June Furunes-Olsen

# Removal of metals and particles from tunnel wash water

A study on the effects of chemical coagulation and precipitation as treatment option

Master's thesis in Civil- and Environmental Engineering Supervisor: Thomas Meyn July 2023

NTNU Norwegian University of Science and Technology Faculty of Engineering Department of Civil and Environmental Engineering

Master's thesis



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# 1 Preface

This master's thesis has been carried out by June Furunes-Olsen during the spring 2023. The thesis constitutes the end of the five-year study programme Civil- and Environmental Engineering at the Norwegian University of Science and Technology (NTNU), with specialization in water- and wastewater engineering. The thesis has been part of the project TreatRW in collaboration with Aquateam COWI and Nye Veier. Thomas Meyn has been the main supervisor for the thesis work.

First, I would like to thank Thomas Meyn, Hanne Vistnes and Nadine Sossalla for all exciting, educational and constructive discussions, inputs and questions during the semester. I am thankful to not only have been included in their project group, but first and foremost for getting to be a part of their team during the past six months. I am grateful to Thomas Meyn and Nadine Sossalla for help with collecting tunnel wash water in Oslo, Hønefoss and Trondheim. Further, I thank Trine Hårberg Ness, Marina Fernandez-Delgado Juarez and once again Hanne Vistnes for guidance, help and patience with my work in the laboratory. I thank Anica Simic and Kyyas Seyitmuhammedov for help with all practicalities related to the ICP-MS analysis, even when the equipment was not on our side. Further, I thank Espen Hoell in Nye Veier and Kai Gundersen in Statens Vegvesen for taking the time to answer my many questions about sludge handling.

Lastly, I would like to greatly thank my family and friends, not only for moral support, but also for patience when all conversations with me during the last months eventually has derailed onto road tunnels and metal contents in wash waters. Especially, I deeply and genuinely thank my mother Laila, my father Karl Petter and my room-mates Vebjørn and Lukas for motivating me, supporting me and believing in me, even when I didn't do so myself. Last but not least I would like to thank my friend Erika who has been going through the ups and downs of writing a thesis along with me. This thesis would not have been possible without either one of you.

# 2 Abstract

Due to challenging topography and sparsely distributed population, tunnels are often proven to be a favorable choice for transport infrastructure in Norway. For traffic safety among other reasons, the tunnels require regular cleaning. Pollutants such as metals, polycyclic aromatic hydrocarbons (PAHs) and salt (NaCl) during wintertime accumulate over time, resulting in severely elevated pollutant concentrations relative to what can be found in regular highway runoff. Many older Norwegian tunnels are completely without any solution for treatment of the water used for cleaning, in turn meaning the polluted water is directly discharged into nature. All newer tunnels are built with sedimentation basins as primary treatment for the wash water. Sedimentation alone can remove a large proportion of the particle bound pollutants, but this process takes time and tunnel wash water also contains a significant share of dissolved pollutants which are not removed through sedimentation alone.

In this study, chemical coagulation and precipitation are both proven to be potentially effective treatment technologies for tunnel wash water. Both treatments involves introducing an external chemical to the wash water, but also opens the door to significantly reducing the required treatment time. Experiments with the iron-based coagulant PIX-318 and alum-based PAX-18 as well as with Metalsorb PCZ and sodium sulfide as precipitants were carried out. The studied raw water was sampled from washes conducted at the Vålerenga tunnel in Oslo and the Grilstad tunnel in Trondheim. The results show low residual turbidities, when the samples were injected with coagulant doses of 5 mmol Me/L the turbidities reached a threshold of <5 NTU. Experiments conducted with adjusted pH imply that higher pH values result in a more efficient turbidity reduction at lower coagulant doses, whereas higher coagulant doses also provide water with a low residual turbidity at lower pH values.

The performance of PIX-318 and PAX-18 in terms of reducing turbidity was found to be rather similar, however a lower coagulant dose was generally required to reach a certain boundary (e.g. 1 NTU) when using PIX-318. For both coagulants a significant reduction occurs during the first 60 minutes of sedimentation, whereas a further increase in sedimentation time post coagulation has little effect on the residual turbidity. After 120 minutes of sedimentation, both coagulants can be said to perform close to equal in terms of turbidity.

Considering metal removal PAX-18 proved to be somewhat more effective, especially in regards of reducing the dissolved fraction. For metals such as zinc which usually is mainly associated with the low molecular mass fraction and thus also is assumed to have an increased mobility and bioavailability, an average reduction of approximately 85% was achieved with PAX-18 as 5 mmol Al/L as coagulant. In comparison, when PIX-318 was injected as 5 mmol Fe/L, the resulting reduction of zinc in the sample was measured to an average of approximately 75%. Similarly for nickel, PAX-18 was perceivably more efficient in reducing the concentrations. When considering cupper, chromium and manganese, the performance of the coagulants was rather similar. For both coagulants, the addition of the coagulant itself resulted in a significant increase in Fe- and Al- contents respectively. After 60 and 120 minutes of sedimentation, the concentrations were however once again reduced to a level deemed accepatable in comparison to existing recommendations for discharge.

# 3 Sammendrag

På grunn av utfordrende topografi og spredt befolkning er tunneler ofte en gunstig løsning for transportinfrastruktur i Norge. Blant annet av hensyn til trafikksikkerhet, krever tunnelene jevnlig rengjøring. Forurensninger som metaller, polyaromatiske hydrokarboner (PAH) og eventuelt salter (NaCl) om vinteren akkumuleres i tunnelene over tid og vaskes for eksempel ikke bort ved nedbørshendelser, noe som resulterer i forhøyede forurensningskonsentrasjoner relativt til det som finnes i vanlig avrenning fra veier. Mange eldre norske tunneler har ingen løsning for behandling av vaskevannet, noe som betyr at det forurensede vannet slippes direkte ut i naturen. Alle nyere tunneler bygges med sedimenteringsbassenger som primær behandlingsløsning for vaskevannet. Sedimentering alene kan med god effekt fjerne partikkelforbundne forurensninger, men denne prosessen er tidkrevende. Tunnelvaskevannet inneholder også en betydelig del oppløste forurensninger, som heller ikke tilstrekkelig lar seg fjerne ved sedimentering alene.

I denne masteroppgaven har kjemisk koagulering og utfelling blitt vist å være potensielt gunstige behandlingsløsninger for tunnelvaskevann. Løsningene innebærer å introdusere kjemikalier i vaskevannet, men muliggjør også en betydelig redusert behandlingstid. I studien har det blitt gjennomført forsøk med den jernbaserte koagulanten PIX-318 og aluminiumsbaserte PAX-18 samt med Metalsorb PCZ og natriumsulfid som fellingsmiddel. Det studerte vannet var tunnelvaskevann som ble hentet fra Vålerengatunnelen i Oslo og Grilstad-tunnelen i Trondheim. Resultatene viser lave turbiditetsverdier, gjenværende turbiditet nådde et nivå <5 NTU for koagulantdose 5 mmol Me/L ved bruk av begge koagulantene. Forsøk med pH-justert råvann indikerer at en mer effektiv reduksjon i turbiditet kan oppnås ved høyere pH verdier og lavere koagulantdoser, mens en høyere koagulantdose også gir lav restturbiditet ved lavere pH-verdier.

Med hensyn til turbiditetsreduksjon var ytelsen til PIX-318 og PAX-18 relativt lik, men generelt var en lavere koagulantdose tilstrekkelig for å nå en bestemt grense (f.eks. 1 NTU) ved bruk av PIX-318. Begge koagulantene hadde produsert en betydelig turbiditetsreduksjon etter 60 minutters sedimentering, mens en ytterligere økning i sedimenteringstid viste seg å ha liten effekt på restturbiditeten. Etter 120 minutter med sedimentering kan begge koagulantene sies å ha omtrent samme ytelse med hensyn til turbiditet.

Når det gjelder fjerning av metaller, viste PAX-18 seg å være noe mer effektiv, spesielt med hensyn til reduksjon av den oppløste fraksjonen. For metaller som sink, som vanligvis antas å være assosiert med den lavmolekylære fraksjonen og dermed også å ha en økt mobilitet og biotilgjengelighet, ble det oppnådd en gjennomsnittlig reduksjon på omtrent 85% med PAX-18 som koagulant ved 5 mmol Al/L. Sammenlignet med dette ble det målt en gjennomsnittlig reduksjon av sink i prøvene på omtrent 75% når PIX-318 ble injisert som 5 mmol Fe/L. Tilsvarende var PAX-18 mer effektiv med hensyn til å redusere konsentrasjonene av nikkel i vaskevannet. Når man vurderer kobber, krom og mangan, var ytelsen til koagulantene ganske lik. For begge koagulantene førte tilsetningen av koagulant selv til en betydelig økning i jern- og aluminiuminnholdet. Etter 60 og 120 minutter med sedimentering ble konsentrasjonene imidlertid igjen redusert til et nivå som anses som akseptabelt i forhold til eksisterende anbefalinger for utslipp.

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# Glossary

Symbol	Definition	
AADT	Annual average daily traffic	
DOC	Dissolved organic carbon	
EQS	Environment quality standard	
Ferric	Iron in its $+3$ oxidation state	
LMM	Low molecular mass	
NPRA	Norwegian public roads administration	
PAH	Polycyclic aromatic hydrocarbon	
PAX	Aluminium-based coagulant	
PIX	Iron-based coagulant	
SS	Suspended solids	
TOC	Total organic carbon	
TWW	Tunnel wash water	

# 4 Introduction

# 4.1 Points of departure

Norway has well over 1000 tunnels with a combined length of >800 km (Meland et al. 2010). Local topography with steep elevation distinctions, fjords and mountains makes tunnels a suitable choice for road infrastructure. Also in urban areas where space is limited and traffic load is relatively high, tunnels have proven to be a preferred solution. Due to emissions from traffic, pollutants accumulate on the road, tunnel walls, ceilings and interior.

For operational reasons and to ensure safety, road tunnels require cleaning with a certain frequency. Most tunnels are cleaned an average of 1-12 times per year, depending on annual average daily traffic (AADT) (Meland et al. 2010). The cleaning process generates a large amount of contaminated water. Most older Norwegian tunnels are without any treatment facilities for wash water, beyond drains and gully pots for removal of larger particles and coarse material. As a consequence, the pollutants accumulated in the tunnel along with any potential detergent used for washing is often discharged directly into nature.

Tunnel wash water contains many of the same substances as regular highway runoff. Due to accumulation of dirt in the tunnel, the concentrations found in tunnel wash water are however often higher. Typical pollutants found in highway runoff and tunnel wash water include metals like lead (Pb), cupper (Cu), nickel (Ni) and zink (Zn) among others, as well as organic environmental toxins such as polycyclic aromatic hydrocarbons (PAHs) and salts (NaCl) during the wintertime (Meland et al. 2016, Garshol 2015).

The Norwegian national transport plan for 2018-2029 states that discharge of damaging substances into nature should be avoided (Avinor et al. 2021). For this reason, several studies on best practice for handling of tunnel wash water has been conducted, e.g. (Garshol et al. 2016, Rathnaweera et al. 2019, Barbosa et al. 2007, Korytář et al. 2022):

- Garshol et. al (2016) reviewed metal removal from tunnel wash water through several treatment technologies such as chemical precipitation & flocculation.
- Rathnaweera et. al (2019) reviewed a variety of treatment technologies, among other coagulation with different coagulants.
- Barbosa et. al (2007) investigated the pollutant load in tunnel wash water.
- Korytár et. al (2022) conducted a study on chemically enhanced sedimentation as treatment and among others reviewed the effect of filtration on granulated activated carbon.

Today, all newer tunnels are built with sedimentation basins as primary treatment solution for the wash water. The efficiency of sedimentation alone as treatment is to a high extent affected by the proportion of particle bound pollutants. The wash water composition is however dependent on a variety of seasonal and operational conditions, such as traffic load, washing procedure, time of year, tunnel length or number of lanes (Meland et al. 2016).

Previous research has found a majority of pollutants in tunnel wash water to be particle bound, which can be relatively easily reduced through sedimentation (Meland 2010, Åstebøl & Coward 2005). However, metals may be present in different species varying in size, charge properties, oxidation state etc. Meland et al. (2010) showed that metals and PAHs were generally present in the discharge at elevated levels after a wash event, and that metals like As, Ca, Mg, Ni and Zn were more associated with low molecular mass species (LMM). The LMM fraction (>10 kDa) is assumed to be more bioavailable and mobile compared to particle bound contaminants.

Additionally, tunnel wash water may contain elevated concentrations of road salts and dissolved organic carbon (DOC), which under oxygen poor conditions may remobilize contaminants in the sedimentation basins and contribute to increased transportation. Thus, sedimentation alone cannot necessarily ensure sufficient water quality of the effluent for discharge into nature. (Meland 2010).

Chemical coagulation is proven effective and is widely used in water treatment, but is not the prevalent practice for tunnel wash water. Studies have found coagulation to have an effect on both smaller particles (>10 um), on dissolved metals such as cupper (Cu) and zinc (Zn) and on polycyclic aromatic hydrocarbons (PAHs) (Nyström et al. 2020, Johnson et al. 2008). The effects of chemical coagulation are however dependent on a variety of water quality parameters, as well as type of coagulant used, coagulation pH etc.

The use of detergents is assumed to increase the concentration of dissolved contaminants. Chemical precipitation is proven effective when it comes to removal of dissolved metals, however the required dosage can be different to predict with changing water quality. The precipitation process itself to a high degree depends on water quality parameters such as pH, and additionally wash water containing detergent is assumed to require a higher dose to efficiently precipitate heavy metals.

Further, treatment of tunnel wash water generates large amounts of sludge which needs to be collected. Depending on traffic load and washing frequencies, the sludge needs to be further treated and disposed of at certain intervals. Implementing coagulation and flocculation as a treatment step is assumed to increase the overall amount of sludge produced during the tunnel wash water treatment, due to significantly improved sludge separation and handling properties of the coagulated sludge. This brings issues related to economy and practicality forth, especially when seen in coherence with implementation of new and more advanced treatment solutions for the wash water.

# 4.2 Objectives

The goal of this thesis is to review the effects and feasability of coagulation and chemical precipitation as a treatment technology for tunnel wash water. The thesis aims to describe the optimized conditions for these processes to take place and to evaluate the robustness of the process with respect to small changes in water quality parameters such as pH. Further, the study aims to review what changes in water quality coagulation and chemical precipitation brings forth, and which practical issues this produces with respect to further treatment. Hence, the research objectives under review are as follows:

- The impact of coagulation and chemical precipitation as treatment for removal of particulate and dissolved metals from tunnel wash water under different coagulation/precipitation conditions
- The robustness of the processes under optimized conditions
- Sludge generation and handling
- Chemical consumption and need for further treatment

# 5 Background

# 5.1 Tunnel operation

For operational and for safety reasons, tunnels require regular cleaning. Accumulation of dirt and pollutants on the walls, ceilings, roadways and interior can worsen visibility and the overall aesthetic expression of the tunnel, provide a harmful work environment for technical staff and decrease the lifespan of the tunnel (Statens Vegvesen 2014). The cleaning is usually carried out through sweeping and by the use of tap water, often with the addition of detergents to increase cleaning efficiency. Prior to a wash, particles such as sand and dust are removed from the roadway through sweeping (Meland & Rødland 2018). The wash is then carried out as specified in guidelines set by the Norwegian public roads administration (NPRA) using water and potentially with the addition of a detergent. According to guideline R610, different washing procedures for tunnels are defined to include the following (Statens Vegvesen 2014):

- Whole wash: cleaning of roof and tunnel walls, doors, barriers, traffic signs and all technical installations, as well as emptying of gullies and cleaning of the roadway and road shoulders.
- Half wash: cleaning of tunnel walls, doors, barriers and all technical installations, as well as cleaning of the roadway and road shoulders
- Technical wash: cleaning of all technical installations, as well as cleaning of the roadway and road shoulders.

Cleaning frequencies are determined based on traffic volume. Common practice is that highly trafficked tunnels with an annual average daily traffic (AADT) > 15.000 veichles per separate tunnel lane should be cleaned 6-12 times per year. Tunnels with a low traffic volume (AADT < 4.000 veichles per separate tunnel lane) should be cleaned < 1-2 times per year. Recommendations for cleaning frequencies according to guideline R610 are presented in full in Table 1.

Traffic-volume: Whole cleaning		Half cleaning	Technical cleaning
AADT/tunnel lane			
0 - 300	Every 5 year	-	Every year without
			cleaning: whole
301 - 4000	1 pr year	-	1 pr year
4001 - 8000	1 pr year	1 pr year	2 pr year
8001 - 12000	1 pr year	2 pr year	3 pr year
12001-15000	2 pr year	3 pr year	5 pr year
>15001	2 pr year	4 pr year	6 pr year

Table 1: Cleaning procedures. Modified after (Statens Vegvesen 2014).

# 5.2 Tunnel wash water

Tunnel wash water contains many similar substances as regular highway runoff. Concentrations of the different substances are however usually higher than compared to regular highway runoff, as the pollutants are not flushed away from rainfall events but rather accumulated on the tunnel walls, ceilings, road and interior.

Water volumes used for washing can vary from 30 to 150 L per m tunnel, depending on AADT and time between washes among others (Hallberg et al. n.d.). The pollutant load will vary accordingly and depend on factors such as water volume or type of detergent used for washing, but also on traffic load, seasonal variations, time in between washes or even type of wash. Pollutants found in tunnel wash water typically originate from wear on veichles, roadways and technical equipment in the tunnels in addition to accidental spills or seasonal strains such as de-icing. An overview of typical pollutants found in TWW and their most likely origins can be found in table 2.

Table 2: Contaminants and their possible origins. Modified after (Meland 2010, Korytář et al. 2022).

Origin	Contaminant
Tires	Al, Pb, Zn, Ca, Cd, Co, Cu, Mn, W, hydrocarbons, PAH (pyrene,
	fluoranthene, benzo(ghi)perylene)
Brakes	Ba, Cu, Cr, Fe, Mo, Ni, Pb, Sb
Catalytic converters	Pt, Pd, Rh
Veichle body	Cr, Fe, Mn, Zn
Combustion	Ba, Ca, Cr, Co, Mo, Ni, V, Sb, Zn, PAH (naphthalene)
Oil, petroleum spill,	Ni, Zn, hydrocarbons, PAH (LMM)
lubricant oil	
Road surface (asphalt,	Al, Ca, Fe, K, Mg, Na, Ph, Si, Sr Ti, PAH (Mix of HMM and
bitumen)	petrogenic LMM species, chrysene)
Winter operation, de-icing	Ca, Cl, Mg, Na, ferro-cyanide
Road equipment (e.g.	Zn (galvanized steel)
crash barriers, traffic signs)	
Detergents used in tunnel	Tensides
wash	

Metals can be present as different speices in the wash water and be associated with either the particulate or the dissolved fraction. This depends on several factors such as pH, water temperature and other compounds present and strongly affects bioavailability and toxicity of the compounds. Metals are generally most toxic as ions such as  $Cu^2+$ ,  $Zn^{2+}$  etc. Complex bindings to detergents or formation of metal hydroxides generally represent less toxic forms (Garshol et al. 2016).

Metals and substances connected to metals contained in the wash water can be categorized according to size. The smallest fraction is referred to as colloids, which are characterized by a small diameter, typically in the range 0,01-1  $\mu$ m, a large specific surface area and a negative surface charge (Ødegaard et al. 2014). Compounds with a larger diameter are referred to as particles, whereas even smaller are considered to be solutes. The LMM fraction is defined according to molecular size (>10 kDa).

In 2015, limit values for concentrations of specified chemicals for in water bodies was implemented into the regulation of water management according to the EU water framework directive (EU-WFD) (Klimaog miljødepartementet 2006). As of 2018, 45 priority parameters are included in evaluation of chemical condition of water bodies. These substances are pollutants such as nickel or lead that are not necessarily hazardous, but that could compose a threat to human health or aquatic ecosystems. For each substance, two different environmental quality standards (EQS) are defined. The limit values represents annual averages (AA) and maximum admissible concentration (MAQ), and are defined for both inland (freshwater) and other (coastal) surface water bodies (European Parliament 2013). In addition, Norway has implemented limit values for region specific substances in water bodies, biota and sediments (Arp et al. 2014). It is important to note that EQS-values are not directly transferable to discharge limits. The concentration of a substance in a discharge will not be equal as the discharge is diluted in the recipient, and several factors will impact the chemical condition of the water body. For this reason, limit values for discharge are determined based on a full-scale evaluation of the discharge (Evensen 1995).

