

Characterization and treatment of tunneling wastewater using natural and chemical coagulants

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ABSTRACT

Particles are a concern regarding tunneling wastewater, but gaps remain in understanding metal(loid)s content and coagulation efficiency. In this research, characterization of the wastewaters before and after treatment was investigated by chemical analysis and various techniques for particle characterization. Then, laboratory work was conducted to evaluate effectiveness of sedimentation and use of coagulants to remove particles and particle-associated contaminants. Both natural (chitosan) and chemical coagulants (ferric chloride sulfates and polyaluminium chloride solution), were applied in a jar test system. The results indicated that short-time sedimentation alone substantially reduced the particle content and particle-associated pollutants, including metal(loid)s, while subsequent chemical coagulation was required to comply with discharge limits. The optimum dosages of chitosan, PIX, and PAX for water 1 after pre-treatment (15 min sedimentation) were 1 mg/L, 3 mg Fe/L, and 3 mg Al/L, respectively, while a dosage of 2 mg Al/L gave the best results in water 3. Furthermore, chemical coagulation significantly decreased the volume density of particles in the diameter range of 1–100 μm , showing that coagulants are efficient for the removal of smaller particles not removed by conventional sedimentation.

Key words: chitosan, metal(loid)s, particle removal, tunneling wastewater

HIGHLIGHTS

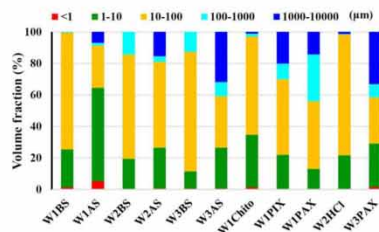
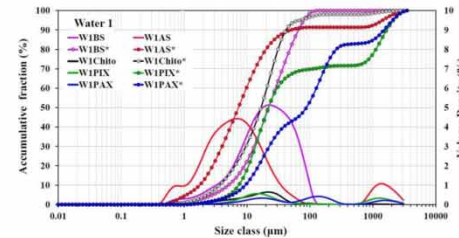
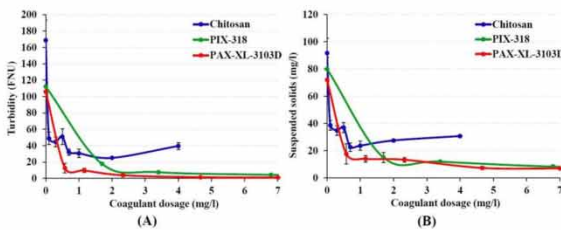
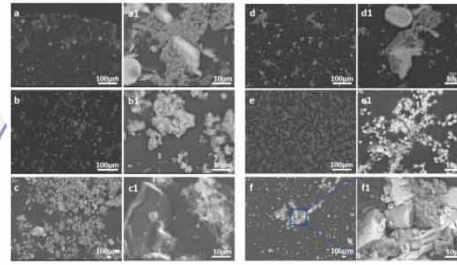
- Characterization and treatment of tunneling wastewater were considered.
- The size distribution, shape, and morphology of the particles were analyzed.
- The application of both natural and chemical coagulants was assessed.
- The proposed method shifted particle size from coarser fractions to finer fractions.
- Chemicals decreased the particle volume showing coagulant efficiency.

GRAPHICAL ABSTRACT

Characterization and Treatment of Tunneling Wastewater Using Natural and Chemical Coagulants

The characterization of tunneling waters before and after 15 minutes sedimentation

Constituent	Tunnel water 1	Tunnel water 1	Tunnel water 2	Tunnel water 2	Tunnel water 3	Tunnel water 3	Concentration limits ^a
	before 15 min sedimentation	after 15 min sedimentation	before 15 min sedimentation	after 15 min sedimentation	before 15 min sedimentation	after 15 min sedimentation	
pH	6.4	6.4	12.7	12.7	12.2	12.2	6-8.5
Alkalinity (mmol/L)	0.2	0.04	32	44	38	15	-
Turbidity (NTU)	534	267	6520	640	3824	60	50-100
TSS (mg/L)	317	143	6140	500	8530	124	100
Total P (mg/L)	0.22	-	2.4	0.17	16	0.14	-
Total N (mg/L)	130	150	35	61	180	160	-
TOC/NPOC ^b (mg/L)	16	8.9	41	46	110	15	-
Hg ²⁺ (µg/L)	-	0.048	0.014	<0.005	0.008	<0.005	0.070
As ³⁺ (µg/L)	2.3	1.85	45	3.7	35	0.35	8.5
Pb ²⁺ (µg/L)	7.7	3.5	350	46	210	1.4	14
Cd ²⁺ (µg/L)	1.2	0.068	4.8	0.45	0.86	0.039	0.45
Cu ²⁺ (µg/L)	41	46	1400	190	1200	110	7.8
Cr ³⁺ (µg/L)	39	32	510	125	270	90	3.4
Ni ²⁺ (µg/L)	8.2	4.6	170	11.8	190	2.6	34
Zn ²⁺ (µg/L)	130	165	1300	65	1200	6.5	11



1. INTRODUCTION

Tunnel construction is one of the most critical activities for surrounding surface water quality in road construction, which may produce several manufactured and natural water-borne sources of pollutants.

The characterization of tunneling wastewater highly varies by site-specific bedrock, weather, construction activity, applied machinery, and the use of chemicals and spray concrete (Vikan & Meland 2013; Rustøen 2015). The tunneling wastewater will consist of water used by drilling rigs for cooling and removal of cuttings (approximately 300 L/min per rig), and infiltration of water from fractures in surrounding rock structures (Vikan & Meland 2013). Tunneling wastewater is generally characterized by high and varying concentrations of total suspended solids (TSS) and turbidity (Lee *et al.* 2014; Liltved *et al.* 2014), elevated and varying ammonia concentrations, pH imbalances (Yi-Wen *et al.* 2012), and oil contaminants originating from mineral oil in explosives, diesel spills, and hydraulic oil from machinery (Vikan & Meland 2013). During drilling operations, high concentrations of TSS are experienced, in the range of 300 to 10,000 mg/L, while the Norwegian discharge limit usually is restricted to 100 mg/L (Vikan & Meland 2013). Tunneling wastewater with large amounts of suspended particles may also have amounts of particle-bound contaminants, such as metalloids, hydrocarbons (e.g., polycyclic aromatic hydrocarbon (PAH)), and oils. Suspended solids and turbidity can cloud receiving water bodies, affect light penetration and food chains, and damage fish gills. According to the European Inland Fisheries Advisory Commission (EIFAC), 400 mg/L of suspended solids will reduce fish populations in rivers and lakes (Vikan & Meland 2013). Metalloids, hydrocarbons, and oils pose toxic risks to the environment, dependent on concentrations. Acute toxic concentrations of metalloids to aquatic life in freshwater bodies are shown in Table 1 as discharge limits (Pettersen 2020). Ammonia from undetonated explosives will be the main contributor to total nitrogen (TN) in tunneling wastewater from Norwegian tunneling projects (Vikan 2013). Ammonium nitrate (NH_4NO_3) is the main constituent of explosives used. In contact with water, NH_4NO_3 is easily

Table 1 | Characterization of the three tunnel waters before and after 15 min of sedimentation

Constituent	Tunnel water 1	Tunnel water 1 after 15 min sedimentation	Tunnel water 2	Tunnel water 2 after 15 min sedimentation	Tunnel water 3	Tunnel water 3 after 15 min sedimentation	Discharge limits ^a
pH	6.4	6.4	12.7	12.7	12.2	12.2	6–8.5
Alkalinity (mmol/L)	0.2	0.04	32	44	38	15	–
Turbidity (NTU)	534	267	6,520	640	3,824	60	50–100
TSS (mg/L)	317	143	6,140	500	8,530	124	100
TP (mg/L)	0.22	–	2.4	0.17	16	0.14	–
TN (mg/L)	130	150	35	61	180	160	–
TOC/NPOC ^b (mg/L)	16	8.9	41	46	110	15	–
Hg ^c (µg/L)	–	0.048	0.014	< 0.005	0.008	<0.005	0.070
As ^c (µg/L)	2.3	1.85	45	3.7	35	0.35	8.5
Pb ^c (µg/L)	7.7	3.5	350	46	210	1.4	14
Cd ^c (µg/L)	1.2	0.068	4.8	0.45	0.86	0.039	0.45
Cu ^c (µg/L)	41	46	1,400	190	1,200	110	7.8
Cr ^c (µg/L)	39	32	510	125	270	90	3.4
Ni ^c (µg/L)	8.2	4.6	170	11.8	190	2.6	34
Zn ^c (µg/L)	130	165	1,300	65	1,200	6.5	11

^aRegarding pH, turbidity, and TSS: discharge limits (Environmental Department of the Governor of Agder County, Norway); Regarding metal(loid)s: concentrations for acute toxic effects in freshwater bodies (Pettersen 2020).

^bTotal organic carbon/nonpurgeable organic carbon.

^cMetals (total).

dissolved into ions, and a pH-dependent equilibrium between NH_4^+ and NH_3 will be established. When spray concrete is used in the construction work, the pH of the tunneling wastewater may reach high values ($\text{pH} > 10$). At such high pH values, most of the ammonia in water will prevail as NH_3 , which is a dissolved gas with high toxicity to aquatic life. For good water quality, a concentration below 10 µg/L of NH_3 in inland water bodies is recommended by the EU Water Framework Directive (Vikan 2013). The pH of tunneling wastewater should be adjusted to values in the range of 6.0–8.5 before discharge to reduce environmental impacts and ammonia toxicity (Vikan 2013).

