

FOULING OF UV LAMP SLEEVES: EXPLORING INCONSISTENCIES IN THE ROLE OF IRON

Sehnaoui, Karim

M. Eng.

McGill University, Montreal, Canada

Gehr, Ronald

Associate Professor, PEng, PhD

McGill University, Montreal, Canada

Abstract

This paper investigates the effects of water quality parameters on fouling rates of UV lamp sleeves, through the results of various published studies performed by different groups as well as by the authors. Water quality parameters used as possible predictors of fouling were COD, SS and VSS, temperature, pH, UV transmission, and metal concentrations, mainly Fe and Ca. Fecal coliforms were counted as an indirect indication of foulant accumulation. Metals analyses were performed on the foulant itself. As part of this study, a pilot UV system was installed at the effluent line of two different biological wastewater treatment plants. Iron was added to the UV pilot system influent in both cases to simulate post-precipitation of phosphorus. Ferric chloride concentration was varied from 0 to 6 mg/L. Most runs were limited to no more than 10 days in order to explore the onset of incipient fouling. At both wastewater treatment plants studied herein, practically no foulant accumulation was observed on the UV sleeves, even after 10 days, and the disinfection efficiency was not reduced by the addition of iron. Incipient fouling was not detected in any of the runs. This suggests that iron alone is not at the source of the fouling problem; one cannot correlate fouling with simple measurements of iron concentrations. A comparison of influent water quality parameters obtained from the literature suggests that calcium concentrations could play an important role in the fouling process. Hardness was almost always elevated in systems with high fouling rates. Moreover, a high influent bacterial content of the wastewater appears to accelerate the fouling process. Thus, the type of treatment that precedes the disinfection process, namely physical-chemical or biological, seems to stimulate different responses to UV radiation.

Keywords

Ultraviolet disinfection, iron, fouling, organics, photochemistry

Introduction and background

Fouling of the outside of the quartz sleeves surrounding UV lamps remains one of the limitations of UV disinfection systems. When submerged in the wastewater, foulant has been observed to grow on these sleeves, reducing the treatment efficiency, sometimes after only a few days of operation. The sleeves have to be taken out of service and cleaned, unless the UV system has an in-situ mechanical wiping mechanism. However, the nature of the foulant itself remains unknown. It is well established that an inorganic layer comprised of mainly calcium (always) and iron (not always, but generally), a lesser amount of magnesium, aluminum (when added), sodium, phosphorus (where present) and other metals, makes up a substantial proportion of the foulant (1). There remain, nevertheless, questions concerning the presence of organic and biological compounds, reported by some as being part of the foulant (2). Biological fouling occurs in the form of a biofilm that has a tendency

to stick to surfaces, depending on the type of microorganisms involved, the nutrient content of the wastewaters, wastewater temperature and many other factors. In addition, the fact that fouling is site-specific makes it harder to determine the mechanisms by which fouling is initiated or even the conditions under which it is manifested. Some systems have reported operational periods of two to three months without lamp cleaning (3), whereas others required lamp cleaning twice a day (4, 5). Table 1 summarizes some of the fouling studies found in the literature and their main findings.

Generally, researchers have identified three mechanisms by which foulant may deposit on the sleeves, namely: 1- heat-induced precipitation of metals with inverted solubilities, 2- gravitational settling and impaction of particles and lastly, 3- flocculation (1, 6, 7, 8).

- Most of the metals comprising the inorganic foulant have inverted solubilities, i.e., their solubility decreases with increasing temperature. Thus, it is likely that the temperature gradient at the sleeve-water interface may promote heat-induced precipitation of these metals.
- Gravitational settling, or impaction, was also identified as a fouling mechanism, especially in treatment plants that use alum coagulants. Pre-formed flocs under the effects of Brownian motion and sedimentation, will deposit on the upper surface of the lamps.
- Considerable fouling was found to occur in regions of low velocity zones and where eddies may occur, mainly close to the lamp brackets (current study), and around the lamps themselves.

This site specificity has prompted researchers to attempt to establish correlations between quality parameters of the wastewater entering UV systems and the disinfection efficiency achieved. The parameters most frequently identified as affecting disinfection efficiency are UV transmissivity (UVT), turbidity and suspended solids (SS) (3, 11, 14, 15). However, in all studies published, no general correlations could be drawn. One can state that UV disinfection will be highly dependent on the performance of the preceding treatment processes, but the disinfection efficiency cannot be accurately predicted based on these wastewater characteristics.

Another element that seems to play an important role on fouling is iron. In many of the published studies, the use of iron coagulants (common in North America) resulted in an increase in the rate of accumulation of foulant on the quartz sleeves, with iron being the main constituent of the foulant (6, 8, 12). This is however not always the case, as was observed in this study and in other work (13). There are two aspects to the role of iron: iron is a strong complexing agent which has been identified to co-precipitate with calcium, but on the other hand, iron can also act as a strong inhibitor of calcium precipitation. Of all the metals identified in foulant samples, calcium is the only one identified in all fouling experiments (16, 17). This explains why the role of iron is not well understood. The effects of iron will also be influenced by the pH of the solution, the organic matter available, its concentration in solution, other metals in solution and their concentrations, as well as light and temperature.

Table 1: Summary of fouling studies

Experimental Site	Variables	Conclusions	Notes	Reference
Full-scale work at WWTP	Three coagulants dosed: Alum, Ferric Chloride, Alufer (30% FeCl ₃ / 70% Al ₂ (SO ₄) ₃)	<ul style="list-style-type: none"> Alum experiments observed the highest log disinfection Iron deposits on quartz sleeves when FeCl₃ used 	<ul style="list-style-type: none"> Influent iron to UV system was 2.12, 0.88 and 0.31 mg/L for FeCl₃, Alufer, and Alum respectively. 	Audet, 2000 (9)
Physical-chemical WWTP	Iron: 1 – 5 mg/L	<ul style="list-style-type: none"> Major fouling after one day Strong correlations between turbidity (T) & UVT, T & SS and SS & UVT 	<ul style="list-style-type: none"> Presence of Fe_{tot} above 0.5 mg/L is a major factor Results point to a combination of SS, Fe and small particles 	Gehr and Wright, 1998 (10)
Five biological WWTPs	Influent conditions All plants had at least secondary treatment	<ul style="list-style-type: none"> Fouling varied considerably from site to site High suspended solids decreased disinfection efficiency Poor quality effluent leads to considerable fouling 	<ul style="list-style-type: none"> A non-fouling effluent is one with mean BOD of 10 mg/L and mean SS of 15 mg/L 	Job et al., 1995 (11)
One physical-chemical WWTP One biological WWTP (biofilters)	Physical-chemical used FeCl ₃ Both plants observed very high influent FC counts (order of 10 ⁶ CFU/100mL)	<ul style="list-style-type: none"> Foulant composed of 80% inorganic material Four times more iron in foulant when FeCl₃ used Very rapid fouling at both plants, biological higher Foulant composed mainly of Ca (highest concentration), Fe, Mg and Al. 	<ul style="list-style-type: none"> Fe concentrations in foulant from OFF-lamps was half of that from UV-lamps Shearing and sloughing affect the fouling phenomena 	Jesien, 1998 (5)

