BIOLOGICAL PRETREATMENT FOR MEMBRANE SYSTEMS

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Investigators at the Center for Biofilm Engineering proposed a technology to increase the biological stability of water to be further processed by reverse osmosis membranes. It was proposed to reduce membrane fouling by pretreating feed water using biological reactors to remove organic nutrients that support formation of fouling biofilms. The goal of the work was to determine if biological pretreatment can reduce fouling in membrane systems. Specific objectives included: (1) determine if biological pretreatment using biologically active carbon and iron oxide coated sand as support media will reduce downstream fouling; (2) determine if chlorination as an oxidation step for feed water will influence downstream fouling; (3) determine if filtration of feed water will reduce downstream fouling; and (4) develop evaluation methods of these objectives by developing and testing assays. The results of this work supported the acceptance of all objectives except the use of chlorination as an oxidation step for the organic amendment. The biological treatment coupled with filtration resulted in the best reduction in downstream fouling as measured by the assays employed. The reduction shown in biofilm cell numbers supported the assumptions made in the cost analysis.

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Glossary

BAC - biological activated carbon

BACCL - biological activated carbon with chlorine

BACPF - biological activated carbon with filtration

BACPFCL - biological activated carbon with filtration and chlorine

DAPI - 4',6-diamidino-2-phenylindole, dihydrochloride

EBCT - empty bed contact time

IOCSPF - iron - oxide coated sand with filtration

IOCS - iron-oxide coated sand

CTRL - control

CTRLCL - control with chlorine

CTRLPF - control with filtration

CTRLPFCL - control with chlorine and filtration

IOCSCL - iron-oxide coated sand with chlorine

IOCSPFCL - iron-oxide coated sand with filtration and chlorine

POTW - publicly owned treatment works

SEM - scanning electron microscope

TOC - total organic carbon

1.0 Executive Summary

In light of the problems inherent in suppressing biological activity and fouling of upstream processes and membranes, investigators at the Center for Biofilm Engineering proposed a technology to increase the biological stability of water to be further processed by reverse osmosis membranes. We proposed to reduce membrane fouling by pretreating feed water using biological reactors to remove the organic nutrients that support formation of fouling biofilms. The technology will improve membrane productivity by complementing existing processes in a pretreatment train while reducing dependence on extensive use of disinfectants. The approach is ecologically sound because it uses biological processes to control subsequent biological activity; the emphasis is on controlling biofouling in an engineered system rather than having it occur in undesirable locations. The process is based on the experiences of the drinking water industry where biological filtration is being used to reduce concentrations of natural organic matter, decrease disinfectant demand, and reduce downstream fouling of the distribution system.

The goal of the work proposed was to determine if biological pretreatment can reduce fouling in membrane systems. Specific objectives included:

- Determine if biological pretreatment using biologically active carbon (BAC) and iron oxide coated sand (IOCS) as support media will reduce downstream fouling.
- Determine if chlorination as an oxidation step for feed water will influence downstream fouling.
- Determine if filtration of feed water will reduce downstream fouling.
- Develop evaluation methods for objectives 1-3 by developing and testing the following assays.

    i. Membrane flux measurements
    ii. Membrane fouling layer cell counts
    iii. Membrane fouling layer thickness measured with SEM
    iv. Membrane fouling layer thickness measured with DAPI stained thin sections.
    v. TOC removal from the system.
    vi. Bead assay biofilm cell counts.

The results of this work supported the acceptance of all the objectives except the use of chlorination as an oxidation step for the organic amendment. The biological treatment process coupled with filtration resulted in the best reduction in downstream fouling as measured by the assays employed. The reduction shown in biofilm cell numbers supported the assumptions made in the cost analysis.

The cost analysis indicated that in large plants (30,000 to 36,000,000 GPD)savings that range from $ 0.33 to $ 0.68/1000 gallons could be realized. On a percentage basis, the

annual cost savings ranged from 6% to 8.8% with a mean savings of 8.7%. It is anticipated that small facilities should realize an even greater cost savings benefit.

In summary, the utilization of a biological treatment process in a RO system design should be considered as one of the unit processes that will reduce operation and maintenance costs and provide for a more efficiently run water treatment plant.

2.0 Background and Introduction

The material that fouls reverse osmosis membranes is diverse, and is composed of inorganic particles (precipitated metal oxides, colloids, etc.), natural organic matter, and bacterial/fungal/algal/protozoan cells (DuPont, 1994; Al-Ahmad and Aleem, 1993). The rate and extent of fouling is a strong function of the quality of water applied to the membranes. It has traditionally been held that fouling material is the result of simple concentration and retention of constituents from the bulk. Another mechanism of fouling is the proliferation of organisms in biofilms on the membranes. In fact, a combination of these effects most likely is responsible for the adverse influence on membrane production (Mallevialle et al., 1996; Chapman-Wilbert, 1997).

2.1 Current Methods of Fouling Control

The control of reverse osmosis (RO) membrane fouling has typically been attempted via (1) physical and/or chemical treatment of the water to remove or stabilize particulates and/or ions, (2) periodic direct cleaning of membranes, (3) development of membranes with reduced fouling potential or modification of the surface chemistry with chemical addition to reduce fouling and (4) continuous upstream application of biocides. In the first case, considerable effort has been expended by the industry to identify processes that decrease the load of particulates onto the membranes. As a consequence, there are a wide variety of "pretreatment" options found at locations using reverse osmosis including dual or single media filtration, softening and/or ion exchange, granular activated carbon filters, pH adjustment, etc. (DuPont, 1994). Our observation has been that there is no one general process train; the treatment options are dependent on water quality and the preferences of the consulting company responsible for installation. It is also possible for membranes to be cleaned directly with agents compatible with the membrane chemistry. Since many membranes are sensitive to oxidizing disinfectants (chlorine, for example) the choice of chemicals must be made judiciously. This aspect of membrane maintenance can be very frustrating to the process operators. In many cases, vendors prescribe a specific chemical and application regime without providing the operators with any information on their rationale, presumably because the cleaning process is proprietary. The third point above is the search for membranes with reduced fouling potential. This could be achieved by either creating/modifying polymers that foul at a decreased rate or producing membranes that can be more easily disinfected or cleaned (Chapman-Wilbert, 1997). Our experience suggests that neither of these approaches will eliminate fouling. During long term operation, the surface chemistry of membranes is modified by molecules and microorganisms found in the water, therefore masking the designed or altered surface chemistry features. There have been improvements in surface modification techniques that reduce fouling by the addition of surfactants and work is continuing in this area (Ridgway, 1997). The fourth point brings out the attempts by the membrane industry to control fouling through the use of biocides. This approach will produce a situation where the fouling may be controlled but not eliminated. There is ample evidence to indicate that biofilm communities are considerably less susceptible to

disinfection than suspended cells and that long-term disinfection will not completely control fouling of surfaces. Continuous upstream disinfection, which is commonly practiced, also has limitations. Typically, oxidizing disinfectants are present until immediately before the membrane in an attempt to suppress biological activity in the pretreatment train. The end result is that the environmental pressures that have repressed biofilm growth are removed at the membrane, which creates the opportunity for microbial proliferation at precisely the point where it is least desired. It is also possible that continuous biocide addition will select for resistant strains of bacteria that can then foul the membrane.

2.2 Premise of the Research: Biological Pretreatment

It is obvious that none of the above approaches for mitigating membrane fouling will always work by itself. It is most probable that a combination of methods will be required to increase membrane productivity. In light of the problems inherent in suppressing biological activity and fouling of upstream processes and membranes, investigators at the Center for Biofilm Engineering proposed a technology to increase the biological stability of water to be further processed by reverse osmosis membranes. We proposed to reduce membrane fouling by pretreating feed water using biological reactors to remove the organic nutrients that support formation of fouling biofilms. The technology will improve membrane productivity by complementing existing processes in a pretreatment train while reducing dependence on extensive use of disinfectants. The approach is ecologically sound because it uses biological processes to control subsequent biological activity; the emphasis is on controlling biofouling in an engineered system rather than having it occur in undesirable locations. The process is based on the experiences of the drinking water industry where biological filtration is being used to reduce concentrations of natural organic matter, decrease disinfectant demand, and reduce downstream fouling of the distribution system.

2.3 Supporting Information: Drinking Water

There is a reasonable amount of information available on biological treatment of drinking water. The process was first implemented in France and other western European countries nearly 20 years ago (Sontheimer et al., 1978, 1979a,b). In the most traditional form, separate granular activated carbon (GAC) filters are located downstream from conventional treatment. In conventional treatment, particle removal is optimized through coagulation, flocculation, sedimentation, and filtration. The biological filters are then optimized for microbial utilization of a portion of the natural organic matter remaining in the water. Biological filters are operated with exhausted carbon, that is, the chemisorptive capacity of the GAC has been exceeded. The surfaces of the filter media act as a support for microbial attachment and growth, resulting in a biofilm adapted to using the organic matter found in that particular water. Total organic carbon removals in these filters range from 5 to 75% (Bouwer and Crowe, 1988).

One of the first observations in the full-scale use of biological filtration was that the type of filter media had a substantial influence on the net removal of organic carbon from the water. For instance, although rapid sand filters do have the capacity to biologically remove carbon (Eberhardt et al., 1977; Sontheimer et al., 1978; Borbiogot et al., 1982; van der Kooij and Hijnen, 1985) it has been found that GAC has superior performance (LeChevallier et al., 1992; comparison between DeWaters and DiGiano, 1987 and Hozalski et al., 1995). This is presumed to be the result of a higher amount of biomass that attaches to GAC vs anthracite (Niquette et al., 1998). LeChevallier et al. (1992) demonstrated that there were more bacteria per unit surface area on GAC than sand, and that the TOC removal rates were 51% vs 26%. Another advantage of GAC over other media is that the attached microbial population is less prone to shock from changes in water quality, down time, or accidental application of disinfectant (Bablon et al., 1988; Krasner et al., 1993). This knowledge of the performance of filter media will be strongly considered in our experimental design.

In most cases, improved TOC removal in biological filters can be gained by increasing the empty bed contact time (EBCT). The EBCT is the residence time of the fluid in the filter calculated as though the entire volume occupied by the filter media is occupied by water. Because of the very large volumes of water that a drinking water plant treats, a small reduction in EBCT results in substantial savings in filter volume. Experimental EBCTs in biological filters have varied from two to 30 minutes. Sontheimer and Hubele (1987) demonstrated an increase in dissolved organic carbon removal from 27 to 41% when the EBCT increased from 5 to 20 minutes. LeChevallier et al. (1992) reported a 29% removal of TOC with a 5 minute EBCT and a 51.5% reduction at 20 min. Prevost et al. (1990) suggest that a 20 min. EBCT is required for 90% removal of biodegradable organic carbon. However, there are instances where increased EBCT is not beneficial, which is probably a result of the biodegradability of the organic matter present in the water (Hozalski et al., 1995). Certainly the EBCT required for biological removal of TOC will be temperature dependent. This has been demonstrated at a full-scale biological filtration plant, where 12 minutes of EBCT was required at 0.5^{o} C for the same percent removal obtained in 6 minutes at 10 - 12^{o} C (Niquette et al., 1998). Due to design constraints and stringent regulatory requirements for filtration to meet particle removal standards, most full scale biological filters operate with short (5 min) EBCT In instances where the technology can be applied for pretreatment of water for membrane processing, the EBCT can be optimized for organic removal.

A common treatment step before biological treatment is ozonation. Ozone may be applied to reduce taste and odor compounds, remove color, provide primary disinfection for protozoan cysts, or to reduce disinfection demand/disinfection byproducts by oxidizing some of the organic matter. Water that has been preozonated often has elevated levels of lower molecular weight organic compounds; these compounds have been associated with increased biofilm development downstream (van der Kooij et al., 1989; Price, 1994; LeChevallier et al., 1996). Goel et al. (1995) reported that the fraction of recalcitrant natural organic matter in water made available for microbial growth was

increased after ozonation, but the numerical value varied from site to site. This has also been substantiated by van der Kooij et al. (1982), Werner and Hambsch (1986), Servais et al. (1987) and Speitel et al. (1993). Because biofilms can form either in controlled treatment processes (biological filters) or in uncontrolled deleterious locations (distribution systems), the drinking water industry strongly considers biological filtration after ozonation, regardless of the original intent of ozone application.

Interestingly, chlorination sometimes has the same effect on biodegradability of natural organic matter as ozonation. LeChevallier et al. (1992) showed that chlorination increased the biodegradable fraction of organic matter in water. In a survey of plants in the Netherlands, this increase was as high as 1.75 fold (Cooperative Report, 1988). Chlorine has been found to alter the structure of humic substances in water, which may render it more degradable (Hanna et al., 1991). Paul (1996) reports not only an increase in degradable organic carbon after chlorination, but an increase in culturable cell counts as well. These observations show how the current practice of applying oxidizing disinfectants until immediately before a reverse osmosis membrane actually "pushes" biological activity and associated fouling onto the membrane. Certainly the use of preoxidation to improve the ability of biological pretreatment to remove organic matter from water is a key component of our work plan.

Even though the emphasis in drinking water has been on the use of GAC, there is other evidence to suggest that the iron oxide coated media may be a better choice for removal of natural organic matter that directly causes fouling (Jacangelo et al., 1995; Owen et al., 1995) and/or subsequent biofouling at the expense of the immobilized organics on reverse osmosis membranes. Chang and Benjamin (1996) demonstrated that addition of iron oxide particles to individual ultrafiltration hollow fibers greatly reduced fouling by organic matter. Chang and Benjamin found the organic matter was preferentially bound to the iron oxide particles which could then be removed by backwashing. Iron oxides have a large potential for the sorption of natural organic matter (McCarthy et al., 1993; Parfitt et al., 1977; Zhou et al., 1994). Under abiotic conditions, humic material is irreversibly held on the surface of iron oxides (Gu et al., 1994; Gu et al., 1996). In fact, this property has been used to develop a technique for the removal of NOM from water by coating sand particles used in slow sand filter beds with iron oxides (McMeen and Benjamin, 1997). Circumstantial evidence indicates that the bound organic matter is potentially available for biofilm bacteria when these same investigators mentioned that the iron oxide-coated olivine used in their filtration studies continued to remove NOM for a 16 month time period; they suggested that the adsorption sites were being "bioregenerated."

A classification of the constituents of NOM from surface waters (Malcolm, 1991; Kaplan, 1993) indicates that approximately 50-75% is humic substances. These concentrations are lower in groundwater. Humic substances are generally considered to be poorly biodegradable, because of their large molecular size. However, Namkung and Rittmann (1987) have shown that humic substances are in fact biodegradable. More

recently, Volk et al.(1997) have shown that biofilm bacteria are capable of using humic materials. Because humic substances in the bulk water are poorly degraded, it is probable that bioavailability of the humic substances is enhanced when bound to surfaces. The humic molecules then undergo a conformational change and expose the utilizable functional groups (Beckett, 1990). Immobilization on the surface is also likely to permit the cells to use exoenzymes to attack the bonds between the bound amino acids, sugars, etc. and the backbone of the humic molecule (Wetzel et al., 1991; Jones and Lock, 1991; Munster, 1991). There is strong evidence to suggest that the sorption of humic substances allows them to become available for biofilm use. When an assessment of the growth rates of biofilm bacteria grown on humic materials was made in experiments in our laboratories, it was found that the growth rate was independent of the added humic carbon concentration (zero order kinetics). We believe this may be caused by the large amount of humic materials bound to the biofilm (8.3 - 11 $\mu g\ C/cm^2$); supplementation of additional humic material did not influence the growth rate. There was also visual evidence that the humic material was sorbed, as these biofilms were a characteristic brown color (unpublished data, our laboratories).

This mechanism has profound implications for the biological treatment of water as well as providing a potential explanation for observations of biofouling on membranes. Membrane autopsy data from a polyamide nanofiltration membrane showed that 73% of fouling deposit was organics, of which 34% was humic material. This same deposit also contained 16% iron oxide. In the same report, foulant from a polyamide RO membrane contained 62% organics and 6% iron oxides (Dudley and Fazel, 1997). Similarly, Butt et al. (1997) reported that RO desalination membrane foulant was primarily biomass and iron contributed appreciably to foulant mass.

It is probable that the choice of filter media (GAC vs. iron coated sand) and whether or not to preoxidize prior to biological filtration will depend on the nature of the organic matter in the source water. Although NOM has a high affinity for iron oxides, high molecular weight fractions of NOM are also preferentially removed by GAC binding (Owen et al., 1995). Preoxidation can result in increased biodegradable fractions, but on occasion biofiltration following ozonation only reduces the biodegradable fraction to preozonation levels (LeChevallier et al., 1992). Since iron oxide coated media tend to remove reduced iron from solution, there may be further advantages for these media when the source water is high in dissolved iron. Therefore, we investigated several potential combinations of filter media and the presence/absence of prechlorination under controlled laboratory conditions to bracket ideal design parameters for biological pretreatment.

The premise of biological treatment as described above is to (1) immobilize organic matter that would otherwise accumulate on reverse osmosis membranes and (2) use indigenous organisms to metabolize the sorbed organics as well as other bulk phase carbon compounds. The end result will be the reproduction of bacterial cells within the filter. These organisms will be released from the filter and could then accumulate on the

reverse osmosis membrane. Decay processes would then permit the dead microbial cells to become substrate for surviving bacteria. Although the overall rate of fouling from this process should be significantly less than if no biological treatment was in place, there is still concern from operators that bacterial accumulation is deleterious to membrane performance. For example, Collentro and Collentro (1997) suggest that GAC has a low efficiency for organic carbon removal and that GAC filter effluent contains elevated nutrients and high organism counts that increase membrane fouling (1997). This is probably because these GAC filters were not optimized for either chemisorption or biological activity. This philosophy has lead to reports that GAC should be used for RO feedwater pretreatment only if no other options exist (Kucera, 1997). This prejudice may be unfounded if (1) the biological filter is operated properly and (2) adequate downstream removal techniques for minimizing bacterial and carbon fine particle release are in place.

To minimize the chance for transport of bacteria produced in the biological filter and released sand/GAC particles, we proposed to use a low maintenance particle filter. Prior to performing this work, we collected particle size distribution data on particles released from a biological filter and decided that the best option will likely be a microfiltration unit. We installed a microfilter downstream from a laboratory biological filter, and it operated extremely well. The microfilter was effective for removing the vast majority of the bacteria and shed filter fines. It should be noted, however, that microfiltration alone would not provide the same advantages for reducing reverse osmosis membrane fouling as microfiltration after biological pretreatment. Although MF has been shown to remove significant fractions of iron and reduce colloidal fouling of downstream nanofiltration membranes (Chellam et al., 1997), a portion of the humic substances found in typical surface waters will pass through MF membranes (Jacangelo et al., 1989). Laine et al. (1990, 1989) demonstrated that pretreatment is necessary to reduce the organic matter that may pass through UF membranes, and that activated carbon pretreatment shows the most promise.

Our approach was to incorporate our experience and available information to develop and test biological treatment processes to be integrated in traditional pretreatment trains with the purpose of prolonging membrane performance. We believed that the deliberate encouragement of biological growth within a component of the pretreatment train that is under process control will substantially reduce the undesirable growth on reverse osmosis membranes. Additionally, the biological filters may significantly reduce natural organic matter/humic fouling. Greibe and Flemming, 1998, have produced work that supports these last two points. The design incorporated both a biological treatment step and a method to remove organisms and fines shed from the biological filter. A favorable outcome, as outlined in the section on economic comparisons, would be a cost effective treatment method to reduce fouling and chemical use.

2.4 Experimental Goals and Objectives

As set forth in the previous sections, we proposed to reduce membrane fouling by pretreating feed water using biological reactors to remove the organic nutrients that support formation of fouling biofilms. To this end we state the following overall goal.

GOAL: Determine if biological pretreatment can reduce fouling in membrane systems.

Several objectives were set forth to test the feasibility of utilizing biological pretreatment as a membrane pretreatment step. These objectives were rephrased as hypotheses that were statistically tested with the experimental design.

- Determine if biological pretreatment using BAC and IOCS as support media will reduce downstream fouling.

- Determine if chlorination as an oxidation step for feed water will reduce downstream fouling.

- Determine if filtration of feed water will reduce downstream fouling.

- Develop evaluation methods for objectives 1-3 by developing and testing the following assays.

    i. Membrane flux measurements
    ii. Membrane fouling layer cell counts
    iii. Membrane fouling layer thickness measured with SEM
    iv. Membrane fouling layer thickness measured with DAPI stained thin sections.
    v. TOC removal from the system.
    vi. Bead assay biofilm cell counts.

3.0 Conclusions and Recommendations

The goal of this study was to determine if biological pretreatment could reduce the downstream fouling potential of feed water in membrane systems. The following sections present the results and recommendations.

3.1 Results

- Biological pretreatment will reduce downstream fouling of membranes. The biological pretreatment step reduced the number of biofilm cells in both the membrane and bead assays. In addition, there was a reduction in TOC.

- Chlorination, as an oxidation step, had no impact on downstream fouling potential of the feed water. The results indicated little or no difference as a result of chlorination.

- Microfiltration, after biological treatment, further reduces downstream fouling of membranes. Postfiltration reduced the number of biofilm cells in both the membrane and bead assays. In addition, there was a reduction in TOC.

- All assays were needed to assess the fouling potential of a particular treatment. The combination of assays provided a much more comprehensive body of evidence that fouling potential had been reduced than any single assay.

3.2 Recommendations

1. Use of biological treatment coupled with microfiltration has the most significant impact on reducing the downstream fouling potential of membrane feed water.

2. A variety of assays is necessary to determine the downstream fouling potential of feed water. The assays developed during this study that consistently revealed a reduction of fouling were the membrane biofilm enumeration, bead assay biofilm enumeration, and TOC removal.

3. These results are considered to be conservative, since the organic carbon amendment of humic and fulvic acids is believed to be the most recalcitrant form of dissolve organic material typically found in feed water. In a real situation, the improvements should be even greater.

4. Chlorine may have a more definitive impact if used in greater concentrations. It is possible that long contact times and carrying a residual into the system may promote more changes in the nature of the organic carbon than was observed in this study.

5. The next step is to perform pilot scale studies using reverse osmosis as the final membrane step. The reverse osmosis membranes can then serve as the membrane assay step.

4.0 Materials and Methods

4.1 Experimental Setup and Methods

The tasks set forth for this project proposed to look at biological filtration using biologically active carbon and iron-oxide coated sand, examine the impact of chlorination of the feed water on downstream fouling, and refine and test a membrane fouling assay and a post biological treatment microfiltration step. Combining the factors of chlorination, different packing media, and post filtration resulted in an experimental setup that consisted of 12 separate treatments. See Figure 1 for a schematic of the syste m.

