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Chemical treatment response to variations in non-point pollution water quality: Results of a factorial design experiment

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ABSTRACT

Chemical treatment of non-point derived pollution often suffers from undesirable oscillations in purification efficiency due to variations in runoff water quality. This study examined the response of the chemical purification process to variations in water quality using a 2^k factorial design for runoff water rich in humic substances. The four *k* factors evaluated and the levels applied were: organic matter as dissolved organic carbon (DOC) (20–70 mg/L), suspended solids (SS) (10–60 mg/L), initial water pH (4.5–7), and applied coagulant dosage (ferric sulphate) (35–100 mg/L). Indicators of purification efficiency were residual concentration of DOC, SS and total phosphorus (tot-P). Analysis of variance and factor effect calculations showed that the initial DOC concentration in raw water samples and its interactions with the coagulant dosage applied exerted the most significant influence on the chemical purification process, substantially affecting the residual concentration of DOC, SS and tot-P. The variations applied to the factors SS and pH only slightly affected purification efficiency. The results can be used in the design of purification systems with high organic matter load variation, e.g. peat extraction runoff.

1. Introduction

Large areas of peatland have been drained for agriculture, forestry and peat extraction for energy fuel and horticulture in the Northern hemisphere and in other regions such as South-East Asia (e.g. palm oil cultivation). Peatland drainage results in increased sediment and nutrient transport (Heikkinen, 1994; Kløve, 2001; Marttila and Kløve, 2008) with harmful impacts on recreation, migratory fish and biodiversity (Marja-aho and Koskinen, 1989; Selin et al., 1994; Laine, 2001). The quality of peatland runoff water is known to oscillate depending on drainage intensity, soil geochemistry, and runoff (Kortelainen and Saukkonen, 1998; Aitkenhead et al., 1999; Edén et al., 1999; Joensuu et al., 1999, 2002; Åström et al., 2001; Kløve, 2001; Worrall et al., 2003; Marttila and Kløve, 2008, 2010). In Finland chemical treatment is considered one of the best available technologies for the purification of peat extraction runoff water and this purification method is currently applied in several extraction sites (Ministry of the Environment, 2013). However, variations in runoff water quality and the lack of development of process parameters has led to the application of high chemical dosages, significant and undesirable fluctuations in purification efficiency and high metal concentration in the discharging waters (Heiderscheidt et al., 2013).

Chemical purification via addition of coagulation and flocculation chemicals is widely used for the treatment of different types of wastewaters. The treatment efficiency is known to be influenced by water quality (e.g. SS, DOC, pH) and process parameters such as dosage and mixing (Franceschi et al., 2002; Duan and Gregory, 2003; Liu and Chin, 2009; Zhao et al., 2011; Zhang et al., 2012). The degree to which pollutant substances affect the coagulation and flocculation mechanisms vary according to their concentration and their interaction with other substances in solution (Cheng, 2002; Yang et al., 2010; Cavarelli et al., 2012; Zhang et al., 2012; Tubić et al., 2013; Zhao et al., 2014). Furthermore, the characteristics and concentration of substances contained in the water have a direct effect on process parameters such as coagulant type and dosage, mixing requirements and sedimentation time (Franceschi et al., 2002; Slavik et al., 2012; Zhang et al., 2012; Rong et al., 2013; Wu et al., 2013; Zhao et al., 2014). Due to several complex interactions influencing chemical treatment; ideal purification conditions are normally identified using the jar test and other pilot test methods (Bratby, 2006).

The objective of this study was to evaluate the response of the chemical purification process to the oscillations in water quality

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which are typical of peatland-derived runoff. A full 2^k factorial design was applied on the planning of the jar test experimental procedure. Despite the many advantages of factorial designs (e.g. more effective than one factor at a time method and reduced number of experimental runs) to date they have only been scarcely applied in investigations of coagulation and flocculation processes (Franceschi et al., 2002; Krishna Prasad and Srivastava, 2009; Martín et al., 2011). The four *k* factors evaluated here were selected based on a literature review and results from previous studies (Heiderscheidt et al., 2013). The investigated factors were: organic matter concentration as DOC, SS concentration, initial water pH and applied coagulant ($Fe_2(SO_4)_3$) dosage. The influence of these factors on purification efficiency was assessed by evaluation of observed changes in the residual concentrations of selected response variables: DOC, SS and total phosphorus (tot-P). Natural water samples were used in a novel attempt to ensure the presence of all naturally occurring substances which can affect the purification process. The purification results were evaluated by factor effect determination using orthogonal contrast (OC) calculations and analysis of variance (ANOVA) in the data sets obtained.