Table 1: Summary of fouling studies (cont'd)

Experimental Site	Variables	Conclusions	Notes	Reference
Purdue University Laboratories Four physical - chemical WWTPs	Various metal concentrations Coagulant addition	<ul style="list-style-type: none"> • CaCO₃ played an important role • Iron complexation increased fouling rates and was found in large conc. • Main fouling process heat-induced precipitation; other processes include flocculation due to shear and turbulence in high colloidal water 	<ul style="list-style-type: none"> * <i>Iron</i> concentrations were <i>less than 0.1mg/L</i> in all samples tested * Lower velocities, lower temperatures, lower alkalinities not discussed 	Lin et al. 1999 (6, 7)
Biological WWTP	Iron concentrations 0-4 mg/L Al concentrations	<ul style="list-style-type: none"> • Use of FeCl₃ lead to rapid fouling, alum did not • Fe and Al were found in large percentage when added as coagulants • Calcium and magnesium main foulant constituents • Foulant composition high in organics 	<ul style="list-style-type: none"> * Initial inorganic layer followed by wet gelatinous protein-containing layer * Increasing iron conc. increased all other element concentrations in foulant (co-precipitation a possibility) 	Pinto and Santamaria, 1999 (12)
McGill University Laboratories (Ca in tap water around 35.1 mg/L and Mg around 9.2 mg/L)	Iron, Iron + Phosphorus Organics	<ul style="list-style-type: none"> • Precipitate sedimentation was the main fouling process • Greater fouling rates when phosphorus and iron added simultaneously (3:5) 	<ul style="list-style-type: none"> * No foulant analysis * No fouling during re-circulation * Heat-induced precipitation rejected 	Sheriff and Gehr, 2001 (8)
Univ. of Alberta Laboratories Biological WWTP	Increase of iron concentrations from 0.2 to 0.5 mg/L	<ul style="list-style-type: none"> • No increased fouling observed with the addition of iron • Calcium main fouling element 	<ul style="list-style-type: none"> * Calcium role downplayed 	Topnik et al., 1999 (13)
Two biological WWTPs	Iron (0-6 mg/L)	<ul style="list-style-type: none"> • Iron concentrations did not increase fouling rates • NO foulant appeared at any iron concentrations 	<ul style="list-style-type: none"> * Calcium concentrations less than 75 mg/L at both plants * Low microbial counts 	This study

Iron dissociation in water will yield various species at different pHs, a factor that also affects its solubility. As a trivalent ion, if introduced in high enough quantities, iron will cause less stable divalent calcium and magnesium complexes to dissociate in favor of trivalent ions. Magnesium does not stand out as a major foulant constituent in the studies investigated. Calcium on the other hand seems to be a major player in the fouling process and some authors would go further and affirm that it is a hardness-controlled process (1). Calcium precipitates predominantly as calcium carbonate (CaCO₃) with no crystalline structure (calcite) and at times as amorphous calcium phosphate (18). Also, Diaz (1994) (19), while investigating the solubility of inorganic P as influenced by pH and calcium concentrations, found that 80 mg/L of calcium is a threshold below which one would not expect calcium carbonate or any of its polymers to precipitate at ambient pH values.

Iron forms much stronger complexes than Ca²⁺ with a wide variety of compounds, such as EDTA, fulvic acids, humic acids, etc. Ca²⁺ is thus outcompeted for binding sites (20). Hence, when added as a coagulant, iron is observed to be the main constituent of the foulant in many studies (Table 1). Furthermore, Pinto and Santamaria (1999) (12) observed that an increase in iron concentrations in the foulant was accompanied by an increase of all other foulant constituents. This provides evidence for the complexing role of iron. Furthermore, in the absence of iron, Takasaki et al. (1994) (17) found that calcite growth rate was linearly dependent on calcium concentrations. Hence, the ratio of iron to calcium concentrations could be an important element of the fouling process. Moreover, hydroxyl and phosphate ions were determined to be the two most important ligands in terms of their affinity toward complex formation with Fe³⁺. Thus, Fe(III) in contact with organic matter is reduced to Fe(II), however in the presence of phosphorus, Fe(III) will tend to react first with soluble phosphate and will destabilize organic colloidal matter only after most of the phosphorus has precipitated (21, 22).

Natural Organic Matter (NOM) plays an important role in the solubilization of metal ions in water (23). Organic matter (OM) is mainly comprised of humic and non-humic substances. Humic substances can be defined as organic matter in an advanced state of decomposition. Humic substances will interact with cations in solution that will generate cation-organic complexes. The formation of these complexes can result in the removal of organic matter by enmeshment with insoluble solids made up of iron and NOM or insoluble iron-NOM complexes. Iron complexation with organic matter (OM) is influenced by the iron species in solution and the functional groups present in the NOM. Iron complexation under ambient temperature and pH conditions usually results in the reduction of iron from Fe(III) to Fe(II) (20, 23, 24, 25).

Humic substances and extracellular organic matter have been shown to act as photosensitizers that may induce chemical reactions, producing compounds such as singlet oxygen, hydrogen peroxide and OH-radicals (26). When a species or atom absorbs radiation, its energy level shifts to an excited state. This shift in energy level is observed at the electron phase and facilitates the electron transfer. The irradiation of humic substances in the presence of oxygen has been found to result in a highly unstable radical HO₂/O₂[•]. These radicals will most likely reduce Fe(III) according to the reaction given in Equation 1.



