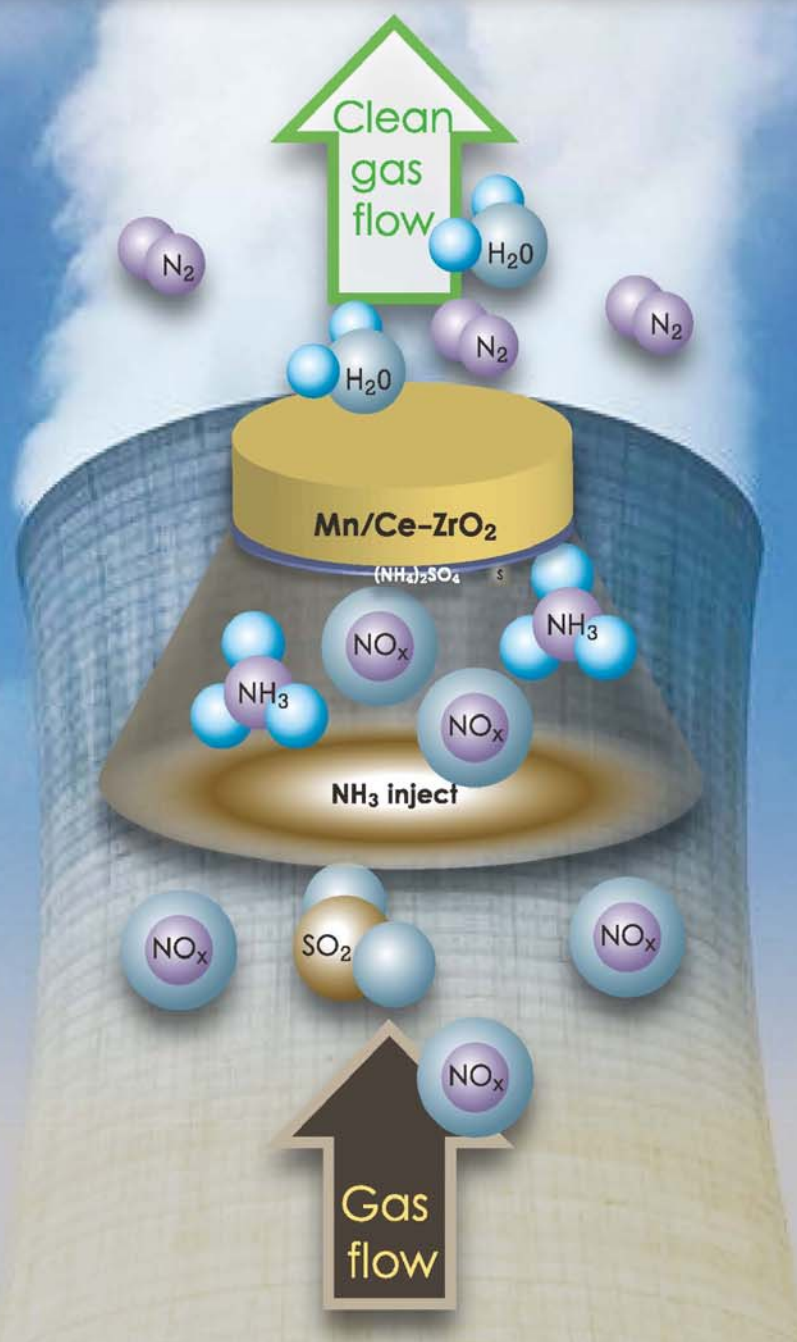


# JES

JOURNAL OF  
ENVIRONMENTAL  
SCIENCES

ISSN 1001-0742  
CN 11-2629/X

April 1, 2013 Volume 25 Number 4  
[www.jesc.ac.cn](http://www.jesc.ac.cn)



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Serial parameter: CN 11-2629/X\*1989\*m\*211\*en\*P\*25\*2013-4



## Optimisation of chemical purification conditions for direct application of solid metal salt coagulants: Treatment of peatland-derived diffuse runoff

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Received 09 July 2012; revised 27 September 2012; accepted 09 October 2012

### Abstract

The drainage of peatland areas for peat extraction, agriculture or bioenergy requires affordable, simple and reliable treatment methods that can purify waters rich in particulates and dissolved organic carbon. This work focused on the optimisation of chemical purification process for the direct dosage of solid metal salt coagulants. It investigated process requirements of solid coagulants and the influence of water quality, temperature and process parameters on their performance. This is the first attempt to provide information on specific process requirements of solid coagulants. Three solid inorganic coagulants were evaluated: aluminium sulphate, ferric sulphate and ferric aluminium sulphate. Pre-dissolved aluminium and ferric sulphate were also tested with the objective of identifying the effects of in-line coagulant dissolution on purification performance. It was determined that the pre-dissolution of the coagulants had a significant effect on coagulant performance and process requirements. Highest purification levels achieved by solid coagulants, even at 30% higher dosages, were generally lower (5%–30%) than those achieved by pre-dissolved coagulants. Furthermore, the mixing requirements of coagulants pre-dissolved prior to addition differed substantially from those of solid coagulants. The pH of the water samples being purified had a major influence on coagulant dosage and purification efficiency. Ferric sulphate (70 mg/L) was found to be the best performing solid coagulant achieving the following load removals: suspended solids (59%–88%), total organic carbon (56%–62%), total phosphorus (87%–90%), phosphate phosphorus (85%–92%) and total nitrogen (33%–44%). The results show that the use of solid coagulants is a viable option for the treatment of peatland-derived runoff water if solid coagulant-specific process requirements, such as mixing and settling time, are considered.

**Key words:** inorganic solid coagulant; peatland runoff; mixing; chemical purification; nutrients; humic substances

**DOI:** 10.1016/S1001-0742(12)60111-9

### Introduction

The deterioration of surface water quality due to non-point source pollution is a significant issue faced all over the world. In regions such as the boreal zone and the tropics with large peat deposits, the drainage of peatland areas for peat extraction, agriculture and forestry has long been cause for concern. Large land areas in Finland, Estonia, Canada, Ireland and Eastern Europe are exploited for peat extraction, mainly for energy purposes. In Finland, peat provides around 7% of the country's primary energy requirements (Energiatollisuus, 2007). Peat extraction activities such as drainage and the exposure of peat layers are known to increase the amount of water discharging from catchment areas (Holden et al., 2006) and also to increase the leaching of pollutant substances

such as suspended solids (SS) dissolved organic carbon (DOC), metals and nutrients into watercourses located downstream (Heikkinen, 1994; Kløve, 2001; Marttila and Kløve, 2008). Leaching of these substances into sensitive water bodies can cause adverse impacts such as eutrophication, siltation and loss of biodiversity (Marja-aho and Koskinen, 1989; Selin et al., 1994). Over recent decades, several treatment methods have been developed to reduce loading from peat extraction runoff, among them are the chemical treatment, overland flow fields and peak runoff control dams (Kløve, 1997; Ronkanen and Kløve, 2007; IPS, 2009). Nevertheless, the advances made with these pollution control measures fall short of sustaining the overall load reduction levels expected by environmental authorities (Rekolainen et al., 2006). Further development of suitable water purification methods is therefore required to reduce the load imposed by the drainage of peatland

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areas and other peat extraction activities.