Studies conducted by Hallberg et al. (2014), Barbosa et al. (2007), Roseth and Meland (2006) and Meland and Rødland (2018) indicate that tunnel wash water often contains significantly elevated concentrations of different pollutants, such as PAHs, particles and metals. The concentrations in many cases strongly exceeded the EQS values. Table 3 presents the results of selected studies and EQS values for fresh water for selected metals. EQS values for As, Cr, Cu, Pb and Zn are according to EU-WFD. For Al, Fe, Mn and Sb, EQS values are not defined. For these substances, comparable limit values from the UK and limit values from the norwegian drinking water regulation are presented as specified as a substitute where such values are available. The EQS values as they are presented should be multiplied with a factor 10 to account for dilution in the recipient as recommended by the directorate of environmental protection (Meland & Rødland 2018). It should also be noted that the EQS value in the case of metals initially refers to dissolved concentration (0.45  $\mu$ m) and not the total concentration.

Name of substance	AA-EQS (In-	MAC-EQS	Values found	Values found
	land)	(Inland)	$(Dissolved)^1$	$(\text{Total})^{2,3}$
Al $[\mu g/L]$	$15 \ (pH>6.5)^4$	$100^{5}$	296-504	-
As $[\mu g/L]$	4.8	8.5	2.7 - 2.9	111259
$ m Cr[\mu g/L]$	3.4	3.4	1.1 - 346	346
$Cu \ [\mu g/L]$	7.8	7.8	9.4-12	195-1020
Fe $[\mu g/L]$	-	$300^{5}$	514-790	5693 - 147418
$Mn[\mu g/L]$	-	-	180	-
Ni $[\mu g/L]$	4	34	7.8 - 8.2	304
$Pb \ [\mu g/L]$	1.2	14	1.6 - 2.2	48-149
Sb $[\mu g/L]$	5	-	-	74
$Zn \ [\mu g/L]$	11	11	476-624	962-4742

Table 3: Comparison of EQS values and metal concentrations found in TWW.

1: (Meland et al. 2010); 2: (Meland & Rødland 2018); 3: (Barbosa et al. 2007); 4: (Crane et al. 2007)

#### 5.3 Sedimentation, coagulation and flocculation

Sedimentation describes the process of removing particle bound pollutants through gravity (settling) (Ødegaard et al. 2014). The sedimentation efficiency is contingent on particle size and shape, density and density of the liquid. Since a majority of pollutants contained in tunnel wash water are considered to be particle bound, sedimentation alone can often provide a sufficient removal rate. Removal of particles in the aqueous phase is however limited (Stotz & Holldorb 2008). Still, sedimentation is per 2023 the standard practice for handling of tunnel wash water. Many newer tunnels are without any treatment beyond drains and gully traps for removal of larger particles such as gravel and other objects, whereas all newer tunnels are built with sedimentation as primary treatment (Garshol 2015).

Due to their negative surface charge, colloidal particles will however be stable and thus unable to sediment. Coagulation describes the injection of a chemical (coagulant) to the solution, in order to destabilize such particles (Ødegaard et al. 2014). Due to affordability and availability, inorganic metal-coagulants are dominant in use. Most common are iron- or aluminum-based salts. Upon addition, charged ions of the coagulants will interact with and neutralize the particles suspended in the liquid. Consequently, the destabilized colloids can effectively be agglomerated in larger flocs that correspondingly can be easier separated and removed. Coagulation is a relatively quick process, rapid mixing is required to prevent the coagulant from reacting with the water and loose effect.

The agglomeration of destabilized colloids into larger flocs is usually referred to as flocculation (Ødegaard et al. 2014). There are two main flocculation mechanisms:

- Perikinetic flocculation: the natural flocculation process where flocs are formed due to Brownian motion of the colloids. This is a highly time consuming process, and thus not commonly used in water treatment.
- Orthokinetic flocculation: addition of energy through stirring in order to increase collision velocity between the colloids. In consequence, larger flocs can be formed more rapidly. This is the mechanism commonly used in water treatment.

Although stirring is required for an efficient flocculation process, a reduced stirring velocity is desired relative to the coagulation process (Ødegaard et al. 2014). This is to avoid dispersing existing flocs while simultaneously ensuring sufficient contact between the colloids. As a compensation for the reduced stirring velocity, the duration of the process itself is often extended. Depending on their properties, the resulting flocs can be removed as sludge in one or multiple steps. Typically, the sludge can then be deposited through processes such as sedimentation or flotation. The coagulation – flocculation process is simplified illustrated in Figure 1.

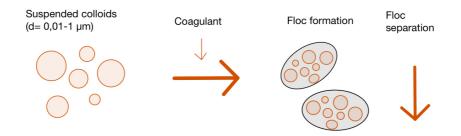


Figure 1: The coagulation - flocculation process. Modified after (Ødegaard et al. 2014).

#### 5.4 Optimalization of coagulation process

Factors such as coagulation pH, temperature, use of detergents, type of coagulant etc. will to various extents impact the coagulation efficiency. The presence of detergents alone in tunnel wash water is shown to increase the concentration of dissolved contaminants and might interfere with coagulation results. Aasum (2013) for instance observed the presence of detergents to increase the mobility of metals, whereas Smith et. Al (1956) found detergents to decrease efficiency of alum floc formation during conventional chemical coagulation.

Several studies have been conducted on the best practice for handling of tunnel wash water. For instance, Rathnaweera et. Al (2019) investigated a variety of treatment combinations, such as filtration, sedimentation, chemical precipitation, aerobic biological treatment and anoxic/anaerobic biological technologies, for treatment of tunnel wash water. Four commonly used coagulants were studied in the experiments, two Al-based and two Fe-based. The coagulants were found to perform quite similarly, however the required doses when using a Fe based coagulant was lower.

The optimal coagulation pH is usually in the range 5-7, depending on the coagulant used. Higher pH ranges (>9-10) can have toxic effects on microorganisms essential for the degradation of organic matter. In lower pH ranges, the solubility of metals is generally increased due to an increased amount of positively charged particles available in the liquid to react with the negatively charged colloids. Consequently, coagulation in a pH range that is considered too low can lead to a higher concentration of remaining metals. In order to reach the optimum pH range and thus optimize the coagulation process, pH adjustments are usually required (Ødegaard et al. 2014).

Compared to sedimentation alone, coagulation can be related to increased costs, among others in form of chemical consumption or possibly the need for additional treatment to reach sufficiently low residual metal concentrations. The treatment time is however significantly reduced and due to better separation of the aqueous and solid phase, the potential for recovery of metals and other resources contained in the water is notably higher.

# 5.5 Electrical double layer

Due to their negative surface charge, repulsive forces will be present between colloids contained in water (Ødegaard et al. 2014). To preserve charge neutrality, the particle will be surrounded by a cluster of ions. A layer of counter-ions (cations) will be fixed to the particle surface. This layer constitutes the Stern layer and is of fixed charge.

Beyond the stern layer, a diffuse ion layer consisting of both positively and negatively charged ions will develop. This is as a direct consequence to the dynamic equilibrium which is established due to the repulsive forces from the cations in the stern layer and the attracting forces from the colloid itself (Ghernaout 2020). Within the diffuse layer, the amount of cations will gradually decrease as the distance from the colloid surface increases, whereas the presence of anions will increase. This dynamic change in cations present proportionate to anions present, is what eventually results in an equilibrium being established. The stern layer and the diffuse layer constitutes the electrical double layer, as illustrated in figure 2. The repulsive forces between similarly charged colloids as well as the presence of the electrical double layer keeps the colloids relatively stable in solution and thus prevents agglomeration into larger and easily removable flocs.

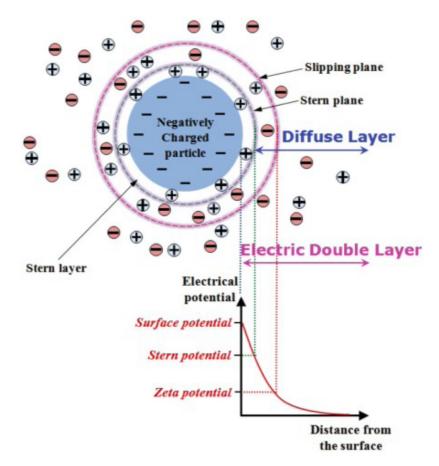


Figure 2: Electric double layer (Park & Seo 2011).

#### 5.5.1 Zeta potential

Particles dispersed in a liquid will have different properties. A hydrophilic particle will be water-seeking, whereas a hydrophobic particle will be water avoidant. Due to such properties and other interactions, particles contained in a fluid will undergo Brownian motion. The particles subject to Brownian motion are assumed to move with both their stern layers to preserve charge neutrality, but also with parts of their diffuse layers. As a consequence, a shear plane materializes within the diffuse layer. This plane represents the interface between the mobile particles of the diffuse layer and the dispersant. The Zeta potential refers to the electrokinetic potential of this shear plane (Ødegaard et al. 2014).

The Zeta potential is a measure for the stability of the dispersed particles. A high absolute value for the Zeta potential corresponds to a high electrokinetic potential and thus stronger repulsive forces between the particles, which means the particles are less probable to collide with each other and form larger aggregates that can be separated later on. A zeta potential close to zero corresponds to weaker repulsive forces between the particles and is thus desirable for an effective coagulation process.

#### 5.6 Coagulation mechanisms

Destabilization of colloids in water is a complex process. Depending on properties of the coagulant as well as the to-be-destabilized colloids, different mechanisms for coagulation occur. Generally, four different coagulation mechanisms are distinguished (Ødegaard et al. 2014) (Ghernaout 2020) (Meyn 2011):

- Destabilization because of double layer compression: In a neutral pH area the colloids in suspension will be negatively charged. Consequently the electric double layer is formed around the colloids, however the thickness of the electric double layer is dependent on charge and concentration of counter-ions in the solution. With a higher charge and/or concentration, the reach of the repulsive forces will decrease to the point where van der Waals forces dominate and coagulation can occur.
- Adsorption and charge neutralization: In a neutral pH-area (6-8), most particles contained in the solution will be negatively charged. When low coagulant concentrations relative to the colloid concentration is added, positively charged hydrolytic intermediates can form and be adsorbed on the colloids. Adsorption of positive ions brings a change of surface charge on the particles forth. Consequently, the colloids can be destabilized from the coagulant and precipitate as metal hydroxides (complexes). It should however be noted that excessive amounts of oppositely charged ions potentially can shift the equilibrium beyond charge neutralization and thus restabilize the particles. As a consequence, there is an optimal concentration (and pH range) where destabilization occurs.
- Adsorption and inter-particle bridging: Long-chained synthetic polymers can be adsorbed on certain particle surfaces and consequently, cross-links (bridges) can be established within the particulate matter. This bridging allows for destabilization to take place. The process is possible because of charge-induced reactions, dipole reactions, hydrogen bonds and/or wan der Waals-forces. Efficiency depends on among others the availability of polymer groups and adsorption capacity of the particles.
- Sweep coagulation: Insoluble precipitation chemicals from Al3+ and Fe3+ are formed at high coagulant dosages. In high or low pH-areas these complexes are charged, whereas they can be precipitated as metal hydroxides within the optimum pH area of the coagulant added. The precipitation process occurs rapidly, so that particulate matter can be 'swept' into the structure and be part of the precipitation product. Thus, this type of coagulation allows for removal of all kinds of colloids contained in the fluid, as floc formation is not affected by the properties of the to-be-removed material.

Within the common operating pH- and coagulant dose intervals, the predominant coagulation mechanisms are usually sweep coagulation and charge neutralization (Aghapour et al. 2016). Ferric ions are usually better suited for these coagulation mechanisms as lower doses are required to achieve comparable effects. This is illustrated by the solubility diagram for trivalent aluminium and iron as presented in Figure 3:

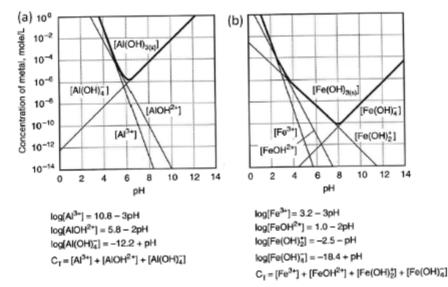


Figure 3: Solubility diagram for (a) Al(3) and (2) Fe(3) at 25 degrees (Ghernaout 2020).

Further, iron based coagulants have a wider operating pH range as can be seen in Figure 4. Studies on several water parameters have found Fe-based coagulation to be more effective. Aghapour et. Al (2016) investigated nitrate removal from wastewater through coagulation and compared the efficiency alum and ferric chloride. Within the common dosage range, ferric chloride was found to be more effective and the working coagulation mechanisms were determined to be adsorption and sweep coagulation. Due to the formation of toxic by-products, the operational range for especially aluminum hydroxide diminished. Nyström et. al (2020) investigated the effects of five different coagulants in terms of reducing particle content, organic carbon, total and dissolved metals, hydrocarbon oil index and PAHs, and also found ferric chloride to be the only coagulant affecting the particle size distribution post treatment. The study did however uncover an increase in dissolved Zn, a change that in conclusion was likely associated with a considerable drop in pH resulting in higher ion mobility. This is in turn related to the coagulant itself being strongly acidic.

PIX-318 has a known optimal pH area in the range 4.5-5.5, and typical dosages in Norwegian water treatment are in the range 3.0-6.0 mg Fe/L (Ødegaard et al. 2014). Although sweep coagulation is likely to be the predominant coagulation mechanism under optimal conditions, there is a narrow interval of co-agulant doses where destabilization of the contained colloids occurs more rapidly than the establishment of equilibrium within the solution after the coagulant is injected. In this interval, destabilization because of adsorption and charge neutralization is the functioning mechanism.

The different coagulation mechanisms rarely occur alone (Ødegaard et al. 2014). As the coagulation process itself is relatively complex and contingent on a variety of parameters, so is which mechanism is dominant. Usually a combination of mechanisms will be present and consequently a change of mechanism can potentially be observed as the coagulant dosage changes for different pH values. As illustrated in the solubility diagram for FeCl<sub>3</sub>, solubility of the coagulant is reliant on pH, subsequently also in which chemical species the ferric occurs. For less optimum pH values, higher coagulant dosages are required. However, as illustrated in figure 3, re-stabilization by overdosing can occur. The maximum coagulant dosage where this occur is referred to as critical stabilization concentration (CSC) (Meyn 2011).

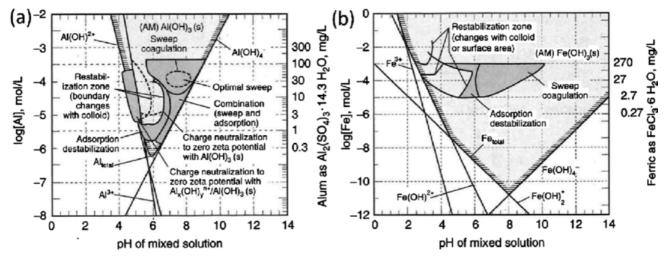


Figure 4: Working coagulation mechanisms for (a) Al(3) and (2) Fe(3) at 25 degrees (Ghernaout 2020).

For the coagulation process to be efficient, a minimum dosage of coagulant is required. This concentration is defined as critical coagulant concentration (CCC) (Meyn 2011). The optimal coagulation dosage is in the range of CCC and CSC and is generally defined as the smallest dosage required to achieve the anticipated treated water quality. Based on present coagulation mechanisms, the optimal coagulation dosage depends on raw water quality such as particle concentration, pH, alkalinity, hardness, temperature, etc. (Ødegaard et al. 2014).

#### 5.7 Chemical precipitation

Chemical precipitation is proven effective to reduce residual metal concentrations in several previous studies (Prokkola et al. 2020, Garshol et al. 2016). The basic principle is converting soluble metal species into relatively insoluble compounds through the addition of precipitating agents such as sodium sulfide (Na<sub>2</sub>S). Naturally, metals that dissolve in water undergo hydrolysis and form hydroxides. In practice, the process of hydrolysis implicates the generation of hydrated complexes through splitting of water molecules (Ghernaout et al. 2015). Hydrolysis provides an increase of available H+ ions in the water and is thus strongly contingent on pH. On a general form, the reaction sequence can be simplified as following (Ødegaard et al. 2014):

 $Me^{3+} \rightarrow Me(OH)^{2+} \rightarrow Me(OH)^{+}_{2} \rightarrow Me(OH)_{3} \rightarrow Me(OH)^{-}_{4} (1)$ 

#### 5.7.1 Speciation of Al and Fe

Since hydrolysis releases protons, these reactions leads to a decrease in alkalinity (Ghernaout et al. 2015). What chemical species is predominant, depends on the hydrolysis equilibrium as illustrated by the speciation diagram for trivalent Fe and Al in Figure 5. Different species has various solubility. For instance, aluminum is found as a variety of different species depending on pH, alkalinity, temperature and dissolved organic carbon (DOC) among others. Aluminum is commonly present as hydroxides in water. The hydroxide ion OH- may however form complexes with present organic or inorganic ligands, leading aluminum to be present in other forms. At pH < 5.5,  $Al^{3+}$  is the prevalent form along with inorganic monomeric complexes such as  $Al(SO_4)$  or  $Al(OH)_x$ . These forms of aluminum have a higher toxicity due to an increased availability. At pH > 8 solubility is increased due to  $Al(OH)_4^-$  being the predominant species (Crane et al. 2007)

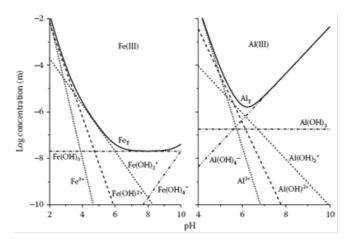


Figure 5: Speciation diagram for Fe(3) and Al(3) (Ghernaout et al. 2015).

Iron can be present in several chemical species. In its ionic form, +2 and +3 are the most common oxidation states. Under most conditions,  $Fe^{2+}$  is oxidized to trivalent ferric ( $Fe^{3+}$ ). Insoluble  $Fe^{3+}$  species adsorb to natural organic compounds and consequently stabilize as colloids. Collodial forms of iron usually include in the dissolved fraction. Dissolved  $Fe^{2+}$  is assumed to be associated with diverse toxic effects to a higher extent than dissolved  $Fe^{3+}$ . Still, diverse precipitates of  $Fe^{3+}$  can also contribute to toxicity in the solution (Johnson et al. 2007).

#### 5.7.2 Sulfide precipitation

Relative to metal hydroxides, the solubility of metal sulfides is low. Consequently, a much more efficient precipitation and thus also removal of the dissolved metals can be accomplished. Simplified, sulfidic precipitation makes use of a sulfide source such as  $H_2S$  or  $Na_2S$  to react with cations in an aqueous solution. The reaction mechanism can be simplified as (Estay et al. 2021):

 $Me^{2+} + S^{2-} - MeS$  (S) (2)

A lowered solubility translates to a number of advantages seen from an environmental point of view. When undesirable metals are removed and disposed of safely, they are less probable to be leached in a wide pH range (Estay et al. 2021). Further, the sludge volumes generated from sulfide precipitation are low compared to comparable technologies. Metal concentrations in the generated sludge are however usually high, which in turn subsidizes recovery of metals from the sludge that can be reused for other purposes.

Due to their solubilities, metal sulfides are usually precipitated as colloids. The amount of sulfide added should be with respect to the contained metals, otherwise excess sulfide will remain in solution or the metals will not be fully removed. However local supersaturation at the injection points will occur and the supersaturation levels virtually cannot be controlled at low levels. Consequently, fine flocs will usually be prevalent (Veeken et al. 2007). As a result, the separation of precipitates from the liquid that can be achieved through technologies such as sedimentation or filtration is usually poor. Under optimal conditions, Zianuddin et.al. (2019) found the sludge generation to be 0.02 g/300 mL sample. Further, the amount of generated sludge decreased as the precipitant dose was either decreased or increased.