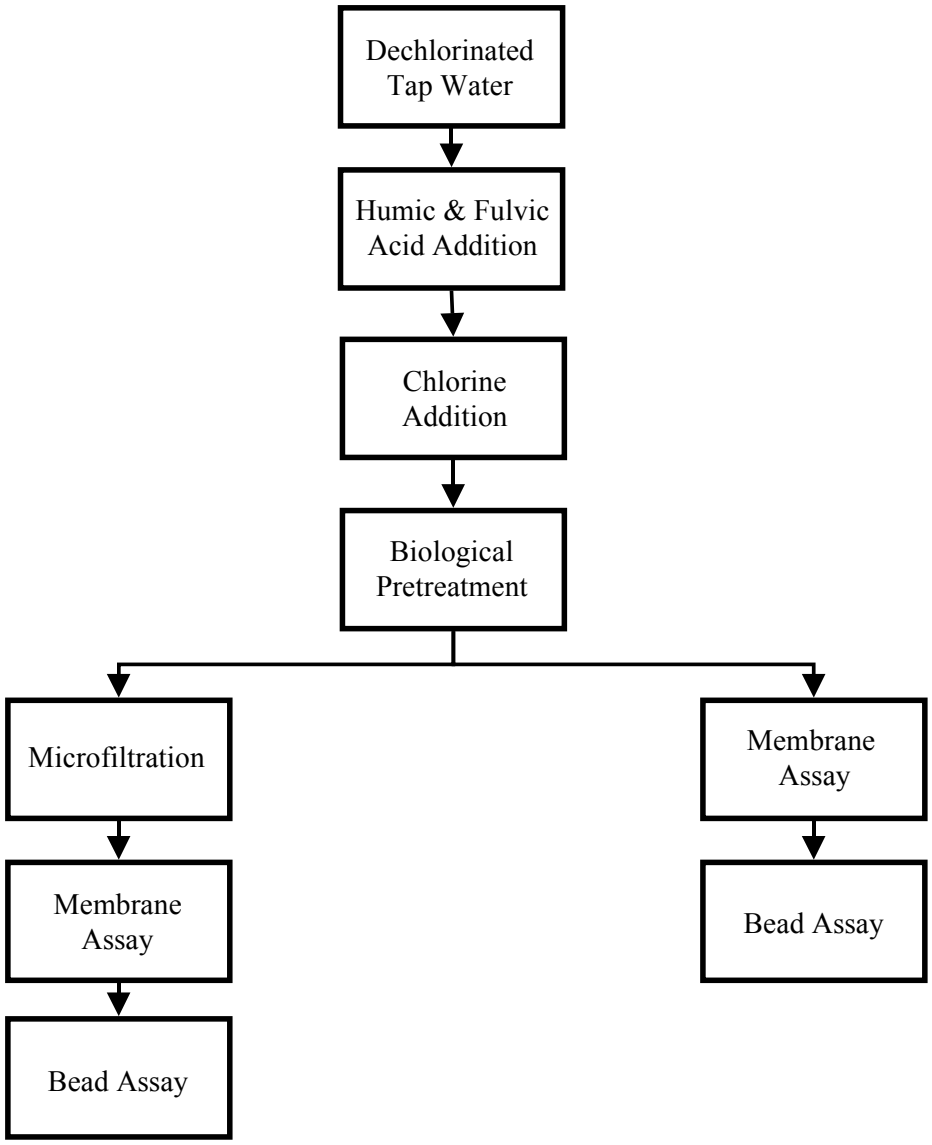

Figure 1. Treatment train schematic.

The experimental setup was run continuously for 10 months. During the first six months, experimental protocols and operational experience were developed. The results presented in this report were obtained from three experimental runs performed after this 6 month break in period. Each run was performed over a period of five days. During each five day run, TOC and flux measurements were taken and at the end of each run, the destructive assays were performed.

The test apparatus for this study was constructed using type 316 stainless steel (ASTM A-213/ASME SA-213 average wall; ASTM A-269,ASTM A-511). The choice of material was based upon the need to limit the amount of leachable organic carbon and still provide sufficient strength for the pressures involved with the operation of the columns. All connecting tubing was 6.35 mm (0.25 in) and 12.7 mm (0.5 in) outside diameter of type 316 stainless steel. The inside diameter of the 6.35 mm (0.25 in) tubing was 4.93 mm (0.194 in) and the inside diameter of the 12.7 mm (0.5 in) tubing was 10.92 mm (0.430 in).

Two types of packing media for the biological pretreatment columns were used; iron-oxide coated sand and biologically activated carbon. These two packing types, along with a control which had no packing, represented three treatments. The effluent of each of these treatments was split and a 0.22 micron (8.66e-6 in) post filtration step was added on one of the streams from each treatment. To assess the effects of chlorination on the system the entire system was duplicated. One side was prechlorinated and the other side was not. Thus, twelve treatments were constructed for sampling and evaluation.

The setup also consisted of several assays that were placed in the effluent streams. One assay was a 4.93 mm (0.194 in) inside diameter tube of 316 stainless steel, 25.4 mm (1 in) in length that was packed with 0.5 mm (0.0197 in) glass spheres. The other assay was an in-line membrane holder that allowed for flux through a 47 mm (1.85 in) diameter membrane swatch and flow across the membrane while the system was under pressure.

Total system pressure was maintained at 207 kPa (30 psi) during the entire time the system was running (>12 months). During this time, the laboratory temperature was maintained at a constant 72° F. The flow rate through each treatment was maintained individually with a stainless steel needle valve. Flows were maintained between 1-2 ml/min.

4.2 Biological Pretreatment Columns

Using information from the drinking water industry, a minimum of 20 minutes empty bed contact time was desired. Studies have shown that a 20 minute empty bed contact time is sufficient to remove the majority of easily assimilable organic compounds commonly found in surface waters.

To assure that this situation was met, the columns were designed to have an empty bed contact time that was approximately 30 minutes.

The columns were constructed with 316 stainless steel tubing and was ordered from Marmon/Keystone Corporation. 25.4 mm (1 in) tubing was specified with a wall thickness of 0.889 mm (0.035 in). The inside diameter was 23.62 mm (0.930 in). The column length was 228.6 mm (9in) and the resulting volume for the columns was approximately 100,170 mm^3 (6.114 in^3).

The average flow rate through the columns was 1.7 ml/min which resulted in an empty bed contact time of 29.46 minutes.

The columns were packed with either iron-oxide coated sand or biologically active carbon. Support screening was 100 mesh 316 stainless steel. All end caps were Swagelock 316 stainless steel compression tube fittings.

Flow configuration was in the upflow mode with the columns mounted vertically.

4.3 Column Packing Media

The support packing for the biological pretreatment columns were iron-oxide coated sand and biologically active carbon.

4.3.1 BAC

The biologically active carbon was PICA brand activated carbon that had been operating in a biological filtration process at a drinking water plant in Laval, Quebec, Canada. The plant was treating surface water and the filtration process was operated to promote biological growth on the activated carbon. The activated carbon had been on line for several years and was never regenerated. The sorptive capacity of the media is due mainly to the microbial activity on the medium.

To acclimate the microbial community to the organic carbon present in Bozeman tap water, the packing material was placed online with Bozeman tap water for more than one year. The material was then used to pack the columns that were used in this study.

4.3.2 IOCS

Iron-oxide coated sand was produced using the method outlined in 'NOM Adsorption Onto Iron-Oxide-Coated Sand', AWWARF, 1993. This coating was developed to be a stable sand coating that could be regenerated in a full scale operation and provide good NOM (natural organic matter) adsorption.

Support sand was industrial quartz sand manufactured by Unmin corporation, Emmett, Idaho. The sand was screened to between 30-40 mesh. The effective size of the sand in this range is 0.45 mm. This sand was soaked in 50% sulfuric acid solution for 24 hours, then rinsed with deionized water, and dried at 110° C for 20 hours in accordance with the method of Chang and Benjamin, 1996.

An iron oxide solution was prepared with 1 gram of ferric chloride per ml of deionized water. A 10 M solution of NaOH was added to this solution until the OH:Fe M ratio was 2.5. This material was then dried in a pan at 110° C for 14 hours. The top crust of salts was scraped off and the iron oxide sludge underneath was used to coat the sand. The iron oxide sludge and the sand were mixed in a ratio of 0.1 g iron oxide sludge per 1 g of sand. The sand was dried at 110° C for 20 hours. The sand was rinsed and the process repeated two more times to obtain a good coating.

Once the sand was coated, it was used to pack two columns; one for the non-chlorinated treatment and one for the chlorinated treatment. All column dimensions and materials were the same as for the biological activated carbon columns described previously.

Using a porosity of 0.35, the iron-oxide coated sand column had a pore volume of approximately 35 ml. This yields a residence time of about 10.3 minutes.

4.4 Bead Assay

The bead assay was put in-line with the effluent stream. Its purpose was to provide an assessment of the biofilm growth potential after each of the treatments.

The apparatus consisted of a short 316 stainless steel tube. The assay column was a 4.93 mm (0.194 in) inside diameter tube of 316 stainless steel, 25.4 mm (1 in) in length that was packed with 0.5 mm (0.0197 in) glass spheres.

At the end of each experimental run (5 days), the assay column was removed and the glass beads extracted into 10 ml of dilution water. This mixture of beads, biofilm, and water was vortexed for 30 seconds and the appropriate dilution series performed to enumerate the bacteria.

4.5 Cell Enumeration

The bacteria were counted using direct count epifluorescent microscopy on a Nikon 8100 at 1000x. The cells were filtered onto a 0.22 micron filter (25 mm black polycarbonate - Poretics) and stained using 4',6-diamidino-2-phenylindole, dihydrochloride (DAPI) obtained from Molecular Probes (catalog number D-1306). 1 ml of the stain was placed on the membrane for 1 minute at a concentration of 10 mg/L.

Ten fields were randomly selected and counted. The appropriate magnification and dilution were then applied to the numbers to arrive at the total cell count per ml. This number was then applied to the appropriate surface area (either total bead area or membrane scrape area) to arrive at the biofilm cells per area number.

4.6 Membrane Assay

The membrane assay holder was constructed out of two plates of aluminum block. The blocks measured 25.4 mm x 76.2 mm x 76.2 mm (1 in x 3 in x 3 in). Two blocks are sandwiched together with four bolts. A membrane, silicon gasket, polycarbonate screen, and filter paper support for the membrane, are placed between the blocks. One block allows the effluent to enter and leave while crossing the membrane swatch. The other block allows for flux through the membrane by providing an outlet for the permeate to atmospheric pressure.

The silicon gasket forms the flow channel for the cross flow of the effluent. The exact dimensions of the channel were always determined at the end of the experiment by electronically measuring the fouled area of the membrane swatch. With the design flow of the system, the Reynold's number is at least 450 and is probably more due to the reduced cross-sectional area produced from compression of the gasket.

The polycarbonate screen was placed over the holes of the block that provides an outlet for the flux through the membrane. Next, an ashless filter paper (47 mm) was placed over the screen to protect the membrane. A polycarbonate, 0.22 micron, 47 mm membrane (Poretics) was placed on top of the filter paper. To seal and create a flow channel, a silicon gasket with the flow area cut out was placed over the screen, filter paper, membrane stack and the other membrane assay block was placed on top. The four bolts were then torqued down to 10 N.m (7.3756 ft.lbs) in a cross hole pattern.

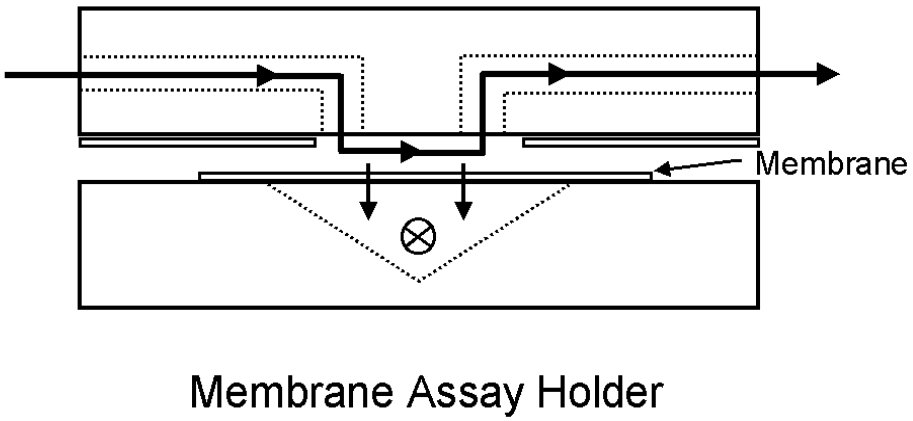

Figure 2.
Membrane assay holder.

During the experimental run, the flux through the membrane was measured periodically to determine the flux reduction as the fouling layer was developing. Flux measurement was determined by collecting the flux effluent from the membrane assay for a known time and then measuring the collected volume. After the final day of the experimental run, the assays were dismantled and the membrane removed for analysis. Immediately, the membrane was photographed for digital analysis of the fouling area for use in the flux calculations. In addition, the fouling layer color could be recorded.

4.7 SEM fouling layer assessment

Scanning electron microscopy was utilized to assess the membrane fouling layer thickness. The scanning electron microscope used in this work was a JEOL Model 6100/NORAN SEM equipped with an Oxford cryostage and cryoprep chamber. The cryostage allows an environmental sample to be frozen quickly to preserve the original structure and then placed under a vacuum for manipulation prior to sputter coating for the SEM. The sample is never removed from vacuum so that it remains intact. The SEM was purchased with a grant from the Murdock Charitable Trust and MSU and is located in the Image and Chemical Analysis Laboratory at MSU-Bozeman.

To determine the thickness of the fouling layer, a small portion of the fouled layer was cut from the membrane. This sample was then mounted vertically in a special stage. The stage is mounted on a post that allows insertion into a flash freezer that utilizes liquid nitrogen and vacuum to quickly freeze the sample. The sample is then introduced into the cryostage where the pressure is reduced to 1e-4 torr and the stage is further cooled to around -190° C. At this point tools within the microscope sample preparation area are used to cut across the membrane and expose a cross section of the membrane and the fouling layer.

After the fouling layer cross section is exposed, the sample can be sputter coated with gold. The coating was 2 nm before moving the sample into the SEM chamber.

Once in the SEM chamber, the pressure is further reduced to 1e-6 torr and the chamber is held at -195° C. Images of the cross section were taken so that an estimate of the fouling layer thickness could be made using the scale bar from the instrument. In addition, bacteria in the fouling layer could be seen. These bacteria were often small (< 1 micron) due to the oligotrophic conditions of the experimental system.

4.8 DAPI fouling layer assessment

Another section of the membrane fouling layer, similar in size to that used for the SEM analysis, was cut out and embedded in TISSUE-Tek™ OCT 4583 compound. This compound is an embedding medium for frozen tissue specimens. The specimen was frozen in a pool of OCT on a block of solid carbon dioxide. The specimen was then mounted on a stage in a Leica CM 1800 Cryostat. This machine enables 5 and 10 micron slices to be made of the cross section of the membrane and fouling layer. These slices were then transferred onto a microscope slide.

The slices were then stained with DAPI. The staining procedure consisted of immersion of the slide in 100 mg/L DAPI solution for 20 seconds and then drying before placing on the microscope for analysis.

The thickness of the fouling layer could then be estimated through several measurements of the fouling layer. To obtain this measurement, the microscope counting grid in the ocular eye piece was first calibrated using a micrometer slide. Then the thickness of the observed fouling layer could be estimated in several places. In addition, images were obtained for some samples and the thickness measurements were performed using ImageTool (http://www.ddsdx.uthscsa.edu/dig/itdesc.html).

4.9 Fouling layer cell counts

To estimate the number of cells per area that were in the fouling layer, a specific area of the polycarbonate membrane from the membrane assay was scraped with a scalpel, placed in 10 ml of dilution water and homogenized. The appropriate dilution was made

and a direct count performed. The direct counts were performed according to the procedure outlined in the glass bead assay section.

A photograph of the membrane was taken with a calibration scale. The photograph was then electronically scanned and the scraped area determined digitally using ImageTool. Imagetool is an image analysis tool that can be downloaded from http://www.ddsdx.uthscsa.edu/dig/itdesc.html.

4.10 TOC amendment

The organic carbon was humic/fulvic acids that were extracted from Elliot Silt Loam Soil. Elliot Silt Loam Soil is a standard soil used for laboratory work on humic/fulvic acids and is obtained from the International Humic Substances Society.

The humic/fulvic acids are extracted from the soil by mixing 75 g of soil in 750 ml of 6N NaOH solution for 24-48 hours. After the mixing is done, the slurry is centrifuged for 20 minutes at 10,000 rpm and 4° C. The supernatant is decanted and used as a stock solution to prepare a feed solution of humic/fulvic acids. This feed solution was fed into the influent of the experimental setup at a 500ppb -2ppm carbon level. Due to the variability of the influent water carbon content and the feed solution input control, the final level of organic carbon fed to the reactors was variable. The amount of organic carbon entering the columns was always measured as a control when TOC measurements were taken.

The feed water is Bozeman tap water that was dechlorinated by passing the water through a column packed with biological activated carbon. The BAC column used to dechlorinate the tap water also may have removed any readily assimilable organic carbon. Therefore, the carbon amendment represents the major carbon source introduced into the system. Since many water systems can be carbon limited, nitrogen and phosphorus were added to ensure that the limiting nutrient would be carbon. The maximum carbon input from the carbon amendment was 2 mg/L and the maximum amount from the tap water after dechlorination was estimated to be around 2 mg/L. Adding these two concentrations yielded an upper bound on the carbon concentration of 4 mg/L of carbon or twice the amount added in the amendment solution. Based upon this concentration and using the common 100:10:1 C:N:P ratio for microbial growth, the amount of nitrogen and phosphorus to be added to the amendment solution was calculated.

The nitrogen and phosphorus was provided in the form of ammonium nitrate and potassium phosphate. The stock carbon amendment solution contained 37.5 mmole of carbon. The necessary nitrogen and phosphorus concentrations were determined to be 7.5 mmole and 0.75 mmole, respectively. The nitrogen and phosphorus were then added to the stock amendment solution when it was prepared. This allowed the nitrogen and phosphorus to be mixed with the carbon source prior to injection into the test apparatus.

4.11 TOC analysis

To determine the amount of carbon that was removed by each treatment, a sample of each treatment effluent was collected daily during the experimental runs. These samples were then tested for carbon content.

To prevent outside carbon contamination, all glass ware was acid washed for 8 hours, triple rinsed with deionized water, triple rinsed with ultrapure water, and baked in an oven at 300° C.

The samples were tested for nonpurgeable organic carbon. First the samples were acidified to a pH below 2 with 2N HCl. The samples were then sparged with medical grade oxygen to remove the dissolved carbon dioxide. A Shimadzu TOC-5000A carbon analyzer with a high sensitivity platinum-palladium catalyst operating at 680° C was used to oxidize the nonpurgeable organic carbon to carbon dioxide and the carbon dioxide was detected with an infrared detector.

The Shimadzu was calibrated using NIST traceable potassium hydrogen phthalate standards obtained from Fisher Scientific.

4.12 Chlorination

A stock solution of 1 mg/L chlorine was injected into the chlorinated treatment train after addition and mixing of the TOC amendment. The chlorine was mixed with a static mixer and a 30 minute contact time with the TOC was allowed prior to application to any columns. The contact time was achieved by building a longer inlet tube to the treatment columns than the inlet tube for the non-chlorinated treatment columns. Chlorine measurements were taken using Iodometric method from AWWA Standard methods.

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5.0 Results

From objectives 1-3 that support the goal of determining if biological pretreatment can reduce fouling in membrane systems, 3 null hypotheses can be stated for statistical testing. They are:

1. The treatment means for the biological treatments are equal to their corresponding control (no biological treatment) treatments.
2. The treatment means for chlorinated treatments are equal to their corresponding non-chlorinated treatments.
3. The treatment means for the filtered treatments are equal to their corresponding non-filtered treatments.

All statistical analyses were performed using SPLUS version 5.1 release 1 for LINUX 2.0.31 : 1999, MathSoft, , Inc., Seattle, WA, http://www.mathsoft.com

A summary of the mean results of the assays is presented in Table 1. The treatments are biological activated carbon (BAC), iron-oxide coated sand (IOCS), and a control (CTRL). If the treatment was prechlorinated then CL is in the name and if the treatment was post-filtered then a PF appears in the name. For example, BACPFCL is the biological activated carbon treatment that was prechlorinated and post-filtered.

Table 1. Summary of results.

	Flux	Cells	SEM	DAPI	TOC	Bead
	mL/ (mm^2 min)	cells/ mm^2	μm	μm	mg/L	cells/ cm^2
BAC	0.00050	7.63	10.51	11.24	22.52	7.70
BACPF	0.0022	7.05	9.99	8.58	37.94	6.09
IOCSPF	0.0010	6.91	8.53	8.38	30.72	5.58
IOCS	0.00044	7.60	9.99	12.47	19.06	7.01
CTRL	0.00046	7.96	21.80	43.72	0.00	8.07
CTRLPF	0.00087	7.32	9.19	19.58	22.72	6.79
IOCSCL	0.00070	7.61	11.60	10.69	3.88	6.16
IOCSPFCL	0.0011	7.05	5.64	12.09	25.12	5.77
BACPFCL	0.0019	7.06	6.97	11.33	36.16	5.87
BACCL	0.00073	7.53	18.94	13.62	16.47	6.52
CTRLCL	0.00058	8.08	31.23	38.88	0.00	7.13
CTRLPFCL	0.0011	7.36	7.52	13.41	21.19	6.83

5.1 Flux

All flux results for each of the three experiments were fit to the model $y=ax^b$. Table 2 contains the fitted model parameters (a,b) for each treatment. Included in the table are the multiple R-squared values which indicate the amount of variability explained by the model and the F-statistic with its corresponding P-value.

Plots of the curve fits are in Figures 21-32. The inside confidence intervals are a point-wise standard error and the outside confidence intervals are a simultaneous confidence interval generated for the whole range using an F-distribution and a 95% confidence interval.

The simultaneous confidence intervals were used to generate the upper and lower flux values at 120 hours for use in the multiple comparison tests across experiments of the hypotheses. These values cover 95% of the predicted values and are considered conservative. The flux results were analyzed using a Monte Carlo based simulation multiple comparison with a control test. The simulation size was 12616 and the simultaneous confidence interval was 95%.

Since the simultaneous confidence intervals were used to generate the range of data found for the flux at 120 hours, the observed differences were hard to detect statistically. Hence, any differences that were detected are noteworthy.

The trends in the flux data revealed that post-filtration and pre-chlorination improved the flux. A combination of the two treatments showed the largest increase in flux.

Figure 9 shows that flux did not change with chlorination between treatments in a manner that was statistically significant. However, the chlorinated treatments did have a greater flux value for all treatments except the BACPF treatment. In the case of the BACPF treatment, the chlorine treatment had a lower flux.

Figure 15 shows there was a significant difference between post-filtration and no post-filtration for both BAC and BACCL biological treatments. In all the treatments, the general trend showed that the flux was greater for the treatments that were post-filtered.

For the biological treatment process, Figure 3 shows that only BAC was significantly better for flux improvement. In both post-filtered treatments, the BAC had a greater flux.

5.2 TOC removal

The percent TOC removal versus amount of TOC added to the system was plotted for each treatment. A linear model (y=mx+b) was then fit to the data to determine a percent removal for all experiments at a TOC addition rate of 800 ppb (Table 3). The choice of 800 ppb (measured humics addition and background tap water carbon) allowed the

experiments to be compared on a common basis. The resulting plots of the data and the curve fit are presented in Figures 33-42. As in the flux data, the inside confidence bands represent a single point-wise standard error and the outside confidence band represents a simultaneous confidence interval generated with an F-distribution and a confidence interval of 95%.

Using the same approach as the flux data, the simultaneous confidence intervals were used to generate upper and lower values for percent TOC removal at 800 ppb TOC added. These numbers were then used for the multiple comparison tests of the hypotheses. The TOC results were analyzed using a Monte Carlo based simulation multiple comparison with a control test. The simulation size was 12616 and the simultaneous confidence interval was 95%.

TOC removal was significantly different when chlorinated for the BAC, IOCS, IOCSPF treatments. In all of these cases, the removal was greater for the treatments where chlorine was not applied. Figure 13 shows the results of this comparison. It should be noted that the means for no chlorination were all greater than the chlorinated treatments.

TOC removal was significantly different for all treatments that were post-filtered from those that were not post-filtered. Figure 19 shows that the post-filtered treatments removed as much as 20% more TOC than the non-filtered treatments.

The ability of biological treatment to remove TOC was significant for all BAC treatments with respect to the control treatment. The IOCS treatment was significant for the non-chlorinated and post-filtered treatments. Figure 7 shows that the BAC and IOCS removal trends were consistent for all the treatments.

5.3 Membrane Cells

The membrane cell counts were analyzed using a Monte Carlo based simulation multiple comparison with a control test. The simulation size was 12616 and the simultaneous confidence interval was 95%.

