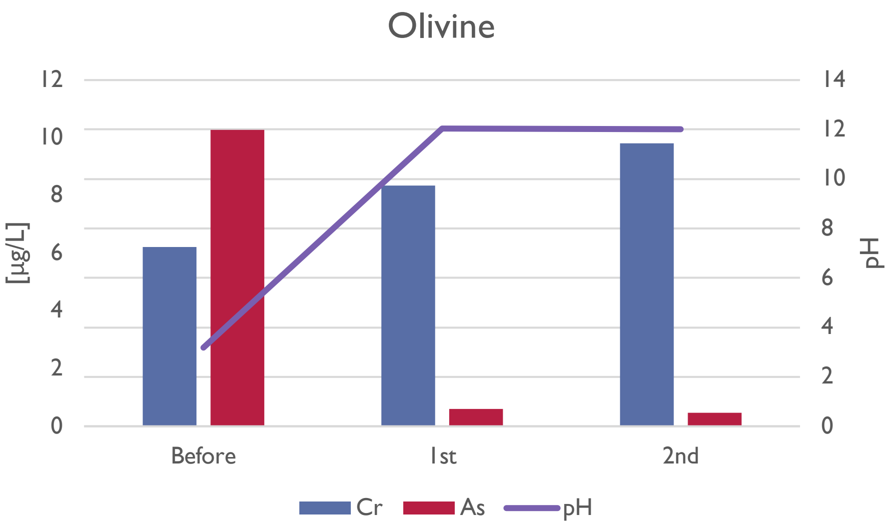

Figure 3.19 Concentrations of Cr and As, and the corresponding pH, before and after treatment with olivine. Two samples were taken during treatment; one after 6 L had passed through the olivine (1st) and one after 12 L had passed through (2nd). The left y axis is concentration [µg/L] and corresponds to the bars, and the right y axis is pH and corresponds to the line.

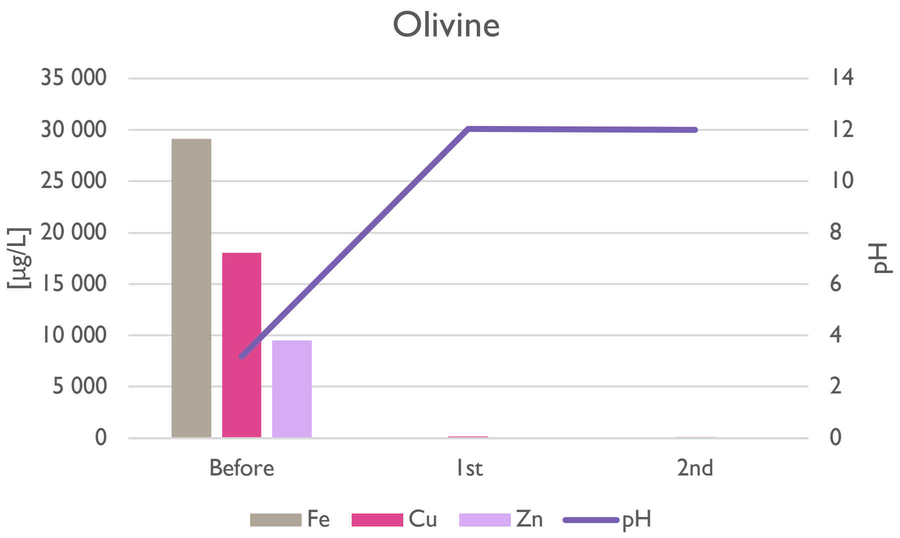

Figure 3.20 Concentrations of Fe, Cu, and Zn, and the corresponding pH, before and after treatment with olivine. Two samples were taken during treatment; one after 6 L had passed through the olivine (1st) and one after 12 L had passed through (2nd). The left y axis is concentration [µg/L] and corresponds to the bars, and the right y axis is pH and corresponds to the line.

By presenting the concentrations before and after treatment in the same diagrams it becomes apparent which elements are being removed in a high degree, and which elements remain or increase in concentration. In figure 3.18 the results for the concentrations of Cd, Pb, and Ni show that the treatment efficiency of olivine on these elements is high, and figure 3.20 shows the same for Fe, Cu, and Zn. Figure 3.19 clearly illustrates that olivine causes an increase in Cr concentration. It also illustrates that As is removed by the treatment. This is in accordance with the results from the first olivine experiment (figure 3.1-3.3). Cr(III) will precipitate as hydroxide when the pH increases, but if the pH increases above 12, the occurrence of Cr(III) hydroxide is negligible, and it will likely be complexed by four hydroxides, a soluble complex.[79]

The purple line in the graphs show the pH of the water at the three points. The pH increases dramatically from the untreated water to the first sample of the treated water, and remains the same during treatment. This increase could be overestimated. It was found that the pH was 3.18 before treatment and 12.0 after treatment with olivine. The pH meter that was used for this experiment was calibrated right before it was used, but the calibration buffers had pH 4 and pH 7. This will result in a good calibration curve in this pH range, but measuring pH outside of this range will have uncertainties. It is therefore not possible to say what the exact pH was at the 1st and 2nd sample of the treated water. It is certain that the pH is higher than 7, and probable that it is considerably higher. The uncertainty of the calibration for samples with a pH above 7 might have caused an overestimation of the pH. A previous study found that olivine raised the pH of a sample from 5.8 to 8.5.[77] This was done with fine olivine powder and not granules, so it is not directly comparable, but it shows that olivine has the ability to raise pH higher than 7. A technical datasheet on olivine products states that the olivine granulate Blueguard G1-3 typically raises the pH to 11-12.[76] This is in agreement with the findings in the experiment.

From this experiment it becomes clear that olivine does have the ability to raise the pH, but from the large scale testing in Trondheim municipality's system, it was observed that the effect does not necessarily last. The adsorption on the olivine would likely have lasted longer if the pH of the initial solution had been raised by means other than olivine, before passing the water through the olivine. This would be an interesting experiment to do in the future.

The results from the recreated lime experiment in combination with the corresponding pH of each sample are presented in figure 3.21-3.23.

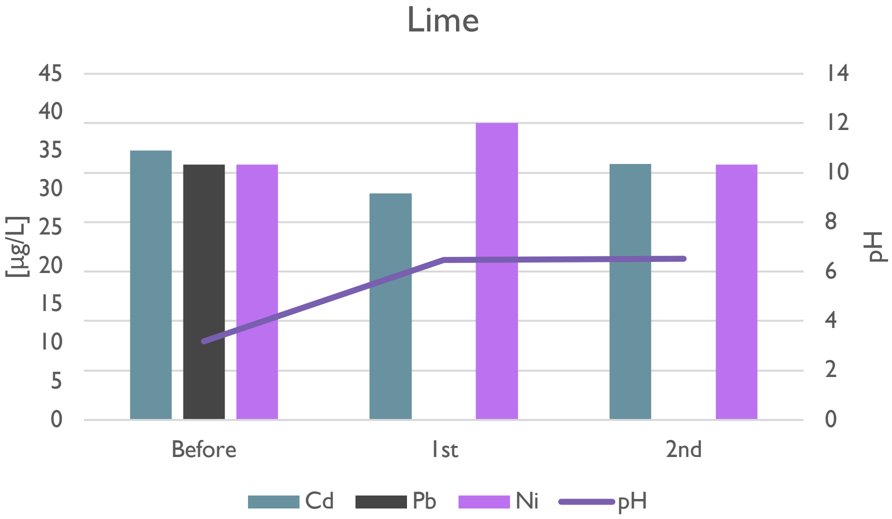

Figure 3.21 Concentrations of Cd, Pb, and Ni, and the corresponding pH, before and after treatment with lime. Two samples were taken during treatment; one after 6 L had passed through the lime (1st) and one after 12 L had passed through (2nd). The left y axis is concentration [µg/L] and corresponds to the bars, and the right y axis is pH and corresponds to the line.

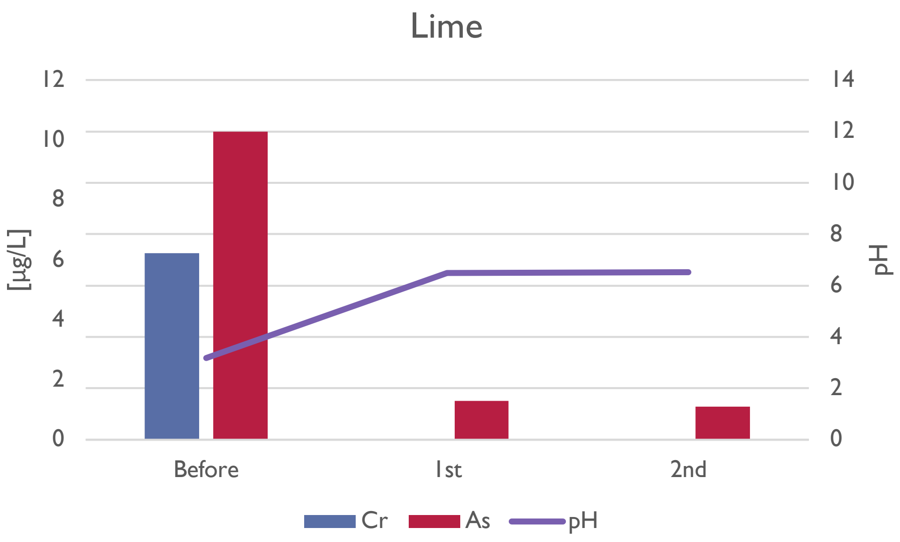

Figure 3.22 Concentrations of Cd and As, and the corresponding pH, before and after treatment with lime. Two samples were taken during treatment; one after 6 L had passed through the lime (1st) and one after 12 L had passed through (2nd). The left y axis is concentration [µg/L] and corresponds to the bars, and the right y axis is pH and corresponds to the line.

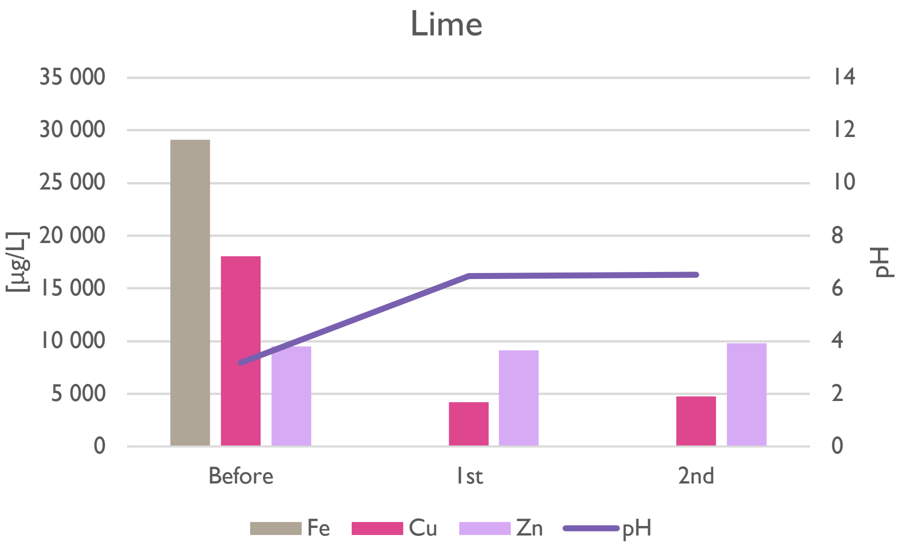

Figure 3.23 Concentrations of Fe, Cu, and Zn, and the corresponding pH, before and after treatment with lime. Two samples were taken during treatment; one after 6 L had passed through the lime (1st) and one after 12 L had passed through (2nd). The left y axis is concentration [µg/L] and corresponds to the bars, and the right y axis is pH and corresponds to the line.

Figures 3.21-3.23 illustrate lime's ability to remove metals from the contaminated water, as well as the corresponding pH of each sample. By keeping the before sample concentrations in the same graph as the treated samples, it becomes clear from figure 3.21 that lime efficiently removes Pb, but not Cd or Ni. A similar effect can be seen in figure 3.22, where it can be observed that lime's ability to remove Cr is better than its ability to remove As, though both decrease in concentration. Finally it can be seen in figure 3.23 that Fe is removed with a much higher efficiency than Cu and Zn, and that Zn is virtually not affected by the lime treatment. Treatment with lime raised the pH of the water from 3.18 to 6.52.

The treatment efficiency of both olivine and the lime from these experiments are presented in table 3.12. The results are presented in % change in metal concentrations after treatment. Positive numbers indicate a decrease in concentration and negative numbers indicate an increase. All values are related to the concentrations in the sample taken before any treatment.

Table 3.12 Treatment efficiency of olivine and lime for the elements Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As as % after 6 L (1st) and 12 L (2nd) of water had passed through the treatment medium. The results are presented in % change in metal concentration after treatment, compared to before, and are based on concentration (µg/L). All values are related to the concentrations in the sample taken before any treatment. The results are color coordinated where green means that the treatment efficiency of olivine on this particular element was >95%, yellow means <95%, but still positive, and red means negative.

	Cd	Pb	Cr	Fe	Ni	Cu	Zn	As
Olivine 1st	100	99.5	-34.3	99.9	99.7	99.1	99.3	94.3
Olivine 2nd	99.9	99.7	-57.7	99.9	99.8	99.4	99.5	95.6
Lime 1st	15.9	99.8	99.6	99.8	-16.3	76.8	3.81	87.5
Lime 2nd	4.79	99.8	99.6	99.9	0.0444	73.8	-3.30	89.2

Olivine displays high efficiency removing most of the elements, with the exception of Cr. Lime does not produce such efficient results. Treatment with lime is efficient for Pb, Cr, and Fe.

The concentrations of all of the elements in all of the samples from these experiments are presented in table 3.13, with corresponding classification of conditions.

Table 3.13 The concentrations, in µg/L, of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As before treatment with olivine and lime, and after 6 L (1st) and 12 L (2nd) had passed through the treatment medium. The table is color coordinated according to the classification of condition for these elements in coastal water, where blue is class I, green is class II, yellow is class III, orange is class IV, red is class V, and gray is no classification.

	Cd [µg/L]	Pb [µg/L]	Cr [µg/L]	Fe [µg/L]	Ni [µg/L]	Cu [µg/L]	Zn [µg/L]	As [µg/L]
Before	35.0	33.2	6.22	29100	33.2	18100	9500	10.3
Olivine 1st	0.0153	0.177	8.35	20.6	0.0834	160	68.6	0.59
Olivine 2nd	0.0224	0.110	9.80	23.3	0.0631	102	42.8	0.457
Lime 1st	29.4	0.0765	0.0253	49.8	38.6	4190	9130	1.29
Lime 2nd	33.3	0.0768	0.0222	17.3	33.2	4740	9810	1.11

Treatment with lime decreases the Cr concentration to background levels. Lime treatment also results in lower Pb concentration than olivine treatment, but the resulting Pb concentrations from both experiments are in class II. Apart from these two elements, olivine has a greater ability to remove the metal contaminants. The treatment with olivine led to Cd and Ni concentrations in background levels and Pb and As in class II. Cr was released, but concentrations still remained in class III. The treatment efficiency of olivine on Cu and Zn was over 99%, as seen in table 3.12. Despite the high treatment efficiency Cu still remained in class V. Zn was found to be in class V half way through the olivine experiment (olivine 1st), but was found to be in class IV at the end of the experiment (olivine 2nd). This is the second time Zn concentrations decreased to below class V, the other time being the first experiment with olivine. This suggests that of the investigated treatment methods, olivine is the only one with potential to remove enough Zn contamination from this water.

The precipitation of Fe and other elements is expected when the pH is increased. Table 1.1 shows the pH range for precipitation, as well as the optimal pH, of these elements as hydroxides. Olivine has properties that affect this water by both raising the pH and by adsorbing elements on its surface. It is therefore difficult to say if the treatment efficiency is due to the increase in pH or adsorption. Lime does not have this adsorbing quality. The treatment efficiency of the lime experiment is therefore due to the increase in pH. The pH of the water before treatment was 3.18, and after treatment with lime it increased to 6.52. This increase in pH is well within the precipitation range of Fe(III) and Cr(III). This was also reflected in the resulting decrease in concentration of these metals. The precipitation range of Cu and Pb, which is > pH 6, has also been exceeded. This seems to have caused the precipitation of both Pb and Cu, but Cu at a lesser degree. This may be due to the competition of OH^- between elements. Another possible explanation is that Pb may be removed due to both precipitation and adsorption onto other precipitates, such as Fe hydroxides. The precipitation range for Ni and Cd is > pH 6.7. This has not been reached in the lime experiment, and this is reflected in the results, where neither of these were efficiently removed.

Fe was removed by 99.9% in this lime experiment, but only by 30.6-56.3% in the first lime experiment (table 3.8) that this one was made to recreate. In the experiment with lime over time, Fe was only removed with 10.8-19.5% (table 3.10). Since the pH increased into the precipitation range of Fe, precipitation and therefore removal of Fe was expected. Because the pH was not measured in the two previous experiments, it is not known if it was raised to the same level as in the experiment where pH was measured, or even raised high enough to expect precipitation of Fe. Since Cr and Pb have been removed efficiently in the two previous lime experiments, and they were also removed efficiently in this one, it is likely that the pH has been increased to a similar level in the previous experiments. One possible reason for the higher Fe removal could be that the Fe in the first two experiments existed mainly as Fe(II), but is mainly Fe(III) in the experiment where pH was measured. Fe(II) does not readily precipitate at pH 6.52, while Fe(III) precipitates at pH below 6.52. Another explanation could be the differences in the condition of the water before treatment. Since these experiments were performed on different dates, and Trondheim municipality made changes in their system between the dates, the condition of the untreated tunnel water was likely different on the different experiment dates.

Precipitation of the metals could also be performed in the tunnel directly. When the metals precipitate, they are removed from the water column, and stay in the tunnel. However, as a part of the sediments in the tunnel they would still represent a potential source of pollution, due to leaching. In a short term perspective precipitation will directly decrease the metal concentrations in the water and therefore also their release into the surrounding environment. However, due to the possibility of leaching, the metals will likely over time enter the water column and consequently the surrounding environment. The precipitation itself may also cause some issues, particularly the removal of the sludge that is left behind. It will be concentrated with metals and need to be dealt with in a safe and clean way. In addition the texture of the sludge may cause issues because it is so lightweight that it is easily stirred up when any type of turbulence occurs.

3.1.9 Turbidity

The turbidity was measured both on the tunnel water from inside the tunnel and on the water at the water's edge. It was measured over a period of 10 minutes. The results from the turbidity measurements are presented in table 3.14.

Table 3.14 The turbidity after 0, 10, 20, 30, 40, and 50 seconds, and after 1, 2, 5, and 10 minutes of two samples of the tunnel water; one taken from Trondheim Municipality's system and one from the water's edge.

Time	Tunnel water [NTU]	Water's edge [NTU]
0 seconds	15.90	12.81
10 seconds	16.22	12.69
20 seconds	15.79	13.15
30 seconds	15.89	12.91
40 seconds	15.88	13.10
50 seconds	15.91	12.84
1 minute	15.88	13.24
2 minutes	15.89	13.14
5 minutes	16.07	12.96
10 minutes	16.04	13.15
Average	15.95	13.00

The results indicate that there are low levels of particulate matter both in the tunnel water from inside the tunnel and the water from the water's edge. It was later discovered that on this day the water from inside the tunnel was most likely taken from the top layer of the water. The sample of the water from the water's edge was also taken from the top layer of water. These results then indicate that there is sedimentation inside the tunnel. The water at the water's edge was in a position where it would not have been disturbed in a while, and therefore the turbidity is low. Inside the tunnel, the water is most likely being taken close to the threshold, i.e. from the top layer. Water will flow towards the threshold and over the threshold when the level is high enough. This flow towards the threshold may explain the higher turbidity, compared to the water's edge, because it might have caused a slight turbulence of the sediments. Even though these results indicate that the particles sediment inside the tunnel, the texture of the sedimented particles is light and fluffy, and a small turbulence stirs up the particles. The sedimentation is not stable enough to be taken advantage of as part of a treatment system.

Table 3.15 A summary of the results from the experiments. The average treatment efficiencies are presented for each element in each experiment in percent. The results are color coordinated where green means that the treatment efficiency of olivine on this particular element was ≥95%, yellow means <95%, but still positive, and red means negative.

Experiment	Average treatment efficiency [%]							
	Cd	Pb	Cr	Fe	Ni	Cu	Zn	As
Olivine	99.9	99.0	-119	99.4	99.8	98.1	98.8	70.0
Olivine (pH experiment)	100	99.6	-46.0	99.9	99.8	99.3	99.4	95.0
Olivine + activated carbon	99.9	98.0	-1490	99.8	99.5	99.4	99.0	-459
Lime	30.0	98.8	99.7	43.5	10.47	42.8	11.0	-2.46
Lime (pH experiment)	10.3	99.8	99.6	99.9	-8.1	75.3	0.255	88.4
Lime over 20 hours	13.2	78.7	93.3	14.8	8.64	-6.65	8.00	-21.4
Aeration	0.81	1.36	-3.16	1.41	1.91	0.976	0.143	-15.9

3.2 Samples from inside the tunnel

Between October 2018 and February 2020 samples were taken at different points inside the tunnel. The sampling points were chosen to get an overview of changes happening at different points inside the tunnel, both in the water in the tunnel itself, and after being treated in Trondheim municipality's system. Samples taken of the water treated with olivine in Trondheim Municipality's system have already been presented in section 3.1.4. Samples were also taken of the water coming into the tunnel through the tunnel walls.

3.2.1 Tunnel water

To investigate how the water inside the tunnel changed over time, samples were taken of the untreated tunnel water with varying regularity. The samples were taken from Trondheim municipality's system. This system evolved over time, and changes were made, but for each sampling date there was a sampling point which represented the water inside the tunnel. This point was not necessarily the same one each time. Samples were taken either from an open tub where untreated water passed through, or a hose taking water in from the tunnel, and once from the tunnel directly. All of the samples represented untreated tunnel water, but they may have been taken from different depths. This may have affected the results, because some precipitation and sedimentation occurs, and samples of the top water will most likely be less contaminated than those deeper in the water column. See appendix A for more information about the changes made in Trondheim Municipality's system.

The concentration of each element in each sample of tunnel water is presented in figures 3.24-3.31. The graphs include the classifications of condition for each element in coastal water. They are color coordinated, and a colored line represents the lowest limit for its class.

