IRON REMOVAL AT REMEDIAL SITES: NEW REGULATIONS DRIVE THE SEARCH FOR MARKET-APPROPRIATE METHODS

Brad A. Horn[1§], Matthew D. Millias[2], Steven P. Stucker[3]
[1]PE, President, Redux Technology, 280 Callegari Drive, West Haven, CT 06516, [2]PE, Principal, CDM Smith, 1 General Motors Drive, Syracuse, NY 13206, [3]CPG,Senior Environmental Engineer, National Grid, 300 Erie Boulevard West, Syracuse, NY, 13202

ABSTRACT

Over the past few years, new regulations limiting the discharge of iron to surface waters have become increasingly common. These regulations are particularly important in the groundwater remediation market, where unique in-situ water chemistry results in elevated levels of iron, as well as other minerals, at a majority of sites with contaminated groundwater. Removal of iron from water tends to be challenging, for a number of reasons, and commercially available methods have not been widely cost-effective in the remedial field.

The assessment criteria for iron removal technology at remedial sites is market-specific, and thus distinct from other markets where iron removal has been practiced. When assessing iron removal in remedial applications, key market-specific technical issues include: 1) space constraints, 2) operator requirements, 3) sludge generation, post-treatment and disposal costs, 4) head loss and operating pressure requirement, and 5) generation of backwash water requiring post-treatment units.

Since 2008, Redux has invested in an R&D effort to identify iron removal methods most appropriate for remedial applications, and to use our chemical knowledge to enhance promising techniques, or develop new ones. This work has involved lab and bench-top studies, as well as pilot work in the field. Techniques studied include bag filtration, settlers, cross-flow micro-filters, sand, greensand and multi-media filters, and other specialty media filters, as well as all of these methods in conjunction with pretreatment chemicals.

This paper provides a review of iron removal technology alternatives, and an assessment of their applicability to remedial applications. It presents bench-top and pilot scale work at many sites where these alternatives have been considered. Finally, it presents new technology, now in commercial application, which shows

[§]Corresponding Author: Brad A. Horn, PE, President, Redux Technology, 280 Callegari Drive, West Haven, CT 06516, Telephone: 203-933-9071, email: bhorn@reduxtech.com

significant promise in being the most appropriate technique, yet discovered by the authors, for this niche treatment application.

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1. INTRODUCTION

Fouling of groundwater remediation systems by iron deposition and iron-related microbial biofilms is a common problem. Dissolved iron, in groundwater contaminated with anthropogenic organic materials, is often elevated, compared to nearby uncontaminated groundwater sources. This is due to a variety of factors, including pH and ORP (oxidation-reduction-potential) depression as a result of natural biodegradation of contaminants, making iron more soluble. During subsequent remedial operations, whether in-situ or ex-situ, iron can become insoluble, creating voluminous inorganic and microbial iron-related solids, interfering with treatment operations.

Control of iron deposition using blended water treatment chemicals has become more common, as appropriate products have become available. However, in recent years, new regulations limiting the discharge of iron to surface waters have become increasingly common. These regulations essentially force site operators to consider alternatives for removal of iron as a pretreatment step. Removal of iron from groundwater is common in drinking water treatment, but methods used in this market are designed for iron levels that would be considered very modest in contaminated groundwater applications. In the drinking water market, total iron levels of one or two milligrams per liter is considered high, while contaminated groundwater often bears ten to a hundred milligrams per liter, and in some cases, more. Alternatively, higher levels of iron are treated in some industrial wastewater applications, but technology used in these applications tends to consume a very large footprint, and is maintenance intensive.

In an effort to identify an iron removal method that is appropriate for the groundwater remediation market, the authors completed various literature reviews, bench-top analyses and pilot studies. This work involved assessing existing iron removal alternatives used in a variety applications in terms of their applicability to the groundwater remediation market. Thus, an important part of this work involved identifying design criteria, performance requirements, and operations requirements specific to the remedial field. Relevant considerations include: 1) space constraints, 2) operator requirements, 3) sludge generation, post-treatment and disposal costs, 4) head loss and operating pressure requirement, and 5) generation of backwash water requiring post-treatment units.