Also, the high pH of tunneling wastewater will influence water treatment processes, for required dosages of acid to comply with pH discharge limits, and when coagulants are used for particle removal (Rizzo *et al.* 2008). Adjustment of coagulation-pH is important to optimize the particle removal efficiency.

pH imbalances disrupt aquatic life, and high turbidity affects light penetration and food chains. The environmental impact varies, from siltation and water quality reduction to bioaccumulation, disrupted ecosystems, and harm to aquatic organisms. Mitigation measures and proper treatment are crucial to minimize these environmental consequences in tunneling projects.

Particles from drilling and blasting might be edgier and sharper than rounded particles (e.g., clay) and thus more harmful for biota by reducing light penetration, changing temperature and spawning conditions, covering the fish foods (e.g., benthic invertebrates), and damaging fish gill tissue (Bilotta & Brazier 2008). TSS is a vital parameter in the characterization of tunneling wastewater. In a biological context, removing particle-bound contaminants should be accompanied by removing dissolved contaminants, including metal(loid)s, since dissolved contaminants are more bioavailable in the aquatic biota (Meland *et al.* 2010). It should be noted that lowering the pH as a part of the treatment process before discharge may mobilize particle-bound metals because metals generally are more water soluble at lower pH. Turbidity is used as a surrogate measure for suspended solids (Bilotta & Brazier 2008), but the correlation value varies between different suspensions due to particle properties such as shape, color, and reflectivity (Rustøen 2015).

Three commonly used methods for particle removal include sedimentation, filtration, and chemical coagulation. Each of these methods plays a crucial role in enhancing water quality by targeting specific types and sizes of particles. Sedimentation

involves allowing suspended particles in water to settle out by gravity. Water is allowed to rest in a tank or basin, during which heavier particles settle to the bottom due to their greater density. Sedimentation is a valuable process for enhancing water quality by reducing the presence of larger suspended particles. Filtration employs physical barriers (porous medium, such as sand, gravel, or a filter cartridge) to remove particles of various sizes, including fine suspended solids and some microorganisms. Chemical coagulation destabilizes and aggregates suspended particles through the addition of coagulants. These coagulants neutralize charges on particles, causing them to form larger flocs that settle more rapidly during sedimentation.

Well-dimensioned sedimentation ponds can purify tunneling water to approximately 400 mg/L TSS (Vikan & Meland 2013). The weakness of sedimentation basins is the limited removal of the finer (<20 µm) particles (Li *et al.* 2006), which poses a risk to the receiving waters (Nyström *et al.* 2020a). Lowering the particle content below 100 mg/L can be obtained by adding coagulants, often in combination with acid (HCl), at the inlet of the sedimentation pond (Vikan & Meland 2013). Coagulation as a process for the destabilization of the colloidal fraction (including organic and inorganic particulates) (Bratby 2016) is widely used in drinking water treatment (Fabris *et al.* 2010; Zemmouri *et al.* 2013; Muthuraman & Sasikala 2014; Choy *et al.* 2016). Also, the coagulation/flocculation processes have been widely applied for the treatment of turbid stormwater and highway construction runoff (Trejo-Gaytan *et al.* 2006; Kang *et al.* 2007; Sansalone & Kim 2008; Rounce *et al.* 2012; Lee *et al.* 2014; Rustøen 2015; Nyström *et al.* 2019, 2020a).

Chemical coagulants, especially inorganic coagulants, are popular owing to their superiority in treating turbid water, wide availability, and cost-effectiveness (Duan & Gregory 2003; Choy *et al.* 2016). The most widely used coagulants for water treatment are aluminum and iron salts (e.g., aluminum sulfate, ferric chloride, ferric sulfate) (Kang *et al.* 2007; Choy *et al.* 2016). However, using chemical coagulants, particularly aluminum, results in excessive amounts of sludge and disposal problems (Renault *et al.* 2009). There are concerns about elevated aluminum concentrations in the aquatic environment due to toxic effects on biota and humans (Mclachlan 1995; Forbes *et al.* 1997). Recently, environmentally friendly coagulants such as natural polymers have been proposed as an important alternative (Rizzo *et al.* 2008; Bina *et al.* 2009). Natural organic polymers (biopolymers) are biodegradable and nontoxic to human health (Muthuraman & Sasikala 2014). Biopolymers are efficient coagulants in low dosage and will therefore reduce the sludge volume (Bina *et al.* 2009; Choy *et al.* 2016), and their impact on pH and alkalinity is insignificant (Zemmouri *et al.* 2013).

Despite the importance of tunneling construction for the deterioration of surface water quality, little research has been published on the characterization and treatment of tunneling wastewater (Yi-Wen *et al.* 2012; Lee *et al.* 2013, 2014). The current study aimed to characterize tunneling wastewaters and explore the feasibility of using precipitating agents for the treatment of such water. The novelty of this research lies in the following items:

- (1) Characterizing tunneling wastewater regarding chemical composition, particle shape, and morphology by use of advanced technologies such as laser particle analyzer, scanning electron microscope, and X-ray energy dispersive spectrometer.
- (2) Assess the effect of various wastewater treatment methods by the same technologies. The treatments included sedimentation and coagulation using a natural biopolymer (chitosan) and chemical coagulants (ferric chloride sulfates and polyaluminium chloride).
- (3) Enhance the understanding of the performance of natural and chemical coagulants in treating tunneling wastewater.

Water samples were collected at different sites during the construction of the new E39 from Kristiansand to Mandal in the southern part of Norway, which is a part of the European road network.

2. MATERIALS AND METHODS

2.1. Tunneling wastewater

The wastewaters were collected from the tunneling project during drilling operations at the new E39 from Kristiansand to Mandal in southern Norway. A mixture of water from the drilling rigs (water used for cooling the drilling rigs and removal of cuttings), and water leaking into the tunnel, were pumped out to a sedimentation basin. After sedimentation, most of the water was pumped back into the tunnel for reuse, while a smaller fraction was pH-adjusted and discharged to surrounding surface water bodies. Three types of representative tunneling wastewater from different sites with various particle contents were collected as composite samples in 25-L polypropylene containers, transported to the laboratory, and stored at room temperature. To achieve the 25-L composite samples, 0.5-L grab samples were collected over a time span of approximately 1 h. The samples were collected during drilling operations. When two or more 25-L containers were collected, the contents

were transferred to a common large container in the laboratory, mixed properly, and homogenous subsamples were withdrawn for characterization and experimental testing.

2.2. Coagulants used

Chitosan, a linear cationic biopolymer of high molecular weight, was applied as a natural coagulant in this research. Due to the powers of static attraction and adsorption in the molecules and its polyelectrolytic nature, chitosan is an efficient coagulant to (i) remove particles (Divakaran & Pillai 2001; Roussy *et al.* 2005), turbidity (Pan *et al.* 1999; Chen *et al.* 2003; Roussy *et al.* 2005; Kang *et al.* 2007; Rizzo *et al.* 2008; Bina *et al.* 2009; Nyström *et al.* 2020a), natural organic matter (NOM), and colored substances (Eikebrokk 1999; Liltved *et al.* 2001; Chiou & Li 2003; Bratskaya *et al.* 2004; Rizzo *et al.* 2008), (ii) bacteria inactivation (Chung *et al.* 2003), and (iii) metals removal (Juang & Shiau 2000; Jeon & Höll 2003; Rae & Gibb 2003; Rizzo *et al.* 2008; Zeng *et al.* 2008; Zemmouri *et al.* 2013; Rustøen 2015). The performance of chitosan has been assessed on industrial wastewaters, and some studies have been conducted concerning the chitosan efficiency for drinking water treatment (Eikebrokk & Saltnes 2002; Fabris *et al.* 2010; Zemmouri *et al.* 2013), and less is published for road construction wastewater (Lee *et al.* 2013). Compared with other commonly used organic coagulants, chitosan requires less dosage and has quicker floc settling velocity, easier sludge treatment, and no secondary pollution (Lee *et al.* 2013). However, it has a higher cost than traditional chemical flocculants (Zeng *et al.* 2008) and may release some organic matter during coagulation (Rizzo *et al.* 2008).

Chitosan stock solution (0.5%) was prepared by adding 0.5 g of chitosan powder to 100 mL of distilled water and then adding drops of a 30% HCl solution under continuous stirring until the chitosan was dissolved (until no powder was visible). Depending on the required amount of chitosan solution, chitosan was transferred to the jar-test experiments by micropipettes (500–5,000 μL or 50–1,000 μL) to compare the treatment efficiency of chitosan with the commercial coagulants PAX-XL-3103D and PIX-318 (Kemira Kemwater, Helsingborg, Sweden). The dosages of coagulants were calculated based on the active substance (Fe/Al) using mass percentage and density according to the manufacturer's product datasheet.

