

SIMULTANEOUS COMPLIANCE OF TOC AND TURBIDITY RELATED TO PATHOGEN BREAKTHROUGH AND THMs CONTROL BY ENHANCED COAGULATION

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ABSTRACT

Since discovery of harmful disinfection by products (DBPs) like trihalomethanes (THMs) and haloacetic acids (HAAs) in chlorinated waters the removal of organic DBPs precursors gained priority in drinking water treatment. Historically, coagulation process was arranged to remove turbidity, typically related to pathogens occurrence, but nowadays it is usually optimized/enhanced for total organic carbon (TOC) reduction.

This work dealt with the removal of TOC and turbidity by means of enhanced coagulation process using $\text{Al}_2(\text{SO}_4)_3 \cdot 18 \text{H}_2\text{O}$ and $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ as coagulants for jar test without pH adjustment according to USEPA procedure. 15% of TOC removal required by USEPA D/DBPs rule (1998) was achieved using 30-50 mg l^{-1} of both coagulants. On the other hand, for compliance with more stringent MCL for total THMs in Italy (30 $\mu\text{g l}^{-1}$) higher TOC removal (>30%) which needed of higher coagulants doses (80 mg l^{-1}), was obviously required. Those high coagulant doses were also to be used for the compliance with the turbidity standard (<1NTU).

KEYWORDS: enhanced coagulation, pathogens control, TOC and turbidity removal, TTHMFP

1. INTRODUCTION

The coagulation process has been primarily used in drinking water treatment to remove turbidity (Eikebrokk, 1999; Volk *et al.*, 2000). Since the discovery of disinfection by products (DBPs) like trihalomethanes (THMs) and haloacetic acids (HAAs) in chlorinated waters (Rook, 1974; Bellar *et al.*, 1974) which are known harmful for human health, the removal of natural organic matter (NOM) has gained growing attention. NOM fraction which is responsible for DBPs formation, is a mixture of humic and fulvic acids; typically characterised by total organic carbon (TOC), dissolved organic carbon (DOC) and UV adsorbance at 254 nm. Edzwald *et al.* (1985) developed the use of specific ultraviolet absorbance (SUVA; the absorbance at 254 nm per unit of DOC in mg l^{-1}) as an operational indicator of the composition of organics. SUVA provides a quantitative measure of the aromatic content (aromatic sites substituted with oxygen- and nitrogen-containing functional groups, i.e., phenolics and aromatic amines) of the organic carbon.

Although DBPs formation shifted attention on NOM control, turbidity is an important parameter with reference to microbial risk management in drinking water treatment because of turbidity spikes are considered potential for breakthrough of pathogens. The

role of turbidity as indicator of pathogen's presence is connected to the electrostatic properties of the surface of some microorganisms (Drozd and Schwartzbrod, 1996; Brush *et al.*, 1998; Butkus *et al.*, 2003) and to possibility that they can be removed by coagulants according to same mechanisms by which turbidity is removed during coagulation process (Bustamante *et al.*, 2001). This explains positive correlations ($r^2=0.93$) between high raw water turbidity (>2 NTU) and the presence of *Mycobacterium avium* (Falkinham *et al.*, 2001), environmental opportunistic human and animal pathogens (Inderlied *et al.*, 1993).

The efficiency of coagulation depends on several parameters some of which closely connected to variation of weather conditions. The rainstorm events can rapidly change the raw water characteristics leading to elevated levels of turbidity, NOM and sometime microbial load (Kistemann *et al.*, 2002). These changes can result in increasing of coagulant demand hindering the capacity of conventional water treatment (coagulation, sedimentation and filtration). In the recent years some studies were focused on this specific problem suggesting different way to improve coagulation efficiency during rainfall events such as increasing alkalinity to reduce turbidity (Tseng *et al.*, 2000), using ion exchange for NOM control (Fearing *et al.*, 2004), describing a turbidity-based index to identify rainy periods and event that adversely effect the treatment process (Hurst *et al.*, 2004).