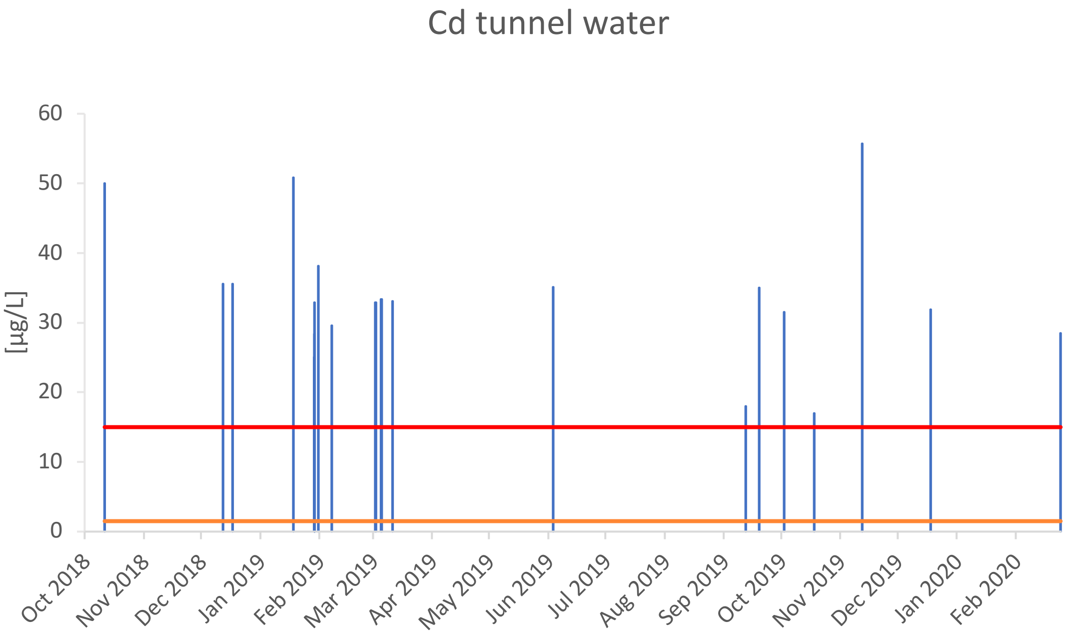

Figure 3.24 The concentration of Cd in water inside the tunnel in the sampling period 11.10.18-24.02.20. Colored lines define the lower limit for a class in the classification of condition for coastal water, where orange is class IV and red is class V.

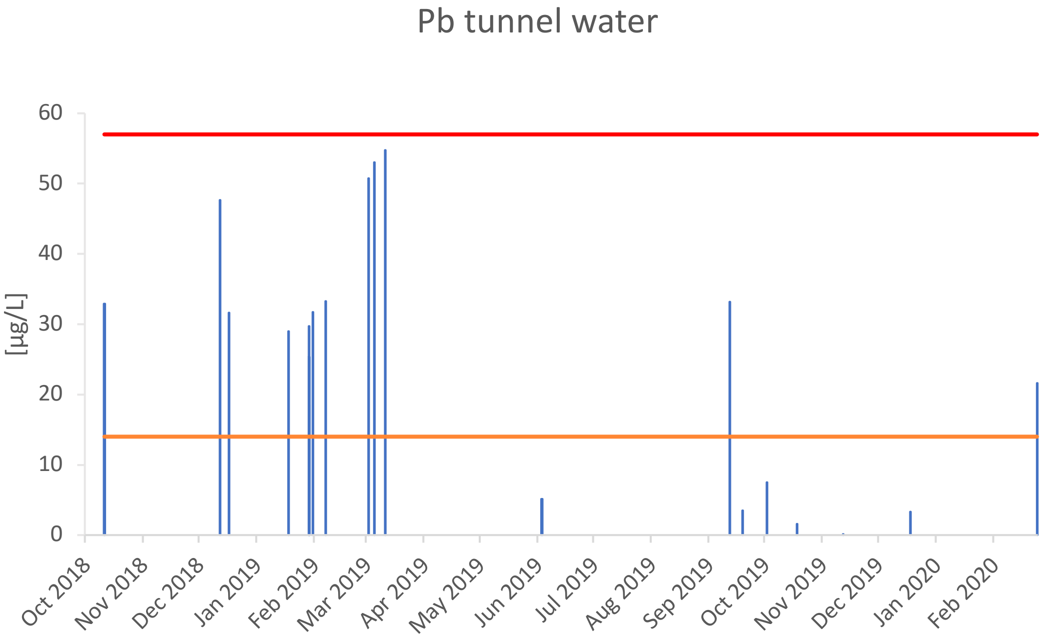

Figure 3.25 The concentration of Pb in water inside the tunnel in the sampling period 11.10.18-24.02.20. Colored lines define the lower limit for a class in the classification of condition for coastal water, where orange is class IV, and red is class V.

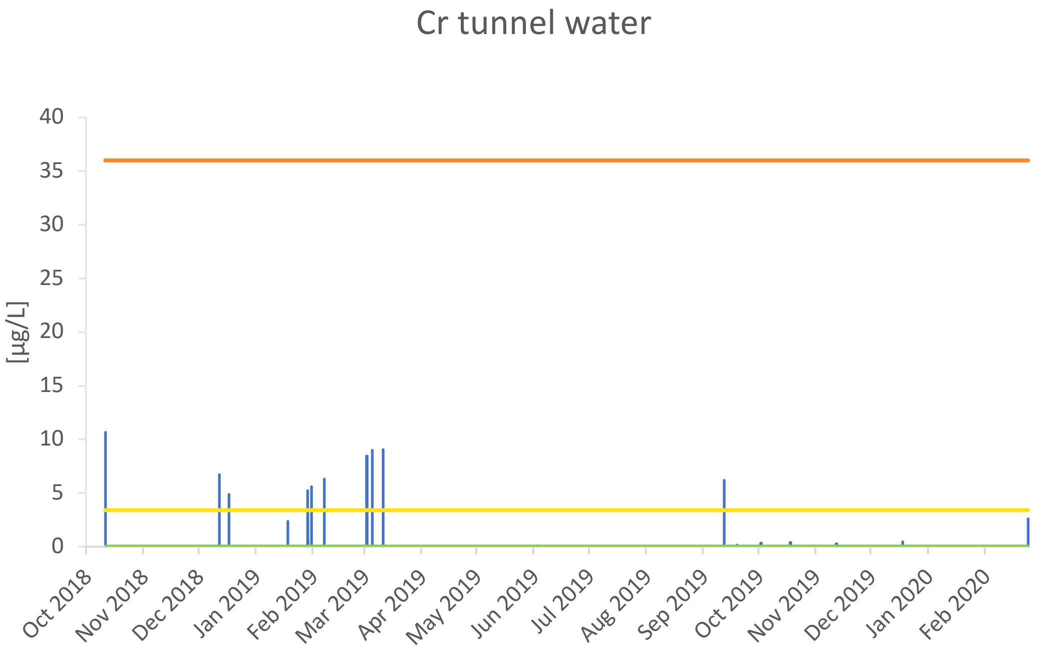

Figure 3.26 The concentration of Cr in water inside the tunnel in the sampling period 11.10.18-24.02.20. Colored lines define the lower limit for a class in the classification of condition for coastal water, where green is class II, yellow is class III, and orange is class IV.

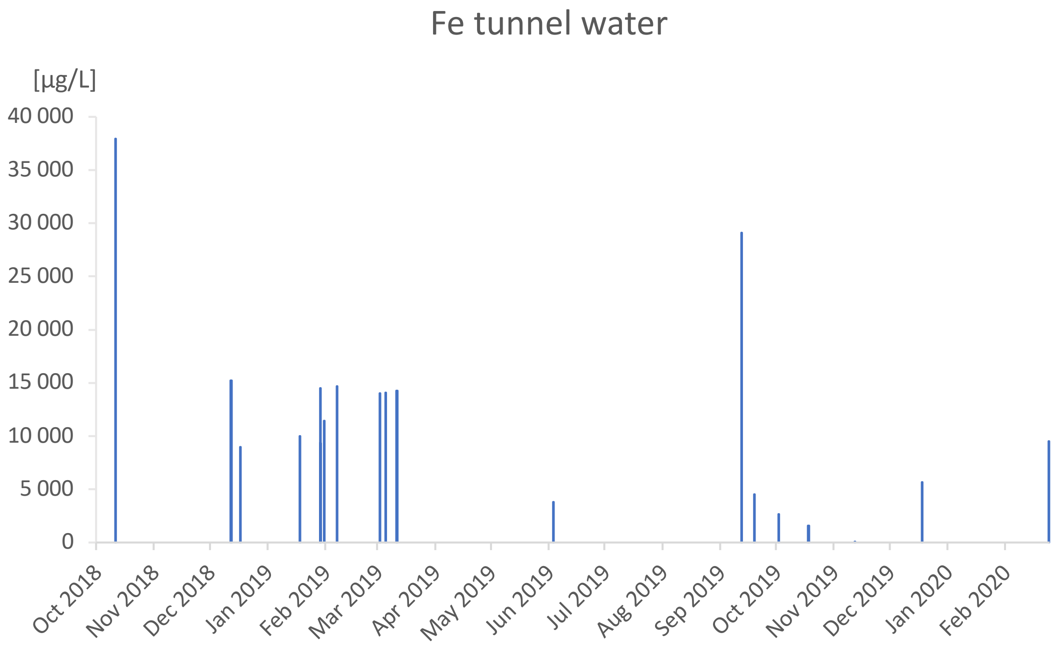

Figure 3.27 The concentration of Fe in water inside the tunnel in the sampling period 11.10.18-24.02.20.

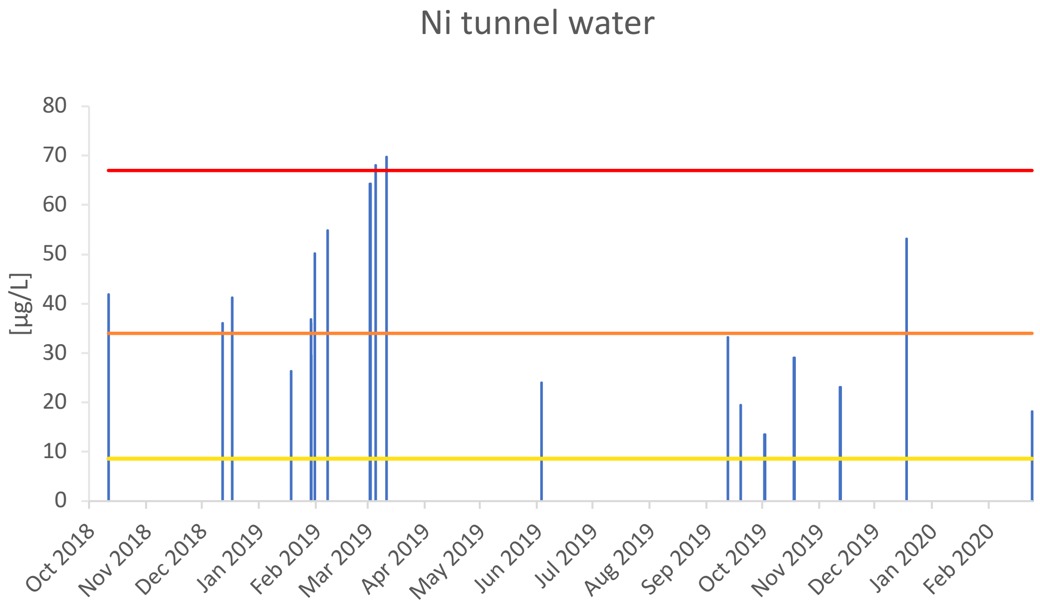

Figure 3.28 The concentration of Ni in water inside the tunnel in the sampling period 11.10.18-24.02.20. Colored lines define the lower limit for a class in the classification of condition for coastal water, where yellow is class III, orange is class IV, and red is class V.

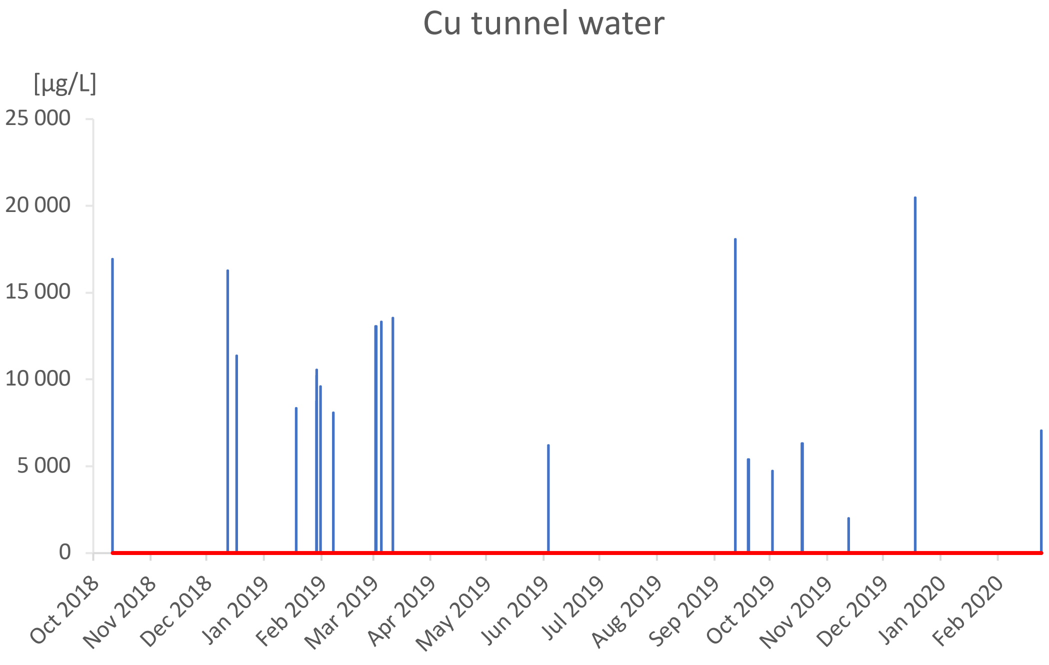

Figure 3.29 The concentration of Cu in water inside the tunnel in the sampling period 11.10.18-24.02.20. The red line defines the lower limit for class V in the classification of condition for coastal water.

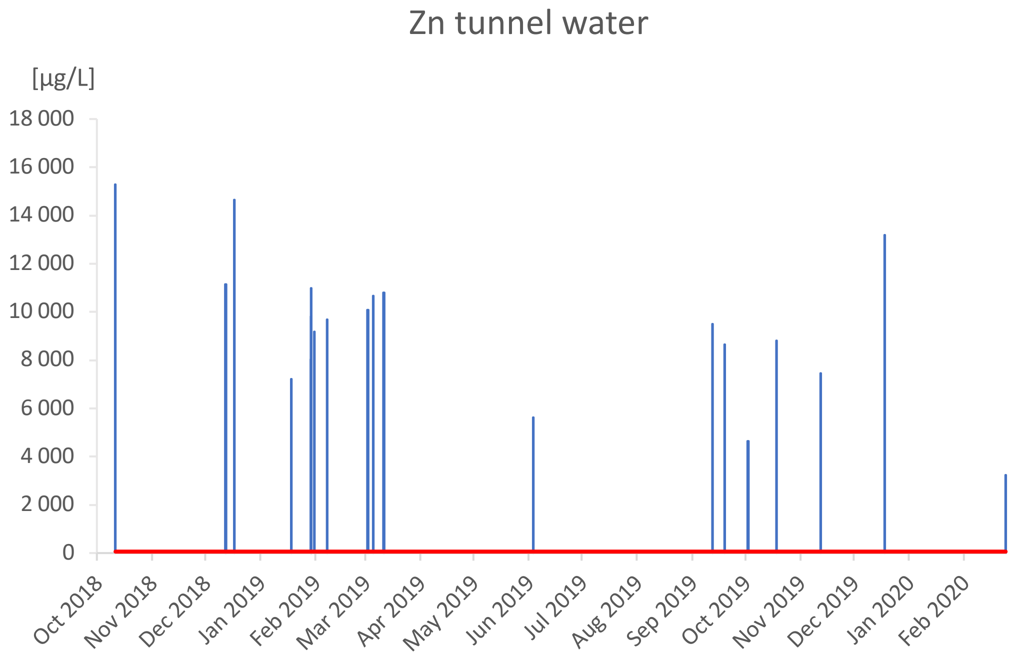

Figure 3.30 The concentration of Zn in water inside the tunnel in the sampling period 11.10.18-24.02.20. The red line defines the lower limit for class V in the classification of condition for coastal water.

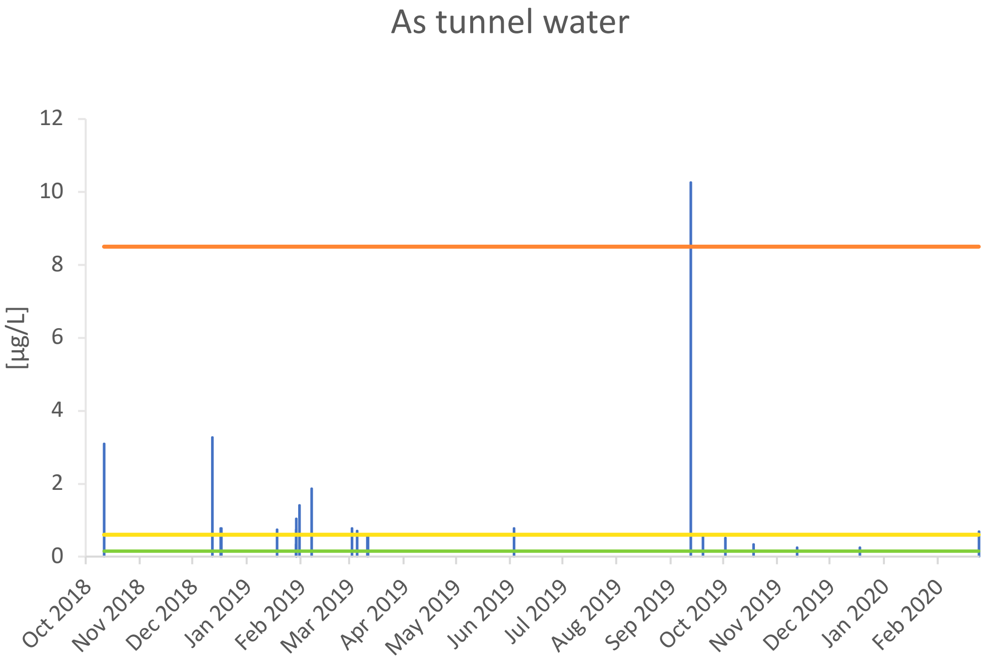

Figure 3.31 The concentration of As in water inside the tunnel in the sampling period 11.10.18-24.02.20. Colored lines define the lower limit for a class in the classification of condition for coastal water, where green is class II, yellow is class III, and orange is class IV.

Figures 3.24-3.31 all show that varying concentrations were found on the different sampling dates. Despite the fluctuations, some observations stay constant. Cd, Cu, and Zn have been found with concentrations in class V throughout the sampling period, which can be seen in figure 3.24, 3.29, and 3.30. Both Cr and As stay within classes I-III, with one exception where

As was found in class IV. The reason for the high As concentration in this particular sample could be contamination. However because Fe, Pb, and Cu also follow this trend of a clear peak at this particular sampling date, the reason is probably not contamination. As will co-precipitate with and adsorb to Fe, but this removal is not stable. A disturbance in the precipitation of Fe will lead to the desorption and release of As. Since Fe is found in high concentrations on the same date as As has a high peak, it is likely that some of the Fe precipitates have dissolved.

The fluctuations in concentration of all the elements indicate the instability of the system. The samples are taken with varying regularity, so the fluctuations may not be as dramatic as they look. Nevertheless, the variation between highest and lowest concentration is large. Precipitation occurs inside the tunnel, but the texture is light and fluffy, and turbulence easily stirs up the precipitation. When disturbed, the adsorbed metals on the precipitates may be released. The instability of the precipitation emphasizes the need for a treatment system. Leaving the water alone and letting the precipitates sediment will not be secure enough. Removing the sedimented precipitates would also pose problems due to the texture. Not all the metals precipitate or adsorb to precipitates. The need for treatment steps is evident.

3.2.2 Water entering the tunnel

To investigate the level of contamination of the water entering the tunnel through the tunnel walls, samples were collected along the wall at different locations inside the tunnel. In addition samples were taken on days where water was found seeping through the tunnel wall close to the entrance. Samples inside the tunnel were taken on two different dates at four locations. An additional sample was taken the first sampling date, but there was no water found at this location on the second sampling date. Samples of water coming through the tunnel wall close to the entrance were taken on the three days water was discovered there. All of these sampling points are shown in figure 2.3, and pictures of the different sampling points can be found in appendix A. The concentrations of each element in each sample are presented in table 3.16.

Table 3.16 The concentrations, in µg/L, of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As at different sampling dates and points along the tunnel wall. The table is color coordinated according to the classification of condition for these elements in coastal water, where blue is class I, green is class II, yellow is class III, orange is class IV, red is class V, and gray is no classification.

		Cd [µg/L]	Pb [µg/L]	Cr [µg/L]	Fe [µg/L]	Ni [µg/L]	Cu [µg/L]	Zn [µg/L]	As [µg/L]
Tunnel wall	03 Jun. 2019	7.80	2.58	31.3	374000	44.1	23800	2080	25.2
Tunnel wall	19 Sep. 2019	8.82	3.56	27.0	297000	39.0	19600	2090	26.4
Tunnel wall	02 Oct. 2019	14.1	2.34	21.2	192000	33.9	14400	3510	12.9
Left tunnel wall	19 Sep. 2019	8.67	0.235	0.963	3880	5.89	1730	2330	0.206
Colorful wall	06 Jun. 2019	0.521	2.17	0.246	202	2.59	122	132	0.621
Colorful wall	18 Dec. 2019	0.193	1.24	0.753	90.1	1.78	96.6	40.8	0.434
First collection	06 Jun. 2019	0.759	0.0242	0.445	15.7	6.55	44.5	215	0.247
First collection	18 Dec. 2019	0.433	0.151	0.844	97.0	4.85	78.3	113	0.245
Second collection	06 Jun. 2019	1.54	1.47	0.400	309	3.24	102	558	0.298
Second collection	18 Dec. 2019	1.76	0.869	0.596	456	4.30	86.7	521	0.364
Dripping from ceiling	06 Jun. 2019	0.657	0.029	0.223	13.8	1.97	9.71	284	0.413
Threshold	06 Jun. 2019	2.17	0.451	0.214	105	2.35	43.8	671	0.157
Threshold	18 Dec. 2019	18.4	0.113	0.210	109	13.6	1230	4600	0.159

There are several differences between the sampling points, dates, and elements within the samples. It is known that residue from the previous mining activity was covered in membranes and is buried beneath the park that is at Killingdal today. Looking at the level of some of the contaminants, it is evident that either the membranes have not been successful, or not all of the residue has been covered. Cu, Zn, Cd, and Fe are entering the tunnel in high concentrations, and this reflects the high levels of these contaminants in the tunnel water, as shown in figure 3.24, 3.27, 3.29, and 3.30. Pb was found in higher levels inside the tunnel than in the water coming through the wall, suggesting that it may exist as less soluble species when entering the tunnel than in the tunnel water.

In general, the concentrations of the elements are higher in the tunnel water than in the water entering the tunnel. Elements inside the tunnel will accumulate, and higher levels are expected. The sampling point “tunnel wall” presented in table 3.16 is an exception. It has higher levels of concentrations of Cr, Fe, Cu, and As than the tunnel water inside the tunnel. This point is close to the entrance and is less covered by ground above. The other samples are further underground, under both old tunnel structures that have been closed off by cement and membrane covered residue. The ground above “tunnel wall” looks like it was not processed during the cleaning in 2010/2011. This suggests that there may be some mining residue left untreated in the ground above this point. Another possible explanation for the higher concentrations could be that the water seeping through the tunnel wall near the entrance was only observed three times from October 2018 to February 2020, i.e. not very often. This was on, or right after, very rainy days. If the water seeping through the tunnel walls inside the tunnel is more of a continuous flow, then it may have washed out more of the metal contaminants it passes through.