This is in agreement with the findings of Fukushima and Tatsumi (1999) (25), which indicate that iron is reduced in the presence of organic matter at neutral or higher pH. They suggest that inorganic species of Fe(III) first form complexes with humic acids (HA), which are then reduced to Fe(II) – HA complexes when irradiated. The final reduction of Fe(III) by irradiation can be written according to Equation 2:



Reduction was found to occur under both dark and light conditions. However, UV irradiation accelerated the reduction of Fe(III) to Fe(II). Fukushima and Tatsumi (1999) (25) studied closely the effects of UV irradiation and reported that the highest percentages of Fe(III) reduction occurred at low wavelengths; studies were conducted between 280 nm and 520 nm, however a linear increase in iron reduction with decreasing wavelength was evident. Furthermore, they established that the reducing effects of UV occur over a wide range of pH. Fukushima and Tatsumi (1999) (25) also established that the greater the concentrations of humic acids, the greater the amount of reduced Fe(II) species.

It is thus established that iron is reduced in the presence of NOM at neutral pH values and that UV irradiation accelerates this process.

In its reduced form, iron is very soluble and not prone to precipitation (27). However, Banwart (1999) (24) studied the precipitation of iron in natural systems. He found that Fe(II) precipitated with inorganic carbon. In his evaluation of possible carbon sources and precipitation species of iron, he found that Fe(II) co-precipitation on calcite was an important sink for ferrous iron. Thus, the role of calcium in water or wastewater becomes apparent and it can be expected that iron precipitation and deposition will be closely associated with the concentrations of calcium in solution.

It is therefore important that iron be in its oxidized form in order for it to precipitate as a separate species. Vaughan and Ord (1994) (28) have identified bacteria as a strong oxidizing agent of iron. This is in agreement with studies performed by Wheatly (1988) (29), which found that bacteria are a major biotic factor involved in the precipitation of iron. Therefore, in high microbial content wastewaters, iron precipitation - hence fouling - could be expected to occur.

Objectives

Specific objectives of this study were (a) to evaluate the correlation of fouling with a coagulant, ferric chloride and (b) to determine the nature of the foulant. A pilot study was performed at two different biological wastewater treatment plants to study the effects of various coagulant concentrations on fouling rates. Foulant samples were collected and analyzed for suspended solids and metals. Fecal coliform counts were used to assess the decrease in disinfection efficiency associated with foulant accumulation, along with UVT, iron concentrations and other wastewater quality parameters. The results of these experiments and a review of published works were then used to provide a comprehensive interpretation of the role of iron in fouling.

Materials and Methods

The Trojan UV System 3000 pilot plant consisted of three independent channels, each containing two lamps (Figure 1). The lamps used were low-pressure mercury arc lamps (1.63 m long, 26.7 W rated UV output). Plexiglas baffles separated each of the modules. An inlet box containing a perforated screen preceded the channels to allow for equal flow distribution between the channels. An outlet box followed the channels; it contained a weir that ensured that there would be a minimum of 76 mm of water over the top lamp in each module. The wastewater was supplied to the system at an average flow of 1-1.5 L/s per channel. Iron was added to the treated wastewater to simulate post-coagulation and precipitation of phosphorus. A 2,500 L tank was used to mix the coagulant with the wastewater before it underwent disinfection. The coagulant was supplied in the form of highly concentrated liquid ferric chloride (FeCl₃) (approximately 600 g/L). An independent peristaltic dosing pump allowed the final FeCl₃ concentrations to be varied. Final doses ranged from 0 to 6 mg/L of iron (Fe). Most runs were limited to ten days to study the onset of fouling. The coagulant was added at the top of the mixing tank to ensure adequate mixing time and flocculation, after which the water would exit from the bottom of the tank. This allowed for an approximate mixing time of 9 minutes.

The experimental work was realized at two different wastewater treatment plants. It was conducted in the summer periods of 1999 and 2000. Table 2 summarizes the various conditions of each experimental run.

St-Eustache

In the summer of 1999, the pilot plant was located at the St-Eustache WWTP, Quebec. This plant, which is entirely indoors, provides pre-treatment (flow measurement, screening, grit removal), primary clarification, biological filtration (using biofilters supplied by Degremont), and uses UV lamps as a final disinfection step

before the water is discharged into the Rivière des Milles Iles. This plant treats the wastewater of a population of almost 40,000, the flow of which can reach approximately 20,000 m³/day. Alum is added during the summer prior to the sedimentation process for phosphorus removal. Feed to the UV pilot plant was extracted immediately upstream of the full-scale UV units.

All six lamps used in the pilot system were low-pressure UV lamps. The concentration of iron added was maintained at approximately 3 mg/L during that summer.

Beauharnois

The pilot plant was moved to the Beauharnois WWTP in the summer of 2000. This plant is located approximately 25 Km outside of Montreal. The town's population is 10,172. The plant does not treat wastewater from industrial sites. The average daily flow is 8,126 m³. The process used at this plant is fairly simple: screening and two grit removal units (indoors) followed by two oxidation ditches and two secondary clarifiers in parallel (outdoors). No coagulants are added. The effluent is discharged directly into the St-Lawrence River.

During the first month of experiments, runs were limited to two or three days to investigate incipient fouling using six UV lamps. The rest of the summer was dedicated to studying the role of iron by gradually increasing iron concentrations from 0 up to 6 mg/L. Then, only one module (Channel 1) contained UV lamps, whereas the other two served as controls with OFF-lamps in Channel 2 and NON-UV (i.e. visible light only) lamps in Channel 3. The purpose of the controls was to separate the effects of UV photochemistry and temperature.

