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

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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 author.

As part of this study, a pilot UV system was installed at the effluent line of two different biological wastewater treatment plants. The first plant included biofilters supplemented by alum addition during the summer for phosphorus removal. The second plant used an oxidation ditch and no coagulants were added. Iron was added to the UV pilot system influent in both cases to simulate post-coagulation of phosphorus. The Trojan UV System 3000 pilot consisted of three separate channels each containing 2 lamps. Two channels were used as controls by having just the sleeves (without UV lamps) in one of them and non-UV lamps (i.e. lamps with visible light output only) in the other. The third channel contained sleeves with two low-pressure UV lamps. The flow in each channel was maintained around 1-1.5 L/s and the 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. 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 and proteins analyses were performed on the foulant itself.

In many of the published studies, the use of iron coagulants resulted in considerable amounts of foulant accumulated on the quartz sleeves, with iron being the main constituent of the foulant. However, 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 the source of the fouling problem; one cannot correlate fouling with simple measurements of iron concentrations. Other parameters need to be investigated. A comparison of influent water quality parameters 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 physico-chemical or biological, seems to stimulate different responses to UV radiation.

RESUME

Cette étude explore les effets des paramètres des eaux usées sur les taux d'encrassement de lampes UV, à travers un receuil de divers études entreprises dans ce domaine par differents groupes de recherche ainsi que par l'auteur lui-même.

Une usine pilote fut installée à l'aval de deux usines différentes a procédés biologiques. La première usine employait des biofiltres et ajoutait de l'alum en été pour controler les rejets de phosphore, alors que la seconde employait des bassins d'aérations externes et n'ajoutait guère de coagulants. Du fer fut ajouté dans les deux usines afin de simuler la poste-coagulation du phosphore. Le pilote, un système Trojan UV 3000, était muni de trois canaux contenant chacun deux lampes. Deux de ces canaux étaient employés en tant que canaux de contrôle, l'un contenant des lampes éteintes et l'autre des lampes a lumière visible. Le débit de chaque canal était maintenu à environ 1 - 1.5 L/s et les concentrations de fer variyaient entre 0 et 6 mg/L. La durée des expériences ne dépassait pas dix jours afin d'explorer les phases initiales de l'encrassement. Les coliformes fécaux etaient utilisés pour determiner indirectement les dépots d'encrassement. La DCO, les MES et MVES. la température, la turbidité, le pH, la transmission UV et les concentrations en métaux étaient aussi mesurés. Enfin, des analyses de protéines et de métaux étaient performés sur les échantillons d'encrassement receuillis.

Dans la plupart des études publiées, l'ajout de coagulants à base de fer engendrait des augmentations de dépot inorganic et organic sur les lampes UV. le fer constituant la majorité de cet encrassement. Cependant, dans les deux usines étudiées. l'ajout de fer n'a généré presque pas d'encrassement. Ceci suggère alors que le fer seul ne serait pas à la base du problème d'encrassement ; l'on ne pas relier le taux d'encrassement à la concentration de fer utilisée. Une comparaison des paramètres de certaines eaux étudiées suggère que le calcium jouerait un rôle important dans le procédé d'encrassement. De plus, il semblerait qu'un taux élevé de bactéries accélèrerait le taux d'encrassement. Ainsi, le type de procédé precédant la désinfection, qu'il soit biologique ou physique/chimique, semble engendrer des résultats différents au traitement UV.

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Chapter 1 Introduction

Ultraviolet wastewater disinfection has proven its benefits on the market for more than a decade. Its advantages include the absence of disinfection byproducts release, its ease of installation and operation and finally, the practically non-existent danger it presents to the plant operators (Mishalani et al., 1996; Le et al., 1996). Professionals in the field have questioned its capability to reach similar levels of disinfection as chlorine or ozone and also its applicability in large-scale wastewater treatment plants. Another disadvantage of UV lamps is their constant need to be cleaned. Foulant that accumulates on the quartz sleeves of UV lamps inhibits the passage of UV light to the surrounding waters, thus reducing the efficiency of the treatment. The fouling phenomenon is not yet well understood and few studies have been initiated to study it in detail.

In many of the published studies, the use of iron coagulants resulted in the accumulation of considerable amounts of foulant on the quartz sleeves, with iron being the main constituent of the foulant (Lin et al, 1999; Pinto and Santamaria, 1999; Sheriff and Gehr, 2001). The nature of the foulant itself remains unknown. Some affirm it is of organic nature while others postulate it is of inorganic nature or a combination of both. Biological material has also been identified as a constituent of foulant. Determining the nature of the foulant could give an insight into why fouling occurs and how it could be reduced or even eliminated.

Hence, specific objectives of this thesis were (a) to study 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, metals and proteins. 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 complete review of published works were then used to provide a comprehensive interpretation of the role of iron in fouling.

Chapter 2 Literature review

2.1 UV Disinfection

2.1.1 General

UV systems depend on four main parameters: the emission spectrum of the UV source, the intensity of the irradiation and the exposure time (fluence), the sensitivity of microorganisms involved and the reactor performance (Meulemans, 1987).

There are two types of lamps currently being used in wastewater disinfection: lowpressure lamps and medium pressure lamps. Low-pressure mercury lamps emit 85% of their light at a wavelength of 253.7 nm, which falls within the optimal germicidal range (Le et al., 1996). The emission spectrum of a medium-pressure lamp is much wider than that of a low-pressure lamp. Overall, they have a poorer effective efficiency when compared to low-pressure lamps, however the convenience of being able to treat higher volumes using fewer lamps compensates for that deficiency (Bircher and Simms, 1998).

Disinfection of water and wastewater with ultraviolet (UV) light is a physical process based on the photochemical alteration of bacteria's DNA. Microorganisms have to absorb the UV radiation in order for disinfection to take place. Moreover, different microorganisms respond differently to UV radiation. Thus, it is important that microorganisms receive the fluence necessary for their deactivation (Bolton, 1999).

As for the performance of the reactor, it depends on the hydraulics of the reactor and the disposition of the lamps. Currently, most wastewater disinfection systems are open channel systems with submerged lamps. Lamps are placed either in the direction of the flow or perpendicular to the flow. The performance of both systems is generally the same, although vertical systems have been observed to fail more often than horizontal systems (Kwan et al., 1996; Correia and Snider, 1993).

2.1.2 Effects of water quality parameters on UV disinfection

UV disinfection efficiency has been reported to vary from site to site. This has prompted researchers to attempt to establish correlations between quality parameters of the wastewater entering UV systems and the disinfection efficiency. The parameters most frequently identified as affecting disinfection efficiency are UV Transmissivity (UVT). turbidity and suspended solids (SS) (Correia and Snider, 1993; Job et al., 1995; James et al., 1996; Mishalani et al., 1996).

• UV Transmissivity

Transmissivity is the amount of light available to irradiate organisms at a certain distance from the lamp. Many compounds in water absorb UV radiation, thus limiting the amount of light reaching the organisms. Such compounds include metals, sulfates, nitrates, humic substances and many more. Absorbance was also found to vary with the type of process that preceded UV disinfection (Loge et al., 1999). Light absorbance has been correlated with the porosity of the absorbing compound. UV light can penetrate porous particles, depending on its porosity, but not solid material. Of interest for this study, chemically induced flocs, especially iron-induced flocs, were found to be less porous than those that were biologically induced (Emerick et al., 1999).

Mishalani et al. (1996) have suggested that there is no reduction in UVT at iron concentrations below 0.3 mg/L. Above that threshold, reduction of as much as 30% in the UVT of distilled water was observed at concentrations of 3 mg/L of Fe. The effect of iron was evaluated in this study and is presented later in Section 4.0. Organic compounds were also reported to reduce UVT (Mishalani et al., 1996; James et al., 1996). Measured as total organic carbon (TOC), UVT decreased with increasing TOC. James et al. (1996) assign more importance in their study to TOC effects than iron effects.

• Suspended Solids (SS)

Since UV light does not penetrate solid material, suspended solids (SS) can shield microorganisms against UV irradiation. Mignot (2000) describes three scenarios possible for bacteria in treated waters: (1) when bacteria are directly exposed to UV rays, complete penetration of UV light will de-activate those bacteria. (2) SS can absorb some of the UV light as mentioned in the previous paragraph. SS can also cause scattering of the UV light. Scattered light, of reduced intensity. will only partially penetrate microorganisms. which will incur limited or no damage. (3) Light penetration will also be incomplete or negligible if bacteria are adsorbed to SS particles. Bacteria shielding is favored by the increased concentration of SS. These will have similar dimensions as bacteria $(1-5 \ \mu m)$ or will be slightly larger. This is translated in the results of collimated beam studies that show an asymptotic tailing at high UV fluences: an increase in dose beyond a certain point does not have any effect on bacteria because of the SS shielding. When high UV fluences are used, residual bacteria can be correlated with SS concentrations (Emerick et al., 1999 a, b: Mignot, 2000). Job et al. (1995) recommend SS concentrations less than 15 mg/L for adequate disinfection.

Turbidity

Turbidity is a measure of light diffusion. It accounts for both dissolved and suspended solids. Generally, the higher the turbidity, the greater the absorbance of the water body and thus the lower the UV transmissivity (Mishalani et al., 1996; Correia and Snider, 1993).

Thus one can state that UV disinfection will be highly dependent on the performance of the preceding treatment processes. The poorer the water quality of the treated wastewater, the wider the range of data obtained and generally the lower the disinfection efficiency (Correia and Snider, 1993). Because of the wide range of data obtained from poor quality effluent treatment plants, many studies resulted in poor or no correlation

between water quality parameters and disinfection efficiencies (Le et al., 1993; Blatchley et al., 1996; Jesien, 1998).