Cu and Zn are found in high concentrations in all of the samples. These elements are also found in high concentrations in the water in the tunnel. Sphalerite and chalcopyrite provide galvanic protection for pyrite due to lower rest potential than pyrite. This causes them to corrode first. This will lead to a high degree of oxidation of sphalerite and chalcopyrite, causing high levels of Zn and Cu in the AMD.

The water at the sampling points “first collection” and “second collection” was at the time of sampling being collected and led past the threshold, being directly released into the fjord. This water is coming through the areas that are thought to be cleaned, and it is therefore assumed that the water is clean. Looking at the concentrations in samples from these sampling points (table 3.16), the levels of Cu and Zn are in class V, and this is considered too high to be released into the fjord. The concentration of Cd at “second collection” is in class IV, which is also too high. This water should therefore be released into the tunnel so that it will go through Trondheim Municipality’s treatment system before reaching the fjord.

3.3 Quality assurance

Due to budget at time restraints, only one sample has been taken at each sampling point and date. Several steps have been taken to assure quality in this thesis. Standard sampling methods have been used and gloves have been worn during sampling to avoid contamination. Standard sampling method includes sample being drawn into the syringe and expelled three times before taking the actual sample, and in addition the sampling syringe was rinsed before and after use, and was switched out regularly. When the same syringe was used for several samples, the (assumed) least concentrated sample was taken first.

During the ICP-MS analysis repeating tests were performed regularly to check for instrument reproducibility. The ICP-MS reproducibility and accuracy is regularly checked using certified reference material. Blanks were made for each batch of acid used for preservation. These blanks are method blanks and will correct for any influence of the method on the results.

4 Conclusion

Monitoring of the water inside the tunnel at Killingdal showed that the water is heavily contaminated, especially in regard to Cd, Cu, and Zn, which were in class V in all of the samples taken from October 2018 to February 2020. This indicates that the water inside the tunnel needs to be treated to avoid it negatively impacting the aquatic ecosystem near its outlet.

Water coming into the tunnel through the tunnel walls was found to be less contaminated than the water inside the tunnel. Cu and Zn concentrations were in class V in all samples except for one sample where Zn was in class IV. The sampling point with the highest concentration was the tunnel wall near the entrance of the tunnel. This indicates that the clean-up of the mining residue in 2010/2011 was not sufficient, especially in the area close to the entrance of the tunnel. The high levels of Cu and Zn, and to some degree Cd, in the water coming into the tunnel indicate that contaminants are being transported by water from the mining residue and into the tunnel. Some of this water was at the time of sampling being collected and led out of the tunnel, but should be let into the tunnel so that it will be treated by the treatment system that will treat the tunnel water.

Two experiments were performed with olivine as the only treatment medium. Olivine removed more than 99% of Cd and Ni in both experiments. Pb, Fe, and Zn were removed with at least 98.0% efficiency in the first experiment and at least 99.5% in the second experiment, while the treatment efficiency of Cu was 96.9%-99.0% in the first experiment and 99.1%-99.4% in the second experiment. Up to 95.6% of As was removed in the second experiment, but only up to 78.3% was removed in the first experiment. In both experiments the Cr concentration increased; up to 211% in the first experiment and up to 57.7% in the second. It was discovered that olivine raised the pH of the solution from 3.18 to 12.00. Olivine over time was not successful. The experiment with olivine over time failed due to precipitation clogging the system, and when olivine was tested in Trondheim municipality's system, its treatment effect quickly wore off. Because of olivine's high treatment efficiencies in the other experiments it should be investigated further over time to see if it can be a potential treatment medium in a system treating the water at Killingdal. The unfortunate Cr release must be monitored, and olivine in combination with other treatment steps should be considered.

Olivine combined with activated carbon resulted in lower treatment efficiencies than olivine alone, and the mixture should therefore not be considered as a treatment medium. Aeration was tested as a possible pre-treatment step, but did not yield any positive results regarding metal removal through precipitation.

Treatment with lime was tested in three different experiments, including one testing lime over time. It was found in all of the three experiments that in a short-term perspective lime removed Pb and Cr with at least 95.8% efficiency. Over time it was found that this effect diminished, and the treatment effects of Pb and Cr were found to be 60.0% and 87.8% after 20 hours. Lime caused the pH to increase from 3.18 to 6.52, which is less than olivine, but still high enough to cause the precipitation of several metals. The treatment efficiencies of lime, especially for Cr, should be investigated further in combination with olivine, either as a mixture of lime and olivine, or using lime as a treatment step before or after olivine.

All of the experiments ended with Cu and Zn in class V with a few exceptions with Zn concentrations in class IV. Future studies should focus on the removal of Cu and Zn from the water. Olivine has high treatment efficiencies for these two elements and can be considered as a treatment medium, but needs further investigation to find a solution that results in acceptable Cu and Zn concentrations that can be released into the fjord. Olivine in combination with other steps such as pre-treatment with aeration or lime, or polishing steps, should be explored. The treatment system and its scale needs further investigation to find a way to optimize olivine and lime’s treatment efficiencies so that the resulting concentrations of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As are all in acceptable levels that can be released into the fjord.

5 Future suggestions

These experiments show the potential of olivine and lime as treatment mediums for the contaminated water at Killingdal. Olivine shows promising results, but the negative impact of Cr needs to be investigated further. Olivine also seems to have a good ability to raise pH, but this effect should be further investigated. Because of olivine's promising results for the retention of the investigated elements, except for Cr, its efficiency over time should be investigated. A pre-treatment step removing particles should be considered to avoid precipitation clogging the system over time. Lime's promising treatment efficiency of Cr should be explored further, and a combination of lime and olivine should be tested, both as mixtures and consecutive steps. Different potential polishing steps should be looked into further, and another master student is doing this.[82]

These experiments have been performed on a medium scale, and larger scale experiments would give a better indication of the potential of the treatment mediums. In this thesis the scale of the treatment medium per water has not been considered, and this should be focused on in future experiments to find the right amount of treatment medium that does not saturate too soon. In general the treatment capacity of the treatment mediums per water should be looked into, as well as technical considerations like pump capacity in a potential treatment system. Safe waste management of the used treatment mediums should also be considered, and a method for changing out used treatment medium for new treatment medium, without causing desorption or leaching, should be looked into.

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Appendix A – Trondheim municipality's system and the tunnel

Throughout the entire sampling and experiment period of this thesis Trondheim municipality has been testing their own system inside the tunnel. This system evolved over time, and several changes were made, both in the system and in what treatment mediums they tested. In 2018 and the beginning of January 2019 the system comprised of open tubs and a pipe filled with lime. A picture of the system is presented in figure A.1.

Figure A.1 Trondheim municipality's system in 2018 and the beginning of January 2020. The lime green pump is taking water in from the tunnel and into the first tub. It passes on further through lime in the metal cylinder in the middle, before it goes into the final open tub and finally exits back into the tunnel.

In January 2019 the system was altered; the metal cylinder was exchanged for a traffic cone placed upside-down in a tub. January 29th the cone was filled with olivine, and water was pumped from the bottom of the cone and spilled over the edges and into the tub, where it eventually passed on through the final open tub and back into the tunnel. A picture of this system is presented in picture A.2.

Figure A.2 The system after 29th of January where the treatment medium, in this case olivine, was placed in an upside-down traffic cone in the middle tub.

After testing this system for a while, a new system was tested which consisted of a large plexiglass pipe filled with lime where water entered through the bottom and exited through the top into the open tub on the end and finally into the tunnel. This system was installed sometime between March 22nd and May 24th 2019, and is shown in figure A.3.

Figure A.3 Trondheim municipality's system that was installed sometime between March 22nd and May 24th 2019, with a plexiglass pipe filled with lime.

This system was tested for a while and in the fall of 2019 another change was made. Three more plexiglass pipes were added so that the system consisted of a total of four plexiglass pipes where treatment mediums such as olivine or lime could be filled into the pipes. The system, without any treatment mediums, is presented in figure A.4.

Figure A.4 Trondheim municipality's system used from the fall of 2019, here shown without any treatment mediums.

This system was made so that more than one treatment medium could be tested at one time. During the fall of 2019 pumps were added to the system that pumped NaOH and polymer into the pipes, and a sedimentation basin was added both in the start and end of the system. This was the final system in the sampling period of this thesis.

Samples that were taken inside the tunnel were taken at the sampling points "tunnel wall", "left tunnel wall", "colorful wall", "first collection", "second collection", "dripping from ceiling", and "threshold", and the location of these is presented in figure 2.3. Pictures of the sampling sites are shown in figure A.5-A.

Figure A.5 Water coming through the tunnel wall. This is the sampling point "tunnel wall", and samples were taken of a weak stream approximately in the middle of this picture, not visible due to the shadow. The sampling point "left tunnel wall" was not photographed, but was similar to this.

Figure A.6 The sampling point “colorful wall”, where water was sampled from the thin stream dripping below the gray-ish stripe in the middle of the picture.

Figure A.7 The sampling point “first collection” where water is collected into pipes and being led directly to the threshold. Samples were taken of a thin stream of water, not visible in this picture, running into the box.

Figure A.8 The sampling point “second collection” where water is collected into pipes and being led directly to the threshold. Samples were taken of the water running along the wall and into the box.

Figure A.9 The sampling point "dripping from ceiling". This was only sampled once because there was no water dripping from the ceiling the second sampling date.

Figure A.10 The threshold where water from inside the tunnel runs over the edge when the water levels are high enough. This is also where the water from the sampling points "first collection" and "second collection" are led. Samples here were taken just over the threshold.

With all of the changes in Trondheim municipality's system, the sampling point for water taken in from the tunnel also changed. In the first and second systems (figure A.1 and A.2) untreated water was sampled from the first open tub (labeled 1). After the third system was installed, the tunnel water was sampled through a hose bringing water in from inside the tunnel. This hose is pictured in figure A.11.

Figure A.11 The hose bringing untreated tunnel water in from inside the tunnel. This was the sampling point of the tunnel water when there was no sampling possibility in Trondheim municipality's systems due to closed pipes.

Appendix B – Calculations

B.1 Hardness of coastal water

The classification of condition of Cd depends on the hardness in the water of the recipient (coastal water). This was calculated based on results from ICP-MS analyses of coastal water samples taken in Hanne B. Olsen's thesis. The calculations were based on the calcium concentrations in eight water samples taken of the sea near the entrance to the Killingdal tunnel, where water has been observed seeping out from the tunnel. The concentrations were:

Ca [μg/L]
288 025
340 551
324 286
318 279
346 175
306 820
396 459
374 431

The average was found by using the average function in excel, and it was found to be 336 878 [μg/L].

The hardness of water in the Norwegian Environment Agency's classification system is based on $CaCO_3$. The weight% if Ca in $CaCO_3$ was calculated based on molar mass. The molar mass was found in SI Chemical Data[84] to be:

Ca	40.08	[g/mol]
C	12.01	[g/mol]
O*3	48	[g/mol]
Tot	100.09	[g/mol]

Then the weight% of Ca in $CaCO_3$ was calculated:

$$\frac{m_{Ca} \times 100}{m_{tot}} = \frac{40.08\, {}^{g}/_{mol} \times 100}{100.09\, {}^{g}/_{mol}} = 40.04\% \tag{B.1.1}$$

The Ca concentration from the coastal water samples were then converted to $CaCO_3$

$$\frac{[\text{Ca in coastal water samples}] \times 100}{\text{weight\% Ca in CaCO}_3} = \frac{336\,878\, {}^{\mu g}/_{L} \times 100}{40.04} \tag{B.1.2}$$

$$= 841\,354\, {}^{\mu g}/_{L} = 841\, {}^{mg}/_{L}$$

This is > 200 mg/L, and therefore 1.5 μg/L was chosen for class III for Cd and 15 μg/L for class IV.

B.2 Treatment efficiency

The treatment efficiency in the different experiments were calculated using this formula:

$$100 - \frac{[\text{after treatment}]}{[\text{before treatment}]} \times 100\% = \text{treatment efficiency in } \% \qquad \text{(B.2.1)}$$

Where [ ] means concentration.

Appendix C – ICP-MS data

C.1 ICP-MS results

The instrument used for HR-ICP-MS analysis was ELEMENT 2, but due to a malfunction in the instrument, some samples were analyzed with Agilent 8800. The analysis in the Agilent 8800 instrument was performed in two different modes; H2 and O2. In addition several different isotopes were analyzed for some of the elements. The mode and isotope with the lowest average RSD was chosen as the main result in this thesis. Table C.1.1 shows the concentrations and corresponding RSD of Cd, Pb, Cr, Ni, Fe, Cu, Zn, and As in the samples used in this thesis. All samples are corrected with blanks. The average concentrations in the blanks that the samples are corrected for are shown in a green row above the samples.

The concentrations of all elements analyzed for in all samples are presented in table C.1.2-C.1.5, with corresponding RSD.

The concentrations in the blanks are presented in table C.1.6 and C.1.7.

Table C.1.1 The concentrations and corresponding RSD of Cd, Pb, Cr, Fe, Ni, Cu, Zn, and As in all samples. There are two different blanks used to correct for samples. These blanks are presented in green rows, and the concentrations of the elements found are presented. When an element was not found in the blank, the cell is left empty. The blanks are used for correction in the samples presented under them. The instrument ELEMENT 2 was used for the samples under the first heading, and Agilent 8800 was used for the samples under the second heading. In the second heading the selected mode (either [H2] or [O2]) is shown.

				Cd114(LR)		Pb208(LR)		Cr53(MR)		Fe56(MR)		Ni60(MR)		Cu63(MR)		Zn66(MR)		As75(HR)	
				Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD
Sample-number	Sample name	Date	Sample ID	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%
Average of two blanks, used for batch 1-61 and 1-35										0,46	8,8					0,2	24,1		
2	Tunnel water	12.12. 2018	Steen-1-61-2	35,562	2,2	47,644	1,4	6,74	3,0	15 235,4	0,3	36,1	5,1	16 299,6	1,2	11 156,9	0,8	3,28	9,1
8	Tunnel water	17.12. 2018	Steen-1-61-8	50,855	1,6	31,624	1,9	4,86	1,2	8 963,8	0,7	41,3	1,1	11 370,0	4,1	14 644,0	2,1	0,77	10,6
14	Tunnel water	18.01. 2019	Steen-1-61-14	25,119	1,2	29,017	1,2	2,40	4,7	9 998,7	2,1	26,3	1,2	8 357,9	1,7	7 223,4	1,0	0,75	9,2
20	Tunnel water	29.01. 2019	Steen-1-61-20	28,381	1,9	18,423	0,7	0,70	9,2	1 255,4	1,0	29,6	2,3	8 750,2	0,9	8 041,1	2,7	0,35	16,0
25	Tunnel water	29.01. 2019	Steen-1-61-25	32,952	1,2	25,361	2,2	3,57	1,5	9 324,7	0,7	36,1	1,0	10 228,1	1,3	9 832,3	3,8	0,75	9,3
28	Tunnel water	29.01. 2019	Steen-1-61-28	38,171	2,3	29,684	1,6	5,27	3,0	14 488,4	1,3	36,8	1,2	10 552,5	2,0	10 978,3	1,8	1,05	19,2
31	Tunnel water	31.01. 2019	Steen-1-61-31	29,619	1,0	31,685	0,8	5,59	1,3	11 421,8	0,8	50,2	0,7	9 618,9	1,3	9 193,7	1,1	1,42	12,7
35	Tunnel water	07.02. 2019	Steen-1-61-35	32,949	0,6	33,265	0,3	6,35	2,6	14 709,0	1,4	54,9	1,7	8 115,3	1,5	9 687,0	2,0	1,87	13,9
40	Tunnel water	02.03. 2019	Steen-1-61-40	33,390	1,7	50,759	1,3	8,47	1,2	13 996,3	0,7	64,4	1,1	13 064,1	0,2	10 092,3	0,2	0,78	32,6
46	Tunnel water	05.03. 2019	Steen-1-61-46	33,080	1,2	52,996	0,8	8,99	2,2	14 097,0	0,1	68,1	4,2	13 321,0	2,0	10 660,3	0,6	0,71	15,5
51	Tunnel water	11.03. 2019	Steen-1-61-51	35,101	1,5	54,717	0,4	9,04	2,8	14 256,6	2,5	69,8	1,4	13 539,0	2,2	10 815,8	3,7	0,62	25,5
8	Tunnel water	03.06. 2019	Steen-1-35-8	17,976	1,2	5,126	1,2	0,11	20,8	3 812,7	2,9	24,02	4,0	6 210,02	0,3	5 632,6	1,9	0,78	12,6
26	Treated in TM's system	29.01. 2019	Steen-1-61-26	5,778	1,9	0,026	6,0	0,32	7,4	2,7	2,3	14,3	5,3	60,7	1,5	746,6	1,7	0,29	12,4
29	Treated in TM's system	29.01. 2019	Steen-1-61-29	32,971	0,9	3,614	1,9	0,22	10,8	2 416,9	0,6	29,9	0,1	7 010,9	3,0	9 385,5	0,9	0,30	26,8
32	Treated in TM's system	31.01. 2019	Steen-1-61-32	29,928	1,1	31,403	2,4	5,64	2,2	10 439,7	0,2	56,0	0,8	9 563,6	1,6	9 268,5	2,2	1,29	9,1
36	Treated in TM's system	07.02. 2019	Steen-1-61-36	31,535	1,4	32,387	1,4	6,11	2,7	13 674,8	1,8	57,2	0,3	7 926,5	0,5	9 490,2	1,2	1,91	17,5

41	Treated in TM's system	02.03.2019	Steen-1-61-41	29,508	0,1	28,773	2,4	4,01	4,0	18 538,9	1,4	23,5	2,1	6 598,1	1,4	7 748,0	1,0	3,50	13,0
47	Treated in TM's system	05.03.2019	Steen-1-61-47	21,414	1,3	38,787	1,8	5,78	1,9	28 401,5	1,2	25,1	0,7	7 247,1	1,1	6 388,1	1,3	5,04	14,6
52	Treated in TM's system	11.03.2019	Steen-1-61-52	30,252	1,2	47,857	2,1	7,54	3,0	33 519,6	2,8	34,4	4,4	7 962,4	1,6	8 802,5	2,8	7,24	2,9
58	Treated in TM's system	22.03.2019	Steen-1-61-58	25,382	1,5	50,042	3,7	8,30	3,1	34 650,8	1,8	32,8	1,4	8 420,2	0,6	7 562,1	1,6	8,13	8,8
13	Tunnel wall	03.06.2019	Steen-1-35-13	7,795	0,9	2,583	1,1	31,29	3,1	373 901,9	3,8	44,11	3,8	23 756,57	3,2	2 078,8	1,8	25,21	9,4
16	Colorful wall	06.06.2019	Steen-1-35-16	0,521	2,1	2,169	3,4	0,25	4,6	201,6	1,5	2,59	5,7	121,99	3,1	131,8	1,7	0,62	52,3
17	First collection	06.06.2019	Steen-1-35-17	0,759	4,1	0,024	9,6	0,45	5,3	15,7	0,5	6,55	4,2	44,53	1,9	214,8	1,0	0,25	88,0
18	Second collection	06.06.2019	Steen-1-35-18	1,540	1,5	1,471	1,5	0,40	18,8	309,4	2,9	3,24	2,4	101,55	2,3	558,4	0,9	0,30	33,3
19	Dripping from ceiling	06.06.2019	Steen-1-35-19	0,657	1,4	0,029	8,7	0,22	9,3	13,8	3,1	1,97	8,2	9,71	3,4	284,0	2,9	0,41	31,7
20	Threshold	06.06.2019	Steen-1-35-20	2,169	1,0	0,451	2,5	0,21	8,5	104,7	2,8	2,35	15,6	43,76	1,6	671,1	1,2	0,16	35,5
21	Before rinse	06.06.2019	Steen-1-35-21	28,363	1,6	22,862	1,5	3,08	4,6	10 360,3	2,1	36,89	6,6	11 699,28	2,0	8 119,4	2,9	0,62	11,8
22	After gravel rinse	06.06.2019	Steen-1-35-22	28,387	1,6	22,806	2,6	3,29	3,7	9 723,5	1,7	37,52	4,6	11 659,42	4,3	8 072,5	3,2	0,76	23,9
23	Olivine exp. 1	06.06.2019	Steen-1-35-23	0,053	6,3	0,321	1,7	9,57	5,6	210,9	2,6	0,17	9,8	362,20	2,9	58,8	12,1	0,14	30,1
24	Olivine exp. 1	06.06.2019	Steen-1-35-24	0,031	17,7	0,202	4,0	5,65	2,9	12,4	1,6	0,09	32,7	235,58	1,2	155,5	3,5	0,20	10,5
25	Olivine exp. 1	06.06.2019	Steen-1-35-25	0,017	27,2	0,210	1,5	6,71	3,1	18,3	1,6	0,06	93,0	195,45	1,2	115,1	5,4	0,23	9,0
26	Olivine exp. 1	06.06.2019	Steen-1-35-26	0,016	7,2	0,168	0,8	5,00	7,8	10,3	2,1	0,03	0,0	115,12	1,6	81,9	6,7	0,19	43,5
28	Lime exp. 1	08.06.2019	Steen-1-35-28	16,516	1,5	0,209	6,0	0,00	41,8	4 531,5	1,8	32,00	2,3	4 001,07	2,1	6 917,7	0,6	0,64	23,7
30	Lime exp. 1	08.06.2019	Steen-1-35-30	23,197	0,4	0,351	2,3	0,02	43,9	7 188,4	0,6	34,03	3,2	9 387,31	0,2	7 528,0	1,0	0,64	39,8
27	Lime exp. 2	08.06.2019	Steen-1-35-27	24,930	0,6	0,969	2,7	0,12	26,4	8 343,9	1,1	34,74	1,8	10 661,24	0,5	7 717,8	0,8	0,73	49,4
29	Lime exp. 2	08.06.2019	Steen-1-35-29	24,792	1,5	4,388	1,8	0,21	23,4	9 115,5	0,9	34,66	3,3	12 559,57	0,9	7 492,3	0,7	0,75	7,5
31	Lime exp. 2	08.06.2019	Steen-1-35-31	25,117	0,4	5,026	0,8	0,11	28,8	9 243,6	0,7	33,75	3,5	12 880,44	0,8	7 550,6	1,7	0,78	25,4