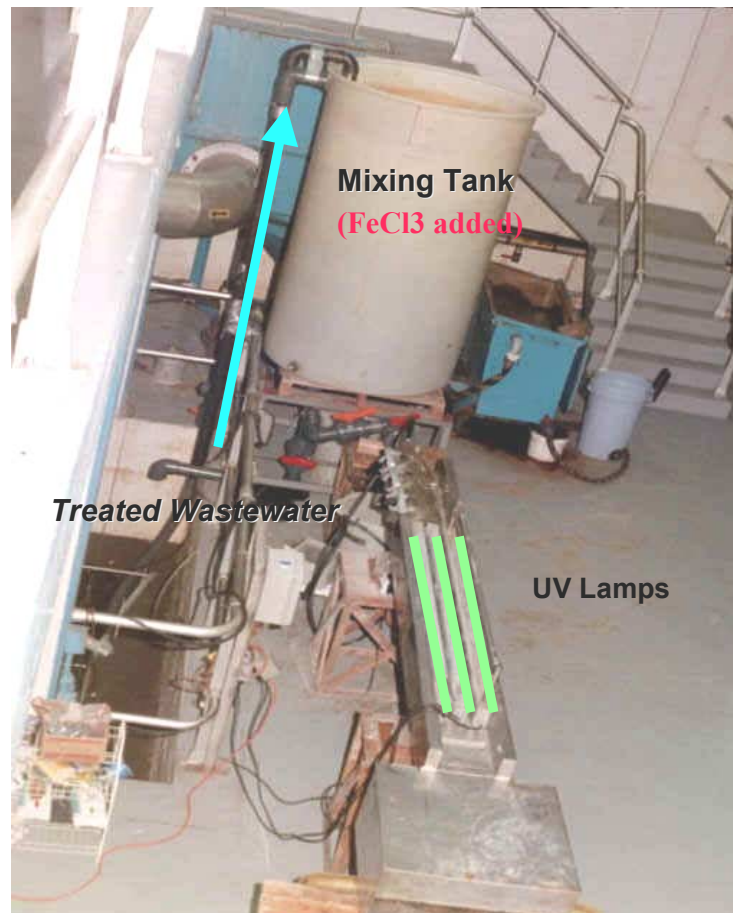


Figure 1: Pilot plant setup

Table 2: Experimental conditions

	Location	Test Number	Iron concentration	Fouling time	Channel Setting
Jun-99	St-Eustache	1	3 mg/L	1-8 days	No controls
Aug-99	St-Eustache	2	3 mg/L	1-8 days	No controls
Jun-00	Beauharnois	3	0 mg/L *	2-3 days	No controls
Jul-00	Beauharnois	4	2 mg/L	2-3 days	No controls
Aug-00	Beauharnois	5	4-6 mg/L	1-8 days	Controls

* Background concentrations below detection limits

Sampling and analysis

A similar grab sampling approach was adopted at each research site. Each sampling event included sampling from downstream of the lamps at the end of each channel (effluent), followed by a sample taken just upstream of the dividing plate (influent). Sampling was done in this order to eliminate any disturbances (iron concentration variation or particulate release downstream) induced by sampling in the reverse sequence. This allowed the disinfection performance of the lamps to be evaluated. The sample bottles were washed and disinfected prior to each sampling event, then rinsed in their respective wastewater to ensure a maximum reduction of bias errors. Collected samples were immediately placed in coolers to minimize bacterial growth and photo-reactivation. Samples were refrigerated below 4°C upon arrival at the Environmental Engineering Laboratory, in the department of Civil Engineering and Applied Mechanics of McGill University (referred to hereafter as “laboratory”; any other testing facilities will be specified). Microbiological tests were always performed within 24 hours of sampling. All other tests were performed within 48 hours. *Standard Methods* (APHA et al., 1995) (30) was used for analyses of all parameters (for fecal coliforms, the membrane filtration procedure was adopted).

The performance and impact of fouling of the UV system was evaluated through the disinfection efficiency achieved. Fecal coliforms were used as the indicator bacteria.

Bottles destined for metals analysis were soaked in a 50% acid solution for at least 24 hours prior to use. Each bottle was then spiked with 2 mL of nitric acid prior to sampling to ensure metal digestion. Metals analysis for samples taken during the summer of 1999 was performed in the Chemistry department laboratory of McGill University using an inductively coupled plasma mass spectrometer (ICP-MS) for low concentrations and inductively coupled plasma atomic emission spectrometer (ICP-AES) for high concentrations. For samples taken during the summer of 2000, only iron and calcium were measured in the laboratory using atomic absorption (AA).

Tracer studies

Tracer studies were conducted to assess and, if necessary, modify flow conditions in each channel at a flow of 1.5 L/s. It is important that all elements of flow reside the same amount of time in the reactor. The dispersion plate in the inlet box helps reduce short-circuiting. It is acknowledged that some dispersion would occur. Kreft et al. (1986) (31) have suggested a criterion to measure the spread from plug flow conditions, termed as the “dispersion number” (31; 32). Approximate flow conditions were assessed using food color dye. The flow was adjusted by modifying the openings in the perforated dispersion plate. The tracer study was then conducted by injecting a pulse salt solution upstream of the dispersion plate and measuring the change of conductivity with

time in each channel. Dispersion curves and hydraulic indices as described by Kreft et al. (1986) (31) could then be obtained for each channel to check flow conditions.

Dose estimation and collimated beam test

The collimated beam test was the approach taken to estimate the fluence delivered by the UV lamps (4).

Results

Tracer studies

The results of the tracer studies were interpreted using both residence time distribution curves obtained for each channel and hydraulic indices suggested by Levenspiel (1972) (32). These studies were repeated at the beginning of each month to ensure that hydraulic conditions remained unchanged. Dispersion numbers obtained for each channel suggested that moderate dispersion was occurring (0.046, 0.045, 0.038 for channels 1, 2 and 3 respectively). Conditions in all channels and the indices calculated were practically all within the suggested guidelines for near plug-flow conditions. Figure 2 shows a typical residence time distribution for Channel 1.

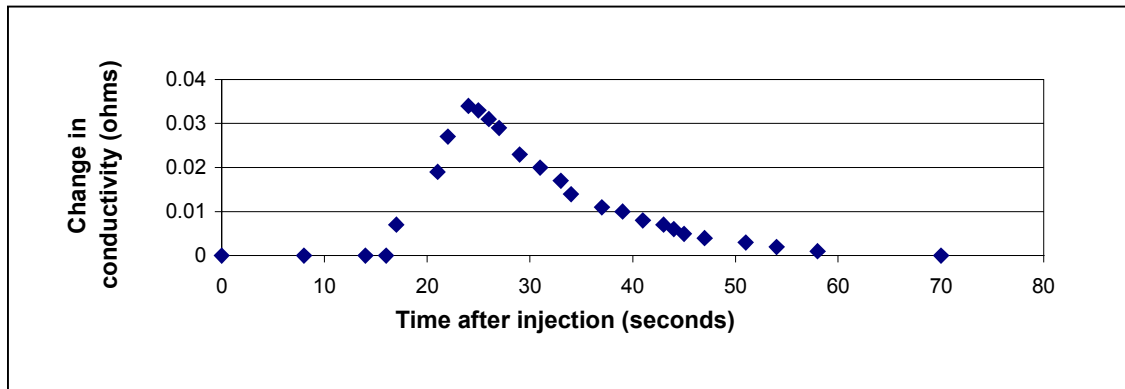


Figure 2: Channel 1 residence time distribution curve.

Collimated beam tests

Collimated beam tests were performed at the beginning of each month to monitor the microbial quality of the influent wastewater. Using the N/N_0 ratio, it was possible to estimate the UV fluence while accounting for water quality fluctuations. These results of these tests showed that microbial quality varied significantly from June to August at St-Eustache, most probably due to their technical failures, whereas that of Beauharnois remained constant. Comparing both plants, a higher disinfection was obtained at Beauharnois for a similar fluence, however, in general, both plants achieved 3- log disinfection at around 20 mJ/cm^2 (Figures 5 and 6).