The precipitation of metals as sulfides usually occurs at lower pH values than the precipitation of metals as hydroxides. Still, the pH must not be too low as some sulfide precipitates are acid soluble. The process is typically carried out at alkaline pH values to obtain minimal solubilities, but more importantly to avoid formation of toxic  $H_2S$  gas. Precipitation pH of diverse metal hydroxides and sulfides is shown in Figure 6, which illustrate how the sulfides start to precipitate at significantly lowered pH values. Theoretically, metal sulfides precipitate according to pH which in turn implies that selective metal removal is possible.

Compared to conventional hydroxide precipitation, sulfide precipitation is not only more efficient, but can also achieve significantly higher removal rates. Zianuddin et. Al. (2019) found zinc removal of >90%with the use of sulfidic precipitation compared to >60% using hydroxide precipitation. Li et. Al. (2019) found removal efficiensies of 99%, 97%, 99% and 98% for Cd, Pb, Cu and Zn respectively through sulfidic precipitation combined with Fenton oxidation, whereas removal efficiencies of 94%, 99% and 99% were achieved for Cu, Zn and Cr respectively by Yatim et. Al. (2021). Further, Garshol et. Al. (2016) investigated metal removal from tunnel wash water by the use of several technologies and achieved the best removal through metal sulfide precipitation in combination with chemical coagulation with PIX. Concentrations of certain metals after sulfide precipitation as found by Zianuddin et. Al (2019) and Li et. Al (2019) are presented in Table 4.

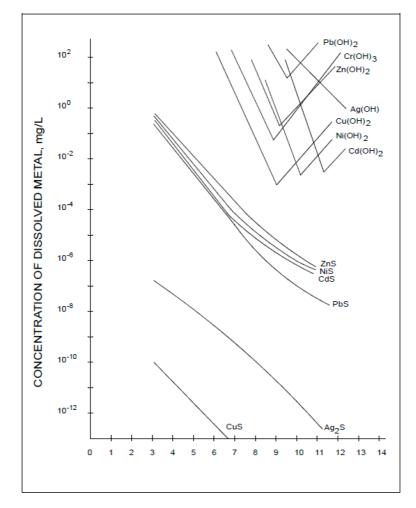


Figure 6: Precipitation pH and concentrations of hydroxides and sulfides (Prokkola et al. 2020).

Table 4: Metal contents  $[\mu g/L]$  measured after treatment with sulfide precipitation. Modified after (Zainuddin et al. 2019,<br/>Li et al. 2019)

Metal	Zianuddin et. al (2019)	Li et. al (2019)	
Cd	<13.4		
Cu	< 39.6	< 0.2	
Ni	< 5.32		
Pb	< 357	< 400	
Zn		< 363	

#### 5.7.3 Metalsorb PCZ

Metalsorb PCZ is an effective precipitant when it comes to removal of ionized metals, including Cd, Cu, Ni, Pb and Zn. For instance, pilot project Hill Canyon wastewater treatment plant achieved a 45,7% reduction in copper concentrations in industrial wastewater with the use of Metalsorb PCZ alone (California Regional Water Quality Control Board 2016), whereas Fraceto et. Al (2023) achieved removals of >90% for divalent Cu, Zn and Co when implementing Metalsorb. Metal concentrations in the untreated wastewater and water treated with Metalsorb as found by Fraceto et. Al. are presented in Table 5.

Table 5: Metal contents [mg/L] after treatment with Metalsorb PCZ. Modified after (Fraceto et al. 2023)

Metal	Metal content in untreated water [mg/L]	Metal content in water treated with
		Metalsorb PCZ $[mg/L]$
Cu	50.83	1.25
Co	47.14	0.61
Zn	52.31	1.67

The working principle behind the precipitation is Metalsorb functioning as a chelating agent, as Metalsorb in principle is a polymer. The chelating properties originate in a sulfur derivative grafted onto an organic molecule, as shown in Figure 7. Subsequently, divalent metal ions (cationic) contained in the solution form bonds with the anionic Metalsorb, and thus form insoluble salts that can easily be precipitated and thus removed from the solution (Watersolve LLC 2023).

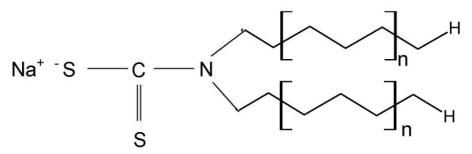


Figure 7: The chemical structure of Metalsorb (Fraceto et al. 2023).

Precipitation with metalsorbents has advantages with respect to sludge generation and operating conditions in comparison to conventional hydroxide or sulfide precipitation (Kansas water tech. n.d.). The sludge density is significantly improved, which in turn enables a higher metal recovery. The chelated complexes are however stable, and further treatment is usually necessary for metal recovery. Hence, for economic reasons, the sludge is usually stored and destroyed. The treatment efficiency of Metalsorb is to little extent vulnerable to small changes in water quality. Metalsorb is in theory applicable over a pH range of 3 to 10, which results in reduced operating costs as the need for e.g. pH adjustments are diminished. One major disadvantage is the excess of organic molecules, not bonding with metal ions, which results in increased chemical oxygen demand (COD) as the effluent is contaminated with organic substances (Fraceto et al. 2023).

#### 5.8 Sludge treatment

Treatment of tunnel wash water, whether through coagulation, sedimentation alone or comparable technologies, generates large amounts of sludge that needs to be handled. The sludge itself contains resources like water, energy potential and nutrients. Sludge from tunnel wash water is however highly polluted and needs to be treated accordingly prior to deposition. Typical sludge treatment includes putrefaction and pasteurization through for example heating, drying or long-term storage to decrease the contaminant load in the sludge. In addition, sludge contains large water volumes which it is necessary to separate to achieve a homogenous sludge quality. This has practical applications, among others to significantly reduce the necessary transport- and storage volumes.

Sludge that is stored long term will undergo consolidation. This means, the sludge volume will to some extent be reduced over time due to compression and decomposition of the sludge. According to Johannessen et. Al. (2017), a volume reduction of 30-50% due to consolidation can be observed if the sludge is stored for two years compared to a storage time of six months. As a consequence, increased sludge ages will result in a much more stabilized sludge (Gray 2010).

Conditions for storage, transportation and deposition of sludge are set by the county governor. For potential reuse of the sludge, such as for agricultural or energy purposes, the municipality is the relevant authority. Permissions are only granted where pollution related-, agricultural-, health- and hygienic conditions are satisfactory attended to. Maximum metal contents in sludge to be reused are presented in Table 6. Contents are presented as mg Me/kg TS. Sludge containing elevated concentrations of metals, environmental toxins and oils, such as sludge from tunnel wash water, is however usually delivered to appropriate landfills and disposed of (Gray 2010).

The sludge accumulated by tunnel wash water is typically collected from the oil separator(s) in combination with cleaning of the separator(s). In principle, clean water is used to flush out the basin. The polluted water used for cleaning out the basin is collected along with the emptied sludge and transported to approved depositions. The process itself is relatively quick, with durations ranging from 4 to 9 hours.

Metal	Agricultural areas	Recreational areas	
Cd	2	5	
Pb	80	200	
Hg	3	5	
Ni	50	80	
Zn	800	1500	
Cu	650	1000	
Cr	100	150	

Table 6: Limit values for metal contents in sludge [mg Me/kg TS]. Modified after (Miljøverndepartementet 1995)

The generated sludge is usually stored prior to transportation to deposition and potential further treatment. The emptying frequencies are determined based on sludge production and volumes available for sludge storage. Previous studies indicate that the performance of the storage facilities is affected by the sludge level in the facility, and thus also sludge production and emptying frequency (Johannessen et al. 2017). Emptying is recommended when the storage facility reaches a filling degree of 70% to prevent loss of SS, and thus increased values of SS in the discharge.

# 5.8.1 The Bjørnegårdstunnel

The Bjørnegårdstunnel is located in Akershus county and has a length of 1900 m. The tunnel has two tubes with in total four lanes, and an estimated AADT of 35.000 veichles (NCC 2023). Information on sludge handling in general and at the Bjørnegårdstunnel was acquired through personal communication with Espen Hoell in Nye Veier and Kai Gundersen in NPRA respectively.

During emptying of one out of two oil separators present at the Bjørnegårdstunnel, samples were collected by the NPRA and analyzed for among other metal contents by Eurofins. The analysis showed adequate contents of pollutants such as metals and PAHs relative to discharge permits. During emptying, it was however necessary to fully pump down the water in the separator in order to fully empty the basin. The samples taken as the water was pumped down showed significantly elevated concentrations of certain metals such as Cu and Zn, which indicates strong local variations. The analyzed samples were taken from the water used to flush out the oil separator. Metal contents found in the representative normal sample and in a sample with poor water quality are presented in Table 7, whereas full results from the analysis are attached in Appendix A. All metal contents are given as  $\mu g/L$ . This was the first time the oil separator was flushed out since the tunnel was opened for normal operation in 2019. According to personal communication, a total 15.26 tons of polluted water was delivered to deposition after flushing out the basin.

Metal	Representative normalsample	Sample with elevated values
As	3.9	2.6
Cd	0.11	0.16
$\operatorname{Cr}$	2.8	1.0
Cu	16	410
Ni	4.3	5.6
Pb	0.59	3.7
Zn	81	2100

**Table 7:** Metal contents  $[\mu g/L]$  found in sludge accumulated at the Bjørnegårdstunnel

The samples previously analyzed from the Bjørnegårdstunnel have been stable and showed pollutant contents similar to the normal sample in the past. For this reason, samples have commonly been exctracted around three times per year. Due to the elevated concentrations found when pumping the basin down, it could however be relevant to extract samples after each wash during draining of the basin in the future.

#### **5.8.2** Costs

Since sludge originating from tunnel wash water often is heavily polluted and containing for instance metals and oil, it needs to be transported to appropriate landfills. Deposition is usually priced per kg contaminated water that is to be disposed of, and is unrelated to degree of pollution. The polluted water to be disposed of from the Bjørnegårdstunnel is delivered to Esval, with a price/kg of 970 NOK ex. VAT. The suction truck used for emptying of the basin and transportation has an hourly cost of 1620 NOK ex. VAT.

Further, additional costs for basins located in the day zone such as cleaning of the surroundings and surrounding vegetation can occur. Cost estimates from the Nordbytunnel indicate hourly costs of approximately 1000 NOK for this kind of maintenance work.

# 6 Materials and Methods

# 6.1 Sampling

Fresh wash water was sampled from the Nestunnel in Buskerud, the Vålerenga tunnel in Oslo and the Grilstad tunnel in Trondheim during cleaning. The samples were taken directly from the sedimentation basins and transferred into 20L water tanks by the use of a submerged pump (Metabo skittenvannspumpe, PS 18000SN). From the Vålerenga tunnel, 300L of wash water was sampled into 15 water tanks and from the Grillstad tunnel, 200L was sampled into 10 tanks. The setup is shown in Figures 8 and 9. The samplings took place on the 26.01.2023, 09.02.2023 and 24.02.2023 for the Nestunnel, Vålerenga tunnel and Grillstad tunnel respectively. All the samples were transported to the Norwegian University of Science and Technology (NTNU), where the experiments was carried out in the drinking water laboratory.



Figure 8: Collection of wash water at the Vålerenga tunnel



Figure 9: The sedimentation basin at the Vålerenga tunnel

During sampling of water at the Nestunnel, the wash water visually appeared to be very clear and with limited contents of large particles. Lack of particles would make it challenging to conduct coagulation experiments on this water. For this reason, a smaller water volume than planned was collected from the Nestunnel and this water was primarily used for initial experiments with chemical precipitation.

# 6.2 Water preparation

In order to ensure a homogenous water quality, the sampled water was mixed. The 20L cans were turned 3 times to resuspend settled particles, and then transferred into a 200L tank for mixing. With the use of a submerged pump and recycled flow from the pump, the sampled water was then mixed again. All of the 20L-cans were then refilled until  $\frac{1}{4}$  full and the remaining water in the 200L-tank was remixed. Then the cans were filled until half-full before mixing again,  $\frac{3}{4}$  full before mixing and then remaining water was refilled into the tanks. A representative sample for water quality measurements was taken from the 200L tank. The re-filled cans were then stored in a cooling room to preserve the water quality as much as possible.

The experiments were carried out within one week of sampling to ensure fresh wash water. All experiments were conducted as standard jar-tests in 2L plexiglass beakers using a Platypus jar tester apparatus.

# 6.3 Analyzed water quality parameters

Several parameters were analyzed during the experiments to obtain knowledge about the overall performance of the procedures. All measurements were conducted three times, before average values were calculated. The following water quality parameters were of interest:

• Turbidity measured using HACH 2100N laboratory turbidity meter from filling approximately 20 ml of sample into a glass vial. The working principle is light diffraction. The measurements were conducted according to ISO 7027-1:2016.

- PH was measured using a Hach sensION+ PH3 pH-meter according to ISO 10523:2008.. The pH meter works like a voltmeter, with a pair of electrodes measuring the hydrogen-ion activity in the solution.
- Conductivity was measured using a conductivity meter according to NS-ISO 7888:1985. The conductivity meter works similar to the pH meter.
- The particle size distribution was delivered from using a Mastersizer3000 from Malvern Panalytical. The working principle is laser diffraction.
- Zeta potential was measured using Zetasizer Nano-ZS ZEN 3600 from Malvern Panalytical.
- Total suspended solids (TSS) was measured according to NS-EN 872:2005.
- Dissolved Organic Carbon (DOC) was measured according to ISO 8245:1999. Approximately 20 ml of sample was used, and the samples was conserved in the freezer in the time following the sampling up to analysis. The technical analysis was carried out by Trine M. H. Næss at NTNU.
- Metal concentrations were measured using ICP-MS. A detailed description of the sample preparation follows in section 6.7.
- Suspended solids was measured as described below.

# 6.4 Suspended solids

Approximately 100 ml of sample was measured and filtered through a 1.2  $\mu$ m filter. The filters were weighed with four decimals beforehand and only touched with clean tweezers and gloves to avoid pollution. Three blanks were included, these were filtered with 100 ml mq-water. After filtration of the samples, the filter cake was allowed to dry before the used filters were placed in aluminum bowls. The filters were heated in the oven at 105 °C and weighed again, then heated at 550 °C and weighed one last time. Due to time limitations with the fresh water in the laboratory, the heating was carried out the following week. This allowed the filter cake to be properly dried before the heating took place. The filtration setup (equivalent to the one used for precipitation) can be seen in Figure 10. The filter cakes can be seen in Figure 11.



Figure 10: Filtration setup used for precipitation and TSS.



Figure 11: Dried filtercake.

# 6.5 Particle removal

The coagulation experiments were conducted with the goal to find optimal coagulation conditions and identify functioning mechanisms. In total, 9 different coagulant doses in the range from 0 mg Me/L to 25 mg Me/L were tested without pH adjustment. One iron based coagulant (PIX-318) and one alum based coagulant (PAX-18) were used for all different doses. Data sheets for PIX-318 and PAX-18 are attached as Appendices A.3 and A.4 respectively. All samples were done in duplicates. The doses tested was a result of existing research conducted on the Nordbytunnel (Garshol et al. 2016) and experiments conducted during the fall 2022 with wash water from E18 Bamble and Kjørholt tunnels as part of the preliminary project work preceding this master thesis.

The jar tests were performed with initial rapid mixing at 200 rpm for 1 min, followed by a slow mixing at 25 rpm for 20 min. The coagulant was injected to the sample right after initializing rapid mixing. The suspension was then left undisturbed for 20 min, allowing the flocs to settle. After settling, samples of 15 ml were collected from approximately 5 cm beneath the surface and measured for pH and remaining turbidity. The setup of the jar tests can be seen in Figure 12.



Figure 12: Setup of the jartest used for coagulation and precipitation.

Preliminary measurements from the coagulation experiments were used to determine a minimal dose for efficient turbidity reduction. Three different doses in this range was then selected, and the experiments were repeated and left undisturbed to settle for 2 hours. Turbidity was measured after 20, 40, 60 and 120 minutes. Samples for metal analysis were extracted after 60 and 120 minutes. The samples were prepared for total metal content (unfiltered) and filtered through a 0.45  $\mu$ m filter using a plastic syringe for the dissolved fraction.

After the full 120 minutes sedimentation and after the samples for metal analysis were taken, the settled particles were resuspended by stirring for approximately 30 seconds. Depending on coagulant dose, approximately 100 ml of the sample was filtered and sampled for suspended solids.

To evaluate the potential or effect of additional pH adjustment, the same three coagulant doses were repeated with pH adjusted to 9. NaOH in 1M solution was injected into each sample to reach the desired pH. The coagulation experiments were then repeated before pH and turbidity was again measured.

# 6.6 Chemical precipitation

The precipitation experiments were conducted with the goal to find optimum precipitation conditions. Two different precipitants were used, Metalsorb PCZ and hydrated sodium sulfide (Na<sub>2</sub>S x H<sub>2</sub>O). In order to determine a range of doses, initial experiments were carried out on tap water injected with a known metal content of 800 ug Cu/L. The same dose was then tested on wash water from the Nestunnel, where the metal contents was unknown. Although the tunnel wash water was assumed to be very polluted, 800 ug Cu/L is a relatively high metal concentration to assume. For this reason, the observed results were compared to findings in literature in order to determine a range of doses (Garshol et al. 2016, Marchioretto et al. 2002). It was determined to move forwards with doses corresponding to metal concentrations in the range of 10 – 10000 ug Me/L, to include both higher concentrations as found in the literature as well as lower concentrations that would be more reasonable to assume.

Prior to the precipitation experiments, the wash water was filtered through a 1.0  $\mu$ m filter in order to remove particles. To resuspend settled particles, each can containing wash water was shaken before filtration. Representative samples from each can as well as from each batch of filtered water was taken to measure pH and unfiltered and filtered turbidity. Precipitation with sodium sulfide needs to take place in neutral or basic pH ranges to prevent the formation of H<sub>2</sub>S, however the investigated water had an initial pH which was suited for this purpose and did therefore not require additional adjustments.

Metalsorb PCZ comes as a liquid that could be directly dosed. The sodium sulfide was in hydrated form and the desired concentrations were relatively low, hence stock solutions of 0.1 g Na<sub>2</sub>S/L and 10 g Na<sub>2</sub>S/L were prepared and used for the smaller and larger doses respectively to ensure precision when dosing.

The precipitation experiments were carried out using standard jar tests in 1L plexiglass beakers using a Platypus jar tester apparatus. Initial rapid mixing was preformed at 100 rpm followed by a slow mixing at 25 rpm for 30 minutes. The precipitant was injected to the sample right after initializing rapid mixing. 25 ml of sample were collected from approximately 5 cm beneath the surface and measured for pH and remaining turbidity. Additionally, 15 ml of sample was collected for analysis of total metal (unfiltered) and filtered through a 0.45  $\mu$ m filter. The suspension was then left undisturbed for 30 minutes, allowing the precipitate to settle. After 30 minutes, new measurements of turbidity was taken.

# 6.7 Metal analyses

The samples were analyzed for concentrations of Al, As, Cr, Cu, Fe, Mn, Ni, Pb, Sb and Zn through ICP-MS. Unfiltered samples and samples filtered through a 0,45  $\mu$ m filter was analyzed. This was done to obtain information on both total metal contents and the dissolved fraction respectively. The instrumental analysis was conducted by Anica Simic at the department of chemistry at NTNU.

# 6.7.1 Decomposition of samples using UltraClave

Prior to the metal analyses, unfiltered samples was decomposed using UltraClave. Precisely 4.0 ml of sample was measured using a pipette and injected with precisely 2.0 ml concentrated HNO3. The samples was turned 3 times prior to extraction to ensure a well-mixed sample. Two samples containing reference material and two blank samples containing only 2 ml of HNO3 was also included to evaluate precision. The samples was then put in a baseload consisting of 300 ml mq-water, 30 ml H2O2 and 2 ml H2SO4 in the UltraClave and was digested for approximately 3 hours at 40 °C. Prior to decomposition, the UltraClave chamber was pressurized at 50 bar using Nitrogen gas. After decomposition, the samples were diluted to approximately 48.8 g using mq-water.