Chemical treatment is considered by the Finnish authorities as one of the best available technologies for the purification of peat extraction runoff water. However, little research has been applied to the development of this treatment method for the purification of non-point source pollution. Due to the high costs involved in the implementation and maintenance of chemical treatment, field application of this method has been restricted to large peat production areas or sites located upstream of sensitive water bodies (Kløve, 1997). The high costs attributed to chemical purification are mostly associated with the implementation and maintenance of conventional treatment stations using liquid pre-hydrolysed coagulants. However, direct dosage of solid metal salt coagulants can greatly reduce the costs of chemical purification. The total costs of the implementation of small scale treatment facilities using solid coagulants were found to be from 50% to 75% lower than for the implementation of facilities using liquid pre-hydrolysed coagulants (Alatalo and Peronius, 2004). Solid coagulants are cheaper and require fewer and simpler dosing equipment and consequently fewer maintenance working hours than liquid coagulants. Currently, solid metal salts of iron are being used in several water treatment facilities at Finnish peat extraction sites. However, the direct addition of solid coagulants restricts dosage control. This, together with variations in runoff water quality and the lack of improvement in rudimentary treatment facilities, has led to the application of high coagulant dosages, fluctuations in purification efficiency and an increase in metal concentration in the discharge waters (Heiderscheidt, 2011). The lack of studies and consequently of information relating to the process requirements of solid coagulants and the influence of inline dissolution of applied chemicals have prevented improvements to purification conditions to date.

Chemical purification is based on the ability of the chemicals to precipitate SS and dissolved substances present in the water (Gregory and Duan, 2001; Demirata et al., 2002; Duan and Gregory, 2003; Sinha et al., 2004; Pernitsky and Edzwald, 2006). It consists of three individual but well-interconnected processes: coagulation, flocculation and sedimentation. The larger suspended particulates present in the runoff water can be easily removed via natural sedimentation. However, dissolved substances and very fine particles or colloids cannot be removed via sedimentation alone and their removal is the main objective of chemical treatment (Koohestanian et al., 2008). Factors known to influence the efficiency of chemical purification include water quality characteristics such as temperature, alkalinity, pH and the concentration and type of pollutant substances (Volk et al., 2000; Franceschi et al., 2002; Sinha et al., 2004; Sansalone and Kim, 2008; Yan et al., 2008) as well as process parameters such as coagulant type (Lee et al., 1998; Liu and Chin, 2009; Zhao et al., 2009, 2012),

dosage and mixing during and after coagulant addition (Dentel, 1988; Rossini et al., 1998; Demirata et al., 2002; Exall and vanLoon, 2003; Zhan et al., 2011).

The aim of the present study was to develop the chemical treatment of non-point source pollution. More specifically, it examined the use of solid inorganic coagulants as purification agents in the treatment of peatland-derived runoff water. Three solid metal salts were tested (aluminium sulphate ( $\text{Al}_2(\text{SO}_4)_3$ ), ferric sulphate ( $\text{Fe}_2(\text{SO}_4)_3$ ) and ferric aluminium sulphate ( $\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$ ). The objective was to study, through a series of laboratory experiments, the purification efficiency achieved by direct addition of the solid coagulants to water samples collected from different peat extraction sites. The evaluation of obtained purification efficiency was based on the percentage removal of concerning substances such as phosphorous, nitrogen, SS and organic matter. Furthermore, the goal was to evaluate the influence of water quality, temperature and process parameters on coagulant performance, and the influence of coagulant dissolution on treatment efficiency and process requirements such as mixing and settling time.

## 1 Materials and methods

Water samples were collected between August and October 2010 from two peat extraction sites in Finland: Sample 1: Vaala (3475877N, 7138725E); Sample 2: Haapavesi (3431984N, 7116451E). The observed water quality characteristics of the samples (Table 1) were typical for peat-derived runoff (Kløve, 2001). Three commercial quality solid metal salts (Kemira Oyj, Kemwater; 90% purity) were studied:  $\text{Al}_2(\text{SO}_4)_3$ ,  $\text{Fe}_2(\text{SO}_4)_3$ , and  $\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$  (commercially available product composed of a mixture of 15% ferric sulphate and 85% aluminium sulphate). Purification conditions were also evaluated for pre-dissolved (pd, stock solutions 10 g/L) aluminium sulphate and ferric sulphate with the objective of evaluating the effects of coagulant dissolution on treatment performance.

The purification efficiency achieved under different process parameters was first monitored and evaluated via

**Table 1** Water quality characteristics of samples used in purification tests

Water quality parameter	Sample 1	<i>n</i>	Sample 2	<i>n</i>
pH	6.5–6.8	5	5.5–6.3	8
EC (mS/m)	6.2–7.4	5	7.2–7.4	8
SS (mg/L)	17.1 ± 3.9	9	1 8.8 ± 1.5	8
tot-N (µg/L)	1720 ± 148	9	2000 ± 0	6
tot-P (µg/L)	58 ± 5.6	9	61 ± 2.4	8
PO <sub>4</sub> -P (µg/L)	24 ± 2.5	9	21 ± 2.8	8
TOC (mg/L)	27 ± 1.5	9	27.5 ± 0.5	8
Fe (µg/L)	3830 ± 132.3	9	2150 ± 50	4
Al (µg/L)	423 ± 59.6	9	730 ± 59.1	4

Data expressed as mean ± standard deviation; *n* is the number of analyses.

measurements of colour, turbidity, pH, temperature and electric conductivity (EC). Samples of the purified water were then further analysed for SS, total phosphorous (tot-P), phosphate phosphorous (PO<sub>4</sub>-P), total nitrogen (tot-N) and total organic carbon (TOC). Standardized (SFS and ISO) analytical methods were used.

Jar tests were performed using the six-jar programmable paddle stirrer equipment Flocculator 2000 (Kemira Kemwater). Laboratory experiments were carried out in 4 stages: (1) Determination of optimum dosage and purification efficiency at 20 ± 2°C (3 replications). Two additional replications were carried out for sample 2 after a pH increase from 5.80 to 6.85 via the addition of slaked lime (Ca(OH)<sub>2</sub>). (2) Settling characteristics at 20 ± 2°C of the flocs formed: Water samples were collected during the sedimentation period for turbidity analyses (2 replications). (3) Influence of temperature on purification efficiency and on settling characteristics: The jar test equipment was transferred to a temperature-controlled (5 ± 1°C) insulated water tank (2 replications). (4) Influence of the four mixing parameters (slow mixing time, slow mixing speed, fast mixing time and fast mixing speed) on purification efficiency was evaluated using the single factor method of optimization (Cochran and Cox, 1957; Rossini et al., 1999). The tests were performed in sequential stages where in each stage three parameters were kept constant while the fourth parameter was systematically varied (**Table 2**). The value corresponding to the parameter variation which achieved the highest removal of turbidity was selected and kept constant in the stages that followed. Two replications of each test were executed where turbidity and colour measurements were performed. Samples from the replication with the highest removal of turbidity were further analysed for SS, tot-P, PO<sub>4</sub>-P, tot-N and TOC.