Techniques studied include bag filtration, settlers, cross-flow micro-filters, sand, greensand and multi-media filters, and other specialty media filters, as well as all of these methods in conjunction with pretreatment chemicals. Ultimately, pilot work, and subsequent commercialization work was performed on several sites using an upflow media filter. This involved site-specific chemical pretreatment and subsequent solids separation. The solids separation unit is commonly called a bead filter, and employs a floating plastic media, which can be treated to enhance removal.

This paper will provide a brief review of iron removal technology alternatives, with a focus on their applicability to remedial applications. Following this review, it presents data and analysis from three separate pilot studies performed using the upflow media filter.

2. MATERIALS AND PROCEDURE

2.1. Technology Review

The corresponding author's special focus on iron fouling since 1990 has enabled visits to many sites where iron deposition is a problem. This process has enabled observations of the various methods of iron removal. A brief description of the common techniques observed in use at remedial sites, along with obvious advantages and disadvantages (relevant to remedial applications), follows.

2.1.1. Chemical Treatment

All common methods of removing iron from water involve chemical treatment. The goal of chemical treatment is to convert any dissolved, or ferrous, iron to insoluble ferric iron salts, generally iron oxides and hydroxides. While treatment for each application is unique, this is typically achieved by chemically increasing ORP (using oxidizers), pH (using alkaline reagents), or both. Common oxidizers include oxygen, sodium hypochlorite, hydrogen peroxide and chlorine dioxide. Common alkaline reagents include sodium hydroxide, hydrated lime, magnesium hydroxide and sodium carbonate.

Iron solids initially formed are notoriously small (a fraction of a micron) and difficult to separate from the bulk water. Thus, in many cases, precipitant aids are used to build particle size and stability. Common precipitant aids include coagulants and flocculants. Coagulants serve to destabilize the charges that surround particles in suspension, and which repel one another to form stable particulate suspensions. Historically, coagulants have been simple low molecular weight inorganics, such as alum, but newer, high performance organic coagulants are now common. Flocculants are polymers which essentially sorb (attach to) and "bridge" between particles formed by the coagulation process, creating larger agglomerated solids which settle faster and are easier to filter.

Typically, some method of solids separation is used in conjunction with chemical treatment. There are a variety of iron "oxy-hydroxides", all of which have slightly different physical characteristics. Chemical treatment can be "designed", using various reagents, to produce those salts with characteristics which favor their removal by any particular solids separation technique. Similarly, precipitant aids can be selected from a great variety available to create the most desirable particulates. While common reagents work in many different applications, the optimum chemical treatment program tends to be specific to a particular groundwater.

Depending upon reagents used, chemical treatment can involve the handling of hazardous materials. Typically reagent addition requires several stages, involving multiple tanks, pumps and mixers, which can be space consumptive, and operations intensive. In remedial applications, footprint and operations efforts can be reduced using static mixers, pipe flocculators and proper controls.

2.1.2. Gravity Settling

Gravity settling, or sedimentation, is the most common method of separating iron solids from the bulk water. It is typically done in a specialized tank referred to as a settler, with various design features that minimize tank footprint, but allow quiescent flow for settling to occur. If space is not limited, large, open rectangular tanks are the simplest type of settler. Accommodations must be made to remove sludge from the bottom of the settler tank. In many cases, sludge removed from a settler may be further treated to increase solids concetration, and thus reduce disposal costs. This may be done by gravity thickening, in a tall tank, or by various methods of active dewatering such as filter press, belt press, rotary filter or centrifuge.

The primary disadvantage of gravity settling in remedial applications is the large space requirement. In addition, sludge withdrawn for a gravity settler has a low solids content. This increases disposal costs (assuming no sludge post-treatment occurs), particularly if sludge is considered hazardous.

2.1.3. Bag Filtration

Bag filtration is not commonly used in other markets for removal of iron, and is not, by any means, an ideal method of removing iron solids. However, bag filters are widely used at remedial sites in general, usually to prevent solids from fouling granular activated carbon (GAC) beds. GAC units and their associated bag filters often are used in a treatment train after an air stripper or oil-water separator. In cases where recovered groundwater is iron-bearing, bag filters often end up removing some or all of this iron. In these situations, filter bags tend to bind with iron solids rapidly (iron sludge is notoriously impermeable), requiring frequent bag replacements. In bags that are not replaced often enough, it has been widely observed that the resulting high pressure drop through the filter bags forces iron to "bleed" through the filter, fouling downstream units.