# 7 Results and discussion

# 7.1 Raw water quality

# 7.1.1 Water quality parameters

Water quality parameters as they were measured in the wash water from Vålerenga and Grilstad respectively are presented in Table 8. The values are averaged based on measurements taken from each single can used in the experiments and are presented with their respective errors. The raw data measurements are listed in Appendices A.6 and A.7.

Parameter	pН	Turbidity	Conductivity	
		[NTU]	[ms/cm]	[mg/L]
Vålerenga (average)	7.48	490	13.86	24.22
Vålerenga (standard deviation)	0.045	1.70	0.017	0.17
Grilstad (average)	7.91	617	3.05	5.82
Grilstad (standard deviation)	0.046	2.94	0.014	0.20

 Table 8: Measured raw water quality

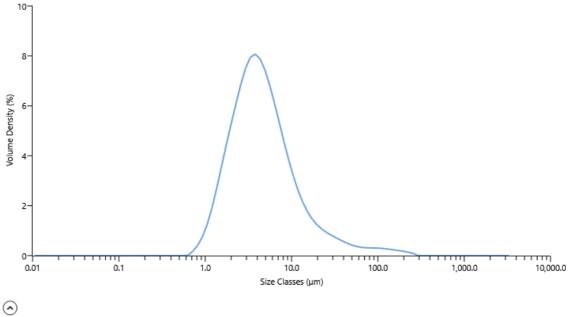
PH in tunnel wash water is typically in the range 7-9. In (Garshol et al. 2016) values in the range 7.3-8.9 are reported or 7.3-9.3 in (Meland 2010). Here, both washes are measured to a pH <8, in other words in the lower tier of what is typically found. The conductivity in the wash water sampled from Vålerenga is perceivably higher than that of Grilstad. This indicates a higher ionic content in the water. Relative to typical Norwegian surface water, both values are however high. Conductivity measured in representative Norwegian surface waters is normally in the range of >5 ms/m, which would indicate that both wash waters are likely to contain elevated metal concentrations and need to be treated accordingly.

# 7.1.2 Particle size distribution

The turbidity measured in the Vålerenga water is lower but the conductivity is higher than that of Grilstad, which indicates a larger proportion of fines or dissolved pollutants. This is supported by the particle size distribution, in which the particles of the Vålerenga water can be clearly seen to be finer.

Particle size distribution for Vålerenga is presented in Figure 47 and for Grilstad in Figure 48. The particle size distributions shows that the majority of particles contained in both wash waters are in the range 1.0-30.0  $\mu$ m. The 10-, 50- and 90-percentile fractions are presented in Table 9. The measurements for particle size distributions were conducted 3 times. The results presented here are the median results, whereas results from the full measurement series are listed in Appendix A.5 Generally, the particles in the wash water sampled at Vålerenga appear to be somewhat finer compared to Grilstad. For the Vålerenga water, the volume density reaches its peak at diameter 4.26  $\mu$ m, whereas the peak for the Grilstad water is reached at almost double this value at 6.92  $\mu$ m.

As presented in section 5.2, a significant proportion of pollutants contained in the wash water can be assumed to be particle bound. Sedimentation can be assumed to have a rather good effect on the larger particles contained in the Grilstad wash water, whereas the percentage of particles  $> 1\mu$ m contained in the Vålerenga water should be further accounted for. These are categorized as colloids, and will likely not be removed by sedimentation alone.



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Figure 13: Particle size distribution - Vålerenga.

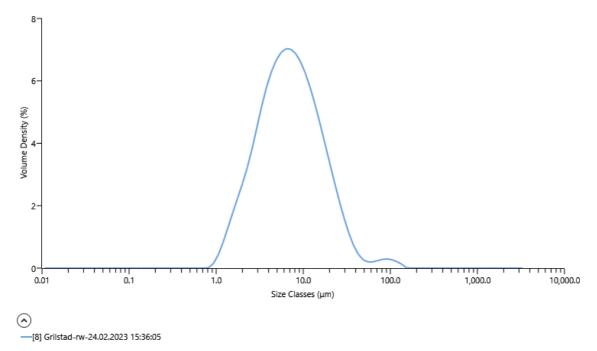


Figure 14: Particle size distribution - Grilstad.

The 10-, 50- and 90-percentile fractions are presented in full in Table 9. These represent the filter size that 10, 50 and 90 % of the particles would pass through. D50 represents the peak of the volume density distribution. The 10%-percentiles of 1.72 and 2.37  $\mu$ m (Vålerenga and Grilstad respectively) represent a significant proportion of the contained particles. Although these are not necessarily collodial particles, they still represent a fraction which would require a substantially increased residence time in order to sediment.

#### 7 Results and discussion

Sample Name	Dx (10) ( $\mu m$ )	Dx (50) ( $\mu m$ )	Dx (90) ( $\mu m$ )
Vålerenga 1	1.71	4.24	14.9
Vålerenga 2	1.71	4.22	15.3
Vålerenga 3	1.73	4.33	16.3
Vålerenga mean	1.72	4.26	15.5
Vålerenga stdev	0.009	0.048	0.589
Grilstad 1	2.35	6.85	19.9
Grilstad 2	2.36	6.86	20.4
Grilstad 3	2.40	7.04	22.2
Grilstad mean	2.37	6.92	20.83
Grilstad stdev	0.022	0.087	0. 989

Table 9: Particle size fractions

#### 7.1.3 TSS

The wash water sampled from Grilstad contained a larger amount of suspended solids compared to that from Vålerenga. The inorganic fraction was however higher in the water from Vålerenga. In the raw water without addition of any coagulant the inorganic fraction was 31 % in Grilstad and twice as high at 62 % in Vålerenga. The addition of a coagulant did however appear to have little effect on the inorganic fraction. With the injection of PAX the inorganic fraction was reduced slightly, but remained relatively stable at around 28 % and 40 % in Grilstad and Vålerenga respectively, although the coagulant dose was increased. The situation when injected with PIX is similar, however the decrease is smaller. The fractions remain relatively stable at around 30% and 50% respectively. The total suspended solids and combustion residue from both washes are presented in Figures 15 and 16 respectively.

TSS is closely related to turbidity, but the sizes are not directly comparable. For both samples, an increase in TSS volumes can be observed for increasing coagulant doses. This could be attributed to the fact that not all contributors to turbidity are possible to weigh, such as for example color. Thus, a reduction in color could entail a precievable turbidity reduction, whereas the TSS volume would still be the same. Thus, it is reasonable to assume that the measured values would significantly decrease over time as the particles have had sufficient time to settle.

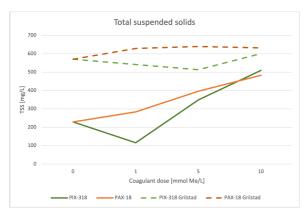


Figure 15: Total suspended solids - both washes

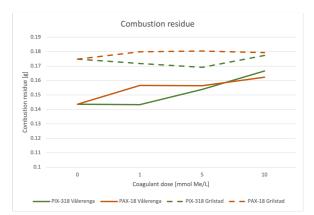


Figure 16: Combustion residue - both washes

# 7.2 Particle removal

#### 7.2.1 Residual turbidity

The coagulation experiments are subject to a distinct pattern. In general, the lowest residual turbidity was achieved for the highest injected coagulant doses. Although the residual turbidity generally was measured to somewhat higher values for the doses in the middle of the tested range (1, 5 and partly also 10 mmol Me/L), these also produced water with a significantly reduced turbidity. For both waters and both coagulants, the lowest coagulant doses produced little to no reduction in turbidity. Residual turbidity as a function of coagulant dose is illustrated in Figures 17 and 18 for the Vålerenga tunnel and the Grilstad tunnel respectively. Raw data from the measurements is available in Appendices A.6 and A.7 respectively.

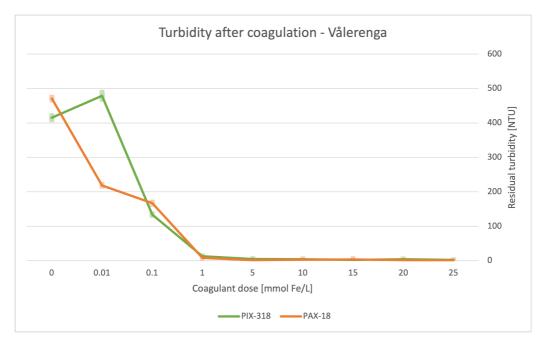


Figure 17: Residual turbidity after chemical coagulation (all doses) - Vålerenga.

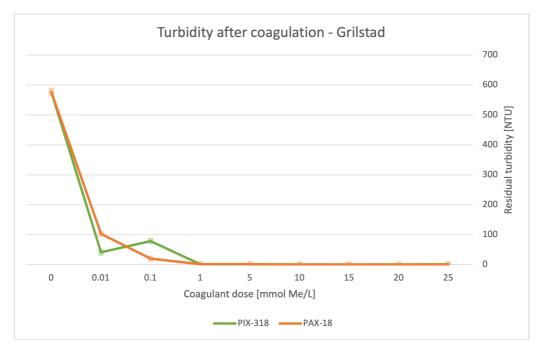


Figure 18: Residual turbidity after chemical coagulation (all doses) - Grilstad.

At a closer review, the largest reduction in turbidity is seen at 25 mmol Me/L for both PIX and PAX in the Vålerenga samples. The additional decrease compared to that achieved by smaller doses is however slim. PAX appears to produce water with a generally lower turbidity. The difference compared to PIX is perceptible, but both coagulants produce water with a residual turbidity < 5 NTU at doses exceeding 5 mmol Me/L. The distinct decrease in residual turbidity between doses 1 and 5 mmol Me/L is however interessting to make note of and will be further discussed in section 7.2.4. The residual turbidities as they were measured for coagulant dose > 1 mmol Me/L can be seen in Figure 19.

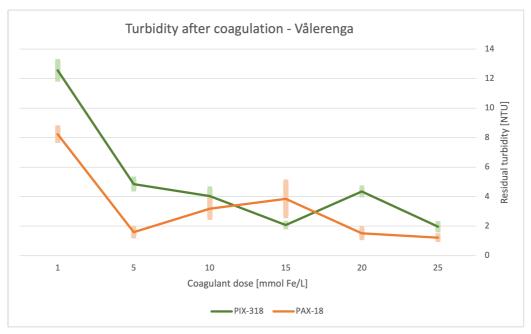


Figure 19: Residual turbidity after chemical coagulation (coagulant dose > 1 mmol Me/L) - Vålerenga.

The samples from Grilstad are generally subject to the same tendencies. The largest reduction in turbidity is seen at a slightly lower dose, around 15 and 20 mmol Me/L, for both coagulants, whereas an increase in dose to 25 mmol Me/L appears to result in a slight increase. The difference is however slim as the measured values are all < 2.5 NTU. For both PIX and PAX, all doses > 1 mmol Me/L result in a significant turbidity reduction and a residual turbidity of < 2.5 NTU. The residual turbidity for coagulant dose > 1 mmol Me/L is presented in Figure 20.

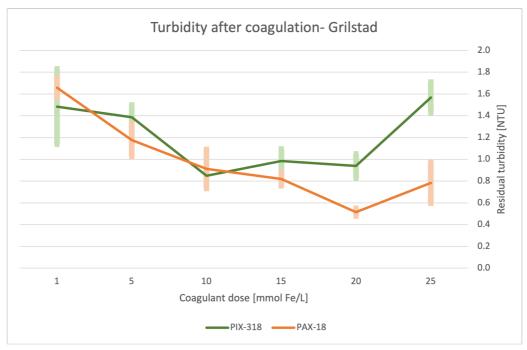


Figure 20: Residual turbidity after chemical coagulation (coagulant dose > 1 mmol Me/L) - Grilstad.

#### 7.2.2 Time series

Figures 21 and 22 represent the timewise turbidity reduction of an untreated sample compared to samples injected with a coagulant. The turbidity decrease over time is evident when considering the untreated sample, however the residual turbidity is still relatively high after 120 minutes of sedimentation.

It is evident that introducing a coagulant to the wash water induces a much more efficient sedimentation. It should however also be noted that the samples injected with a coagulant reach a much more level quality after sedimenting for 120 minutes. Relative to the Vålerenga water, the Grilstad water contained generally larger particles. These sediment more rapidly and thus the sedimentation in this sample appears to be more effective. Natural distinctions do however occur in tunnel wash water. As clearly illustrated by the Vålerenga sample where the turbidity is still > 300 NTU after 120 minutes of sedimentation, this is nowhere near a sufficient residence time. Compared to the untreated sample, all of the treated samples does however result in water with virtually no residual turbidity after 120 minutes.

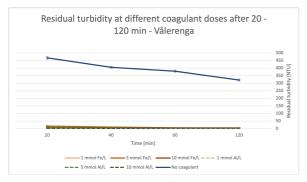


Figure 21: Turbidity reduction 0-120 minutes after coagulation compared to sample without coagulant - Vålerenga

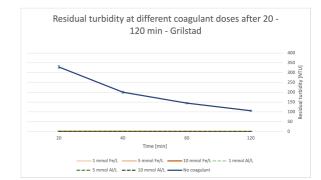


Figure 22: Turbidity reduction 0-120 minutes after coagulation compared to sample without coagulant - Grilstad

Under closer review, a further reduction in turbidity can be observed as the residence time is increased. When injected with PIX, a sedimentation time of 60 minutes is necessary to achieve water with a residual turbidity < 5 NTU in the samples from Vålerenga. In the Grilstad samples the turbidity after 20 minutes was already < 5 NTU, however a further decrease can still be observed within the first hour.

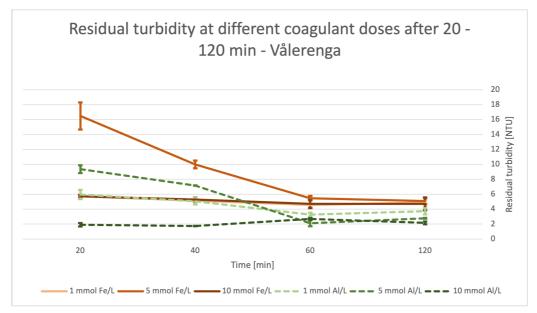


Figure 23: Turbidity reduction 0-120 minutes after coagulation - Vålerenga

Compared to 1 and 5 mmol Me/L, the samples injected with 10 mmol Me/L behaved relatively stable over time. Based on the developed time series, only a small additional reduction in turbidity could be achieved from sedimenting for 120 minutes compared to 20 minutes. For 1 and 5 mmol Me/L, a certain effect could however be observed within the first 60 minutes. This indicates smaller flocs at the smaller doses and is further supported by visual observations during the experiments as will be further described in section 7.2.5. Further, this should be seen in coherence with residual turbidity achieved from the different doses. After the initial 20 minutes of sedimentation 10 mmol Me/L appears to be generally favorable compared to the lower doses, whereas the differences in residual turbidity produced by the different doses are close to eliminated after 120 minutes of sedimentation.

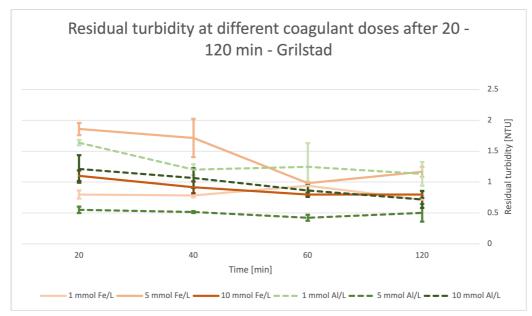


Figure 24: Turbidity reduction 0-120 minutes after coagulation - Grilstad

Relative to todays practice, both 20 and 120 minutes of sedimentation represent a significant decrease in required residence time. However, 120 minutes as opposed to 20 minutes also enables water with a low (<10 NTU) residual turbidity to be produced from a relatively low coagulant dose.

### 7.2.3 pH

For both coagulants and both tunnels, the pH post coagulation was relatively stable. This is generally favorable with regards to the mobility of metals, as described in Section 5.6. The observed pH drop caused by adding the coagulant was comparable for both coagulants, but more distinct in the water sampled at Grilstad. Injecting PIX as 25 mmol Fe/L resulted in an average reduction of 0.54 in the Vålerenga sample and 0.95 in the Grilstad sample. As the coagulant dose was decreased, the corresponding reduction in pH decreased in a close to linear manner accordingly. Injecting PIX as 0.01 mmol Fe/L produced water with a pH reduction of 0.015 and 0.025 for Vålerenga and Grilstad respectively. With PAX the comparable measurements read 0.25 and 1.17 when injecting 25 mmol AL/L and 0.035 and 0.025 for the smallest dose. The observed relationship was again close to linear. All pH measurements from Vålerenga and Grilstad are listed in Appendices A.6 and A.7 respectively.

Adjustment of pH generally appears to have been more favorable to the smaller doses. This corresponds well with the solubility diagram as presented in section 5.6, as higher coagulant doses are usually required in less optimum pH areas.

Turbidity after sedimentation as a function of coagulant dose with and without pH adjustment is presented in Figures 25 and 26. Without any pH adjustment, a lower residual turbidity was achieved for the highest doses whereas the performance of the lowest dose perceptibly improved as the initial pH was adjusted to 9. Adjusting the pH had minor effects on the residual turbidity achieved by the higher dose. The samples with adjusted pH seems to have reached a generally lower residual turbidity, with the exeption of 10 mmol Me/L in the Grilstad sample. Still, it is a distinct tendency that turbidity to a greater extent has been removed at initial pH 9.

# 7 Results and discussion

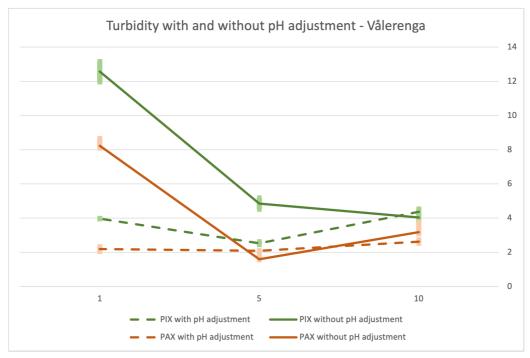


Figure 25: Residual turbidity with and without pH adjustment - Vålerenga

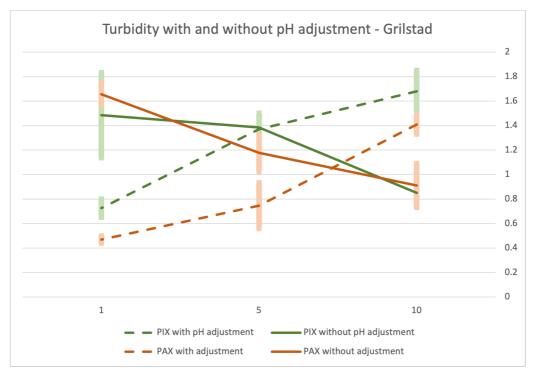


Figure 26: Residual turbidity with and without pH adjustment - Grilstad

### 7.2.4 Zeta potential

Overall, the zeta potential in the samples injected with PAX reach a lower absolute value at higher doses than those injected with PIX. This implies that the coagulation is more effective at lower doses when injected with PIX, and at higher doses when injected with PAX. The zeta potential measurements as a function of coagulant dose are presented in Figures 27 and 28. Raw data from the measurements are available in Appendices A.6 and A.7.