For a raw water with high turbidity (10-20 NTU) and relatively high TOC (5-6 mg l⁻¹) with a pH range of 7.2-8.5, and alkalinity between 46 and 50 mg CaCO₃ l⁻¹, the simultaneous removal of TOC and turbidity was demonstrated by means of TOC and turbidity removal domains overlapping in alum coagulation diagram (Amirtharajah's diagram) (Seung-Hyun *et al.*, 2001). If turbidity of water is not excessive the coagulant demand is considered to be dependent on the NOM content (Edzwald and Van Benschoten, 1990; Gregor *et al.*, 1997; Ratnaweera *et al.*, 1999). For hard waters with high alkalinity and pH levels, it is often a matter of discussion from economic and practical perspectives whether to use inorganic acids or coagulant overdosing in order to depress coagulation pH to optimum levels with respect to NOM removal. Low temperature affects the organic matter removal negatively resulting in increasing residual coagulant concentration in finished water (Exall and Vanloon, 2000).

The Long Term 1 *Enhanced Surface Water Treatment Rule* (LT1ESWTR) of USEPA (2002) set very stringent limit on effluent turbidity in conventional and direct filtration plants. According to that regulation, turbidity values of 95% of samples taken each month must be less than or equal to 0.3 NTU and never >1 NTU to improve control of microbial pathogens, including specifically the protozoan *Cryptosporidium*. On the other hand, to protect drinking water consumer from DBPs risk USEPA (1998) promulgated Stage 1 of *Disinfectants and Disinfection By-Products* (D/DBP) Rule establishing both maximum contaminant levels (MCLs) for four DBPs categories (TTHM, HAAs, bromate and chlorite), in particular 80 µg l⁻¹ for TTHMs, and TOC removal requirements for water systems that use conventional filtration treatment. If water treatment plants can not met criteria set in D/DBP Rule, they are allowed to establish alternative TOC removal requirements implementing jar test procedure according to a guidance manual for enhanced coagulation and precipitative softening (USEPA, 1999).

According to the drinking water quality rule of Italy (Official journal of Italy, 1988), any explicit limit for DBPs (except a not better specified limit for organic halogenated compounds) was set up to December 25, 2003. However, recent established legislation (Official journal of Italy, 2001) set very stringent limits for three species of DBPs (TTHMs, chlorite and bromate), in particular for TTHMs (30 µg l⁻¹). But no TOC removal was established while 1 NTU turbidity level has to be met.

This work gives the results of NOM (in term of TOC, DOC and UV₂₅₄) and turbidity removal by means of enhanced coagulation with the aim of simultaneous control of THMs formation and pathogen breakthrough. Experiments were performed on surface water

samples taken from an artificial basin in southern Italy. Two coagulants ($\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ and $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) were used for jar test without pH adjustment according to USEPA procedure for enhanced coagulation.

2. MATERIALS AND METHODS

2.1 Sampling and conservation

Raw water samples were taken from artificial basin located in province of Salerno (Southern Italy). The basin was constructed to compensate water deficit in tourist season in the region where usually groundwater is consumed. Eleven water samples (S1,.. S11) were collected between February 2002 and March 2003 representing different water conditions and water characteristics, in particular TOC and turbidity parameters.

The raw water samples were collected in 10 L containers and delivered in to the laboratory in 1 h to be stored at 4 °C.

2.2 Jar test procedure

All samples were brought to room temperature prior to jar testing. Coagulation and flocculation experiments were carried out using ISCO jar test apparatus in 1 l beakers. Following the addition of the coagulants, the samples were subjected to rapid mixing for 1 min at 100 rpm, than flocculation for 30 min at 30 rpm and last settling for 60 min. Alum ($\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$) and ferric chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) were used as coagulant up to reaching a final coagulant dose of 80 mg l⁻¹ yielding around 7.0 pH which was referred target value by USEPA(1999). The coagulants were added at 10 mg l⁻¹ increases calculated in terms of equivalent to $\text{Al}_2(\text{SO}_4)_3 \cdot 14\text{H}_2\text{O}$ according to USEPA procedure for enhanced coagulation (USEPA, 1999).