2.2 Lamp fouling in UV systems

Fouling of the outside of the quartz sleeves surrounding UV lamps remains one of the limitations of UV disinfection systems. These sleeves are submerged in the wastewater. Foulant has been observed to grow on these sleeves, rendering the treatment efficiency low, 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 whiping mechanism.

The addition of ferric chloride (a common coagulant used in North America) was observed to intensify this problem (Gehr and Wright, 1998; Jesien, 1998; Lin et al., 1999 a. b; Pinto and Santamaria, 1999; Sheriff and Gehr, 2001). This is however not always the case, as was observed in this study and in other work (Topnik et al., 1999). Iron has been shown to form complexes with dissolved organic carbon in aqueous systems, which may result in increased fouling (Lin et al., 1997). This is covered in more depth later on in this section.

One reason why the fouling problem has not yet been solved is the fact that the exact nature of the foulant itself remains unknown. It is well established that an inorganic layer comprised of mainly aluminum, calcium, magnesium, iron, sodium, phosphorus (where present) and other metals, makes up a substantial proportion of the foulant (Lin et al., 1997). There remain, however, questions concerning the presence of organic and biological compounds, reported by some as being part of the foulant (Black et al., 1993). 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 wastewaters, wastewater temperature and many other factors.

The fact that fouling is site-specific makes it harder to determine the mechanisms by which the foulant 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 (James et al., 1996; Stafford et al., 1996), whereas others required lamp cleaning twice a day (Gehr et al., 1993; Jesien, 1998). Table 2-1 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 (Lin et al., 1997, Lin et al., 1999 a, b; Sheriff and Gehr, 2001).

- 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 heatinduced precipitation of these metals.
- Gravitational settling, or impaction, was also identified as a fouling mechanism, especially in treatment plants that use alum coagulants. Preformed flocs under the effects of Brownian motion will deposit on the upper surface of the lamps.
- Considerable fouling was found to occur in regions of low flow zones and where eddies may occur, mainly close to the lamp brackets (current study), and around the lamps themselves.

Fouling has been attributed to a combination of temperature, photo-chemistry and physical processes of aquatic chemistry. It would be useful to separate individually each of these elements in order to obtain a general overview of what might be occurring in UV disinfection systems. The rest of Section 2.2 covers those aspects of aquatic chemistry and photo-chemistry that might have a bearing on fouling.

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)	 Aluminum batch observed highest log disinfection Iron deposits on quartz sleeves when FeCl₃ used 	 Influent iron to UV system was 2.12, 0.88 and 0.31 mg/L for FeCl₃ Alufer, and Alum respectively. 	Audet, 2000
Physico-chemical WWTP	Iron: 1 – 5 mg/L	 Major fouling after one day Strong correlations between Turbidity (T) & UVT, T & SS and SS & UVT 	 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
Five biological WWTPs	Influent conditions All plants had at least secondary treatment	 Fouling varied considerably from site to site High suspended solids decreased disinfection efficiency Poor quality effluent leads to considerable fouling 	• A non-fouling effluent is one with mean BOD of 10 mg/L and mean SS of 15 mg/L	Job et al., 1995
One physical- chemical WWTP One biological WWTP	Physical-chemical used FeCl ₃ Both plants observed very high FC counts (order of 10 ⁶ CFU/100mL)	 Foulant composed of 80% inorganic material Four times more iron in foulant when FeCl₃ used Very rapid fouling at both plants, <i>biological higher</i> Foulant composed mainly of Ca (highest concentration), Fe, Mg and Al. 	 Fe concentrations in foulant from OFF-lamps was half of that from UV-lamps Shearing and sloughing affect the fouling phenomena 	Jesien, 1998

.

Table 2-1: Summary of fouling studies

Experimental Site	Variables	Conclusions	Notes	Reference
Purdue University Laboratories Four physical - chemical WWTP	Various metal concentrations Coagulant addition	 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 	 <i>Iron</i> concentrations were <i>less</i> <i>than 0.1mg/L</i> in all samples tested Lower velocities, lower temperatures, lower alkalinities not discussed 	Lin et al. 1999 a, b
Biological WWTP	Iron concentrations 0-4 mg/L Al concentrations	 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 	 Initial inorganic layer followed by wet gelatinous protein-containing layer Increasing iron conc. increased all other element concentrations in foulant 	Pinto and Santamaria, 1999
McGill University Laboratories (Ca in tap water around 35.1 mg/L and Mg around 9.2 mg/L)	Iron, Iron + Phosphorus Organics	 Precipitate sedimentation was the main fouling process Greater fouling rates when phosphorus and iron added simultaneously (3:5) 	 No foulant analysis No fouling during re-circulation Heat-induced precipitation rejected 	Sheriff and Gehr, 2000
Univ. of Alberta Laboratories Biological WWTP	Increase of iron concentrations from 0.2 to 0.5 mg/L	 No increased fouling observed Ca main fouling element 	 Calcium role downplayed No influent water quality 	Topnik et al., 1999
Two biological WWTPs	Iron (0-6 mg/L)	 Iron concentrations did not increase fouling rates NO foulant appeared at any iron concentrations 	 Calcium concentrations less than 75 mg/L at both plants Low microbial counts 	This study

Table 2-1b: Summary of fouling studies (cont'd)

2.2.1 Aquatic solubility of metals

It is worthwhile noting that the fouling mechanisms observed in most heat transfer studies are substantially different from those observed in UV systems. Temperature gradients. hydraulic conditions and other parameters make these two systems too different to enable any comparison to be made between them (Lin et al., 1999 a, b). Thus the situations discussed hereafter are those that could be found under normal wastewater treatment plant conditions.

In naturally occurring systems, metals are found in very small quantities, except for hardness ions, mainly calcium and magnesium. Under suitable conditions, these ions will precipitate most likely as phosphates or carbonates. Musvoto et al. (2000) have determined the species most likely to be precipitated first under normal pH and temperature conditions.

The form of magnesium phosphate most likely to precipitate is magnesium ammonium phosphate, otherwise known as struvite (MgNH₄PO₄.6H₂O). It was found to precipitate at neutral (or higher) pH and at Mg/Ca molar ratios greater than 0.6. Magnesium carbonate (MgCO₃) is not a stable compound below a pH of 11 and thus is not considered an important factor in precipitation.

As for the calcium phosphate species, hydroxyapatite (HAP, $Ca_5(PO_4)_3OH$) was found to be the most stable one by many scientists. However, there are precursors to this mineral, mainly ACP, amorphous calcium phosphate, which in time may transform to HAP. This process usually takes months or even years, therefore for wastewaters it is more important to study ACP precipitation rather than HAP.

These precursors and their transformations are greatly influenced by interactions between pH. alkalinity, organic material present and Ca and Mg concentrations. In solutions highly supersaturated in Ca, Mg and P, ACP is amongst the first species to precipitate at

a pH of 7.0 and above (Musvoto et al., 2000). Furthermore, the presence of Mg in solution strongly affects the transformation process. ACP transformation was found not to occur when Mg/Ca molar ratios were greater than 4.0.

Calcite (CaCO₃, with no crystalline structure) is the most stable calcium carbonate and thus the most abundant species at ambient temperature and atmospheric pressure. The rate of precipitation of calcium carbonate is heavily influenced by the presence of Mg. Fe, phosphates and dissolved organics, which could increase its solubility and thus decrease its precipitation rates.

Under conditions observed in wastewater treatment plants, the aforementioned salts are the most important and most intensively studied salts that deposit in aqueous systems. Analysis of foulant material shows that the most abundant constituents are calcium, iron, aluminum, magnesium and where present, phosphorus. It is crucial to understand the interactions between these minerals in order to get a better understanding of what may happen in wastewater treatment systems. It should be noted that the following discussion is based on studies performed in most cases without UV photo-effects.

Of all the metals identified in foulant samples, calcium is the only one identified in all fouling experiments. Calcium precipitates predominantly as the calcite form rather than as calcium carbonate. Diaz (1994), while investigating the solubility of inorganic P as influenced by pH and calcium concentrations found that when calcium was below 50 mg/L the solubility of P would not be decreased; furthermore no precipitation was observed at a pH of 8.0 and below. The pH had to be increased to around 10.0 so as to induce some precipitation. However, at calcium concentrations above 115 mg/L, precipitation occurred at lower pH values (around 9.0). Other authors quoted in Diaz's paper, such as Ferguson et al. (1970, 1973) reported similar results where Ca concentrations of at least 80 mg/L were needed to induce P precipitation. Higher pH values were also found to increase the precipitation rates. Musvoto et al. (2000) had to drastically increase calcium concentrations from a range of 50 - 60 mg/L to about 5 times the initial concentration in order to obtain any form of precipitation. Thus, it appears that

80 mg/L is a threshold below which one would not expect calcium carbonate or any of its polymers to precipitate at ambient pH values.

Magnesium does not stand out as a major foulant constituent in the studies investigated. Diaz et al. (1994) restricted its role to the formation of a less-soluble precipitate that would not dissolve easily back into solution. Musvoto et al. (2000) found considerable deposits of struvite in their experiments. Struvite, as mentioned above, is a form of magnesium phosphate. However, these deposits are attributed to the complexing effects of phosphorus contained in the struvite polymer rather than on the magnesium characteristics. In contrast, they reported that precipitation rates of CaCO₃ were reduced in solutions with high Mg²⁺ concentrations. In fact, they correlated the deposition rate of CaCO₃ with the Mg/Ca ratio; the higher the ratio, the lower the precipitation rate. This is in accordance with the findings of Benjamin et al. (1977) quoted in their paper, who report Mg²⁺ as a weak inhibitor of calcite precipitation and who note that its inhibiting effects are enhanced by phosphorus at a Mg/P molar ratio of 1.5 to 2.0.