2. Materials and methods

Humic and sediment rich natural water samples were collected from peat extraction runoff. The water samples were manipulated using techniques such as dilution, centrifugation and base addition to produce analytical samples with the required water quality characteristics. A full 2^4 factorial design (four factors each at two levels) was used in the planning of performed experiments. The response variables (residual concentration of DOC, SS and tot-P) or indicators of purification efficiency were selected based on monitoring requirements established by Finnish environmental authorities (Ministry of the Environment, 2013).

2.1. Experiment design

The four factors investigated were: organic matter concentration as DOC (factor A); SS concentration (factor B); initial water pH (factor C); and applied coagulant ($Fe_2(SO_4)_3$) dosage (factor D). The influence of variations in these factors on the response variables, residual DOC, SS and tot-P, was evaluated using a 2^4 factorial design. A total of 16 experimental runs were conducted (randomly) for which 3 replicates were performed. The outputs of the experimental design were analysed using SPSS statistical software. The magnitude and direction of factor effect were determined via OC calculations and the significance of the factor effect was evaluated using ANOVA.

Purification tests were performed using the six-jar (1 L) programmable paddle stirrer jar test equipment Flocculator 2000 (Kemira Kemwater). Previously identified optimum mixing parameters (Heiderscheidt et al., 2013) were applied: 300 rpm for 10 s followed by 50 rpm for 25 min and 30 min of sedimentation time. Stock solution (10 g/L) of the commercial quality solid metal salt (Kemira Oyj, Kemwater; 90% purity) $Fe_2(SO_4)_3$ was used as the coagulant agent.

2.2. Identification of factors levels

Statistical analysis of water quality monitoring data from ten Vapo Oy peat extraction sites in Finland (biweekly or monthly sampling for a period of at least 2 years from each site) was used to identify the range of factor variations to be applied. The 10th and 90th percentile of observed values for DOC, SS and pH were then selected as the low and high concentration (or level), respectively, of each factor (Table 1). High and low levels of the fourth factor, applied dosage (Dos), were selected via dosage requirement identification tests in which increasing dosages of coagulant were applied to two water samples. First, increasing dosages were added to samples containing low concentrations of DOC, SS and pH (sample (1), Table 3) and subsequently to samples containing high concentrations of these factors (sample abc, Table 3). Removal of colour and turbidity were used as parameters in the identification of the optimum dosages to purify both samples. The optimum dosages were then selected as the low and high level of the applied coagulant dosage (D) factor (Table 1).

Table 1
Selected high and low level values for the factors evaluated in the 2^4 factorial design.

Water quality parameter	Factor	Level	
		Low	High
DOC (mg/L)	A	20	70
SS (mg/L)	B	10	60
pH	C	4.5	7
$Fe_2(SO_4)_3$ (mg/L)	D	35	100

2.3. Preparation of water samples and statistical analyses

Two water samples were collected from peat extraction sites between May and July 2012 (Table 2). Sample 1 was collected from Navettarimpi, a site located near Vaala, Finland. Sample 2 was collected from Verkanneva, a site close to Vihanti, Finland. Two samples were used due to the fact that at the time of sampling, no single sample containing high concentrations of both DOC and SS was obtained. Sample 2, containing high DOC and low SS concentrations, was selected as the basis for the preparation of the eight samples required (containing the low and high concentration combinations of DOC, SS and pH). A diagram describing the samples preparation process is presented in Fig. 1. To prepare samples with high SS, sample 1, which contained high SS concentration, was centrifuged (4000 rpm, 4 min) and the sediment added to sample 2 while the supernatant was discarded. Samples with low concentration of all factors were prepared by diluting sample 2 (plus added sediments) with deionised water. Samples with high pH (7.0) values were prepared by addition of sodium hydroxide (NaOH, 0.1 M).

Water quality characteristics of the prepared samples are presented in Table 3. In addition to the controlled substances or factors, the water samples were also analysed for a series of other water quality parameters such as: phosphate phosphorus (PO_4–P), total iron (Fe), sulphates (SO_4^{2-}), UV_{254} abs., Cq and total nitrogen (tot-N). All water quality analyses, apart from UV absorbance at 254 nm (UV_{254} abs.), pH and charge quantity (Cq) measurements, were performed by a FINAS (Finnish Accreditation Service) certified laboratory using the following standard methods: DOC SFS-EN 1484:1997 (0.45 μm filtration); SS (GFC) SFS-EN 872:2005; tot-P SFS-EN ISO 6878:2004 FIA technique; PO_4–P SFS-EN ISO 6878:2004, FIA technique; Fe in-house method IC207 and IC209, microwave oven combustion preparation, ICP-OES technique ISO

Table 2
Characteristics of sample 1 and 2 used as basis for preparation of the eight water samples required.

Substance	Sample 1	Sample 2
DOC (mg/L)	18	72
SS (mg/L)	240	11
pH	6.5	4.6
tot-P (μg/L)	280	190
PO_4–P (μg/L)	150	57