To evaluate the coagulation efficiency, raw and treated samples were analyzed without pH adjustment for pH, turbidity, TOC, DOC, UV₂₅₄, SUVA₂₅₄, alkalinity parameters. Bromide was also measured on raw water samples. Iron and aluminium were monitored in raw and coagulated samples.

2.3 Analytical measurements

Raw and coagulated samples were filtered through 0.45 µm pre-rinsed GMF filter (25 mm GD/X, Whatman Inc, USA) for DOC and UV₂₅₄ adsorbance measurements. TOC and DOC were measured by SHIMADZU analyser (model 5000A). UV-Vis spectrophotometer was used to measure UV₂₅₄ absorbance (PerkinElmer, model Lambda12, USA). Turbidity was measured using Hach turbidimeter (model 9024, USA) and pH was monitored by Hach portable meter (Model 2100N). Iron (AAS, Varian, spectraAA 10 Plus, USA) and aluminium (Metrohm polarograph, 757VA, USA) were also measured.

3. RESULTS AND DISCUSSION

3.1 General

The raw water characteristics of each sample are given in Table 1. Bromide was below detection limits (<10 µg l⁻¹). The raw water samples were characterized by relatively high alkalinity (190.3 ± 2.79 mg CaCO₃ l⁻¹) and low TOC (2.81 ± 0.19 mg l⁻¹) and DOC (2.74 ± 0.18 mg l⁻¹). A 3.88 ± 0.28 l mg.m⁻¹ mean value was obtained for of SUVA indicating that the composition of NOM consisted of the mixture or mostly of aquatic humics (Edzwald *et al.*, 1985) which means the coagulation can be effective in NOM removal (Randtke, 1999). Relatively high turbidity was measured during investigation period (10.66 ± 6.74 NTU) characterized by strong variations from 4.7 to 29.0 NTU.

According to TOC removal criterion set by USEPA (1998), a 15% of TOC removal is targeted because raw waters were characterized by both a higher alkalinity (> 120 mg CaCO₃ l⁻¹) and lower TOC (< 4 mg l⁻¹).

Table 1. Characteristics of the samples taken from Alento constructed basin and coagulant used for jar test

Sample	pH	Alkalinity (mg l ⁻¹)	Turbidity (NTU)	TOC (mg l ⁻¹)	DOC (mg l ⁻¹)	UV ₂₅₄ (1 cm ⁻¹)	SUVA (l mg. m ⁻¹)	Coagulant
S1	7.4	183.4	29.0	3.32	3.22	0.123	3.82	Alum
S2	7.4	190.9	15.0	2.80	2.79	0.108	3.87	Alum
S3	7.4	190.9	10.0	2.80	2.72	0.099	3.64	Alum
S4	7.4	188.5	9.0	2.76	2.60	0.100	3.85	FeCl ₃
S5	7.5	189.4	8.0	2.71	2.66	0.102	3.83	Alum
S6	7.4	193.5	8.0	2.80	2.77	0.128	4.62	FeCl ₃
S7	7.3	192.2	8.0	2.90	2.86	0.112	3.92	Alum
S8	7.5	189.7	12.4	2.84	2.75	0.098	3.56	FeCl ₃
S9	7.7	191.9	8.0	2.66	2.59	0.102	3.94	Alum
S10	7.7	193.2	4.7	2.61	2.56	0.102	3.98	Alum
S11	7.7	190.0	5.3	2.72	2.64	0.097	3.67	FeCl ₃
Average	7.49±0.14	190.3±2.79	10.66±6.74	2.81±0.19	2.74±0.18	0.11±0.01	3.88±0.28	
Range	7.3-7.7	183.4-193.5	4.7-29.0	2.61-3.32	2.56-3.22	0.1-0.13	3.56-4.62	