There are two aspects to the role of iron: iron is a strong complexing agent, but it is also a strong inhibitor of calcium precipitation (Johannsen et al., 1997: Takasaki et al., 1994). Therefore its role is not well understood. Iron complexation depends on the pH of the solution, the organic matter available, its concentration in solution, other metals in solutions, light and temperature.

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, it will cause less stable divalent calcium and magnesium complexes to dissociate in favor of trivalent ions. Iron forms much stronger complexes than Ca^{2+} with a wide variety of compounds, e.g., EDTA, fulvic acids, humic acids, etc. Ca^{2+} is thus outcompeted for binding sites (Nowack and Cannon, 1997). Hence, when added as a coagulant, iron is observed to be the main constituent of foulant in many studies (Table 2-1). Furthermore, Pinto and Santamaria (1999) 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) found that calcite growth rate was linearly dependent on calcium concentrations. However, when iron was added, the authors observed an increased inhibition of calcite formation with an increase in ferric ion concentration. At very high iron concentrations, calcite growth was completely inhibited.

2.2.2 Effects of natural organic matter

Natural Organic Matter (NOM) plays an important role in the solubilization of metal ions in water (Rose et al., 1998). 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 include humic and fulvic acids. Humic acids are known to have a higher UV absorbance and a higher molecular weight range than fulvic acids. On the other hand, they tend to have lower charge densities that make them harder to remove if charge neutralization is the main coagulation mechanism (Nowack and Cannon, 1997).

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. In water and wastewater treatment, the concentration of organic matter can be reduced by coagulation (Nowack and Cannon, 1997). The impact of coagulation on fouling is discussed in Section 2.3.

Iron complexation with organic matter (OM) is influenced by the iron species in solution (which can be predicted from pH - pC diagrams) and the functional groups present in the NOM. Functional groups can be either hydroxyl (R – OH), carboxyl (C=O) or methyl (R – CH₃) groups, the first two being the most reactive. Iron complexation under ambient temperature and pH conditions usually results in the reduction of iron from Fe(III) to Fe(II) (Banwart, 1999; Fukushima and Tatsumi, 1999; Nowack and Cannon, 1997; Rose et al., 1998).

2.2.3 Photo-chemistry

Unless otherwise stated, the following information is taken from Wayne (1988, 1996). In polar solvents, photochemical reactions often involve oxido-reduction steps. Many ions, one of which is iron, show a strong absorbance of ultraviolet light, usually at wavelengths of 230-285 nm. This is mainly due to charge transfer transitions, in which an electron is transferred from one ion to another, or from an ion to a solvent or vice versa. 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. Charge transfer in cations can occur either from or to a solvent (Equations 2-1 and 2-2), depending on the wavelength of irradiation and the composition of the solvent (in our case, wastewater).

$$Fe^{2+} - H_2O + hv \rightarrow Fe^{3+} - H_2O^{-} \qquad (\lambda \max \sim 285 \text{ nm})$$

$$Fe^{3+} - H_2O + hv \rightarrow Fe^{2+} - H_2O^{+} \qquad (\lambda \max \sim 230 \text{ nm})$$
(2-1)
(2-2)

The change of importance in most reactions occurring in nature is the reduction of Fe^{3+} to Fe^{2+} . From the following reaction, it was found that the quantum yield at a wavelength of 253.7 nm is less than 0.1, compared to about 4.0 for the reduction of Fe(III). It is thus not expected that oxidation of Fe^{2+} would be an important reaction (Equation 2-3).

$$Fe^{2+} + hv \rightarrow Fe^{3+} + e_{aq}$$
(2-3)

Typical processes involving Fe^{3+} complexes in solution are:

$$Fe^{3+}Cl^{-} + hv \rightarrow [Fe^{2+}Cl^{-}] \rightarrow Fe^{2+} + Cl^{-}$$
(2-4)

$$Fe^{3+}OH^{-} + hv \rightarrow [Fe^{2+}OH] \rightarrow Fe^{2+} + OH^{-}$$
(2-5)

It has been shown that ferric chloride reduction is preceded by hydrogen abstraction that results in the release of highly reactive hydrogen radicals. These radicals were reported to interact with organic matter in reducing iron, which is in agreement with the previous section, i.e., that the reduction of Fe(III) is affected by the interaction of OM with UV irradiation. This is discussed further in the next section.

2.2.4 Effect of light on iron reduction in the presence of NOM

As mentioned previously, the irradiation of an atom or molecule brings about a shift in energy levels that makes them less stable (i.e., places them in an exited state). 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 (Bin Alam et al., 2001). The irradiation of humic substances in the presence of oxygen has been found to result in a highly unstable radical HO₂/O₂^{••}. This radical may also originate from the reduction of O₂ by the primary photooxidation product of ligand-to-metal charge transfer (LMCT) reactions of Fe(III) – humate complexes (Bin Alam et al., 2001; Voelker et al., 1997). The end product of this radical intermediate is hydrogen peroxide according to the following stochiometric equation

$$2 \operatorname{HO}_2/\operatorname{O}_2^{\bullet^*} \to \operatorname{H}_2\operatorname{O}_2 + \operatorname{O}_2$$
 (2-6)

This last reaction was found to be catalyzed by the presence of iron in solution following these two reactions:

$$HO_2/O_2^{\bullet^*} + Fe(II) \rightarrow Fe(III) + H_2O_2$$
(2-7)

$$HO_2/O_2^{\bullet} + Fe(III) \rightarrow Fe(II) + O_2$$
 (2-8)

Voelker et al. (1997) found that the reaction in Equation 2-7 was the dominant reaction taking place under acidic conditions (pH 3 and 5). Fe(III) in solution is supplied via reoxidation of Fe(II) by the radicals produced from other reactions. Dissolved Fe(III) and Fe(II) are continuously and rapidly cycled back and forth and equilibrium is reached. It has already been established that pH affects the speciation of iron in solution and thus will have an effect on iron-organic complex formation. In fact, Voelker et al. (1997) report that at a neutral pH, the ability of humic substances to form complexes with dissolved Fe(III) would diminish as the free-ion activity of Fe(III) is lowered by hydrolysis. LMCT reactions were also reported to play a less important role in metalorganic complexation at higher pH values. Hence, the extent of formation of Fe(III) – humate complexes at the surface of iron oxides is expected to diminish at increased pH values (above 5.0). This lead the authors to suggest that if Fe(III) is not organically complexed at high pH values, the radicals which are usually expected to oxidize Fe(II) at low pH values (Equation 2-7) will most likely reduce Fe(III) according to the reaction given in Equation 2-8. This is in agreement with the findings of Fukushima and Tatsumi (1999), which indicate that iron is reduced in the presence of organic matter at neutral or higher pH. 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) also established that the greater the concentrations of humic acids, the greater the amount of reduced Fe(II) species.

Fukushima and Tatsumi (1999) 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 2-9:

$$Fe(III) - HA_{red} \rightarrow^{h\nu} Fe(II) - HA_{ox}$$
(2-9)

Fukushima and Tatsumi 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. At higher wavelengths. Fe(III) reduction was considerably decreased. Furthermore, they established that the reducing effects of UV occur over a wide range of pH.

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 (Howard, 1998). However, Banwart (1999) 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. Vaughan and Ord (1994) have identified bacteria as a strong oxidizing agent of iron. This is in agreement with studies performed by Wheatly (1988). which found that bacteria are a major biotic factor involved in the precipitation of iron. Hence, in high microbial content wastewaters, iron precipitation and thus fouling can be expected to occur.

2.3 Coagulation mechanisms using iron salts

Unless otherwise stated, most of the information included in this section is taken from Weber (1972). The main purpose of coagulation in wastewater treatment is to agglomerate particles in order to increase their settling rates. Most of the nonsettleable particles fall into the colloidal range. These colloids have electrostatic forces that keep them dispersed in waters. They can differ in charge depending on the type of surrounding water and on the particle type, however most naturally occurring colloids. organic or inorganic, are negatively charged at neutral pH or above (Reynolds and Richards, 1996).

Iron and aluminum salts, lime and organic polyelectrolytes are commonly used in wastewater treatment for coagulation as well as for phosphorus precipitation. Because of the acidic nature of iron and alum, they are very effective at destabilizing colloids. There are four main mechanisms by which destabilization can take place. However, Weber

(1972) affirms that in wastewater treatment, enmeshment of particles within hydroxide or carbonate precipitates and adsorption of coagulant species are the dominant processes controlling coagulation.

When a coagulant such as ferric chloride (FeCl₃), is introduced into water, it dissociates into hydrated metal ions. In an aqueous solution, these metal ions react with other hydroxyl ions to yield positively charged hydroxometallic ion complexes. Ferric chloride in wastewater releases ferric ions which are surrounded with six (6) hydrated water molecules (Fe₃(H₂O)₆³⁺). It is common to refer to this compound as Fe(III), however it should be kept in mind that in reality it is never in this simple ionic form.

Fe(III) is a metal with trivalent metal ions. It hydrolyzes in water to release mineral acidity:

$$FeCl_3 + 3H_2O \rightarrow Fe(OH)_3\downarrow + 3HCl$$
(2-10)

Coagulants are generally added in amounts that exceed the solubility of their metal hydroxide (i.e., Fe(OH)₃), which is necessary for the formation of hydroxo-metallic complexes. These excess complexes continue to polymerize until the formation of an insoluble metal hydroxide precipitate.

At equilibrium, the maximum solubility of Fe(III) is 10^{-10} mol/L at neutral pH and 15 °C. Solubility of the hydrolyzed-ferric compound (Fe₃(H₂O)₆³⁺) is even lower, at 10^{-17} mol/L. This indicates that these simple species do not play an important role in particle destabilization. Rather, it is brought about by Fe(III) polymers that form by adsorbing to colloidal particles, an intermediate step prior to the formation of an insoluble metal hydroxide precipitate. Galarneau and Gehr (1996) reached similar conclusions with aluminium: they found that phosphorus removal was due to aluminum-hydroxide-phosphate precipitate rather than Al(PO₄) precipitation when dosing with conventional alum.