32	Lime exp. 2	09.06. 2019	Steen-1-35-32	23,598	0,4	9,152	0,3	0,38	8,0	8 611,6	1,0	31,66	1,4	13 802,05	0,7	7 122,0	0,7	0,77	11,9
Average of five blanks, used to for batch 36-81 and 82-162								0,02	17,7	0,3	5,5								
36	Tunnel water (hose on wall)	12.09. 2019	Repeatingtest -133	34,992	2,6	33,187	1,0	6,22	1,2	29 131	3,5	33,22	5,3	18 074,2	2,2	9 496,8	0,5	10,272	7,2
45	Tunnel water (hose on wall)	19.09. 2019	Stine-Steen-36-81-45-	31,543	0,3	3,467	1,5	0,16	8,0	4 512	1,2	19,49	1,5	5 410,6	1,3	8 645,6	1,3	0,623	15,8
51	Tunnel water (hose on wall)	02.10. 2019	Stine-Steen-36-81-51-	17,034	1,1	7,535	0,4	0,37	9,2	2 678	3,1	13,60	4,6	4 755,5	0,2	4 642,8	0,8	0,510	7,9
74	Tunnel water	11.10. 2018	Stine-Steen-36-81-74	55,765	0,8	32,899	1,3	10,70	1,5	37 934,4	3,4	41,93	1,4	16 950,2	2,7	15 277,5	1,9	3,097	9,9
46	Tunnel wall	19.09. 2019	Stine-Steen-36-81-46-	8,815	3,7	3,563	2,4	27,02	2,9	296 863	0,7	39,01	2,9	19 638,3	0,9	2 087,4	2,2	26,421	7,8
47	Left tunnel wall	19.09. 2019	Stine-Steen-36-81-47-	8,673	0,6	0,235	2,4	0,96	3,6	3 880	2,2	5,89	4,6	1 726,5	2,6	2 333,3	1,7	0,206	21,1
54	Tunnel wall	02.10. 2019	Stine-Steen-36-81-54-	14,058	1,5	2,341	1,1	21,15	1,9	192 205	4,0	33,89	4,3	14 403,1	4,6	3 505,4	1,5	12,851	10,1
36	Before rinse	12.09. 2019	Repeatingtest -133	34,992	2,6	33,187	1,0	6,22	1,2	29 131	3,5	33,22	5,3	18 074,2	2,2	9 496,8	0,5	10,272	7,2
37	After gravel rinse	12.09. 2019	Stine-Steen-36-81-37-	33,850	0,6	31,063	1,8	6,33	2,4	27 867	1,3	33,88	4,2	18 602,3	0,6	9 681,5	2,8	9,758	8,2
38	After gravel rinse	12.09. 2019	Stine-Steen-36-81-38-	32,931	1,4	28,899	0,9	6,24	1,5	27 391	1,7	35,24	3,6	18 007,1	0,9	9 693,3	0,6	9,133	9,4
39	Olivin exp. 2	12.09. 2019	Repeatingtest -134	0,015	14,0	0,177	8,4	8,35	2,9	20,6	0,6	0,08	22,7	159,8	1,4	68,6	2,6	0,590	1,7
40	Lime exp. 3	12.09. 2019	Stine-Steen-36-81-40	29,438	2,1	0,076	1,5	0,03	7,3	49,8	2,9	38,62	3,5	4 188,1	3,3	9 134,9	1,6	1,286	5,1
41	Olivine exp. 2	12.09. 2019	Stine-Steen-36-81-41	0,022	8,8	0,110	2,7	9,80	1,5	23,3	2,4	0,06	6,3	101,7	2,4	42,8	1,9	0,457	4,9
42	Lime exp. 3	12.09. 2019	Stine-Steen-36-81-42	33,316	0,7	0,077	6,2	0,02	14,1	17,3	1,2	33,20	1,1	4 737,5	3,8	9 810,4	1,4	1,110	8,4

				114 -> 114 Cd [H2]		208 -> 208 Pb [O2]		52 -> 52 Cr [H2]		56 -> 56 Fe [H2]		60 -> 60 Ni [O2]		63 -> 63 Cu [O2]		66 -> 66 Zn [H2]		75 -> 91 As [O2]	
				Conc. [µg/l]	RSD %	Conc. [µg/l]	RSD %	Conc. [µg/l]	RSD %	Conc. [µg/l]	RSD %	Conc. [µg/l]	RSD %	Conc. [µg/l]	RSD %	Conc. [µg/l]	RSD %	Conc. [µg/l]	RSD %
85	Tunnel water (hose on wall)	18.10. 2019	batch 82-162	31,88	0,88	1,56	4,1	0,422	19,9	1596	2,1	29,08	0,5	6339	2,1	8822	1,6	0,335	4,8
89	Tunnel water	12.11. 2019	batch 82-162	28,51	0,28	0,13	20,8	0,291	16,5	92,4	1,1	23,09	1,9	2029	1,6	7450	0,7	0,247	32,5
95	Tunnel water	18.12. 2019	batch 82-162	49,99	0,98	3,35	1,8	0,492	3,5	5698,7	1,9	53,22	2,1	20492	0,9	13188	1,6	0,258	23,6

91	Colorful wall	18.12. 2019	batch 82-162	0,19	12,09	1,24	3,0	0,753	13,6	90,1	2,1	1,78	19,7	97	1,0	41	4,1	0,434	13,9
92	First collection	18.12. 2019	batch 82-162	0,43	4,16	0,15	6,5	0,844	2,6	97,0	0,4	4,85	6,5	78	1,9	113	2,0	0,245	37,0
93	Second collection	18.12. 2019	batch 82-162	1,76	2,41	0,87	3,1	0,596	1,3	456,1	1,0	4,30	5,0	87	1,3	521	1,9	0,364	21,1
94	Threshold	18.12. 2019	batch 82-162	18,35	1,34	0,11	45,4	0,210	12,5	108,7	0,8	13,57	4,5	1234	3,2	4596	1,2	0,159	8,5
95	Before rinse	18.12. 2019	batch 82-162	49,99	0,98	3,35	1,8	0,492	3,5	5698,7	1,9	53,22	2,1	20492	0,9	13188	1,6	0,258	23,6
97	After gravel rinse	18.12. 2019	batch 82-162	47,60	1,35	3,22	5,8	0,560	3,5	5378,8	2,9	50,70	0,3	18658	0,7	12428	4,3	0,178	40,2
99	Olivine+activated carbon mix	18.12. 2019	batch 82-162	0,06	10,44	0,08	19,3	9,826	1,4	16,3	2,6	0,50	8,5	165	0,7	140	0,9	2,027	9,3
101	Olivine+activated carbon mix	18.12. 2019	batch 82-162	0,04	34,46	0,06	24,2	9,639	4,5	9,3	1,6	0,14	12,0	153	1,3	159	4,1	1,722	4,7
102	Olivine+activated carbon mix	18.12. 2019	batch 82-162	0,04	16,02	0,06	20,7	3,943	6,4	6,3	1,2	0,16	5,9	45	1,4	78	4,3	0,576	15,0
157	Before aeration filtered	24.02. 2020	batch 82-162	12,07	2,89	21,57	0,5	2,600	5,5	9534,8	1,7	18,22	2,8	7064	0,8	3228	1,8	0,692	8,2
159	Aeration 10 min filtered	24.02. 2020	batch 82-162	11,83	1,07	21,04	3,9	2,729	0,8	9368,1	1,6	17,40	4,4	6908	2,3	3198	2,3	0,784	22,0
160	Aeration 10 min unfiltered	24.02. 2020	batch 82-162	11,60	0,58	21,54	3,2	3,323	5,2	12283,0	1,7	17,31	2,9	6556	2,2	3093	0,9	3,237	2,2
161	Aeration 40 min filtered	24.02. 2020	batch 82-162	12,11	2,62	21,52	0,1	2,635	4,8	9432,9	2,2	18,34	5,4	7082	2,0	3249	1,9	0,820	3,5
162	Aeration 40 min unfiltered	24.02. 2020	batch 82-162	12,37	0,40	21,77	2,4	3,170	4,3	13036,4	2,0	17,02	1,9	6868	2,3	3285	1,0	3,259	4,7

Table C.1.2 The concentrations of all of the different elements analyzed for in all of the samples in batch 1-61 with corresponding RSD. The description of the samples is only given on the first page. When the table continues to the next pages only the sample number is given.

				Li7(LR)		Be9(LR)		B11(LR)		Y89(LR)		Zr90(HR)		Nb93(HR)		Cd114(LR)		Mo98(MR)	
				Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD
Sample-number	Sample name	Date	Sample ID	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%
2	Tunnel water	12.12.2018	Steen-1-61-2	12,40	1,9	0,272	3,5	30,4	2,4	13,270	3,2	0,163	36,3	0,005	72,5	35,562	2,2	0,043	59,1
8	Tunnel water	17.12.2018	Steen-1-61-8	13,44	1,2	0,330	11,3	34,3	0,9	16,556	1,9	0,110	22,1	0,007	48,0	50,855	1,6	0,036	60,6
14	Tunnel water	18.01.2019	Steen-1-61-14	10,66	2,1	0,239	5,1	28,3	4,2	11,387	1,0	0,052	16,5	0,002	86,8	25,119	1,2	0,031	46,3
20	Tunnel water	29.01.2019	Steen-1-61-20	11,76	3,2	0,239	11,9	26,5	1,6	13,253	2,6	0,047	47,5	0,004	114,6	28,381	1,9	0,029	89,9
25	Tunnel water	29.01.2019	Steen-1-61-25	12,32	1,7	0,289	8,2	27,8	3,5	15,025	0,8	0,086	9,3	0,009	10,2	32,952	1,2	0,022	24,6
28	Tunnel water	29.01.2019	Steen-1-61-28	12,68	2,6	0,300	4,4	29,4	1,4	16,189	1,0	0,109	23,9	0,010	62,4	38,171	2,3	0,070	12,6
31	Tunnel water	31.01.2019	Steen-1-61-31	11,46	0,6	0,284	17,3	26,7	1,2	14,379	1,7	0,104	36,1	0,006	14,0	29,619	1,0	0,040	1,5
35	Tunnel water	07.02.2019	Steen-1-61-35	11,38	1,3	0,270	17,9	27,9	1,4	12,860	2,6	0,121	13,6	0,010	72,1	32,949	0,6	0,055	40,4
40	Tunnel water	02.03.2019	Steen-1-61-40	12,05	3,2	0,263	4,5	26,9	1,3	14,305	0,5	0,134	4,6	0,008	53,9	33,390	1,7	0,045	33,8
46	Tunnel water	05.03.2019	Steen-1-61-46	12,12	3,1	0,272	10,8	26,4	3,6	14,385	2,3	0,171	14,7	0,009	73,3	33,080	1,2	0,038	32,7
51	Tunnel water	11.03.2019	Steen-1-61-51	12,68	1,5	0,306	8,7	27,2	3,5	15,269	4,6	0,196	19,1	0,008	58,2	35,101	1,5	0,051	73,0
3	Treated in TM's system	12.12.2018	Steen-1-61-3	12,52	1,6	0,306	9,8	30,0	0,8	14,268	1,2	0,028	44,6	0,004	34,6	35,762	1,3	0,025	46,5
9	Treated in TM's system	17.12.2018	Steen-1-61-9	12,95	1,8	0,263	4,6	32,1	1,4	16,476	1,5	8,730	172,7	-0,003	94,4	47,944	0,9	0,055	40,6
15	Treated in TM's system	18.01.2019	Steen-1-61-15	10,13	5,6	0,179	40,6	27,5	3,0	10,725	4,7	0,024	38,2	0,001	173,2	24,815	2,7	0,030	44,3
21	Treated in TM's system	29.01.2019	Steen-1-61-21	12,24	2,8	0,153	8,8	29,2	4,5	13,342	3,1	0,011	45,2	0,004	173,2	28,988	1,6	0,017	61,7
26	Treated in TM's system	29.01.2019	Steen-1-61-26	12,91	2,5	0,002	87,7	25,3	2,7	0,137	3,4	0,005	86,6	0,000	0,0	5,778	1,9	0,176	29,1
29	Treated in TM's system	29.01.2019	Steen-1-61-29	12,31	0,7	0,186	4,5	28,4	3,8	11,566	0,6	0,012	43,3	0,007	60,7	32,971	0,9	0,042	46,2
32	Treated in TM's system	31.01.2019	Steen-1-61-32	11,42	3,0	0,316	11,6	27,5	1,7	14,433	0,6	0,098	8,2	0,004	52,7	29,928	1,1	0,049	26,2
36	Treated in TM's system	07.02.2019	Steen-1-61-36	10,93	1,5	0,268	8,8	26,9	0,8	12,744	1,4	0,083	26,7	0,009	32,7	31,535	1,4	0,032	27,7
41	Treated in TM's system	02.03.2019	Steen-1-61-41	9,03	1,2	0,165	4,5	30,1	2,8	8,422	1,7	0,080	34,6	0,002	173,2	29,508	0,1	0,046	52,8
47	Treated in TM's system	05.03.2019	Steen-1-61-47	9,64	1,1	0,214	16,7	28,1	4,1	10,275	1,7	0,107	21,6	0,012	73,7	21,414	1,3	0,052	43,4
52	Treated in TM's system	11.03.2019	Steen-1-61-52	11,25	1,4	0,287	10,7	29,0	2,3	13,268	3,4	0,132	4,9	0,056	136,3	30,252	1,2	0,054	21,5
58	Treated in TM's system	22.03.2019	Steen-1-61-58	10,81	0,3	0,261	8,7	51,7	2,1	13,391	1,2	0,164	31,6	0,014	5,3	25,382	1,5	0,145	33,9

	Sn118(LR)		Cs133(LR)		La139(MR)		Ce140(LR)		Pr141(LR)		Nd146(LR)		Sm147(LR)		Eu151(LR)	
	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD
Sample-number	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%
2	0,011	5,1	0,200	4,9	25,589	8,1	52,270	0,5	5,200	1,4	21,322	1,5	3,656	2,1	0,486	2,4
8	0,010	41,8	0,211	2,3	27,883	0,6	65,292	1,1	6,533	2,1	26,758	2,6	4,517	2,4	0,584	4,1
14	0,011	4,3	0,186	2,5	20,840	7,5	43,854	2,4	4,354	0,8	17,856	2,1	3,012	3,0	0,370	0,7
20	0,008	22,7	0,205	1,9	22,945	10,3	49,261	0,9	4,808	1,6	19,820	0,7	3,346	1,2	0,419	3,0
25	0,006	32,5	0,204	3,6	25,817	1,0	58,596	1,4	5,822	2,1	24,379	2,8	4,026	7,1	0,495	0,9
28	0,045	14,4	0,203	6,6	27,702	1,6	61,909	2,5	6,131	2,1	25,350	3,6	4,269	2,0	0,542	3,1
31	0,008	18,6	0,214	2,3	24,533	1,3	53,425	0,5	5,380	0,8	21,879	1,2	3,644	2,3	0,475	0,6
35	0,008	6,8	0,213	1,0	21,321	2,5	46,871	2,2	4,692	1,5	19,540	0,4	3,307	1,4	0,431	1,9
40	0,021	23,9	0,202	4,2	27,059	7,4	55,346	0,3	5,678	3,3	23,358	2,0	3,900	2,9	0,497	1,2
46	0,018	7,2	0,210	5,5	25,689	1,8	54,565	2,3	5,550	1,7	23,463	2,1	3,835	2,6	0,498	5,1
51	0,025	23,7	0,223	6,2	26,749	6,2	57,520	3,8	5,748	3,6	24,440	2,9	4,073	0,7	0,512	2,7
3	0,011	7,8	0,204	4,5	22,896	0,7	51,754	0,6	5,155	3,2	21,273	2,5	3,550	1,8	0,474	2,5
9	0,005	24,8	0,205	3,7	27,417	1,4	63,059	1,3	6,038	1,1	25,265	0,7	4,157	1,7	0,545	2,3
15	0,007	34,6	0,186	4,4	20,323	6,5	41,647	1,5	4,079	1,6	16,521	3,1	2,726	5,9	0,341	5,2
21	0,012	23,7	0,206	6,7	22,646	8,3	49,168	1,9	4,781	0,3	19,544	1,2	3,140	3,0	0,395	2,3
26	0,011	16,9	0,179	5,3	0,268	4,4	0,317	6,1	0,030	2,0	0,116	6,0	0,015	26,0	0,004	16,9
29	0,009	21,7	0,191	2,9	20,631	0,9	44,355	3,2	4,379	2,1	17,372	2,8	2,740	3,6	0,352	3,1
32	0,007	28,3	0,208	8,1	24,411	0,8	52,284	1,6	5,272	1,3	21,465	0,5	3,620	0,2	0,454	1,3
36	0,012	19,9	0,207	4,1	20,367	4,3	44,751	1,4	4,582	1,7	19,203	0,4	3,165	3,1	0,421	3,1
41	0,005	28,2	0,151	5,0	15,812	1,3	32,388	2,1	3,702	2,3	14,841	1,7	2,380	4,7	0,303	4,0
47	0,008	9,8	0,175	5,0	18,827	9,6	37,614	2,0	3,902	0,5	16,007	2,0	2,586	1,5	0,344	1,9
52	0,006	46,3	0,205	3,1	22,605	3,2	47,221	0,7	4,731	1,8	19,909	2,6	3,305	0,3	0,422	3,6
58	0,033	15,3	0,209	4,6	22,495	0,6	48,959	3,3	4,905	2,3	20,910	2,5	3,385	3,0	0,430	2,5

	Gd157(HR)		Tb159(LR)		Dy163(LR)		Dy161(LR)		Ho165(LR)		Er166(LR)		Tm169(LR)		Yb172(LR)	
Sample-number	Conc. μg/L	RSD, %	Conc. μg/L	RSD, %	Conc. μg/L	RSD, %	Conc. μg/L	RSD, %	Conc. μg/L	RSD, %	Conc. μg/L	RSD, %	Conc. μg/L	RSD, %	Conc. μg/L	RSD, %
2	2,954	9,2	0,454	5,2	2,369	4,3	2,595	2,6	0,477	2,1	1,392	3,4	0,180	3,0	1,333	2,5
8	3,175	18,5	0,561	1,8	3,000	3,2	3,203	3,0	0,595	3,0	1,808	4,8	0,240	2,3	1,653	2,6
14	2,373	8,0	0,375	2,4	2,020	4,5	2,140	4,3	0,395	5,6	1,191	3,9	0,154	2,0	1,142	3,2
20	2,836	12,9	0,414	2,1	2,257	2,3	2,358	4,0	0,444	4,2	1,346	0,9	0,175	4,0	1,199	2,9
25	3,454	1,0	0,498	2,3	2,673	4,0	2,840	4,7	0,521	2,2	1,565	4,2	0,200	3,4	1,427	1,7
28	3,475	4,4	0,535	1,8	2,911	2,3	3,064	1,4	0,570	2,4	1,666	0,5	0,215	2,6	1,582	0,7
31	3,480	8,7	0,470	4,1	2,505	1,4	2,739	4,8	0,497	2,8	1,470	1,8	0,192	3,3	1,333	4,1
35	3,052	5,7	0,407	1,6	2,262	0,8	2,396	1,4	0,455	3,7	1,378	2,7	0,172	4,3	1,294	2,4
40	3,168	14,1	0,472	1,4	2,567	1,5	2,707	1,3	0,489	2,0	1,519	2,0	0,196	3,6	1,426	4,5
46	3,343	3,3	0,483	2,2	2,479	1,3	2,578	2,4	0,508	6,0	1,512	2,1	0,201	0,8	1,408	5,7
51	3,258	17,4	0,507	3,7	2,703	0,3	2,889	2,4	0,531	1,9	1,594	3,7	0,200	1,6	1,487	6,1
3	2,751	12,4	0,437	1,2	2,383	2,2	2,525	1,1	0,473	0,9	1,441	1,0	0,193	2,5	1,350	2,6
9	3,188	5,8	0,536	1,5	2,852	3,4	3,099	4,0	0,572	2,6	1,704	3,2	0,217	2,7	1,566	0,8
15	2,275	5,9	0,338	2,8	1,831	1,5	1,893	2,9	0,375	1,3	1,103	1,1	0,142	4,1	1,014	5,8
21	2,613	6,8	0,408	1,8	2,130	3,7	2,288	2,6	0,423	3,9	1,266	1,2	0,166	4,4	1,128	2,8
26	0,000	0,0	0,002	22,2	0,013	17,2	0,013	6,9	0,003	12,1	0,008	8,5	0,001	17,8	0,008	0,0
29	2,248	12,7	0,366	3,0	1,922	0,5	2,028	1,1	0,373	4,9	1,121	4,3	0,147	3,5	1,064	2,6
32	2,998	3,2	0,451	2,1	2,516	2,0	2,608	5,8	0,493	2,7	1,522	1,2	0,195	3,5	1,383	3,6
36	2,459	11,9	0,401	3,6	2,189	3,2	2,321	1,3	0,434	0,2	1,298	1,6	0,169	3,8	1,191	2,2
41	1,814	10,2	0,287	3,1	1,526	3,7	1,634	1,4	0,306	2,6	0,910	3,9	0,119	1,7	0,859	5,2
47	2,287	7,1	0,335	0,9	1,742	1,6	1,871	2,2	0,346	2,4	1,040	2,3	0,134	4,1	0,964	2,4
52	2,851	8,5	0,413	1,6	2,184	1,1	2,415	1,8	0,423	1,8	1,272	1,9	0,170	4,1	1,201	4,5
58	2,709	11,1	0,436	1,8	2,280	2,1	2,457	1,8	0,474	1,6	1,396	0,8	0,181	4,0	1,294	3,0

	Lu175(LR)		Hf178(LR)		Ta181(LR)		W182(LR)		Ir193(LR)		Pt195(LR)		Au197(LR)		Hg202(LR)	
	Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.	
Sample-number	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %
2	0,183	3,9	0,0083	49,9	0,0013	23,1	0,006	27,8	0,0003	86,6	0,0004	34,6	0,0075	37,5	0,064	3,3
8	0,238	2,3	0,0074	16,5	0,0009	45,3	0,004	17,2	0,0000	0,0	0,0013	56,8	0,0080	10,1	0,054	5,1
14	0,157	1,2	0,0062	15,8	0,0005	41,7	0,005	18,3	0,0002	89,2	-0,0001	173,2	0,0060	3,8	0,060	5,5
20	0,171	3,1	0,0050	33,5	0,0014	13,8	0,004	6,6	0,0001	94,4	0,0001	94,4	0,0033	32,9	0,046	5,3
25	0,202	0,7	0,0048	3,9	0,0010	57,9	0,006	14,6	0,0004	173,2	0,0000	173,2	0,0053	58,2	0,031	9,3
28	0,217	3,6	0,0077	27,9	0,0012	25,1	0,007	21,6	0,0000	0,0	0,0004	28,9	0,0033	32,7	0,043	2,4
31	0,200	3,8	0,0077	26,1	0,0014	14,9	0,004	16,6	0,0002	87,7	0,0006	114,6	0,0058	13,3	0,057	10,3
35	0,180	4,2	0,0072	17,5	0,0016	13,3	0,003	29,8	0,0001	173,2	0,0001	173,2	0,0045	16,8	0,033	5,1
40	0,199	0,9	0,0064	16,3	0,0011	16,2	0,013	19,0	0,0001	173,2	0,0000	173,2	0,0050	27,9	0,011	2,4
46	0,203	2,1	0,0090	31,6	0,0018	36,4	0,007	26,9	0,0000	0,0	0,0008	87,0	0,0029	48,0	0,062	2,3
51	0,214	8,5	0,0101	27,8	0,0007	75,8	0,005	31,8	0,0000	0,0	0,0008	114,6	0,0041	26,9	0,055	7,0
3	0,189	2,8	0,0032	0,1	0,0007	37,6	0,006	18,9	0,0000	0,0	0,0003	100,0	0,0071	8,3	0,006	4,9
9	0,215	1,0	0,0040	51,2	0,0012	51,0	0,005	10,2	0,0000	0,0	0,0004	56,8	0,0054	23,5	0,023	10,0
15	0,146	3,8	0,0046	28,4	0,0010	65,0	-0,002	53,7	0,0001	173,2	0,0010	36,7	0,0052	17,1	0,026	7,8
21	0,163	3,2	0,0030	47,7	0,0005	59,7	0,005	41,8	0,0001	173,2	0,0011	51,5	0,0039	35,3	0,033	7,4
26	0,001	11,6	0,0008	43,4	0,0000	173,2	0,074	9,0	0,0001	173,2	0,0005	22,9	0,0035	21,6	0,056	2,5
29	0,150	2,4	0,0047	35,3	0,0008	100,0	0,003	35,4	0,0001	173,2	0,0001	91,7	0,0077	14,8	0,062	9,5
32	0,203	0,8	0,0076	5,3	0,0015	39,0	0,002	16,9	0,0001	173,2	0,0005	26,6	0,0043	21,6	0,014	1,4
36	0,170	5,2	0,0083	13,7	0,0012	47,4	0,003	26,3	0,0000	0,0	0,0008	21,7	0,0045	26,0	0,023	8,3
41	0,125	1,2	0,0046	22,2	0,0011	28,4	0,004	43,5	0,0001	173,2	0,0007	32,8	0,0110	24,2	0,149	5,8
47	0,141	3,1	0,0041	39,1	0,0008	10,2	0,005	28,6	0,0000	0,0	-0,0001	173,2	0,0084	44,7	0,114	2,6
52	0,178	2,8	0,0073	25,0	0,0017	21,6	0,003	13,6	0,0004	100,0	0,0002	100,0	0,0066	11,3	0,083	5,0
58	0,186	1,9	0,0081	31,5	0,0021	29,6	0,006	32,4	0,0001	173,2	0,0009	57,0	0,0047	24,2	0,798	10,9