3.2 The pH

Although final pH was dependent on the coagulant type and alkalinity, no significant difference in final pH values of alum (Figure 1a.) and FeCl₃ (Figure 1b.) experiments were observed. Around 7.0 of target pH was reached in the range of 30-50 mg l⁻¹ of equivalent dose for alum and 30-40 mg l⁻¹ of equivalent dose for FeCl₃ of which residual Al and Fe ion concentrations did not exceed the limit (< 0.2 mg l⁻¹)

3.3 Organic DBPs precursors removal

The results of the jar test (in terms of TOC, DOC and UV₂₅₄) showed high NOM removal efficiencies (>30%) for both coagulants for all samples (Figure 2a, b) using 80 mg l⁻¹ as equivalent dose which allowed to remove almost 33 and 36% of TOC for alum and FeCl₃, respectively (Table 2). However, 16% of TOC removal was obtained at target pH.

Alum proved highest TOC removal for S1 due to its high initial TOC value (3.32 mg l⁻¹) while for other samples the NOM removal were similar due to a narrow range of TOC and SUVA (3.8-3.9 l mg.m⁻¹) (Figure 2a). FeCl₃ yielded the best NOM removal efficiency for S6 characterized by 4.62 l mg.m⁻¹ of SUVA which was significantly higher than S4, S8 and S11 samples (Figure 2b). The composition of NOM by means of SUVA consists of the mixture or mostly of aquatic humics as hydrophobic character with moderate/high molecular weight (Edzwald *et al.*, 1985) for which coagulation is very effective for NOM removal (Randtke, 1999).

Our previous study on the same raw water showed that TTHM formation potential (TTHMFP) concentrations in raw water varied between 120-200 µg l⁻¹ (Bekbolet *et al.*, 2004) and it could be lowered to 30 µg l⁻¹ using 30-40 mg l⁻¹ of alum and FeCl₃ for initial 2.7 mg l⁻¹ of TOC and 185 mg l⁻¹ of alkalinity (Rizzo *et al.*, 2004). Hence, the present results given in Table 2 are indicative for compliance with required TOC removal and for lowering TTHMFP.