PAX-XL-3103D is a low-dose, pre-polymerized aluminum coagulant, which can be used for flocculating most waters (e.g., raw water, process water, and wastewater). Compared with traditional coagulants, PAX-XL-3103D is claimed to be more efficient in removing phosphorus and particles with lower sludge volume production and alkalinity consumption and improves the filterability of the settled water (Ghazy *et al.* 2001). On the other hand, PIXs are effective trivalent iron (Fe^{3+}) based coagulants, which are excellent for drinking water production and wastewater treatment (e.g., color, phosphate, metal(loid)s removal, and sludge conditioning). PIX-318 is approved as a precipitating agent in drinking water purification by the Norwegian Food Safety Authority (Rustøen 2015). Both PAX-XL-3103D and PIX-318 were provided by Kemira (Helsinki, Finland). Notably, chitosan, PAX-XL-3103D, and PIX-318 were stored in a refrigerator in sealed glass bottles, and the required concentrations were applied directly in the experiments.

2.3. Particle removal

To assess particle removal efficiency, jar tests with flocculation and sedimentation were performed. A portable jar testing apparatus (Flocculator, Kemira Kemwater, Helsingborg, Sweden) was used for all jar tests. Six 1,000 mL beakers, including one beaker as control, were filled with tunnel water and placed in the jar-test apparatus. Each jar test had a protocol consisting of an initial 45-s rapid mixing phase (200 rpm) in which coagulant was added, followed by 10 min of slow mixing (30 rpm) and finally sedimentation for 45 min. This standard protocol was followed and kept unchanged throughout all experiments. Coagulants (either chitosan, PIX-318, or PAX-XL-3103D) were added during rapid mixing (200 rpm), and pH was adjusted to 6 ± 0.2 by diluted HCl solutions. After completing the jar-test cycle (rapid mixing, flocculation, and sedimentation), 500 mL samples were carefully siphoned from the clear phase for analysis. The removal efficiency (%) was defined as the decrease in effluent concentration after using a precipitating agent, compared to the influent concentration and calculated as in the following equation (Choy *et al.* 2016):

$$E (\%) = \frac{C_1 - C_2}{C_1} \times 100 \quad (1)$$

where C_1 is influent concentration and C_2 is effluent concentration after treatment with the coagulant.

2.4. Analytical methods

Before and after treatment (sedimentation and coagulation), analyses were performed to characterize the different tunnel waters and evaluate the treatment efficiency. pH, electrical conductivity (EC), turbidity, TSS, and particle size distribution (PSD) analysis were measured in the laboratory of the University of Agder according to Norwegian (<https://vanmiljokoder.miljodirektoratet.no/AnalysisMethod.aspx>) and European standards (ISO 11885:2007). For pH measurements, a pH-meter (VMR pH110) with Ross–Sure flow combination pH electrode (Termo Scientific) was used. The meter was calibrated against two standard buffer solutions (pH 4 and pH 7). EC was measured using a calibrated Hach HQ40d instrument (Loveland, CO, USA), while turbidity measurements were conducted using a Hach 2100Q turbidimeter (Loveland, CO, USA). The TSS was determined according to the standard method by filtering known amounts of water through pre-weighed 0.45 µm Whatman GF/C glass microfiber filters. After filtration, the filter was dried at 105 °C for 1 h, and TSS was determined by calculating the difference in filter weight after and before filtration.

TN, total phosphorous (TP), total organic carbon (TOC), and metal(loid)s (Hg, As, Pb, Cd, Cu, Cr, Ni, Zn) were analyzed by Eurofins laboratory, Norway. Samples for TN, TP, and metal(loid)s were pretreated by nitric acid microwave extraction for mobilization of particle-associated compounds and analyzed using inductively coupled plasma atomic emission spectrometry (ICP-OES) according to European standards (ISO 11885:2007).

In this study, three distinct techniques were utilized for particle characterization, including PSD analysis, scanning electron microscopy (SEM), and X-ray energy-dispersive spectrometry (EDS). Each method served a unique purpose, with PSD revealing size distribution, SEM providing insights into particle shapes and morphology evaluation, and EDS identifying particle-associated metals and elemental composition. Combining SEM with EDS enables us to link the visual information obtained from SEM images with the elemental composition provided by EDS, offering the possibility to not only identify any contaminants but also provide information on their potential sites for attachment on the particles. Together, these techniques enhance understanding of particle characteristics, their behavior, potential environmental impact, and their interactions with other materials. Particle size distributions were conducted by light scattering using a Mastersizer 3000 (Malvern v3.72) instrument. For each sample, 10 measurements were performed, and the average values were presented. The samples were analyzed in the range of 0.01–10,000 µm. In volume-based distribution, the volume contributions of different particle size classes to the total particle volume were assessed.

For particle morphology study, a field emission scanning electron microscope (JEOL 7200F, Tokyo, Japan) equipped with an energy-dispersive X-ray spectrometer (Octane Elect EDS system from EDAX[®]-AMETEK[®]) was employed. The microscope was operated at 15 kV (accelerating voltage), at a working distance of 10 mm. Representative samples for microscopy were collected, and subsamples for analysis were siphoned from the upper water layer. This methodology allowed for the settling of larger particles, while samples with the remaining dispersed particles were used for analysis. These particles will also be more typical for particles discharged to surface water bodies after treatment of tunneling water. Water containing dispersed particles was drop-casted on a carbon sticky tape. Once the water evaporated, leaving behind dried particles adhered to the carbon tape, samples were coated with a carbon film using a carbon coater (JEOL JEC-530). For assessing particle-associated metals, samples were analyzed using X-ray EDS equipped with TEAM[™] software. For each tunnel water type, at least three samples were analyzed.

2.5. Statistics

The jar-test experiments were repeated three times. Average values with standard deviations are presented. Where standard deviation bars are not observable on the graphs, they do not extend beyond the dimensions of the symbols. For chemical and physical water analysis, duplicate samples were collected and analyzed. For particle characterization, 3–10 samples were analyzed. Average values are presented.

3. RESULTS AND DISCUSSION

3.1. Characterization

The chemical characteristics of the three tunneling wastewater before and after 15 min sedimentation are shown in Table 1. In published research, the environmental impact of tunnel water has been focused on high pH values, suspended solids, and ammonia, not on metals. In our study, we conducted a comprehensive analysis of tunneling wastewater, with a specific focus on metal(loid)s content both before and after the sedimentation process. Initially, the wastewater exhibited varying levels of metal(loid)s contamination, raising concerns about its potential environmental and health impacts. For the metal(loid)s, the

maximum admissible concentrations for acute toxic effects on aquatic biota in freshwater bodies have been given (Pettersen 2020), while discharge limits for other parameters released by the Environment Department of Agder County, Norway, are also presented in Table 1. Water 1 was from a period of low drilling and blasting activity with low concentrations of substances compared to water 2 and water 3; e.g., the TSS content in water 1 was only 4–5% of the TSS of the two other waters. The pH value of water 1 was also different from water 2 and water 3 (pH value of 6.4 compared to pH values of 12.7 and 12.2, respectively), which may have influenced other characteristics, such as metal solubility. Higher solubility is expected at lower pH.

Water 2 and water 3 carried high amounts of mineral solids with high TSS and turbidity values. The mineral nature of the solids in all waters was evident by the low content of organic carbon (TOC/ non purgeable organic carbon (NPOC)) compared to the TSS concentrations. The majority of the solids, particularly in water 2 and 3, were easily removed by short-time sedimentation (15 min) as shown in Tables 1 and 2. The characterization also shows considerable contents of metal(loid)s in all three waters. It was evident that most metal(loid)s were associated with particles in the waters with high solid content and high pH (water 1 and 2). By removal of solids by sedimentation, most of the metal(loid)s were also removed. However, after undergoing sedimentation, we observed a significant reduction in metal(loid)s concentrations. This reduction highlights the effectiveness of sedimentation as a wastewater treatment method for mitigating metal(loid)s pollution. It also underscores the importance of implementing such treatment strategies in tunneling projects to promote sustainable practices. The removal efficiencies for metals were in the range of 66.7–99.5% (Table 2). The particle association of metals was also confirmed by the EDS analysis, which revealed the presence of As, Pb, S, Cd, Cu, Cr, Ni, Fe, and Ti in particles (supplementary material). The particle association of metals was also confirmed by the EDS analysis, which revealed the presence of As, Pb, S, Cd, Cu, Cr, Ni, Fe, and Ti in particles (see the supplementary material), transferred to the wastewater from the drilling and blasting operations.

In spite of the removals by sedimentation, the residual concentrations were still high for some of the metals, particularly Cu and Cr. The concentrations of Cu and Cr exceeded the acute toxicity levels by factors of 6 and 9 in water 1, and by factors of 24 and 36 in water 2, respectively. This means that dilution factors of the same magnitude should be implemented if such water is discharged to local freshwater bodies to avoid toxicity to aquatic biota. However, the risk of toxicity is reduced due to metal association with particles, which limits bioavailability. It should be taken into consideration that the pH value must be reduced by several units by CO₂-dosing before discharge to comply with requirements. By lowering the pH, increased metal solubility may also take place, which will increase the toxicity of the metals and the risk to aquatic biota. More research should be conducted to highlight the potential metal toxicity of tunnel water, with and without pH adjustment.

Among the nutrients, TP was present in untreated tunnel waters, ranging from 0.22 to 16 mg/L, while N was in the range of 35–180 mg/L (Table 1). As for the metals, the majority of the P-content was removed with the solids during sedimentation, while TN concentrations were more or less unaffected by sedimentation (Table 2). This can be explained by the presence of ammonia-ion (NH₄⁺) as the main N-compound in these waters, released from dynamite during blasting operations. This dissolved form of nitrogen will not be reduced by sedimentation. The percentage removal of the various constituents after 15 min sedimentation in tunnel waters 1, 2, and 3 are shown in Table 2.