Though bag filters are not be a very efficient way to remove iron solids, their pervasiveness at remedial sites has lead the authors (and others) to experiment with employing chemical pretreatment to improve their viability for iron removal. The authors have found that chemical pretreatment can improve the usefulness of bag filters under certain situations. For example, precipitant aids (specifically, "dewatering" polymers) can reduce bag filter change-outs, while retaining iron solids effectively. In spite of this, bag filtration does not compare favorably with other methods for iron removal at remedial sites.

2.1.4. Sand and Multimedia Filtration

Sand, and multimedia, filtration typically involves packed beds operating in down-flow mode under gravity or pressure. When employed with chemical pretreatment, they are effective at removing iron to low mg/l levels. Iron sludge accumulates on the sand or media until pressure drop due to sludge cake reaches some unacceptable level, at which point, the beds are backwashed and fluidized to remove iron solids. Backwash is typically collected and treated further, or it may be discharged to sanitary sewers, in some cases.

Packed bed filters can be effective for iron removal, and consume less space than gravity settlers, of the same flow rating. While this smaller footprint is a big advantage in remedial applications, the requirement for clean backwash water, and the resultant generation of dilute iron-bearing backwash water creates an operations challenge. Many remedial sites do not have clean water available, unless treated water is stored, which requires large footprint storage tanks. Treatment of backwash water also creates an operations challenge, requiring storage tanks and further solids treatment of the dilute and voluminous backwash.

2.1.5. Greensand

Greensand is a naturally occurring, fine-grained material, which can be treated to remove iron in a packed bed arrangement. It is also referred to as manganese greensand since the treatment process to enable iron removal involves the application of potassium permanganate oxidizing agent. This oxidizer sorbs on the sand, oxidizing the iron to create insoluble ferric salts which then sorb to, and are physically filtered by, the media. The greensand media requires regular backwashing to remove iron solids, as well as subsequent regeneration by addition of potassium permanganate.

Greensand has been widely used in drinking water treatment for many years, and has been proven to be very effective in that application. Its use in remedial applications has been limited for several reasons: The requirement for clean backwash water, and for treatment of dilute iron bearing backwash water create challenges for many remedial sites. In addition the economics of greensand regeneration, and backwash, at high iron loadings limit its window of appropriate application to what are considered low levels of iron (less than 5 mg/l) in the remedial market.

2.1.6. Specialty Media Filters

There are various specialty and proprietary media marketed specifically for iron removal. Most of these media are applied in packed beds and may require regeneration or backwash. One of the most common of these is BIRM (a tradename), widely used for iron removal in small drinking water applications. It requires a certain level of dissolved oxygen in influent water in order to insure iron oxidizes to the ferric state: many groundwater remediation system influent waters do not contain appreciable amounts of dissolved oxygen, requiring the introduction of air or oxidizers to employ BIRM. Siemens (formerly US Filter) markets a specialty iron removal media which has been shown to be effective for removal of low levels of iron (several mg/l), but the authors do not have direct experience with this material.

2.1.7. Crossflow Microfilters

Several manufacturers offer filtration systems which avoid (or minimize) the common problem of membrane fouling by creating high shear rates parallel to the membrane surface. These are termed by some as cross-flow microfilters, and have been shown to be effective for removal of iron at the higher concentrations common on remedial sites with iron fouling. This method is necessarily used in conjunction with chemical pretreatment, and involves recirculation of solids bearing water at a high ratio compared to the system throughput. While this technology is effective for iron removal at remedial sites, and its space requirement is less than most alternatives, the capital and operating costs are higher than alternatives.