The impact of chlorination on the concentration of cells on the membrane was not significant for any of the treatments. Figure 10 shows the trends in the data were mixed. Post-filtration was found to significantly change the cell counts for all treatments. Figure 16 shows that all non-post-filtered treatments had at least a one-half log greater cell count on the membrane.

The impact of the different biological treatments on the cell counts was found to be significant for all treatments. The biological treatments showed a reduction of at least 0.3 log. Figure 4 summarized these results.

5.4 Fouling layer thickness (SEM)

The fouling layer thickness as measured by the SEM method was analyzed using the Dunnett method of multiple comparison with a control and a 95% confidence interval.

Chlorination had an impact on the BAC and CTRL treatments only. Figure 11 shows that in the two cases where there was a statistical difference the fouling layer thickness was thicker for the chlorinated treatment. In all other treatments, the fouling layer thickness was less than 5 microns different between the chlorinated and non-chlorinated treatments.

When post-filtration was applied, the treatments were significantly different for all treatments except BAC and IOCS which were within 2 microns of each other. Figure 17 shows that the non-post-filtered treatments had greater fouling layer thicknesses than the post-filtered treatments. The difference in these thicknesses ranged from 5 microns to greater that 20 microns.

The impact of biological treatment on fouling layer thickness was significant for BAC, IOCS, BACCL, and IOCSCL. Figure 5 shows that the non-post-filtered treatments showed a thinner fouling layer (> 10 microns) than the control, while the post-filtered treatments were the same as the control.

5.5 Fouling layer thickness (DAPI)

The fouling layer thickness as measured by DAPI stained thin sections under a microscope provided another measure for thickness. These results were analyzed using a Monte Carlo based simulation multiple comparison with a control test. The simulation size was 12616 and the simultaneous confidence interval was 95%.

The fouling layer thickness was not significantly different for the prechlorinated treatments. Figure 12 shows that the trends were mixed.

Post-filtration reduced the fouling layer thickness for CTRL and CTRLCL only. Figure 18 shows that in the other treatments the impact of post-filtration on the fouling layer thickness was not significant.

Biological treatment showed a significant reduction in fouling layer thickness as compared to the control for all treatments except the post-filtered treatments that were prechlorinated. Figure 6 shows a difference of as much as 30 microns difference from the control.

5.6 Bead Assay

The bead assay cell counts were analyzed with a Monte Carlo based simulation multiple comparison with a control test. The simulation size was 12616 and the simultaneous confidence interval was 95%.

Prechlorination reduced the bead assay cell counts significantly for BAC, IOCS, and CTRL. Figure 14 shows that in the cases where there was a close to a one log reduction for the prechlorinated treatments. It should be noted that there is no chlorine residual carried thought the system.

For post-filtered treatments, all differences were significant except for CTRLCL. Figure 20 shows that the bead assay cell counts were higher for the non-post-filtered treatments and that this trend continued with the CTRLCL treatment even thought the difference with and without the post-filter was not statistically significant.

Biological treatment reduced the bead assay cell counts significantly for all treatments. The biological treatments resulted in as much as a log reduction in biofilm cells on the beads. In addition, as seen in Figure 8, the difference between the BAC and IOCS treatments was significant (> 0.5 log) except for the filtered treatments that were prechlorinated.

5.7 Summary

To pool together the results of the various assays to determine if the hypotheses are to be accepted, the assays were scored with a +1 if the treatment was statistically better than the control treatment and a -1 if it was statistically worse than control treatment. If the treatment was not statistically different than the control treatment, then it received a score of zero. For the flux, a higher number means a greater flux and hence, better performance. Therefore, a statistically higher flux would score a +1. For membrane cells, a lower cell number is considered a reduction in fouling and hence a lower number would score +1. For both fouling layer thickness measurements, a thinner thickness is considered a reduction in fouling and would receive a +1. Similarly, a greater TOC removal is considered a +1 and a lower cell count on the bead assay is considered good and receives a +1.

The assay scores were then added together to obtain an overall score for the treatments. These summaries are then used to determine if the hypothesis in question is accepted.

The impacts of prechlorination on the downstream fouling potential of water are summarized in Table 4. In this table, the assays were scored with a +1 if the treatment was statistically better than no chlorine and a -1 if it was statistically worse than no chlorine. In most cases, we fail to reject the null hypothesis. Overall, the results were minimal and mixed. As a result, prechlorination did not appear to be a factor in down stream fouling.

Table 4. Summary of chlorination impacts on assay measurements.

	BAC	BACPF	IOCSPF	IOCS	CTRL	CTRLPF
Flux	0	0	0	0	0	0
Cells	0	0	0	0	0	0
SEM	-1	0	0	0	-1	0
DAPI	0	0	0	0	0	+1
TOC	-1	0	-1	-1	0	0
Bead	+1	0	0	+1	+1	0
Total	-1	0	-1	0	0	+1

The impacts of post-filtration are summarized in Table 5. Here if post-filtration was statistically better it scored a +1 and if it was statistically worse it scored a -1. A score of zero was used if post-filtration was not statistically different from the non-post-filtration treatment. Overall, we failed to accept the null hypothesis in at least three of the assays and therefore we may conclude that post-filtration reduces the downstream fouling potential of the water.

Table 5. Summary of filtration impacts on assay measurements.

	BAC	IOCS	CTRL	BACCL	IOCSCL	CTRLCL
Flux	+1	0	0	+1	0	0
Cells	+1	+1	+1	+1	+1	+1
SEM	0	0	+1	+1	+1	+1
DAPI	0	0	+1	0	0	+1
TOC	+1	+1	+1	+1	+1	+1
Bead	+1	+1	+1	+1	+1	+1
Total	+4	+3	+5	+5	+4	+4

The effectiveness of biological treatment with respect to the assays is summarized in Table 6. Here the biological treatment scores a +1 if it was statistically better than the control and a -1 if it is statistically worse than the control. The treatment will score a zero if it is not statistically different from the control. Note that only one of the biological treatments must work for the score to be +1 and that both must fail for a score of zero. In this case, we fail to accept the null hypothesis in at least three assays for the prechlorinated/post-filter treatment and in five assays for the other combinations. Therefore, we may conclude that biological treatment reduces the downstream fouling potential of the water.

Table 6. Summary of biological treatment impacts on assay measurements.

	NPFCL	PF	CL	PFCL
Flux	0	+1	0	0
Cells	+1	+1	+1	+1
SEM	+1	0	+1	0
DAPI	+1	+1	+1	0
TOC	+1	+1	+1	+1
Bead	+1	+1	+1	+1
Total	+5	+5	+5	+3

To pick out the difference between the IOCS and BAC, the rankings for the biological treatment were changed to +1 if BAC was statistically "better" than IOCS, zero if there was no difference and -1 if IOCS was statistically "better" than BAC. Table 7 shows that the results were mixed with BAC performing better than IOCS for the post-filtered treatments, IOCS performing "better" than BAC for the prechlorinated treatment and no difference for the non-post-filtered, non-prechlorinated treatment. The two assays that showed a preferential trend were TOC and Bead with BAC removing more TOC than IOCS and IOCS reducing the biofilm cells on the glass beads more than the BAC.

Table 7. Summary of BAC versus IOCS biological treatments

	NPFCL	PF	CL	PFCL
Flux	0	+1	0	+1
Cells	0	0	0	0
SEM	0	0	-1	0
DAPI	0	0	0	0
TOC	+1	+1	+1	+1
Bead	-1	-1	-1	0
Total	0	+1	-1	+2

To qualitatively rank the treatments, the treatments were scored from 1 to 12 without regard to statistical differences. Many scores could differ by more than one and still be statistically the same. However, this provides another view of the overall trends of the results of the experiments.

Table 8 shows how the treatments ranked in order from the most effective treatment (lowest score) to the least effective treatment (highest score) without regard to statistical differences. This provides a qualitative measure of the overall effectiveness of each treatment.

Table 8. Qualitative ranking of assay measurements. The ranking presented here uses the convention that a lower number is 'better' than a higher number.

	Flux	Cells	SEM	DAPI	TOC	Bead	Ave
IOCSPF	3	1	4	1	3	1	2.17
BACPF	1	2	7	2	1	4	2.83
BACPFCL	2	4	2	5	2	3	3
IOCSPFCL	4	3	1	6	4	2	3.33
CTRLPF	5	5	5	10	5	7	6.17
CTRLPFCL	7	6	3	8	7	7	6.5
BAC	6	10	8	4	6	11	7.5
IOCS	8	8	6	7	8	9	7.67
IOCSCL	10	9	9	3	10	5	7.67
BACCL	9	7	10	9	9	6	8.33
CTRLCL	11	12	12	11	12	10	11.33
CTRL	12	11	11	12	11	12	11.5

The ranking reveals that the combination of biological treatment and post-filtration proved to be the most effective in reducing the effects as measured by the assays. Filtration alone appears to do better than biological treatment alone and any treatment appears to reduce downstream fouling except for prechlorination.

6.0 Cost Analysis

6.1 Introduction

To assess the potential cost savings from the proposed research, it was necessary to establish a cost model that could be used to estimate existing costs for membrane treatment of water. While there are many cost models in the literature, they were usually geared toward parameters (e.g. comparisons to multistage flash distillation, Wade, 1993) other than those necessary for this analysis.

The uses of membrane processes in the world range from fairly large scale desalination facilities in the Middle East and treatment of brackish water in the U.S. for potable water to highly specialized applications found in the pharmaceutical, semiconductor, and boiler feedwater industries that require ultrapure water. These industries will have source waters that range from seawater (40,000 mg/L TDS) to water from a publicly owned treatment works (POTW) that is drinking water quality (<500 mg/L TDS). In addition, the economy of scale is evident in the range of sizes of membrane facilities that are operating.

To determine the possible cost savings for the biological pretreatment unit process, it was decided to examine the RO seawater desalination industry. Desalination poses higher requirements on membranes due to higher operating pressures and higher TDS content. The literature also provided a good cross section of cost values with respect to both size and location of desalination plants. The costs for this operation should represent a lower bound for savings using biological pretreatment technology since the cost in $/1000gallons in the ultrapure water industry can be significantly higher. Therefore, any savings shown by this analysis should extrapolate to higher savings in other areas of membrane water treatment processes.

6.2 Approach and assumptions

A sampling of the literature and industries involved with seawater desalination revealed cost data that covered plant sizes from 20,000 GPD to 36,000,000 GPD (Darwish, et. al., 1990, Leitner, 1998). The studies used data from 1985 until 1997 (El-Rehaili, 1991, Leitner, 1998) and covered plants with source waters in the Red Sea, the Arabian Gulf, the Mediterranean Sea, the Caribbean Sea, and the Atlantic Ocean (DuPont, 1996). To account for the variation in costs found in the literature due to the year of the source data, the Building Cost Index from the Engineering News Record (http://www.enr.com) was used to bring the costs up to a common point in time of December 31, 1999.

The cost model was an empirical model fit for production capacity in gallons per day (GPD) versus total annual cost in dollars per thousand gallons of produced water. Once the empirical model was obtained, there was enough information to assess the portion of the total annual cost that was amortized capital cost. The remaining portion of the total

annual cost was therefore the total annual operation and maintenance cost. The literature data revealed a percentage of the total annual O&M cost that was due to chemicals. The chemical costs included those for cleaning membranes, biocides, coagulation, scale control, disinfection, etc. The fraction of the total annual O&M costs that are chemical costs as a function of plant production size were modeled with a first-order polynomial in a least squares sense. A similar approach was utilized for the annual membrane replacement costs fraction of total O&M costs It was assumed that labor cost was not going to change as a result of a change in membrane replacement rates or cleaning cycles, so they were not addressed. This assumption would need to be addressed for small scale desalination plants and ultrapure water plants where labor costs can be significant.

Once the existing costs were modeled, an estimate of the savings from implementing biological pretreatment was determined. Since the biological pretreatment unit process proposes to use microfiltration as a post filtration step, the coagulation/flocculation step before the gravity filters could be relaxed but not eliminated. The RO cleaning cycles were reduced by one cycle annually and the biocide and disinfection steps were assumed to be reduced by 10%. Overall, it was assumed that the annual chemical costs at the RO plant were to be reduced by 25% annually. For the membrane replacement, it was assumed that instead of the 20% annual replacement rate used in most of the literature cost numbers, a 15% replacement rate might be achieved. This amounts to a 25% reduction in membrane costs. This is still conservative since with proper pretreatment, DuPont recommends an estimate of a 12% replacement rate for membranes.

To determine the cost of implementing a biological pretreatment unit process, the process was broken into determining the increased capital cost due to a longer EBCT for a BAC filter versus a gravity mixed media filter, and determining the cost of post microfiltration of the BAC filter. To this end, the annual capital cost for existing gravity filters was determined from the plant production size in the model and cost equations available from Qasim, et.al., 1992 which were then amortized for 20 years at an interest rate of 8% and then adjusted with the Building Cost Index from the Engineering News Record. In addition, these cost equations, which depend upon area of the filtration, allowed for calculation of the BAC filter size and subsequent cost increase. The cost of microfiltration was determined from Ebrahim, et.al., 1997, which compared MF with traditional filtration for RO desalination plants. The cost comparison in this study allowed for a determination of the annual capital and O&M costs of the MF implementation along with the BAC filtration. These costs were then combined and subtracted from the estimated savings from the chemicals and membrane replacements to determine an overall annual estimated cost savings per thousand gallons due to the implementation of the biological pretreatment unit process.

6.3 Results

The total annual cost for RO desalination of seawater as a function of plant production size was fit to the curve y=ax^b. For the data in the literature search, a = 16.6232 and b = -0.1403. The multiple R-squared value for this data set was 0.284. This low value is to be expected due to the variable nature of cost numbers usually found in the literature. The F-statistic was 5.554 with a p-value of 0.03353. The model did, however, capture the economy of scale found in these plants. The modeled costs ranged from over $10/1000 gallons for plants less than 30,000 GPD down to $3.81/1000 gallons for a plant production size of 36 MGD.

The estimated cost savings from the chemical and membrane reduction costs were found to be approximately a 9.3% to 9.6% of the total estimated annual cost in $/1000gallons. The biological pretreatment unit process cost model yielded an total estimated annual cost that ranged from 0.027 $/1000 gallons to 0.36 $/1000 gallons. Combining this estimated cost with the estimated cost savings yields an estimated cost savings that ranges from 0.33 $/1000 gallons to 0.68 $/1000 gallons for plant production sizes that range from 30,000 GPD to 36,000,000 GPD. It should be pointed out that on a percentage basis, the estimated cost savings ranged from 6% to 8.8% of the total estimated annual cost with a mean savings of 8.7% of the total estimated annual cost.

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Appendix 1 - Tables

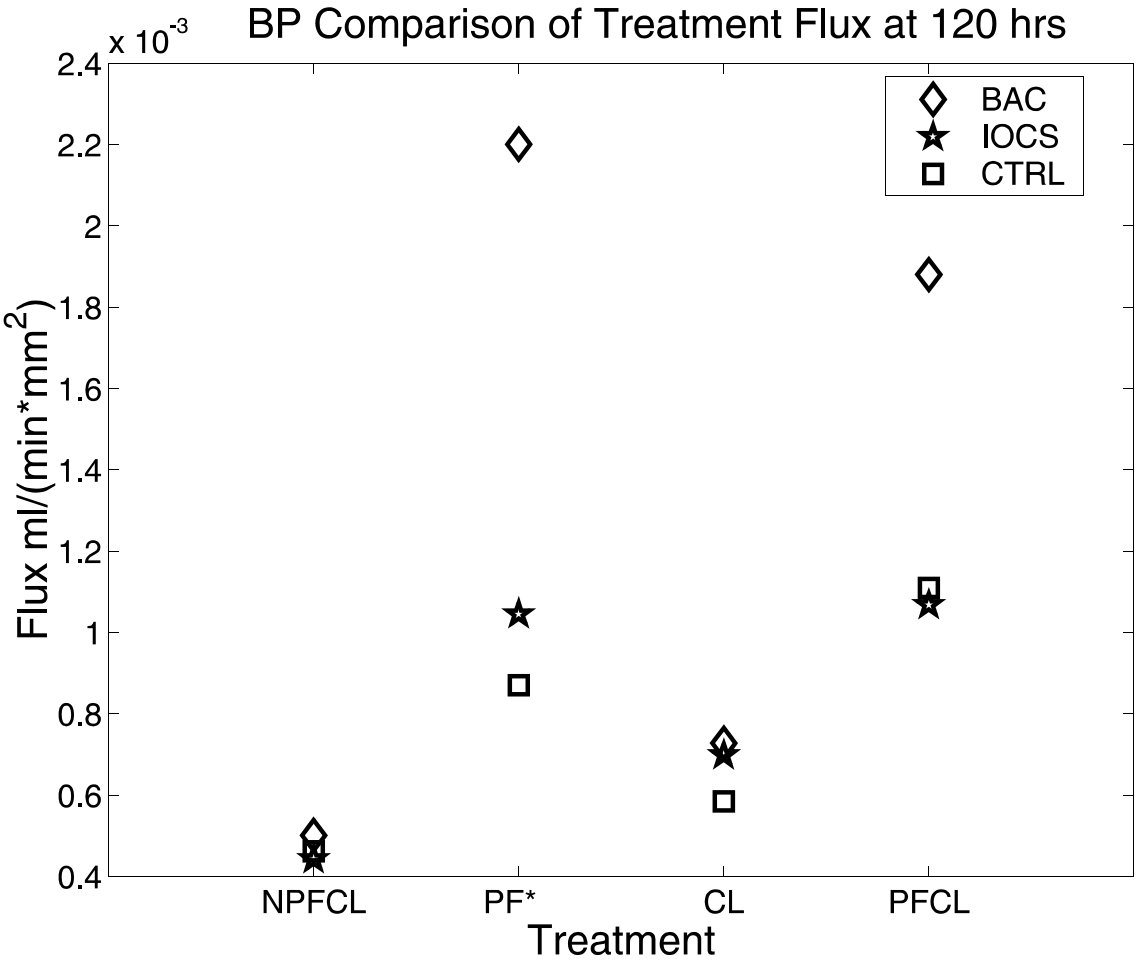
Table 2. Flux prediction model parameters

Treatment	a	b	R^2	F-stat	P-value
BAC	4.88e-3	-0.4757	0.7203	41.21	8.482e-6
BACPF	7.33e-2	-0.7323	0.7764	55.56	1.371e-6
IOCSPF	2.84e-2	-0.6901	0.8522	92.24	4.805e-8
IOCS	1.18e-2	-0.6859	0.7317	43.63	6.048e-6
CTRL	2.94e-3	-0.3869	0.6662	31.93	3.613e-5
CTRLPF	9.56e-3	-0.5006	0.6917	35.89	1.883e-5
BACCL	6.39e-3	-0.4539	0.6261	26.79	9.193e-5
BACPFCL	2.69e-2	-0.557	0.5809	22.17	2.365e-4
IOCSPFCL	2.15e-2	-0.6273	0.6072	24.73	1.382e-4
IOCSCL	7.66e-3	-0.5004	0.5529	19.78	4.051e-4
CTRLCL	8.409e-3	-0.5571	0.5278	17.88	6.387e-4
CTRLPFCL	2.83e-2	-0.6765	0.768	52.95	1.853e-6

Table 3. TOC prediction model parameters

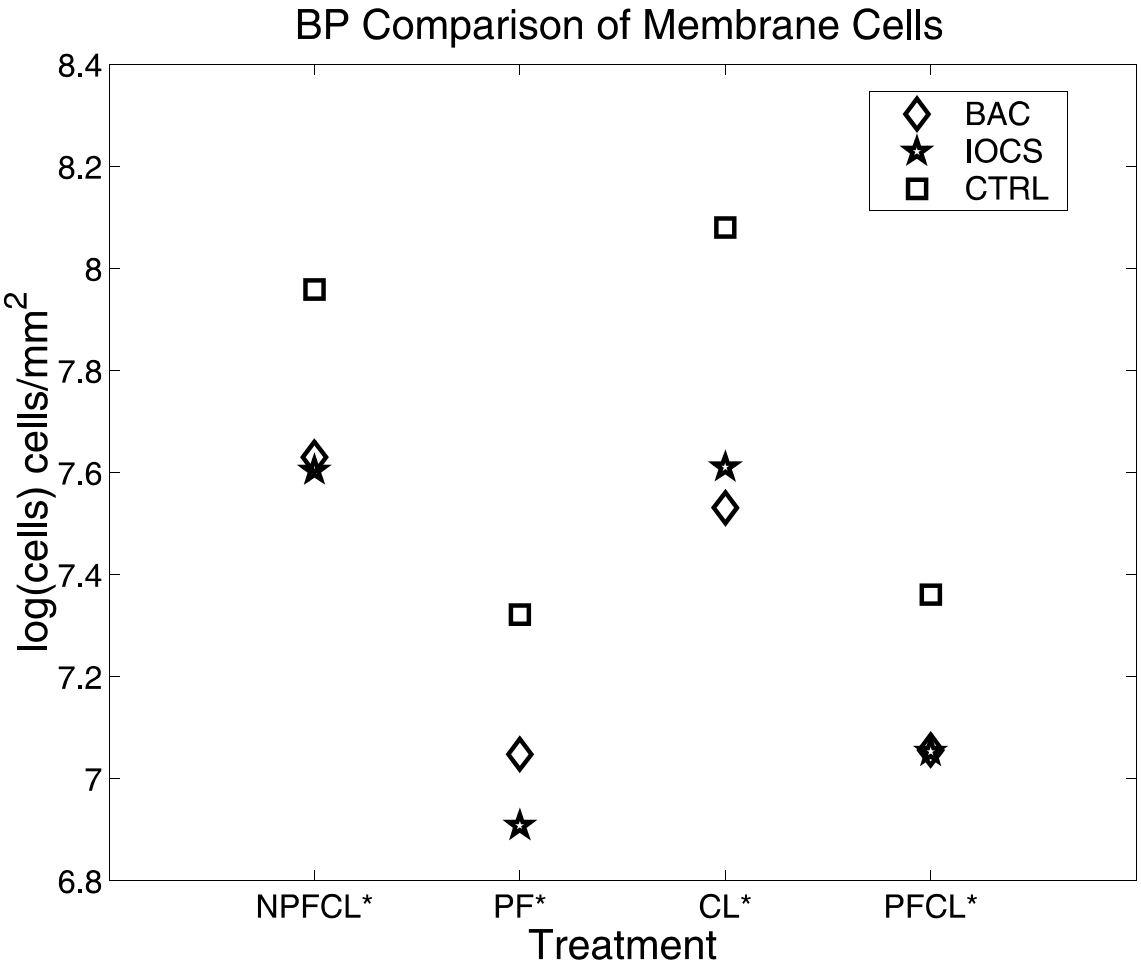
Treatment	m	b	R^2	F-stat	P-value
BAC	0.0283	-0.1600	0.894	194.1	1.066e-12
BACPF	0.0386	7.0781	0.9566	485.1	2.22e-16
BACCL	0.0104	8.1912	0.6014	33.19	8.538e-6
BACPFCL	0.0310	11.3549	0.9539	454.9	3.331e-16
CTRLPF	0.0155	10.3283	0.8995	196.9	1.862e-12
CTRLPFCL	0.0309	-3.5511	0.9567	486.1	2.22e-16
IOCS	0.0306	-5.4254	0.8736	152.1	2.348e-11
IOCSPF	0.0344	3.2004	0.8859	170.8	7.578e-12
IOCSCL	0.0005	3.5192	0.01242	0.2767	0.6041
IOCSPFCL	0.0309	0.4006	0.944	371	2.887e-15

Appendix 2 - Figures



**** Statistically different - 95% Simultaneous Confidence Interval - BAC only***
Tukey's Multiple Comparison Test

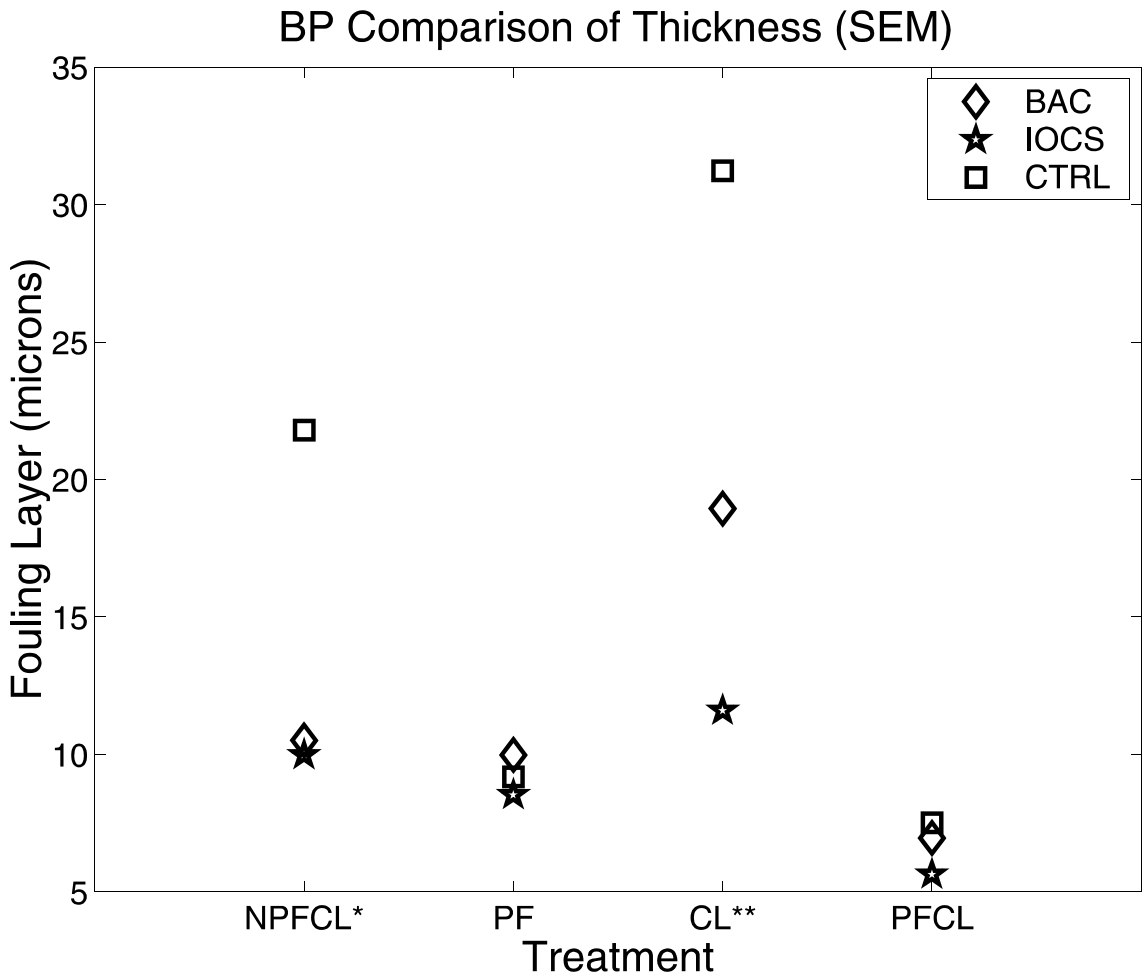
Figure 3.