Sample-number	Tl205(LR)		Pb208(LR)		Bi209(LR)		Th232(LR)		U238(LR)		Na23(MR)		Mg25(MR)		Al27(MR)	
	Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.	
	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %
2	1,0022	0,9	47,644	1,4	0,0473	12,8	1,65	3,3	1,612	2,5	50 103	2,3	12 601	2,5	5 148	1,3
8	0,9804	2,3	31,624	1,9	0,0279	4,8	1,29	3,8	1,758	2,9	49 551	2,0	14 969	1,1	5 435	2,1
14	0,7725	1,5	29,017	1,2	0,0167	9,8	0,53	6,1	1,167	0,9	52 239	0,6	11 155	1,6	3 680	1,2
20	0,8370	2,7	18,423	0,7	0,0185	11,7	0,22	3,7	1,211	3,2	47 642	1,9	11 672	1,0	3 703	1,5
25	0,7800	0,7	25,361	2,2	0,0202	4,2	1,17	3,6	1,417	1,0	45 851	2,5	12 297	1,5	4 929	1,2
28	0,7843	1,4	29,684	1,6	0,0238	2,0	1,72	3,8	1,524	1,3	43 968	1,9	12 643	0,9	5 344	1,9
31	0,8010	2,8	31,685	0,8	0,0306	4,0	1,26	7,8	1,477	1,6	46 961	2,1	13 126	1,9	5 569	2,0
35	0,7828	1,0	33,265	0,3	0,0247	17,2	1,25	5,8	1,433	0,2	47 282	0,7	13 410	2,2	5 272	0,8
40	0,8381	4,4	50,759	1,3	0,0222	15,2	2,42	4,9	1,535	0,9	47 089	1,1	13 897	0,3	5 541	1,0
46	0,8532	4,5	52,996	0,8	0,0206	10,1	2,50	5,6	1,534	2,5	48 467	0,8	14 058	2,3	5 736	0,3
51	0,8923	2,2	54,717	0,4	0,0218	6,3	2,69	4,0	1,629	3,8	50 976	0,6	14 310	1,4	5 831	1,1
3	1,0532	0,5	16,008	2,4	0,0315	7,3	0,15	5,5	1,373	2,1	49 937	1,6	12 404	1,8	4 042	0,3
9	0,9346	2,9	12,503	2,0	0,0135	4,9	0,13	5,9	1,384	1,0	46 912	3,0	14 221	2,3	3 552	0,2
15	0,7539	1,4	9,523	2,0	0,0118	15,9	0,06	18,7	0,795	1,8	51 571	1,7	11 254	1,4	1 646	1,5
21	0,8738	3,3	3,707	2,9	0,0040	33,1	0,03	18,0	0,711	0,5	48 400	0,2	11 917	2,5	610	1,0
26	0,5418	2,5	0,026	6,0	0,0049	17,8	0,10	9,9	0,297	0,8	48 725	3,1	11 741	2,7	43	2,1
29	0,5661	1,2	3,614	1,9	0,0047	7,5	0,13	9,1	0,859	3,1	44 690	1,6	12 899	0,7	2 359	0,5
32	0,7948	3,3	31,403	2,4	0,0245	5,6	1,30	3,7	1,483	2,2	47 432	0,5	13 602	1,6	5 560	0,5
36	0,7591	3,1	32,387	1,4	0,0244	19,0	1,22	4,1	1,375	1,8	46 403	1,9	13 044	2,0	5 170	2,0
41	0,6311	1,3	28,773	2,4	0,0117	7,5	0,99	4,5	0,930	1,0	74 832	1,4	11 468	1,9	2 762	1,8
47	0,7080	1,9	38,787	1,8	0,0181	12,5	1,50	3,0	0,998	0,6	82 607	2,8	11 701	1,8	3 655	2,2
52	0,8204	2,3	47,857	2,1	0,0319	2,3	2,00	2,9	1,227	0,1	73 479	1,2	13 513	1,6	4 780	0,8
58	0,8088	3,9	50,042	3,7	0,0728	8,0	1,88	6,4	1,392	1,8	67 227	3,3	12 890	2,1	5 026	2,0

Sample-number	Si29(MR)		P31(MR)		S34(MR)		Cl35(MR)		K39(MR)		Ca44(MR)		Sc45(MR)		Ti49(MR)	
	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %
2	11 417	2,1	6,80	1,3	197 890	2,8	54 783	2,8	6 232	2,7	145 765	1,2	0,637	2,0	2,54	6,2
8	11 938	1,3	5,26	4,2	210 664	0,9	55 375	2,0	6 268	1,0	166 356	1,6	0,664	6,4	0,92	7,2
14	10 215	2,6	3,57	2,6	176 458	0,5	74 235	3,6	7 197	4,5	153 017	1,0	0,423	4,8	0,80	4,4
20	10 443	0,8	2,78	4,8	186 938	1,6	61 594	3,9	6 857	2,3	165 421	0,7	0,331	7,1	0,08	55,6
25	11 218	2,7	4,09	3,8	192 053	2,2	57 202	1,8	6 684	1,9	155 253	0,9	0,534	5,8	1,16	12,3
28	11 232	1,2	5,82	4,1	192 656	1,6	54 758	3,3	6 453	0,8	150 236	1,0	0,602	1,8	1,85	1,8
31	11 757	1,3	6,18	4,7	195 646	1,6	60 396	10,7	6 851	1,2	153 938	0,8	0,605	2,5	2,38	2,4
35	12 042	0,7	9,33	4,8	191 609	1,7	58 823	9,6	6 975	1,8	147 919	1,3	0,578	1,2	3,04	7,1
40	12 334	1,7	4,61	5,9	193 945	1,2	61 834	4,4	6 823	2,3	133 564	1,9	0,714	3,1	1,33	1,9
46	12 575	1,7	5,76	9,7	198 354	1,0	58 360	0,4	7 018	2,1	138 218	2,4	0,755	3,5	1,42	7,2
51	13 647	1,7	5,59	3,1	201 321	0,7	60 124	3,7	7 284	3,0	139 598	2,5	0,744	4,8	1,16	6,5
3	10 968	0,1	4,69	4,7	194 939	1,8	56 644	1,6	5 960	2,4	173 709	1,8	0,383	6,7	0,08	29,8
9	11 121	1,2	3,71	7,8	197 874	1,2	54 061	5,0	5 735	3,1	173 429	0,6	0,302	1,7	0,08	75,7
15	10 221	1,0	2,64	5,6	177 923	1,7	73 655	5,0	7 374	1,4	168 974	1,1	0,191	4,1	0,06	26,6
21	11 193	2,7	2,92	2,8	186 497	2,3	63 674	7,6	6 916	3,6	177 223	3,2	0,095	10,3	0,06	0,2
26	6 557	1,0	1,64	6,3	192 313	2,9	55 632	3,1	7 769	2,4	202 505	1,9	0,007	47,2	0,07	19,6
29	10 462	2,2	2,79	6,1	191 025	2,2	55 984	10,2	6 518	1,8	178 569	3,0	0,138	2,4	0,05	45,0
32	11 878	2,7	6,87	4,6	192 620	2,1	59 155	7,6	6 849	1,8	151 468	1,5	0,609	4,7	1,86	7,8
36	11 723	1,6	8,65	0,9	187 848	1,6	59 738	7,3	6 850	3,0	139 968	1,9	0,562	1,6	2,95	6,7
41	8 388	0,3	12,22	5,3	150 732	3,5	159 188	4,9	6 713	3,8	130 690	1,9	0,349	0,8	6,35	5,0
47	10 305	1,8	17,89	2,3	181 448	1,8	177 684	4,6	7 627	4,0	139 488	1,3	0,473	1,4	10,39	4,7
52	11 046	2,7	24,81	2,4	194 157	0,7	133 510	1,8	8 175	7,3	148 017	0,6	0,542	2,2	9,74	2,8
58	10 903	2,6	21,59	2,6	191 308	0,2	219 195	7,1	7 822	4,6	137 971	3,1	0,608	2,1	11,16	1,1

	V51(MR)		Cr53(MR)		Mn55(MR)		Fe56(MR)		Co59(MR)		Ni60(MR)		Cu63(MR)		Zn66(MR)	
	Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.	
Sample-number	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %
2	0,026	40,8	6,74	3,0	481,99	0,9	15 235,4	0,3	81,26	0,9	36,1	5,1	16 299,6	1,2	11 156,9	0,8
8	0,045	9,3	4,86	1,2	623,99	2,6	8 963,8	0,7	84,84	2,4	41,3	1,1	11 370,0	4,1	14 644,0	2,1
14	0,026	16,0	2,40	4,7	396,54	1,4	9 998,7	2,1	62,51	0,5	26,3	1,2	8 357,9	1,7	7 223,4	1,0
20	0,014	31,2	0,70	9,2	480,93	1,7	1 255,4	1,0	66,09	1,8	29,6	2,3	8 750,2	0,9	8 041,1	2,7
25	0,028	18,0	3,57	1,5	520,06	1,5	9 324,7	0,7	76,47	1,0	36,1	1,0	10 228,1	1,3	9 832,3	3,8
28	0,024	24,7	5,27	3,0	539,67	2,0	14 488,4	1,3	77,25	0,3	36,8	1,2	10 552,5	2,0	10 978,3	1,8
31	0,053	22,6	5,59	1,3	532,91	0,5	11 421,8	0,8	74,94	0,9	50,2	0,7	9 618,9	1,3	9 193,7	1,1
35	0,091	13,8	6,35	2,6	514,01	1,4	14 709,0	1,4	72,91	1,9	54,9	1,7	8 115,3	1,5	9 687,0	2,0
40	0,025	49,7	8,47	1,2	570,39	1,3	13 996,3	0,7	79,66	1,1	64,4	1,1	13 064,1	0,2	10 092,3	0,2
46	0,025	13,8	8,99	2,2	592,51	3,2	14 097,0	0,1	82,09	1,0	68,1	4,2	13 321,0	2,0	10 660,3	0,6
51	0,029	26,6	9,04	2,8	592,14	0,9	14 256,6	2,5	84,19	4,4	69,8	1,4	13 539,0	2,2	10 815,8	3,7
3	0,012	36,5	0,57	6,5	476,08	2,1	2 924,5	1,0	75,25	1,3	33,7	0,8	15 746,3	1,2	10 349,5	1,4
9	0,012	38,3	0,43	14,5	595,89	2,2	4 060,8	2,2	76,98	2,8	39,7	2,1	10 833,3	1,1	13 691,7	0,7
15	0,012	25,4	0,15	9,2	400,50	1,2	8 025,0	1,2	61,58	0,4	27,0	3,4	7 863,5	0,6	7 417,4	1,9
21	0,011	19,5	0,04	23,3	481,75	1,1	5 808,7	0,4	68,30	3,9	29,3	1,9	7 905,0	0,6	8 024,8	1,8
26	0,125	14,2	0,32	7,4	254,63	1,0	2,7	2,3	28,09	0,4	14,3	5,3	60,7	1,5	746,6	1,7
29	0,013	5,1	0,22	10,8	479,87	1,0	2 416,9	0,6	60,72	2,2	29,9	0,1	7 010,9	3,0	9 385,5	0,9
32	0,094	5,3	5,64	2,2	529,96	1,2	10 439,7	0,2	73,99	0,9	56,0	0,8	9 563,6	1,6	9 268,5	2,2
36	0,154	15,7	6,11	2,7	509,94	3,2	13 674,8	1,8	71,09	1,7	57,2	0,3	7 926,5	0,5	9 490,2	1,2
41	0,054	19,6	4,01	4,0	233,49	1,6	18 538,9	1,4	47,13	3,3	23,5	2,1	6 598,1	1,4	7 748,0	1,0
47	0,121	6,8	5,78	1,9	308,56	0,6	28 401,5	1,2	54,58	1,5	25,1	0,7	7 247,1	1,1	6 388,1	1,3
52	0,130	7,5	7,54	3,0	444,06	1,5	33 519,6	2,8	63,50	3,1	34,4	4,4	7 962,4	1,6	8 802,5	2,8
58	0,142	6,3	8,30	3,1	476,66	1,6	34 650,8	1,8	67,46	3,2	32,8	1,4	8 420,2	0,6	7 562,1	1,6

	Ga69(MR)		As75(HR)		Rb85(MR)		Sr88(MR)		Ag109(MR)		Sb121(MR)		Ba137(MR)		Br81(HR)	
Sample-number	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %
2	0,475	4,4	3,28	9,1	6,97	2,0	458,8	2,9	0,129	9,1	0,082	18,3	13,83	10,9	62,4	0,6
8	0,226	1,9	0,77	10,6	7,21	2,1	497,6	2,3	0,066	4,3	0,064	7,7	13,73	2,4	60,5	6,9
14	0,127	12,2	0,75	9,2	6,45	2,8	457,8	1,4	0,045	19,5	0,076	52,7	11,29	9,0	48,3	11,8
20	0,010	50,8	0,35	16,0	6,51	3,1	471,3	1,6	0,037	0,0	0,556	13,8	11,05	8,6	44,8	4,5
25	0,296	18,9	0,75	9,3	6,75	4,4	471,1	1,3	0,037	12,1	0,047	16,4	11,08	4,3	56,7	5,5
28	0,450	6,7	1,05	19,2	6,98	2,2	460,5	0,8	0,054	8,2	0,580	2,6	11,69	0,6	60,0	12,4
31	0,350	2,7	1,42	12,7	7,23	1,3	497,0	0,6	0,062	12,6	0,076	41,6	16,19	3,0	47,1	16,6
35	0,471	7,7	1,87	13,9	7,29	1,5	489,9	0,9	0,050	25,6	0,071	31,1	15,50	1,9	52,4	12,9
40	0,606	8,5	0,78	32,6	7,26	3,2	460,8	1,5	0,133	10,3	0,122	18,2	13,84	12,3	59,7	15,7
46	0,679	4,9	0,71	15,5	7,56	0,9	465,1	2,0	0,105	21,6	0,058	43,5	12,81	2,2	64,8	8,6
51	0,770	9,8	0,62	25,5	7,83	5,5	480,6	0,9	0,133	20,2	0,060	49,5	13,53	8,9	62,4	24,6
3	0,025	40,6	2,03	6,6	6,81	5,9	459,4	1,0	0,102	1,6	0,072	7,9	12,50	1,8	52,4	4,1
9	0,018	39,8	0,69	23,6	6,88	0,8	478,6	1,1	0,070	28,9	0,039	33,1	14,14	3,0	51,1	15,7
15	0,010	49,5	0,73	10,8	6,45	1,9	469,0	1,4	0,045	32,1	0,066	24,1	11,79	10,2	54,4	14,8
21	0,020	56,3	0,43	20,0	6,67	3,1	469,8	2,5	0,038	46,2	0,040	33,1	11,22	6,5	51,8	15,9
26	0,044	11,6	0,29	12,4	7,13	1,6	860,5	2,6	0,006	24,7	0,063	50,8	30,36	1,6	53,5	6,7
29	0,013	41,1	0,30	26,8	6,96	0,7	609,1	1,9	0,022	71,8	0,032	45,6	20,68	1,5	53,4	22,4
32	0,343	8,3	1,29	9,1	6,99	1,9	491,6	1,1	0,053	14,7	0,059	35,7	16,71	0,3	59,6	2,8
36	0,427	10,6	1,91	17,5	7,17	0,4	469,4	0,4	0,053	19,2	0,059	17,8	15,61	0,6	58,3	6,3
41	0,420	10,5	3,50	13,0	5,98	1,3	428,7	2,0	0,115	21,5	0,076	46,7	16,04	0,3	82,3	17,2
47	0,676	8,5	5,04	14,6	6,70	2,1	469,3	1,6	0,090	43,7	0,114	11,3	13,32	9,8	80,5	9,6
52	0,827	4,3	7,24	2,9	7,02	2,0	504,8	1,6	0,099	22,9	0,099	11,9	13,11	1,9	87,0	22,9
58	0,914	2,2	8,13	8,8	7,21	3,0	478,9	1,9	0,088	20,0	0,151	16,8	11,99	2,4	76,5	11,3

Table C.1.3 The concentrations of all of the different elements analyzed for in all of the samples in batch 1-35 with corresponding RSD. The description of the samples is only given on the first page. When the table continues to the next pages only the sample number is given.

				Li7(LR)		Y89(LR)		Zr90(LR)		Cd114(LR)		Mo98(MR)		Sn118(LR)	
				Conc.		Conc.		Conc.		Conc.		Conc.		Conc.	
Sample-number	Sample name	Date	Sample ID	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %
8	Treated in TM's system	03.06.2019	Steen-1-35-8	8,37	0,3	6,295	1,5	0,010	25,8	17,976	1,2	0,000	9,6	0,009	19,8
1	Treated in TM's system	24.05.2019	Repeatingtest-176	11,39	1,2	12,333	1,0	0,013	26,3	32,840	1,1	0,031	48,1	0,009	8,3
4	Treated in TM's system	28.05.2019	Steen-1-35-4	11,08	1,6	6,414	0,9	0,005	26,3	27,867	0,3	0,019	72,7	0,006	43,9
9	Treated in TM's system	03.06.2019	Steen-1-35-9	8,71	1,8	6,176	1,4	0,006	21,6	18,408	2,1	0,021	52,9	0,003	26,8
13	Tunnel wall	03.06.2019	Steen-1-35-13-fersk	16,35	1,3	68,637	0,6	0,548	10,4	7,795	0,9	11,323	1,7	0,009	9,1
16	Colorful wall	06.06.2019	Steen-1-35-16-fersk	2,04	1,7	0,614	0,5	0,436	6,3	0,521	2,1	1,006	8,8	0,008	72,2
17	First collection	06.06.2019	Steen-1-35-17-fersk	2,90	1,9	0,445	4,3	0,026	3,0	0,759	4,1	0,550	7,7	0,004	33,5
18	Second collection	06.06.2019	Steen-1-35-18-fersk	2,15	0,9	0,358	5,1	0,071	10,8	1,540	1,5	0,167	8,9	0,005	31,3
19	Dripping from ceiling	06.06.2019	Steen-1-35-19-fersk	2,98	1,8	0,109	4,5	0,031	22,6	0,657	1,4	0,995	10,6	0,006	30,8
20	Collected ceiling water (at threshold)	06.06.2019	Steen-1-35-20-fersk	1,05	3,7	0,472	2,3	0,095	5,3	2,169	1,0	0,121	11,2	0,003	41,7
21	Before rinse	06.06.2019	Steen-1-35-21	10,74	2,3	14,306	1,7	0,080	1,8	28,363	1,6	0,032	121,4	0,007	37,8
22	After rinse	06.06.2019	Steen-1-35-22	10,81	1,6	14,648	3,7	0,084	12,8	28,387	1,6	0,049	55,1	0,012	21,6
23	Olivine	06.06.2019	Steen-1-35-23	7,25	0,5	0,072	5,9	0,011	13,4	0,053	6,3	3,544	11,0	0,008	22,6
24	Olivine	06.06.2019	Steen-1-35-24	10,55	1,4	0,023	3,4	0,006	18,0	0,031	17,7	1,698	8,4	0,005	36,2
25	Olivine	06.06.2019	Steen-1-35-25	11,00	1,3	0,019	12,1	0,008	12,9	0,017	27,2	1,959	5,3	0,009	20,1
26	Olivine	06.06.2019	Steen-1-35-26	11,34	0,5	0,015	13,9	0,006	17,1	0,016	7,2	1,483	9,8	0,007	15,4
28	Lime	08.06.2019	Steen-1-35-28	10,74	2,1	3,405	1,6	0,008	7,6	16,516	1,5	0,157	20,0	0,005	29,2
30	Lime	08.06.2019	Steen-1-35-30	11,60	1,0	8,471	1,0	0,010	14,5	23,197	0,4	0,034	14,6	0,007	29,6
27	Lime over time	08.06.2019	Steen-1-35-27	11,49	2,4	10,742	0,5	0,011	27,9	24,930	0,6	0,039	84,3	0,024	20,3
29	Lime over time	08.06.2019	Steen-1-35-29	11,08	1,8	11,702	1,3	0,017	11,1	24,792	1,5	0,012	97,5	0,009	44,5
31	Lime over time	08.06.2019	Steen-1-35-31	11,31	1,2	12,190	0,9	0,014	16,8	25,117	0,4	0,045	63,8	0,008	8,9
32	Lime over time	09.06.2019	Steen-1-35-32	10,47	2,0	10,631	0,2	0,018	19,2	23,598	0,4	0,038	25,0	0,009	20,0