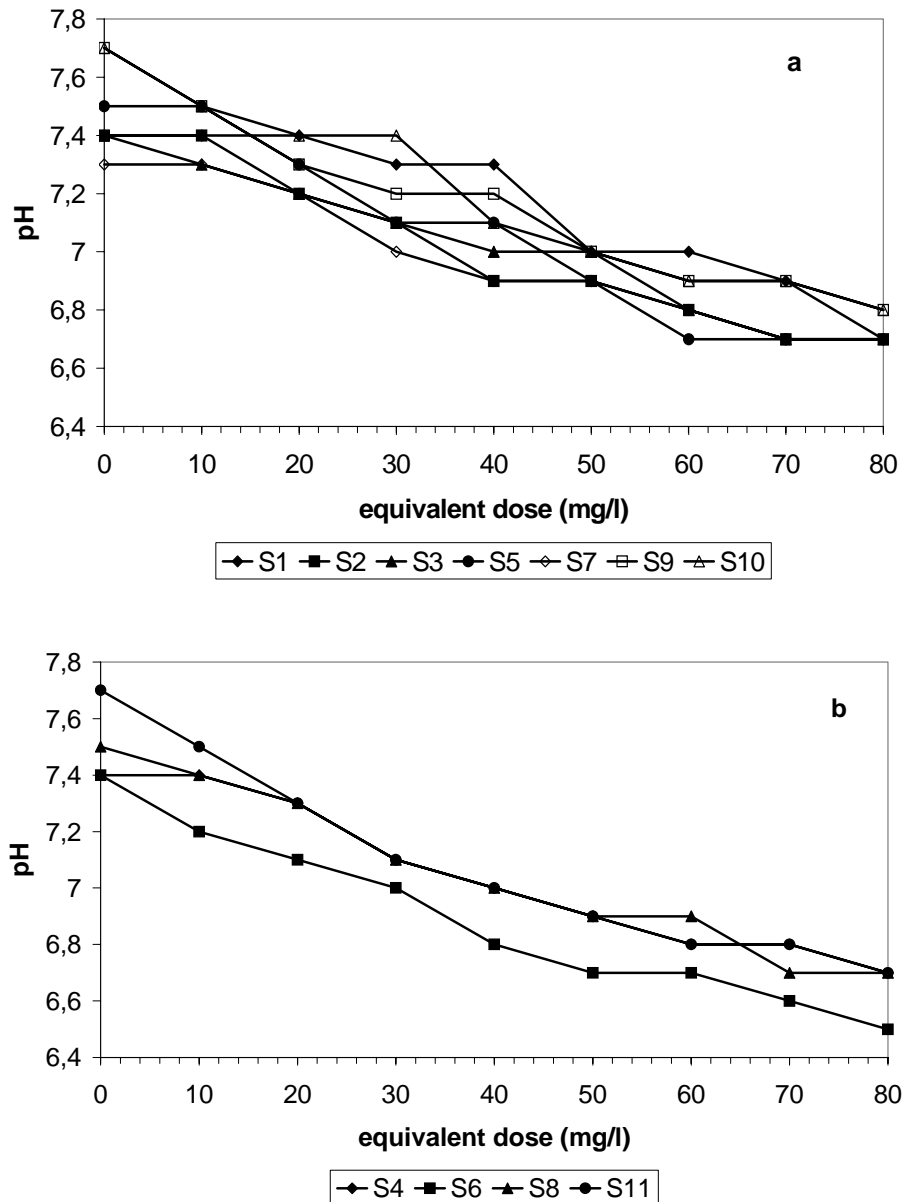


Figure 1. pH behaviour for alum (a) and ferric chloride (b) experiments

3.4 Simultaneous TOC and turbidity compliance

Although high NOM removal efficiencies were obtained by enhanced coagulation, coagulation experiments did not provide lower turbidity values than 2.0 NTU for alum and 1.2 NTU for FeCl_3 at target pH. However, using 80 mg l^{-1} of equivalent coagulant doses turbidity could be lowered to lesser than 1 NTU (Table 3).

For controlling of pathogens simultaneously to turbidity removal, the capacity of filtration unit to reduce turbidity up to 0.3 NTU according to standard set in LT1ESWTR (USEPA, 2002) should be controlled very strictly. If filtration unit cannot provide this result, coagulation process should be enhanced for optimal removal of turbidity rather than TOC, increasing coagulant dose above target pH as well as corresponding residual iron or aluminium salts in the effluent.

In Italy, where higher TOC removal efficiencies are required due to more stringent TTHMs limit, pH target removal could not provide to reduce TOC which requires the use of higher

coagulant doses. However, rather than TOC turbidity removal could be limiting factor in the evaluation of coagulation process.