2.2. Pilot Work

2.2.1. Technology Identification

Shortcomings of existing methods guided the author's search for alternative techniques that might be adapted for remedial work. These criteria include compact footprint, minimal generation of backwash, maximum concentration of solids, and a general minimum of process sub-units. The use of packed bed separators offers small footprint, but complications in separating solids from bed media. Literature searches suggested that packed beds employing floating beads might offer the benefit of small footprint, without the challenge of managing backwash: Certain configurations allow the use of air, to stir and separate media from solids entrained in the bed. Bead filters have been widely used in the US in aquaculture water treatment, and to a more limited extent, overseas for municipal wastewater treatment.

After literature review and various discussions with academic investigators involved in bead filter work, the Authors toured beadfilter installations to better assess feasibility in remedial applications. Based upon this work, the Authors determined that adaptation to remedial work would require 1) different bead geometry than that used in prior applications, 2) pretreatment chemistry to create particles large enough to make bead size practical, targeting particle size of 50 microns, and 3) agglomerated particulates with a stability to endure the shear forces expected in packed bed flow. With these challenges in mind several pilot study candidate sites were identified. Three pilot studies were run to assess feasibility of beadfilters for iron removal at two separate sites.

2.2.2. Pilot Study Site A

2.2.2.1. Site A – Phase I

This site is a very large superfund site in Upstate New York. Two sequential pilot studies were run here, with the results of the first informing a redesign of the pilot apparatus for the second. The subject water is recovered from a single well which is one of two maintaining an inward gradient through a sheet-pile wall surrounding a fill area. The driving force for iron removal is expected discharge to surface waters, with an iron discharge limit of 0.5 mg/l. Various site specific conditions make the pilot work more challenging than would be expected at almost any remedial site: Total iron levels ranged from 350 mg/l to 400 mg/l with a good portion of this being dissolved. Iron removal is complicated by the addition of a blended deposit control agent to the well, to prevent very rapid fouling of the recovery pump and transmission piping. Several bench-top studies were completed to determine if a chemical pretreatment program would be able to "break" the sequestered iron complexes and degrade the dispersants to allow creation of required iron particulates. These studies resulted in the development of a treatment protocol which included 1) Fenton's chemistry to degrade deposit control components, 2) addition of a blended alkaline reagent to raise pH slightly from approximately 3.5 to about 6.5, 3) a high performance organic coagulant, and 4) a cationic flocculating polymer.

Using this pretreatment scheme, two sequential pilot studies were run. The first utilized a simple bead bed arrangement depicted below. The schematic on

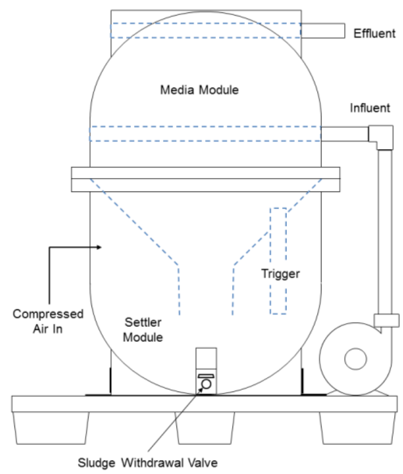

Figure 1. Photographic and schematic representations of initial pilot configuration

the right is useful for understanding the basic operating concepts of this method. Various subsequent designs utilize the same basic components in different arrangements according to the performance criteria desired. The media module contains, and is partially filled with, the floating bead media. The feed pump introduces pretreated water into the bottom of the bead bed and water flows upward through the bed to the effluent pipe, essentially slotted well screen. Solids are entrained in the media by flocculation, sorption and physical exclusion. Meanwhile, air is introduced slowly into the air chamber (in this case toroidally shaped, wrapping around the internal "sludge funnel"), lowering the water level in this chamber until it reaches the patented trigger, at which point air is rapidly transferred into the bead bed above, stirring it aggressively. The sludge/water mixture flows downward through the "sludge funnel" to fill the settler module completely (including the air chamber). Influent flow is uninterrupted during this bed purge, which has an adjustable cycle, based upon the rate of compressed air introduced into the air chamber. After a bed purge, influent water refills the top of the unit, and another operating cycle begins, during which sludge settles in the bottom of the unit, and can be periodically withdrawn.