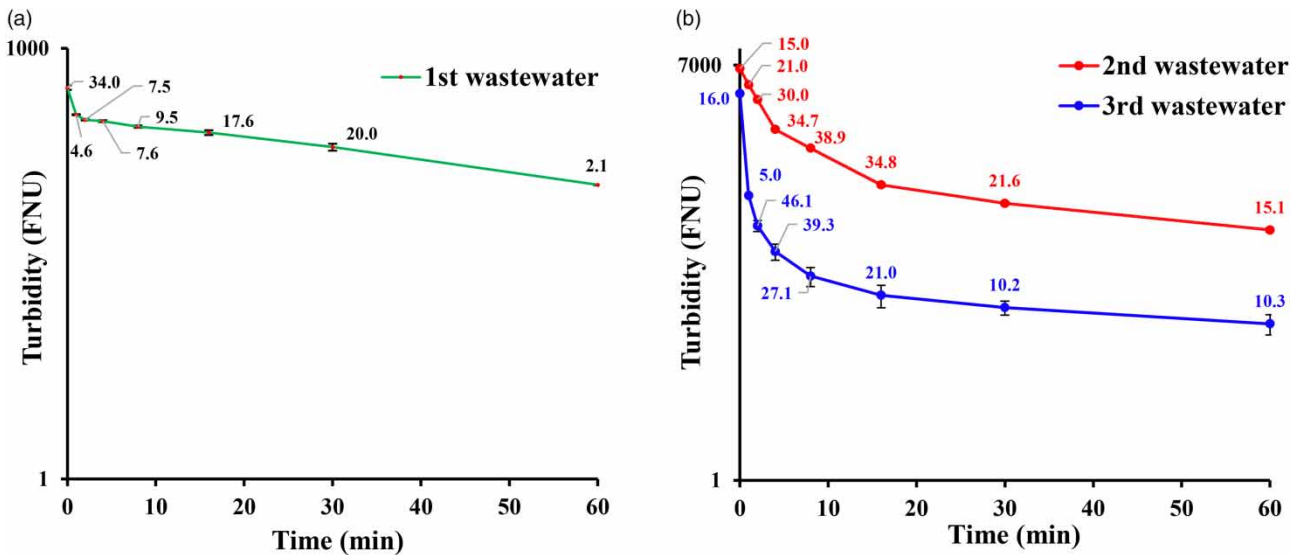
To have an indication of possible emission of persistent organic pollutants, 16 different priority polycyclic aromatic hydrocarbons (PAHs) were analyzed in water 1 and 3 after 15 min of sedimentation. The sum of the 16 PAHs ($\sum\text{PAH}_{16}$) was below the detection limit in water 1, while a low concentration of 0.110 µg/L was found in water 3, which consisted of phenanthrene and pyrene in concentrations of 0.091 and 0.019 µg/L, respectively.

The settling velocity of particles expressed as a reduction in turbidity with the time of the untreated tunnel waters (1, 2, and 3) are presented in Figure 1 (Figure 1(a) shows water 1, while water 2 and 3 are shown in Figure 1(b)). The shape of the curves indicates rapid sedimentation rates. For water 2 and 3 (Figure 1(b)), most of the turbidity was removed after 15 min, from where the shape of the curves leveled off. After 60 min of sedimentation, the reduction in turbidity was 79.0, 96.8, and 99.3% for water 1, 2, and 3, respectively. As shown in Table 2, the corresponding reductions after 15 min were 50.4, 90.2, and 90.4%. From these numbers, the shape of the curves in Figure 1, and reductions in TSS after 15 min of sedimentation (Tables 1 and 2), it was indicated that sedimentation is a highly effective treatment process for tunnel water, in particular for waters carrying high loads of suspended solids (water 2 and 3). However, additional treatment is needed to comply with discharge limits regarding particle content (TSS) and metal concentrations, e.g., Cu and Cr.

Figure 2 displays the SEM images of particles present in tunnel water 1, 2, and 3 before and after sedimentation. SEM analysis indicated a reduction of the solid content after sedimentation, particularly in water 3, where the density of dispersed

Table 2 | Percentage removal of various constituents after 15 min of sedimentation

Constituent	% Removal of constituents after 15 min sedimentation		
	Tunnel water 1	Tunnel water 2	Tunnel water 3
Turbidity (NTU)	50.0	90.2	98.4
TSS (mg/L)	54.9	91.9	98.5
Total P (mg/L)	–	92.9	99.1
Total N (mg/L)	–	–	11.1
TOC/NPOC (mg/L)	44.4	0	86.4
Hg ($\mu\text{g/L}$)	–	>64.3	>37.5
As ($\mu\text{g/L}$)	19.6	91.8	99.0
Pb ($\mu\text{g/L}$)	54.5	86.9	99.3
Cd ($\mu\text{g/L}$)	94.3	90.6	95.5
Cu ($\mu\text{g/L}$)	–	86.4	90.8
Cr ($\mu\text{g/L}$)	17.9	75.5	66.7
Ni ($\mu\text{g/L}$)	43.9	93.1	98.6
Zn ($\mu\text{g/L}$)	–	95.0	99.5

**Figure 1** | Settling velocity, as turbidity values versus sedimentation time, (a) for tunnel water 1 and (b) tunnel water 2 and 3 (both figures in logarithmic scale, and labels indicate standard deviation).

particles after sedimentation (Figure 2(f)) appeared significantly lower compared to the water without treatment (Figure 2(c)). To highlight particle morphology, high-magnification images are shown in images a1–c1 (before sedimentation) and d1–f1 (after 15 min of sedimentation). For water 2, there was no clear reduction in particle density before and after sedimentation when comparing images b and e in Figure 2. The apparent modest differences in particle densities can be explained by the sampling procedure, which allowed for some settling of larger particles before sampling, thereby reducing the differences between untreated and treated waters. This is a limitation of the sampling procedure that can lead to a potential underestimation of the actual differences between the two water samples, particularly in relation to smaller particles that might have remained suspended for longer periods. Additionally, the time delay between sampling and analysis further compounds this limitation, as it allows for ongoing settling during the interval. To address this limitation comprehensively, future studies could explore alternative sampling methods or provide statistical adjustments to account for the settling effect, ensuring a more accurate assessment of the treatment's impact on particle densities. It is also important how the SEM images are

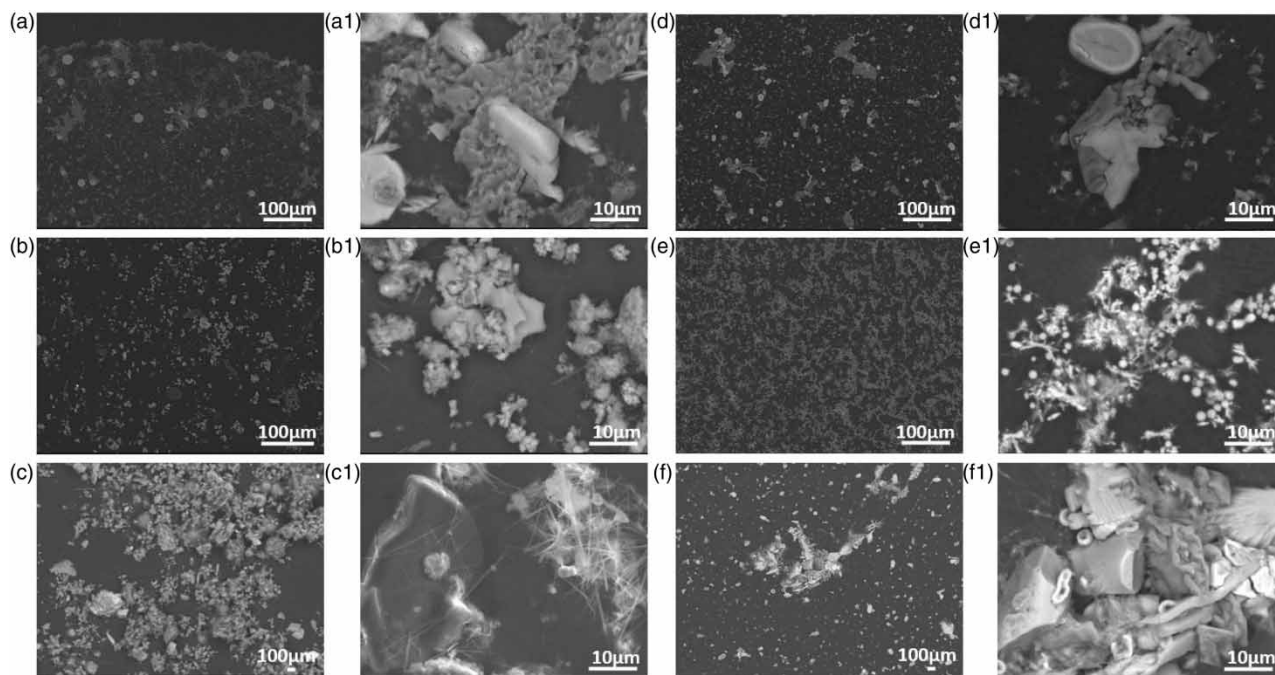


Figure 2 | (a–c) shows particles in tunnel water 1, 2, and 3, respectively, before sedimentation. Corresponding high-magnification images are shown in (a1–c1). (d–f) shows particles from tunnel water 1, 2, and 3, respectively, after 15 min of sedimentation, with corresponding high-magnification images shown in (d1–f1).

interpreted before concluding particle density. At first glance, the density of the particles in water 2 appeared to be higher after sedimentation than before sedimentation (Figure 2(e) compared to Figure 2(b)). However, a closer inspection revealed that untreated water consisted of large clusters of particles, as indicated in Figure 2(b1). After sedimentation, the majority of the particles were smaller, and they were distributed to give an impression of the high density of particles despite an overall lower volume density in the sample (Figure 2(e) and 2(e1)).