The addition of sufficient quantities of ferric chloride will result in the rapid precipitation of large amounts of ferric hydroxide $Fe(OH)_{3(S)}$. Colloidal particles will be enmeshed in this precipitate as it is formed; this is referred to as "sweep floc" coagulation. There exists however a correlation between the dose of coagulant used and the concentration of colloids present in solution. The rate of precipitation of metal hydroxide will actually depend on the degree of oversaturation of coagulant. It has been determined that for low colloid concentrations, it is necessary to bring about a considerable excess of amorphous hydroxide precipitate. On the other hand, if colloids are in sufficient concentration, they can serve as nuclei for precipitate formation. The rate of precipitation then increases with increased colloidal concentration; there is thus an inverse relationship between rate of coagulation with coagulant concentrations.

Table 2-2 shows the reactions that take place in the formation of hydroxocomplexes for alum, iron III and iron II.

Reaction				log K
1)	Al3+		Al(OH) ²⁺ + H ⁺	-5.02
2)	2.AJ 3+		$Al_2(OH)_1^{4+} + 2H^+$	-6.27
3)	6A]3+		$Al_{6}(OH)_{15}^{3} \div 15H^{+}$	-47.00
4)	8Al3+		$Al_{B}(OH)_{20}^{++} + 20H^{+-}$	-68.7
5)	13A]3+		$AI_{13}(OH)$; + 34H+	-97.39
6)	Al3+		$AI(OH)_{4}^{-} + 4H^{+}$	-23.57
7)	Fe ³⁺	====	$FeOH^{2+} + H^+$	-3.0*
8)	Fe ³⁺		$Fe(OH)^{+}_{2} + 2H^{+}$	-6.4*
9)	2Fe ³⁺		$Fe_2(OH)_2^{+} + 2H^+$	-3.1*
10)	Fe ³⁺		Fe(OH) ³ + 3H ⁺	-13.5*
11)	Fe ³⁺		$Fe(OH)_{4}^{-} + 4H^{+}$	23.5*
12)	Fe ²⁺	,	$FeOH^+ + H^+$	-8.3
13)	Fe ²⁺		Fe(OH) ⁹ + 2H ⁺	-17.2
14)	Fe ²⁺	╤═╧	$Fe(OH)_{1}^{-} + 3H^{+}$	-32.0
15)	Fe ²⁺	4	$Fe(OH)^{-}_{4} + 4H^{+}$	-46.4

Table 2-2: Iron and alum hydroxide reactions (Source: Benefield et al., 1982)

It can be seen that hydrogen ions are almost always liberated when metal hydroxocomplexes are formed. Thus pH will have an effect on the metal hydroxocomplex formation and on the solubility of ferric. A log[species] vs. pH diagram (Figure 2–1) illustrates that effect.



Figure 2-1: Ferric hydroxide solubility diagram (Source: Benefield et al., 1982)

It is clear from this diagram that the solubility of ferric hydroxide is increased under both acidic and basic conditions. Around neutral pH, the solubility is at its minimum, i.e., most of the precipitate can be expected at this pH (Benefiled et al., 1982). Under these conditions, "sweep floc" coagulation can be expected to be the dominant coagulation process. Adsorption remains nevertheless an important process, but below a pH of 7 and above a pH of 8, it plays a more significant role. This is very important in the sense that the effect of coagulant dose is different depending on the principal coagulation mechanism; destabilization by adsorption usually requires a lower coagulant dosage than sweep floc coagulation, with increasing dosage with increasing colloidal concentration. However, this relationship is inversed if "sweep floc" is the main mechanism.

Figure 2-2 shows the solubility of ferrous hydroxide with respect to pH. The minimum solubility of ferrous hydroxide occurs at a pH of 9. Since the pH at a wastewater treatment plant seldom exceeds 8, it is expected that most of the coagulation and precipitation will be due to the ferric hydroxide complexes.



Figure 2-2: Ferrous hydroxide solubility diagram (Source: Snoeyink and Jenkins, 1980)

2.3.1 Phosphorus removal:

Phosphorus is considered by many to be the key nutrient in controlling algal growth. Much attention is paid to its removal. Phosphorus in municipal wastewaters is found in three different forms: organic phosphorus, orthophosphorus and condensed phosphorus. Condensed phosphate must first undergo enzymatic hydrolysis to orthophosphates before precipitation can occur (Benefield and Randall, 1980).

Galarneau and Gehr (1997) found that condensed phosphates were more efficiently removed than orthophosphates. Solutions that include phosphates will produce precipitates as long as there is sufficient dissolved phosphate. If not enough phosphates are present, the only other precipitate that would form is that binding the coagulant and dissociation products of phosphates. Orthophosphate removal is apparently achieved through sorption of orthophosphates to the hydroxide of the coagulant. such as $(Al(OH)_3)$. This corresponds with the aforementioned suggestion that sweep floc is generally the main mechanism of coagulation. 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, iron will tend to react first with soluble phosphate and will destabilize organic colloidal matter only after most of the phosphorus has precipitated (Popel, 1969: Prested et al., 1977).

Chapter 3 Materials and methods

3.1 Pilot plant setup

As part of this study, a pilot plant was installed at two wastewater treatment plants. The Trojan UV System 3000 consisted of three independent channels, each containing two lamps (Figure 3.1). 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 0.076 m of water over the top lamp in each module. The water 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 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. Variations ranged from 0 to 6mg/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 mainly in the summer periods of 1999 and 2000. Table 3-1 summarizes the various conditions of each experimental run.



Figure 3-1: Pilot plant setup

3.1.1 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, North of Montreal. This plant treats the wastewater of a population of almost 40,000, the flow of which can reach approximately $20,000 \text{ m}^3$ /day. Alum is added during the summer prior to the sedimentation process for phosphorus removal.

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

3.1.2 Beauharnois

The pilot plant was moved to the Beauharnois WWTP in the summer of 2000. The WWTP is located approximately 25 Km South-West 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. Only one module contained UV lamps, whereas the other two served as controls
with OFF-lamps in one channel and NON-UV (i.e., visible light only) lamps in the other. The purpose of the controls was to separate the effects of UV photochemistry and temperature.

	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

Table 3-1: Sampling schedule and fouling days

* Background concentrations below detection limits

3.2 Sampling and analysis

3.2.1 Sampling

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 and 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 according to Standard Methods (APHA et al., 1995).

The performance of the UV system was evaluated through the disinfection efficiency achieved. Fecal coliforms were used as the indicator bacteria. The fecal coliforms bacterial group is currently the most common indicator organism used to measure disinfection efficiency for surface water discharge (Emerick et al. 1999; Correia and Snider, 1993). Some states have recommended using more persistent organisms as surrogates, mainly enterococcus. However a comparative study of both organisms indicated that they behaved similarly and that fecal coliforms remained an effective surrogate organism (Correia and Snider, 1993)

Water quality was characterized by measuring the following parameters for each sample: suspended solids (total (SS) and volatile (VSS)), turbidity, chemical oxygen demand (COD), pH, temperature, UV transmissivity (UVT), particle count and lastly the chemical composition of the influent water, mainly calcium, magnesium, phosphorus, iron, sodium and aluminum.

Table 3-2 lists the methods used from Standard Methods (1995):

Analytical test	Method number
Fecal coliforms	9221 E
Suspended solids (SS)	2450 D
Volatile suspended solids (VSS)	2450 E
Chemical oxygen demand (COD)	5220 D
Turbidity	2130
Iron digestion and measurement using AA	3111 C

 Table 3-2: Analytical method numbers (Standard Methods, 1995)

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).

Foulant samples were collected using a large rubber cone with an inner diameter equal to that of the quartz sleeves. The cone was passed along the sleeve and foulant fell directly into polyethylene containers. Each sampling event provided three samples: one sample was sent to the Lady Davis Institute for Cancer Research (Montreal) where protein analysis was performed under the direction of Dr. Bluma Brenner. Another sample was sent to the chemistry laboratory where metal concentrations would be ascertained using an ICP-MS/AES. Samples were first digested by reflux in an "aqua regia" solution, composed of a 3:1 volume-based mixture of concentrated HCl to HNO₃. Liquid samples were then analyzed using a Perkin-Elmer SCIEX ELAN 5000 ICP-MS (SCIEX,

Thornhill, ON, Canada). The last sub-sample was conserved in the laboratory to determine total solids and total suspended solids. All samples remained frozen until analyzed.

Protein analyses were performed to give an insight as to the amount of biomass that could accumulate on the lamps. A number of extraction procedures were used to isolate constituents from what appeared to be principally inorganic matrices. Procedures included detergent extraction, trichloric acid precipitation to concentrate proteins, and NaOH that solubilized the foulant material.