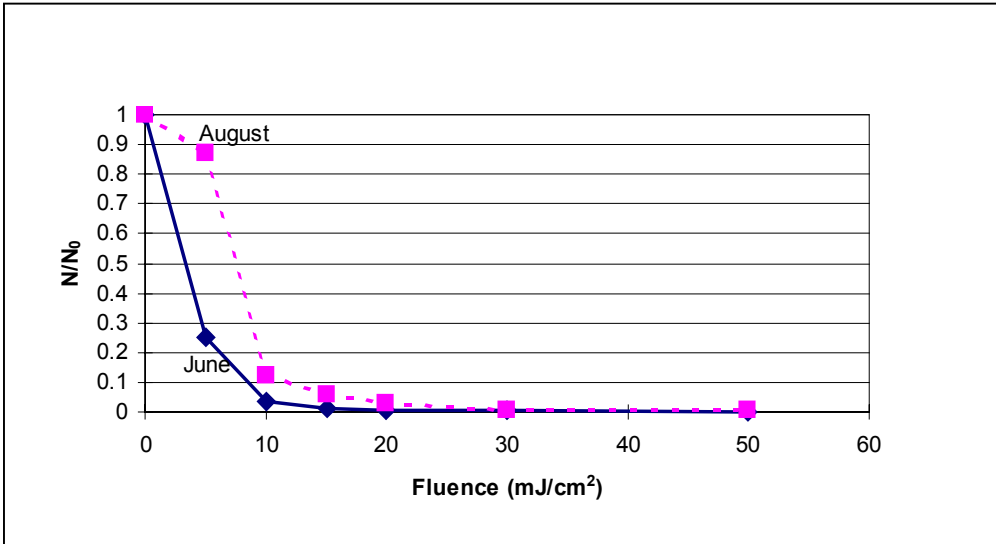


Figure 3: Collimated beam results (N/N₀), St-Eustache

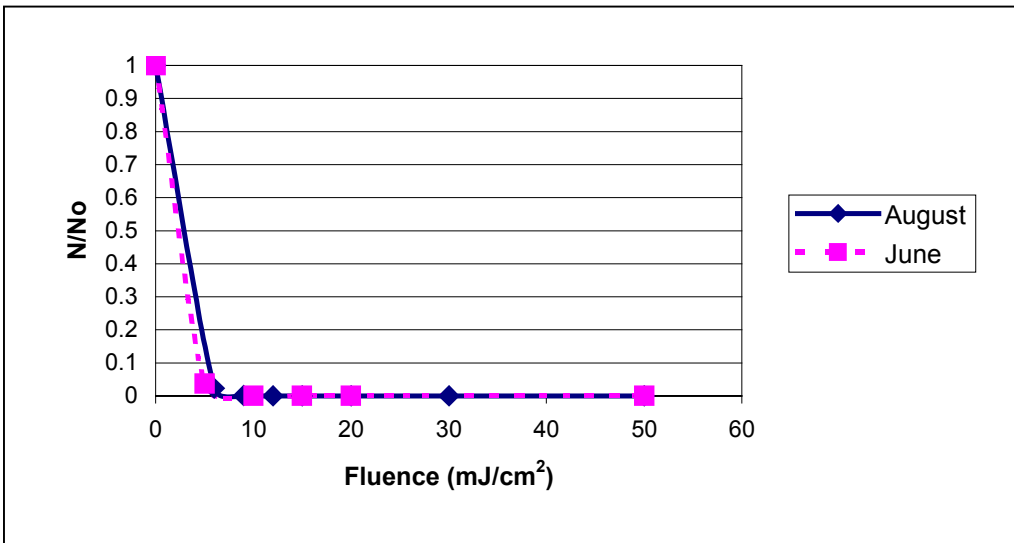


Figure 4: Collimated beam results (N/N₀), Beauharnois

Wastewater quality

Standard wastewater quality parameters are presented in Tables 3 and 4.

Table 3: Pilot plant influent wastewater quality (St-Eustache)

Parameters	June, 1999		August, 1999	
	Range	Average	Range	Average
Fecal Coliforms (CFU/100mL)	7,000- 116,000	47,500	13,000-54,000	29,300
COD (mg/L)	28-90	51	24-78	49
UVT (%T)	40-61	51	58-64	61
Run SS (mg/L) *	12-18	16	9-17	13
VSS (mg/L) *	3-11	7.5	3-9	7
Iron (mg/L) *	1.92-2.4	2.1	1-3.7	2.1
Calcium (mg/L)	33-38	35.3	24-35	31
Alkalinity (mg CaCO ₃ /L)	--	--	--	115
Hardness (mg CaCO ₃ /L)	--	--	--	160

*Outliers were removed in these cases

Table 4: Pilot plant influent wastewater quality (Beauharnois)

Run Parameters	June, 2000		July, 2000		August, 2000	
	Range	Average	Range	Average	Range	Average
Fecal coliforms (CFU/100mL)	40,000-217,000	135,000	33,000-130,000	70,300	30,000 - 120,000	60,000
COD (mg/L)	--	--	23-34	28	9.8-29.5	20
UVT (%T)	75-80 *	77	60-68	64	44-57	50
Turbidity (NTU)	1.5-4.8 *	3.24	2.3-4.2	3.1	6.3-7	6.6
SS (mg/L)	7-38	16	8-18	12	5-33	20
VSS (mg/L)	1-23	10	0.67- 6.67	3.5	3-21	9
Iron (mg/L)	0 **	0	1.9-2.4	2.2	3.9-6.3	5.2
Calcium (mg/L)	--	--	--	--	73-75	74
Alkalinity (mg CaCO ₃ /L)	--	--	--	--	131-160	144
Hardness (mg CaCO ₃ /L)	--	--	--	--	200-240	220

*Outliers have been removed in these cases

**Background values were below the detection limits

Pilot plant influent water quality

St-Eustache

Suspended solids (SS) in the effluent averaged 16 mg/L throughout the runs, varying between 10 and 20 mg/L. It is interesting to note that VSS varied consistently, with SS representing on average slightly more than 50% of the TSS. VSS gives an indication of the organics content of the treated wastewater.

COD, SS and VSS were observed to weakly follow the same pattern over this period although no correlation could be drawn between these parameters. COD averaged 50 mg/L but fluctuations were within a wide range (30 – 80 mg/L).