EDS analysis, presented in the supplementary material, showed the presence of As, Pb, S, Cd, Cu, Cr, Ni, Fe, and Ti in all three waters. However, Zn was not found in the EDS in any of the samples, even though the presence was shown in all three waters in Table 1. Tunneling water comprises an inhomogeneous mixture of both larger particles and dispersed particles. To conduct SEM/EDS analysis, we selected representative samples from the upper water layer, primarily composed of dispersed particles. A small quantity of this water was carefully drop-casted onto a carbon sticky tape and subsequently subjected to air-drying. This sampling method promotes a more uniform distribution of particles, facilitating comprehensive morphological examinations. However, it does come with a limitation, potentially leading to an underestimation of detected particles when dealing with an inhomogeneous sample. The absence of Zn in the EDS analysis can be attributed to this limitation inherent in our SEM/EDS sampling procedure.

3.2. Chemical coagulation and sedimentation

The results of the sedimentation study indicated that additional treatment was required to remove small residual particles with low settling velocity. To achieve this and comply with discharge limits, we introduced a two-step treatment process, with (1) pre-sedimentation to remove the majority of the suspended solids (here 15 min of sedimentation) followed by (2) chemical coagulation, flocculation, and precipitation to remove the finer solids.

To follow this two-step treatment strategy, all tunnel waters were subjected to 15 min sedimentation, and the clear phase was used for further coagulation experiments. Due to the higher ratio of finer particles in water 1 after 15 min of sedimentation, compared to water 2 and 3, water 1 was chosen for extensive studies of the effects of coagulants. Three different coagulants were tested (PIX-318, PAX-XL-3103D, and chitosan). HCl was used for adjustment of the coagulation-pH to 6.0 ± 0.2 . Turbidity values and TSS concentrations versus increasing dosages of the three coagulants are presented in Figure 3(a) and 3(b). It was shown that both PAX and PIX coagulation resulted in lower turbidity and TSS values than the use of chitosan; however,

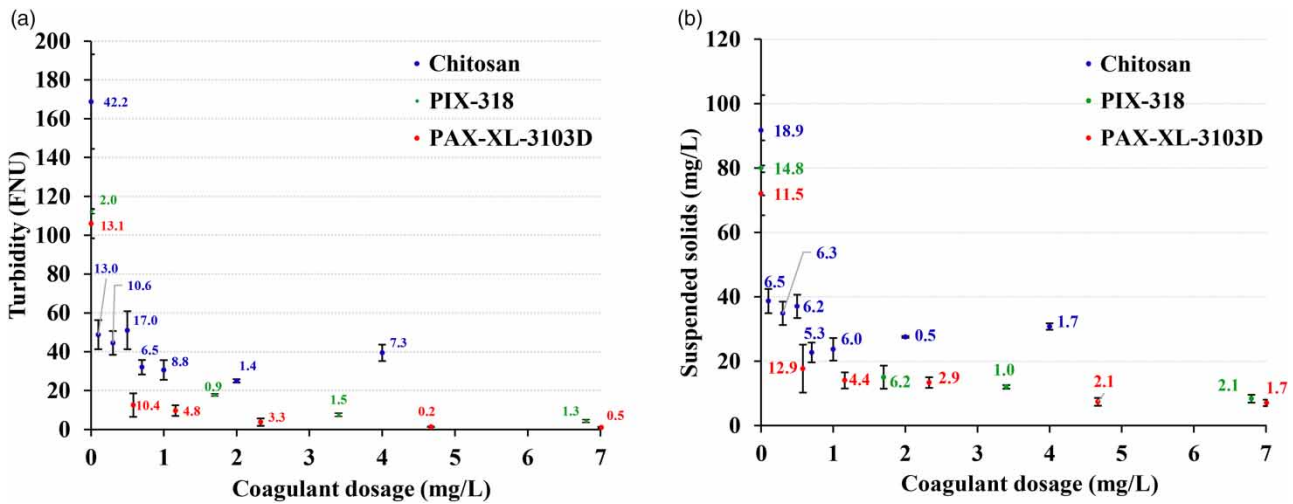


Figure 3 | Turbidity values (a) and TSS concentrations (b) in tunnel water 1 versus increasing dosages of chitosan, PIX, and PAX (Note: labels indicate standard deviation).

higher dosages were required to obtain the best removal efficiencies. Regarding chitosan, the optimum dosage was approximately 1 mg/L, which resulted in residual turbidity of 30.6 FNU and a TSS concentration of 23.7 mg/L, and removal efficiencies of 93.7 and 92.5%, respectively. At higher dosages, reduced removals were observed. For PAX and PIX, very low residuals for turbidity (3.0 and 9.9 FNU) and TSS (11.6 and 12.7 mg/L) were obtained at dosages of 3 mg Al/L and 3 mg Fe/L, respectively. Marginal additional removals were observed by applying higher dosages. PAX was regarded as slightly superior to PIX, particularly at lower dosages, and was therefore selected for additional testing with water 3.

As indicated in Figure 4, low residuals of turbidity and TSS were shown by low amounts of PAX in water 3, with an optimum dosage of 2 mg Al/L. The results show poorer removal efficiencies at higher dosages (3 mg Al/L), indicating that overdosing is possible and should be avoided.

Figure 5 displays the SEM images of water 3 after 15 min of sedimentation (left image) and after coagulation with PAX followed by sedimentation (right image). The SEM analysis showed a reduction in both large clusters and finer particles after coagulation, if we ignore the large brighter structures on the right image in Figure 5. These structures can be explained by voluminous unsettled aluminum hydroxide flocs formed as a result of the PAX treatment.

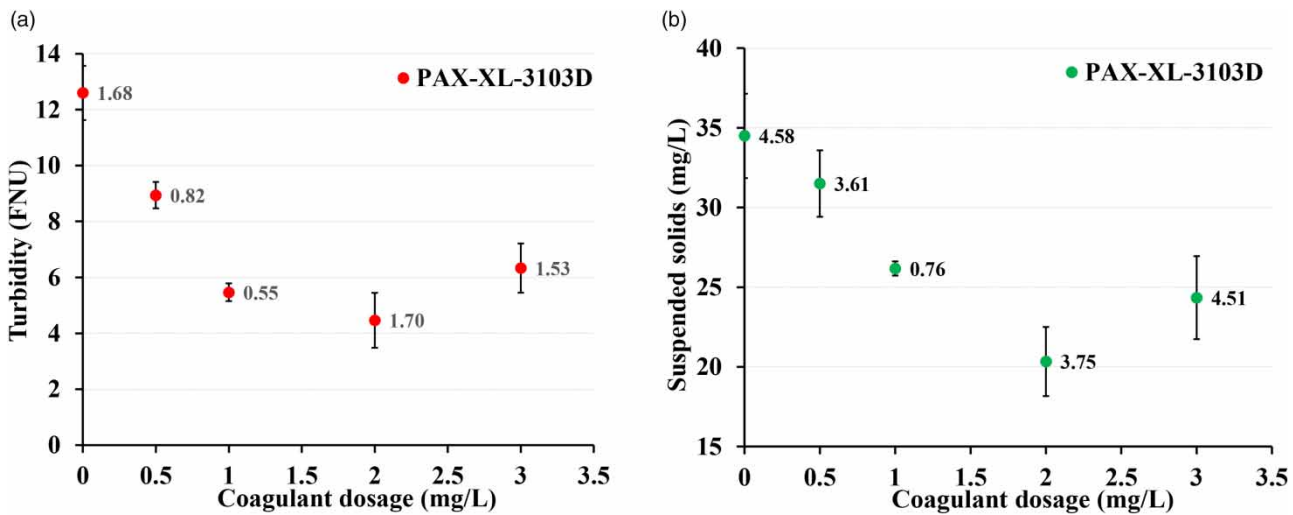


Figure 4 | Turbidity values (a) and TSS concentrations (b) in tunnel water 3 versus increasing dosages of PAX. (Note: labels indicate standard deviation).