Constant mixing parameters, here referred to as nominal mixing conditions, were applied during experiment phases 1, 2 and 3. Solid coagulants: 400 rpm for 60 sec, 70 rpm for 15 min followed by 30 min of sedimentation; pre-dissolved coagulants: 300 rpm for 10 sec, 50 rpm for 25 min followed by 30 min of sedimentation. The root mean square velocity gradient ( $G$ ) induced by the mixing parameters applied was determined using Eq. (1) (Bratby, 2006).

$$G = \sqrt{\frac{W}{\mu}} \quad (1)$$

**Table 2** Variations applied to mixing parameters and selected values.

Parameter	Applied variations	Selected values according to turbidity removal	
		Solid	Pre-dissolved
Slow mix speed (rpm)	20, 50, 70, 90	70	50
Slow mix time (min)	5, 10, 15, 25	25	25
Fast mix speed (rpm)	100, 200, 300, 400	60	10
Fast mix time (sec)	10, 60, 120	400	300

where,  $W$  (N/m<sup>2</sup>) =  $P/V$  is the power dissipation function,  $\mu$  (NS/m<sup>2</sup>) is liquid viscosity,  $V$  (m<sup>3</sup>) is volume of mixing tank. For a mechanical mixer with rotating blades, the power  $P$  (Nm/sec) drawn by the device is determined by its rotational speed and the geometry of the tank in which it operates ( $P = \phi \times \rho \times n^3 \times D^5$ ), where  $\phi$  is the mixer dimensionless power number related to the type and dimension of mixer as well as tank geometry,  $\rho$  (kg/m<sup>3</sup>) is liquid density. For the equipment used  $\phi = 3.5$  extracted from Bratby (2006);  $n$  is the mixer rotational speed in revolutions per second; and  $D$  (m) is the diameter of the mixer impeller.

## 2 Results

### 2.1 Purification efficiency and dosage requirements

Based on the results obtained, under the laboratory test conditions applied (nominal mixing and 20°C); all coagulants achieved high load reduction levels (**Fig. 1**). Among the coagulants tested, Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> was the best performing salt and produced the highest removal levels both in solid and pre-dissolved form. The direct addition of solid Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> to both water samples produced excellent removal of tot-P, PO<sub>4</sub>-P (> 85%) and SS (> 80%). When applied to water sample 2, even at 50% lower dosages than the other solid coagulants, Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> achieved considerably higher purification efficiency. It removed around 20% more tot-P and PO<sub>4</sub>-P, 15% more tot-N, and over 30% more TOC than the other solid coagulants. The purification efficiencies achieved while treating water sample 1 were generally higher than those achieved while treating sample 2, especially for aluminium-based coagulants.

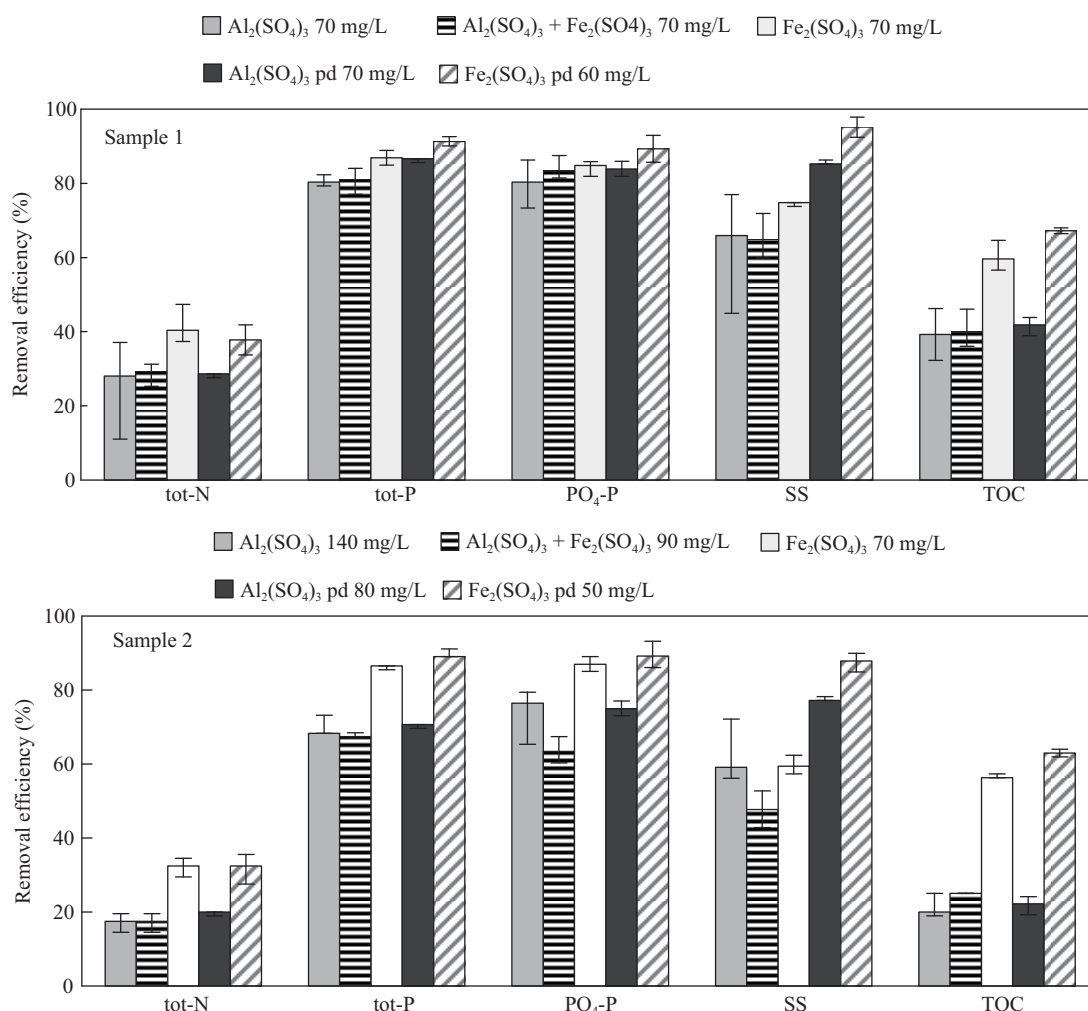
Generally, the removal of SS was significantly higher when pre-dissolved coagulants were applied (< 30%), while the removal of the other substances analysed was only slightly higher (< 5%). The identified optimum dosages of pre-dissolved coagulants were up to 30% lower than those identified for solid coagulants (**Table 3**). Pre-dissolved coagulants also displayed lower variations in purification efficiencies between experiment replications (**Fig. 1**).