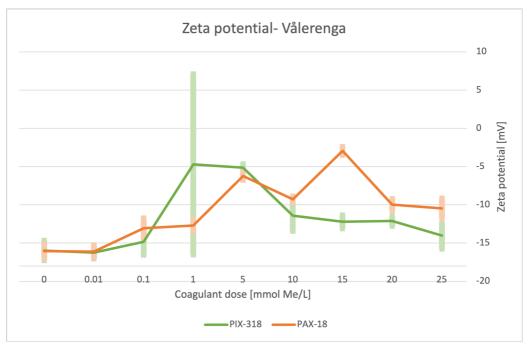


Figure 27: Zeta potential - Vålerenga

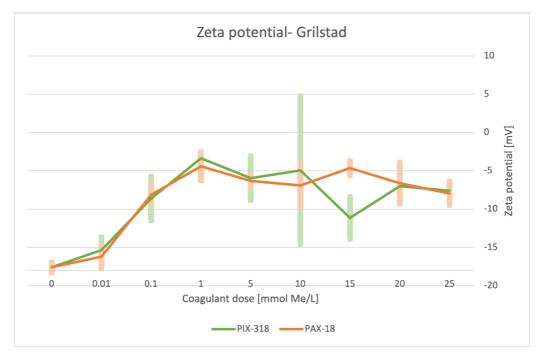


Figure 28: Zeta potential - Grilstad

For both washes, the zeta potential is low for the lowest coagulant doses, at around -15 mV for both coagulants. As the coagulant dose increases, the increased turbidity reduction for doses >1 mmol Me/L as discussed in section 7.2.1 can be clearly recognized in the zeta potential. For both coagulants, the zeta potential reaches its lowest absolute value in the range 1-5 mmol Me/L and partly also when injected with 10 mmol Me/L. With basis in the solubility diagrams for alum and iron as presented in Section 5.6, this is the interval where sweep coagulation should occur. Higher doses still induce formation of the insoluble precipitation products  $Fe^{3+}$  (PIX) and  $Al^{3+}$  (PAX), which would explain the significant reduction in turbidity. The smaller doses (<1 mmol Me/L) fall outside of normal operating conditions, which is reflected in the low zeta potential values and higher residual turbidity as seen for these doses.

One exemption to make note of is 0.1 mmol Al/L injected into the wash water from Grilstad. This water had a slightly higher initial pH than that of Vålerenga, leaving 0.1 mmol Al/L to be a coagulant dose theoretically included in the area giving optimal conditions for sweep. However, due to unstable measurements, the pH meter in use was changed after measuring pH of the samples injected with 0.1 mmol Al/L. The error margin of this particular value is therefore larger than it appears. Since the residual turbidity is still rather high and since the interval for optimal sweep coagulation is rather narrow, this has likely not transpired. The turbidities measured for this coagulant dose in the Grilstad water are still perceivably lower than those at Vålerenga and the tendency is similar for 0.01 mmol Al/L. Generally, a higher pH appears to be favorable for the lower alum-doses.

Further, the zeta potential as discussed above is with the exemption of 15 mmol Al/L measured in the Vålerenga sample. Neither the turbidity measurements nor the metal analysis provides a reasonable explanation for this particular measurement being deviant from the trend as it was discussed. Hence, this particular measurement is accounted for as a probable outlier.

### 7.2.5 Observations

Overall, floc formation could be observed in most of the samples after various amounts of time. In the samples injected with PIX, flocs were observable the quickest when injected with 25 mmol Fe/L (after about 1-2 minutes of slow stirring). As the coagulant dose decreased, the duration until the first flocs were visible increased. At 10 mmol Fe/L, flocs were observable first after about 5 minutes of slow stirring. The flocs also appeared to be decreasing in size as the coagulant dose decreased, which is well in coherence with them not being as visible. In the samples injected with PAX, flocs were observable the quickest when injected with doses 20 and 15 mmol Al/L, and this occurred already during rapid mixing. As the dose was increased, the flocs once again appeared to be decreasing in size.

The flocs formed when injected with PAX appeared to be generally larger than those from PIX, which in turn resulted in a thicker layer of sedimented sludge. This is also mirrored in the turbidity reduction, as coagulation with PAX in both cases resulted in water with a lower residual turbidity. Due to the flocs formed by coagulation with PAX being larger, and thus also presumably heavier, a distinct pattern was observable in the sedimentation for most of the samples. This is assumably due to gravity, and some of the larger flocs sedimenting already during the slow mixing. Overall for both coagulants, the thickness of the layer of sedimented particles increased with an increasing coagulant dose.

In the samples injected with PIX as concentrations in the middle of the investigated dose interval (<20 mmol Fe/L and >1 mmol Fe/L), particles were also settled as foam on the water surface. Formation of a foam layer could also be observed in the samples injected with PAX, however in a much more narrow interval (<10 mmol Al/L, >1 mmol Al/L). The amount of foam formed generally increase as the coagulant dose decreased for both coagulants until the point where the dose was too low to achieve any distinct coagulation.

### 7.2.6 Metal removal through coagulation

The samples were analyzed for contents of Al, As, Cr, Fe, Mn, Ni, Pb, Sb and Zn. These 10 metals were chosen as previous research implies that they are commonly present in tunnel wash water. Certain substances like Pb are typically present at rather low concentrations not exceeding EQS-values. As previously stated, substances like Zn and Cu have however often proven to be problematic in the past. Further, it is necessary to observe concentrations of Al and Fe in the samples to evaluate to what extent the water quality is affected or possibly degraded from injecting the coagulants.

Overall relatively high removal rates (>90%) were achieved through both coagulants. The lowest residual metal concentrations were in general achieved through coagulation with PIX, with respect to both dissolved and total metal fractions. The greatest percentwise reduction was achieved in the dissolved fraction. The poorest reduction in dissolved concentration was observed at >70% for Ni and >40% for Mn, whereas the total concentration in comparison was measured to increase with around 10% for Mn and 5% for Ni. A significant reduction in both Mn and Ni was however produced through chemical precipitation, which will be further addressed in Section 7.3.4. The observed increase in total concentrations could as described in section 5.6 be due to increased ion mobility, consequent to the acidic character of the coagulant resulting in a drop in pH.

The sample from Vålerenga containing 10 mmol Fe/L was measured to unrealistically high values of virtually all the metals tested for. It is assumed that this particular sample was contaminated prior to analysis, most probably due to a procedural error. This sample is therefore exempt from the further discussion and presented results.

The concentration of Pb was below the detection limit in most of the samples. This corresponds well with previous research, where Pb rarely has been observed to be present at concentrations that are problematic with regards to the EQS-values (14  $\mu$ g/L). For this reason, Pb will not be further discussed.

The initial concentration of As was measured to 1.94  $\mu$ g/L and 1.775  $\mu$ g/L in the Vålerenga and Grilstad samples respectively. This is already well below the MAC-EQS value at 8.5  $\mu$ g/L. The concentration of As post coagulation was measured to be < 1  $\mu$ g/L in all samples in both the dissolved and total fraction. Consequently a good reduction is achieved through the treatment, but as the initial concentrations are not problematic with respect to discharge this will not be further discussed.

Sb was measured present at 14.32  $\mu$ g/L and 6.34  $\mu$ g/L in the Vålerenga and Grilstad samples respectively. For Sb no EQS value is defined, but comparable values from the UK implies that concentrations no higher than  $\mu$ g/L should be present to prevent unwanted toxic effects. All concentrations of Sb post coagulation treatment were measured well below 5  $\mu$ g/L, and will therefore not be further discussed. It should however be noted that the samples from Vålerenga generally showed higher concentrations relative to Grilstad, and was measured at <  $\mu$ g/L. This could be of concern if the initial concentrations were higher.

Cr was measured at initial concentrations of 32.2  $\mu$ g/L and 38.84  $\mu$ g/L in the Vålerenga and Grilstad samples respectively. After having undergone coagulation, Cr is almost completely removed in all samples, measuring dissolved concentrations at <0.6  $\mu$ g/L and total concentrations <1  $\mu$ g/L. This implies that Cr has been strictly associated with particles. Although the initial concentrations measured in both waters exceeds the EQS-values (3.4  $\mu$ g/L), the good reduction as seen post treatment indicates that Cr is of little concern in these samples. It should however be noted that higher concentrations have been reported in literature (e.g. 346  $\mu$ g/L in (Meland et al. 2010)). When accounting for dilution in the recipient (assumed to be equivalent to a factor 10), it is however reasonable to assume that Cr in most cases would be sufficiently reduced through the treatment. In this case, percentwise removals (dissolved fraction) of >96 % was achieved in all samples, which would represent acceptable concentrations even if the initial concentrations were more on the higher end of what is reported in literature.

The concentrations of Fe that was measured were unrealistically high and without any particular pattern in the samples injected with both PIX and PAX. Although the concentrations were high, the RSD errors were also high implying unstable measurements. The concentrations were for this reason measured repeatedly, but still without any distinct pattern or reasonable concentrations. The measurements from each duplicate were also strongly differing, and the RSD errors were still high. For this reason, it is difficult to conclude on how well Fe has actually been removed from the samples.

Initial Cu concentrations were measured at 102.62  $\mu$ g/L and 44.96  $\mu$ g/L in the samples from Vålerenga and Grilstad respectively. This is comparable to what is reported in the literature, although on the lower end of the scale. Cu is present in the tunnel wash water through a variety of different pollution sources, and thus often source of concern. As seen in e.g. the Bjørnegårdstunnel as presented in section 5.8.1, Cu can be present in elevated values in severly polluted samples, which could result in toxic effects in the recipient if not treated accordingly. The EQS-value for Cu is 7.8  $\mu$ g/L, which the initial concentrations in both waters exceed.

Cu concentrations post coagulation were measured to be  $< 10 \ \mu g/L$  for all coagulant doses, which suggests an overall good reduction. When accounting for dilution in the recipient, these values are acceptable relative to the EQS-values. Concentrations post treatment are shown in Figures 29 and 30. Lower concentrations of the dissolved fraction can be observed, which suggests that the treatment has had the desired effect on the dissolved fraction. This is however with the exeption of the sample from Vålerenga containing 10 mmol Fe/L, which as previously stated is assumed to have been contaminated. For illustration, it is still included in the plots below. The good reduction in the sample not injected with a coagulant should also be noted. This could suggest that theinitial Cu has been strongly associated with particles.

In general, a good percentwise reduction of the dissolved fraction was achieved through coagulation. This is a promising result, as the dissolved fraction is assumed to be more bioavailable than the particulate fraction and thus is desireable to reduce. The percentwise reduction in Cu is shown in Figures 31 and 32.

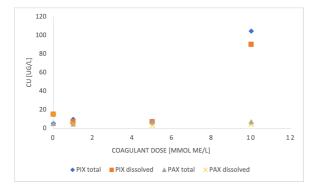
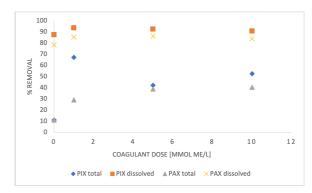


Figure 29: Cu concentrations post coagulation - Vålerenga



**Figure 30:** Percentwise reduction in Cu post coagulation - Vålerenga

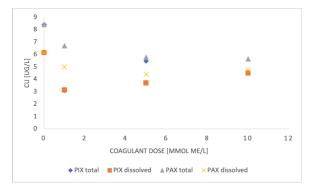


Figure 31: Cu concentrations post coagulation - Grilstad

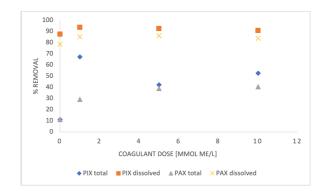


Figure 32: Percentwise reduction in Cu post coagulation - Grilstad

Relative to what is usually reported in literature, low concentrations of Zn were measured in the samples at 418.4  $\mu$ g/L and 374.8  $\mu$ g/L in Vålerenga and Grilstad respectively. As the EQS value for Zn is 11  $\mu$ g/L, Zn still represent a significant treatment need in the samples. Compared with other investigated metals, a sparse reduction (at worst >70 %) is achieved through the treatment. Contradictory to e.g. Cu, the residual concentrations are measured to be the highest within the dissolved fraction and when injected with PIX. This implies that Zn to a higher extent has been present as colloids or solutes, and would suggest that coagulation with PIX is not the most favorable treatment option to reduce this fraction. This is however well corresponding with what has been reported in previous studies. Among other metals, Zn is generally assumed to be more associated with the LLM fraction. For this reason Zn often poses as problematic and desirable to reduce when treating tunnel wash water, as this fraction is assumed to be more bioavailable compared to particulate fractions.

In general, it appears that treatment with PAX has a better effect on reducing both total and dissolved Zn concentrations. All samples injected with PAX achieve residual concentrations < 66  $\mu$ g/L, which is acceptable with regards to the Zn EQS-value of 110  $\mu$ g/L. However, according to Rathnaweera et. al. (2019) the discharge limit for Zn is 50  $\mu$ g/L suggesting that also treatment with PAX cannot necessarily produce a sufficient to reduction in Zn. This concentration is only met when injected with 1 and 5 mmol Al/L, and for none of the samples injected with PIX. The concentrations post treatment can be seen in Figures 33 and 34.

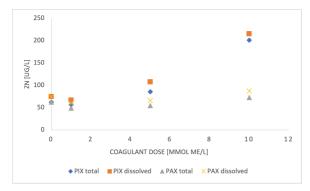


Figure 33: Zn concentrations post coagulation - Vålerenga

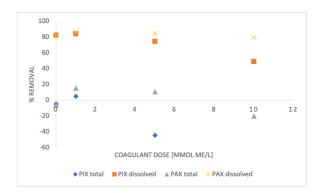


Figure 34: Percentwise reduction in Zn post coagulation - Vålerenga

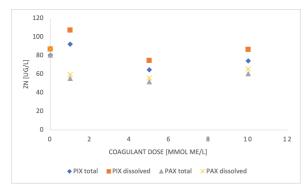


Figure 35: Zn concentrations post coagulation - Grilstad

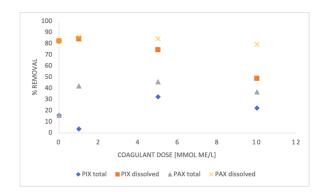
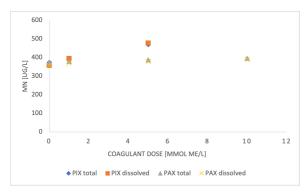


Figure 36: Percentwise reduction in Zn post coagulation - Grilstad

The concentrations of Mn that were measured are comparable to those found in the literature, at 612.6  $\mu$ g/L and 322.5  $\mu$ g/L in Vålerenga and Grilstad respectively. For Mn no EQS value is defined, but comparable values of 5  $\mu$ g/L suggest that Mn represent a significant treatment need. The best percentwise reduction rates were achieved for the dissolved fraction, whereas the total fraction was increased post treatment. This for one implies that Mn was initially present as colloids or solutes and that coagulation to some extent is efficient in reducing these, but also that the coagulation is not an effective treatment in sufficiently reducing the total concentration.



**Figure 37:** Mn concentrations post coagulation - Vålerenga

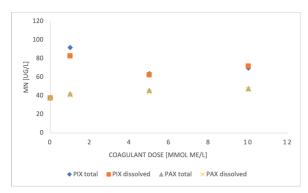


Figure 39: Mn concentrations post coagulation - Grilstad

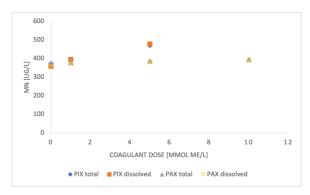


Figure 38: Percentwise reduction in Mn post coagulation - Vålerenga

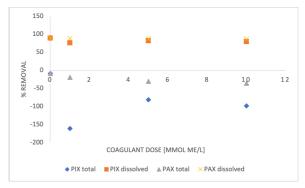
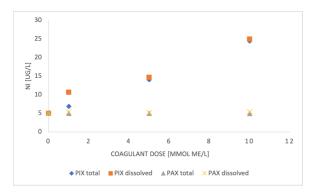


Figure 40: Percentwise reduction in Mn post coagulation - Grilstad

#### 7 Results and discussion

The concentrations measured of Ni in the samples were high compared to those previously found in literature (e.g. (Meland et al. 2010)), with dissolved concentrations of 18.6  $\mu$ g/L and 20.7  $\mu$ g/L in Vålerenga and Grilstad respectively. Post coagulation the dissolved fraction was however significantly reduced as can be seen in figures 42 and 44. With regards to the recommended EQS value of 34  $\mu$ g/L, both coagulants provides water with a sufficiently low Ni content. However, as can be clearly seen in figures 41 and 43, PAX is by far more effective. This is interessting to consider in combination with the measured Zn concentrations, as Zn and Ni often are observed to behave rather similar. Both Zn and Ni are generally assumed to be affiliated with the LMM fraction to a greater extent than metals such as Cu, Cr or Al, where PIX visibly has provided a superior removal. This indicates that PAX could be favorable for metals which to a higher degree are present as solutes, whereas PIX is more efficient in removing metals affiliated with particles or colloids.



**Figure 41:** Ni concentrations post coagulation - Vålerenga

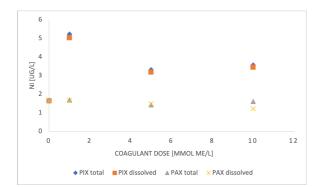


Figure 43: Ni concentrations post coagulation - Grilstad

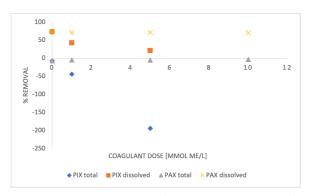


Figure 42: Percentwise reduction in Ni post coagulation - Vålerenga

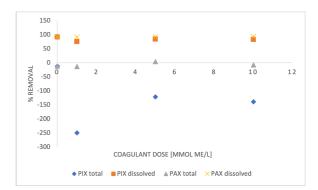


Figure 44: Percentwise reduction in Ni post coagulation - Grilstad

All values as presented above are measured in samples taken 120 minutes post coagulation. Metal contents were also measured after 60 minutes, however a significant reduction could only be observed when increasing the sedimentation time to 120 minutes for the metals Cr, Zn and Fe. Following the general trend, the concentrations measured after 120 minutes was however comparable to those after 60 minutes, indicating that a rather short sedimentation time would be sufficient to achieve acceptable metal contents in combination with chemical coagulation. For this reason, the measurements taken after 60 minutes are not further discussed. Given the amount of measurements that were made and the size of the resulting data series, it was not deemed reasonable to present the raw data in any other format than excel. The raw data is therefore not directly attached to the thesis, but is available in the separate excel file *metal-results.xls*.

# 7.3 Chemical precipitation

### 7.3.1 Pre-experiments

Preliminary experiments with chemical precipitation were conducted on tap water spiked with a known metal content of 800  $\mu$ g Cu/L and on tunnel wash water sampled from the Nestunnel with both Metalsorb PCZ and sodium sulfide. The dose of 16.25  $\mu$ l Metalsorb/L resulted in visible precipitation when injected to the tap water spiked with copper. Also when the same dose was tested on tunnel wash water and was doubled, visible precipitates could be seen. The residual turbidity was significantly lower for the highest dose of Metalsorb PCZ, although both doses resulted in visible precipitates also for the tunnel wash water.

## 7.3.2 Precipitation with Metalsorb PCZ

Overall, precipitation with Metalsorb PCZ showed promising results. Visible precipitates could be observed after some extent of time for all the three smaller doses  $<32.50 \ \mu$ l/L. The highest dose appears to have been too high to achieve an efficient precipitation. Leaving the solution to sediment for 30 minutes appears to have had only a marginal effect on the residual turbidity. This corresponds with visual observations done during the jar tests, the precipitates appeared to be large and heavy and a significant part of them had already sedimented during the slow stirring. The samples from both washwaters were generally subject to the same tendencies, and the measurements were conducted with relatively small standard deviations in all the measurements.

Turbidity does not provide a sufficient measure on effect of precipitation. The samples were filtered prior to injection of precipitant, and hence the turbidity is on the contrary expected to increase as the dissolved pollutants are precipitated. For this reason, the smaller doses are still described as more successful even though the water produced from these doses were with a higher turbidity after sedimentation.

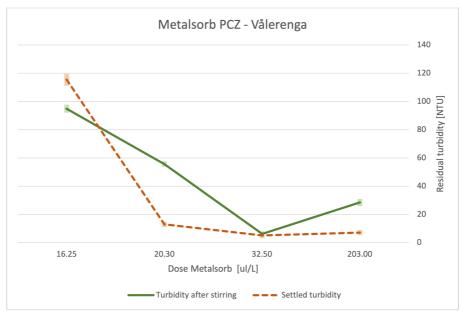


Figure 45: Residual turbidity after precipitation with Metalsorb PCZ - Vålerenga.