3.2.2 Experimental methods

a) Tracer studies

Tracer studies were conducted to ensure near-plug flow conditions in each channel at a flow of 1.5 L/s, but more importantly, to ensure that all channels were similar. 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 reduces short-circuiting. It is acknowledged that some dispersion would occur. Kreft et al. (1986) have suggested a criterion to measure the spread from plug flow conditions, termed as the "dispersion number" (Le et al., 1996; Kreft et al., 1986). 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) could then be obtained for each channel to check flow conditions.

b) Dose estimation and collimated beam test

The collimated beam test was the approach taken to estimate the fluence delivered by the UV lamps. The collimated beam test, or "bioassay" approach consists of developing a

The collimated beam test was the approach taken to estimate the fluence delivered by the UV lamps. The collimated beam test, or "bioassay" approach consists of developing a fluence response-curve using pre-irradiated influent wastewater. Sub-samples of wastewater are exposed to different fluences of UV in the collimated beam unit and finally a curve showing disinfection ratio (N/N_0) vs. fluence can be traced. Sub-samples. typically 50 mL, are placed in dishes of known depth under a collimating beam of UV light of known intensity. By varying the time of exposure, it is possible to vary the UV fluence. The fluence (F) applied is a function of the time of exposure of the sample and the average intensity of the UV light (Equation 3-1). The incident intensity can be measured with a radiometer (model: IL 1400A; with cosine diffuser lean and solar blind lens). The average is then obtained using the depth-integrated form of Beer's law (Equation 3-2) (Morowitz, 1950).

$$F = I_{avg} \times t \tag{3-1}$$

where F = UV fluence (mJ/cm²), I_{avg} = average intensity (mW/cm²), t = average time of exposure (seconds)

$$I_{avg} = I_{a} \frac{1 - \exp(-d\ln(T^{-1}))}{d\ln(T^{-1})}$$
(3-2)

where I_0 = incident intensity (mW/cm²), d = depth of sample under UV irradiation (cm). T = transmittance (when cell path length = 1.0 cm) (dimensionless)

Chapter 4 Results

This study was conducted at two different wastewater treatment plants. The results obtained at each experimental site will be discussed separately. It is however interesting first to notice the effects of iron on UVT.

4.1 Absorbing effects of iron

Iron is known to absorb UV light and was thus expected to affect UVT. Tests were performed in the laboratory to assess UVT variation with iron concentrations. Figure 4-1 illustrates the titration of deionized water with ferric chloride.



Figure 4-1: UV absorption of Fe solutions using deionized water

Similar tests were performed with wastewater samples from Beauharnois, where iron concentrations were varied. Figure 4-2 illustrates the variation of UVT with iron concentrations observed in the pilot plant. Again, excellent correlations were obtained between these two parameters.



Figure 4-2: UV absorption of Fe solutions in wastewater (Beauharnois)

It should be noted that wastewater with no iron does not have a turbidity and UVT of 0 or 100% respectively, due to the presence of other particulates in the wastewater.

4.2 St-Eustache

The St-Eustache wastewater treatment plant experienced numerous breakdowns during the summer of 1999, which limited most experimental runs to less than a week and reduced the number of results that could be obtained. It should also be noted that technical problems imposed a lengthy shutdown of the plant during the month of July. Experiments therefore took place during the months of June and August only. Ultraviolet lamps (UV) were used in the experiments and the iron concentration was maintained at approximately 3 mg/L. Summary tables of all results are produced in Appendix B.

4.2.1 Tracer studies

The results of the tracer studies can be interpreted using both the residence time distribution (RTD) curves obtained for each channel (Figures 4-3, 4-4 and 4-5) and hydraulic indices suggested by Levenspiel (1972) which are summarized in Table 4-1. Detailed calculations as well as theoretical and mean residence times are presented in Appendix A. These studies were repeated both in June and in August to ensure that conditions were unchanged after the plant had not been operating for a month.

The dispersion number (d) as proposed by Levenspiel (1972) is a function of a dispersion coefficient (D) obtained by regression, the fluid flow in the reactor (U) and the reactor dimensions (L) in the direction of the flow. It is expressed in the following equation:

$$d = D/UL \tag{4-1}$$

Reactor dispersion can be assessed by the relative spread of the RTD: the wider the spread, the greater the dispersion. The following curves show a similar pattern in all

three channels: the relative spread around the mean residence time is similar in all channels.



Figure 4-3: Channel 1 RTD curve



Figure 4-4: Channel 2 RTD curve



Figure 4-5: Channel 3 RTD curve

Numerically, the dispersion number should approach infinity in completely mixed systems and 0 in ideal-plug flow conditions. As observed in the table below, dispersion numbers in all three channels were in the moderate dispersion range (0.01 - 1.0). The indices calculated are practically all within the guidelines suggested for hydraulic conditions resembling plug-flow (Kreft et al., 1986).

Indices		Channel		Guidelines		
	Channel 1	Channel 2	Channel 3	(Kreft et al., 1986)		
t90/t10	2.1	2.1	1.95	<2		
t _p /T	6	6	6.25	>0.9		
t _f /T	3.75	4.25	4	>		
t ₅₀ /t _m	0.97	0.91	0.89	0.9 - 1.1		
d	0.0403	0.0363	0.0405	Moderate dispersion: 0.01 - 0.1		

Table 4-1: Hydraulic indices of reactor channels

 $t_{00'}t_{10}$: ratio of the time for 90% of the tracer to pass to the time for 10% of tracer to pass $t_{p'}T$: ratio of the time at which the peak concentration occurs to the theoretical residence time $t_{t'}T$: ratio of the time when the tracer is first detected to the theoretical residence time $t_{s0'}t_{m}$: ratio of the time for 50% of the tracer to pass to the mean residence time d: dispersion number

4.2.2 Collimated beam tests

Collimated beam tests were performed at the beginning of each month to monitor the microbial quality of the influent wastewater. Figures 4-6 and 4-7 show that the water quality in August was not the same as in June. Colliform counts in the beginning of August were much higher than those in June. This is partially due to the incomplete recovery of the biofilters that had failed in July. However, on an average basis, colliform counts were lower in August.



Figure 4-6: Collimated beam results (total counts), St-Eustache

Using the N/N_0 ratio (effluent to influent coliform count) as opposed to just the coliform counts, it was possible to estimate the UV fluence while accounting for water quality



Figure 4-7: Collimated beam results (N/N₀), St-Eustache

fluctuations.

4.2.3 Pilot plant influent water quality

Standard wastewater quality parameters previously mentioned were recorded throughout both runs. Table 4-2 summarizes the water quality conditions of the St-Eustache wastewater treatment plant in the months of June and August.

Suspended solids (SS) in the effluent averaged 16 mg/L throughout the runs, varying between 9 and 18 mg/L. It is interesting to note that VSS varied consistently with SS, representing on average slightly more than 50% of the SS. VSS can give an indication of the organics content of the treated wastewater.

COD, SS and VSS are observed to weakly follow the same trends over this period although no correlation could be drawn from these parameters. COD averaged 50 mg/L but fluctuations were within a wide range (30 - 80 mg/L) (Figure 4-8).

Run	June, 1999		August, 1999		
	Range	Average	Range	Average	
Parameter					
Fecal Coliforms (/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	
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)			(taken in 2000)	115	
Hardness (mg CaCO ₃ /L)			(taken in 2000)	160	

Table 4-2: Pilot plant influent wastewater quality (St-Eustache)

*Outliers were selected in these categories



----- COD mg/L - -= - SS mg/L --- 🛓 -- VSS mg/L

Figure 4-8: SS, VSS and COD in St-Eustache effluent

The variation of metals concentrations in the pilot plant influent is represented in Figure 4-9. 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 is mainly due to the fact that the water level in the mixing tank, and hence the flow rate to the UV system, was observed to vary considerably from day to day. This problem was corrected in 2000 by installing two valves to better regulate the flow. Also, concentrated FeCl₃ was diluted in a larger tank before mixing it with wastewater. Precipitation could have occurred in the dilution tank. Dilution was done approximately every three days. Poor mixing of the diluted solutions would also have contributed to the inconsistencies in iron concentrations. This was corrected in 2000 by eliminating the dilution tank and transferring the FeCl₃ to the mixing tank immediately using micro-tubes.

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.



Figure 4-9: Pilot plant influent metals concentrations (St-Eustache)

4.2.4 Coliform counts and disinfection achieved

The main parameter used to study fouling rates was the fecal coliform count. Any disinfection efficiency decrease of the lamps could be attributed largely to foulant deposition on the quartz sleeves.

Figure 4-10 below shows the variation of UV transmissivity and influent fecal coliform counts from June to August. Coliform counts varied considerably during the month of June due to instabilities in the system. Performance was more stable in August. nevertheless. collimated beam studies showed that the treatment plant's effluent was more difficult to disinfect then.

This figure also shows how both fecal coliforms and UVT can indicate poor raw water quality; this is translated as a lower UV transmittance and a higher coliform count.



Figure 4-10: UVT and fecal coliform variations (St-Eustache)

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 are presented as percentage disinfection instead of total effluent coliform counts. Figures 4-11 and 4-12 show the variation of disinfection efficiency with days of operation.



Figure 4-11: Fecal coliform disinfection efficiency (St-Eustache, June, 1999)

It should be noted that channel 2 was out of service during the month of June.



Figure 4-12: Fecal coliform disinfection efficiency (St-Eustache, August, 1999)

Disinfection efficiency dropped after about a week of fouling in June. In August, after one week of operation, disinfection efficiency had not changed. All three channels demonstrated similar disinfection behavior hence only one line is visible on Figure 4-12. This difference in behaviour is probably due to the poor water quality observed in June: double the coliform counts and slightly higher SS.

4.2.5 Foulant analysis

It was difficult to quantify the foulant because the sample sizes from a run with no disturbances were very small. Protein and metals analysis were performed, but the size of the samples made it impossible to quantify suspended and volatile solids in the foulant, which would have allowed a more appropriate foulant characterization by unit area.

• Inorganic contents

The relative proportions of inorganic contents of foulant material collected off the lamps are presented in Figures 4-13 and 4-14. 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 4-13: Relative proportions of inorganic foulant constituents (channel 1) (St-Eustache)



Figure 4-14: Relative proportions of inorganic foulant constituents (channel 2) (St-Eustache)

• Protein analysis

Protein analyses were performed under the supervision of Dr. Brenner. Figure 4-15 shows protein concentrations as a function of fouling days.