While identifying the coagulant's optimum dosage, the removal of turbidity and colour with increasing dosage was evaluated. The observed patterns of turbidity and colour removal with increasing dosages differed greatly among the salts tested (**Fig. 2**) but were very similar among individual coagulants when applied to water samples 1 and 2. When increasing dosages of Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> were applied, the samples turbidity and colour steadily decreased until the optimum dosage was reached. After this point, colour and turbidity values remained mostly constant. For Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>, constant or slightly higher than the initial colour and turbidity values were observed until the optimum dosage was applied when a sharp decrease in the sample's colour and turbidity occurred. However, high removal efficiencies

**Table 3** Optimum dosage range identified and selected optimum dosage for tested coagulants

Water sample	Coagulant	Optimum dosage range (mg/L)	Selected optimum dosage (mg/L)	Resulting pH (optimum dosage)
1	$\text{Al}_2(\text{SO}_4)_3$	60–80	70	4.6–4.7
1	$\text{Al}_2(\text{SO}_4)_3$ pd	60–80	70	4.6–4.8
1	$\text{Fe}_2(\text{SO}_4)_3$	60–80	70	3.8–3.9
1	$\text{Fe}_2(\text{SO}_4)_3$ pd	50–70	60	4.0–4.2
1	$\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$	70–90	70	4.5–4.6
2	$\text{Al}_2(\text{SO}_4)_3$	90–140	140	4.1–4.2
2	$\text{Al}_2(\text{SO}_4)_3$ pd	70–90	80	3.9–4.0
2	$\text{Fe}_2(\text{SO}_4)_3$	50–70	70	3.5–3.6
2	$\text{Fe}_2(\text{SO}_4)_3$ pd	50–70	50	3.7–3.8
2	$\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$	80–90	90	4.0–4.2

pd: pre-dissolved coagulant.

**Fig. 1** Average removal efficiency (including maximum and minimum values of experiment replications represented by error bars) for optimum dosages of solid and pre-dissolved coagulants in the purification of water samples 1 and 2.

were only observed for a narrow dosage range of about 20 mg/L. When the dosage was increased above the observed optimum range, it resulted in a sharp increase in turbidity and to a lesser extent in colour values. The mix coagulant  $\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$  displayed a combination of the behavioural patterns of its constituent aluminium- and iron-based coagulants.

The behaviour of the pre-dissolved coagulants, ( $\text{Al}_2(\text{SO}_4)_3$  and  $\text{Fe}_2(\text{SO}_4)_3$ ), mostly reflected that of their solid counterparts. However, the removal efficiencies obtained were higher overall and were achieved at lower dosages for pre-dissolved coagulants. It was also noticeable that while  $\text{Fe}_2(\text{SO}_4)_3$  in solid form presented a narrow optimum dosage range, with a sharp increase in



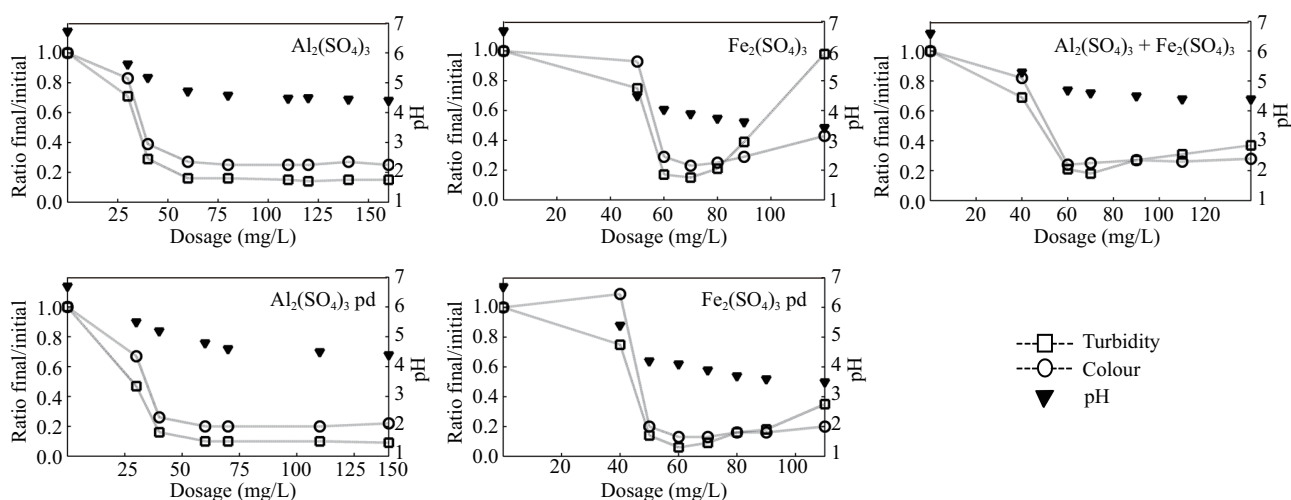


Fig. 2 Removal efficiency of turbidity and colour with increasing dosage of solid and pre-dissolved (pd) coagulants in the purification of water sample 1.

turbidity and colour above optimum dosage, the increases in turbidity and colour observed for pre-dissolved ferric sulphate could be described as steady, but not sharp.

## 2.2 Settling characteristics

The settling characteristics of the individual coagulants represent their ability to coagulate the pollutants present in the water and also their ability to form suitable flocs, which provide short sedimentation times and clarified supernatant water. Among the solid coagulants,  $\text{Fe}_2(\text{SO}_4)_3$  produced the fastest sedimentation rates and overall higher clarification of the supernatant water, achieving a value of turbidity at time  $t$ /initial turbidity = 0.2 or 80% of initial turbidity removal after 4 min of sedimentation for samples 1 and 2 (Fig. 3). While  $\text{Al}_2(\text{SO}_4)_3$  and  $\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$  required between 11 and 25 min. The settling characteristics of the pre-dissolved salts mostly reflected the patterns observed for the solid coagulants. However, some differ-

ences were apparent at the beginning of sedimentation, when the solid coagulants appeared to display somewhat faster settling rates, and at the end of sedimentation, when the pre-dissolved coagulants displayed slightly better supernatant water clarification (Fig. 3).

## 2.3 Influence of temperature

Decreasing test temperature from 20 to 5°C had a negative influence on the removal efficiencies or purification levels achieved by coagulants in solid and pre-dissolved form (Fig. 4). The magnitude of this influence depended on the coagulant type, its physical form and applied dosage, the substance being removed and the water quality. Generally, low temperature had only a small influence on the removal of phosphorous, nitrogen and organic matter for pre-dissolved and solid coagulants (Fig. 4) however; the removal of SS was significantly affected specially for coagulants in solid form. The detrimental effects of