Using this initial pilot arrangement, a three month pilot effort was completed. During the first two months of study the pilot unit was run during individual two or three day site visits, occurring every week or two. This phase of work involved extensive debugging of the pretreatment chemistry, recovery and transfer pumping systems, and various bead bed operating parameters. During each site visit, system operations were observed, monitored and documented in the site field book. With each visit, changes were made in configuration and operations as needed.

During the last month of field work the system operated continuously and periodic iron removal data was collected, along with extensive additional data reflecting status of operations.

2.2.2.2.Site A – Phase II

The second phase of pilot work at this site was conducted to incorporate lessons learned in the first phase. Figure 2 below depicts the entire process set up for this phase of pilot work, including the pretreatment chemical feed, mixing and

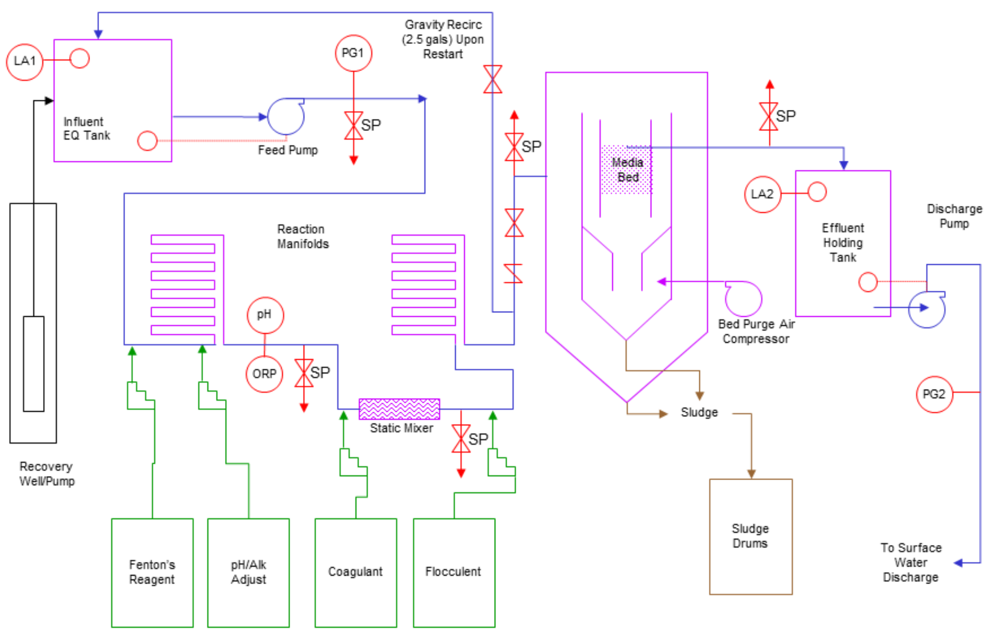

Figure 2. Entire process diagram for Site A – Phase II pilot work

reaction manifolds, and a recirculation loop to recycle "stale" water upon each restart (added in the middle of this second phase). This second phase of pilot work lasted approximately seven months. During the first two months, data was collected using the same apparatus used in the first phase, in order to enable design of an improved apparatus. Subsequently, this new bead filter, along with necessary controls to allow continuous operation, was installed, debugged and operated for about five months.

There were two major improvements in the new pilot design: First, the use of a gear pump (rather than a centrifugal pump used in the first phase) enabled very accurate control of groundwater flow rate. This, in conjunction with fixed-feed chemical reagent pumps, eliminated the need for complicated chemical feed controls, while still enabling accurate dosing. Second, the new design incorporates a "roughing chamber" for removing the largest solids, those which can be easily settled, rather than running all solids generated through the bead bed. This reduces unnecessary solids loading to the bead bed, allowing it to flocculate and remove smaller solids, or those that float. Figure 3 shows the top of the new pilot bead filter, showing the outer "roughing chamber", with an inner weir over which water flows, to subsequently rise through the bead bed, in the center section.

Figure 3. Top interior of redesigned Site A pilot bead filter

During the five months of operation, a dedicated site operator visited the site for several hours three times per week to collect data regarding system performance and operation parameters.