Figure 4-15: Protein analysis of foulant

4.2.6 Qualitative results (pictures)

Quantitative analysis of the foulant was limited due to the fact that very little fouling accumulated on the quartz sleeves. At times when disinfection efficiency was reduced, a semi-transparent layer was visible but it was impossible to collect enough material for proper analysis.

In general, fouling was observed to initiate on the steel brackets and make its way to the quartz sleeves. Figure 4-16 shows what is referred to herein as a "fingering" process.



Figure 4-16: Fingering process (St-Eustache)

It is interesting to note that the minute foulant that appeared on the lamp covered both ends of the quartz sleeves identically. There was no foulant on the middle third of the lamp sleeves. Lin et al. (1997) also reported foulant growth on the lamp brackets. Turbulent shear leading to particle agglomeration was hypothesized to cause such foulant deposition. This is arguable because foulant was also observed to deposit on the downstream edge of the bracket, where eddies and regions of flow stagnation would be expected to occur. Lin et al. (1997) observed that fouling on the lamp sleeves increased along the quartz surface in the direction of the flow. This was believed to be the result of thermally-induced co-precipitation of polyvalent metals such as calcium, iron, magnesium and aluminum. This was not the case at St-Eustache. The fouling pattern observed at St-Eustache does not support the theory that thermally-induced coprecipitation is the main foulant deposition mechanism: the middle third of the sleeve had no foulant. Other mechanisms, such as gravitational settling may have played an important role. This is supported by the fact that more foulant material was found to deposit on the upper lamp than on the lower one (Figure 4-17)



Figure 4-17: Upper lamp fouling (St-Eustache)

Similar work performed by Santamaria and Pinto (1999) concluded that the fouling mechanisms involved in the control channels were different than those observed in the UV channel. In fact, control channels observed rapid fouling rates, mainly organics in nature. The control lamps in all runs were also found to contain a protein mass much greater than that found on the UV lamp. At Beauharnois, control lamps did accumulate some foulant. The lower lamp of the non-UV (i.e., visible) lamp pair was rapidly covered with brown gelatinous foulant, suggesting gravitational sedimentation of preformed flocs. As to the OFF lamps, both lamps in the channel accumulated large quantities of foulant. Most of it was clearly algae material.

4.3 Beauharnois

Experiments at the Beauharnois wastewater treatment plant took place during the months of June, July and August 2000. In the months of June and July, incipient fouling was investigated; fouling was monitored hourly (during the day) for up to four days to account for the typical induction period of two to three days reported in the literature (Lin et al. 1999 a,b). All channels contained low-pressure UV lamps to increase the likelihood of fouling. In the first run of experiments (June), no iron was added and background concentrations of iron were below the detection limits of the apparatus used. In July (run 2), iron was added at an approximate concentrations were thus increased to 4-6 mg/L and control channels were inserted. One control channel contained visible light (VIS), and in a second one the lamps were turned off.

4.3.1 Tracer studies

Tracer studies were conducted in a similar manner as at St-Eustache and plug flow conditions were again achieved. However, within one day of operations, algae covered parts of the perforated screen thus rendering it ineffective. Most of the openings were clogged after 24 hours and wastewater flowed above the screen. It was impossible to ensure that plug flow conditions occurred in the channel. This inconvenience was mitigated by the fact that only one channel included UV lamps and that identical conditions were not crucial for the control channels. Nonetheless, the screen was cleaned every day.

4.3.2 Collimated beam tests

The Beauharnois wastewater treatment plant proved to be a reliable plant. It did not experience any downtime during the summer and thus collimated beam tests were only performed in June and August. The results are shown in Figures 4-18 and 4-19. It is obvious that the wastewater quality in June and August was quite similar.



Figure 4-18: Collimated beam results (total counts), Beauharnois



Figure 4-19: Collimated beam results (N/No), Beauharnois

4.3.3 Pilot plant influent wastewater quality

Table 4-3 below summarizes the influent wastewater quality to the pilot plant.

Run	June, 2000		July, 2000		August, 2000	
Parameter	Range	Average	Range	Average	Range	Average
Fecal coliforms	40,000-	135,000	33,000-	70,300	30,000 -	60,000
(CFU/100mL)	217,000		130,000		120,000	
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					131-160	144
(mg CaCO ₃ /L)						
Hardness					200-240	220
(mg CaCO ₃ /L)						

 Table 4-3: Pilot plant influent wastewater quality (Beauharnois)

*Outliers have been removed in these categories

**Background values were below the detection limits

SS varied on average between 12 and 20 mg/L throughout the summer. Table 4-3 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 (Figure 4-20). However, it should be noted that this decrease in UVT also corresponds to an increase in iron concentrations.



Figure 4-20: UVT and turbidity variations, Beauharnois

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 (Appendix C).

4.3.4 Coliform counts and disinfection achieved

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 studies have shown that similar UV fluences were required in June and August to achieve the similar levels of disinfection (Section 4.2.2).

As was observed at St-Eustache, effluent coliform counts followed similar trends as influent coliform counts. Figure 4-21 represents the coliform count variation in the month of August. This figure also presents the UVT variation for that same period.



Figure 4-21: UVT and coliform counts variations (August 2000)

As for the disinfection efficiency, it was never observed to go below 99%. In fact, a 3 to 4 log disinfection was achieved most of the time at all iron concentrations, even at 6 mg/L after one week of operations. There was no foulant deposition on the quartz sleeves of the UV lamps. Detailed tables of coliform counts and disinfection efficiencies achieved are shown in Appendix D.

Chapter 5 Discussion

The results obtained at both treatment plants will be discussed in this section. Because experiments conducted at both plants produced similar results, the results will be discussed concurrently. The first part of this discussion will focus on wastewater quality parameters and their correlation with disinfection efficiency. The second part will focus on disinfection efficiency and the fouling process.

5.1 Physical wastewater quality parameters

UV disinfection performance is obviously highly dependent on the UV fluence applied. As mentioned in Section 2.1.2, UV transmittance (UVT), suspended solids (SS) and turbidity are the parameters most often 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 the laboratory studies (Section 4.1), 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 decreases by approximately 20% (Figure 4-1). According to Figure 4-2, and daily comparisons of UVT from the wastewater treatment plant's effluent, the actual reduction 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. Turbidity was also shown to correlate slightly with UVT. Correlation coefficients 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. Corresponding graphs and data are included in Appendix E. However, combining results from all runs in the summer of 2000, the UVT and turbidity correlation coefficient drops to 0.39 (no correlation). This suggests that the complexing role of iron was considerable. Also, noting that VSS constituted at times more than 50% of the total SS, substantial amounts of organic matter (OM) could have been present in the wastewater. Moreover, both treatment plants included biological processes that usually release effluents with high OM concentrations. This is in agreement with the findings mentioned in Section 2.0, where iron was found to be reduced in the presence of OM and form complexes with them. These iron-OM complexes would then be responsible for the increase in UVT absorbance.

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. Gehr et al. (1993) came to similar conclusions where disinfection efficiencies did not correlate with UVT. Generally, at high SS concentrations (20 mg/L and above), the negative effects of SS could 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): 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 increase dSS, albeit with a weak correlation of 0.6 (Appendix E).

5.2 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. It is rather a complex interaction between the organic and inorganic contents of the wastewater, the UV radiation, the microbial population and the various iron species in solution.

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. none 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, 74 mg/L at Beauharnois
- Double the COD concentrations at St-Eustache: 51 mg/L, 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 (Popel. 1969: Prested et al., 1977). Sheriff and Gehr (2001) found that greater fouling rates occurred when phosphorus and iron were added simultaneously. Figures 4-13 and 4-14 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). 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 of 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 (Lin et al., 1997; Pinto and Santamaria, 1999). 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 4-13 and 4-14, 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 the literature presented in Section 2, a minimum threshold 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 2-1). This also supports Banwart's (1999) 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 (Section 2), 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) (Howard, 1998). Iron is more likely to precipitate as Fe(III)-OM complexes. These latter complexes are the oxidized products of Fe(II)-OM complexes (Vaughan and Ord, 1994). 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 coprecipitation 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. It is impossible to establish what could have happened in this study because of the lack of information on the nature of the organic matter present in the wastewater as well as its concentrations.

COD concentrations in 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) 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) and Jensien (1998) in high bacterial count wastewaters. In these studies, coliform counts were up to two orders of magnitude higher than those observed at Beauharnois or St-Eustache.

The Beauharnois wastewater treatment plant had slightly higher coliform counts than St-Eustache and would accordingly be expected to experience more rapid fouling rates. However, disinfection efficiency decreased faster at St-Eustache than at Beauharnois, probably due to an initial deposit of inorganic fouling. Not enough information is available to determine which mechanism, biotic oxidation of iron or co-precipitation of iron with calcium, occurs at a dominant rate in order to justify these results obtained. Moreover, it is also likely that a comparison of bacterial counts by simply comparing the averages of two periods would not represent accurately the real conditions.

Chapter 6 Conclusions and Recommendations

6.1 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 important factors in 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 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 add iron!

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 a hardness controlled process.

Iron is most soluble as Fe(II) and will be found most likely in species of Fe(II). At neutral pH values and under aerobic conditions, iron is oxidized to Fe(III), which may precipitate as ferric hydroxide $Fe(OH)_3$. 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 process is further enhanced by UV irradiation at low wavelengths.

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.

This study did not support the theory of heat-induced co-precipitation, based on the irregular deposition of foulant on the quartz sleeves. However, not enough foulant was generated in order to draw any firm conclusion about fouling mechanisms. Sedimentation and shearing did however affect fouling rates. Hydraulic conditions, such as flow rates (0.7 L/s - 1.7 L/s) and turbulence, were observed to affect the extent to which foulant deposited and remained on the sleeves. A simple observed change in flow $(\pm 0.2 \text{ L/s})$ 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. COD could to a certain extent give some insight as to the microbial content of the wastewater. One might attempt to correlate COD with fouling rates, or further yet, with disinfection efficiency.