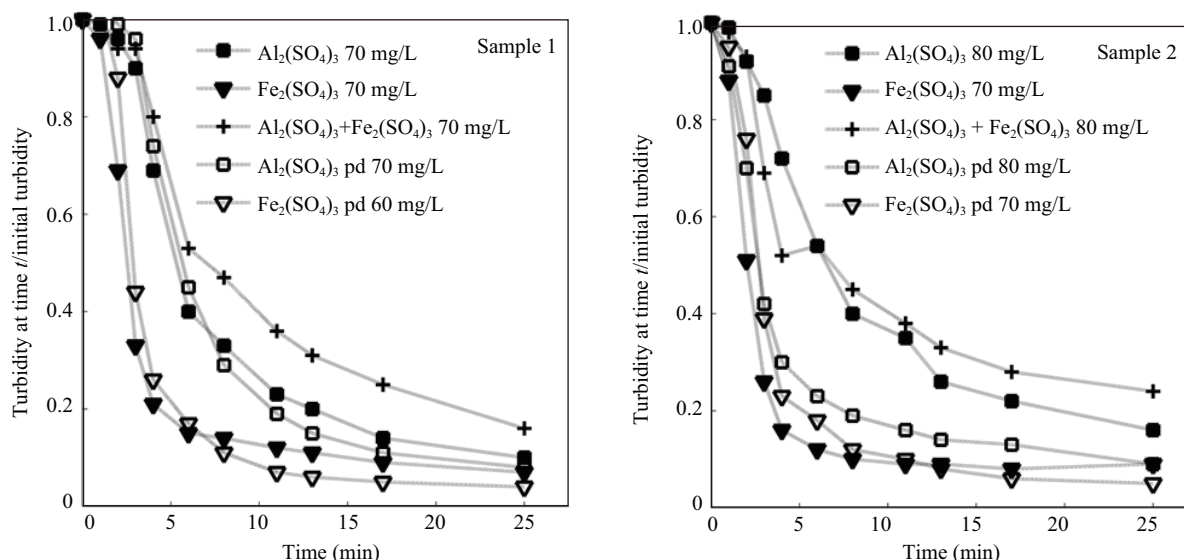
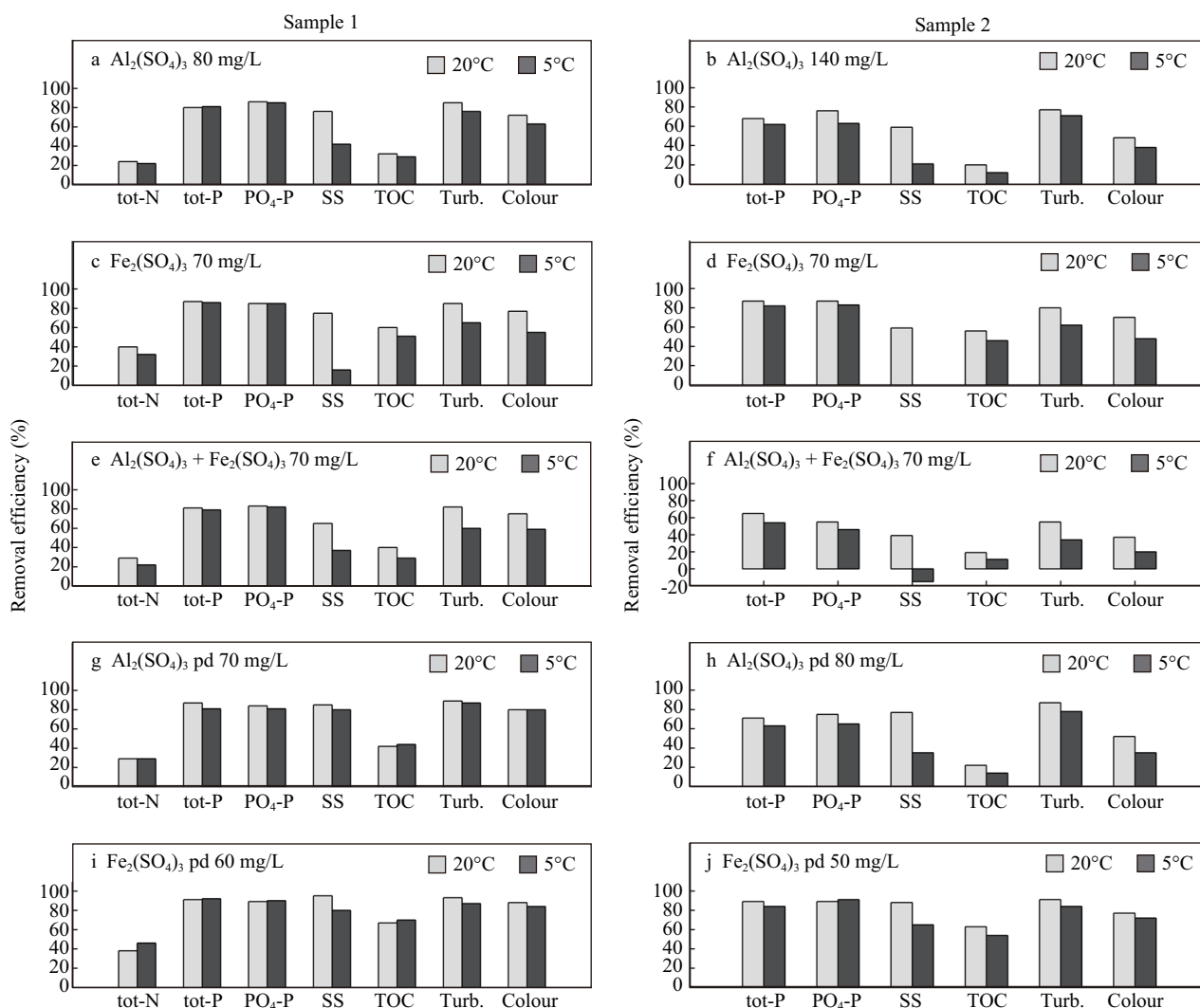


Fig. 3 Settling characteristics of coagulants (removal of turbidity as a function of time) during the purification of water samples 1 and 2.





**Fig. 4** Influence of temperature on removal efficiencies achieved by coagulants tested in solid and in pre-dissolved form in the purification of water samples 1 and 2.

temperature were more pronounced in the purification of water sample 2 than sample 1.

The removal of SS by solid  $\text{Fe}_2(\text{SO}_4)_3$  was significantly impaired at 5°C, where it was about 60% lower than at 20°C for both water samples. When applied to water sample 2,  $\text{Fe}_2(\text{SO}_4)_3$  presented 0% removal efficiency, meaning that it did not reduce the initial SS concentration of the sample. The addition of  $\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$  to water sample 2 at 5°C actually increased the initial SS concentration of the sample by 15% (Fig. 4f). The SS removal by  $\text{Al}_2(\text{SO}_4)_3$  at 5°C was about 40% lower than at 20°C for both water samples. Pre-dissolved coagulants remove between 15% and 30% less SS at low temperature (Fig. 4g–j).