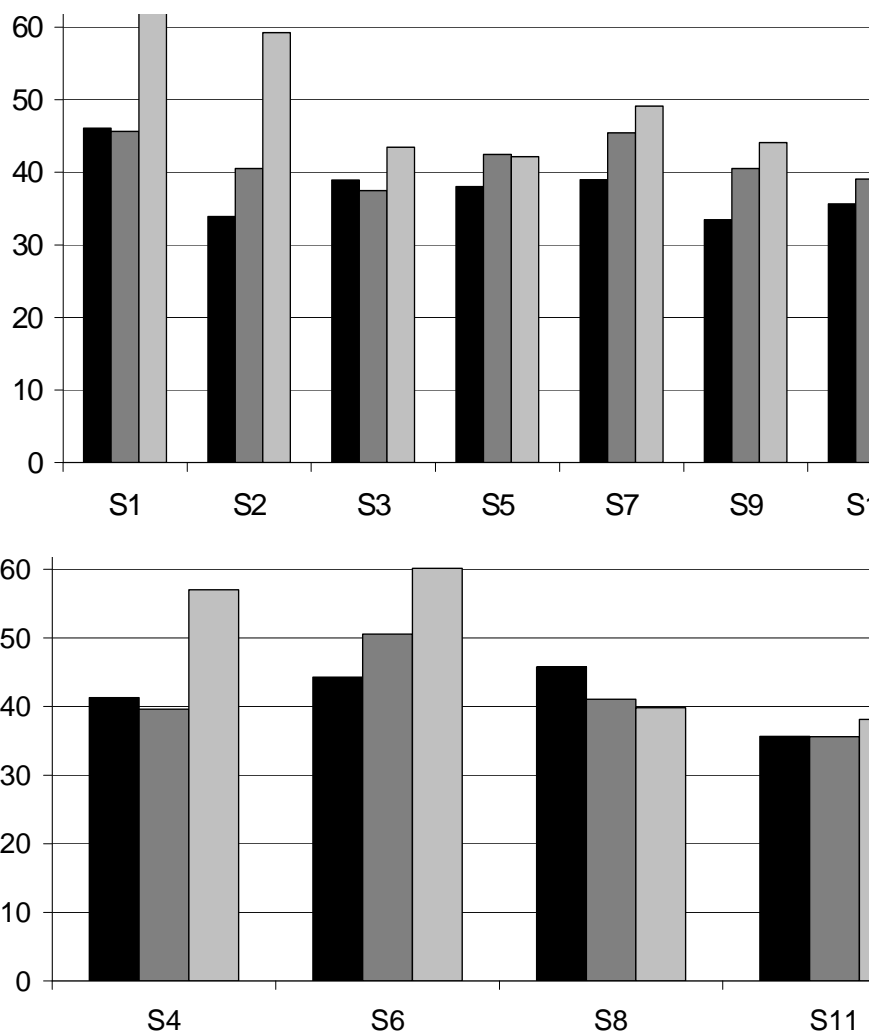


Figure 2. organic THMs precursors removals using alum (a) and ferric chloride (b).

Table 2. TOC removal at both target pH and maximum coagulant dose.

Coagulant	Samples	target pH removal		Max. dose removal	
		Dose (mg l^{-1})	TOC removal (%)	Dose (mg l^{-1})	TOC removal (%)
Alum	S1-S3, S5, S7, S9, S10	30-50	16-36	80	33-46
FeCl_3	S4, S6, S8, S11	30-40	22-30	80	36-46

Table 3. Turbidity removal at both target pH and maximum coagulant dose

Type	Samples	target pH removal			Max. dose removal		
		Dose (mg l^{-1})	NTU (%)	Final NTU	Dose (mg l^{-1})	NTU (%)	Final NTU
Alum	S1-S3, S5, S7, S9, S10	30-50	50-83	1.9-6	80	77-93	0.8-2
FeCl_3	S4, S6, S8, S11	30-40	37-81	1.2-5	80	75-95	0.6-2