**** Statistically different - 95% Simultaneous Confidence Interval***

Tukey's Multiple Comparison Test

Figure 4.

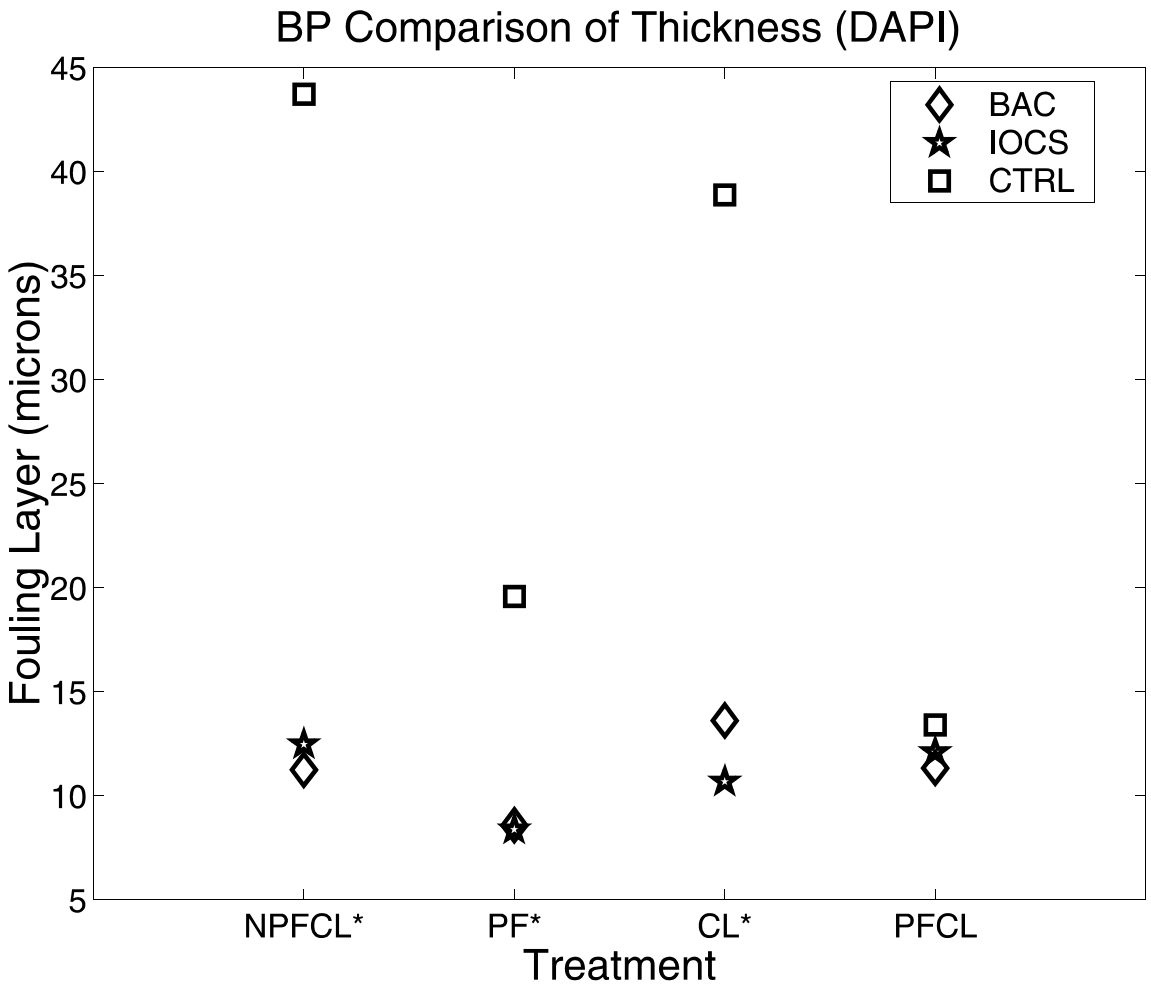


**** Statistically different from control - 95% Simultaneous Confidence Interval***

***** BAC and IOCS are statistically different from each other***

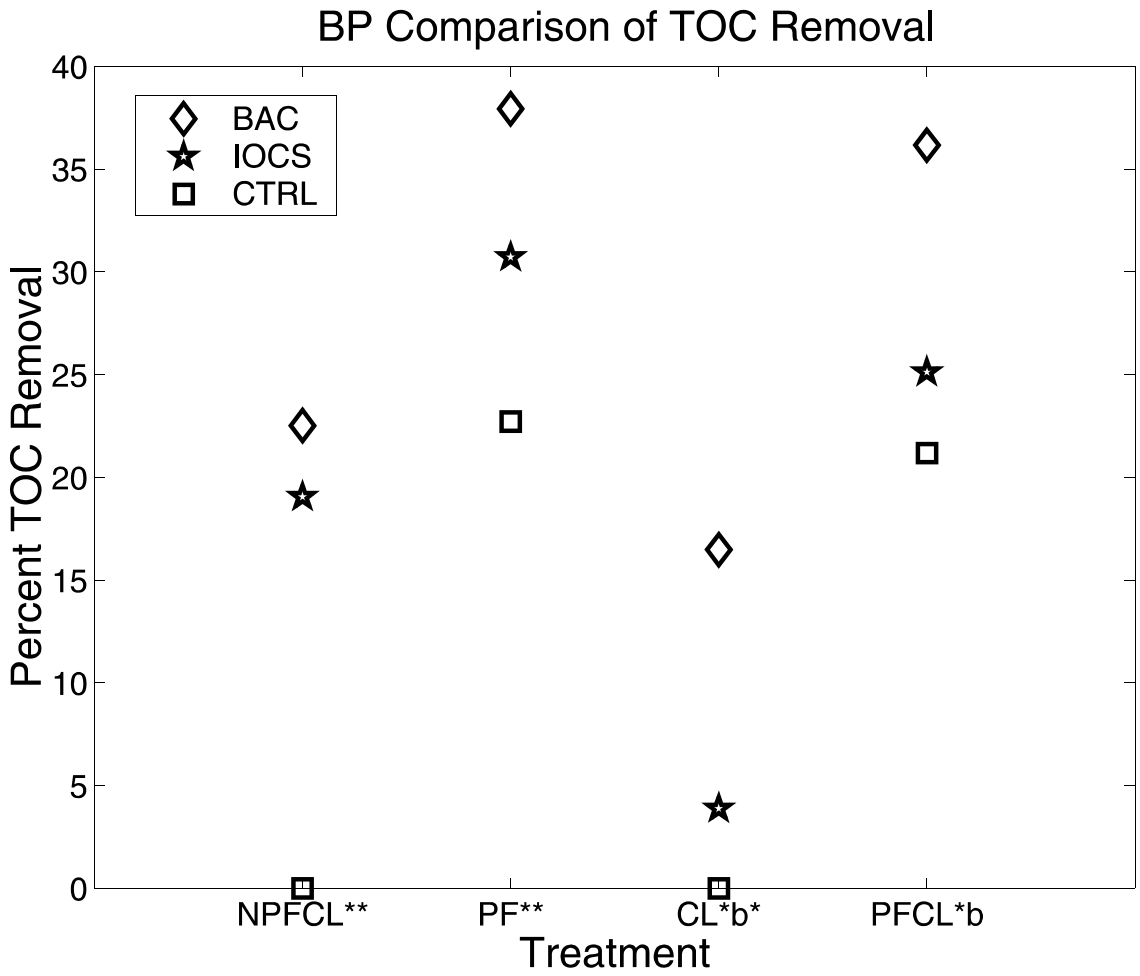
Tukey's Multiple Comparison Test

Figure 5.



**** Statistically different - 95% Simultaneous Confidence Interval***
Tukey's Multiple Comparison Test

Figure 6.



**** Statistically different from control - 95% Simultaneous Confidence Interval***

***** BAC and IOCS are statistically different from each other***

Tukey's Multiple Comparison Test

Figure 7.

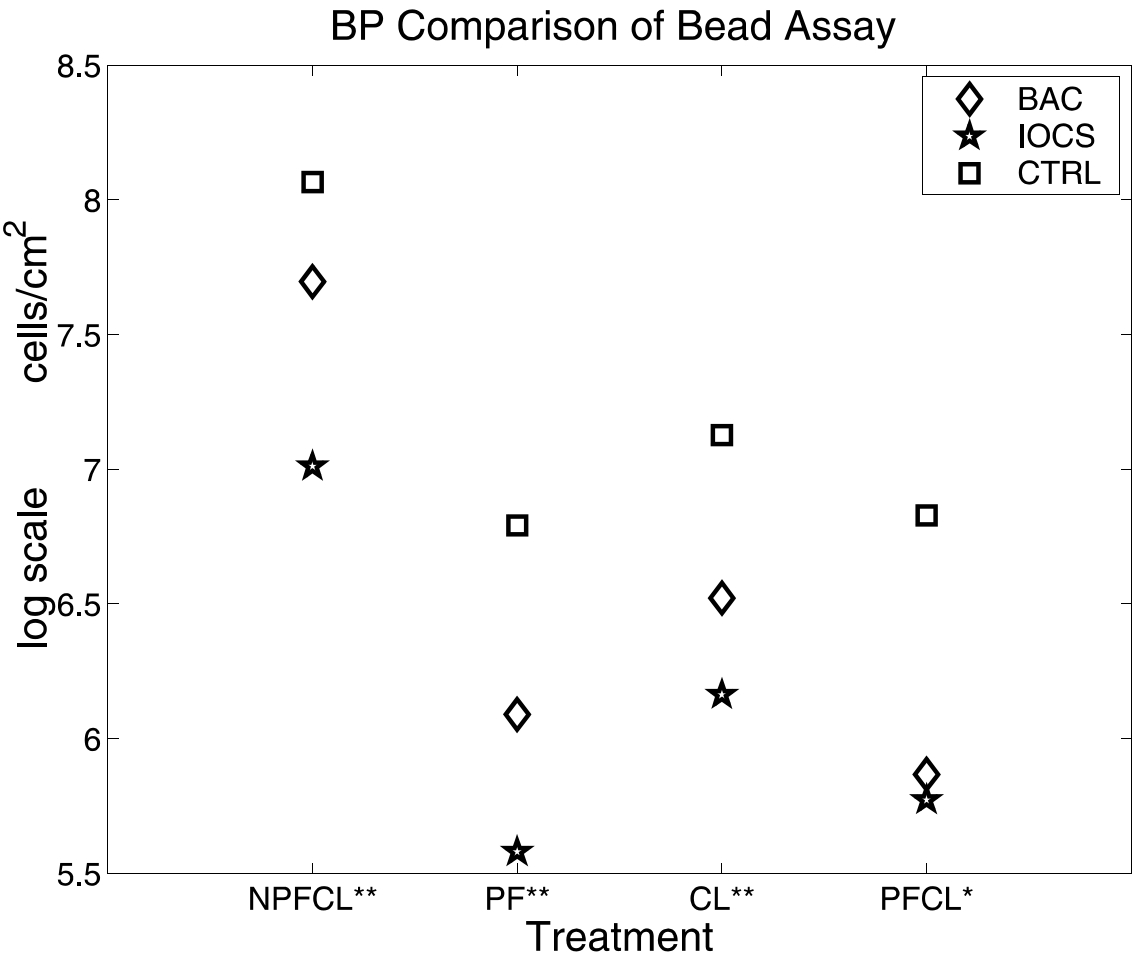
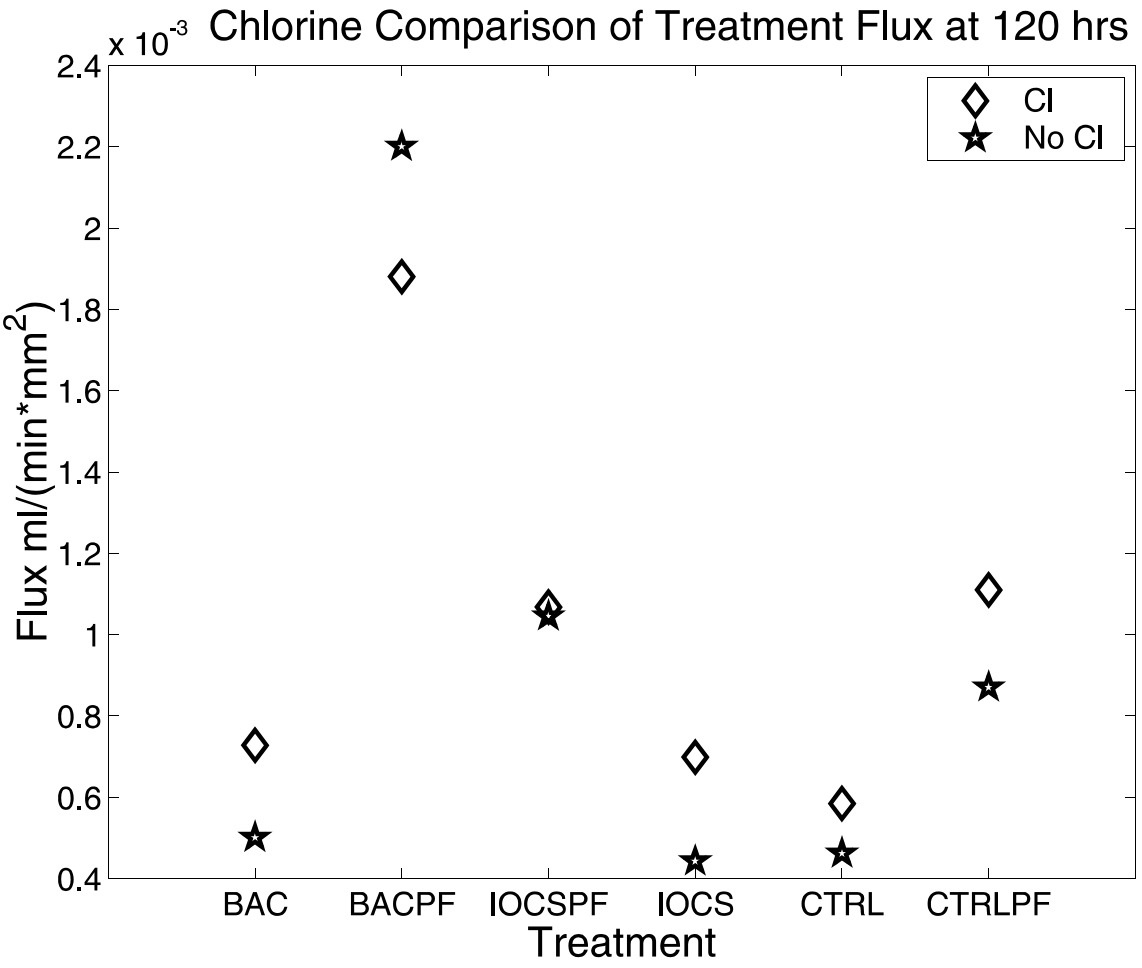

BP Comparison of Bead Assay
BAC
IOCS
CTRL
8.5
8
7.5
7
6.5
6
5.5
log scale cells/cm2
NPFCL**
PF**
CL**
PFCL*
Treatment
* Statistically different from control - 95% Simultaneous Confidence Interval
** BAC and IOCS are statistically different from each other
Tukey's Multiple Comparison Test

Figure 8.



**** Statistically different - 95% Simultaneous Confidence Interval***
Tukey's Multiple Comparison Test

Figure 9.

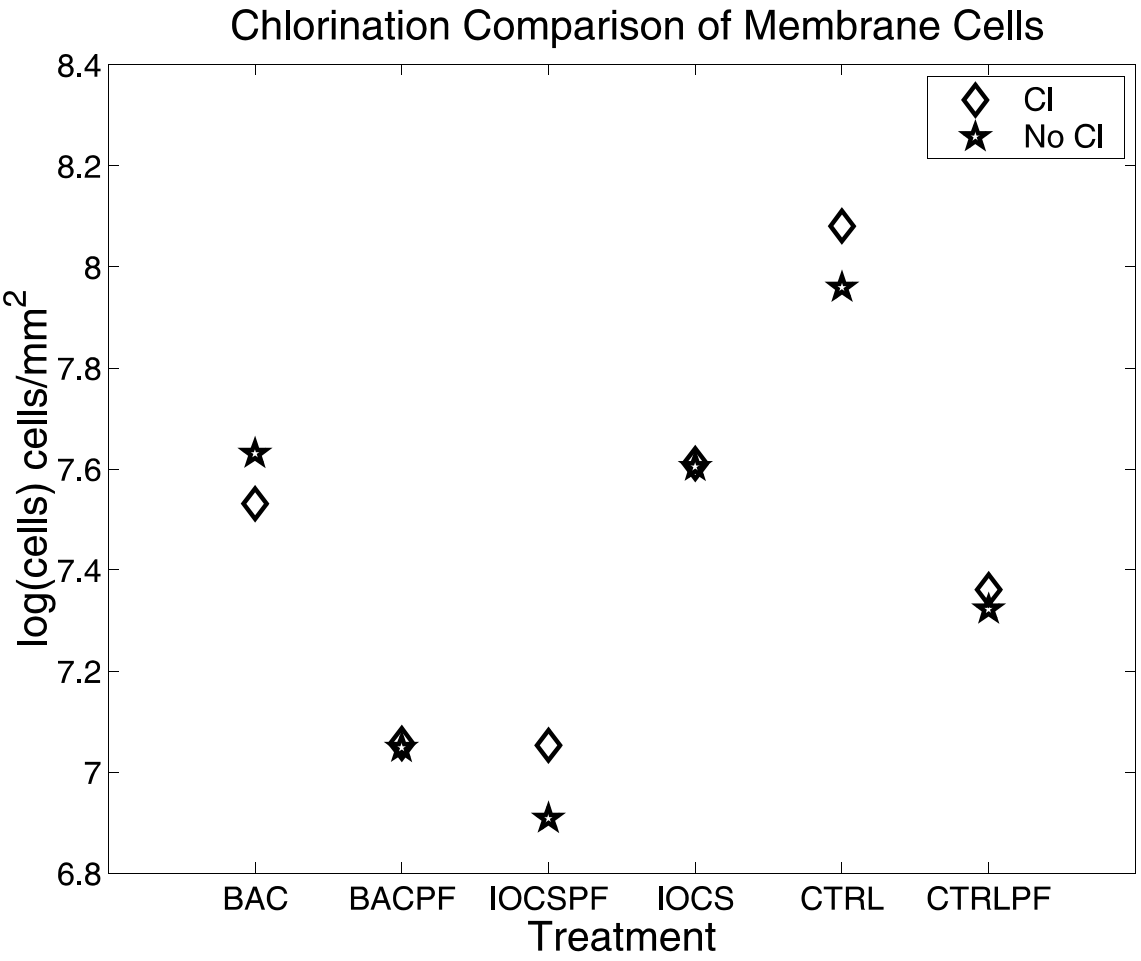
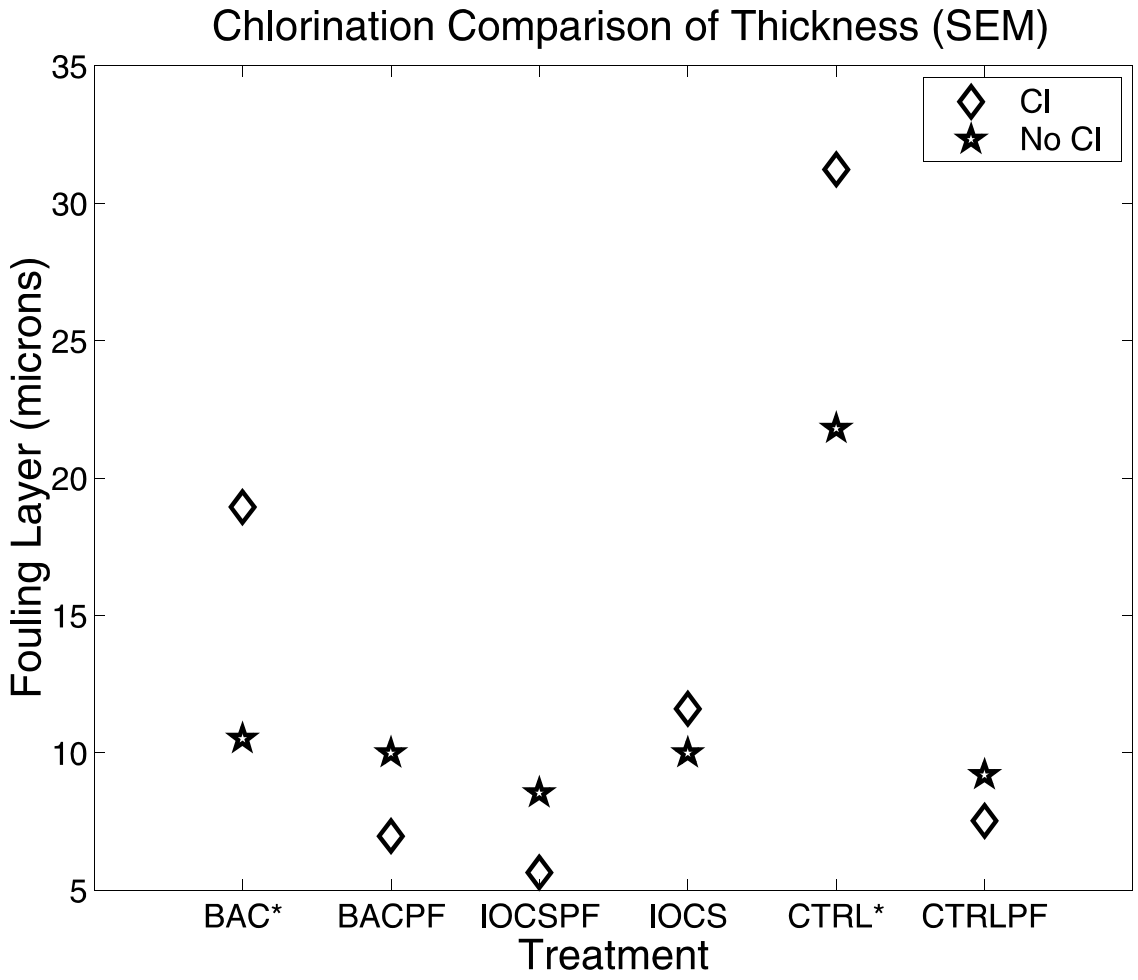