The scope of this study did not cover what could be happening at a micro-scale. In fact, studies conducted by Bouhairie (2001) 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. Mathematical 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. Also, the low flow occurring in these regions will experience high UV doses that may enhance the action of light. Further research is required to identify and quantify the effects of these micro-scale events.

6.2 Recommendations

In light of the studies examined in this paper, it is clear that future fouling studies should depart from inorganics and examine more closely the role of other wastewater constituents, namely biological contents and NOM. Inorganic fouling is certain, however, 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, that could be a key to reducing the amounts of iron deposits in UV systems.

Biological contribution to fouling occurs because of the oxidizing capabilities of bacteria. The rate at which this oxidation may take place is probably dependent on the magnitude of bacterial content of the wastewater since poor quality wastewaters experienced rapid fouling. This can probably be translated in laboratory work as COD and would be very important to include in any future fouling study.

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

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Appendix A Tracer studies

EQUATION UTILIZED FOR TRACER ANALYSIS

$$tm = \frac{\sum (dC \times dt \times t)}{\sum (dC \times dt)}$$

$$\sigma^{2} = \frac{\sum (dC + dt \times t^{2})}{\sum (dC + t)} - tm^{2}$$
$$\sigma^{2} = \frac{\sigma^{2}}{tm^{2}}$$
$$-\zeta = 2 + \frac{D}{UL}$$

Where

df : change in time

t: time

DTL: dispersion number

tm : mean residence time

:(seconds)	C1	dc ,	dt	dC * dt * t	dC 1 dr	5 1 dC 1 dr
	665				Same Constant	
	645		— <u>-</u> -			
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Fracer study data and sample calculation for channel 1

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I racer study data and sample calculation for channel 2

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Fracer study data and sample calculation for channel 3

RTD data

	tm	t10	t90	:50	тр	ſ	
Channel I	28,93	10	- 38	20	22		:
Channel 2	28.28	14	30	25	•	•	
Channel 3	28.4	;	;-	12			

main actual residence time (seconds).

1709 (nine for 10%) of fracer to pass (seconds).

many time for 90% of tracer to pass (seconds).

(5) mine for 50% a of tracer to pass (seconds)

(p) mine at which peak concentration occurs (second -)

(i) time when tracer is first detected (seconds).

mentionl tesidence time (seconds).

Summary of tracer studies: Beauharnois

<u>June 14th, 2000</u>

Channel	tm	t10	t50	t90	tp	tf	D/UL
Channel 1	27.7	17	23	39	21	15	0.063
Channel 2	31.3	17	27	44	22	15	0.0616
Channel 3	28.7	17	26	40	21	15	0.0646







Appendix B St-Eustache results

Summary of suspended solids (mg/L) :

Date	Influ	uent	Lan	np 1	Lan	ւթ 2	Lan	np 3
	SS	VSS	SS	VSS	SS	VSS	SS	VSS
3-Jun	18.7	5.3	21.3	16	12	12	20	12
8-Jun	12	2.7	14.7	4	11.3	4.7	23.3	10.7
15-Jun	-	-	-	-	22	18.7	-	-
23-Jun	18	10.7	15.3	15.3	20.6	12	18	12
25-Jun	17.3	9.3	17.3	9.3		-	14	7
AVG	16.5	7	17.1	11.1	16.5	11.8	18.8	10.4
5-Jul	30.6	16.7	23.3	2	16.6	2.7	18	3.33
23-Jul	6	2.7	7.3	4	7.3	0	8.7	2.7
5-Aug	12.7	4.7	11	5	6.7	6.7	13.3	4.7
6-Aug	9.3	8.7	16	15.3	-	-	9	5
10-Aug	13.7	3.3	14.3	6.7	13.7	7.3	14.3	5.3
11-Aug	17	9.3	13	9	15.7	6.7	-	-
13-Aug	14	8	17.3	9	16.7	8	16	9.3
AVG	13.3	6.8	14.3	9	13.1	7.2	13.2	6.1

UVT and COD

.

		U	VT		COD mgO2/L				
					Lamp				
DATE	Influent	Lamp 1	Lamp 2	Lamp 3	Influent	1	Lamp 2	Lamp 3	
					Averag	Avera	Averag	Averag	
	%	%	%	%	е	ge	<u>e</u>	<u>e</u>	
4/6/1999	44.4	47.2	47.2	47.2	90.4	65.4	48.1	81.7	
8/6/1999	48.4	45.2	45.2	45.2	28.3	45.2	38.1	38.3	
10/6/199									
9	54.5	54.5	54.5	54.5	64.9	45.5	41.0	68.4	
13/06/99	-	-	-	-	42.2	61.0	27.8	46.7	
14/06/99	61.3	61.3	61.3	61.3	6.6	8.1	14.5	7.7	
17/06/99	-	-	-	-	31.3	34.7	-	18.0	
23/06/99	60.4	60.4	58.5	58.5	35.9	37.6	-	23.8	
25/06/99	40	40	40	40	60.8	54.5	52.2	55.6	
30/06/99	41.7	41.7	41.7	41.7	53.0	56.8	50.3	50.9	
2/7/1999	36.4	27.3	27.3	27.3	62.6	71.5	71.1	75.6	
5/7/1999	45.8	50	50	45.8	42.0	-	34.0	46.8	
23/07/99					27.1	18.1	28.4	2.8	
5/8/1999					24.2	20.0	2.8	19.8	
6/8/1999	64	61	62.5	64	66.9	65.4	52.8	62.9	
9/8/1999	57.8	58	58.6	57	28.2	53.3	49.7	46.0	
10/8/199									
9	59.5	60.5	60	61	78.3	51.2	70.2	67.2	
11/8/199									
9					46.7	68.8	56.8	68.8	
13/09/99	59.5	60	61	60	57.5	64.3	62.9	63.9	

Influent Metals Concentrations

		Influent Metal Concentrations											
	AI	Ca	Fe	Mg	Mn	Na	Р						
Date	ppm	ppm	ppm	ppm	ppm	ppm	ppm						
14/6/99	0.83	33.30	0.34	12.10	0.04	6.77	0.36						
18/6/99	0.78	38.54	1.92	14.43	0.03	7.79	0.72						
21/6/99	1.03	37.69	2.07	12.73	0.04	6.70	0.93						
23/6/99	0.99	33.13	2.42	10.28	0.04	5.61	0.55						
25/6/99	1.09	33.87	1.98	10.01	0.04	4.91	0.50						
5/8/99	0.74	31.18	2.21	10.85	0.04	6.24							
6/8/99	0.42	24.35	0.97	8.22	0.02	5.05							
11/8/99	1.01	33.84	2.17	10.46	0.42	6.55							
13/8/99	0.84	35.15	3.74	11.15	0.04	6.20							
16/08/99	0.75	34.62	0.18	11.20	0.03	6.44							
18/08/99	0.771	27.21	1.25	7.48	0.03	5.32							

Summary Disinfection Efficiency

	Fecal Co	liform cou	nts /100ml	% disinfection				
Day	Inf	Lamp 1	Lamp 2	Lamp 3	Day	Lamp 1	Lamp 2	Lamp 3
2	19000	800	600	750	2	95.79	96.84	96.05
2	21000	1230	1100	1038	2	94.14	94.76	95.06
6	25000	1450	1433	1013	6	94.20	94.27	95.95
1	23000	900	900	700	1	96.09	96.09	96.96
2	13000	-	1100	895	2	-	91.54	93.12
4	26000	2000	2300	1200	4	92.31	91.15	95.38
1	54000	2000	1550	1500	1	96.30	97.13	97.22
2	43000	15000	14500	13600	2	65.12	66.28	68.37

	Influent	Day	Lamp 1	% Dis.	Lamp 3	% Dis.
15-Jun	116600	1	2433	97.90	2333	98.00
17-Jun	44300	3	157	-	163	-
21-Jun	7300	4	233	96.80		-
23-Jun	19000	6	577	-	313	-
25-Jun	50300	8	8533	83.00	6550	87.00

Foulant Analysis

	Fouling								
	days	AI	Ca	Fe	Mg	Mn	Na	Р	protein
		ppm	mg/L						
14-Jun	4	0.649	0.941	1.367	0.144	0.002	0.111	0.589	0.095
5-Jul	5	2.093	1.299	7.415	0.133	0.006	0.011	4.287	0.088
25-Jun	8	0.587	0.302	1.530	0.011	0.001	0.021	0.149	0.108
	Fouling						_		
	days	AI	Ca	Fe	Mg	Mn	Na	P	protein
		mg/L							
10-Jun	2	0.360	1.023	1.012	0.082	0.006	0.096	0.208	0.238
14-Jun	4	2.119	2.077	6.616	0.404	0.013	0.112	4.309	0.165
5-Jul	5	2.345	1.476	8.651	0.222	0.005	0.000	2.077	0.118
	Fouling	i							
	days	AI	Ca	Fe	Mg	Mn	Na	Ρ	protein
		ppm	mg/L						
14-Jun	6	0.612	0.605	1.225	0.108	0.001	0.131	0.437	0.114
25-Jun	8	0.114	0.144	0.153	0.030	0.002	0.110	0.000	0.109

Appendix C Phosphorus official results

Appendix D Beauharnois results

Incipient Fouling Data sheet

Sampling Date:June $6^{th} - 16^{th}$ [FeCl_3] =0 mg/l

Fecal Coliform (counts / 100ml):