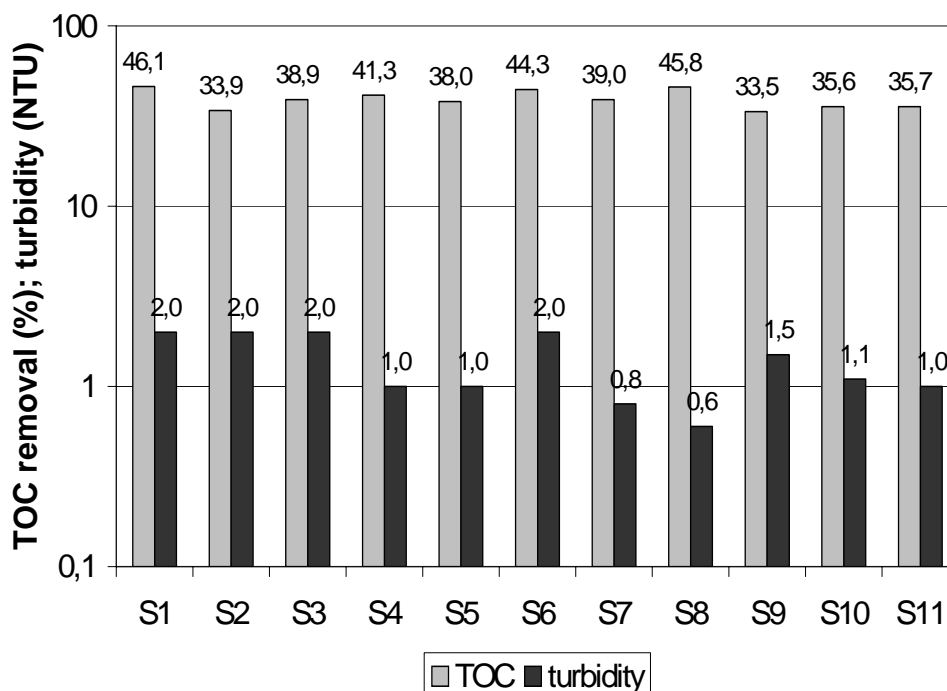


Figure 3. Simultaneous TOC and turbidity removal at 80 mg/l as equivalent coagulant dose

Figure 3 shows the resulting turbidity and TOC removals obtained using 80 mg l⁻¹ of equivalent coagulant dose for all samples. TOC removals ranged between 33-46%.

Keeping in mind that previous studies established that to obtain effective removal of *Giardia* and *Cryptosporidium* the turbidity of filtered water has to be kept in the range of 0.1-0.2 NTU (Logsdon *et al.*, 1985; Al-Ani *et al.*, 1986; Niemiski and Ongerth, 1995) Italian standard for turbidity (<1 NTU) seems to be relative high to control the risk of pathogen breakthrough. A further integrative treatment to coagulation/filtration, e.g., granular activated carbon adsorption or enhanced sand/micro filtration would be proper to decrease the over coagulant doses which could result in high residual iron or aluminium ion in finished water (Srinivasan *et al.*, 1999) as well their toxicity risk (Pagano *et al.*, ; Melluso *et al.*, 2004).

4. CONCLUSIONS

Coagulation process can be effective for simultaneous control of organic precursors of THMs formation and pathogen breakthrough. Both the correlation between NOM and THMs formation and the positive evidence found between turbidity and some pathogens force to evaluate the approach of simultaneous compliance for THMs precursors and turbidity removal. The following results were obtained in this study:

The TOC removal was always higher than 16% at target pH in compliance with TOC removal requirement (15%) set by USEPA (1999) enhanced coagulation procedure for raw water with low TOC (< 4.0 mg/l) and high alkalinity (> 120 mg CaCO₃/l) by using 30-40 mg l⁻¹ of alum and FeCl₃ doses. However, at target pH turbidity removal was rather limiting parameter than TOC removal for compliance with both Italian turbidity limit (<1 NTU) and USEPA limit (2002) for pathogen control.

For TOC removal requirement according to Italian low limit for TTHM, 80 mg l⁻¹ equivalent dose of alum and FeCl₃, yielding 33 and 36% of TOC removal as well as <1 NTU for alum and FeCl₃ respectively, were to be used.

On the other hand, to reach turbidity values in the range of 0.1-0.2 NTU for pathogen breakthrough control, integrative treatment such as granular activated carbon adsorption, sand/micro filtration could be necessary to avoid from using overdoses of coagulants.

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