Chlorination Comparison of Membrane Cells
Cl
No Cl
8.4
8.2
8
7.8
7.6
7.4
7.2
7
6.8
log(cells) cells/mm2
BAC
BACPF
IOCSPF
IOCS
CTRL
CTRLPF
Treatment
* Statistically different - 95% Simultaneous Confidence Interval
Tukey's Multiple Comparison Test

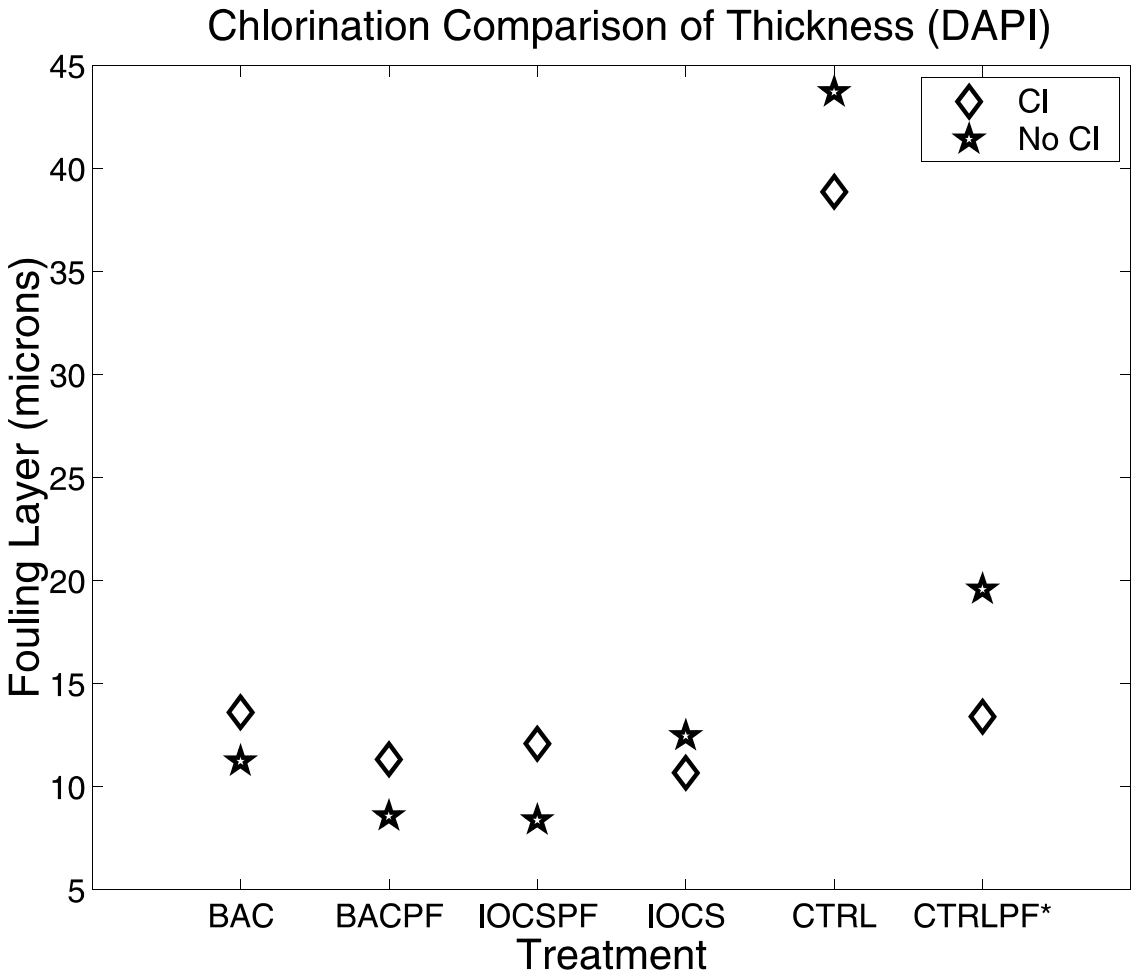
Figure 10.



**** Statistically different - 95% Simultaneous Confidence Interval***

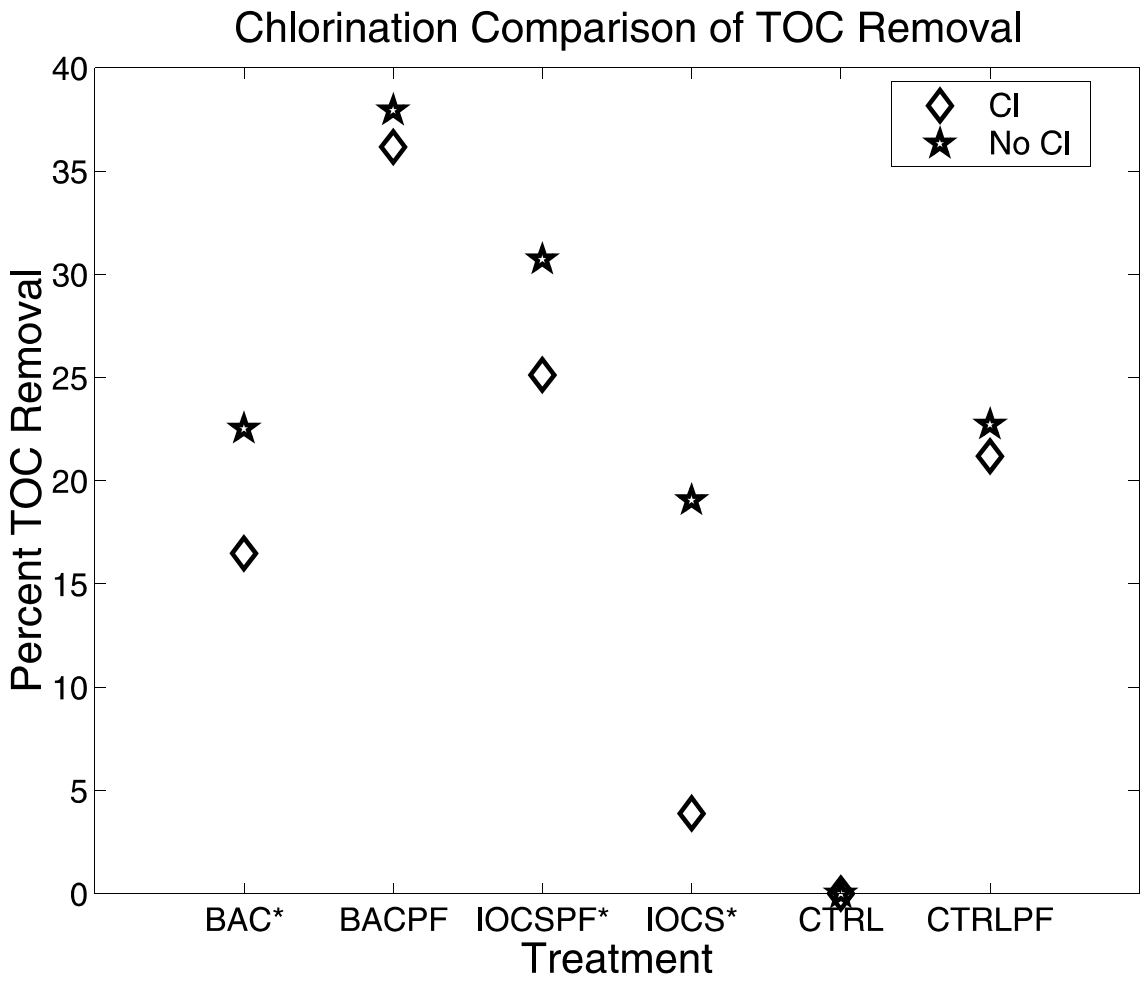
Tukey's Multiple Comparison Test

Figure 11.



**** Statistically different - 95% Simultaneous Confidence Interval***
Tukey's Multiple Comparison Test

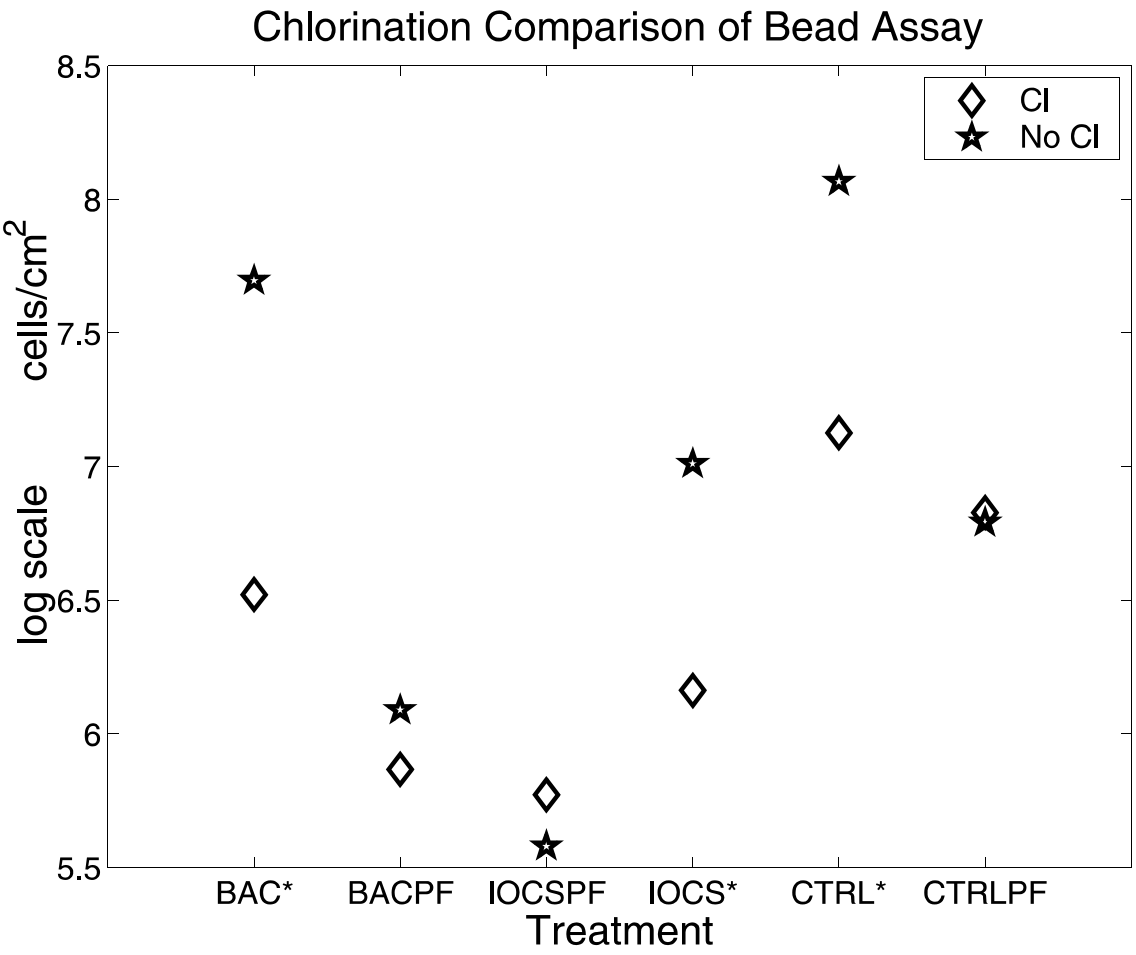
Figure 12.



**** Statistically different - 95% Simultaneous Confidence Interval***

Tukey's Multiple Comparison Test

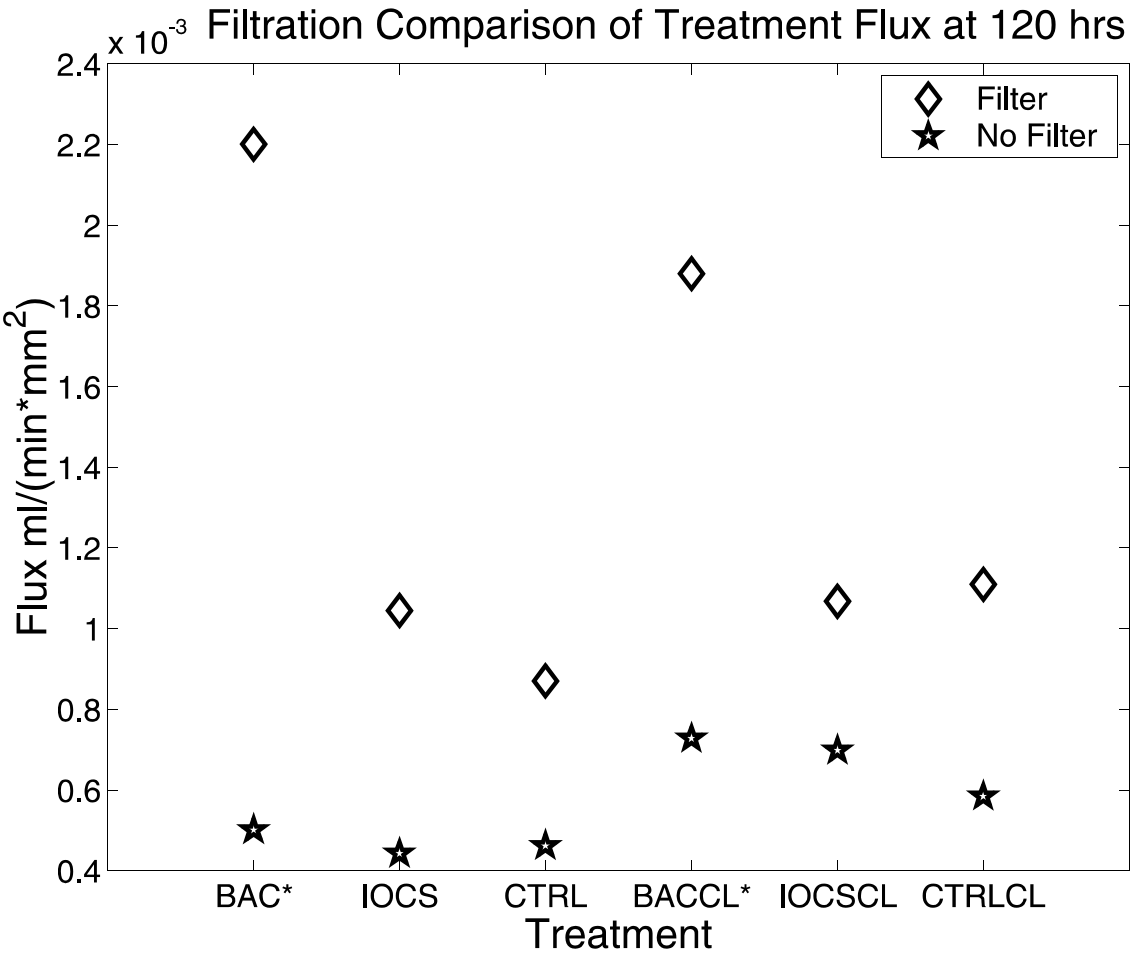
Figure 13.



**** Statistically different - 95% Simultaneous Confidence Interval***

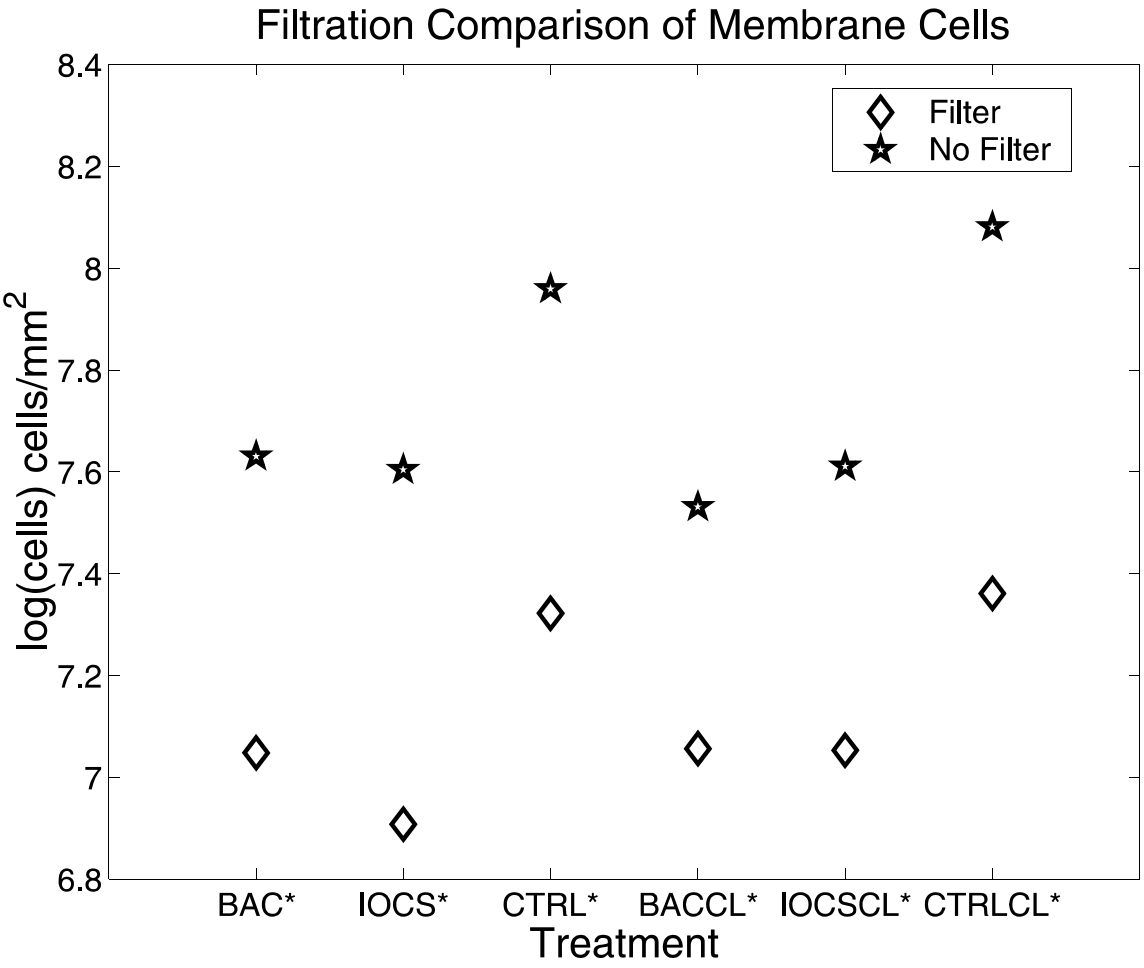
Tukey's Multiple Comparison Test

Figure 14.



**** Statistically different - 95% Simultaneous Confidence Interval***
Tukey's Multiple Comparison Test

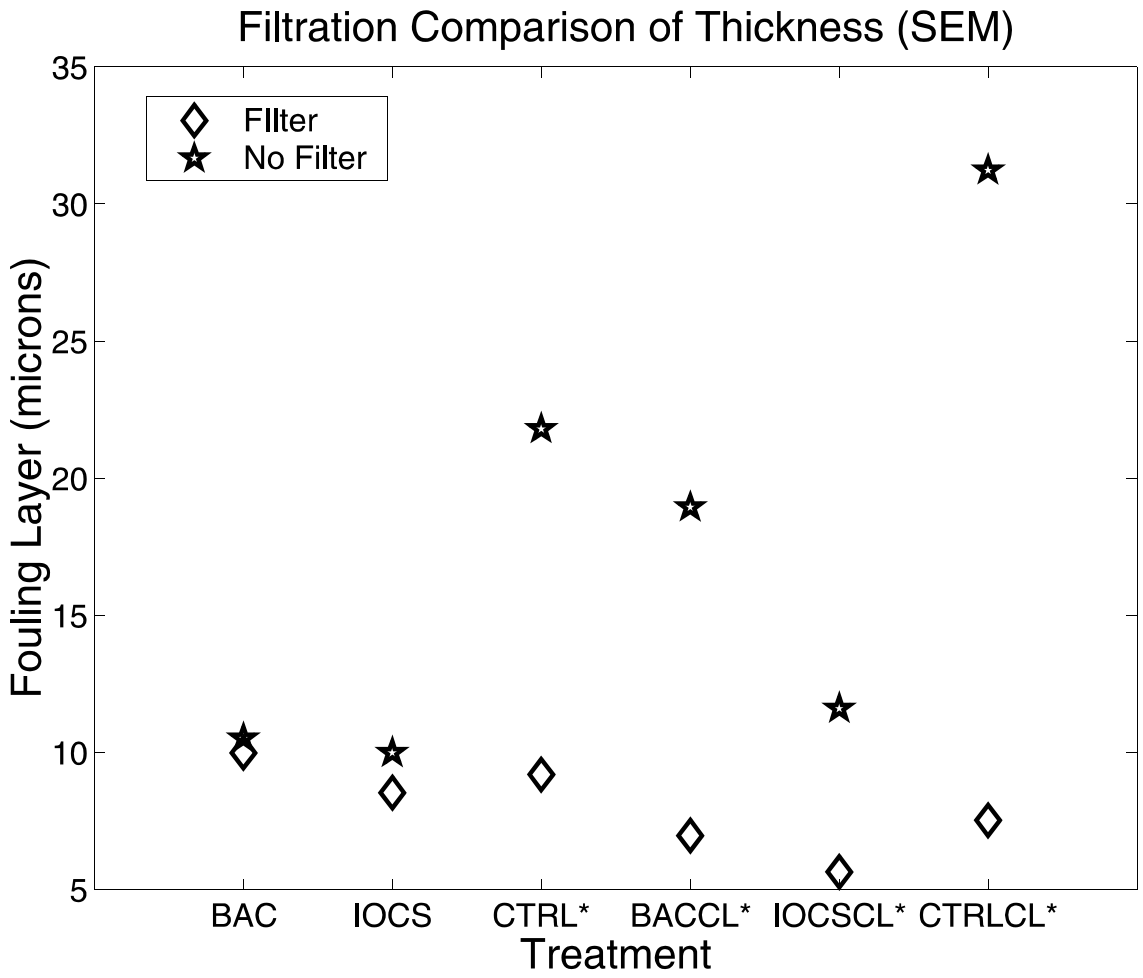
Figure 15.



**** Statistically different - 95% Simultaneous Confidence Interval***

Tukey's Multiple Comparison Test

Figure 16.



**** Statistically different - 95% Simultaneous Confidence Interval***

Tukey's Multiple Comparison Test

Figure 17.