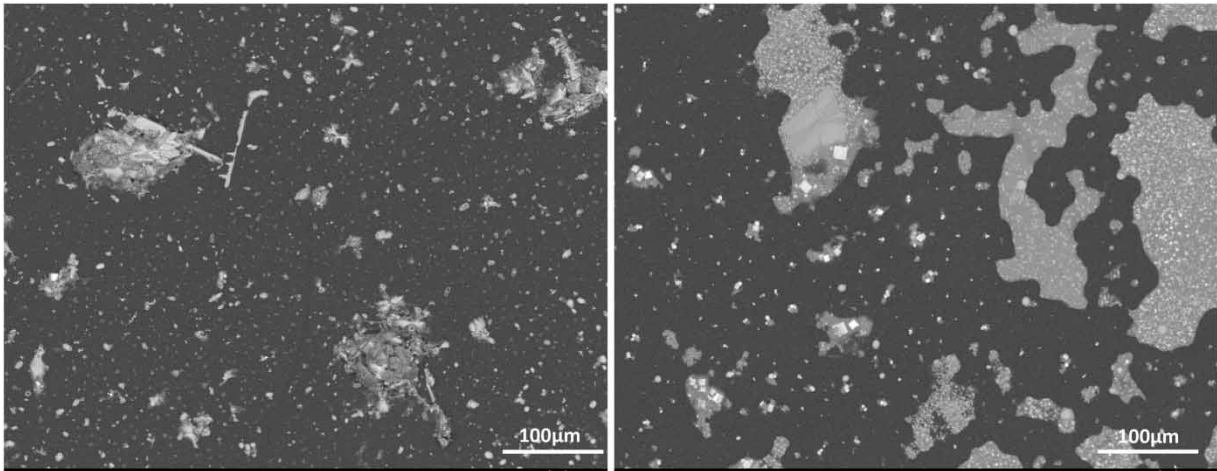


Figure 5 | SEM images of the tunnel water 3 after 15 min of sedimentation (left panel), and two-step treatment (15 min sedimentation followed by coagulation with PAX (2 mg Al/L)) (right panel).

Due to the high initial pH value of tunnel water 2 and 3, it was observed that substantial amounts of acid (HCl) were required to reduce the pH for the best coagulation performance, which was approximately 6 for the chemicals used. It was also observed that the addition of acid alone resulted in good particle removal in jar tests. As shown in Figure 6, by applying a dosage of 2 mL/L of 32% HCl, the residual TSS was lower than the result obtained in water 3 with the optimum dosage of PAX (2 mg Al/L) (Figure 3(b)). At higher acid dosages, both turbidity values and TSS concentrations increased.

Notably, the TSS reduction was highly correlated with the turbidity removal, with a Pearson correlation coefficient of 0.97 based on all the jar-test results. The optimal dosage and removal efficiencies regarding turbidity and TSS from all jar tests are presented in Table 3.

The two-step treatment (15 min sedimentation followed by chemical coagulation and 45 min sedimentation) showed high removal efficiencies for both turbidity (>93%) and TSS (>92%) for all chemicals tested, with residual concentrations substantially below discharge limits. The results obtained with PAX and PIX in water 1 were similar, with a slightly lower final turbidity value with PAX. Dosages of 2–3 mg Al/L of PAX (tunnel water 1 and 3) resulted in effluent turbidity and TSS

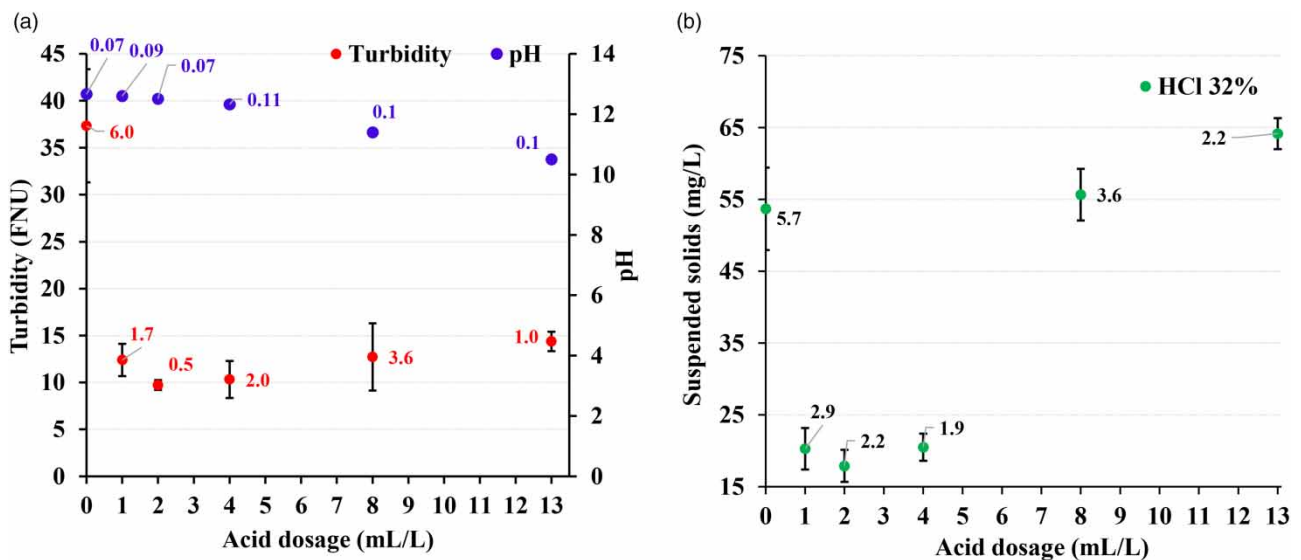


Figure 6 | Turbidity values (a) and TSS concentrations (b) in tunnel water 2 versus increasing dosages of HCl (Note: labels indicate standard deviation).

Table 3 | Optimal chemical dosages for coagulation and removal efficiencies for the three tunnel waters

Tunnel water	Chemical	Optimal dosage	Turbidity (FNU)			TSS (mg/L)			pH
			Initial	Final	reduction	Initial	Final	Reduction	
1	Chitosan	1 (mg/L)	534	30.6	93.7	317	23.7	92.5	6 ± 0.2
	PIX	3 (mg Fe/L)	534	9.9	98.1	317	12.7	95.9	6 ± 0.2
	PAX	3 (mg Al/L)	534	3	99.4	317	11.6	96.3	6 ± 0.2
2	HCl(32%)	2 (mL/L)	6,520	9.7	99.8	6,140	17.9	99.7	12.5 ± 0.2
3	PAX	2 (mg Al/L)	3,824	4.5	99.8	8,530	20.3	99.7	6 ± 0.2

concentrations in the range of 3.0–4.5 FNU and 11.6–20.3 mg/L, respectively, corresponding to removal efficiencies of 96.3–99.7%. The performance of chitosan was slightly lower, but still sufficient to comply with discharge limits. The low chitosan dosage required (1 mg/L) implies low sludge production, which is beneficial when it comes to the environmental impact and operational costs of treatment facilities. Also, the acid treatment with 2 mL/L of 32% HCl resulted in an outstanding performance regarding overall turbidity removal (99.8%) and TSS removal (99.7%). Low sludge production is also expected with acid treatment compared to Al- and Fe-based coagulants.

3.3. Particle size distribution

Besides turbidity and TSS, characterization of tunnel water in terms of the PSD and volume density is important to predict environmental impacts and evaluate the effect of treatment. The PSD curves are shown in Figure 7, in which the continuous solid lines show the volume density fraction of PSD of the different tunnel waters. The corresponding lines with symbols indicate the accumulative fraction of PSD.

Regarding water 1, comparing the volume density curves before (W1BS) and after sedimentation (W1AS), suggests that 15 min of sedimentation shifted PSDs from coarser particles (mean diameter = 22.1 µm) to finer particles (mean diameter = 7.3 µm). Interestingly, after sedimentation, some few unsettled larger particles (>1,000 µm) made substantial contributions to the total particle volumes. This was particularly pronounced in water 3 (W3AS), which was the water with the best sedimentation performance (98.5% of the TSS was removed after 15 min sedimentation, as shown in Table 2).

The application of coagulants (chitosan, PAX, and PIX) significantly decreased the volume density of particles (%) in the measured size classes (Figure 7(a) and 7(c), water 1 and water 3). When comparing the volume densities (%) after the use of coagulants (W1Chito, W1PIX, and W1PAX) to after sedimentation (W1AS) in water 1 (Figure 7(a)), the volume density contribution of different size classes, particularly in the range of 1 µm to 100 µm, were significantly reduced. A similar pattern was observed in water 3 when PAX was used as a coagulant, and also a substantial removal of the larger and lighter particles in the size range of 500–2,000 µm (W3PAX versus W3AS, Figure 7(c)).

The cumulative volume (%) of a specific size indicates the cumulative volume of particles in the sample that has an equivalent diameter less than the specific size. In water 1 (Figure 7(a)), the contribution (percentage) of smaller particles to the total particle volume is higher after sedimentation than before sedimentation, i.e., particles with diameter <20 µm will contribute to 80% of the total particle volume after sedimentation (W1AS*), while the same particle size class (<20 µm) will contribute to only 47% of the total particle volume before sedimentation (W1BS*). This indicates the removal of larger particles and a shift toward finer particles by sedimentation. The cumulative volume distribution after sedimentation (W1AS*) shows an almost flat curve in the particle size range of approximately 50–1,000 µm, indicating that the majority of particles in this size range were removed by sedimentation, resulting in a marginal contribution of these particles to the total particle volume. Further, when comparing the cumulative volume after sedimentation (W1AS*) to the cumulative volume when coagulants were used, the smaller particle contributed less to the total cumulative volume. This was especially pronounced for PIX and PAX (W1AS* versus W1PAX* and W1PIX* in Figure 7(a)), indicating these coagulants' efficiency in removing finer particulate matter.