2.2.3. Pilot Study Site B

This two week pilot study was completed at a landfill site located on a large air force base in Georgia. Work from the above pilot studies helped with redesign of a pilot apparatus most appropriate for the expected water characteristics. Iron removal was required, in this case, in order to effectively operate an ozonation unit without fouling it, and to prevent iron from increasing oxidant demand. Influent water contained approximately 60 mg/l of total iron, virtually all dissolved. Preliminary bench-top studies derived an unusual treatment protocol: it involved addition of an inorganic poly-aluminum chloride coagulant followed by pH increase using caustic soda, followed by addition of high molecular weight anionic flocculating polymer. It is rare that coagulant is added before solids generation (in this case by increasing pH), but in this case it made a very significant difference in particle size development.

The apparatus used for solids separation is depicted in Figure 4. While this design is similar to that used in Phase I work at Site A described earlier, it utilizes a bead size which is about one-third that used in the Site A work.

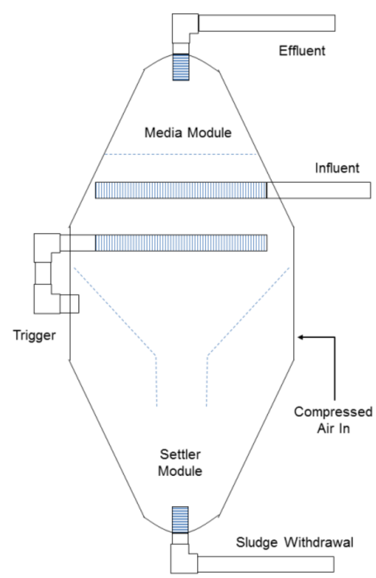

Figure 4. Photographic and schematic representation of Site B apparatus. Note in photo, bead filter is on right, along with recirculation and discharge tanks respectively to its left.

Site work for the pilot study involved an initial set-up and debug stage for several days, prior to the initiation of ozonation work. Iron removal and ozonation together operated for about two weeks, at a flow rate of one gpm. Iron removal and other operations parameters associated with filter operations were recorded regularly.

3. DATA AND ANALYSIS

This section provides data relating to iron removal for all three pilot efforts described previously. Though much additional data was collected relating to operations parameters, all of which ultimately relate to improving iron removal, the data presented here includes only measurements specific to iron concentrations in influent and effluent, and certain sludge characteristics. This data is most critical to determining whether or not the subject iron removal technique merits further study. Other data which was collected in all three studies, but which is not directly germane to the iron removal performance and sludge production, includes bed pressure drop, iron particle size and stability measurements, settling tests, jar tests of samples taken at various points in the treatment train, reagent dosing tests, bed purging tests, and “upset” testing (where the bed was challenged, intentionally or unintentionally with poor pretreatment conditions), among other data. While this latter data constitutes the great majority of the testing data collected, and it enabled regular improvement in iron removal, detailed reporting and explanation of it is beyond the scope of this paper.

With the above caveat in mind, Table 1 is presented below, giving iron measurements taken during Phase I testing at Site A, along with relevant brief comments. In assessing this data set it is important to understand that the influent contained blended deposit control agents (containing sequesterants and dispersants) which complicated proper pretreatment. During the initial test work of this phase, the groundwater fed to the filter was pumped directly from the well using the down-hole electric submersible well pump, through the pretreatment manifold and filter. The recovery pump had been originally set up to cycle, feeding a relatively high flow rate periodically, with a ratio of on-time to off-time of about ten to one. This short period, high flow situation made accurate chemical dosing difficult, as compared to longer runs at lower flows. For various reasons, changing the pumping cycle was not feasible, so at mid study, an equalization tank was installed to allow a more desirable cycling pattern, greatly improving chemical dosing, and subsequent iron removal.

By the end of this phase, enough sludge was generated to estimate sludge production and solids content. Sludge directly from the filter ranged around 3.5% solids content. Gravity thickening produced a sludge solids content in the range of 4.5%, with a final production rate of this material estimated to be about 1.6% of the groundwater volume treated.