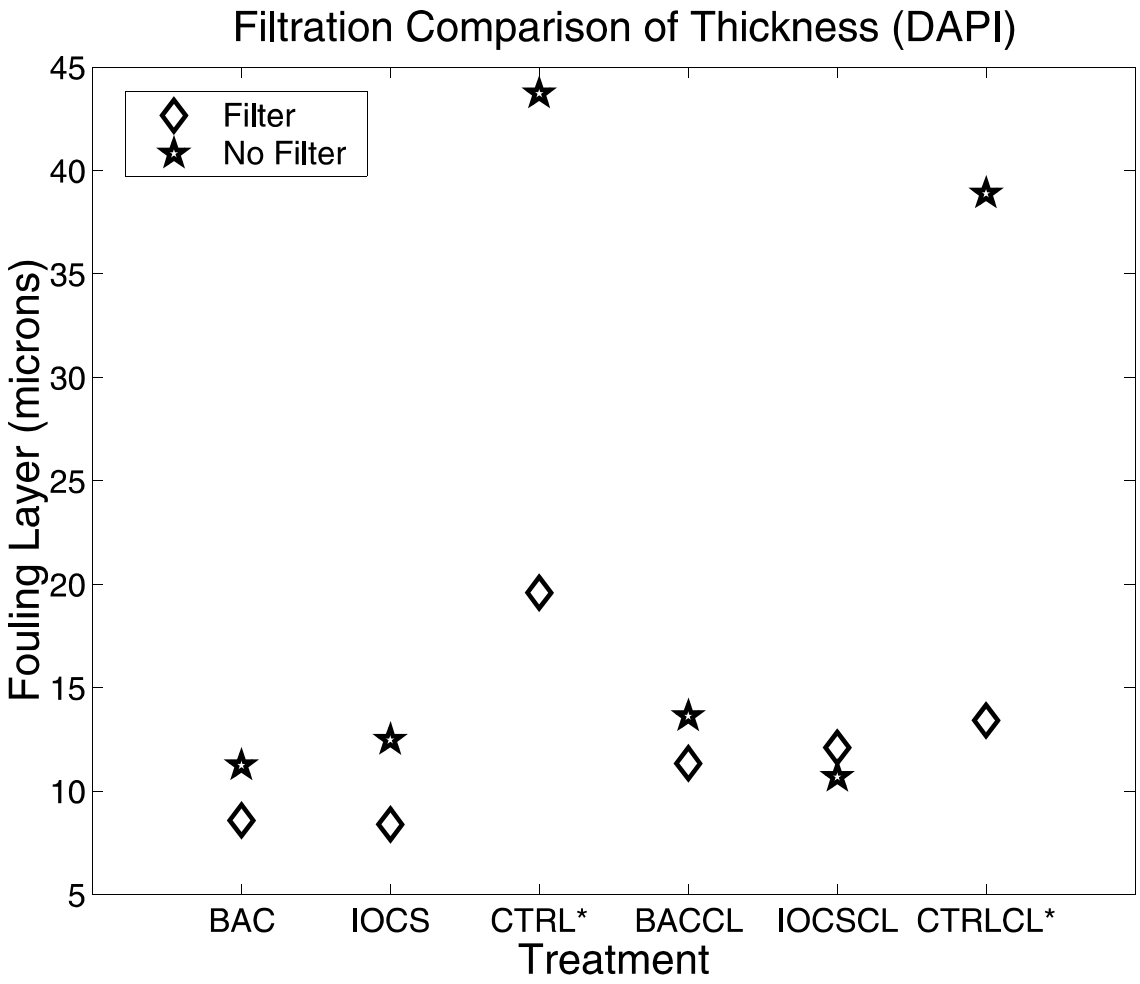

Filtration Comparison of Thickness (DAPI)
Filter
No Filter
Fouling Layer (microns)
45
40
35
30
25
20
15
10
5
BAC
IOCS
CTRL*
BACCL
IOCSCL
CTRLCL*
Treatment
* Statistically different - 95% Simultaneous Confidence Interval
Tukey's Multiple Comparison Test

Figure 18.

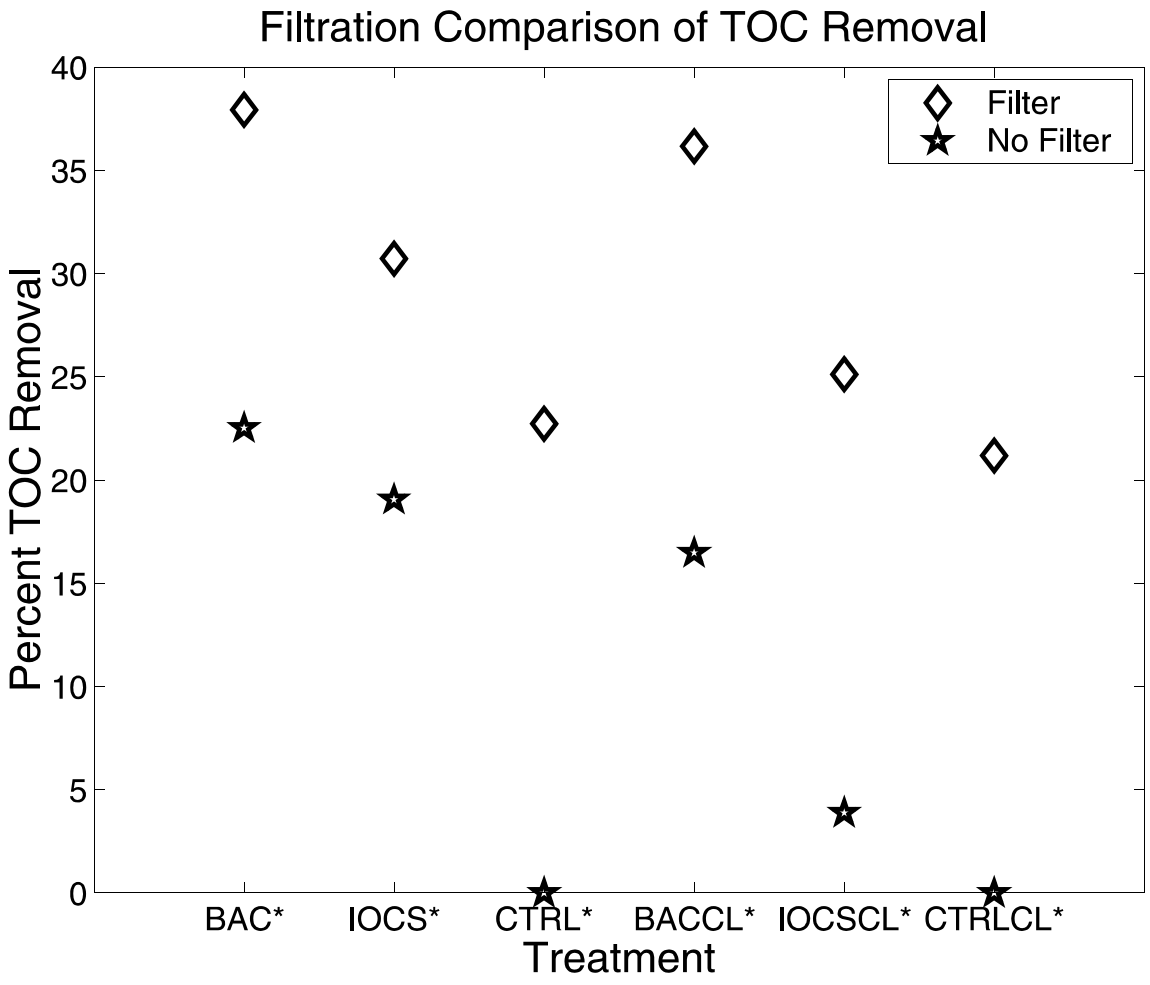

Filtration Comparison of TOC Removal
Filter
No Filter
Percent TOC Removal
0
5
10
15
20
25
30
35
40
BAC*
IOCS*
CTRL*
BACCL*
IOCSCL*
CTRLCL*
Treatment
* Statistically different - 95% Simultaneous Confidence Interval
Tukey's Multiple Comparison Test

Figure 19.

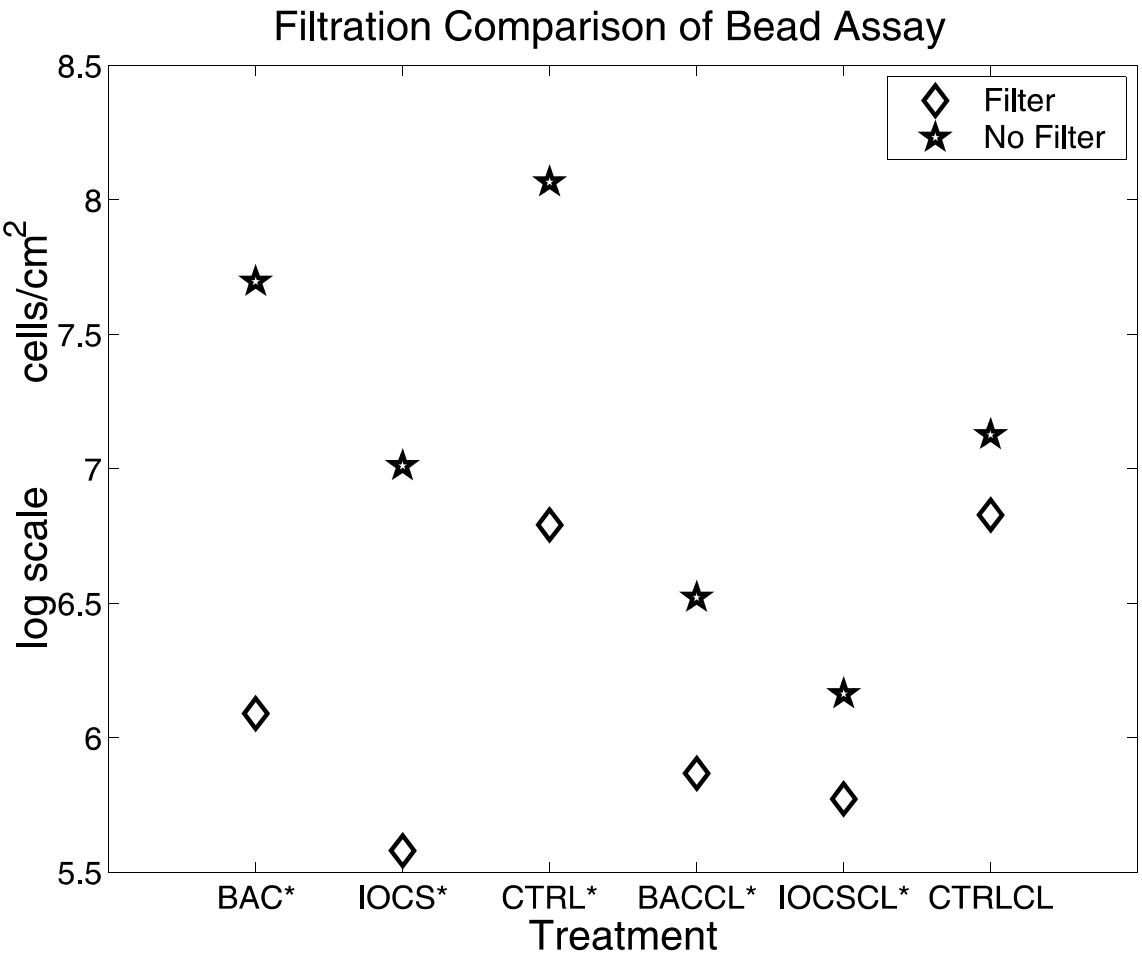

Filtration Comparison of Bead Assay
Filter
No Filter
log scale cells/cm2
8.5
8
7.5
7
6.5
6
5.5
BAC*
IOCS*
CTRL*
BACCL*
IOCSCL*
CTRLCL
Treatment
* Statistically different - 95% Simultaneous Confidence Interval
Tukey's Multiple Comparison Test

Figure 20.

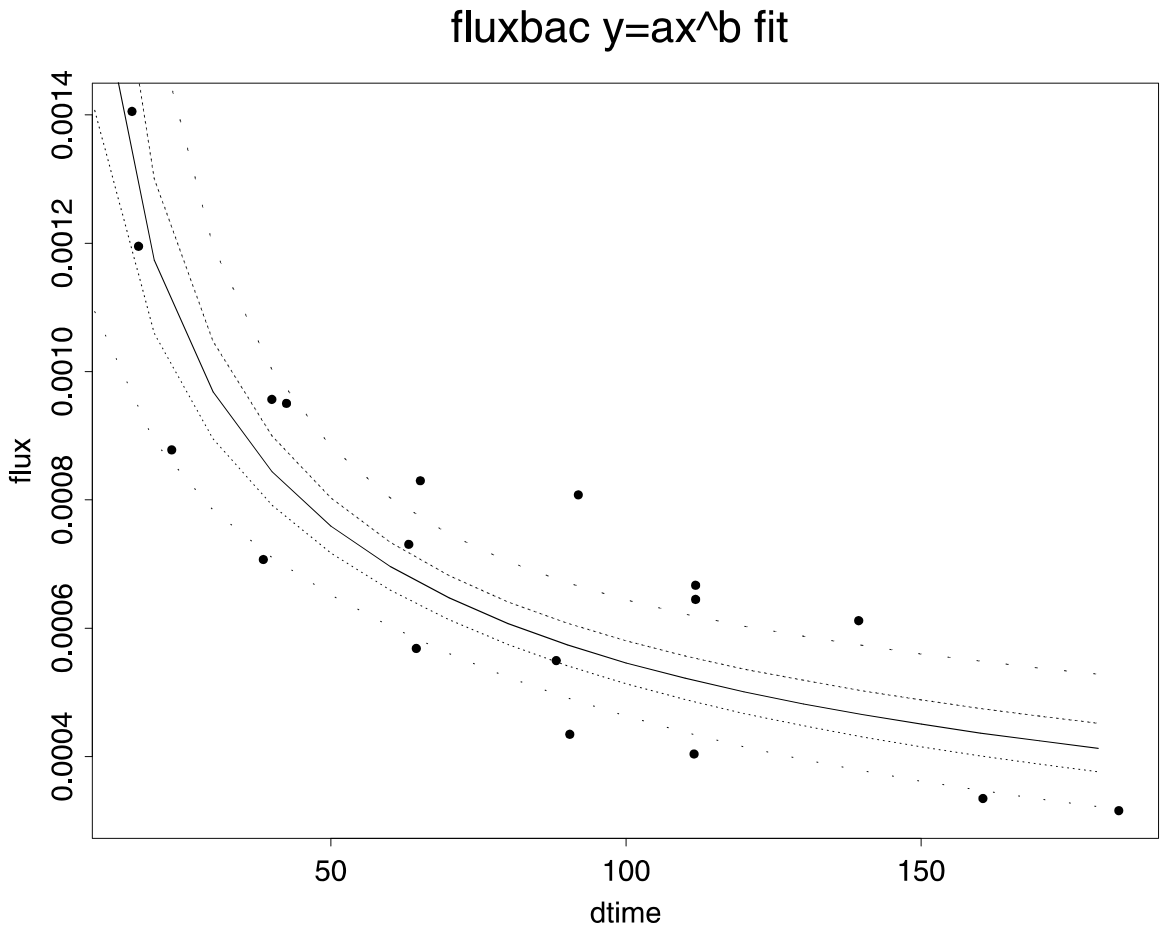


Figure 21. Flux data using biological activated carbon pretreatment. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

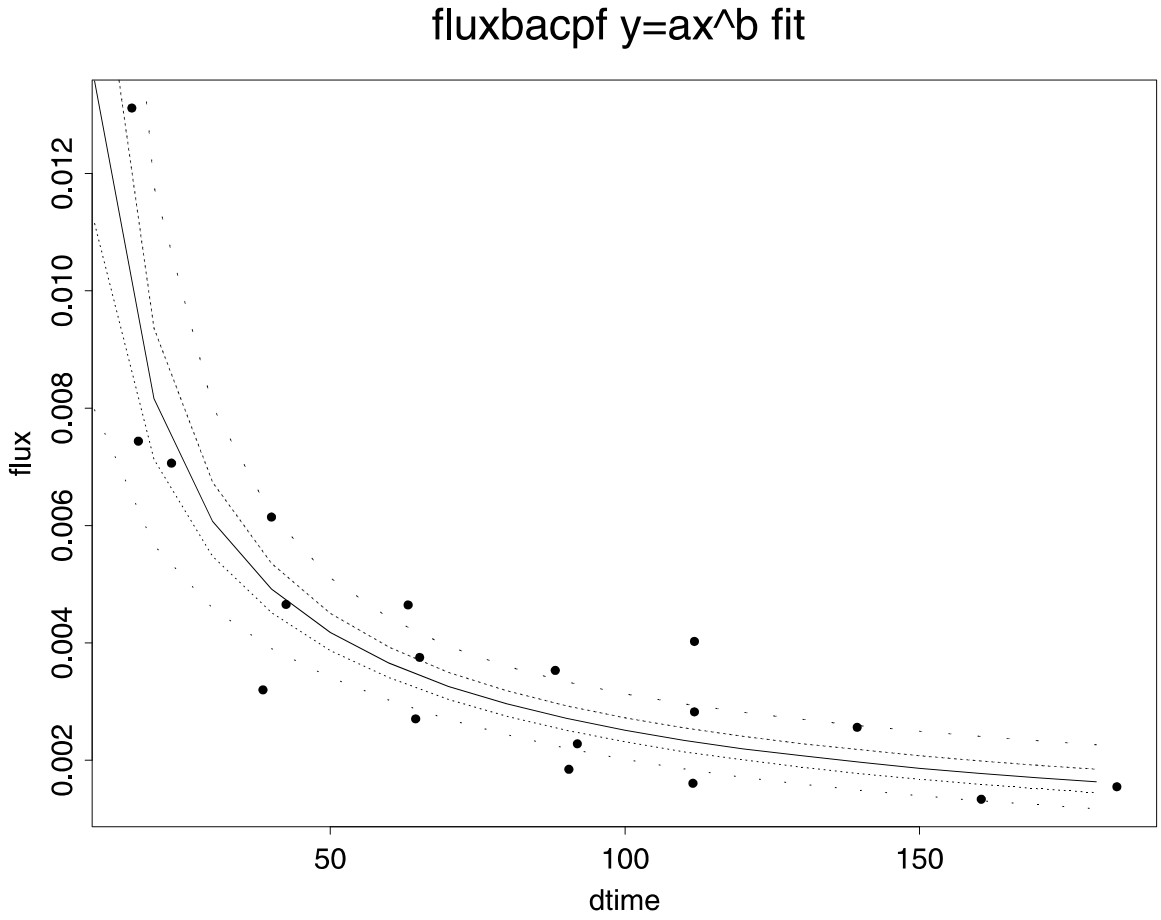


Figure 22. Flux data using biological activated carbon pretreatment with post filtration. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

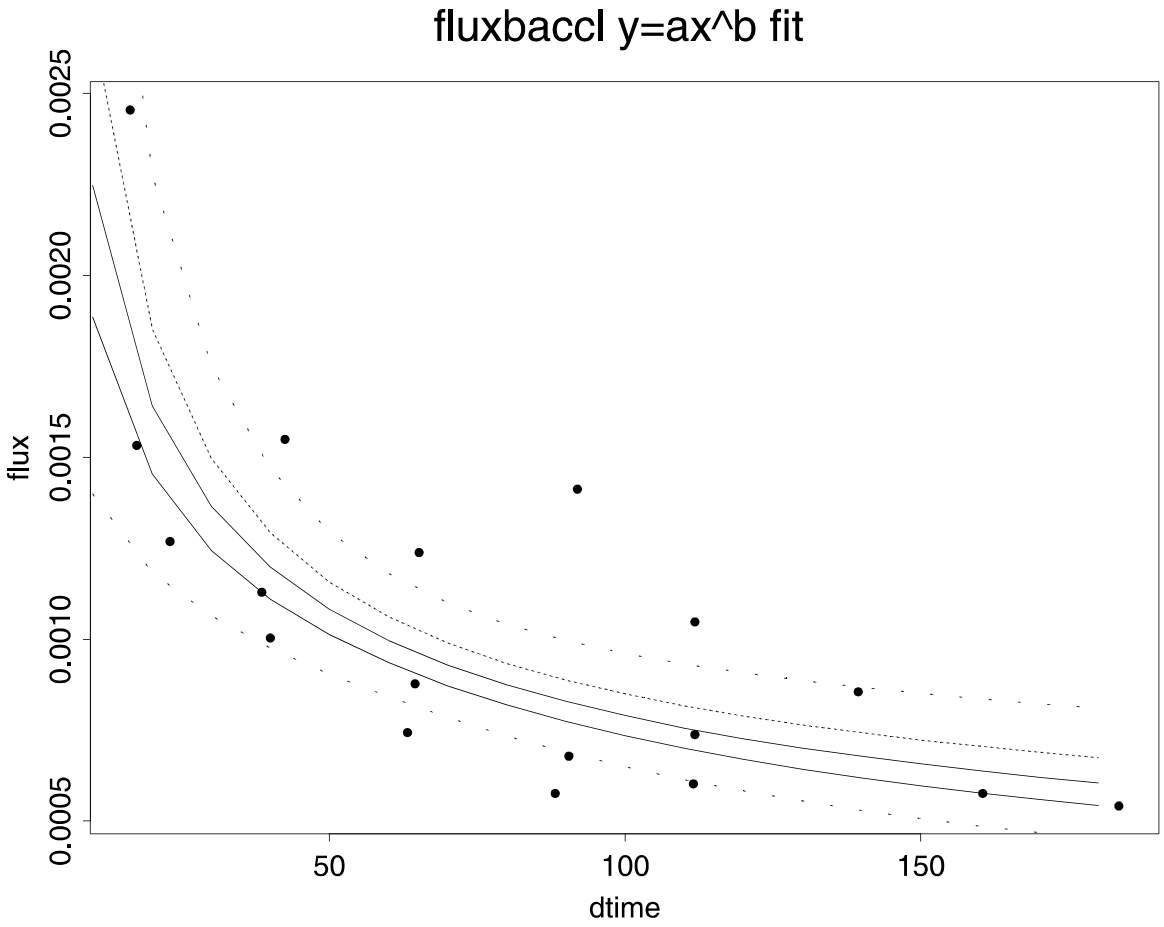


Figure 23. Flux data using biological activated carbon pretreatment with prechlorination. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