#### 2.4 Optimisation of mixing parameters

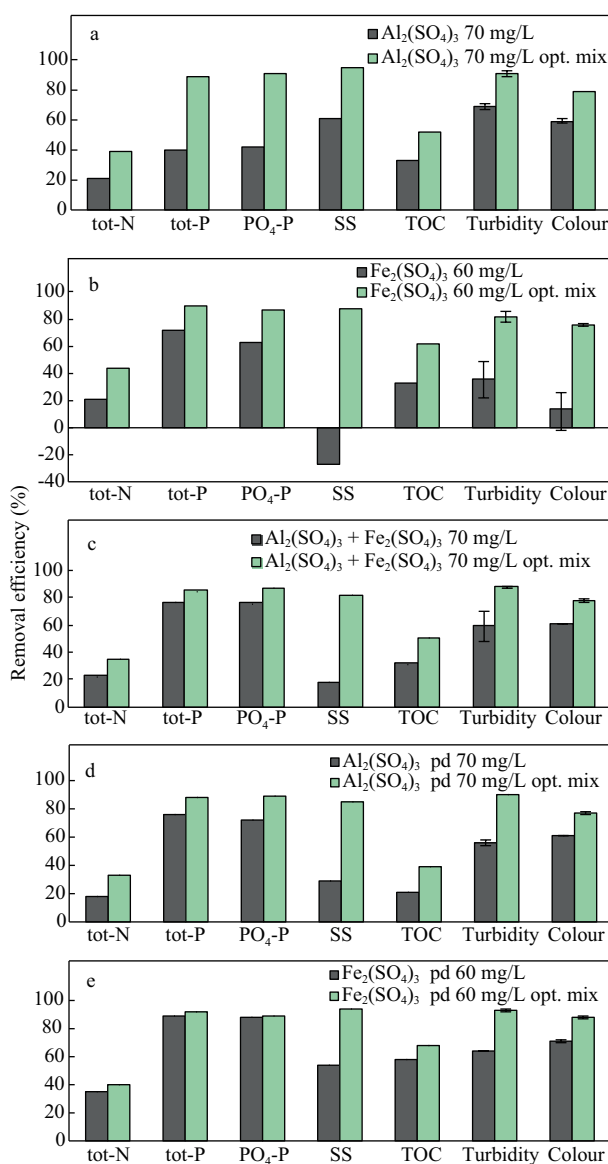
Varying mixing time and speed during the coagulation and flocculation stages of the purification process had a significant influence on the removal efficiencies achieved.

The presented results (Fig. 5) represent a comparison between the purification efficiency obtained in the treatment of water sample 1 when optimum and most unsatisfactory mixing parameters were applied (Table 4). Herein, optimum mixing refers to the combination of parameters that achieved the highest removal efficiencies, while unsatisfactory mixing refers to the parameters that resulted in the lowest removal efficiencies. Generally the lowest removal efficiencies were observed when either short mixing time or low mixing speed was applied during the flocculation stage of the purification process (Table 4).

Regarding the results obtained while testing the solid coagulants, low mixing speed of 20 rpm, even when combined with mixing times inside the optimum range, produced removal efficiencies up to 80% lower than with the optimum speed of 70 rpm. Short flocculation mixing times of 5 min appeared to have an even greater influence on purification, especially in the removal of SS, where the fluctuations in removal efficiency compared with optimum

**Table 4** Optimum and most unsatisfactory mixing parameters identified (among tested) and calculated velocity gradient ( $G$ ) values

Coagulant	Mixing conditions	Dosage (mg/L)	Rapid mixing time (sec)	Rapid mixing speed (rpm)	$G$ ( $\text{sec}^{-1}$ )	Slow mixing time (min)	Slow mixing speed (rpm)	$G$ ( $\text{sec}^{-1}$ )
$\text{Al}_2(\text{SO}_4)_3$	Optimum	70	10	400	756	25	70	55
	Unsatisfactory	70	60	400	756	15	20	9
$\text{Fe}_2(\text{SO}_4)_3$	Optimum	60	10	400	756	25	70	55
	Unsatisfactory	60	60	400	756	5	70	55
$\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$	Optimum	70	10	400	756	25	70	55
	Unsatisfactory	70	60	400	756	15	20	9
$\text{Al}_2(\text{SO}_4)_3$ pd	Optimum	70	10	100	95	25	50	34
	Unsatisfactory	70	60	400	756	5	50	34
$\text{Fe}_2(\text{SO}_4)_3$ pd	Optimum	60	10	300	491	25	50	34
	Unsatisfactory	60	60	400	756	5	50	34

**Fig. 5** Comparison of removal efficiency values achieved for the purification of water sample 1 following the application of the optimum (op. mix) and unsatisfactory mixing parameters identified.

mixing conditions were greater than 100% (**Fig. 5**). The mixing requirements or the optimum mixing conditions identified for the pre-dissolved coagulants differed greatly

from those identified for the solid coagulants (**Table 4**). However, low mixing speeds and short mixing times in the flocculation stage also produced significant fluctuations in the removal efficiencies of pre-dissolved coagulants (**Fig. 5d** and **e**); the fluctuations were less pronounced than those observed for solid metal salts.

### 3 Discussion

Chemical treatment is considered a suitable purification method for peatland-derived runoff water due to the potentially high removal of dissolved organic carbon, phosphorus and SS. The removal of particulate and dissolved organic compounds is important to prevent oxygen consumption in incomplete spring overturn mixing or long winters (Keskitalo and Eloranta, 1999). De-oxygenation can cause internal eutrophication and decimate e.g. fish populations in the receiving water bodies. The removal of phosphorus and SS is particularly desirable in regions of sensitive freshwater systems. Phosphorus is often the growth-limiting factor for plants and microorganisms in most rivers and lakes in Finland and, together with the deposition of SS, is the main factor causing eutrophication, siltation and water quality deterioration.

However, as confirmed by our results, chemical treatment is highly dependent on a series of process and water quality parameters. It was found that high purification levels, in particular for coagulants dosed in solid form, can only be achieved when process parameters such as mixing and dosage are tightly controlled and when the influence of physicochemical characteristics of the incoming water, such as pH and temperature, is known and taken under consideration during optimisation of the purification process.

#### 3.1 Purification efficiency

The purification efficiencies obtained by all coagulants (20°C and nominal mixing conditions) were high (20%–45% tot-N; 70%–92% tot-P; 65%–92% PO<sub>4</sub>-P; 20%–68% TOC; 50%–95% SS) and inside the load removal levels expected for chemical purification of peat-derived runoff water (30%–90% SS; 30%–60% tot-N and 75%–95% tot-P) (IPS, 2009). However, significant variations in

purification efficiencies were observed among the water samples being purified, as well as between coagulant type and physical form (solid or stock solution).

The addition of the coagulants optimum dosages resulted in treated water samples with low pH values: sample 1: Aluminium-based coagulants pH 4.6–4.8, iron-based pH 3.8–4.2; sample 2: Aluminium-based coagulants pH 3.9–4.2, iron-based pH 3.5–3.8. The observed resurgence of turbidity and colour in the samples treated with increasing dosages of  $\text{Fe}_2(\text{SO}_4)_3$  and  $\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$ , during the determination of coagulant dosage requirements was most likely the result of coagulant over dosage and low purification pH which lead to surface charge reversal and re-stabilisation of the colloidal suspension (Poon and Chu, 1999; Sansalone and Kim, 2008). The higher observed removal of TOC (**Fig. 1**) by  $\text{Fe}_2(\text{SO}_4)_3$  (60%–70%) than by  $\text{Al}_2(\text{SO}_4)_3$  (40%–50%) can be attributed to the slightly favourable pH conditions for the removal of organic matter by iron salts (Aguiar et al., 1996; Huang and Shiu, 1996; Gregor et al., 1997; Volk et al., 2000; Cheng, 2002; Matilainen et al., 2010). Furthermore, a significant number of studies have found that iron salts perform somewhat better (10%–15%) in the removal of organic matter than aluminium salts (Volk et al., 2000; Matilainen et al., 2005, 2010).