A target concentration of 3 mg/L of iron was aimed for, but iron concentrations fluctuated considerably between 0.5 and 3.8 mg/L. This was mainly due to the fact that the water level in the mixing tank, and hence the flow rate to the UV system, varied considerably from day to day.

As expected, hardness metals (Ca and Mg) constituted the majority of the metals in solution, although calcium concentrations never exceeded 38 mg/L and Mg fluctuated between 7 and 14 mg/L. Thus the wastewater treated can be categorized as very soft water. The St-Eustache WWTP adds alum for phosphorus removal during the summer periods. This resulted in residual Al concentrations of approximately 1 mg/L or less in the influent of the pilot plant.

Beauharnois

SS varied on average between 12 and 20 mg/L throughout the summer. Table 4 indicates a wider range of values compared to that observed at St-Eustache. This is mainly due to the attached algae on the lamp brackets close to the sampling points that released small particles when disturbed. VSS comprised a high proportion of the SS in June (approximately 62.5%), but were well below 50% in July and August.

COD measurements were well below those observed at St-Eustache (around 20 mg/L compared to 50 mg/L). Turbidity and UVT were observed to vary in opposite directions throughout the summer. This decrease in UVT also corresponded to an increase in iron concentrations.

Metals analysis was initially restricted to iron at Beauharnois. This was subsequently observed to have been a poor decision, as iron concentration variations did not seem to initiate any fouling. Thus calcium and phosphorus were also analyzed in August. The calcium concentration was approximately 74 mg/L while phosphorus concentrations were below detection limits, i.e., less than 0.1 mg/L.

Coliform counts and disinfection achieved

St-Eustache

Coliform counts varied considerably during the month of June due to instabilities in the system. Performance was more stable in August, nevertheless, collimated beam results (Figure 3) showed that the treatment plant's effluent was more difficult to disinfect then.

Effluent fecal coliform counts were taken regularly to monitor the efficiency of the lamps. In order to take into account the effects of water quality, results were compared on a percentage disinfection basis instead of total effluent coliform counts. Disinfection efficiency dropped after about a week of fouling in June. In August, after one week of operation, disinfection efficiency had not changed. This difference in behaviour is probably due to the poor water quality observed in June: double the coliform counts and slightly higher SS.

Beauharnois

Coliform counts were elevated in June (average 135,000 CFU/100mL) and decreased in July and August by about half (60,000 – 70,000 CFU/100mL). Nevertheless, collimated beam results (Figure 4) have shown that similar UV fluences were required in June and August to achieve the similar levels of disinfection. Moreover, as was observed at St-Eustache, effluent coliform counts followed similar trends as influent coliform counts.

As for the disinfection efficiency, it was never observed to go below 99%. In fact, 3 to 4 log disinfection was achieved most of the time at all iron concentrations after one week of operations (even as high as 6 mg/L). There was no foulant deposition on the quartz sleeves of the UV lamps.

Foulant analysis

No foulant whatsoever appeared on the lamps at Beauharnois. At St-Eustache, some foulant appeared on the lamp brackets but very little on the lamps themselves. The minute foulant that appeared on the lamps covered both ends of the quartz sleeves identically. There was no foulant on the middle third of the lamp. This fouling pattern does not support the theory that thermally-induced co-precipitation of polyvalent metals is the main fouling mechanism. Other mechanisms, such as gravitational settling may have played an important role. This is supported by the fact that more foulant was observed to deposit on the upper lamp than on the lower one.

It was difficult to quantify the foulant because the sample volumes were very small. Metals analysis were performed, but the size of the sample made it impossible to quantify suspended solids and volatile solids in the foulant.

The relative proportions of inorganic contents of foulant material are presented in Figures 5 and 6. Iron and calcium were found in the highest concentrations, followed by alum and phosphorus. Iron concentrations increased with time, but similar patterns were not observed for the rest of the polyvalent metals.

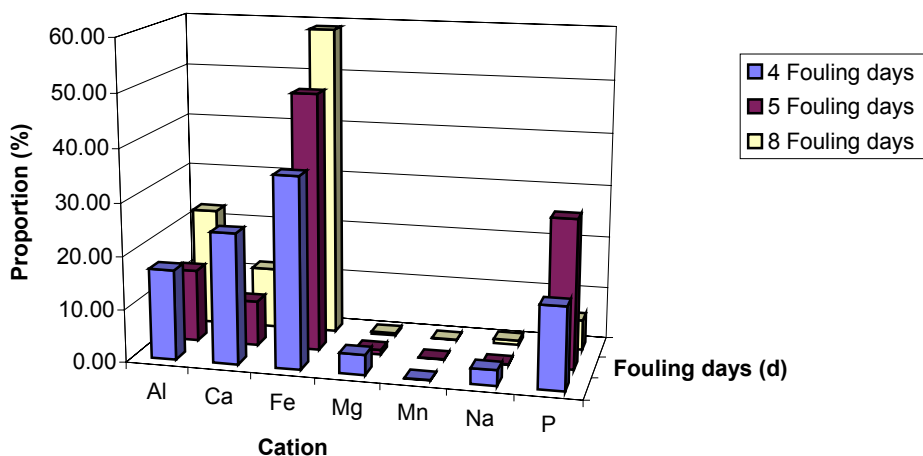


Figure 5: Relative proportions of inorganic foulant constituents (channel 1) (St-Eustache)

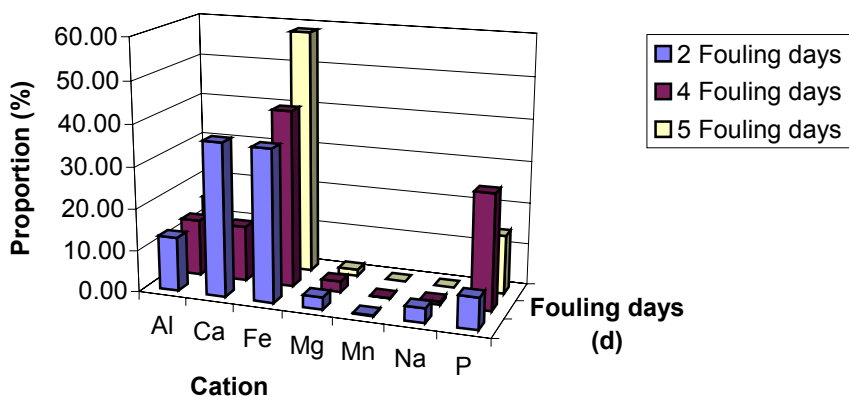


Figure 6: Relative proportions of inorganic foulant constituents (channel 2) (St-Eustache)

Discussion

Physical wastewater quality parameters

UV disinfection performance is obviously highly dependent on the UV fluence applied. As mentioned previously, UV transmittance (UVT), suspended solids (SS) and turbidity are the parameters most frequently studied by researchers. UVT however is in itself affected by turbidity and SS. To what extent each of these parameters can be correlated to one another was found to be dependent on the quality of the incoming wastewater.