Date	Time (hrs)	Influent	Lamp 1	Lamp 2	Lamp 3	E1	E2	E3
6/6/2000	0	156670	0	15	13	100	99.99	99.99
6/6/2000	1	-	30	0-300	10	-	-	-
6/6/2000	3	200000	6	6	3	99.99	99.99	99.99
6/6/2000	4	160000	6	13	16	100.00	99.99	99.99
6/7/2000	23	67000	3	0	3	100.00	100.00	100.00
6/7/2000	24	110000	0	0	0	100.00	100.00	100.00
14-Jun	0	126600	22	8	6	99.97	99.99	99.99
14-Jun	1	216600	40	14	2	99.94	99.98	100.00
14-Jun	2	183300	4	6	8	99.99	99.99	99.99
15-Jun	25	-	-	-	-	-	-	-
16-Jun	45	40000	10	10	17	99.99	99.99	99.97
19-Jun	113	90000	8	0	5	99.99	100.00	99.99

	[13.16	5.846	5.785
Std dev.	58398.58	1	2	6

<u>UVT (%T):</u>

	Time	Influe	Lamp	Lamp	
Date	(hrs)	nt	1	2	Lamp 3
6/6/2000	0		73.5	72.6	73.5
6/6/2000	1				
6/6/2000	3	ł			
6/6/2000	4	}			
6/7/2000	23	75	75.4	75.1	75
6/7/2000	24		74.8		74.5
14-Jun	0	78.3	78.5	78	75.5
14-Jun	1	76	76.5	76.5	76.6
14-Jun	2	76	76	76	75.5
15-Jun	25	60	60	58.6	59
16-Jun	45	80	80	80	80.3

Std dev. 7.20 6.62 7.75 6.79

Turbidity (NTU):

	Time	Influe	Lamp	Lamp)
Date	(hrs)	nt	1	2	Lamp 3
6/6/2000	0	3	3.5	4	2.6
6/6/2000	1			4.5	5.2
6/6/2000	3		7	8.5	7
6/6/2000	4		5.3	6	6.5
6/7/2000	23	4.8	4.2	4.8	3.4
6/7/2000	24	3.3	3.5	3.5	3.5
14-Jun	0	3.7	4	4	4
14-Jun	1	3.4	3	3.2	3.7
14-Jun	2	3	3	3	3.5
15-Jun	25	10	10	10	10
16-Jun	45	1.5	1.8	2.3	2
	Std dev.	1.27	1.52	1.80	1.90

Incipient Fouling Data sheet

Sampling Date:June 28-29th of June[FeCl_3] =2 mg/l

Fecal Coliform (counts / 100ml):

Date	Time (hrs)	Influent	Lamp 1	Lamp 2	Lamp 3	E1	E2	E3
6/28/2000	0	95000	1000	700	900	98.95	99.26	99.05
6/28/2000	1	130000	100	130	200	99.89	99.86	99.79
6/28/2000	2	100000	100	130	90	99.90	99.87	99.91
6/29/2000	24	40000	0	30	25	100.00	99.93	99.94

Ctd dov	27500	AGO 205 2 AOA
j Slu dev.	3/300	409 303.3 404

<u>UVT (%T):</u>

		Influe	Lamp	Lamp	
Date	Time (hrs)	nt	1	2	Lamp 3
6/28/2000	0	65	67	72	66
6/28/2000	1	68	65	64	66
6/28/2000	2	66	64	64	65
6/29/2000	24	65	_63	62	62

Std dev.	1.41	1.71	4.43	1.89

<u>Turbidity</u> (NTU):

		Influe	Lamp		
Date	Time (hrs)	nt	1	2	Lamp 3
6/28/2000	0	3	2.8	2.4	2.3
6/28/2000	1	3.2	3	3.1	2.7
6/28/2000	2	3.3	3.8	2.6	2.5
6/29/2000	24	2.3	2.4	0.4	2.7
	Std dev.	0.45	0.59	1.19	0.19

Incipient Fouling Data sheet

Sampling Date:July 5th-12th[FeCl_3] =2 mg/l

Fecal Coliform (counts / 100ml):

	· · · · · · · · · · · · · · · · · · ·		Lamp	Lamp	Lamp			
Date	Time (hrs)	Influent	1	2	3	E1	E2	E3
7/5/2000	0.1	33000	290	270	450	99.12	99.18	98.67
7/5/2000	1	40000	63	64	130	99.81	99.81	99.61
7/5/2000	2.5	120000	85	85	260	99.93	99.93	9 9.78
7/5/2000	4	80000	60	65	200	99.93	99. 92	99.75
7/6/2000	25	50000	50	30	90	99. 94	99.96	99.89
7/7/2000	40	45000	120	70	180	99.85	99.91	99.78
7/10/2000	115	40000	125	65	150	99.84	99.92	99.81
7/12/2000	165							

	31180.7			119.3
Std dev.	3	83.25	79.89	6

<u>UVT (%T):</u>

1	Time	Influe	Lamp	Lamp	
Date	(hrs)	nt	<u>1</u>	2	Lamp 3
7/5/2000	0.1	65	65.6	65	64.6
7/5/2000	1	63.5	63	64	63
7/5/2000	2.5	62	60.5	61	61
7/5/2000	4	61	60.3	60	60
7/6/2000	25	63.2	63.3	63.3	63
7/7/2000	40	64	63.6	64	64
7/10/2000	115	62.5	61.3	62	62
7/12/2000	165	60	58.5	59	59

Std	dev.	1.62	2.27	2.13	1.95

Turbidity (NTU):

Date	Time (hrs)	Influent	Lamp 1	Lamp 2	Lamp 3
7/5/2000	0.1	3	3	2.5	2.5
7/5/2000	1	3.2	3	3	3
7/5/2000	2.5	3.2	3.2	3.25	3.2
7/5/2000	4	3.1	3.1	3.1	3.1
7/6/2000	25	2.5	2.5	2.5	2.6
7/7/2000	40	3.2	2.8	3	3
7/10/2000	115	3	3	3	3.3
7/12/2000	165	4.2	4	4.2	4
	Std dev.	0.47	0.43	0.53	0.46

Results of August 15th -29th

[FeCl3] = 4-6 mg/l

Fouling days	Fecal Collform	而且此论而		UVT	Turbidity	COD mgO2/L				
				And here a more derived		DATE	Influent	Off	UV lamp	Visible
				For the line	13. A. A.		Average	Average	Average	Average
0		表已可以同世		÷a 57	,z 6,5	8/15/2000	29.534	10.720	10.980	19.183
1				· · · · · · · · · · · · · · · · · · ·	6.3	··· 8/23/2000 ···	14.886	5.903	6.033	5.512
3	新加加的 11-1			44	6.6	, 8/18/2000 x	9.809		10.590	
6	Per la			50	S. 7. 5	8/28/2000	26.084	4.340	1.346	1.997
max	120000.00	500.00	99.83	57.00	7.00	36766.00	29.53	10.72	10.98	19.18
min	30000.00	90.00	99.12	44.00) 6.30	36753.00	9.81	4.34	1.35	2.00
avg	60000.00	260.00	99.47	50.50	<u> </u>	36759.00	20.08	6.99	7.24	8.90

				Suspen	nded solids		
Fouling days 0 1 1 3 3 3	Iron	S	F	Ratio			
	Concentration	ł	UV		UV	1	UV
0	3.9	15.3	18	10.67	10	69.7	55.6
1	4.9	16.7	23.3	6.7	8	40.1	34.3
1	5.7	32.7	23.3	20.7	18.7	63.3	80.3
3	4.3			{			
3	5.4	22.7	30	4.7	5.3	20.7	17.7
4	4.6	19	11.3	9	1	47.4	8.8
6	6.3	32	16.67	10.7	10.7	33.4	64.2
7	6.1	5.3	1.3	2.7	6.7	50.9	-
max	6.3	32.7	30.0	20.7	18.7	69.7	80.3
min	3.9	5.3	1.3	2.7	1.0	20.7	8.8
avg	5.2	20.5	17.7	9.3	8.6	46.5	43.5

COD Results (2000)

	COD mgO ₂ /L							
DATE	Influent	Lamp 1	Lamp 2	Lamp 3				
	Average	Average	Average	Average				
6/14/2000	18.7	0.0	0.0	0.00				
6/16/2000	64.9	45.4	40.9	68.4				
6/17/2000	0.0	4.7	10.8	11.5				
6/21/2000	17.6	18.7	19.5	8.2				
6/21/2000	2.3	5.9	8.2	35.5				
6/21/2000	12.5	29.3	29.9	15.6				
6/28/2000	11.7	0.8	7.4	10.5				
6/28/2000	1.9	15.6	10.1	2.7				
6/28/2000	0.8	10.9	0.0	2.3				
7/5/2000	34.0	13.7	22.6	25.4				
7/5/2000	23.0	27.3	16.8	17.6				
7/5/2000	8.8	2.7	14.4	18.4				
7/6/2000	18.0	13.7	12.5	14.4				
Average	16.5	14.5	14.1	17.7				
Min	0.0	0.0	0.0	0.0				
Max	64.9	45.5	40.9	68.4				

COD mgO ₂ /L						
DATE	Influent	Influent Off UV lamp				
	Average	Average	Average	Average		
8/15/2000	29.5	10.7	10.9	19.1		
8/18/2000	9.8		10.5			
8/21/2000	22.6	16.1	22.3	26.0		
8/22/2000	18.0	0.0	0.0	0.0		
8/23/2000	14.8	5.9	6.0	5.5		
8/25/2000	0.8	0.0	0.0	0.0		
8/28/2000	26.0	4.3	1.3	1.9		
8/29/2000	5.7	0.0	4.6	14.1		
Average	15.9	5.3	6.9	9.5		
Min	0.8	0.0	0.0	0.0		
Max	29.5	16.1	22.	26.0		

Appendix E

Wastewater quality parameters correlations Beauharnois

UVT and Turbidity