Sample-number	La139(LR)		Ce140(LR)		Pr141(LR)		Nd146(LR)		Eu153(LR)		Gd160(LR)		Ho165(LR)		Er166(LR)		W182(LR)		Hg202(LR)	
	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %
8	12,389	0,2	21,264	0,9	2,254	0,5	8,849	1,6	0,179	3,9	1,32	4,3	0,198	2,8	0,608	3,2	0,005	9,1	0,093	10,9
1	25,514	1,3	44,623	0,7	4,531	1,2	17,649	1,5	0,330	4,1	2,57	5,0	0,374	2,6	1,129	0,7	0,006	34,7	0,088	5,1
4	14,410	0,6	20,376	0,2	1,896	1,4	7,225	2,0	0,129	1,1	1,05	2,4	0,153	4,5	0,436	3,4	0,001	28,6	0,064	6,7
9	12,838	2,3	20,859	0,4	2,142	0,7	8,220	0,9	0,158	5,6	1,18	0,6	0,170	6,0	0,527	4,0	0,003	25,9	0,083	4,8
13	87,220	1,3	179,735	2,4	19,765	1,1	89,818	0,8	2,129	1,7	14,18	1,3	2,482	1,2	7,959	2,2	0,028	13,9	0,062	17,7
16	0,954	1,5	1,255	1,3	0,210	5,0	0,953	4,4	0,019	6,2	0,06	9,1	0,019	5,4	0,061	1,3	0,173	2,7	0,118	5,6
17	0,769	5,0	1,120	2,1	0,146	8,2	0,631	6,6	0,019	5,0	0,00	5,3	0,015	9,8	0,043	10,6	0,023	9,1	0,048	3,3
18	0,713	1,8	1,055	3,3	0,146	1,1	0,620	7,4	0,017	12,4	0,00	7,1	0,012	9,3	0,034	9,0	0,066	7,1	0,071	5,3
19	0,079	4,1	0,043	8,0	0,017	10,9	0,057	3,8	0,007	17,6	-0,07	13,1	0,003	25,2	0,009	18,4	0,014	28,7	0,071	11,0
20	0,534	0,7	0,529	3,1	0,108	2,2	0,491	2,1	0,015	11,2	0,00	3,1	0,012	10,6	0,047	14,4	0,055	5,1	0,071	4,2
21	27,939	0,7	54,228	0,8	5,824	0,9	24,353	1,0	0,503	1,2	3,52	2,4	0,503	2,2	1,572	3,5	0,008	17,0	0,048	4,2
22	28,513	0,2	54,619	1,0	5,745	5,1	23,952	4,0	0,502	3,7	3,41	5,7	0,491	5,9	1,541	6,2	0,007	0,9	0,042	15,5
23	0,005	7,3	0,003	21,1	0,001	89,3	0,004	52,3	0,029	4,0	-0,06	17,2	0,000	128,9	0,000	94,4	0,953	2,6	0,027	4,9
24	0,003	52,4	0,001	54,3	0,000	87,7	0,001	89,2	0,014	24,9	-0,06	87,5	0,000	173,2	0,001	173,2	0,450	6,0	0,059	6,4
25	0,003	29,1	0,001	37,7	0,000	173,2	0,003	50,0	0,015	15,7	-0,06	54,7	0,000	89,2	0,000	0,0	0,472	4,6	0,039	11,6
26	0,003	31,1	0,004	40,0	0,001	69,4	0,001	89,2	0,012	13,1	-0,06	36,2	0,000	79,9	0,000	86,6	0,451	5,4	0,058	5,9
28	7,986	1,3	9,727	1,0	0,849	5,1	3,162	3,1	0,053	5,8	0,38	4,7	0,070	6,2	0,207	2,5	0,004	28,5	0,184	5,4
30	18,498	1,4	30,201	0,9	2,988	0,5	11,460	1,2	0,208	5,1	1,55	1,7	0,227	3,2	0,712	3,3	0,006	30,7	0,079	9,6
27	21,847	1,7	38,624	1,2	4,002	0,5	16,091	0,9	0,318	0,6	2,26	1,7	0,322	2,8	0,926	3,4	0,006	26,7	0,051	16,2
29	23,439	2,0	43,036	1,0	4,595	1,0	18,517	1,3	0,372	0,3	2,69	3,6	0,381	1,8	1,166	2,9	0,009	25,3	0,053	8,5
31	23,959	1,3	44,709	0,8	4,782	0,0	19,744	1,4	0,395	0,9	2,71	1,5	0,381	2,4	1,191	2,1	0,005	27,3	0,073	0,5
32	20,912	0,2	38,907	0,6	4,174	1,2	17,087	0,4	0,354	2,1	2,49	3,8	0,358	2,3	1,089	1,9	0,006	51,6	0,101	13,9

Sample-number	Tl205(LR)		Pb208(LR)		U238(LR)		Dy161(LR)		Na23(MR)		Mg25(MR)		Al27(MR)		Si29(MR)		P31(MR)		S34(MR)	
	Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.	
	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %
8	0,684	5,4	5,126	1,2	0,465	3,3	1,000	2,2	54 104	1,4	9 658	1,3	730,0	1,6	9 678	0,8	1,98	19,3	166 070	0,3
1	0,661	1,1	0,485	1,7	0,170	0,9	1,886	2,3	50 661	3,2	13 015	2,3	322,9	2,1	12 373	2,1	2,19	13,6	193 183	2,3
4	0,616	1,8	0,148	5,3	0,091	3,4	0,731	4,7	54 455	2,0	12 662	3,8	33,3	3,0	10 913	1,1	1,10	17,7	207 644	1,0
9	0,779	3,4	0,785	2,6	0,138	0,1	0,846	6,8	57 437	1,4	9 788	0,7	69,6	1,0	9 549	1,4	1,23	15,1	168 463	2,7
13	0,047	1,9	2,583	1,1	3,361	2,0	12,515	1,3	16 771	2,5	20 004	3,7	24 050,5	3,8	32 263	1,6	360,23	1,8	627 136	3,7
16	0,049	5,1	2,169	3,4	0,900	2,6	0,087	8,6	83 330	1,7	5 262	2,8	50,4	2,7	12 044	3,3	15,28	9,8	29 008	2,6
17	0,031	15,7	0,024	9,6	0,188	2,7	0,068	16,1	14 204	1,9	3 233	1,4	80,1	1,9	3 962	1,1	1,26	28,3	28 959	0,8
18	0,144	11,5	1,471	1,5	0,129	2,4	0,068	5,1	28 486	3,9	2 214	1,7	51,0	4,5	4 212	2,8	1,23	15,6	33 278	2,3
19	0,055	12,3	0,029	8,7	1,769	2,4	0,010	15,8	53 901	2,2	8 956	2,5	4,5	7,5	5 451	2,1	4,87	4,4	58 962	2,8
20	0,072	10,1	0,451	2,5	0,084	3,7	0,057	10,3	25 225	1,1	2 393	1,0	44,5	2,2	3 871	0,6	1,39	16,9	19 803	1,5
21	0,802	2,3	22,862	1,5	1,316	0,3	2,729	1,7	57 070	2,9	12 456	4,4	4 338,0	1,6	11 132	3,2	2,14	14,4	199 396	3,8
22	0,780	4,2	22,806	2,6	1,318	3,2	2,716	3,4	56 009	2,4	12 419	3,5	4 311,4	3,4	11 012	3,6	1,65	3,6	197 742	3,3
23	0,045	16,4	0,321	1,7	0,005	28,1	0,000	173,2	156 243	2,2	13	1,2	126,2	3,0	1 035	0,9	6,55	8,8	41 051	1,1
24	0,110	2,4	0,202	4,0	0,003	20,7	0,000	0,0	66 110	2,2	116	4,2	169,0	2,1	2 354	2,2	2,32	16,0	129 943	2,5
25	0,127	0,7	0,210	1,5	0,002	37,7	0,001	86,6	69 856	1,3	159	1,5	176,5	1,2	2 796	1,4	3,03	19,4	160 284	1,0
26	0,170	4,0	0,168	0,8	0,002	19,2	0,001	86,6	63 940	1,7	549	2,4	159,9	0,2	3 691	1,5	2,66	12,5	199 871	0,5
28	0,791	4,0	0,209	6,0	0,939	1,8	0,344	9,4	62 759	1,9	12 682	1,3	20,5	0,7	11 731	1,4	1,36	23,7	216 337	0,5
30	0,907	0,7	0,351	2,3	0,260	2,0	1,159	2,9	63 717	0,3	12 789	0,5	86,2	1,9	12 365	0,5	1,59	10,6	220 617	1,2
27	0,924	1,6	0,969	2,7	0,489	2,3	1,693	2,5	62 690	0,9	12 755	2,6	233,0	0,7	12 439	0,8	2,13	14,2	221 489	2,1
29	0,928	0,1	4,388	1,8	0,571	1,5	2,026	4,1	62 657	1,6	12 467	1,3	926,6	1,9	12 491	2,0	1,87	39,2	216 751	0,8
31	0,930	1,5	5,026	0,8	0,639	1,9	2,104	2,4	64 107	0,9	12 681	1,9	1 066,0	1,5	12 554	1,5	2,17	2,6	218 694	0,6
32	0,914	2,2	9,152	0,3	0,700	2,6	1,918	2,6	62 834	2,1	11 485	1,6	1 415,8	0,9	11 964	1,1	2,35	9,0	205 832	1,1

	Cl35(MR)		K39(MR)		Ca44(MR)		Ti49(MR)		V51(MR)		Cr53(MR)		Mn55(MR)		Fe56(MR)		Fe57(MR)		Co59(MR)	
Sample-number	Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.	
	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %
8	69 777	2,0	5 683	2,8	157 974	1,2	0,04	29,7	0,011	54,8	0,11	20,8	362,64	1,9	3 812,7	2,9	3 712,7	2,3	58,5	1,2
1	66 221	2,6	5 453	3,2	189 302	2,0	0,08	55,6	0,009	53,9	0,15	71,7	634,83	2,1	4 854,7	1,8	4 745,8	2,5	75,0	1,8
4	70 728	0,8	5 969	1,4	213 759	1,9	0,08	33,6	0,006	64,0	0,02	30,5	622,57	0,9	991,5	0,5	1 082,3	1,3	85,8	2,8
9	73 633	2,0	5 749	0,2	167 656	2,3	0,09	43,3	0,016	49,5	0,06	33,5	340,46	2,8	4 035,3	1,8	3 933,5	2,5	58,9	5,1
13	8 887	1,6	654	3,5	180 308	4,0	443,62	2,9	14,289	4,0	31,29	3,1	904,18	2,4	373 901,9	3,8	410 880,6	2,2	319,162	3,1
16	112 880	2,5	4 025	4,2	53 029	3,5	2,38	1,7	0,128	20,8	0,25	4,6	17,81	2,2	201,6	1,5	225,4	4,8	2,114	11,3
17	18 458	1,6	2 123	4,3	35 929	1,0	0,14	39,3	0,119	33,5	0,45	5,3	66,74	0,5	15,7	0,5	17,7	7,1	7,339	1,9
18	37 168	1,2	2 655	0,3	41 218	4,1	0,32	15,2	0,065	31,6	0,40	18,8	16,67	1,7	309,4	2,9	346,3	1,6	2,043	5,4
19	64 485	1,6	6 345	1,2	83 426	2,8	0,04	53,9	0,098	26,5	0,22	9,3	40,19	2,7	13,8	3,1	15,1	11,9	0,099	18,9
20	35 409	1,7	1 494	3,8	30 696	3,3	0,36	24,2	0,045	30,8	0,21	8,5	8,20	5,9	104,7	2,8	116,8	1,0	0,872	5,5
21	72 837	2,8	5 535	0,9	173 166	3,9	0,57	27,4	0,009	53,9	3,08	4,6	521,44	2,4	10 360,3	2,1	10 610,2	2,5	79,3	2,6
22	72 131	3,5	5 585	1,9	169 015	3,2	0,47	22,4	0,003	82,9	3,29	3,7	524,31	3,0	9 723,5	1,7	9 918,7	4,3	80,7	4,7
23	39 288	1,3	39 041	1,8	386 139	1,6	0,13	66,8	1,543	3,7	9,57	5,6	1,38	1,6	210,9	2,6	245,0	5,9	3,5	7,1
24	62 928	1,6	10 880	2,3	334 722	2,8	0,07	68,4	2,665	4,2	5,65	2,9	0,94	2,3	12,4	1,6	15,1	12,2	0,91	3,4
25	75 134	1,5	10 770	2,5	354 136	1,9	0,57	167,5	3,284	4,3	6,71	3,1	0,86	3,7	18,3	1,6	19,4	10,5	0,99	5,1
26	76 501	2,1	8 540	2,7	331 724	1,9	0,08	44,9	3,694	2,6	5,00	7,8	1,29	3,3	10,3	2,1	13,5	13,5	0,58	5,8
28	81 914	2,2	6 114	2,0	224 458	0,5	0,07	11,4	0,006	28,9	0,00	41,8	513,72	0,2	4 531,5	1,8	4 356,0	0,3	78,1	2,3
30	83 359	1,3	6 331	1,6	217 581	2,5	0,08	33,2	0,012	27,0	0,02	43,9	533,62	0,3	7 188,4	0,6	7 096,8	1,3	85,4	1,4
27	82 446	2,6	6 447	3,2	210 164	1,6	0,09	66,2	0,018	19,9	0,12	26,4	531,08	2,5	8 343,9	1,1	8 282,1	1,2	85,6	2,0
29	82 284	0,9	6 172	4,4	200 819	1,2	0,14	73,6	0,012	59,6	0,21	23,4	514,41	0,6	9 115,5	0,9	9 082,3	2,7	85,2	1,2
31	83 519	0,7	6 279	3,0	201 301	1,8	0,09	28,9	0,004	33,3	0,11	28,8	525,36	1,9	9 243,6	0,7	9 300,2	1,7	86,7	2,2
32	82 633	1,0	6 218	3,0	186 508	1,2	0,17	76,2	0,015	8,7	0,38	8,0	466,57	0,6	8 611,6	1,0	8 581,3	1,5	79,9	1,1

Sample-number	Ni60(MR)		Cu63(MR)		Zn66(MR)		Rb85(MR)		Sr88(MR)		Ba137(MR)		As75(HR)		Se78(HR)		Br81(HR)	
	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %
8	24,02	4,0	6 210,02	0,3	5 632,6	1,9	6,29	3,6	391,5	1,7	12,54	4,1	0,78	12,6	<1.0	47,4	54	11,7
1	38,55	4,2	9 708,87	1,4	9 621,2	2,2	6,95	3,8	441,7	2,6	13,83	1,3	0,66	8,6	<1.0	69,7	59	6,5
4	40,81	2,4	5 363,09	0,9	8 862,3	0,5	7,69	5,3	478,0	3,0	13,54	2,9	0,24	8,9	<1.0	66,2	61	2,6
9	22,59	2,7	5 587,76	2,1	5 435,4	2,4	6,36	1,0	403,5	1,8	13,38	5,7	0,52	21,7	<1.0	37,0	43	3,1
13	44,11	3,8	23 756,57	3,2	2 078,8	1,8	2,96	6,7	265,1	2,9	0,36	30,9	25,21	9,4	<1.0	7,9	12	8,9
16	2,59	5,7	121,99	3,1	131,8	1,7	3,48	4,7	133,0	0,9	16,66	4,0	0,62	52,3	<1.0	52,2	62	15,3
17	6,55	4,2	44,53	1,9	214,8	1,0	3,54	0,7	110,6	2,6	7,69	6,1	0,25	88,0	<1.0	139,3	14	10,0
18	3,24	2,4	101,55	2,3	558,4	0,9	3,18	1,5	142,8	4,2	6,07	2,2	0,30	33,3	<1.0	46,5	35	7,8
19	1,97	8,2	9,71	3,4	284,0	2,9	3,50	1,3	297,2	0,3	21,94	9,4	0,41	31,7	<1.0	58,5	59	4,9
20	2,35	15,6	43,76	1,6	671,1	1,2	2,14	5,7	90,1	1,1	11,81	4,7	0,16	35,5	<1.0	31,9	33	8,2
21	36,89	6,6	11 699,28	2,0	8 119,4	2,9	6,74	3,6	432,2	3,7	12,90	7,0	0,62	11,8	<1.0	20,1	48	1,3
22	37,52	4,6	11 659,42	4,3	8 072,5	3,2	6,73	3,1	424,2	3,6	13,33	1,4	0,76	23,9	<1.0	30,6	48	8,1
23	0,17	9,8	362,20	2,9	58,8	12,1	12,77	0,7	10 230,3	1,8	304,49	2,3	0,14	30,1	<1.0	32,7	38	4,7
24	0,09	32,7	235,58	1,2	155,5	3,5	6,86	1,9	3 030,7	2,3	138,63	1,2	0,20	10,5	<1.0	48,4	47	10,2
25	0,06	93,0	195,45	1,2	115,1	5,4	7,58	1,4	2 888,8	2,1	131,78	0,3	0,23	9,0	<1.0	29,6	53	7,9
26	0,03	0,0	115,12	1,6	81,9	6,7	8,38	2,6	1 827,5	0,6	98,81	0,5	0,19	43,5	<1.0	38,1	53	13,2
28	32,00	2,3	4 001,07	2,1	6 917,7	0,6	7,11	1,1	503,2	1,0	18,45	3,4	0,64	23,7	<1.0	16,1	50	12,7
30	34,03	3,2	9 387,31	0,2	7 528,0	1,0	7,49	3,5	499,0	1,1	15,27	4,6	0,64	39,8	<1.0	20,8	57	8,0
27	34,74	1,8	10 661,24	0,5	7 717,8	0,8	7,43	2,9	491,8	1,3	15,33	4,6	0,73	49,4	<1.0	14,5	51	6,3
29	34,66	3,3	12 559,57	0,9	7 492,3	0,7	7,28	3,7	474,9	2,9	14,86	5,0	0,75	7,5	<1.0	64,5	51	12,3
31	33,75	3,5	12 880,44	0,8	7 550,6	1,7	7,43	0,9	480,8	0,5	14,81	2,1	0,78	25,4	<1.0	43,0	59	9,3
32	31,66	1,4	13 802,05	0,7	7 122,0	0,7	7,06	3,1	458,1	2,7	14,49	2,1	0,77	11,9	<1.0	31,8	54	5,0

Table C.1.4 The concentrations of all of the different elements analyzed for in all of the samples in batch 36-81 with corresponding RSD. The description of the samples is only given on the first page. When the table continues to the next pages only the sample number is given.

				Li7(LR)		Y89(LR)		Cd114(LR)		Mo98(MR)		Sn118(LR)		La139(LR)		Ce140(LR)	
				Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD	Conc.	RSD
Sample-number	Sample name	Date	Sample ID	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%	[µg/L]	%
36	Tunnel water	12.09.2019	Repeatingtest-133	10,06	1,5	11,721	2,0	34,992	2,6	0,039	20,3	0,014	26,9	23,474	1,6	46,823	3,1
45	Tunnel water	19.09.2019	Steine-Steen-36-81-45-f	7,46	0,3	4,506	1,8	31,543	0,3	0,008	11,2	0,004	21,5	9,387	3,5	16,399	2,5
51	Tunnel water	02.10.2019	Steine-Steen-36-81-51-f	5,32	0,3	4,726	1,5	17,034	1,1	0,006	74,7	0,002	9,2	9,699	2,8	17,740	3,0
74	Tunnel water	11.10.2018	Steine-Steen-36-81-74-usikker	15,13	1,4	20,555	1,5	55,765	0,8	0,035	78,0	0,044	20,1	42,873	2,0	84,919	1,6
53	Treated in TM's system	02.10.2019	Steine-Steen-36-81-53-f	8,25	0,7	9,021	3,9	23,013	1,7	0,002	83,2	0,004	17,0	19,314	1,5	36,529	2,0
46	Tunnel wall	19.09.2019	Steine-Steen-36-81-46-f	16,50	3,6	57,880	4,2	8,815	3,7	11,798	3,1	0,006	45,9	83,578	5,2	173,287	3,1
47	Left tunnel wall	19.09.2019	Steine-Steen-36-81-47-f	4,35	1,6	8,099	1,8	8,673	0,6	0,050	37,2	0,002	32,0	17,225	1,1	31,835	2,2
54	Tunnel wass	02.10.2019	Steine-Steen-36-81-54-f	12,42	1,5	43,282	2,2	14,058	1,5	6,605	1,7	0,004	15,5	69,467	1,7	140,437	1,8
36	Before rinse	12.09.2019	Repeatingtest-133	10,06	1,5	11,721	2,0	34,992	2,6	0,039	20,3	0,014	26,9	23,474	1,6	46,823	3,1
37	After rinse	12.09.2019	Steine-Steen-36-81-37-f	10,09	2,9	12,102	2,2	33,850	0,6	0,033	22,4	0,014	10,6	23,130	0,9	47,357	1,9
38	After rinse	12.09.2019	Steine-Steen-36-81-38-f	10,02	1,3	11,735	1,2	32,931	1,4	0,034	28,6	0,016	13,9	23,051	0,7	45,213	1,4
39	Olivin (pH exp.)	12.09.2019	Repeatingtest-134	8,82	2,1	0,034	6,8	0,015	14,0	1,756	5,9	0,009	9,5	0,003	23,1	0,003	14,3
40	Lime (pH exp.)	12.09.2019	Steine-Steen-36-81-40	10,77	2,1	2,326	2,5	29,438	2,1	0,090	16,2	0,014	28,9	6,177	4,0	6,203	3,2
41	Olivin (pH exp.)	12.09.2019	Steine-Steen-36-81-41	9,33	1,6	0,029	8,3	0,022	8,8	1,852	10,1	0,016	15,1	0,009	8,6	0,015	6,7
42	Lime (pH exp.)	12.09.2019	Steine-Steen-36-81-42	11,39	2,6	2,680	1,3	33,316	0,7	0,040	32,8	0,011	20,7	6,867	2,5	7,133	3,6

Sample-number	Pr141(LR)		Nd146(LR)		Eu151(LR)		Eu153(LR)		Gd157(LR)		Dy163(LR)		Ho165(LR)		Er166(LR)		W182(LR)		Hg202(LR)		Tl205(LR)	
	Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.	
	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %
36	4,864	1,5	20,235	1,8	0,491	5,2	0,457	3,8	2,932	0,6	2,210	2,2	0,427	3,1	1,297	2,4	0,004	19,200	0,015	9,7	0,823	0,1
45	1,760	0,7	6,995	1,7	0,150	3,6	0,147	1,8	1,003	4,6	0,751	0,9	0,150	3,3	0,452	2,2	0,001	40,100	0,009	4,7	0,493	4,0
51	2,004	3,5	7,960	0,9	0,165	5,6	0,166	7,2	1,133	0,5	0,856	1,7	0,166	3,5	0,497	1,0	0,003	29,300	0,009	5,9	0,384	1,2
74	8,759	1,4	36,489	2,4	0,745	4,9	0,725	0,7	4,930	1,7	3,743	2,0	0,733	1,8	2,219	1,3	0,005	39,800	0,020	11,4	0,826	2,3
53	3,590	2,8	14,569	1,2	0,306	1,7	0,296	1,4	2,083	1,9	1,541	4,4	0,305	3,6	0,931	3,1	0,003	24,700	0,013	7,9	0,669	3,9
46	19,295	3,6	82,896	1,7	1,937	3,6	1,808	2,2	12,110	3,5	10,547	3,5	2,117	1,9	6,718	1,2	0,012	29,700	0,011	3,3	0,065	3,9
47	3,334	2,2	13,745	3,3	0,248	3,6	0,249	5,1	1,991	3,6	1,528	2,1	0,294	0,8	0,877	2,5	0,000	10,900	0,001	9,9	0,098	7,2
54	15,053	1,1	64,315	1,1	1,414	5,0	1,364	2,9	9,641	1,7	7,938	1,2	1,601	1,9	5,088	0,3	0,007	24,000	0,014	0,5	0,119	7,8
36	4,864	1,5	20,235	1,8	0,491	5,2	0,457	3,8	2,932	0,6	2,210	2,2	0,427	3,1	1,297	2,4	0,004	19,200	0,015	9,7	0,823	0,1
37	4,838	0,7	20,135	2,0	0,476	0,6	0,458	2,6	2,929	2,2	2,225	3,1	0,429	1,5	1,317	3,9	0,002	60,000	0,012	10,1	0,800	1,5
38	4,748	2,9	19,247	2,2	0,449	2,6	0,430	2,6	2,817	2,2	2,198	0,9	0,427	3,3	1,284	1,3	0,003	33,400	0,016	15,4	0,773	3,3
39	0,001	66,9	0,001	86,6	0,003	44,0	0,006	23,0	0,001	61,9	0,000	93,6	0,000	173,2	0,000	173,2	0,524	2,900	0,011	8,6	0,130	4,8
40	0,489	2,5	1,638	5,4	0,028	8,3	0,025	2,6	0,240	5,5	0,163	5,0	0,037	3,2	0,121	5,2	0,004	21,600	0,013	14,4	0,809	2,3
41	0,002	11,4	0,007	26,2	0,003	11,7	0,005	26,1	0,002	90,7	0,001	45,6	0,000	25,0	0,001	54,7	0,568	3,500	0,024	11,3	0,123	1,6
42	0,561	2,1	1,783	4,2	0,031	5,0	0,029	5,5	0,261	3,0	0,199	4,3	0,046	10,9	0,147	7,0	0,003	44,800	0,015	8,9	0,834	3,2