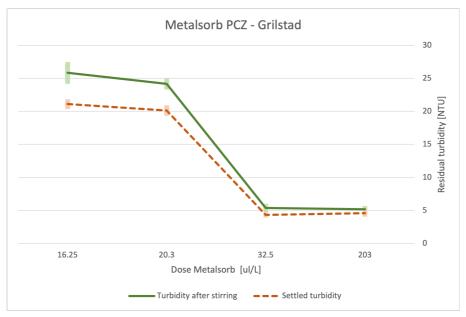


Figure 46: Residual turbidity after precipitation with Metalsorb  $\mathrm{PCZ}$  - Grilstad.

#### 7.3.3 Precipitation with sodium sulfide

In all cases, introducing sodium sulfide into the solution resulted in an increase in turbidity after the stirring. The samples behaved rather different apart from this. For the Vålerenga wash water the lowest residual turbidity was achieved with the lowest sodium sulfide doses, whereas the Grilstad wash water was subject to an opposite trend. As with Metalsorb, leaving the samples to sediment for 30 minutes appears to have had only a marginal effect on the residual turbidity with an additional reduction of only a few NTU. This was the case at all measurement points and the error margins were generally low.

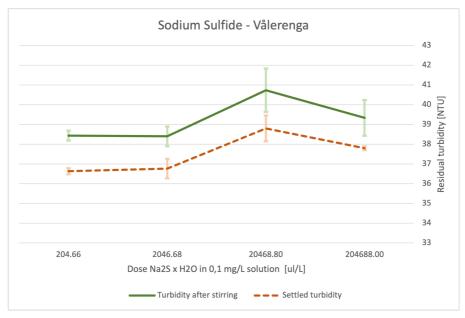
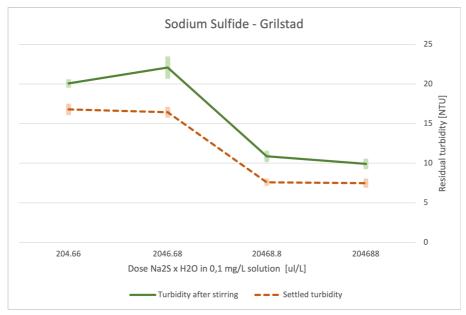


Figure 47: Residual turbidity after precipitation with sodium sulfide - Vålerenga.



 ${\bf Figure \ 48:} \ {\rm Residual \ turbidity \ after \ precipitation \ with \ sodium \ sulfide \ - \ Grilstad.$ 

### 7.3.4 Metal removal through precipitation

Relative to the metal contents measured post coagulation, the percentwise reduction rates achieved from precipitation with both Metalsorb PCZ and sodium sulfide was generally poorer. It should however be noted that for certain metals, such as Mn and Fe which was poorly reduced through coagulation alone, significantly higher reduction rates was achieved when implementing precipitation.

For Fe, all samples injected with sodium sulfide reached a reduction of >99% of the dissolved fraction. The reduction in the total fraction was however nowhere near comparable, but this could be attributed the fact that the water had been filtered prior to the actual precipitation experiments and thus that a good amount of the particle bound Fe had already been removed. After undergoing precipitation with sodium sulfide, the highest measured iron concentration was 162  $\mu$ g/L which is well below the recommended limit of 300  $\mu$ g/l. Relative to the coagulation experiments, the samples injected with Metalsorb PCZ also achieved a good reduction in dissolved Fe. At >98%, this was however still not as effective as sodium sulfide. Nevertheless, all samples was measured at <196  $\mu$ g Fe/L, which equally fulfills the recommended (dissolved) values for discharge.

With regards to removal of Mn, the greatest reduction was without doubt observed in the samples injected with Metalsorb PCZ. Excluding the sample from Vålerenga which was injected with 203 ul/L and where the reduction rate was measured to 54%, reduction rates >80% was measured in all the further samples. This corresponds to (dissolved) concentrations <105  $\mu$ g Mn/L, which is relatively low compared to what has previously been reported in the literature such as 180  $\mu$ g/L in (Meland et al. 2010).

For Al, the dissolved concentrations was measured up to 95  $\mu$ g/L when injected with Metalsorb PCZ. In the samples injected with sodium sulfide, concentrations up to 186  $\mu$ g/L was measured. For the metals Pb, Sb and Cr the initial concentrations were as described in section 7.2.6 sufficiently low for these metals not to be considered problematic for discharge.

Both total and dissolved concentrations of Ni was low after precipitation compared to those achieved by coagulation. When injected with Metalsorb PCZ, the highest residual (dissolved) contentration was measured at the highest dose (203  $\mu$ L/L) at 26.5  $\mu$ g/L and 1.7  $\mu$ g/L in the samples from Vålerenga and Grilstad respectively. For the lower doses and in the samples injected with sodium sulfide, concentrations <5.2  $\mu$ g/L were measured. This is similar to values found in the literature as presented in sections 5.2 and 5.7, and well below the recommended EQS value even when accounting for dilution in the recipient. The good reduction achieved through precipitation relative to that achieved through coagulation can likely be attributed to Ni generally being more present in dissolved form, and thus more difficult to remove through coagulation or similar technologies without aiding the formation of colloids that can later on be agglomerated into larger flocs.

#### 7.4 Optimalization of process

Although the lowest turbidities overall were achieved through the highest coagulant doses tested, doses in the middle of the range (1, 5 and 10 mmol Me/L) also showed promising results. Zeta potential measurements implies that the coagulation process itself is more efficient at lower doses when injected with PIX, which also corresponds well with previous reported research (Aghapour et al. 2016, Nyström et al. 2020) as presented in Section 5.6.

Overall, precipitation with Metalsorb PCZ showed promising results in comparison to sodium sulfide. For the smallest doses with Metalsorb, visible precipitates could be clearly observed after a short amount of time when injecting the precipitant. When considering the sulfide precipitation however, it is reasonable to conclude that the flocs were too small to properly settle and thus remained in the aqueous phase. This implies that processes such as coagulation could significantly aid the separation process, but would mean introducing the addition of yet another chemical into the treatment.

#### 7.4.1 Chemical consumption and toxicity

As seen in section 7.2.3, a higher pH value appears to be favorable for lower coagulant doses. Introducing pH adjustment into the treatment process thus enables a reduction in the coagulant dose itself but introduces other issues such as adding another chemical into the mix. Further, pH values as typically found in tunnel wash water are already relatively high (7-9) compared to what is usually seen in coagulation (>5). In other words, the margin to permissible discharge limits must be considered carefully. Another aspect is that higher pH ranges (>9-10) are potentially favorable for introducing toxic effects on microorganisms that are essential to degrade organic matter. Still, pH adjustment provides a desired element of control with respect to small changes in water quality. As clearly illustrated in e.g. section 6.1 or 7.2.2, tunnel wash water can be subject to distinct variations that to different extents can impact coagulation efficiency. In such, pH adjustment is a relatively simple control element with respect to reaching the desired effect also at lower coagulant doses.

Iron is likely to be present as trivalent ferric ( $Fe^{3+}$ ) under the conditions seen, which is assumed to be associated with toxic effects to a lesser degree than  $Fe^{2+}$ . Still, potential precipitates contributing to toxicity to a higher extent should be accounted for.

In (Garshol et al. 2016), treatment through precipitation is reported to have had little effect on the toxicity of tunnel wash water. In several cases the toxicity was increased after precipitation with sodium sulfide, which could be attributed to the presence of very fine sulfide flocs. The toxicity is related to several factors including metals, but also oil- and PAH-content among others. As the metal contents were still relatively high in the water samples injected with sodium sulfide, it is reasonable to assume that also oil- and PAH contents have not been sufficiently reduced and thus that the toxicity still is rather

high. Further, precipitation with Metalsorb PCZ as opposed to sodium sulfide eliminates the potential of having toxic by-products such as  $H_2S$  gas form. On the contrary, the excess of organic molecules has been observed to result in contamination of the effluent and an increased chemical oxygen demand.

#### 7.4.2 Robustness

A significant reduction in turbidity was achieved for both water samples, which implies that the process is not overly sensitive to small changes in water quality. The overall residual turbidity reached in the Grilstad samples is somewhat lower compared to Vålerenga, which could indicate some discrimination towards finer, lighter particles. On the contrary, this could also be result of insufficient sedimentation time as is supported by the developed time series.

Although the coagulation experiments appears to favor a higher pH at lower coagulant doses, the residual turbidity is still sufficiently reduced without any form of adjustment. The reduction in metal contents appears to be rather stable inspite of small changes in water quality parameters such as pH.

Regarding the precipitation experiments, Metalsorb PCZ appear to have induced an efficient precipitation process regardless to small changes in initial water quality. One major advantage with Metalsorb PCZ as precipitating agent is the wide range of operating conditions, especially with respect to pH. Although the pH values commonly found in tunnel wash water (>7) does not imply that pH adjustment is deemed necessary for precipitation with sodium sulfide, monitoring would still be required in a large scale setting.

### 7.4.3 Sludge handling

Overall, the amount of settled sludge is increased with increasing coagulant dose. Consequently, it is desirable that the coagulant dose in use is as low as possible to reduce costs entailed to sludge volume. Based on cost estimates from the Bjørnegårdstunnel as presented in 5.8.2, 15.26 tons of polluted water to be disposed of over a duration of in total 6 hours would entail a total of 14 811 920 NOK ex. VAT. In comparison, a 10% increase in sludge volume would represent increased deposition costs alone of 1 480 220 NOK ex. VAT. In addition, it is reasonable to assume that the increased sludge volume would result in increased handling time, thus increasing the total costs related to handling even further.

Based on Zianuddin et. Al (2019), a 10% increase in generated sludge is not an unlikely assumption. A sludge generation of 0.133 g/2L sample corresponds to a total of 1015 kg, or an increase of just under 7%, when considering the volumes from the Bjørnegårdstunnel. Under optimized conditions, it is reasonable to assume that the sludge volume would be further increased due to improved separation of the solid phase.

Realistically, there is little potential for reuse or resource extraction associated with sludge from tunnel wash water. As shown through analyses in previous sections, the sludge is heavily polluted and contains a number of metals to be removed, additional to other pollutants such as oil and PAHs. Consequently, minimizing the sludge production should be of priority within the sludge handling and within the treatment of the tunnel wash water altogether. Although introducing chemical coagulation or precipitation as treatment has been shown to both significantly improve the water quality of the effluent and simultaneously reduce the required treatment time consumption to a great extent, implementing such a treatment step would imply an increase in overall sludge generation and thus also entail increased costs from the treatment altogether. Still, due to the possibility of considerably improving the separation properties of the coagulated sludge, the overall waste volume would not necessarily need to be increased.

# 8 Conclusion

In general, coagulation and chemical precipitation show promising results as a potential treatment technology for tunnel wash water. Efficient turbidity removal is attainable through relatively low coagulant doses under optimal conditions and the effects with regards to turbidity removal are stable in spite of small changes in the initial water quality. Without any pH adjustment, lower residual turbidities was measured in the samples injected with higher coagulant doses. The initial pH of the sampled water was within the lower tier of what is typically found in tunnel wash water. Adjusting the pH to represent the higher tier proved favorable for the lower coagulant doses, indicating that the coagulant dose required to reach sufficiently low turbidities could be decreased in combination with pH monitoring and/or necessary adjustments. Increasing the pH and thus decreasing the required coagulant dose should be desireable with regards to reducing the generated sludge volumes. Under the optimal conditions provided after pH adjustment, sweep coagulation is plausibly the working coagulation mechanism.

The performance of the investigated coagulants PIX-318 and PAX-18, was rather similar with regards to turbidity removal. Overall, a lower coagulant dose was required when using PIX-318, however PAX-18 was observed to have a better effect on reducing metals from the wash water. Especially when considering the dissolved fraction, PAX-18 was observed to provide a more stable and efficient removal. Although the concentrations of Al were increased from addition of the coagulant, these were after having undergone 120 minutes sedimentation measured at acceptable levels relative to recommended discharge concentrations. A similar conclusion could not be drawn for Fe. For certain metals, such as Zn and Ni, which are assumed to be more associated with the LMM fraction, chemical precipitation resulted in the lowest residual concentrations. However for the majority of metals measured, the greatest removal percentage was achieved through coagulation with PAX-18, although at a higher coagulant dose than with the use of PIX-318.

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#### Appendices Α

#### The bjørnegårdstunnel - representative normalsample A.1



Mesta AS Postboks 253 1326 LYSAKER Attn: Olav Øien Eurofins Environment Testing Norway

(Moss) F. reg. NO9 651 416 18 Møllebakken 50 NO-1538 Moss

TIf: +47 69 00 52 00 miljo@eurofins.no

#### AR-23-MM-002651-01

#### EUNOMO-00360816

Prøvemottak: Temperatur: Analyseperiode: 05.01.2023 05.01.2023 02:05 -12.01.2023 02:22

Referanse:

200958-1209

## **ANALYSERAPPORT**

Prøvenr.: Prøvetype: Prøvemerk	ι	<b>39-2023-01050324</b> Jrent vann Bjørnegård		Prøvetakingsdato: Prøvetaker: Analysestartdato:			.2023 zien .2023
Analyse			Resultat	Enhet	LOQ	MU	Metode
pH målt v	ed 23 +/- 2°C		8.0		1	0.2	NS-EN ISO 10523
Suspende	rt stoff		15	mg/l	2	20%	Intern metode
a) Arsen (As	), oppsluttet		3.9	µg/l	0.2	30%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Bly (Pb), o	oppsluttet		0.59	µg/l	0.2	20%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Kadmium	(Cd), oppslutte	t	0.11	µg/l	0.01	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Kobber (C	u), oppsluttet		16	µg/l	0.5	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Krom (Cr)	, oppsluttet		2.8	µg/l	0.5	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Nikkel (Ni	), oppsluttet		4.3	µg/l	0.5	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Sink (Zn),	oppsluttet		81	µg/l	2	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Totale hy	drokarboner (1	HC)					
a) THC >C5-	C8		< 5.0	µg/l	5		Intern metode
a) THC >C8-	C10		< 5.0	µg/I	5		Intern metode
a) THC >C10	)-C12		< 5.0	µg/I	5		Intern metode
a) THC >C12			< 5.0		5		Intern metode
a) THC >C1				µg/I	20	35%	Intern metode
	(>C5-C35)		140	µg/l		35%	Intern metode
a) PAH(16) E	PA						
a) Naftalen			< 0.010		0.01		Intern metode
a) Acenaftyle			< 0.010		0.01		Intern metode
a) Acenafter a) Fluoren			< 0.010 < 0.010		0.01 0.01		Intern metode Intern metode

Tegnforklaring:

 Kko emfattet av akkrediteringen LOQ: Kvantifiseringsgrense MU: Måleusikkerhet
 Kinder enn >: Større enn nd: likke påvist, Bakteriologiske resultater angitt som <1,<50 e.l. betyr ikke påvist, Måleusikkerhet er angitt med dekningsfaktor k=2. Måleusikkerhet er ikke tatt hensyn til ved vurdering av om resultatet er utenfor grenseverdi/ -området. For mikrobiologiske analyser oppgis konfidensintervallet. Ytterligere opplysninger om måleusikkerhet fås ved henvendelse til laboratoriet. Rapporten må ikke gjengis, unntatt i sin helhet, uten laboratoriets skriftlige godkjennelse. Resultatene gjelder kun for de(n) undersøkte prøven(e). Resultater gjelder prøven slik den ble mottatt hos laboratoriet.

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#### AR-23-MM-002651-01

# **eurofins**

#### EUNOMO-00360816

	Se curonnis				
a)	Fenantren	< 0.010 µg/l	0.01		Intern metode
a)	Antracen	< 0.010 µg/l	0.01		Intern metode
a)	Fluoranten	< 0.010 µg/l	0.01		Intern metode
a)	Pyren	< 0.010 µg/l	0.01		Intern metode
a)	Benzo[a]antracen	< 0.010 µg/l	0.01		Intern metode
a)	Krysen/Trifenylen	< 0.010 µg/l	0.01		Intern metode
a)	Benzo[b]fluoranten	< 0.010 µg/l	0.01		Intern metode
a)	Benzo[k]fluoranten	< 0.010 µg/l	0.01		Intern metode
a)	Benzo[a]pyren	< 0.010 µg/l	0.01		Intern metode
a)	Indeno[1,2,3-cd]pyren	< 0.0020 µg/l	0.002		Intern metode
a)	Dibenzo[a,h]antracen	< 0.010 µg/l	0.01		Intern metode
a)	Benzo[ghi]perylen	0.0023 µg/l	0.002 4	40%	Intern metode
a)	Sum PAH(16) EPA	0.0023 µg/l	4	10%	Intern metode

Utførende laboratorium/ Underleverandør: a) Eurofins Environment Sweden AB (Lidköping), Box 887, Sjöhagsg. 3, SE-53119, Lidköping ISO/IEC 17025:2017 SWEDAC 1125,

Moss 12.01.2023

Kjetil Sjaastad Kjetil Sjaastad Kundeveileder (ASM)

Tegnforklaring:         LOQ: Kvantifiseringsgrense         MU: Måleusikkerhet           * Ikke omfattet av akkrediteringen         LOQ: Kvantifiseringsgrense         MU: Måleusikkerhet           <: Mindre enn >> Større enn nd: Ikke påvist. Bakteriologiske resultater angitt som <1,<50 e.l. betyr 'ikke påvist'.           Måleusikkerhet er angitt med dekningsfaktor k<2. Måleusikkerhet er ikke tatt hensyn til ved vurdering av om resultatet er utenfor grenseverdi/ -området.           For mikobiologiske analyser oppgis konfidensintervallet. Ytterligere oppysninger om måleusikkerhet fås ved henvendelse til laboratoriet.           Rapporten må ikke gjengis, unntatt i sin helhet, uten laboratoriets skriftlige godkjennelse. Resultaten gjelder kun for de(n) undersøkte prøven(e).           Resultater gjelder prøven slik den bie mottatt hos laboratoriet.	Side 2 av 2	AR-001 v 189
Resultater gjelder prøven slik den ble mottatt hos laboratoriet.	Side 2 av 2	

# A.2 The Bjørnegårdstunnel - sample with deteriorated water quality



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miljo@eurofins.no

#### AR-23-MM-020211-01

#### EUNOMO-00366799

Prøvemottak: Temperatur: Analyseperiode:

Referanse:

200958-1209

01.03.2023 02.03.2023 12:00 -08.03.2023 09:12

### **ANALYSERAPPORT**

Prøvenr.: 439-2023-03010314 Prøvetype: Urent vann Prøvemerking: Bjørnegård			Prøvetakingsdato: Prøvetaker: Analysestartdato:			2.2023 Kartnes 3.2023
Analyse		Resultat		LOQ	MU	Metode
pH målt ved 2	3 +/- 2°C	7.7		1	0.2	NS-EN ISO 10523
Suspendert st	off	< 2.0	mg/l	2		Intern metode
a) Arsen (As), op	ppsluttet	2.6	hð\I	0.2	30%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Bly (Pb), opps	luttet	3.7	hð\I	0.2	20%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Kadmium (Cd	), oppsluttet	0.16	hð\I	0.01	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Kobber (Cu), o	oppsluttet	410	hð\I	0.5	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Krom (Cr), op	psluttet	1.0	hð\I	0.5	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Nikkel (Ni), op	psluttet	5.6	hð\I	0.5	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Sink (Zn), opp	isluttet	2100	hð\I	2	25%	SS-EN ISO 15587-2:2002/SS-EN ISO 17294-2:2016
a) Totale hydrok	arboner (THC)					
a) THC >C5-C8		< 5.0	µg/l	5		Intern metode
a) THC >C8-C10	)	< 5.0	µg/l	5		Intern metode
a) THC >C10-C1	2	< 5.0		5		Intern metode
<ul> <li>a) THC &gt;C12-C1</li> </ul>		< 5.0		5		Intern metode
a) THC >C16-C3	35	< 20	µg/l	20		Intern metode
a) Sum THC (>C	5-C35)	nd				Intern metode
a) PAH(16) EPA						
a) Naftalen		< 0.010	µg/l	0.01		Intern metode
a) Acenaftylen		< 0.010	µg/l	0.01		Intern metode
a) Acenaften		< 0.010	µg/l	0.01		Intern metode
a) Fluoren		< 0.010	µg/l	0.01		Intern metode

Tegnforklaring:

Kikke onfattet av akkrediteringen
 LOQ: Kvantifiseringsgrense
 MU: Måleusikkerhet
 S: Mindre enn >: Større enn nd: Ikke påvist. Bakteriologiske resultater angitt som <1,<50 e.l. betyr 'ikke påvist'.