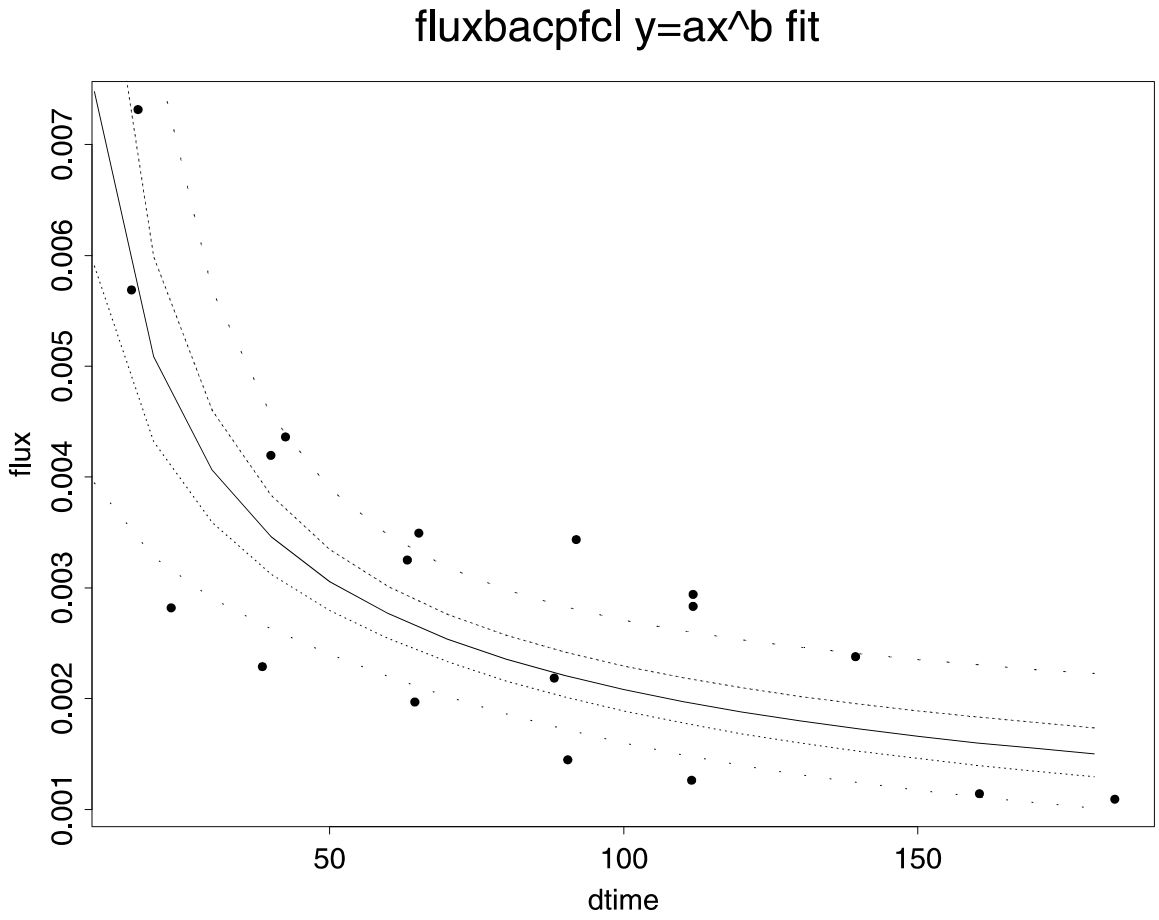


Figure 24. Flux data using biological activated carbon pretreatment with post filtration and prechlorination. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

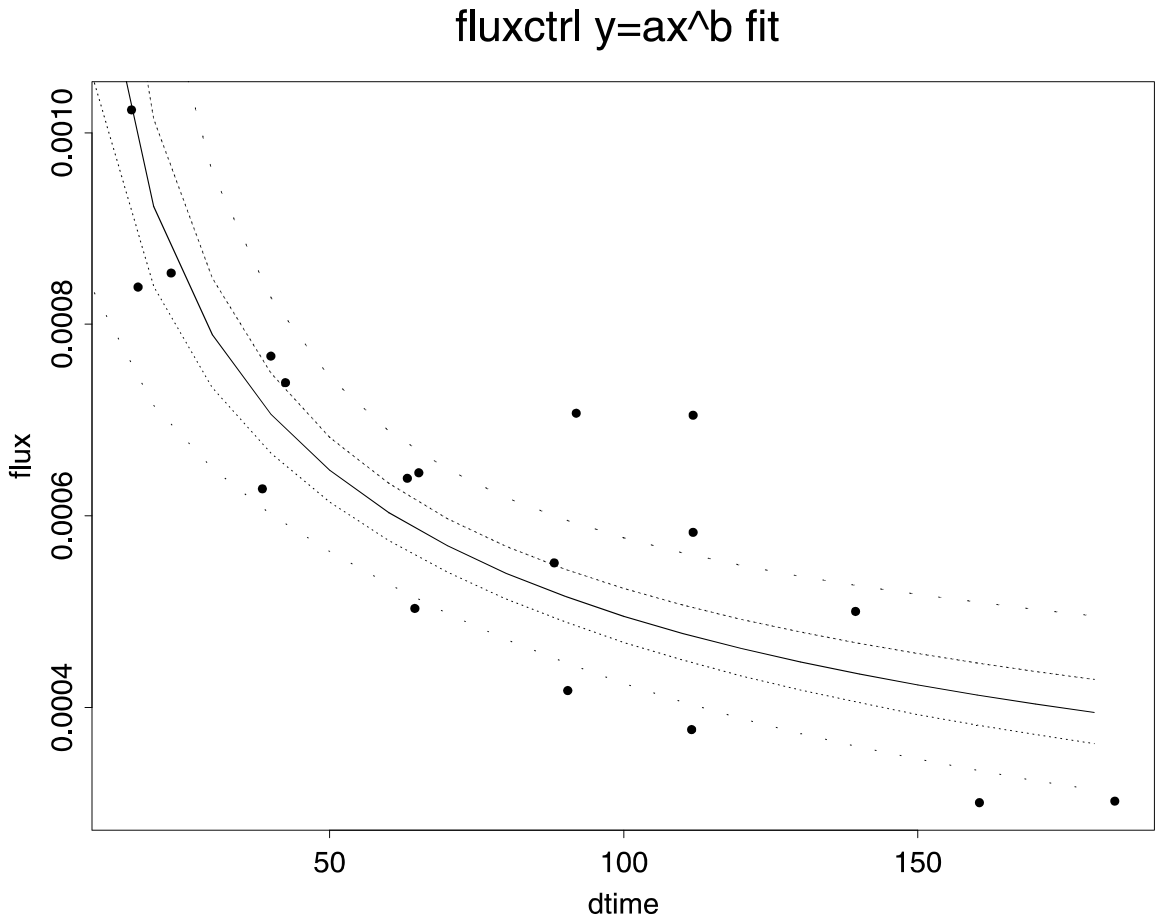


Figure 25. Flux data using the control treatment (no treatment). Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

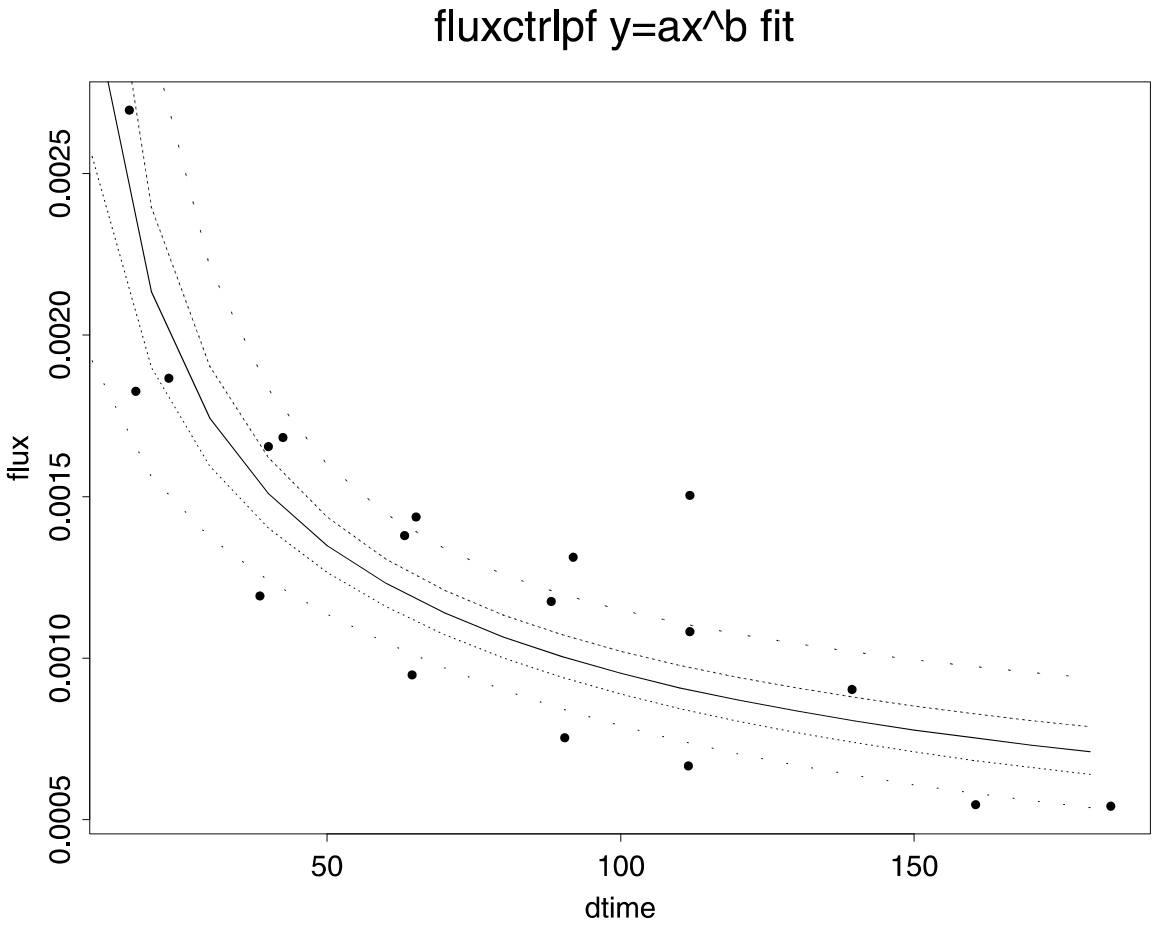


Figure 26. Flux data using post filtration only. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

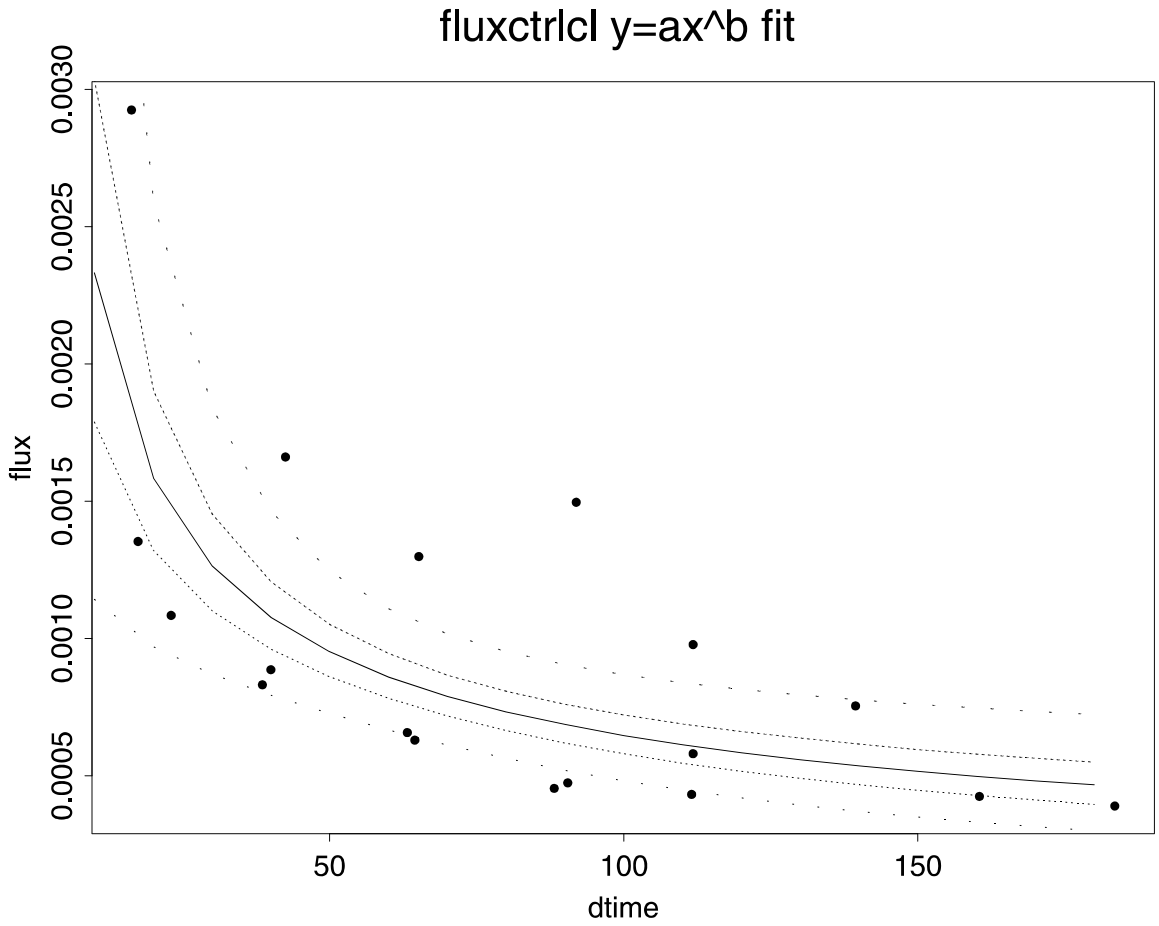


Figure 27. Flux data using prechlorination only. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

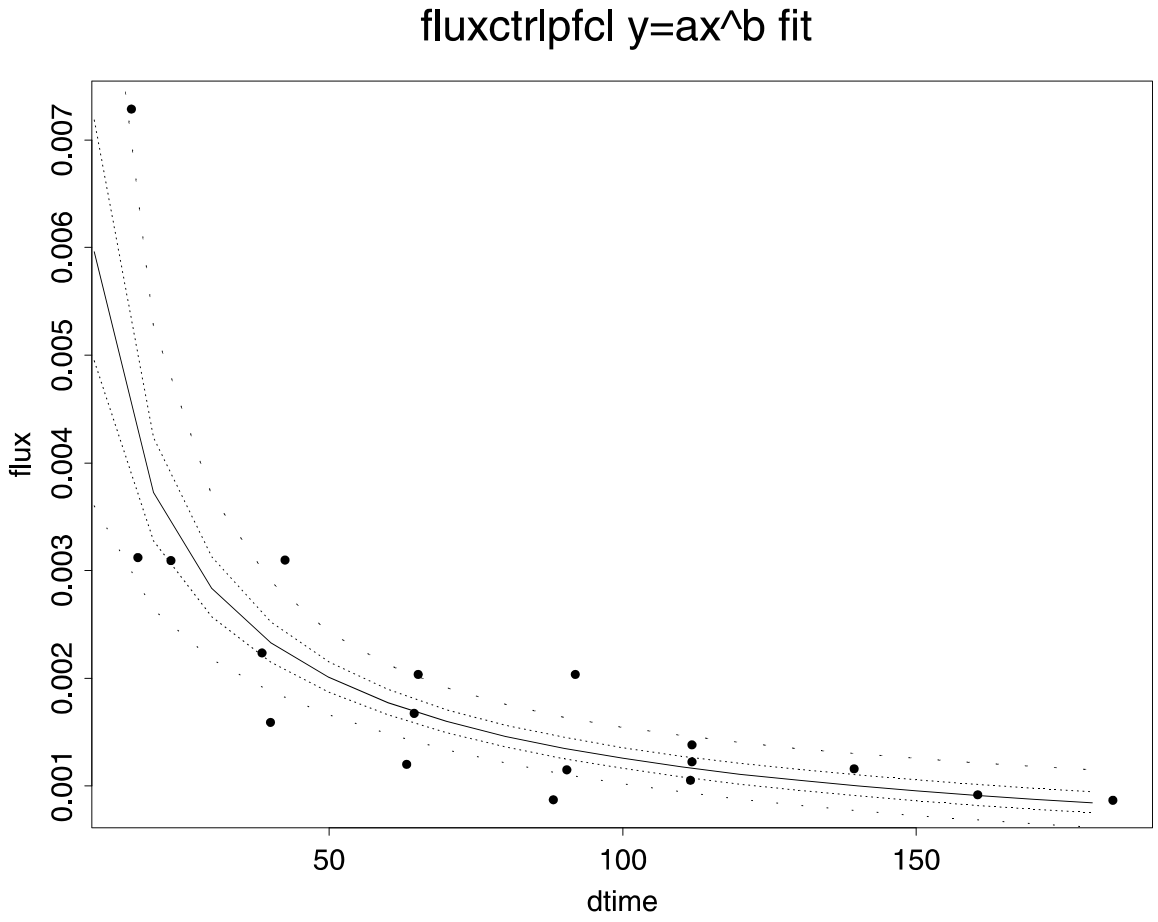


Figure 28. Flux data using post filtration with prechlorination. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

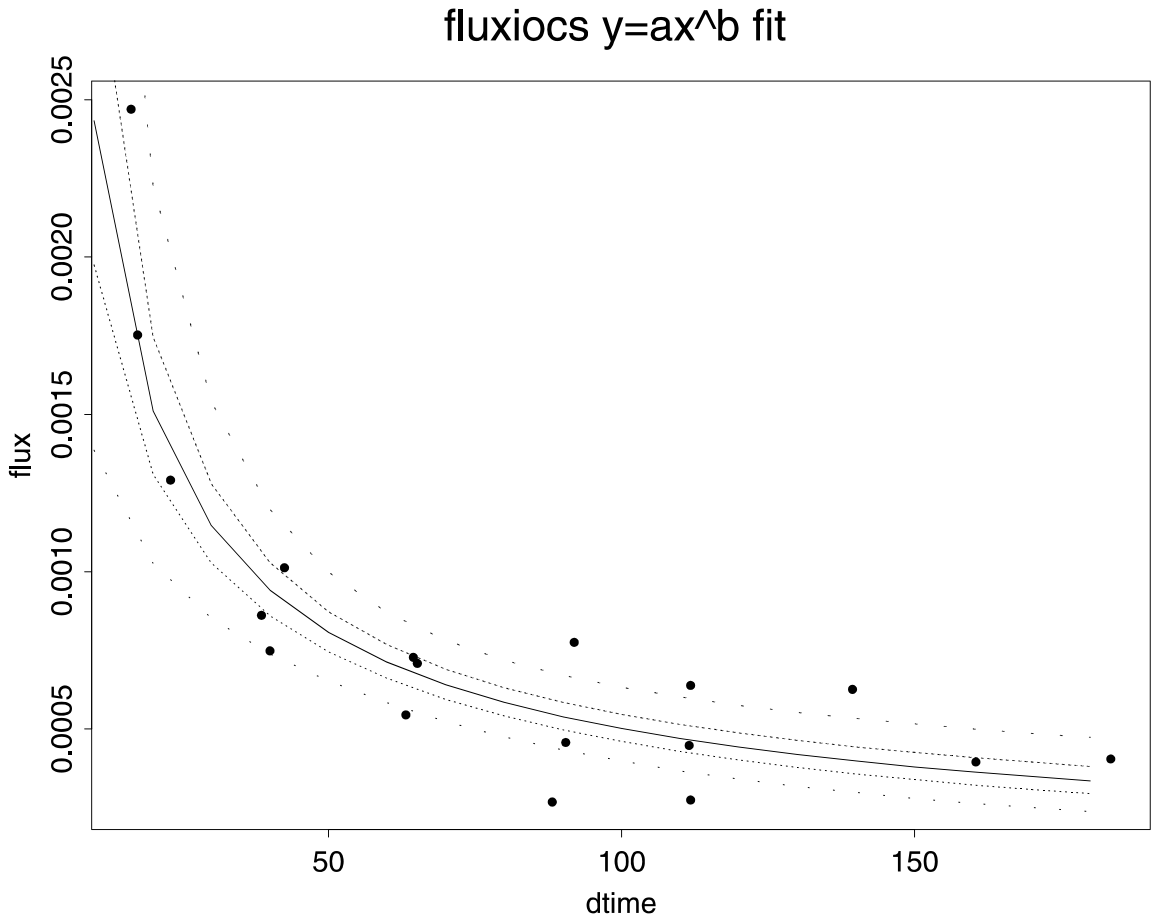


Figure 29. Flux data using iron-oxide coated sand pretreatment. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

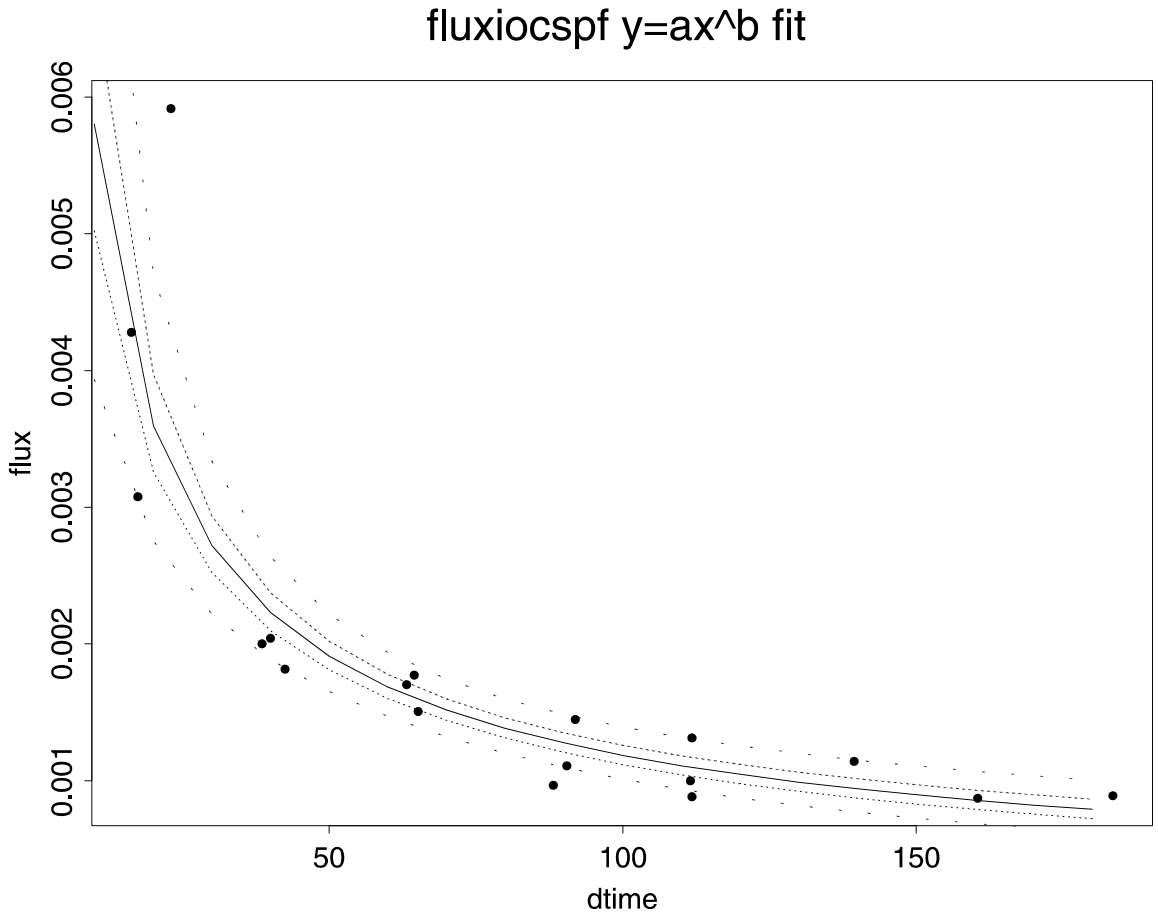


Figure 30. Flux data using iron-oxide coated sand pretreatment with post filtration. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