Sample-number	Pb208(LR)		U238(LR)		Na23(MR)		Mg25(MR)		Al27(MR)		Si29(MR)		P31(MR)		S34(MR)		Cl35(MR)		K39(MR)	
	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %
36	33,187	1,0	1,587	2,2	46 283	1,1	11 945	2,2	5 124,3	1,9	10 393	0,4	11,49	2,1	216 377	2,2	41 466	2,9	6 665	3,3
45	3,467	1,5	0,490	2,3	36 624	1,0	10 406	1,2	789,0	2,3	7 938	0,9	2,39	2,4	147 100	1,0	32 778	1,7	5 686	2,4
51	7,535	0,4	0,561	2,0	24 859	0,7	6 520	0,6	1 281,5	2,0	6 356	1,7	2,06	10,1	101 465	0,8	20 404	2,2	4 015	1,7
74	32,899	1,3	1,932	1,0	60 149	2,2	15 434	1,6	6 769,8	1,7	12 797	1,5	27,77	4,4	259 673	2,2	62 299	3,2	7 343	3,2
53	2,734	0,6	0,543	2,5	36 120	1,2	9 412	1,8	615,1	2,0	9 645	0,6	2,33	3,8	173 412	2,5	27 062	2,0	6 059	2,7
46	3,563	2,4	3,022	2,6	12 465	1,5	15 785	1,6	17 815,2	1,2	29 555	0,7	221,46	1,4	580 149	3,1	6 031	3,1	1 036	6,2
47	0,235	2,4	0,471	4,9	9 954	2,0	4 896	1,8	2 050,6	2,0	9 104	2,0	2,14	4,4	73 005	1,8	6 468	2,8	2 319	4,3
54	2,341	1,1	2,425	2,1	10 837	2,6	14 220	2,1	14 029,6	1,9	25 297	0,6	145,69	1,7	429 409	4,7	4 819	3,6	1 493	3,1
36	33,187	1,0	1,587	2,2	46 283	1,1	11 945	2,2	5 124,3	1,9	10 393	0,4	11,49	2,1	216 377	2,2	41 466	2,9	6 665	3,3
37	31,063	1,8	1,605	1,2	46 982	1,1	12 080	1,6	5 311,9	1,4	10 463	1,1	12,83	3,7	218 611	0,9	40 893	3,2	6 569	2,4
38	28,899	0,9	1,564	1,7	46 676	2,2	12 157	1,7	5 318,6	1,1	10 659	1,6	14,33	2,9	222 251	0,6	40 446	1,1	6 440	3,8
39	0,177	8,4	0,003	11,0	58 997	0,6	202	2,1	208,4	0,8	3 347	0,8	6,65	4,5	180 770	1,9	43 633	4,8	11 988	0,3
40	0,076	1,5	0,172	3,0	47 423	2,5	12 811	2,2	11,4	2,2	10 052	1,6	2,89	4,5	218 059	0,0	40 979	1,5	6 711	1,6
41	0,110	2,7	0,004	12,3	50 577	2,0	211	0,8	139,7	0,7	3 323	1,6	4,82	3,7	194 475	2,4	43 143	3,4	11 062	3,0
42	0,077	6,2	0,166	2,2	49 602	3,3	13 068	2,2	18,5	1,4	10 599	2,0	2,31	5,4	228 276	3,9	43 375	2,2	7 098	2,9

Sample-number	Ca44(MR)		Ti49(MR)		V51(MR)		Cr53(MR)		Mn55(MR)		Fe56(MR)		Co59(MR)		Ni60(MR)		Cu63(MR)		Zn66(MR)	
	Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.		Conc.	
	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %
36	133 222	1,5	5,849	3,7	0,064	22,1	6,22	1,2	525,83	2,2	29 131	3,5	69,814	4,6	33,22	5,3	18 074,2	2,2	9 496,8	0,5
45	117 441	1,2	0,085	21,4	0,020	20,0	0,16	8,0	221,04	1,1	4 512	1,2	32,116	1,5	19,49	1,5	5 410,6	1,3	8 645,6	1,3
51	79 812	2,1	0,278	102,8	0,028	16,5	0,37	9,2	171,78	1,3	2 678	3,1	26,505	1,4	13,60	4,6	4 755,5	0,2	4 642,8	0,8
74	151 964	3,2	7,166	3,4	0,073	15,4	10,70	1,5	611,33	1,4	37 934,4	3,4	94,901	0,6	41,93	1,4	16 950,2	2,7	15 277,5	1,9
53	143 094	2,8	-0,006	73,0	0,007	48,1	0,08	18,0	329,90	0,8	5 639	0,4	55,374	1,9	23,54	2,7	9 528,9	0,6	6 697,3	1,5
46	176 891	1,0	355,200	0,6	13,171	0,5	27,02	2,9	777,60	1,6	296 863	0,7	246,403	1,9	39,01	2,9	19 638,3	0,9	2 087,4	2,2
47	43 044	1,1	3,077	1,5	0,012	37,6	0,96	3,6	158,57	2,4	3 880	2,2	21,855	1,0	5,89	4,6	1 726,5	2,6	2 333,3	1,7
54	149 905	3,9	230,615	0,4	8,536	2,2	21,15	1,9	672,90	4,0	192 205	4,0	179,059	2,0	33,89	4,3	14 403,1	4,6	3 505,4	1,5
36	133 222	1,5	5,849	3,7	0,064	22,1	6,22	1,2	525,83	2,2	29 131	3,5	69,814	4,6	33,22	5,3	18 074,2	2,2	9 496,8	0,5
37	135 495	1,2	5,610	2,4	0,063	2,2	6,33	2,4	623,44	1,3	27 867	1,3	74,915	4,1	33,88	4,2	18 602,3	0,6	9 681,5	2,8
38	138 327	4,6	5,628	2,8	0,064	27,0	6,24	1,5	695,86	1,8	27 391	1,7	78,106	4,7	35,24	3,6	18 007,1	0,9	9 693,3	0,6
39	266 011	0,2	0,016	34,7	3,502	4,1	8,35	2,9	0,81	2,2	20,6	0,6	0,767	1,8	0,08	22,7	159,8	1,4	68,6	2,6
40	206 854	0,7	0,051	40,4	0,010	30,0	0,03	7,3	530,06	1,8	49,8	2,9	72,261	3,3	38,62	3,5	4 188,1	3,3	9 134,9	1,6
41	272 692	2,6	0,011	86,7	3,765	2,6	9,80	1,5	0,84	2,0	23,3	2,4	0,777	8,5	0,06	6,3	101,7	2,4	42,8	1,9
42	219 389	2,6	0,030	48,4	0,003	35,3	0,02	14,1	546,34	2,2	17,3	1,2	73,373	1,1	33,20	1,1	4 737,5	3,8	9 810,4	1,4

	Rb85(MR)		Sr88(MR)		Ba137(MR)		As75(HR)		Br81(HR)	
Sample-number	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %	Conc. µg/L	RSD, %
36	7,85	1,0	469,8	0,4	13,66	2,9	10,272	7,2	60,1	12,0
45	6,06	3,0	371,0	0,4	17,45	4,7	0,623	15,8	49,2	5,9
51	4,07	3,3	250,5	2,2	13,05	0,9	0,510	7,9	34,7	3,1
74	8,15	3,5	538,4	0,3	16,53	2,4	3,097	9,9	64,3	7,1
53	6,06	3,5	400,0	3,4	13,04	3,4	0,858	8,7	35,4	6,5
46	3,83	4,7	270,8	0,6	0,51	10,8	26,421	7,8	18,0	7,2
47	4,92	1,5	115,9	1,4	2,36	11,0	0,206	21,1	12,1	6,2
54	4,07	3,5	277,9	1,5	1,57	7,3	12,851	10,1	11,0	11,5
36	7,85	1,0	469,8	0,4	13,66	2,9	10,272	7,2	60,1	12,0
37	7,73	0,8	462,2	1,5	24,22	1,5	9,758	8,2	57,9	9,0
38	8,25	2,4	472,8	1,1	34,84	2,0	9,133	9,4	59,8	8,9
39	8,22	2,9	2 385,4	3,2	88,50	0,5	0,590	1,7	52,5	10,3
40	7,57	4,1	522,6	0,6	15,74	2,2	1,286	5,1	48,1	5,2
41	8,63	2,0	1 955,7	1,6	89,15	1,2	0,457	4,9	56,6	7,4
42	8,20	2,1	554,0	0,4	16,74	1,5	1,110	8,4	50,3	5,2

Table C.1.5 The concentrations of all of the different elements analyzed for in all of the samples in batch 82-162 with corresponding RSD. The description of the samples is only given on the first page. When the table continues to the next pages only the sample number is given.

			7 -> 7 Li [H2]		23 -> 23 Na [O2]		24 -> 24 Mg [H2]		24 -> 24 Mg [O2]		27 -> 27 Al [H2]	
Sample-number	Sample name	Date	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD
85	Tunnel water	18.10.2019	12,06	12,6	41871	0,6	13217	2,8	12954	1,5	256	0,1
89	Tunnel water	12.11.2019	10,95	21,5	64997	1,5	12220	2,7	12027	4,0	13	4,4
95	Tunnel water	18.12.2019	14,24	11,0	100794	0,9	14804	1,9	14844	1,0	2508	2,3
157	Before aeration	24.02.2020	5,24	18,0	56466	0,2	7859	0,2	8115	4,3	2438	0,9
86	Treated in TM's system	18.10.2019	14,32	3,4	48498	0,9	14044	2,6	13971	0,9	1124	0,3
88	Treated in TM's system	12.11.2019	13,04	29,4	105984	1,8	12580	0,5	13015	1,8	8	7,9
91	Colorful wall	18.12.2019	0,65	37,0	117948	0,3	7248	1,3	7372	2,0	27	3,8
92	First collection	18.12.2019	2,63	8,4	10007	1,4	2333	1,6	2457	3,2	337	0,9
93	Second collection	18.12.2019	3,40	30,6	40155	1,6	3074	2,9	3242	4,1	31	1,9
94	Threshold	18.12.2019	7,73	23,1	70348	1,2	9222	2,8	9399	3,5	6	12,4
95	Before rinse	18.12.2019	14,24	11,0	100794	0,9	14804	1,9	14844	1,0	2508	2,3
97	After rinse	18.12.2019	11,02	21,6	98060	1,3	13709	2,0	14374	1,7	2195	0,5
99	Olivine + activated carbon	18.12.2019	15,13	21,3	110905	0,4	194	1,1	200	1,1	157	2,1
101	Olivine + activated carbon	18.12.2019	16,16	10,6	110330	1,9	330	1,3	344	1,3	120	1,7
102	Olivine + activated carbon	18.12.2019	14,12	5,3	108789	2,2	3574	3,9	3666	2,8	37	4,5
157	Before aeration	24.02.2020	5,24	18,0	56466	0,2	7859	0,2	8115	4,3	2438	0,9
159	aeration	24.02.2020	5,31	22,1	55363	0,8	7947	3,2	8034	3,1	2377	0,9
160	aeration	24.02.2020	6,83	19,2	52453	2,2	7627	0,5	7562	0,9	2394	0,1
161	aeration	24.02.2020	5,91	10,5	56537	1,1	8117	0,8	8311	1,8	2412	1,1
162	aeration	24.02.2020	7,91	13,8	54290	0,3	7972	0,1	7822	1,5	2481	1,2

	29 -> 45 Si [O2]		31 -> 47 P [O2]		32 -> 48 S [O2]		35 -> 51 Cl [O2]		39 -> 39 K [H2]		39 -> 39 K [O2]		40 -> 40 Ca [H2]		44 -> 60 Ca [O2]	
	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD
85	9902	7,8	2,18	18,5	163857	1,3	27913	7,6	7095	1,6	7264	0,7	164286	1,5	167777	4,7
89	7912	6,5	1,61	25,1	149776	1,1	21179	5,1	6711	1,8	7102	1,5	135742	1,5	136760	3,5
95	10708	8,3	2,54	16,6	192936	1,1	52235	3,1	5867	1,7	6566	0,8	143216	0,8	154534	6,0
157	9186	9,7	4,03	21,5	107865	0,6	67221	4,6	5029	1,3	5554	1,0	95111	1,5	95630	7,0
86	11633	7,5	2,94	7,1	201885	1,2	32331	5,0	7647	1,7	8241	1,8	198137	0,1	215346	6,8
88	9108	8,3	3,02	19,7	195051	1,9	29997	7,2	7813	1,2	8470	2,3	155560	2,6	164486	7,9
91	9936	8,6	21,42	8,3	17012	1,6	172157	1,7	4375	3,8	4774	0,5	50464	1,6	48202	6,9
92	2947	9,2	4,50	4,9	13447	1,0	10287	10,6	1300	1,3	1359	2,4	21353	1,4	19542	6,5
93	4523	8,8	3,29	10,9	39743	2,3	60785	2,6	3566	2,5	3999	1,7	58534	1,3	56748	6,4
94	6055	6,5	1,11	21,4	112403	1,7	46938	4,2	5609	3,2	6062	1,5	106686	2,4	100449	5,7
95	10708	8,3	2,54	16,6	192936	1,1	52235	3,1	5867	1,7	6566	0,8	143216	0,8	154534	6,0
97	10179	9,3	2,60	36,6	189424	0,8	50710	3,3	5747	0,3	6581	1,3	136426	1,0	147580	6,8
99	3190	8,2	7,25	10,9	141044	0,5	48238	4,0	12138	1,3	12785	0,6	291127	1,7	307132	6,9
101	3248	9,0	4,57	18,9	146641	3,1	50338	4,3	10869	1,5	11944	1,5	278024	0,7	295057	7,2
102	4557	8,0	2,98	23,9	197991	1,7	52079	6,7	8079	1,3	8847	1,8	250082	2,6	267382	6,8
157	9186	9,7	4,03	21,5	107865	0,6	67221	4,6	5029	1,3	5554	1,0	95111	1,5	95630	7,0
159	9169	8,0	2,72	13,4	108329	1,2	68491	2,3	5018	1,5	5435	1,7	94389	1,4	96516	7,1
160	8739	8,9	12,88	13,1	101573	0,2	62676	2,3	4776	2,7	5331	2,2	91501	1,5	91487	7,4
161	9250	6,5	3,75	9,1	107808	2,1	68771	1,6	5020	2,4	5539	2,2	97727	1,3	96456	5,8
162	8998	9,3	9,90	12,1	107554	2,4	67476	2,5	5110	1,7	5493	1,1	98893	1,8	95093	7,0

	47 -> 63 Ti [O2]		51 -> 67 V [O2]		52 -> 52 Cr [H2]		52 -> 52 Cr [O2]		55 -> 55 Mn [O2]		56 -> 56 Fe [H2]		59 -> 59 Co [O2]		60 -> 60 Ni [H2]	
	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD
85	0,141	24,2	0,037	43,3	0,422	19,9	0,171	2,7	376	2,3	1596	2,1	53,9	1,8	30,16	8,5
89	0,047	72,2	0,024	40,4	0,291	16,5	0,170	50,1	296	1,8	92,4	1,1	41,5	1,3	22,39	10,8
95	0,074	23,4	0,030	38,3	0,492	3,5	0,301	20,8	747	0,9	5698,7	1,9	117,4	0,5	52,83	2,2
157	1,895	1,8	0,045	16,2	2,600	5,5	2,702	1,8	276	0,3	9534,8	1,7	42,6	1,4	17,66	12,4
86	0,105	19,6	0,032	43,1	0,420	23,2	0,264	17,7	528	2,3	2411	2,0	81,0	0,3	34,73	1,2
88	0,128	35,9	0,055	23,7	0,325	9,7	0,167	25,1	458	1,6	5,7	3,4	70,0	1,5	26,56	5,3
91	1,334	16,0	0,165	22,0	0,753	13,6	0,548	12,1	7	0,9	90,1	2,1	0,9	12,1	1,77	25,6
92	0,805	6,1	0,170	30,8	0,844	2,6	0,727	9,3	44	1,5	97,0	0,4	4,7	8,3	4,52	11,4
93	0,248	29,6	0,053	17,1	0,596	1,3	0,349	17,7	15	0,8	456,1	1,0	1,6	3,8	4,14	10,3
94	0,102	4,7	0,027	43,4	0,210	12,5	0,127	35,7	165	0,3	108,7	0,8	19,2	0,2	13,67	3,1
95	0,074	23,4	0,030	38,3	0,492	3,5	0,301	20,8	747	0,9	5698,7	1,9	117,4	0,5	52,83	2,2
97	0,084	39,7	0,040	50,7	0,560	3,5	0,314	9,5	732	1,4	5378,8	2,9	108,6	1,5	47,88	4,8
99	0,078	50,0	7,617	3,4	9,826	1,4	9,592	1,8	2	4,5	16,3	2,6	0,9	3,0	0,51	25,9
101	0,096	57,0	7,204	2,9	9,639	4,5	9,931	0,0	1	6,1	9,3	1,6	0,6	16,9	0,40	7,3
102	0,074	17,0	4,967	1,9	3,943	6,4	3,904	2,9	1	13,7	6,3	1,2	0,5	4,0	0,39	31,6
157	1,895	1,8	0,045	16,2	2,600	5,5	2,702	1,8	276	0,3	9534,8	1,7	42,6	1,4	17,66	12,4
159	1,844	6,1	0,050	19,0	2,729	0,8	2,742	6,8	269	2,0	9368,1	1,6	42,2	2,4	16,57	3,6
160	5,045	3,4	0,134	6,6	3,323	5,2	3,258	1,7	255	1,6	12283,0	1,7	40,0	0,9	17,36	4,0
161	1,891	6,8	0,042	44,6	2,635	4,8	2,707	3,4	275	0,8	9432,9	2,2	42,1	1,0	17,51	10,4
162	4,493	3,2	0,157	15,6	3,170	4,3	3,241	8,0	268	1,0	13036,4	2,0	41,3	1,1	18,35	7,2

	60 -> 60 Ni [O2]		62 -> 62 Ni [H2]		62 -> 62 Ni [O2]		63 -> 63 Cu [O2]		66 -> 66 Zn [H2]		75 -> 91 As [O2]		79 -> 79 Br [O2]		79 -> 95 Br [O2]	
	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD
85	29,08	0,5	32,25	8,3	27,51	8,7	6339	2,1	8822	1,6	0,335	4,8	46,7	21,0	63,7	12,6
89	23,09	1,9	21,86	5,3	22,23	6,9	2029	1,6	7450	0,7	0,247	32,5	43,9	11,5	47,5	14,1
95	53,22	2,1	52,06	6,4	51,56	2,1	20492	0,9	13188	1,6	0,258	23,6	46,3	11,1	53,4	13,7
157	18,22	2,8	14,53	19,1	17,81	6,4	7064	0,8	3228	1,8	0,692	8,2	51,2	9,2	57,8	24,2
86	36,47	3,2	35,88	4,8	34,85	3,3	10211	1,5	7479	1,2	0,737	16,7	57,3	22,5	62,0	12,5
88	29,62	4,9	27,99	11,5	28,87	2,4	1360	2,5	5962	1,1	0,423	9,9	45,7	3,7	53,3	12,4
91	1,78	19,7	1,83	43,9	1,87	13,5	97	1,0	41	4,1	0,434	13,9	161,8	9,6	158,2	3,2
92	4,85	6,5	6,43	14,6	4,66	6,3	78	1,9	113	2,0	0,245	37,0	38,3	33,5	40,5	7,3
93	4,30	5,0	4,09	20,2	4,02	13,1	87	1,3	521	1,9	0,364	21,1	49,2	14,6	56,7	9,0
94	13,57	4,5	10,78	13,1	12,97	5,0	1234	3,2	4596	1,2	0,159	8,5	47,8	19,5	48,9	12,2
95	53,22	2,1	52,06	6,4	51,56	2,1	20492	0,9	13188	1,6	0,258	23,6	46,3	11,1	53,4	13,7
97	50,70	0,3	47,76	2,8	49,66	5,3	18658	0,7	12428	4,3	0,178	40,2	52,6	29,5	48,3	15,6
99	0,50	8,5	-0,01	226,3	0,29	6,1	165	0,7	140	0,9	2,027	9,3	42,0	33,1	44,0	12,8
101	0,14	12,0	0,47	48,9	0,21	29,7	153	1,3	159	4,1	1,722	4,7	44,5	19,3	50,9	30,9
102	0,16	5,9	-0,51	N/A	0,33	58,8	45	1,4	78	4,3	0,576	15,0	46,6	24,0	45,2	11,2
157	18,22	2,8	14,53	19,1	17,81	6,4	7064	0,8	3228	1,8	0,692	8,2	51,2	9,2	57,8	24,2
159	17,40	4,4	14,81	23,7	18,25	4,3	6908	2,3	3198	2,3	0,784	22,0	54,2	8,4	59,7	7,5
160	17,31	2,9	15,91	15,6	16,51	7,5	6556	2,2	3093	0,9	3,237	2,2	42,0	15,4	55,1	15,9
161	18,34	5,4	17,60	5,0	18,37	3,0	7082	2,0	3249	1,9	0,820	3,5	48,7	19,7	49,8	23,0
162	17,02	1,9	17,76	8,5	17,22	4,1	6868	2,3	3285	1,0	3,259	4,7	50,8	16,1	55,3	6,7

	85 -> 85 Rb [H2]		85 -> 85 Rb [O2]		88 -> 88 Sr [H2]		88 -> 88 Sr [O2]		98 -> 98 Mo [H2]		98 -> 130 Mo [O2]		114 -> 114 Cd [H2]		114 -> 114 Cd [O2]	
	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD
85	8,50	3,3	7,99	1,8	488,57	1,0	472	2,7	0,034	14,8	0,045	56,75	31,88	0,88	31,17	0,9
89	7,92	3,2	7,60	1,7	445,45	0,8	444	0,7	0,058	41,3	0,080	35,58	28,51	0,28	28,24	2,1
95	7,68	2,2	7,29	3,5	460,67	2,6	467	2,4	0,023	76,6	0,022	67,15	49,99	0,98	50,75	1,6
157	5,58	1,1	5,75	1,9	304,69	1,8	332	0,3	0,022	145,7	0,002	237,12	12,07	2,89	12,21	2,1
86	9,08	0,4	8,78	1,8	549,26	1,6	541	2,3	0,020	24,0	0,015	136,87	27,28	0,94	26,84	1,0
88	8,63	3,7	8,40	1,0	510,19	1,0	511	1,3	0,002	916,6	0,022	62,48	22,54	1,43	22,54	2,7
91	3,93	1,0	3,73	0,2	138,94	1,8	141	1,1	0,937	17,0	0,899	14,30	0,19	12,09	0,20	10,9
92	2,13	2,4	1,94	7,3	63,75	0,2	63	0,1	0,296	16,7	0,337	14,43	0,43	4,16	0,45	12,0
93	4,84	1,1	4,50	4,3	220,87	1,9	231	1,5	0,183	12,4	0,186	10,01	1,76	2,41	1,79	3,2
94	6,47	3,3	6,14	1,5	348,53	3,0	358	1,1	0,071	43,2	0,063	19,27	18,35	1,34	17,94	1,8
95	7,68	2,2	7,29	3,5	460,67	2,6	467	2,4	0,023	76,6	0,022	67,15	49,99	0,98	50,75	1,6
97	7,34	0,5	7,27	3,6	443,14	1,4	470	1,8	0,001	1061,1	0,011	44,28	47,60	1,35	47,79	1,0
99	8,78	1,6	8,21	1,3	2824,28	1,8	2866	0,7	3,098	8,9	3,282	7,58	0,06	10,44	0,05	20,3
101	8,56	1,5	8,41	2,8	2543,21	1,4	2586	0,3	2,914	5,4	2,934	5,69	0,04	34,46	0,04	48,2
102	9,43	3,1	9,32	3,3	1342,04	3,0	1363	2,9	1,083	3,5	1,181	2,47	0,04	16,02	0,04	12,0
157	5,58	1,1	5,75	1,9	304,69	1,8	332	0,3	0,022	145,7	0,002	237,12	12,07	2,89	12,21	2,1
159	5,60	2,6	5,70	2,9	304,92	1,1	316	2,5	0,025	67,1	-0,001	N/A	11,83	1,07	12,23	1,6
160	5,43	2,3	5,35	1,6	296,01	1,2	311	1,4	0,078	32,9	0,117	3,25	11,60	0,58	11,70	1,3
161	5,62	0,9	5,84	3,1	316,51	4,4	335	3,3	0,018	125,2	0,015	15,04	12,11	2,62	12,49	1,7
162	5,83	4,2	5,64	4,3	314,30	3,4	318	2,1	0,135	26,6	0,082	17,14	12,37	0,40	12,24	1,3