Table 1. Iron Removal Data for Site A – Phase I Pilot Work

Date & Sample ID	Iron (mg/l)		Removal	Comment
	Total	Dissolved	(total iron)	
11/3/2010				
Influent	430	310	95.3%	High flow, short duration runs, 6-8 gpm
Effluent	20	4		
11/16/2010				
Influent	390	300	99.0%	flow stabilized at 1-2 gpm
Effluent	4	2		
11/30/2010				
Influent	400	320	99.5%	Sludge Analysis: 4.5% solids w/w
Effluent	2	1		

Phase II of the Site A Pilot Work incorporated many lessons learned from the Phase I work. Treatment system upgrades included the incorporation of the equalization tank and positive displacement gear pump to enable long runs at low flow, with consistent reagent dosing. Once design data was collected and the redesigned pilot unit was installed, along with aforementioned changes, iron removal to low mg/l levels was consistently achieved, with complete oxidation of iron.

During Phase II pilot work, several process improvements incrementally increased average iron removal and improved sludge quality. These improvements included 1) the incorporation of a timer-controlled recirculation loop to send “stale” water back to the influent equalization tank for several minutes upon each cycle restart, and 2) better control of sludge level in the “roughing chamber” (aka influent atrium).

After installation of the redesigned pilot unit, influent water chemistry changed somewhat (as it does periodically at Site A), which resulted in a sudden increase in floating iron solids. This upset highlighted one advantage of the bead bed approach over settlers depending upon gravity separation, as overall removal remained relatively stable. Subsequently, influent chemistry shifted back to original conditions (as indicated by the success of original optimized reagent feeds) and overall iron removal reached its best yet.

Table 2 presents iron removal data and relevant comments collected during Phase II work at Site A.

Table 2. Iron Data for Site A – Phase

Date & Sample ID	**Iron (mg/l)**		**Comment**
	Total	**Dissolved**	
3/16/2011			install gear pump
3/14/2011			
Influent	380	280	optimize chemistry
Effluent		<1	
3/28/2011			
Influent	390	300	1st full tote runs
Effluent		ND	
4/17/2011			
Influent	400	330	using settler only, removal of all particulate iron greater than 5 microns, <8 ppm colloidal
Effluent	5-8	ND	
5/22/2011			Install new bead filter, restart
5/30-6/6/11			system upset, floating sludge
6/13/2011	390		
Effluent	3-4	ND	particulates 5-25 microns
7/1-9/13/11			
Effluent	1-2	ND	particulates 5-25 microns

While regular improvements in treatment occurred throughout both phases of the Site A work, the best effluent consistently produced contains one or two milligrams per liter of total iron.

Pilot work at Site B occurred subsequent to both phases of pilot work at Site A, and incorporated adjustments in an attempt to improve iron removal. Media used in the Site B work was smaller and geometrically different than that used in the Site A work. Iron removal data collected from the Site B work consistently gave an influent total iron level of sixty mg/l: All pilot work at this site was completed on a fixed volume of influent water contained in a single tank, resulting in very consistent influent quality. Once chemical pretreatment dosing was adjusted, iron filter effluent contained less than one milligram per liter of total iron in all cases. Influent and effluent iron measurements were collected at least twice per day over the two week period of operation.

While iron removal was very consistent during most of the Site B operations (allowing a very successful ozonation study to occur), particular attention was paid to bead bed operations parameters affecting iron removal. These included hydraulic loading, the advantages and effect of recycling through the iron filter, and loss of iron removal efficiency immediately after purging the bead bed of iron solids. From this work, various plans for improvement were borne, most significantly relating to loss of iron removal efficiency after bed purge: Iron removal suffers after bed purge, for some period of flow (twenty to thirty minutes after bed purge). This effect can be minimized by recycling effluent back to the treatment system headworks, or by adding chemical "retreatment" to a recycle loop at the filter itself. These alterations are to be studied in upcoming pilot work.

4. CONCLUSION

The pilot work described in this paper proved that the use of bead beds for iron removal shows promise. Data collected helped greatly improve removal throughout the course of these three studies, as changes were implemented in response to data collected. The Site B pilot work results enabled the site owner to make the decision to proceed to full scale process development, and this system has now been in operation for over a year. Certain process problems identified in this pilot work have been addressed and others are still under study.

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