<: Mindre enn >: Større enn nd: ikke pavist. Bakteriologiske resultater angitt som 11,<br/>>50 et. Detyr ikke pavist.
Måleusikkerhet er angitt med dekningsfaktor k=2. Måleusikkerhet er ikke tat hensyn til ved vurdering av om resultatet er utenfor grenseverdi/-området.
For mikrobiologiske analyser oppgis konfidensintervallet. Ytterligere opplysninger om måleusikkerhet fås ved henvendelse til laboratoriet.
Raporten må kke gjengis, unntatt i sin helhet, uten laboratoriets skriftlige godkjennelse. Resultatere gjelder kun for de(n) undersøkte preven(e).
Resultater gjelder prøven slik den ble mottatt hos laboratoriet.
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#### AR-23-MM-020211-01

# **R**eurofins

#### EUNOMO-00366799

	<b>Curonns</b>				
a)	Fenantren	< 0.010 µg/l	0.01	Intern metode	
a)	Antracen	< 0.010 µg/l	0.01	Intern metode	
a)	Fluoranten	< 0.010 µg/l	0.01	Intern metode	
a)	Pyren	< 0.010 µg/l	0.01	Intern metode	
a)	Benzo[a]antracen	< 0.010 µg/l	0.01	Intern metode	
a)	Krysen/Trifenylen	< 0.010 µg/l	0.01	Intern metode	
a)	Benzo[b]fluoranten	< 0.010 µg/l	0.01	Intern metode	
a)	Benzo[k]fluoranten	< 0.010 µg/l	0.01	Intern metode	
a)	Benzo[a]pyren	< 0.010 µg/l	0.01	Intern metode	
a)	Indeno[1,2,3-cd]pyren	< 0.0020 µg/l	0.002	Intern metode	
a)	Dibenzo[a,h]antracen	< 0.010 µg/l	0.01	Intern metode	
a)	Benzo[ghi]perylen	< 0.0020 µg/l	0.002	Intern metode	
a)	Sum PAH(16) EPA	nd		Intern metode	

Utførende laboratorium/ Underleverandør: a) Eurofins Environment Sweden AB (Lidköping), Box 887, Sjöhagsg. 3, SE-53119, Lidköping ISO/IEC 17025:2017 SWEDAC 1125,

Moss 08.03.2023

Kjetil Sjaastad Kjetil Sjaastad Kundeveileder (ASM)

Tegnforklaring:		
* Ikke omfattet av akkrediteringen	LOQ: Kvantifiseringsgrense MU: Måleusikkerhet	
<: Mindre enn >: Større enn nd: Ikke på	avist. Bakteriologiske resultater angitt som <1,<50 e.l. betyr 'ikke påvist'.	æ
Måleusikkerhet er angitt med dekningsfa	faktor k=2. Måleusikkerhet er ikke tatt hensyn til ved vurdering av om resultatet er utenfor grenseverdi/ -området.	× 18
For mikrobiologiske analyser oppgis kor	nfidensintervallet. Ytterligere opplysninger om måleusikkerhet fås ved henvendelse til laboratoriet.	6
Rapporten må ikke gjengis, unntatt i sin	n helhet, uten laboratoriets skriftlige godkjennelse. Resultatene gjelder kun for de(n) undersøkte prøven(e).	AR
Resultater gjelder prøven slik den ble m	nottatt hos laboratoriet.	Side 2 av 2

#### A.3Kemira PIX 318 product information

# Kemira PIX-318

Kemira POK-318, jemikloridsuklat, er et flytende fellingsmiddel for vanmeuning og inneholder aktive 3-verdige jemitutkindelser. PIK-318 ogner sog for avlapsvannar, nænäng, overfølse og grunnvannsrensing og alamiondisjonering i de fioste muneprosasser. PIK-318 er effektiv ved bekjempelae av hydrogensuffid.

Kjemisk data		
Jem Fe <sup>2+</sup>	$11,6 \pm 0,3$	%
pH	<1.0	
Tethet	$1,50 \pm 0,03$	g/cm <sup>2</sup>
Frisyne	<20	gikg PIX-318
Sulfat (SO42)	<230	grkg PIX-318
Klorid (CI)	<100	gikg PIX-318
Uløst stoff	<0,05	%
Krystallisasjonstemp	0°C	
Aktiv stoff (Me)	ca 2,1	mol/kg (3,1 mol/l)

#### Typiske analyseverdier <1,00 mg/kg PIX-318 <0.05 mg/kg PIX-318 <20.00 mg/kg PIX-318 Sporstoff Bly (Pb) Kadmium (Cd) Kobolt (Co)

Kobber (Cu)	<1.00	mg/kg PDC-318
Krom (Cr)	<5,00	mg/kg PIX-318
Kvikksølv (Hg)	<0.05	mg/kg PIX-318
Nikkel (Ni)	<40,00	mg/kg PIX-318
Sink	<70,00	mg/kg PIX-318

Desering Desering av PIX-318 skjør med pumper i korrosjonsbeskyttet utførelse. Rørtedninger og ventiler skal være utført i plast eller gummiert stäl. PIX-318doseres uten fortynning direkte fra lagertank.

Godkjenning PDC-518 er godkjent av SNT som fellingsmiddel ved drikkevannsrensing.

Kvalitet og miljø Kemira Chemicals er sertifisert iht ISC9001/PR2 og 14001.

#### Omregningstabell for dosering

mi PIX- 318/m <sup>3</sup>	g PIX- 318/m <sup>5</sup>	g Fe /m²
20	29.8	3.46
40	59,6	6,91
60	89,4	10,37
80	110.2	13,83
100	149.0	17,28
120	174.8	20,74
140	208.6	24,20
160	238.4	27,65
180	268.2	31,11
200	298.0	34,57
300	447.0	51,85
400	696.0	69,14
500	745.0	86,42

#### Händtering

Beskyttelsesbriller skal benyttes ved åpen håndtering av PIX-318. Personlig vemeutstyr benyttes ved behov.

Lagertanker og beholdere skal merkes:

ETSENDE VÆSKE JERNKLORIDSULFAT

Lagring PIX-318 ber ikke lagres mer enn 12 måneder. Lagertank ber utføres i glassfberammert polysster eller gummiert stältank. Lagertank ber inspiseres og rengjøres 1 gang per år.

Vernetiltak PIX-318 er svakt etsende. Kroppsdeler som kommer i kontakt med væsken skal skylles med rikelig mangder vann. Øynene skylles godt med øyen-skyllevant ha spyklatske i minst 5 minutter. Ved varig infrasjon i øynene eller på huden ber lege kontaktes. Ved sel ber det spyles med vann Og nøytraliseres med kalk eller kalkstensmel.

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NCM8

## KENIRA CHEMICALS AS, ØRAVN. 14, 1630 GAMLE FREDRIKSTAD, TELEFON: 69 35 85 85, TELEFAX: 69 35 85 95

OSLOKONTOR: HAMANG TERASSE 55, 1338 SANDVIKA, TELEFON: 69358585 TELEFAX: 69358577 e-mail: kemira.no@kemira.com, webelle: www.kemira.ro

PRODUKTINFORMASJON

# A.4 Kemira PAX 18 product information

# Kemira Water Solutions, Inc.

# PAX-18 Polyaluminum Chloride

Kemira's PAX-18 is a high performance liquid polyaluminum chloride coagulant that generally offers superior clarification in either potable or wastewater. The aluminum in PAX-18 is highly charged, enabling less of it to do more. Advantages available to many end users are **Reduced Sludge**, **Minimized pH Adjustment**, **Longer Filter Runs**, **Superior Finished Water Quality**, and **Optimized Cold Water Performance**. PAX-18 is a general-purpose coagulant, versatile enough to handle any type of challenge.

#### PRODUCT SPECIFICATION

Appearance	Yellowish Liquid
Aluminum (Al)	$9.0\pm0.2\%$
Al <sub>2</sub> O <sub>3</sub>	$17.1\pm0.4\%$
Iron (Fe)	< 0.01%
Specific Gravity (25°C)	$1.37\pm0.03$
pН	$0.9 \pm 0.3$
Basicity	$42 \pm 2\%$
Active Material	3.33 moles/kg
Viscosity (25° C)	$30 \pm 5 \text{ cP}$
Freezing Point	-20° C / -4° F

#### **CERTIFICATION / APPROVAL**

PAX-18 meets or exceeds all AWWA standards for polyaluminum chloride. PAX-18 is ANSI/NSF Standard 60 certified for use in potable water treatment up to 200 mg/l.

#### DOSING

PAX-18 should be fed straight without dilution. A diaphragm-metering pump of non-corrosive material is suitable.

#### STORAGE

Storage tanks and piping should be constructed of suitable non-corrosive material such as fiberglass or cross-linked polyethylene. PAX-18 is mildly corrosive and will attack most metals over a period of time. PAX-18 has a recommended shelf life of 8 months. As with any chemical, it is recommended to clean the storage tank every 1-2 year.

#### HANDLING / SAFETY

The handling of any chemical requires care. Anyone responsible for using or handling PAX-18 should familiarize themselves with the full safety precautions outlined in our Material Safety Data Sheet.

#### DELIVERY

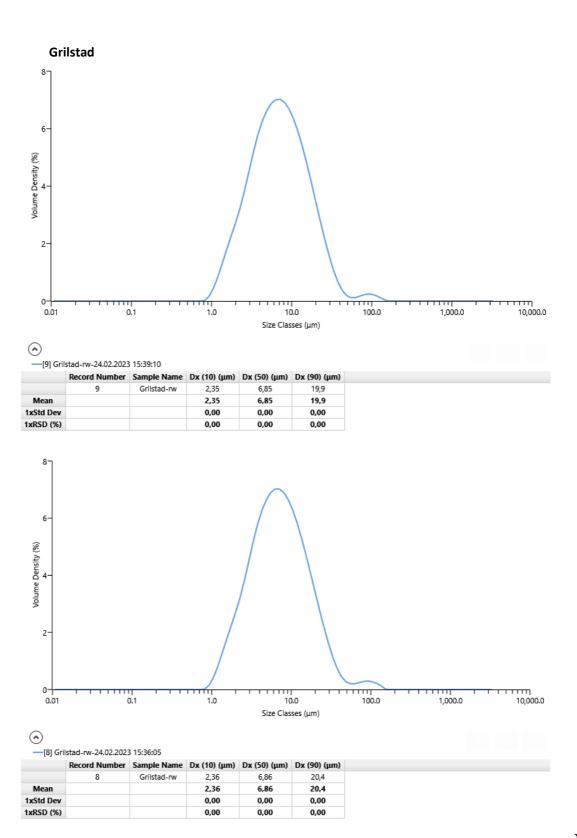
55 gal. plastic drums / 275 gal. tote bin Bulk tank trucks, Railcar Corrosive Liquid, Acidic, Inorganic, n.o.s. 8, UN, 3264, P.G. III

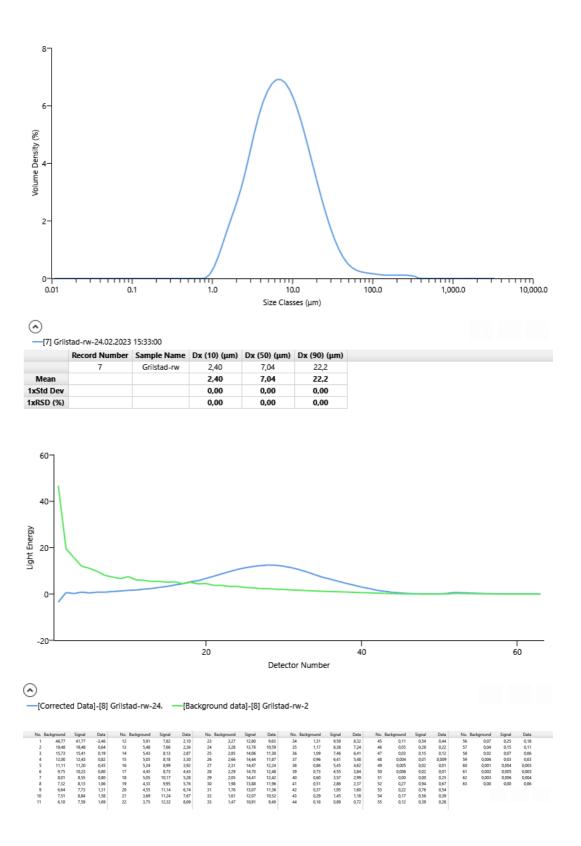
#### PRODUCTION

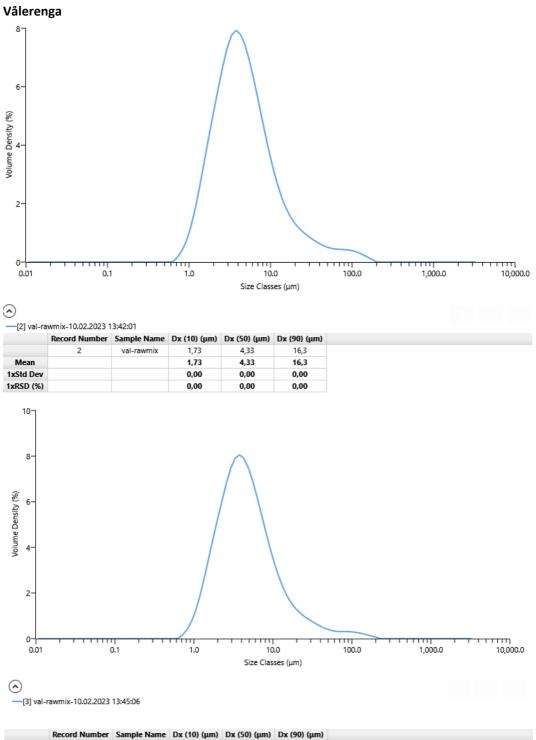
Kemiron has coagulant production plants in

Bartow, FL Fontana, CA Houston, TX Mojave, CA Rowley, UT Saint Louis, MO Savannah, GA Spokane, WA East Chicago, IN

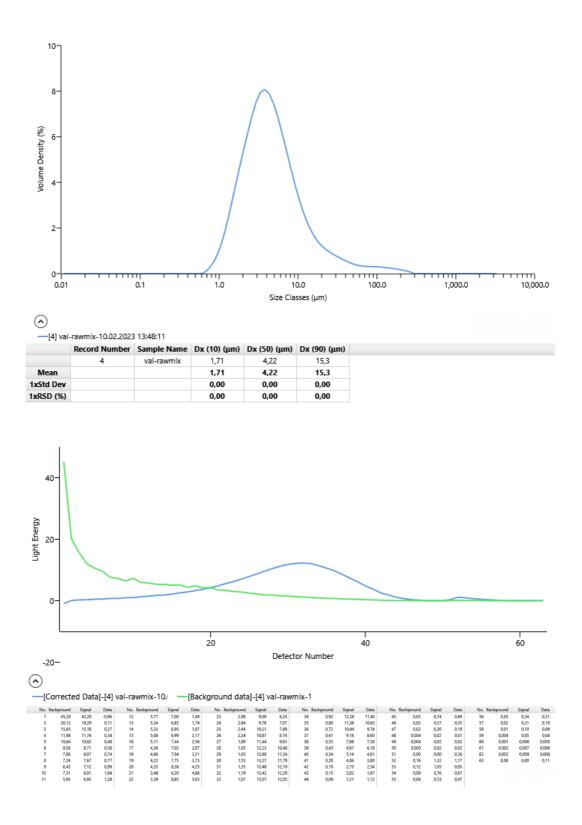
# A.5 Particle size distribution - full measurement







	Necola Namber	Sample Hame	DX (10) (pill)	DX (50) (µm)	DX (50) (µm)
	3	val-rawmix	1,71	4,24	14,9
Mean			1,71	4,24	14,9
1xStd Dev			0,00	0,00	0,00
1xRSD (%)			0,00	0,00	0,00



# A.6 Vålerenga - Raw data particle removal

# A.6.1 Vålerenga - PIX-318 coagulation raw data

Sample ID	Coagulant dose [mmol Fe/L]	pH #1 p	H#2 pH	#3	oH mean	pH stdev	Turbidity #1	Turbidity #2	Turbidity #3	Turbidity mean	Turbidity stdev
F1A	25	6.55	6.60	6.63	6.59	0.04	1.45	1.50	1.28	1.41	0.12
F1B	20	6.93	6.93	6.94	6.93	0.01	3.95	4.07	4.25	4.09	0.15
F1C	15	6.84	6.80	6.93	6.86	0.07	2.09	2.20	2.50	2.26	0.21
F1D	10	6.89	6.90	6.92	6.90	0.02	3.66	4.55	4.61	4.27	0.53
F1E	5	6.49	6.41	6.38	6.43	0.06	3.08	2.88	3.31	3.09	0.22
F1F	1	7.07	7.17	7.16	7.13	0.06	10.20	9.40	10.70	10.10	0.66
F1G	0.1	7.19	7.31	7.36	7.29	0.09	151.00	149.00	152.00	150.67	1.53
F1H	0.01	7.35	7.21	7.23	7.26	0.08	486.00	493.00	475.00	484.67	9.07
F1I	0	6.94	7.24	7.51	7.23	0.29	383.00	387.00	385.00	385.00	2.00
F2A	25	6.82	6.79	6.79	6.80	0.02	2.33	2.85	2.38	2.52	0.29
F2B	20	7.02	7.01	7.02	7.02	0.01	4.70	4.29	4.87	4.62	0.30
F2C	15	7.00	6.96	6.87	6.94	0.07	1.87	1.90	1.90	1.89	0.02
F2D	10	7.01	7.02	6.97	7.00	0.03	3.33	3.96	4.14	3.81	0.43
F2E	5	6.50	6.52	6.51	6.51	0.01	7.12	6.40	6.34	6.62	0.43
F2F	1	7.28	7.25	7.06	7.20	0.12	14.60	15.60	14.90	15.03	0.51
F2G	0.1	7.42	7.46	7.49	7.46	0.04	115.00	119.00	116.00	116.67	2.08
F2H	0.01	7.20	7.26	7.55	7.34	0.19	470.00	465.00	485.00	473.33	10.41
F2I	0	7.55	7.56	7.55	7.55	0.01	437.00	455.00	445.00	445.67	9.02
F1R	5	6.34	6.31	6.30	6.32	0.02	1.28	1.38	1.53	1.40	0.13

# A.6.2 Vålerenga - PAX-18 coagulation raw data

Sample ID	Coagulant dose [mmol Fe/L] pH #1	pH #2	2 pH #3		pH mean	pH stdev	Turbidity #1	Turbidity #2	Turbidity #3	Turbidity mean	Turbidity stdev
A1A	25	6.85	6.93	6.88	6.89		0.49	0.64	0.63		0.08
A2A	20	6.89	6.85	6.81	6.85	0.04	1.16	1.23	1.38	1.26	0.11
A3A	15	6.85	6.91	6.82	6.86	0.05	1.24	1.51	1.56	1.44	0.17
A4A	10	7.04	7.08	6.96	7.03	0.06	3.34	4.95	5.12	4.47	0.98
A5A	5	6.76	7.08	6.96	6.93	0.16	1.59	1.85	1.73	1.72	0.13
A6A	1	7.04	7.10	7.12	7.09	0.04	8.61	8.50	7.84	8.32	0.42
A7A	0.1	7.03	7.29	7.33	7.22	0.16	131.00	132.00	134.00	132.33	1.53
A8A	0.01	7.45	7.38	7.50	7.44	0.06	222.00	225.00	223.00	223.33	1.53
A9A	0	7.37	7.47	7.47	7.44	0.06	469.00	474.00	479.00	474.00	5.00
A1B	25	6.89	6.86	6.93	6.89	0.04	1.69	1.91	1.99	1.86	0.16
A2B	20	6.88	6.90	6.91	6.90	0.02	2.12	1.99	1.25	1.79	0.47
A3B	15	6.71	6.95	6.99	6.88	0.15	4.48	8.59	5.69	6.25	2.11
A4B	10	7.03	7.01	6.99	7.01	0.02	2.05	1.88	1.73	1.89	0.16
A5B	5	7.03	6.70	7.09	6.94	0.21	1.11	1.49	1.81	1.47	0.35
A6B	1	7.05	7.07	7.16	7.09	0.06	7.96	8.64	7.86	8.15	0.42
A7B	0.1	7.26	7.32	7.33	7.30	0.04	198.00	203.00	205.00	202.00	3.61
A8B	0.01	7.50	7.42	7.44	7.45	0.04	212.00	213.00	217.00	214.00	2.65
A9B	0	7.27	7.43	7.43	7.38	0.09	468.00	465.00	470.00	467.67	2.52
A1R	15	6.85	6.91	6.82	6.86	0.05	1.24	1.51	1.56	1.44	0.17
A2R	10	7.07	7.07	7.09	7.08	0.01	6.71	8.60	6.66	7.32	1.11