According to preliminary laboratory experiments performed as part of this study, uncomplexed iron at concentrations of 3 mg/L should yield a decrease in UVT of approximately 8 to 10 %. For concentrations of 6 mg/L, UVT should decrease by approximately 20%. However, the decrease in UVT of the wastewater exiting the Beauharnois pilot plant was found to be between 13 and 18 % at 3 mg/L and up to 30% at 6 mg/L of Fe. This could be due to the formation of iron complexes with dissolved or suspended organic or inorganic compounds formed in the wastewater.

Anticipating the absorbing role of iron, both periods (with and without iron addition) were considered separately. Linear correlation coefficients (r^2) between turbidity and UVT were above 0.8; in June, when no iron was added, the correlation coefficient was as high as 0.98 and in July and August, it was approximately 0.82. However, combining results from June, July and August, the correlation coefficient drops to 0.39 (no correlation). This suggests that the complexing role of iron was considerable. Also, noting that VSS in the wastewater constituted at times more than 50% of the total SS, substantial amounts of organic matter (OM) could have been present to form complexes with iron. These iron-OM complexes would then be responsible for the increase in UVT absorbance. This is in agreement with the findings discussed in the introduction, where iron was found to be reduced in the presence of OM and form complexes with them.

No correlation was observed between UVT and SS. This is due to the dispersive and absorptive properties of SS that may reduce the actual transmittance perceived by conventional measurement apparatus. An integrated sphere spectrophotometer can account for these discrepancies. Disinfection efficiency however remained very high at both treatment plants. This may be due to the fact that most of the micro-organisms were not attached to particles and thus bacteria shielding was not considerable at these treatment plants. Generally, at high SS concentrations (20 mg/L and above), the negative effects of SS should be enhanced and this can be observed in the weak correlation coefficient (0.62) obtained for the August run at Beauharnois when SS concentrations averaged 20 mg/L. But that is not always the case as reported by Gehr et al. (1993) (4): kill ratios seemed to increase with increased SS. Furthermore, in June 1999, similar results were obtained at St-Eustache, where UVT seemed to increase with increased SS, albeit with a weak correlation of 0.6.

Fouling process: inconsistencies in the role of iron

The initial goal of this study was to observe the nature of the foulant. According to most work done on UV disinfection, elevated iron concentrations should yield high fouling rates. No fouling was observed at Beauharnois after more than a week of operations, and disinfection efficiency did not drop below 99.9% at any time during the summer of 2000. At the St-Eustache wastewater treatment plant, disinfection efficiencies dropped down to 87% at times (after 8 days of operations). However, practically no fouling was observed on the quartz sleeves. This suggests that iron in itself is not at the source of the fouling problem.

The pilot plant setup at both treatment plants was similar to allow for comparison of wastewater qualities and possible precursors to fouling. Influent fecal coliform counts, SS and VSS were in the same order of magnitude at each plant. Some of the main differences included:

- Phosphorus content of St-Eustache wastewater: 0.3 – 0.9 mg/L, less than 0.1 mg/L at Beauharnois
- Alum addition at St-Eustache: 0.7 – 1.1 mg/L, none at Beauharnois
- Half the calcium content at St-Eustache: 30 – 35 mg/L vs. 74 mg/L at Beauharnois
- Double the COD concentrations at St-Eustache: 51 mg/L vs. 20 mg/L at Beauharnois

Besides these differences in wastewater quality parameters, it should be noted that St-Eustache used an enclosed upstream biofilter system whereas Beauharnois operated open-air oxidation ditches. The open-air system encouraged algal growth in the UV system and other parts of the treatment plant.

In the presence of phosphorus, Fe(III) will first react with the phosphorus before forming any complexes with organic matter or other polyvalent metals (20, 21). Sheriff and Gehr (2001) (8) found that greater fouling rates occurred when phosphorus and iron were added simultaneously. Figures 5 and 6 show that iron concentrations in the foulant were approximately double those of phosphorus. This is close to the complexation ratio of 5 Fe : 3 P suggested by Sheriff and Gehr (2001) (8). Influent wastewater at St-Eustache had up to 1 mg/L of phosphorus whereas that of Beauharnois had less than 0.1 mg/L. This may explain why the UV system in St-Eustache experienced some fouling whereas that at Beauharnois did not.

It is generally agreed that alum addition will result in the formation of flocs that will deposit on quartz sleeves by sedimentation (1, 11). When used as a coagulant, alum constituted a considerable proportion of the accumulated foulant in their studies. This can be seen in this study, Figures 5 and 6, where alum ranks third or fourth as a foulant constituent, behind iron, phosphorus and calcium.

Calcium concentrations averaged 30 - 35 mg/L at St-Eustache compared to 74 mg/L at Beauharnois. Both of these concentrations were probably too low to induce calcite precipitation under the temperature and pH conditions present at the treatment plants. According to Diaz (1994) (19), a minimum threshold for calcite precipitation was established at 80 mg/L. Moreover, the initial concentration of calcium being low, most of the iron added would have acted as an inhibitor of calcite formation. This would explain the absence of foulant at Beauharnois and the prolonged induction period at St-Eustache. Nevertheless calcium remains one of the most important components of fouling material; in this study and most of the other studies examined for this project, calcium is the only polyvalent metal always present in large proportions in fouling material (Table 1). This also supports Banwart's (1999) (24) suggestion that calcite is a significant sink for ferrous iron. If iron were to precipitate due to co-precipitation with calcium and other polyvalent metals, high calcium concentrations would be needed to initiate this reaction and would thus result in relatively high calcium concentrations in foulant material.

Iron is introduced into the wastewater as ferric (Fe(III)) chloride. However, according to the literature, iron is reduced to Fe(II) in the presence of organic matter (OM) and UV irradiation accelerates this process. Fe(II) is much more soluble than Fe(III) (27). Iron is more likely to precipitate as Fe(III)-OM complexes. These latter complexes are the oxidized products of Fe(II)-OM complexes (28). Thus in the form of Fe(II) and in the presence of organic compounds, iron will most likely remain in solution and no precipitate or fouling would be expected except by co-precipitation with calcite and other polyvalent metals or by sedimentation of preformed flocs. Photo-chemical oxidation is not expected to play an important role in the precipitation of iron unless in the presence of enough calcium (in the form of calcite) to co-precipitate the iron.