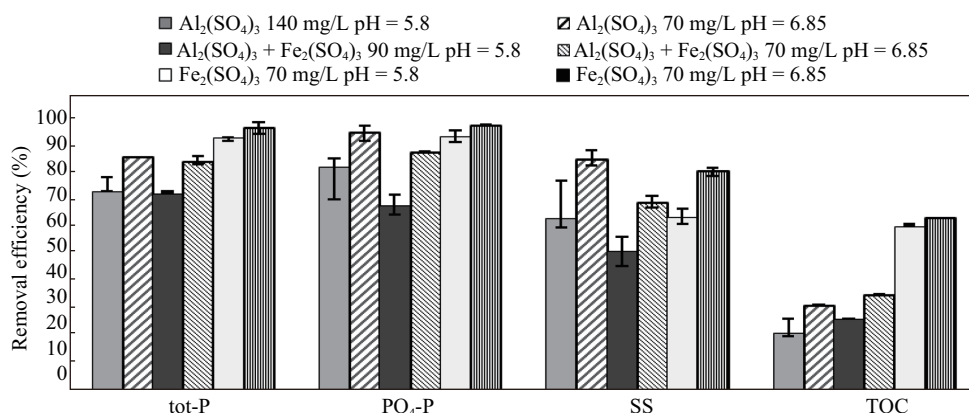
When comparing the removal efficiencies achieved by solid and pre-dissolved coagulants the most distinguishable difference is the removal of SS; which was up to 30% higher when pre-dissolved coagulants were applied. The settling characteristics of solid coagulants differed significantly from those of pre-dissolved. Samples treated with solid coagulants presented slower settling rates and higher turbidity of the supernatant water. These indicate that the required inline dissolution of solid coagulants had a significant influence on the flocculation stage and outcome of the purification process. The addition of solid coagulants appears to have led to the formation of smaller flocs which remained in suspension, resulting in slower settling rates and higher SS concentration in the supernatant water.

### 3.2 Influence of pH

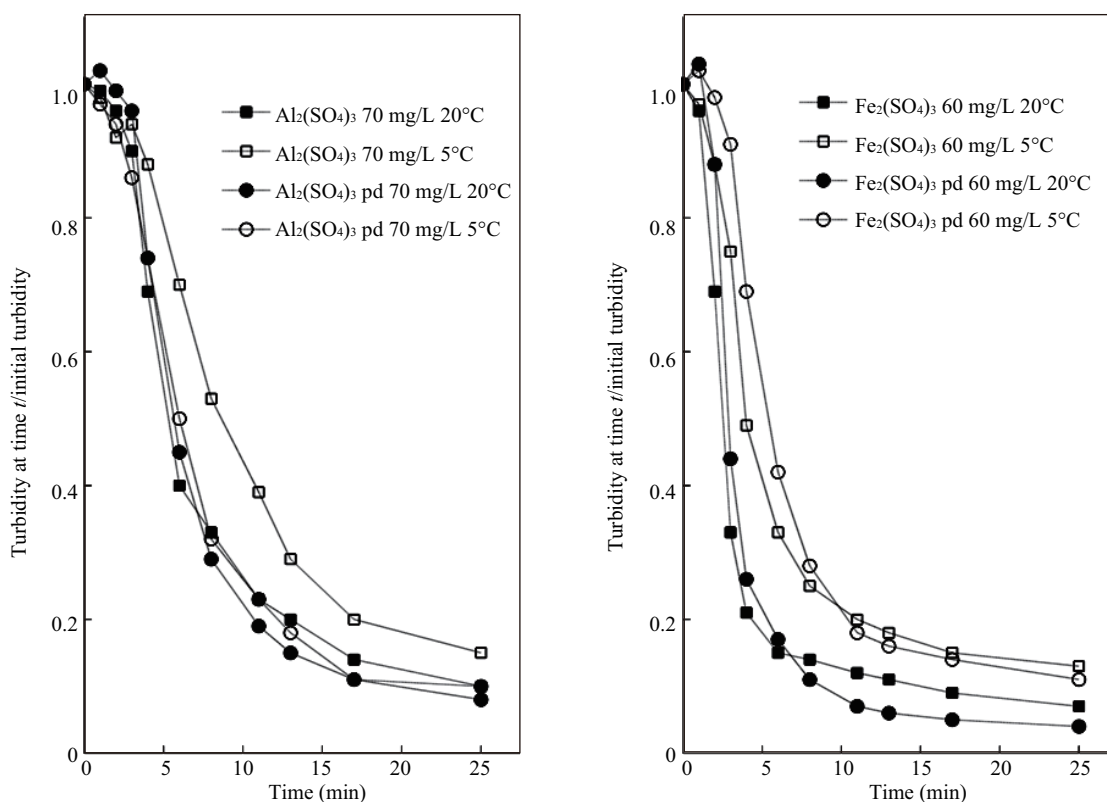
Coagulation pH is considered to be one of the most important variables affecting chemical purification performance. It affects the dissolution and chemistry of the coagulant, as well as the charge presented by the colloidal particles and organic substances in solution (Gregor et al., 1997; Volk et al., 2000; Cheng, 2002; Pernitsky and Edzwald, 2006; Saukkoriipi and Laasonen, 2008; Jiang and Wang, 2009; Wang et al., 2010). Consequently it affects the mechanism of removal of these pollutant substances and purification performance (Dentel, 1988; Demirata et al., 2001; Gregory and Duan, 2002; Sansalone and Kim, 2008; Yan et al., 2008).

It was assumed that the overall lower removal efficiencies achieved, especially by aluminium-based coagulants, in the purification of sample 2 were strongly dependent on the small but significant difference in pH values between the samples (sample 1 pH around 6.80, sample 2 pH around 5.8). To test this assumption, the pH of sample 2 was raised from 5.8 to around 6.80 and purification experiments were then run using the solid coagulants. The results (**Fig. 6**) showed that, the overall removal efficiencies increased, especially for SS, in the purification of sample 2 with increased pH (6.85). The dosage requirements of aluminium-based coagulants were greatly reduced, the optimum dosage of solid  $\text{Al}_2(\text{SO}_4)_3$  decreased from 140 to 70 mg/L. The results indicate a strong influence of pH on the dissolution of the solid coagulants, which had a direct effect on dosage requirements and purification efficiencies achieved. This statement is further confirmed by the fact that pre-dissolved coagulants presented similar performances at similar dosages in the purification of water samples 1 and 2 at initial pH (**Fig. 1**).

Overall, the initially lower pH of water sample 2 had a smaller effect on the purification levels and dosage requirements of iron based coagulants. This might be due to the fact that positively charged hydrolysis species, crucial for an effective coagulation process, are known to be present in a much wider pH range for iron than



**Fig. 6** Comparison of average removal efficiency of tot-P, PO<sub>4</sub>-P, SS and TOC (including maximum and minimum values of experiment replications represented by error bars) for the purification of water sample 2 at field pH (5.8) and raised pH (6.85).