	118 -> 118 Sn [H2]		118 -> 118 Sn [O2]		137 -> 137 Ba [O2]		139 -> 155 La [O2]		140 -> 156 Ce [O2]		146 -> 162 Nd [O2]		151 -> 151 Eu [O2]		151 -> 167 Eu [O2]	
	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD
85	0,076	14,2	0,105	21,9	19,24	1,7	15,407	1,9	28,97	0,8	11,601	1,1	0,262	26,4	0,279	29,3
89	0,060	45,9	0,043	23,2	18,69	3,3	6,089	1,4	8,30	0,7	2,599	9,1	0,039	35,0	0,029	57,7
95	0,126	3,4	0,165	26,5	15,73	3,3	42,645	2,0	79,40	0,1	31,250	0,9	0,717	2,0	0,771	8,5
157	0,048	37,1	0,048	18,4	12,38	4,6	12,840	1,5	25,15	0,4	11,015	5,9	0,229	6,1	0,242	15,9
86	0,032	35,0	0,065	33,2	16,44	4,7	24,452	1,6	47,71	0,3	20,332	1,6	0,444	11,3	0,457	19,9
88	0,065	33,2	0,070	27,3	15,29	1,1	6,126	2,2	6,22	2,2	1,548	5,7	0,020	21,6	0,029	28,5
91	0,047	14,6	0,050	23,3	19,35	3,7	0,737	5,6	0,77	4,5	0,807	8,4	0,020	34,8	0,011	50,2
92	0,030	49,1	0,041	37,8	5,10	12,3	1,319	3,6	2,30	4,2	1,294	3,7	0,043	32,1	0,038	51,1
93	0,041	52,9	0,042	19,0	9,58	3,4	0,420	4,8	0,65	5,5	0,336	17,6	0,009	34,5	0,002	173,2
94	0,036	23,7	0,045	44,5	15,76	1,6	2,324	2,5	2,64	1,7	0,748	9,2	0,014	86,6	0,007	114,7
95	0,126	3,4	0,165	26,5	15,73	3,3	42,645	2,0	79,40	0,1	31,250	0,9	0,717	2,0	0,771	8,5
97	0,036	20,1	0,051	15,9	16,70	1,7	38,352	1,6	70,46	0,6	27,243	2,8	0,645	6,7	0,630	7,4
99	0,038	75,5	0,050	53,3	130,01	1,6	0,008	7,5	0,02	24,3	0,018	75,2	0,011	16,4	0,009	91,8
101	0,038	53,3	0,041	16,5	121,12	1,1	0,005	66,5	0,01	21,6	0,004	86,6	0,008	115,3	0,006	173,2
102	0,034	47,4	0,065	11,5	85,93	1,2	0,007	59,9	0,01	118,4	0,000	N/A	0,007	89,3	0,004	173,2
157	0,048	37,1	0,048	18,4	12,38	4,6	12,840	1,5	25,15	0,4	11,015	5,9	0,229	6,1	0,242	15,9
159	0,159	7,0	0,135	13,2	12,33	3,5	12,803	1,8	25,37	0,7	11,310	2,4	0,255	7,5	0,223	21,0
160	0,867	5,0	0,918	3,5	11,98	1,8	12,262	1,2	24,18	1,6	10,314	1,0	0,252	19,1	0,221	29,2
161	0,199	29,4	0,228	34,9	13,22	5,8	13,022	0,5	25,75	2,0	10,987	3,0	0,263	7,1	0,279	19,8
162	0,730	10,9	0,670	11,3	12,91	5,1	12,782	0,5	25,08	2,3	10,994	1,1	0,256	15,4	0,211	22,2

	153 -> 153 Eu [O2]		153 -> 169 Eu [O2]		157 -> 173 Gd [O2]		163 -> 179 Dy [O2]		165 -> 181 Ho [O2]		166 -> 182 Er [O2]		182 -> 182 W [H2]		208 -> 208 Pb [O2]	
	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD
85	0,236	8,6	0,243	23,7	1,720	10,0	1,387	2,6	0,242	9,4	0,727	5,1		163,3	1,56	4,1
89	0,043	20,0	0,040	33,4	0,359	8,8	0,288	9,2	0,048	8,2	0,164	15,7	0,002	2,2	0,13	20,8
95	0,692	7,1	0,751	20,4	4,828	1,4	3,939	0,8	0,761	6,4	2,153	2,2		79,1	3,35	1,8
157	0,260	7,3	0,277	12,9	1,769	1,7	1,345	4,0	0,236	11,6	0,743	3,6	0,000	N/A	21,57	0,5
86	0,429	6,3	0,509	12,8	3,120	5,2	2,455	7,8	0,437	4,6	1,304	3,2	0,006	45,0	4,89	2,1
88	0,024	56,0	0,018	41,3	0,214	6,7	0,119	28,6	0,030	4,0	0,077	2,8	0,013	28,1	0,14	16,3
91	0,019	22,9	0,018	57,2	0,113	31,7	0,059	24,9	0,011	23,2	0,044	14,4	0,145	3,7	1,24	3,0
92	0,030	26,2	0,043	26,6	0,174	24,9	0,177	33,8	0,036	20,2	0,093	10,0	0,020	48,3	0,15	6,5
93	0,011	65,8	0,005	99,6	0,053	24,7	0,047	5,1	0,011	41,1	0,020	32,4	0,025	36,5	0,87	3,1
94	0,016	43,8	0,022	54,8	0,130	15,4	0,086	11,4	0,021	9,1	0,068	8,8	0,003	170,0	0,11	45,4
95	0,692	7,1	0,751	20,4	4,828	1,4	3,939	0,8	0,761	6,4	2,153	2,2	0,016	79,1	3,35	1,8
97	0,652	9,9	0,661	12,9	4,502	2,7	3,414	5,8	0,671	4,6	1,923	10,2	0,016	79,2	3,22	5,8
99	0,020	60,9	0,010	100,2	0,000	N/A	-0,001	N/A	0,000	N/A	0,000	N/A	0,680	23,9	0,08	19,3
101	0,018	33,9	0,007	173,2	0,000	N/A	-0,001	N/A	0,000	N/A	0,000	N/A	0,703	4,8	0,06	24,2
102	0,009	88,1	0,005	99,2	0,002	173,2	0,004	203,3	0,001	173,2	0,000	N/A	0,447	8,8	0,06	20,7
157	0,260	7,3	0,277	12,9	1,769	1,7	1,345	4,0	0,236	11,6	0,743	3,6	0,000	N/A	21,57	0,5
159	0,227	17,1	0,249	10,9	1,658	9,3	1,323	6,1	0,226	8,0	0,709	9,2	0,008	58,7	21,04	3,9
160	0,211	8,1	0,219	13,3	1,641	7,2	1,157	11,8	0,228	4,9	0,725	6,0	0,009	69,4	21,54	3,2
161	0,231	15,8	0,274	10,6	1,635	3,5	1,289	8,2	0,248	11,2	0,764	10,5	0,004	187,8	21,52	0,1
162	0,244	9,7	0,289	15,1	1,654	9,9	1,312	6,4	0,236	3,0	0,755	8,5	0,013	69,0	21,77	2,4

Table C.1.6 Concentrations of the elements found in their blanks used to correct for batch 1-61 and 1-35, and their average. The elements that are not presented here were not found in the blanks.

				P31(MR)		K39(MR)		Ca44(MR)		Fe56(MR)		Zn66(MR)	
				Conc.		Conc.		Conc.		Conc.		Conc.	
Sample-number	Sample name	Date	Sample ID	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %	µg/L	RSD, %
55	Blank 1	22.03.2019	Steen-1-61-55-blank	1,59	11,3	2	12,6	9	3,4	0,9	7,2	0,1	16,7
56	Blank 2	22.03.2019	Steen-1-61-56-blank	2,15	1,5	4	7,3	8	9,1	0,0	10,4	0,2	31,5
	Average used for correction			2,15	1,5	3,71	7,3	7,67	9,1	0,03	10,4	0,2	31,5

Table C.1.7 Concentrations of the elements found in their blanks used to correct for batch 36-81 and 82-162, and their average. The elements that are not presented here were not found in the blanks.

			23 -> 23 Na [O2]		24 -> 24 Mg [H2]		24 -> 24 Mg [O2]		27 -> 27 Al [H2]		31 -> 47 P [O2]		32 -> 48 S [O2]	
Sample-number	Sample name	Date	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD
152	Blank 1	04.02.2020	2	12,3	0	22,3	0	2,9	0,13	23,3	0,63	22,9	4	20,8
153	Blank 2	04.02.2020	1	7,4	1	1,8	1	2,8	1,00	3,3	0,94	19,7	3	23,1
154	Blank 3	04.02.2020	8	2,1	1	21,9	1	11,6	0,30	15,7	1,73	8,9	4	6,6
155	Blank 4	04.02.2020	31	0,9	2	2,0	2	2,0	0,46	12,7	1,28	16,6	3	8,2
156	Blank 5	04.02.2020	2	5,6	0	8,2	0	5,2	0,41	2,4	1,07	6,9	2	12,6
	Average blanks		2	5,6	0,2	8,2	0,23	5,2	0,41	2,4	1,07	6,9	2	12,6

39 -> 39 K [H2]		39 -> 39 K [O2]		40 -> 40 Ca [H2]		44 -> 60 Ca [O2]		47 -> 63 Ti [O2]		52 -> 52 Cr [H2]		52 -> 52 Cr [O2]		56 -> 56 Fe [H2]	
Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD	Conc. [ug/l]	Conc. RSD
2	4,3	3	1,4	3	2,2	3	2,2	0,027	27,6	0,016	26,4	0,020	1,3	0,10	7,6
3	2,9	3	3,4	5	0,7	5	13,0	0,041	24,2	0,021	9,4	0,015	21,9	0,68	7,8
12	1,7	12	1,9	8	5,1	7	7,5	0,043	29,8	0,022	11,7	0,012	13,4	0,28	2,0
7	2,8	7	0,5	7	0,6	7	6,7	0,026	22,3	0,018	17,9	0,019	13,0	0,18	5,4
6	2,8	6	2,3	4	0,7	4	15,9	0,032	15,1	0,022	22,8	0,016	48,8	0,13	4,6
6	2,8	6	2,3	4	0,7	4	15,9	0,032	15,1	0,022	22,8	0,016	48,8	0,1	4,6

C.2 Detection limits

The detection limits and limits for quantification of different elements in the ICP-MS analysis are presented in table C.2.1.

Table C.2.1 Detection limits and quantification limit for different elements in the analysis.

Sign	Isotope	Element	Resolution	LODs-25% [µg/l]	QL-25% [µg/l]
Al	27	Aluminium	Mr	0,04	0,20
Sb	121	Antimony	Mr	0,0004	0,0020
As	75	Arsenic	Hr	0,005	0,025
Ba	137	Barium	Mr	0,0026	0,013
Be	9	Beryllium	Lr	0,0004	0,0020
Be	9	Beryllium	Mr	0,0016	0,0080
Bi	209	Bismuth	Lr	0,0002	0,0010
B	11	Boron	Lr	0,010	0,050
B	11	Boron	Mr	0,016	0,080
Br	81	Brom	Hr	0,6	3,0
Cd	111/114	Cadmium	Lr	0,0004	0,0020
Cd	111/114	Cadmium	Mr	0,0020	0,0100
Ca	44	Calcium	Mr	0,4000	2,0
Ce	140	Cerium	Lr	0,0000	0,0002
Cs	133	Cesium	Lr	0,0	0,0005
Cl	35	Chlorine	Mr	20,0000	100
Cr	53	Chromium	Mr	0,0040	0,0200
Co	59	Cobalt	Mr	0	0,0040
Cu	63/65	Cupper	Mr	0,0060	0,030
Dy	163	Dysprosium	Lr	0,0002	0,0008
Er	166	Erbium	Lr	0,000	0,0003
Eu	153	Europium	Lr	0,0002	0,0008
Gd	155	Gadolinium	Mr	0,0040	0,020
Ga	69	Gallium	Mr	0,0014	0,0070
Ge	72	Germanium	Hr	0,004	0,020
Au	197	Gold	Lr	0,0000	0,0002
Hf	178	Hafnium	Lr	0,000	0,0010
Ho	165	Hollium	Lr	0,0000	0,0002
In	115	Indium	Lr	0,0001	0,0005
I	127	Iodide	Mr	0,2000	1,0
Fe	56	Iron	Mr	0,004	0,020
Ir	193	Iridium	Lr	0,0001	0,0005
La	139	Lantan	Lr	0,0001	0,0005
La	139	Lantan	Mr	0,0004	0,0020
Pb	208	Lead	Lr	0,000	0,0020
Li	7	Lithium	Lr	0,0010	0,005
Li	7	Lithium	Mr	0,01	0,030
Lu	175	Lutetium	Lr	0,0000	0,0002
Mg	24	Magnesium	Mr	0,0200	0,10
Mg	25	Magnesium	Mr	0,100	0,50
Mn	55	Manganese	Mr	0,0012	0,0060
Hg	202	Mercury	Lr	0,0004	0,0020
Mo	98	Molybdenum	Mr	0,004	0,020
Nd	146	Neodymium	Lr	0,00	0,0002

Ni	60	Nikkel-60	Mr	0,0030	0,015
Nb	93	Niob	Lr	0,0	0,001
Nb	93	Niob	Hr	0,0050	0,025
Pd	104	Palladium	Hr	0,01	0,050
P	31	Phosphor	Mr	0,0800	0,40
Pt	195	Platinum	Lr	0,0002	0,0010
K	39	Potassium	Mr	0,2000	1,0
Pr	141	Praseodymium	Lr	0,0001	0,0003
Re	185	Rhenium	Hr	0,0080	0,04
Rh	103	Rhodium	Lr	0,000	0,0005
Rh	103	Rhodium	Hr	0,00	0,0100
Rb	85	Rubidium	Mr	0,0	0,012
Ru	85	Ruthenium	Lr	0,000	0,0010
Sm	147	Samarium	Lr	0,000	0,0005
Sc	45	Scandium	Mr	0,001	0,0040
Se	82	Selenium	Lr	0,0	0,05
Se	78	Selenium	Hr	0	0,150
Si	29	Silisium	Mr	2,0000	10,0
Ag	109	Silver	Mr	0,004	0,020
Na	23	Sodium	Mr	2,0000	10,0
Sr	88	Strontium	Mr	0,0050	0,025
S	32	Sulphur	Mr	1,0000	5,0
S	34	Sulphur	Mr	4,0000	20
Ta	181	Tantalum	Lr	0,0000	0,0002
Te	125	Tellur	Mr	0,0200	0,100
Tb	159	Terbium	Lr	0,0000	0,0002
Tl	205	Thallium	Lr	0,000	0,0003
Th	232	Thorium	Lr	0,0001	0,0005
Tm	169	Thulium	Lr	0,0001	0,0005
Sn	118	Tin	Lr	0,0002	0,001
Sn	118	Tin	Mr	0,0020	0,010
Ti	47	Titanium	Mr	0,0040	0,020
U	238	Uranium	Lr	0,000	0,0003
V	51	Vanadium	Mr	0,0006	0,0030
W	182	Wolfram	Lr	0,000	0,0010
Yb	172	Ytterbium	Lr	0,00008	0,0004
Yb	89	Yttrium	Lr	0,00008	0,0004
Zn	66	Zink-66	Mr	0,005	0,025
Zn	67	Zink-67	Mr	0,008	0,040
Zr	90	Zirkonium	Lr	0,0001	0,0005
Zr	90	Zirkonium	Hr	0,005	0,025

Appendix D – Statistics

To investigate if the gravel added to the experiments caused a statistically significant difference, concentrations before and after rinsing the gravel were run through statistical tests. A Shapiro-Wilk test was used to check for normality, and a T-test was used when normality was found and a Mann Whitney U test was used when it was not. The tests were performed for each element separately. The concentrations before and after the gravel was rinsed are presented in table D.1.

Table D.1 Concentrations of the different elements found before and after gravel was rinsed. The unit is µg/L.

	Cd	Pb	Cr	Fe	Ni	Cu	Zn	As
Before rinse	28.4	22.9	3.08	10 400	36.9	11 700	8 120	0.624
Before rinse	35.0	33.2	6.22	29 100	33.2	18 100	9 500	10.3
Before rinse	50.0	3.35	0.492	5 700	53.2	20 500	13 200	0.258
After rinse	28.4	22.8	3.29	9 720	37.5	11 700	8 070	0.761
After rinse	33.9	31.1	6.33	27 900	33.9	18 600	9 680	9.76
After rinse	32.9	28.9	6.24	27 400	35.2	18 000	9 690	9.13
After rinse	47.6	3.22	0.560	5 380	50.7	18 700	12 400	0.178
After rinse	48.2	3.07	0.519	5 330	49.9	18 300	12 800	0.264

The resulting p-values for the different elements from the Shapiro-Wilk test are presented in table D.2. The concentrations can be assumed normally distributed when the p is > 0.05.

Tabel D.2 Resulting p-values from Shapiro-Wilk tests for each element.

	Shapiro-W (p)
Cd	0.0783117
Pb	0.0469031
Cr	0.0464153
Fe	0.0171153
Ni	0.0439246
Cu	0.0140252
Zn	0.133531
As	0.00484681

The concentrations of the elements with p-values above 0.05 were tested for statistical significance before and after gravel rinse in a T-test. The resulting p-values for the different elements from the T-tests are presented in table D.3. The concentrations are not statistically different when the p is > 0.05.

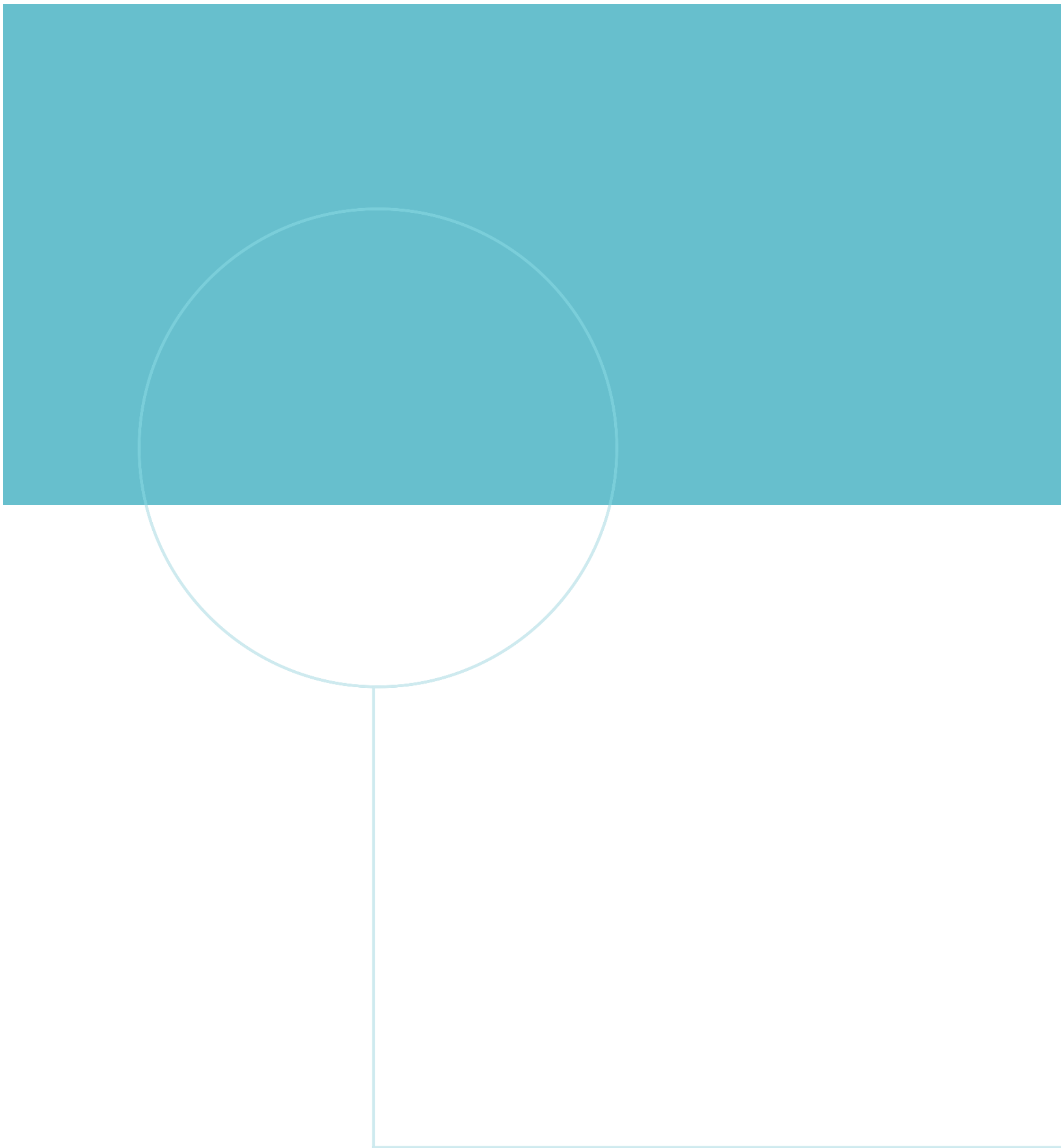
Tabel D.3 Resulting p-values from T-tests Cd and Zn.

	T-test (p)
Cd	0.477586
Zn	0.438405

The elements with p-values < 0.05 in the Shapiro-Wilk tests were tested for statistical significance before and after gravel rinse in a Mann Whitney U test. The resulting P-values for the different elements from the Mann Whitney U tests are presented in table D.4. The concentrations are not statistically different when the P is > 0.05.

Table D.4 Resulting p-values from Mann Whitney U tests for each element.

	Mann Whitney U (P)
Pb	0.571429
Cr	0.571429
Fe	0.571429
Ni	1.000000
Cu	1.000000
As	1.000000



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