COD concentrations at St-Eustache were approximately double those observed at Beauharnois. Influent wastewater to the treatment plant was of poor quality and had relatively high bacterial counts. Vaughan and Ord (1994) (28) stipulate that besides abiotic oxidation, bacteria can be a major biotic factor involved in iron oxidation that results in Fe precipitation. That may explain the fast rates of fouling observed by Gehr et al. (1993) (4) and Jensien, (1998) (5) in high bacterial count wastewaters. In these studies referred to, coliform counts were up to two magnitudes higher than those observed at Beauharnois or St-Eustache. Bacterial oxidation was not expected to have played any role in this research.

Conclusions

This study investigated the role of iron on UV lamp fouling. The results obtained suggest that iron alone is not at the center of the fouling problem observed in UV disinfection systems. Fouling is rather a complex interaction between biological and chemical components that could inhibit or accelerate the fouling process depending on their proportionate concentrations. In this study, the main parameters identified as being important contributors to the fouling process were iron, calcium, phosphorus, natural organic matter, light intensity and last but not least, microbial population. The effects of temperature did not appear to be an important fouling factor for low pressure mercury UV lamps.

Where foulant was present in large amounts and iron was added, even in low amounts (as low as 0.1 mg/L), iron constituted the highest proportion of foulant material. Phosphorus plays a major role in the precipitation of iron. However, it is not possible to simply correlate the amount of foulant to the concentrations of iron in wastewaters. Under a non-fouling environment, increasing iron concentrations will not necessarily increase fouling rates, as was observed in this study. The inhibiting role of iron should be taken into consideration and furthermore, one approach against fouling may indeed be to increase the iron concentration in the wastewater and hence promote its inhibiting effects on calcium precipitation.

Calcium seems to be an indispensable element for foulant deposition. All fouling studies reported substantial amounts of calcium in the foulant, regardless of the presence of iron. It appears that concentrations of at least 115 mg/L of calcium are needed to induce calcite precipitation, which in UV systems would translate into inorganic fouling precipitation, at neutral pH conditions and ambient temperature conditions. Fouling could be therefore be considered to be a hardness controlled process.

Iron is most soluble as Fe(II). At neutral pH values and under aerobic conditions, iron is oxidized to Fe(III), which will most likely precipitate as ferric hydroxide $\text{Fe}(\text{OH})_3$ or possibly $\text{Fe}(\text{PO})_4$. If sufficient amounts of naturally occurring organic matter are present in the wastewater, they will form complexes with Fe(II) species and outcompete the oxidation reaction of Fe(II) to Fe(III). Hence the complexation of Fe(II) with organic compounds, mainly humic acids, will lessen iron precipitation. This reduction/complexation process is further enhanced by UV irradiation. Thus UV irradiation will, in this case, reduce the fouling potential.

Finally, in poor quality wastewater effluents with high microbial concentrations, fouling may occur at accelerated rates because of the oxidizing capabilities of bacteria, as well as the presence of extracellular material, proteins, bacterial debris, etc. The rates of these reactions are not well known, however the time required for a significant effect to occur is expected to exceed the exposure time of the water body to UV irradiance. Hence, this case might be restricted to wastewaters containing iron prior to the addition of iron coagulants or to wastewaters in which iron is added at the beginning of the overall treatment process. Again, the specific role here of UV irradiation is to inhibit the action of microorganisms and hinder the production of foulant material. In practice, this phenomenon is evident in systems where the UV lamps have been switched off; accelerated fouling conditions are experienced then.

This study did not support the theory of heat-induced co-precipitation, based on the irregular deposition of foulant on the quartz sleeves. Sedimentation and shearing did however play a more important role on fouling rates. Moreover, hydraulic conditions, such as flow rates (0.7 L/s – 1.7 L/s at St-Eustache) and turbulence, were observed to affect the extent to which foulant deposited and remained on the sleeves. A simple change in flow (+ 0.2 L/s) was enough to remove accumulated foulant material on the quartz sleeve and increase disinfection efficiency.

Correlations between various wastewater quality parameters and UV disinfection were studied at both treatment plants. Suspended solids, turbidity and UVT proved to be independent from each other. One might attempt to correlate COD with fouling rates, or further yet, with disinfection efficiency, but this was not evident herein.

The scope of this study did not cover what could be happening at a micro-scale. In fact, studies conducted by Bouhairie et al (2000) (33) have shown that within the boundary layer, temperature gradients just above the

surface of the lamp existed. In these regions, heat-induced co-precipitation might play an important role. Turbulence models have also shown that it is possible for heat elements to leave that boundary layer, thus extending the zone in which heat-induced co-precipitation might be important. These effects will be more pronounced in medium-pressure lamp UV systems, which experience much higher temperatures on the lamp surface than low-pressure lamp systems. Also, the lower velocity occurring in these boundary regions will experience high UV doses that may enhance the action of UV light. Further research is required to identify and quantify the effects of these micro-scale events.

Recommendations

In light of the studies examined in this paper, it is clear that future fouling studies should depart from focusing only on inorganics, and should examine more closely the role of other wastewater constituents, namely biological contents and NOM. Although the occurrence of inorganic fouling is almost certain, it is not always the dominant fouling mechanism.

It would be insightful to study any relationship that may exist between iron and organics. Not enough data could be gathered in this study, however an understanding of this relationship could be a key to reducing the amounts of iron deposits in UV systems.

Biological contribution to fouling occurs partly because of the oxidizing capabilities of bacteria. The rate at which this oxidation may take place is probably dependent on the concentration of bacteria in the wastewater (poor quality wastewaters tend to experience more rapid fouling) but also on the exposure of bacteria to iron. The bacteria must be acclimated to iron in order for oxidation to occur, and only certain types of bacteria will be able to perform such oxidations. Hence, it would be worthwhile to study the role of acclimated bacteria on UV lamp fouling.

It is obvious that UV fluence is crucial for efficient disinfection. However, UV irradiation initiates various reactions besides deactivating bacteria. These reactions, such as the release of highly reactive radicals, should be explored in more depth, especially in the presence of substantial amounts of NOM. In this context, one might also consider the effects of varying lamp intensity as opposed to just varying the fluence.

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