Also, in water 2, the PSD analysis demonstrates that 15 min of sedimentation shifted the distribution toward finer particles (W2BS versus W2AS in Figure 7(b)). However, some larger and lighter particles >500 µm contributed to 18.4% of the total particle volume. These particles were also evident in the cumulative volume distribution (Figure 7(b)). The impact of 15 min of sedimentation was shown on the cumulative volume distribution, with the absence of particles in the size range of

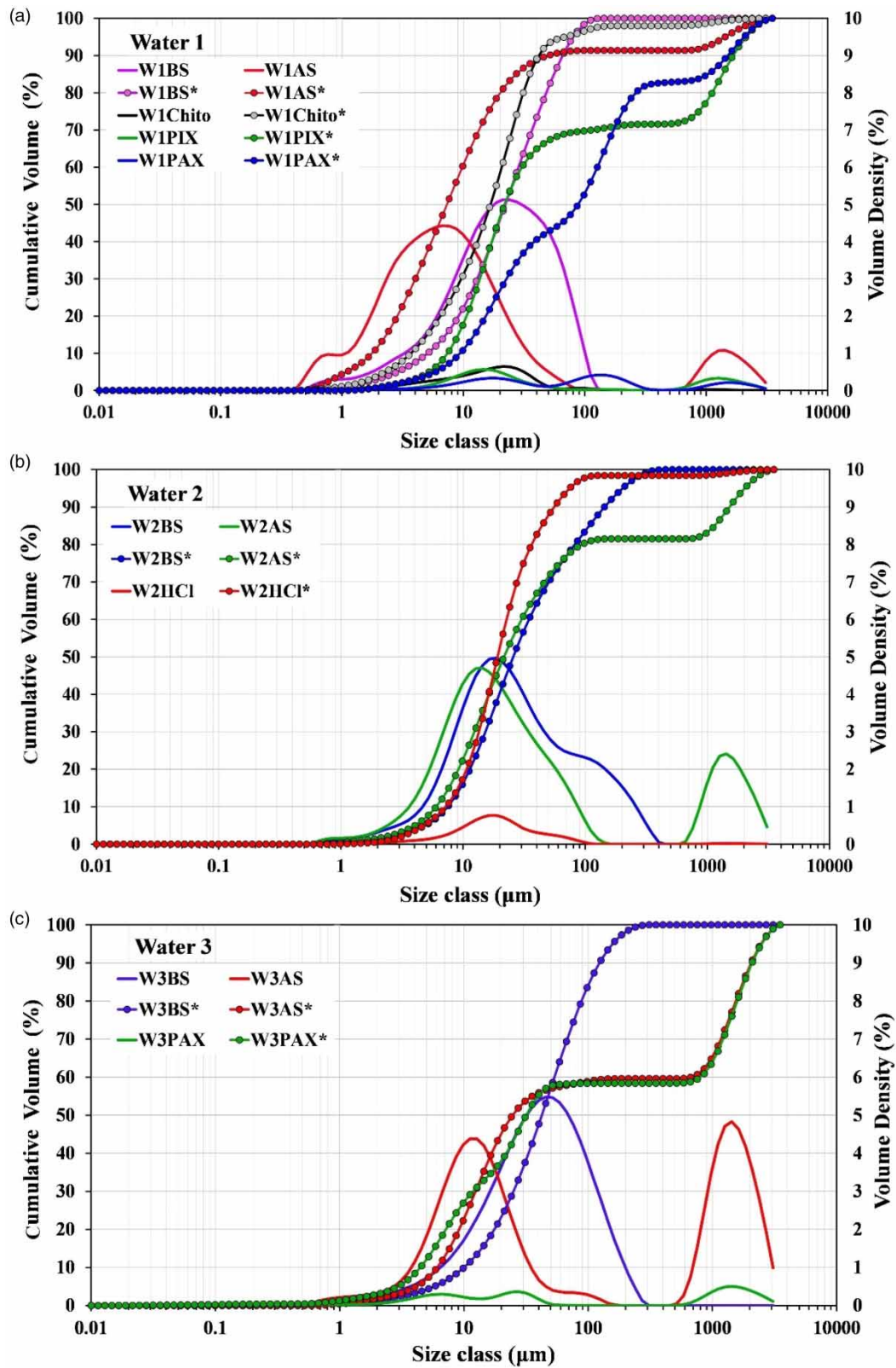


Figure 7 | The cumulative volume distributions (lines with symbols) and the volume density distributions (solid lines) of water 1 (a), water 2 (b), and water 3 (c). Legends: BS, before sedimentation, AF, after sedimentation, HCl, optimum dosage of acid (HCl), Chito, optimum dosage of chitosan, PAX, optimum dosage of PAX, PIX, optimum dosage of PIX, *, cumulative volume distribution.

approximately 100–1,000 μm after sedimentation (W2BS* versus W2AS* in Figure 7(b)). Using 2 mL/L of 32% HCl in the jar tests remarkably decreased the particle volume density (W2AS versus W2HCl in Figure 7(b)). By HCl treatment, the cumulative volume of smaller particles (W2HCl*) increased slightly compared to after sedimentation (W2AS*), while particles $>100 \mu\text{m}$ were removed.

Regarding water 3 (Figure 7(c)), 15 min of sedimentation shifted the PSD toward finer particles. However, a notable portion (approximately 40%) of the particle volume after sedimentation (W3AS) was detected in the particle size range of 500–2,000 μm (W3BS versus W3AS in Figure 7). This percentage of larger and lighter particles, not removed by sedimentation, can be explained by some kind of agglomeration during the 15 min sedimentation period. Similar but more limited peaks in the same particle size range were also detected for water 1 and 2 (Figure 7(a) and 7(b)). The optimum dosage of PAX (2 mg Al/L) sharply reduced the volume density (W3AS versus W3PAX in Figure 7(c)) in the measured particle size classes.

In water 3 (Figure 7(c)), the cumulative volume distribution after 15 min of sedimentation and the distribution of the optimum dosage of PAX were almost identical. This does not indicate a lack of particle removal by PAX treatment, but similar removal in all size fractions. The flattening of the cumulative volume distribution curves after sedimentation (W3AS*) and after PAX coagulation (W3PAX*) in the particle size range of approximately 50–1,000 μm , indicating again that the majority of particles in this size range were removed by sedimentation and PAX coagulation.

In volume-based PSDs, the distributions of particles are often defined by various size classes, e.g., D10, D50, and D90. To each of these parameters, a particle size in μm is specified. These terms are defined as follows:

- D10: 10% of the particle volume in the sample has a diameter smaller than the given diameter.
- D50: 50% of the particle volume in the sample is larger than the specified diameter, while 50% is smaller than the given diameter. This is also known as the median diameter.
- D90: 90% of the particle volume in the sample has a diameter smaller than the given diameter.

The shape of the particle distribution curve can be described by the span, defined as $D90-D10/D50$. In Table 4, values of D10, D50, D90, and the span are given before and after sedimentation and after coagulation. In water 2 and 3, the span was raised substantially after sedimentation, which is in accordance with the distributions shown in Figure 7(b) and 7(c), indicating the presence of larger and lighter particles not removed by sedimentation, which elevated the D90-values.

Regarding water 1, the application of chitosan did not significantly change the span. But PIX and PAX sharply increased the D90 and consequently amplified the span, indicating that smaller particles were removed, and larger particles contributed more to the total particle volumes after coagulation and sedimentation, as shown in Figure 7(a). Contrarily, the acid treatment (addition of 32% HCl) of water 2 decreased both D10 and D90 and consequently reduced the span, which were in accordance with the results shown in Figure 7(b), that most of the particles with diameter $>90 \mu\text{m}$ were removed by acid treatment.

Table 4 | Diameter parameters based on particle volume distribution for the tunneling wastewaters (μm)

Sample ^a	D10	D50	D90	Span ^b
Water 1 before sedimentation	5.07	22.1	65.6	2.7
Water 1 after sedimentation	1.76	7.31	49.8	6.6
Water 1 with chitosan ^c	3.7	16.7	41.8	2.3
Water 1 with PIX ^c	7.38	21.8	1,660	75.8
Water 1 with PAX ^c	9.37	87.9	1,500	17
Water 2 before sedimentation	7.49	25.7	144	5.3
Water 2 after sedimentation	6.03	21.6	1,520	70.1
Water 2 with HCl ^c	7.2	19.2	55.7	2.5
Water 3 before sedimentation	10	42.4	123	2.7
Water 3 after sedimentation	6.10	24.7	2,050	82.7
Water 3 with PAX ^c	4.46	31.8	2,090	65.6

^aAverage of 10 measurements.

^bIn volume-based size distribution, span is defined as $(D90-D10)/D50$.

^cFor the optimum dosage from the jar test.

To analyze the impact of treatment on various particles size ranges, the PSD results were categorized in five intervals, including <1, 1–10, 10–100, 100–1,000, and 1,000–10,000 μm . Figure 8 shows how the treatments (15 min of sedimentation and coagulation and sedimentation) influenced the various particle size ranges.

In water 1, before sedimentation, 74% of the volume density constituted of particles in the size range of 10–100 μm , as shown in Figure 8. By 15 min of settling, the largest portion of the particle volume (59%) was in the range of 1–10 μm . A similar decrease of the particle volume in the size range of 10–100 μm , with an increase in volume in the size range of 1–10 μm , was also shown for water 2 and water 3 after 15 min of sedimentation (Figure 8). In water 3, before sedimentation, the highest volume density was in the range of 10–100 μm (76%). After 15 min of settling, this part was reduced by 43%, while the volume density of particles in the range of 1,000–10,000 μm was 31%, which might be explained by a few large particles with low settling velocity, contributing significantly to the total particle volume. These larger particles were also observed in Figure 7(c).