# A.6.3 Vålerenga - time series raw data

Sample ID	B1	B2	11	12	13	14	15	16	A1	A2	A3	A4	A5	A6
Coagulant	-	-	PIX-318	PIX-318	PIX-318	PIX-318	PIX-318	PIX-318	PAX-18	PAX-18	PAX-18	PAX-18	PAX-18	PAX-18
Coagulant [mmol Me/L]	0	0	1	1	5	5	10	10	1	1	5	5	10	10
pH 20 min #1	7.47	7.35	6.95	6.91	6.60	6.22	4.13	4.14	6.98	7.02	6.85	6.81	6.36	6.32
pH 20 min #2	7.49	7.47	6.99	6.88	6.24	6.21	4.14	4.12	7.02	7.05	6.89	6.83	6.33	6.33
pH 20 min #3	7.52	7.54	6.98	6.96	6.26	6.52	4.13	4.12	7.06	7.09	6.85	6.82	6.34	6.35
20 #1	455.00	473.00	6.34	4.54	25.00	8.86	4.83	6.73	5.44	7.70	10.20	7.56	2.07	1.56
20 #2	470.00	478.00	6.71	5.10	27.80	7.73	4.77	6.39	5.14	5.53	11.50	7.37	2.11	1.43
20 #3	467.00	464.00	6.39	5.24	22.10	7.40	5.10	6.49	5.31	6.62	11.60	7.90	2.43	1.94
20 mean	464.00	471.67	6.48	4.96	24.97	8.00	4.90	6.54	5.30	6.62	11.10	7.61	2.20	1.64
20 std	7.94	7.09	0.20	0.37	2.85	0.77	0.18	0.17	0.15	1.09	0.78	0.27	0.20	0.27
40 #1	413.00	390.00	5.81	3.94	13.50	5.71	5.60	4.63	5.28	5.24	7.89	6.40	2.02	1.45
40 #2	419.00	393.00	6.67	4.05	14.50	6.64	5.61	4.77	4.76	5.75	7.89	6.45	2.05	1.51
40 #3	412.00	403.00	6.38	4.00	13.60	6.03	6.22	4.93	4.18	5.13	7.90	6.46	1.99	1.48
40 mean	414.67	395.33	6.29	4.00	13.87	6.13	5.81	4.78	4.74	5.37	7.89	6.44	2.02	1.48
40 std	3.79	6.81	0.44	0.06	0.55	0.47	0.36	0.15	0.55	0.33	0.01	0.03	0.03	0.03
60 #1	388.00	368.00	5.27	4.24	6.08	4.45	3.02	4.13	4.09	2.72	1.71	2.58	3.99	1.19
60 #2	381.00	376.00	5.09	3.93	5.92	4.93	4.76	4.18	3.56	2.60	1.48	2.19	4.02	1.28
60 #3	394.00	371.00	5.14	3.73	6.57	4.88	4.91	3.98	3.63	3.03	2.57	2.17	4.25	1.40
60 mean	387.67	371.67	5.17	3.97	6.19	4.75	4.23	4.10	3.76	2.78	1.92	2.31	4.09	1.29
60 std	6.51	4.04	0.09	0.26	0.34	0.26	1.05	0.10	0.29	0.22	0.57	0.23	0.14	0.11
120 #1	326.00	307.00	6.66	4.08	7.44	3.50	7.94	2.82	2.25	5.64	3.44	2.22	3.19	1.20
120 #2	331.00	322.00	5.32	3.68	7.28	2.97	5.76	3.32	2.58	5.31	3.26	2.19	2.91	1.10
120 #3	323.00	317.00	5.63	3.92	6.68	2.69	5.16	3.23	1.92	4.61	3.39	2.15	3.04	1.53
120 mean	326.67	315.33	5.87	3.89	7.13	3.05	6.29	3.12	2.25	5.19	3.36	2.19	3.05	1.28
120 std	4.04	7.64	0.70	0.20	0.40	0.41	1.46	0.27	0.33	0.53	0.09	0.04	0.14	0.23

# A.6.4 Vålerenga - TSS raw data

Comple ID	Congulant	Coogulant doca [mmal Ma/1]	Filtered water vel [m]]	Filter initial woight [g] Fil	tor weight ofter besting [g]	Bowlip	Combustion residue [g	TCC [mg/1]
Sample ID	Coagulant	0 1 1					Combustion residue [g	
Blank	-	0	100	0.1250	0.1260		2 0.1241	10
Blank 13	-	0	100	0.1260	0.1256	B9	0.1241	-4
Blank 15	-	0	100	0.1260	0.1250	B7	0.1233	-10
B1	-	0	100	0.1257	0.1499	3-1	0.1446	242.333333
B2	-	0	100	0.1257	0.1473	3-2	0.1425	216.333333
11	PIX-318	1	200	0.1257	0.1428	3-3	0.1386	85.6666667
12	PIX-318	1	200	0.1257	0.1548	4	0.1479	145.666667
13	PIX-318	5	100	0.1257	0.1547	4-1	0.1490	290.333333
14	PIX-318	5	100	0.1257	0.1665	4-2	0.1588	408.333333
15	PIX-318	10	100	0.1257	0.1758	4-3	0.1659	501.333333
16	PIX-318	10	100	0.1257	0.1774	5	0.1672	517.333333
A1	PAX-18	1	200	0.1257	0.1697	0000	0.1608	220.166667
A2	PAX-18	1	100	0.1257	0.1604	v-1	0.1524	347.333333
A3	PAX-18	5	100	0.1257	0.1684	x-1	0.1589	427.333333
A4	PAX-18	5	100	0.1257	0.1622	:	1 0.1540	365.333333
A5	PAX-18	10	100	0.1257	0.1737	B6	0.1617	480.333333
A6	PAX-18	10	100	0.1257	0.1743	x	0.1628	486.333333

# A.6.5 Vålerenga - Zeta potential raw data

Coagulant dose [mmol Me/L]	Coagulant	ZP1	ZP2	ZP3	ZP4	ZP-mean	ZP-stdv
25	PIX-318	-13.2	-12.6	-13.8	-16.5	-14.025	1.72119145
20	PIX-318	-11.7	-12.6	-11.5	-12.7	-12.125	0.61305247
15	PIX-318	-13.4	-12.3	-11.7	-11.5	-12.225	0.85391256
10	PIX-318	-8.61	-12.8	-12.8	-11.5	-11.4275	1.97577622
5	PIX-318	-5.17	-5.64	-4.58	-5.15	-5.135	0.43378182
1	PIX-318	-10.3	-9.82	-11.7	12.9	-4.73	11.7803622
0.1	PIX-318	-14.2	-12.8	-16.3	-16.1	-14.85	1.66232769
0.01	PIX-318	-16.1	-16.9	-15.2	-16.8	-16.25	0.78528127
0	PIX-318	-14.3	-16.5	-17.3	-15.8	-15.975	1.27377392
25	PAX-18	-9.33	-10.1	-10.1	-12.4	-10.4825	1.32886857
20	PAX-18	-9.46	-10.9	-9.29	-10.3	-9.9875	0.75168145
15	PAX-18	-3.05	-2.55	-3.66	-2.57	-2.9575	0.52226271
10	PAX-18	-9.56	-9.09	-8.85	-9.63	-9.2825	0.375
5	PAX-18	-6.1	-6.86	-6.3	-5.69	-6.2375	0.48650968
1	PAX-18	-12.3	-13.3	-13.4	-11.8	-12.7	0.7788881
0.1	PAX-18	-11.7	-13.5	-12.4	-14.6	-13.05	1.27148207
0.01	PAX-18	-15.1	-15.8	-16.7	-16.9	-16.125	0.83416625
0	PAX-18	-15.6	-15.4	-17.3	-15.9	-16.05	0.85829288

# A.7 Grilstad - Raw data particle removal

# A.7.1 Grilstad - PIX-318 coagulation raw data

Sample ID	Coagulant dose [mmol Fe/L] pH #1	pH #2	pH #3	р	H mean	pH stdev	Turbidity #1	Turbidity #2	Turbidity #3	Turbidity mean	Turbidity stdev
F3A	25	6.30	6.27	6.26	6.28	0.02	0.91	0.89	1.07	0.96	0.10
F3B	20	6.61	6.63	6.60	6.61	0.02	1.00	0.73	0.88	0.87	0.13
F3C	15	6.64	6.99	6.98	6.87	0.20	0.45	0.68	0.64	0.59	0.12
F3D	10	7.19	7.20	7.22	7.20	0.02	0.88	0.95	1.03	0.95	0.07
F3E	5	7.26	7.24	7.25	7.25	0.01	1.95	1.66	1.77	1.79	0.15
F3F	1	7.33	7.32	7.30	7.32	0.02	0.95	1.45	1.56	1.32	0.33
F3G	0.1	7.61	7.61	7.64	7.62	0.02	68.90	71.30	70.70	70.30	1.25
F3H	0.01	7.79	7.73	7.74	7.75	0.03	39.20	36.30	39.00	38.17	1.62
F3I	0	7.53	7.72	7.78	7.68	0.13	580.00	570.00	578.00	576.00	5.29
F4A	25	6.50	6.44	6.39	6.44	0.06	2.23	1.98	2.33	2.18	0.18
F4B	20	6.39	6.40	6.40	6.40	0.01	1.11	0.96	0.95	1.01	0.09
F4C	15	6.93	6.69	6.72	6.78	0.13	1.42	1.27	1.45	1.38	0.10
F4D	10	6.79	6.80	6.84	6.81	0.03	0.78	0.71	0.75	0.75	0.04
F4E	5	7.04	7.07	7.14	7.08	0.05	1.07	0.92	0.94	0.98	0.08
F4F	1	7.20	7.30	7.30	7.27	0.06	1.28	2.01	1.66	1.65	0.37
F4G	0.1	7.71	7.83	7.88	7.81	0.09	87.20	89.10	86.00	87.43	1.56
F4H	0.01	7.95	8.03	8.04	8.01	0.05	42.50	43.50	45.00	43.67	1.26
F4I	0	7.53	7.72	7.78	7.68	0.13	580.00	570.00	578.00	576.00	5.29

# A.7.2 Grilstad - PAX-18 coagulation raw data

Sample ID	Coagulant dose [mmol Al/IpH #1	pH #2	2# Hq		pH mean	pH stdev	Turbidity #1	Turbidity #2	Turbidity #3	Turbidity mean	Turbidity stdey
A1A	25	6.24	6.3	6.3		0.03464102	0.555	0.97	0.677		0.213290881
A2A	20	6.73	6.8								0.026857649
				6.85			0.49	0.54	0.532		
A3A	15	6.81	6.83	6.81	6.81666667	0.01154701	0.834	0.905	0.917	0.885333333	0.044859038
A4A	10	6.82	6.88	6.91	6.87	0.04582576	0.984	0.825	0.829	0.879333333	0.090666054
A5A	5	7	7.08	7.13	7.07	0.06557439	1.13	1.37	1.21	1.236666667	0.122202019
A6A	1	7.19	7.18	7.22	7.19666667	0.02081666	2.1	1.79	1.88	1.923333333	0.159478316
A7A	0.1	7.6	7.83	7.84	7.75666667	0.13576941	29.2	29.7	30.9	29.93333333	0.873689495
A8A	0.01	7.77	7.81	7.85	7.81	0.04	108	107	108	107.6666667	0.577350269
A9A	0	7.53	7.72	7.78	7.68	0.13	580.00	570.00	578.00	576.00	5.29
A1B	25	6.19	6.17	6.16	6.17333333	0.01527525	0.952	0.897	0.652	0.833666667	0.159713285
A2B	20	6.85	6.86	6.86	6.85666667	0.0057735	0.478	0.487	0.558	0.507666667	0.043821608
A3B	15	6.75	6.83	6.92	6.83333333	0.08504901	0.794	0.664	0.804	0.754	0.078102497
A4B	10	6.87	6.91	6.92	6.9	0.02645751	0.64	1.13	1.06	0.943333333	0.265015723
A5B	5	7.12	7.12	7.13	7.12333333	0.0057735	1.05	0.982	1.32	1.117333333	0.178777329
A6B	1	7.22	7.25	7.24	7.23666667	0.01527525	1.36	1.4	1.41	1.39	0.026457513
A7B	0.1	7.63	7.71	7.75	7.69666667	0.06110101	10.5	10.8	10.9	10.73333333	0.2081666
A8B	0.01	7.83	7.88	7.89	7.86666667	0.0321455	97.5	98.1	98.4	98	0.458257569
A9B	0	7.53	7.72	7.78	7.68	0.13	580.00	570.00	578.00	576.00	5.29

# A.7.3 Grilstad - time series raw data

Sample ID	B1	B2	11	12	13	14	15	16	A1	A2	A3	A4	A5	A6
Coagulant	-	-	PIX-318			PIX-318	PIX-318	PIX-318	PAX-18				-	PAX-18
Coagulant dose	0	0	1	1	5	5	10			1	5			10
pH 20 min #1	7.53	7.51	7.06	6.90	7.17	7.24	7.12				7.38			
pH 20 min #2	7.72	7.63	7.02	6.90	7.22	7.23	7.10				7.41	7.33		
pH 20 min #3	7.78	7.71	7.03	6.91	7.22	7.22	7.10		7.55		7.39			
20 #1	330.00	321.00	0.90	0.85	1.96	1.55	1.02		2.02		0.52			
20 #2	329.00	331.00	0.89	0.62	2.03	1.74	1.09		2.05		0.56			1.41
20 #3	324.00	341.00	0.90	0.63	2.00	1.87	1.10	1.28	1.95	1.25	0.59	0.58	1.45	0.61
20 mean	327.67	331.00	0.90	0.70	2.00	1.72	1.07	1.13	2.01	1.27	0.56	0.54	1.42	1.01
20 std	3.21	10.00	0.01	0.13	0.04	0.16	0.04	0.13	0.05	0.03	0.04	0.07	0.04	0.40
40 #1	188.00	220.00	1.03	0.58	2.15	1.95	0.90	0.77	1.42	1.13	0.54	0.52	1.15	1.29
40 #2	190.00	204.00	0.94	0.56	1.48	1.50	0.90	0.89	1.37	1.07	0.51	0.51	1.09	0.98
40 #3	190.00	211.00	1.02	0.58	1.46	1.74	1.07	0.95	1.35	0.86	0.50	0.50	0.88	1.01
40 mean	189.33	211.67	1.00	0.57	1.70	1.73	0.96	0.87	1.38	1.02	0.52	0.51	1.04	1.09
40 std	1.15	8.02	0.05	0.01	0.39	0.23	0.10	0.09	0.04	0.14	0.02	0.01	0.14	0.17
60 #1	133.00	152.00	0.93	0.87	1.01	0.90	0.82	0.70	0.96	1.12	0.49	0.44	0.85	1.01
60 #2	136.00	160.00	0.97	1.02	1.04	0.94	0.82	0.65	0.97	1.05	0.51	0.32	0.86	0.83
60 #3	135.00	152.00	0.87	0.99	1.08	0.92	0.73	0.66	1.16	2.22	0.44	0.35	0.91	0.71
60 mean	134.67	154.67	0.93	0.96	1.04	0.92	0.79	0.67	1.03	1.46	0.48	0.37	0.87	0.85
60 std	1.53	4.62	0.05	0.08	0.04	0.02	0.05	0.02	0.11	0.66	0.04	0.06	0.03	0.15
120 #1	111.00	102.00	0.92	0.65	1.20	1.25	0.76	0.93	1.36		0.46	0.27	1.01	0.70
120 #2	107.00	101.00	0.84	0.54	1.23	1.14	0.78		0.79		0.69		0.70	0.71
120 #3	111.00	106.00	0.76	0.57	1.07	1.11	0.69		0.97		0.50		0.48	0.72
120 mean	109.67	103.00	0.84	0.59	1.17	1.17	0.74		1.04		0.55			0.71
120 std	2.31	2.65	0.08	0.06	0.09	0.07	0.04	0.07	0.29	0.10	0.12	0.17	0.27	0.01

# A.7.4 Grilstad - TSS raw data

Sample ID	Bowl ID	Coagulant	Coogulant de Eilte	rod wat Fil	tor initial . Eil	lter weight Skål nr	Gla	derest	TSS [mg/L]
•		'-	0			0			
Blank	Blank	-	0	100	0.122	0.1247	2	0.1232	27
Blank	Blank 12		0	100	0.124	0.1242 B9		0.1231	2
Blank	Blank 14		0	100	0.126	0.1246 B7		0.1234	-14
B21		1 '-	0	100	0.124	0.182 3-1		0.1755	580
B22		2 '-	0	100	0.124	0.1801 3-2		0.1741	561
121		3 PIX-318	1	100	0.124	0.1806 3-3		0.1742	566
122		4 PIX-318	1	100	0.124	0.1757 4		0.1693	517
123		5 PIX-318	5	100	0.124	0.1799 4-1		0.1733	559
124		6 PIX-318	5	100	0.124	0.1708 4-2		0.165	468
125		7 PIX-318	10	100	0.124	0.1838 4-3		0.1767	598
126		8 PIX-318	10	100	0.124	0.1844 5		0.178	604
A21		9 PAX-18	1	100	0.124	0.1871 0000		0.1801	631
A22		10 PAX-18	1	100	0.124	0.1867 v-1		0.1797	627
A23		11 PAX-18	5	100	0.124	0.1885 x-1		0.181	645
A24		12 PAX-18	5	100	0.124	0.1874	1	0.1798	634
A25		13 PAX-18	10	100	0.124	0.1874 B6		0.1796	634
A26		14 PAX-18	10	100	0.124	0.187 x		0.179	630

# A.7.5 Grilstad - Zeta potential raw data

Coagulant dose [mmol Me/L]	Coagulant	ZP1	ZP2	ZP3	ZP4	
25	PIX-318		-7.91	-9.05	-7.17	-6.23
20	PIX-318		-7.88	-7.66	-7.51	-5.04
15	PIX-318		-11.7	-10.9	-7.73	-14.3
10	PIX-318		-9.61	-8.84	9.47	-10.8
5	PIX-318		-4.25	-10.2	-4.45	-4.9
1	PIX-318		-4.42	-2.84	-3.19	-3
0.1	PIX-318		-11.6	-8.64	-9.47	-4.77
0.01	PIX-318		-15	-14.4	-14.2	-17.8
0	PIX-318		-17.8	-17.8	-16.7	-18.1
25	PAX-18		-7.13	-7.05	-10.2	-7.45
20	PAX-18		-10.6	-5	-5.37	-5.43
15	PAX-18		-3.86	-4.15	-5.88	-4.74
10	PAX-18		-11	-4.58	-6.43	-5.63
5	PAX-18		-6.15	-5.75	-7.53	-5.79
1	PAX-18		-5.2	-6.48	-2.3	-3.65
0.1	PAX-18		-9.21	-8.44	-8.76	-6.06
0.01	PAX-18		-14.6	-17.3	-17.4	-15.6
0	PAX-18		-17.8	-17.8	-16.7	-18.1