**Fig. 7** Influence of temperature on settling characteristics of solid and pre-dissolved  $\text{Al}_2(\text{SO}_4)_3$  and  $\text{Fe}_2(\text{SO}_4)_3$  in the purification of water sample 1.

for aluminium salts (Demirata et al., 2001; Gregory and Duan, 2001). As expected, the removal of SS was higher (20%) in the purification of water samples 1 and 2 with increased pH. At pH conditions which are closer to neutral the amorphous iron and aluminium hydroxide precipitates have minimum solubility, favouring the sweep coagulation mechanism and enhancing the removal of SS (Demirata et al., 2002; Sansalone and Kim, 2008; Yan et al., 2008).

### 3.3 Influence of temperature

Low temperatures are known to affect the chemical purification process in several ways by altering coagulant solubility, increasing water viscosity and hindering hydrolysis reaction kinetics and the subsequent particle aggregation (Kang and Cleasby, 1995; Xiao et al., 2008a, 2008b, 2009). The most significant influence of low temperatures is nevertheless believed to occur during the flocculation and sedimentation stages of the chemical purification process. Low temperatures have been found to decrease floc aggregation rates (Kang and Cleasby, 1995; Xiao et al., 2008b) and have a detrimental effect on the physical characteristics of the flocs formed (Xiao et al., 2009). Agreeing with the aforementioned statement, our results showed a significant decrease in the removal of SS at low temperature for all coagulants. Furthermore, smaller and less compact flocs were observed during our experiments at 5°C, particularly in samples treated with solid coagulants. These indicate a clear influence of temperature on floc formation and sedimentation. There

was nevertheless, a distinct difference between the performances of pre-dissolved and solid coagulants. While the pre-dissolved coagulants presented up to 30% decrease in SS removal at 5°C compared to 20°C, solid coagulants presented decreases of up to 70%. This significant difference points to a direct influence of temperature on the solubility of solid coagulants.

The temperature of the water influences the actual settling of the flocs, since it has a direct effect on physical water properties such as density and viscosity. Settling tests carried out at 5°C showed a significant delay in flocs settling rates for both solid and pre-dissolved coagulants (Fig. 7). The effects of low temperature were most evident in the first minutes (0 to 6) and most intense period of settling. Overall, all coagulants required between 50% and 100% longer times to reach 50% removal of turbidity (turbidity at time  $t$ /initial turbidity = 0.5) at 5°C than at 20°C. Samples treated with solid coagulants presented 5% to 15% higher turbidity values at the end of the settling period when compared to the pre-dissolved coagulants.

### 3.4 Mixing

Most studies reporting the influence of mixing on coagulation and flocculation processes relate their results to pre-dissolved or liquid coagulants (e.g. Rossini et al., 1999; Yan et al., 2009). Our results show significant differences between the mixing requirements of coagulants dissolved prior to application and coagulants dosed directly in solid form. Although the identified optimum mixing

times for the fast and slow mixing stages were the same for solid and pre-dissolved coagulants, the mixing intensity or turbulence required in both stages were significantly higher when solid coagulants were applied (**Table 4**). Solid  $\text{Fe}_2(\text{SO}_4)_3$  required around 2 times higher  $G$  values during the fast and slow mixing stages than when dosed in pre-dissolved form. Solid  $\text{Al}_2(\text{SO}_4)_3$  required up to 8 times higher  $G$  values during fast mixing stage and around 6 times higher during the slow mixing stage compared to its stock solution.

Generally, our findings regarding the influence of mixing on the coagulation or rapid mixing stage of the purification process mostly agree with those of previous studies (Rossini et al., 1999; Kan et al., 2002; Yan et al., 2009). The removal of dissolved substances such as organic matter was only slightly affected by the intensity and duration of the rapid mixing stage, while the removal of SS appeared to decrease with increasing mixing times and increase with increasing mixing intensity until a critical value was reached. The mixing parameters applied during the flocculation process had a direct influence on SS removal from suspension. The observations made during our experiments regarding the characteristics of the flocs formed mostly agreed with widely reported characteristics in the literature (AWWA, 1990; Sincero and Sincero, 2003; Zhan et al., 2011). Low velocity gradients of less than 20 ( $\text{sec}^{-1}$ ) and long retention times of 25 min produced large but light flocs, which did not settle well. Short retention times of 5 min and higher mixing intensity produced very small flocs, especially in the case of solid coagulants, which also remained in suspension.

## 4 Conclusions

The purification efficiencies obtained by all coagulants tested (20°C and nominal mixing conditions) were high and inside the purification levels required for chemical treatment of peat derived runoff water. As expected, a clear distinction between the results obtained by pre-dissolved and solid coagulants was observed. More importantly, our results showed that the dissolution of the coagulants influenced not only the purification efficiencies achieved and required dosages, but also the way in which process parameters such as mixing, pH and temperature affected the coagulant's performance. It is possible to confirm that the physical form of the coagulant has a significant influence on parameters which are critical for the design of chemical purification process and thus requires careful evaluation during process optimisation.

Based on the results obtained, the following needs to be taken under consideration: (1) The detrimental effects of low temperature, inadequate mixing and acidic pH conditions were significantly more evident in the purification levels achieved by solid coagulants. Solid coagulants performance is therefore substantially more

susceptible to temperature, mixing and pH variations than the pre-dissolved coagulants. (2) The removal of SS from suspension was greatly reduced at low temperatures, while the removal of other substances such as phosphorus and TOC was only slightly decreased. The removal of SS at 5°C was around 70% lower for solid and 30% lower for pre-dissolved coagulants. (3) Mixing requirements of solid coagulants differed significantly from those of pre-dissolved coagulants. At similar retention times, solid coagulants required up to 8 times higher  $G$  values during the fast and slow mixing stages of the purification process. (4) Under our experiment conditions, small variations of about 1 pH unit resulted in variations of up to 100% in the dosage requirements of solid coagulants as well as a significant decrease in purification efficiencies. The removal of SS was particularly affected and was around 25% lower in the more acidic pH condition. (5)  $\text{Fe}_2(\text{SO}_4)_3$  was the best performing coagulant, requiring lower dosages and achieving higher removal efficiencies than  $\text{Al}_2(\text{SO}_4)_3$  and  $\text{Al}_2(\text{SO}_4)_3 + \text{Fe}_2(\text{SO}_4)_3$ . However,  $\text{Fe}_2(\text{SO}_4)_3$  especially in solid form, had a very narrow optimum dosage range, with colour and turbidity resurgence at dosages above the optimum.  $\text{Fe}_2(\text{SO}_4)_3$ , as the other coagulants, also displayed a significant decrease in purification efficiency at low temperatures and under unsatisfactory mixing conditions.

## Acknowledgments

This research was funded by Vapo Oy and also supported by Maa-ja Vesitekniiikan tuki r.y.

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## Journal of Environmental Sciences (Established in 1989)

Vol. 25 No. 4 2013

<b>Supervised by</b>	Chinese Academy of Sciences	<b>Published by</b>	Science Press, Beijing, China
<b>Sponsored by</b>	Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences		Elsevier Limited, The Netherlands
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<b>Editor-in-chief</b>	Hongxiao Tang	<b>Printed by</b>	Beijing Beilin Printing House, 100083, China
<b>CN 11-2629/X</b>	<b>Domestic postcode: 2-580</b>	<b>Domestic price per issue</b>	<b>RMB ¥ 110.00</b>

ISSN 1001-0742