As discussed earlier, and also shown in Figure 7(a), the application of chitosan, PIX, and PAX in water 1 after 15 min of sedimentation changed the particle distributions. Much of the smaller particles (1–10 μm) were removed, while particles in the range of 10–100 μm , and some non-settled larger particles (100–10,000 μm), contributed more significantly to the total particle volumes (W1AS versus W1Chito, W1PIX, and W1PAX) (Figure 8). In water 3, the optimum dosage of PAX did not change the volume density distribution much (W3PAX versus W3AS) (Figure 8). As pointed out previously, and also seen in Figure 7(c), this does not indicate a lack of particle removal by PAX treatment, but similar removal in all size fractions. Application of diluted HCl (32%) resulted in an increased percentage volume density of particles in the range of 10–100 μm , while larger particles (100–10,000 μm) were removed (W2AS versus W2HCl), as also indicated in Figure 7(b).

The present study indicates some limitations when using PSD and volumetric particle density for evaluating the effect of coagulation and sedimentation. The study indicates significant removal of smaller particles in the range of 1–100 μm by coagulation and sedimentation; however, the samples' volumetric percentage in the range of 100–10,000 μm increased, which might be explained by some few large and lightweight particles not removed by sedimentation. Al- and Fe-coagulants are known to produce voluminous hydroxide particles in water, which are the principal agents for particle removal. However, some of these large unsettled hydroxide flocs may have interfered with the PSD analysis in this study.

The reduced efficiency of conventional sedimentation in removing smaller particles within the 1–100 μm diameter range can be attributed to their lower settling velocities and the presence of stronger repulsive forces between particles. Smaller particles exhibit slower settling rates due to their size and Brownian motion, keeping them suspended for longer periods. Additionally, the Van der Waals forces and electrostatic repulsion between these particles hinder their aggregation into larger flocs, which are crucial for effective sedimentation. Turbulence in the water can further disrupt settling, leading to particle resuspension. In contrast, chemical coagulation addresses these challenges by neutralizing particle charges through coagulants, encouraging the formation of larger and denser flocs that settle more rapidly. This enhanced aggregation process

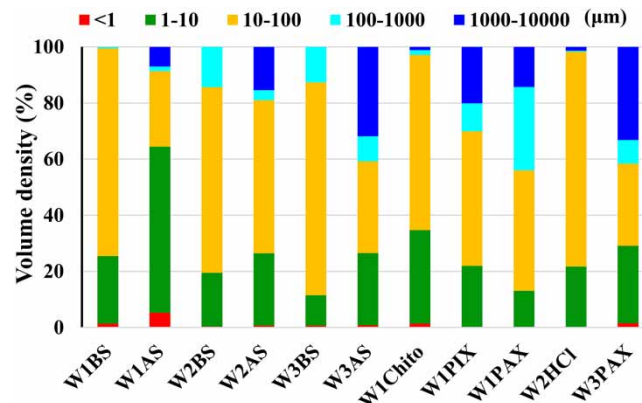


Figure 8 | The effect of water treatment on various particle size ranges in tunneling wastewaters before and after sedimentation, and after coagulation and sedimentation. Legends: W1, water 1, W2, water 2, W3, water 3, BS, before sedimentation, AS, after sedimentation, HCl, optimum dosage of acid (HCl), Chito, optimum dosage of chitosan, PAX, optimum dosage of PAX, PIX, optimum dosage of PIX.

facilitated by chemical coagulation contributes to its superior effectiveness in removing smaller particles compared to conventional sedimentation.

There is a consistency between our results and other studies in the literature. However, the characterization of the tunneling wastewaters are site-specific. For example, *Lee et al. (2013)* used turbidity to evaluate the performance of chitosan in tunneling wastewater treatment. The results showed 98 and 99% removal efficiencies for turbidity and TSS, respectively. Also, *Liltved et al. (2014)*, assessed the chitosan efficiency for the treatment of tunneling wastewater and introduced chitosan as a coagulant with high efficiency at low dosages (82 and 74% for turbidity and TSS), and low sludge production.

4. DISCUSSION

Nyström et al. (2020a) examined the efficiency of coagulation/flocculation treatment on semi-synthetic stormwater characterized by high TSS (averaging 990 mg/L) and turbidity (averaging 800 NTU). Among the five coagulants tested, including alum, two pre-hydrolyzed aluminum coagulants (PAX-215 with a relative basicity of 30% and PAX-XL360 amended with an organic polymer), iron chloride (PIX-111), and chitosan, only iron chloride caused a notable shift in PSD toward larger particles. In our research, we observed that the use of chitosan, PIX, and PAX, in particular, significantly reduced the volume density of particles within the 1–100 μm range, demonstrating the effectiveness of these coagulants in removing particles smaller than 100 μm . Both our findings and *Nyström et al.'s (2020a)* results indicated over 90% removal of particle-based metal(loid)s by the coagulants. Regarding PSD, *Nyström et al. (2020a)* found that sedimentation alone, as we also observed, led to a shift toward smaller particles by removing settleable particles from suspension. When coagulation treatment was applied, it notably decreased the prevalence of smaller particles, especially those smaller than 10 μm in diameter, particularly with iron chloride as the coagulant. Our study showed similar effects, with the application of chitosan, PIX, and PAX leading to a notable decrease in the volume density of particles within the 1–100 μm range, underscoring the efficiency of coagulants in removing particles smaller than 100 μm .

In another study by *Nyström et al. (2020b)*, jar tests were conducted on semi-synthetic stormwater without pH adjustment or alkalinity addition, achieving a high treatment efficiency (over 90% reduction in both turbidity and suspended solids) across various coagulants, including PIX-111, PAX-215, PAX-XL100, PAX-XL360, limewater, alum, drinking water sludge, chitosan, superfloc C491, superfloc C494, superfloc A110 HMW, and superfloc A130 HMW. For low alkalinity waters, the use of chitosan may be advantageous due to minimal alkalinity consumption. The charge neutralization mechanism was identified as the likely coagulation mechanism due to charge reversal observed for all chemicals investigated. In contrast, our research identified PAX-XL-3103D as the most effective coagulant. It's worth noting that *Sun et al. (2019)* suggested that the influence of PSD on particle removal and coagulation mechanisms remains slight at different pH values, with PSD and concentration having more significant impacts on coagulation mechanisms.

5. CONCLUSION

This research's principal goal was to characterize tunneling wastewater and evaluate the performance of sedimentation and different coagulants on particle removal and removal of metals associated with particles. Characterization of the wastewaters before and after treatment was carried out by chemical analysis and various techniques for particle characterization, including PSD, shape, and morphology of particles by a field emission scanning electron microscope (SEM), and particle-associated metals by using X-ray EDS. Laboratory work was conducted to evaluate the effectiveness of coagulants for particle removal from tunneling wastewater. Both natural (i.e., chitosan) and chemical coagulants (i.e., PIX-318 and PAX-XL-3103D) were applied in a conventional jar-test apparatus to remove particles. The following conclusions were derived from the microstructure analysis and jar-test results.

1. Short-time sedimentation alone substantially reduced the particle content (suspended solids and turbidity) and particle-associated pollutants, including metal(loid)s, in the tunneling waters used in this study. However, our results indicate that sedimentation alone is not sufficient as the suspended solid concentration remained higher than the discharge limit of 100 mg/L in all waters, no nitrogen removal was evident, and the concentrations of some metal(loid)s, in particular Cu and Cr, severely exceeded the concentrations for acute toxic effects in freshwater bodies (*Petterson 2020*), indicating that high dilution factors are required when discharged to local freshwater bodies. In addition, pH adjustment is required before discharge.

2. Improved removal efficiencies were demonstrated by chemical coagulation followed by sedimentation. The optimum coagulant dosages in jar tests led to removal efficiencies in the range of 92–99% for turbidity and TSS. The optimum dosages of chitosan, PIX, and PAX for water 1 after pretreatment (15 min sedimentation) were 1 mg/L, 3 mg Fe/L, and 3 mg Al/L, respectively, while a dosage of 2 mg Al/L gave the best results in water 3. It was also evident that acid dosage alone (2 mL/L of 32% HCl) improved particle removal.
3. Fifteen minutes of sedimentation shifted PSDs from coarser-size fractions to finer particles. The size fraction of 10–100 μm was reduced in all three waters, indicating a rapid sedimentation rate for such particles. The application of chitosan, PIX, and PAX particularly decreased the volume density of particles in the range of 1–100 μm showing that coagulants are efficient for the removal of particles smaller than 100 μm .
4. Tunneling water without treatment may contain particles with sharp edges with negative impacts on aquatic biota, especially fishes. Sedimentation and chemical treatment may remove the majority of these particles.

The results of this study suggest that the proposed two-step method (15 min of sedimentation followed by coagulation and sedimentation) is highly efficient for particle removal in tunneling wastewater.

The decision between natural and chemical coagulants depends on a variety of factors including local conditions, water quality requirements, treatment goals, and economic considerations. The choice between natural and chemical coagulants involves a trade-off between cost-effectiveness and environmental sustainability. While natural coagulants offer reduced chemical impact and align well with sustainability goals, chemical coagulants are known for their efficiency and lower immediate costs. Careful consideration of specific project requirements and long-term environmental implications is crucial when deciding which approach to adopt. This study contributes to the characterization of tunneling wastewater and puts a step forward in the understanding of the efficiency of coagulants in particle removal of tunneling wastewater.

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DATA AVAILABILITY STATEMENT

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

CONFLICT OF INTEREST

The authors declare there is no conflict.

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