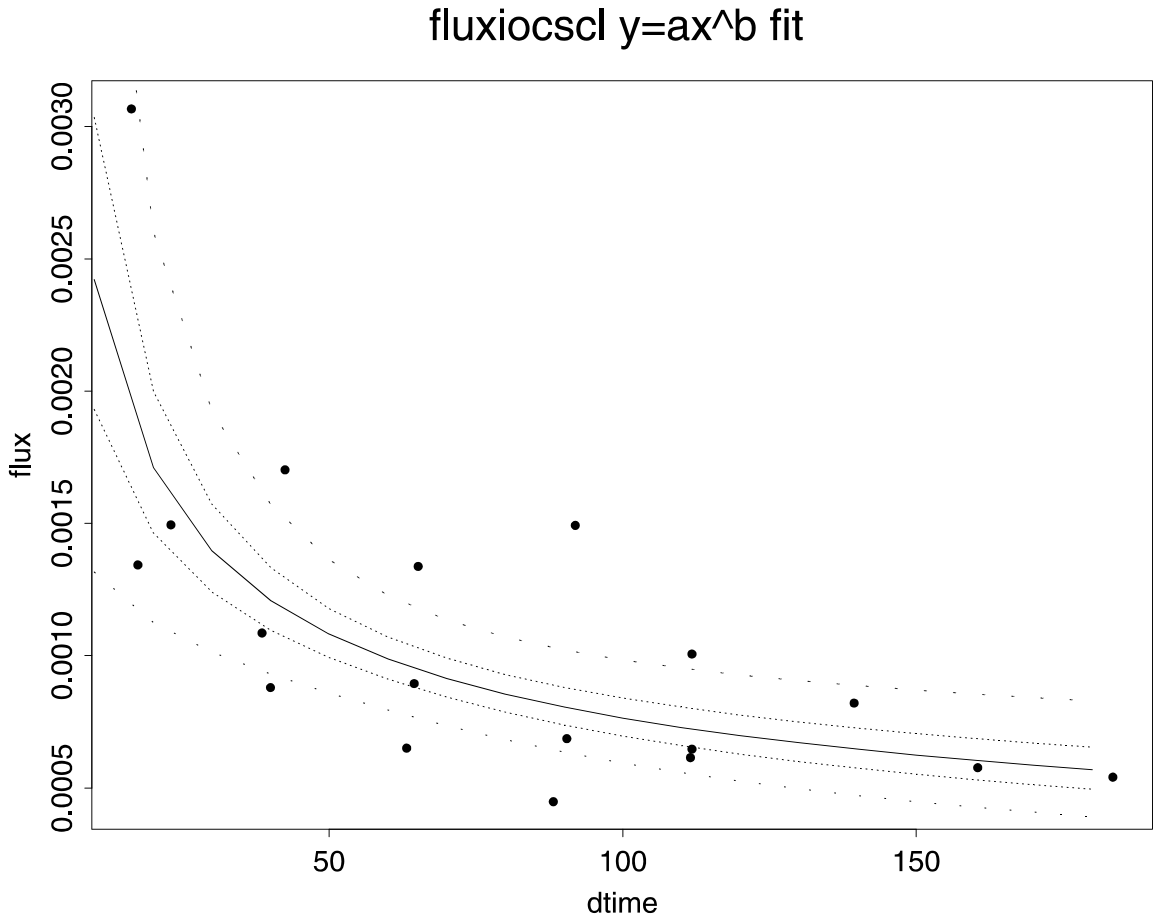


Figure 31. Flux data using iron-oxide coated sand pretreatment with prechlorination. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

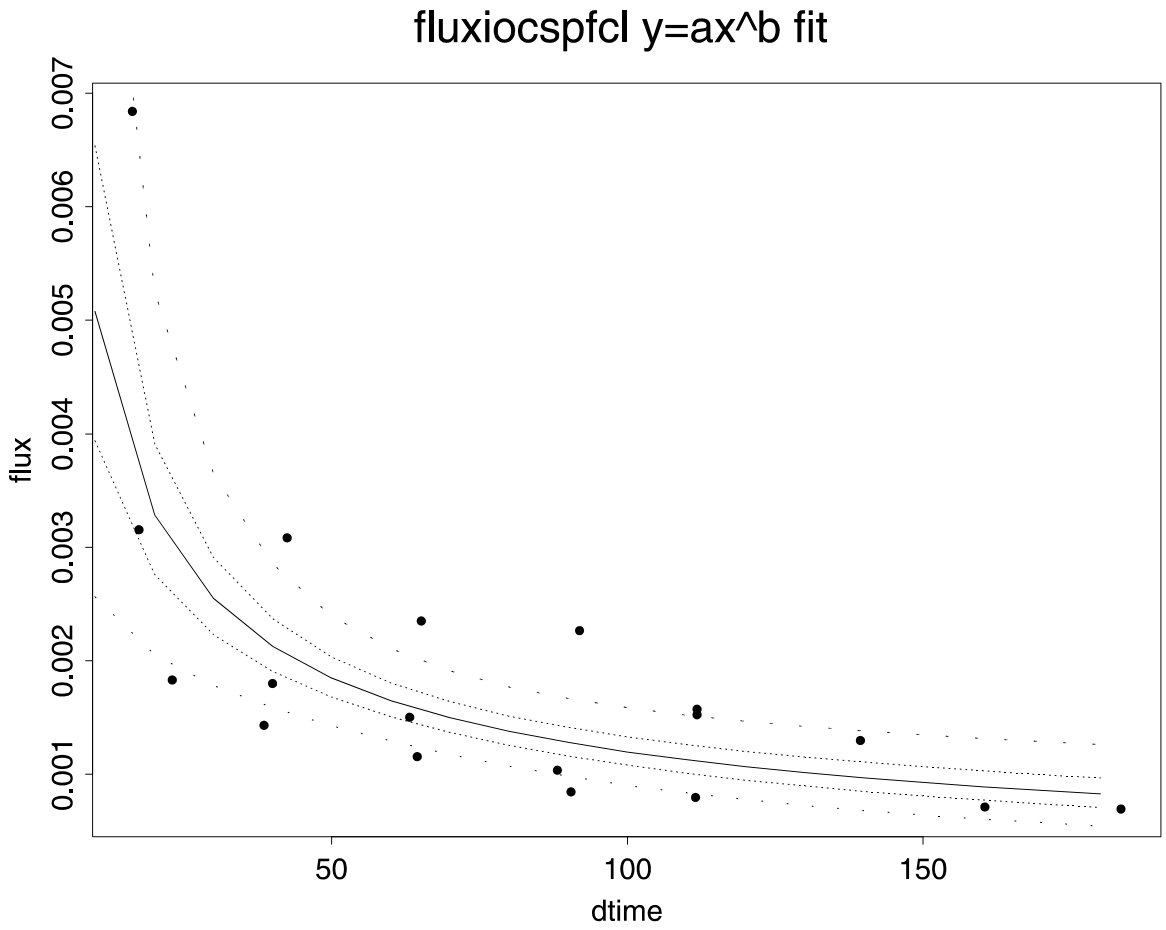


Figure 32. Flux data using iron-oxide coated sand pretreatment with post filtration and prechlorination. Flux has units of mL/(mm^2 min) and dtime is the test run time in hours.

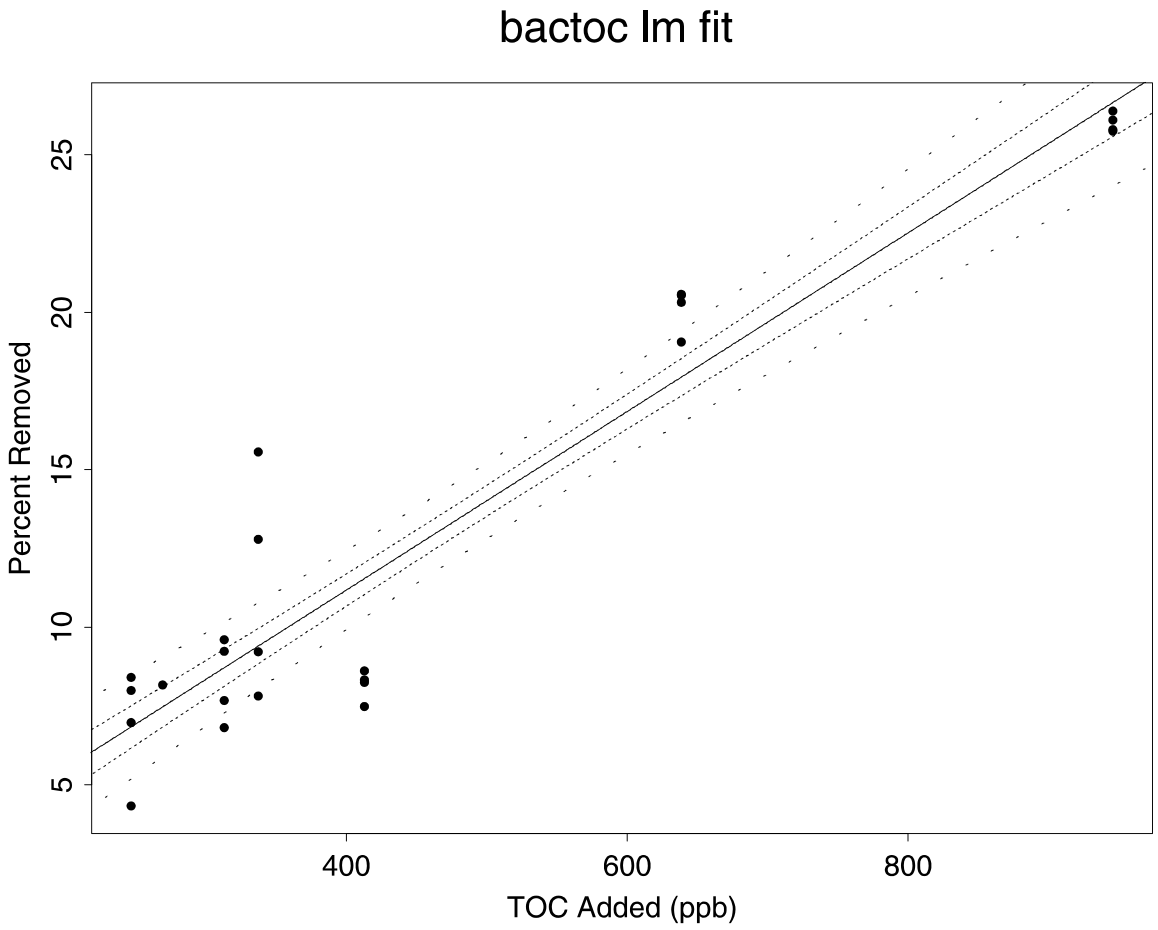

bactoc lm fit
Percent Removed
5
10
15
20
25
400
600
800
TOC Added (ppb)

Figure 33.

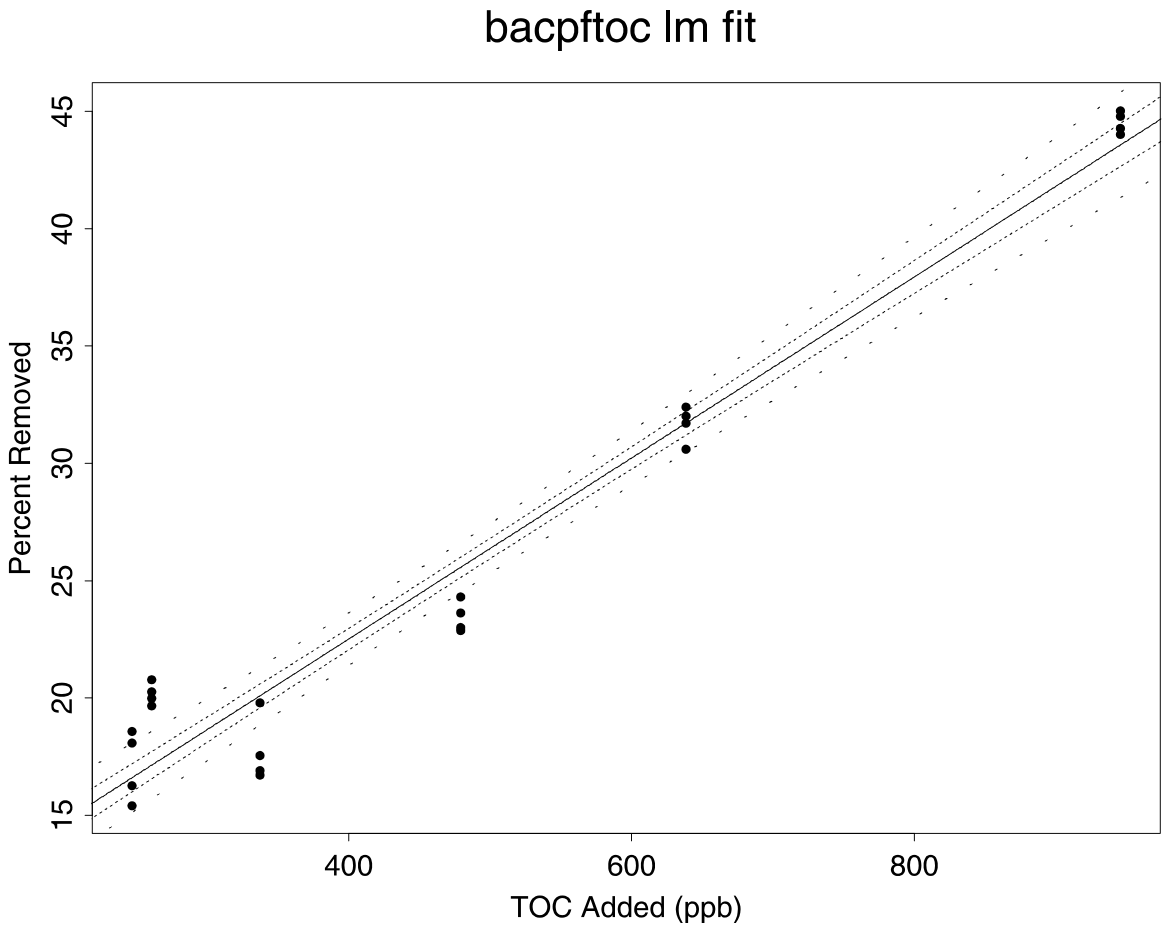

bacpftoc lm fit
Percent Removed
15
20
25
30
35
40
45
400
600
800
TOC Added (ppb)

Figure 34.

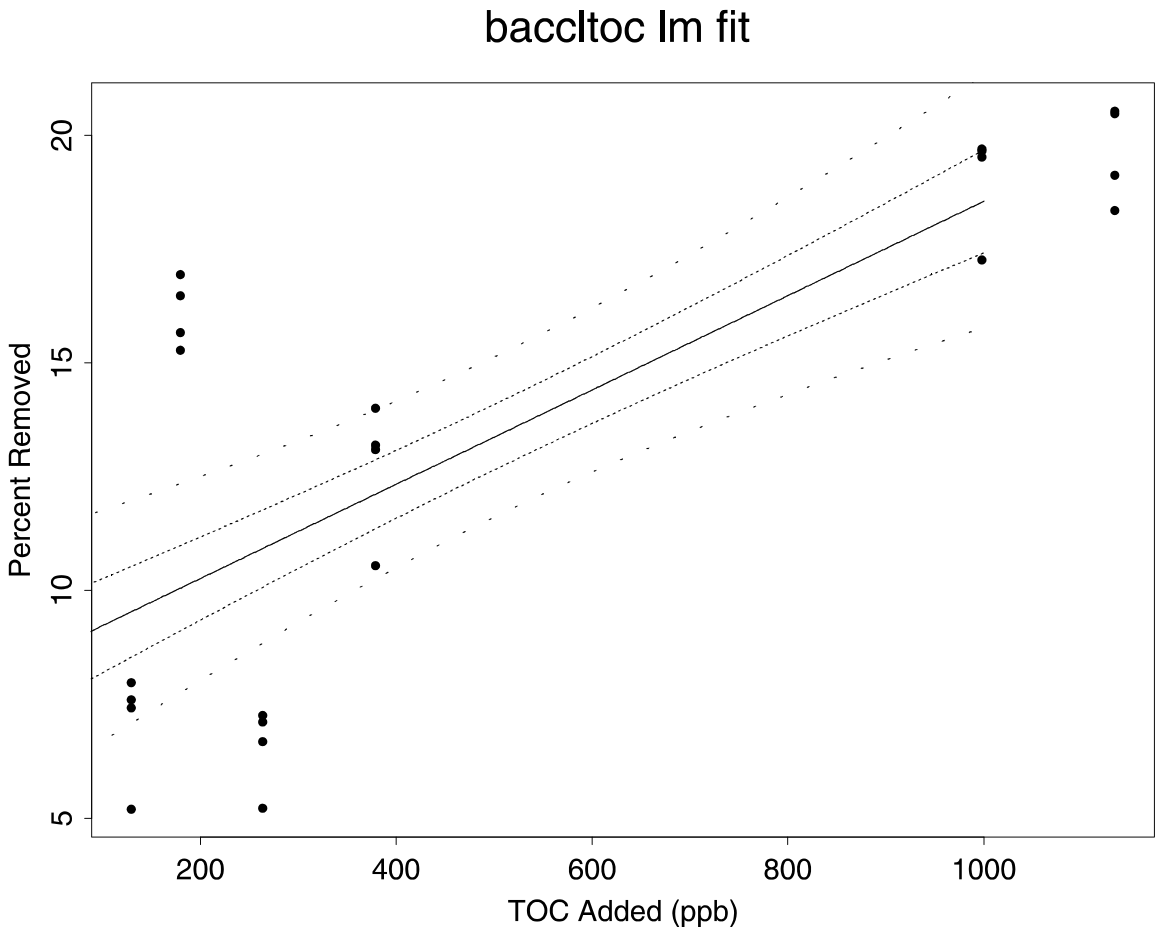

baccltoc lm fit
Percent Removed
TOC Added (ppb)
5
10
15
20
200
400
600
800
1000

Figure 35.

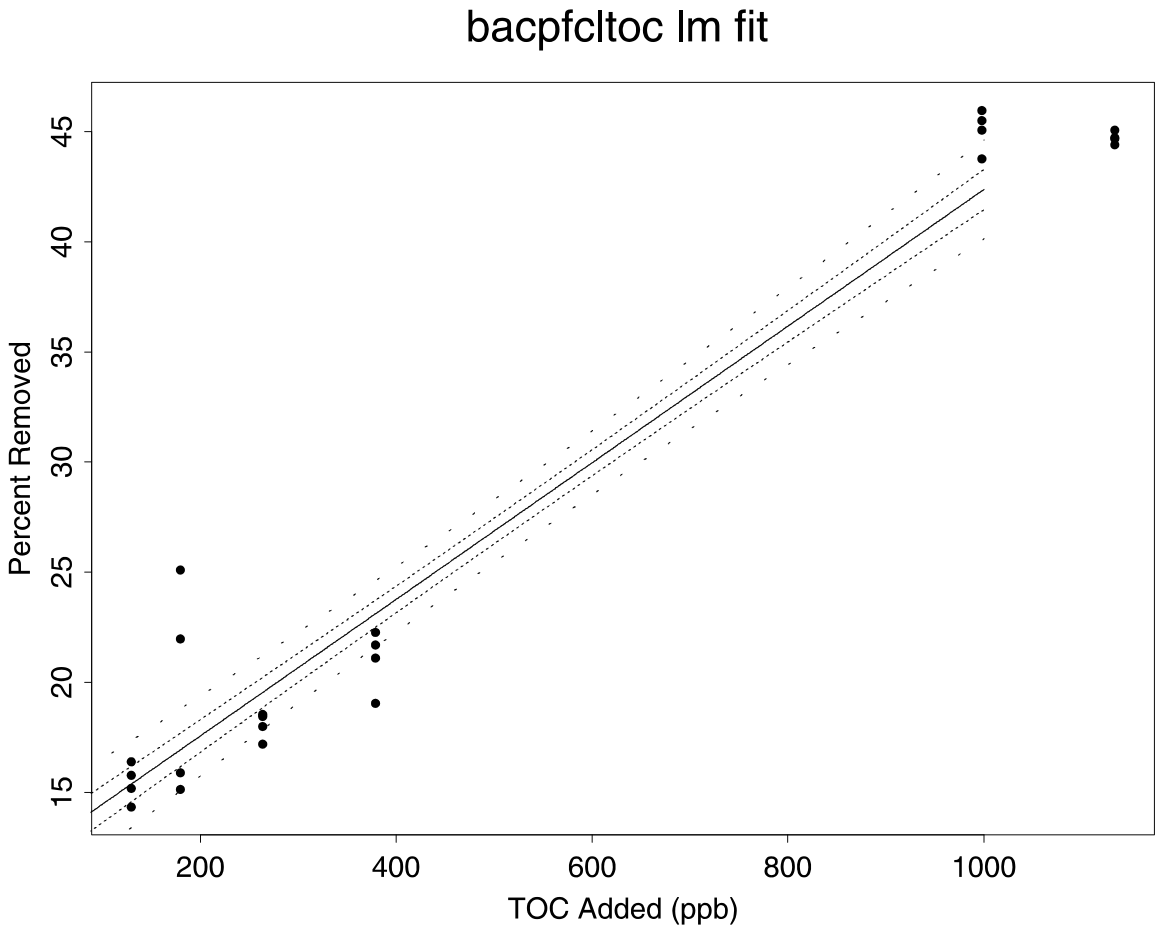

bacpfcltoc lm fit
Percent Removed
15
20
25
30
35
40
45
200
400
600
800
1000
TOC Added (ppb)

Figure 36.

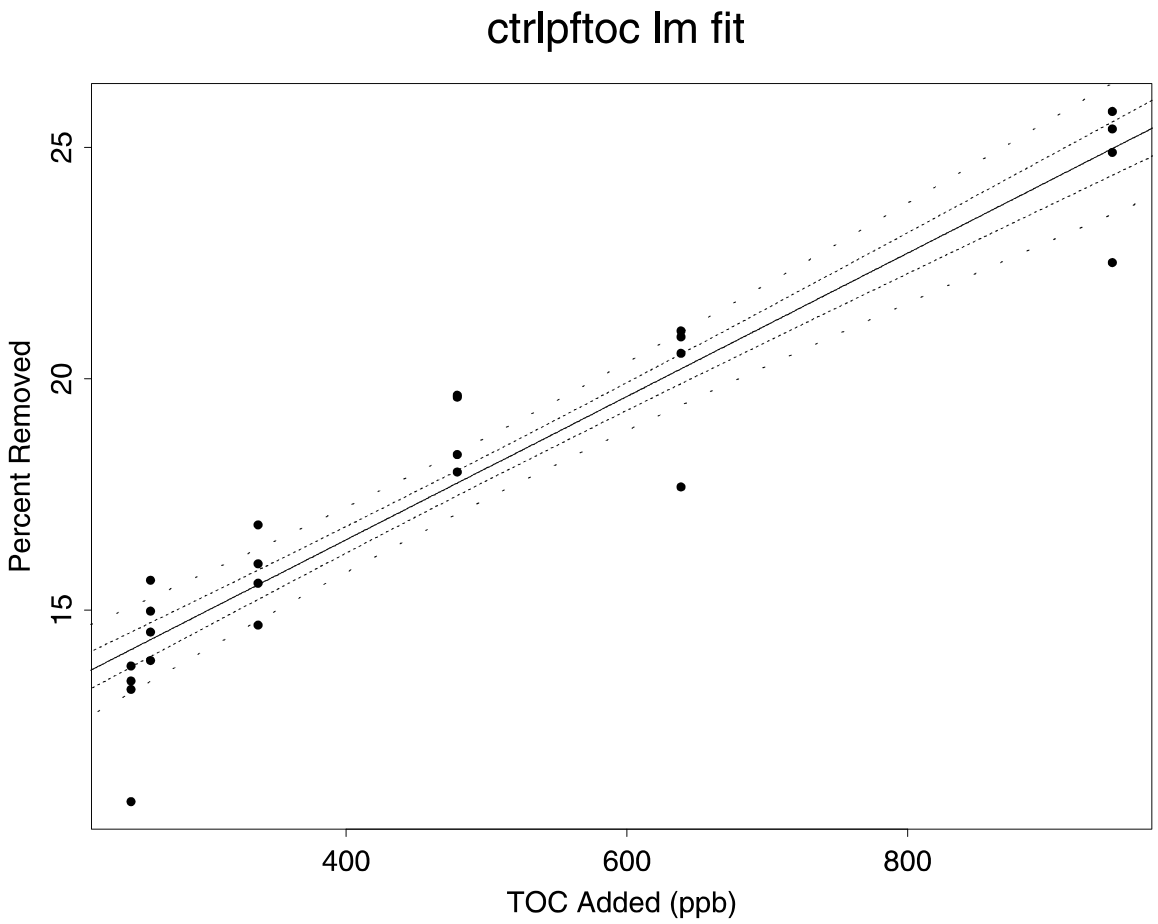

ctrlpftoc lm fit
Percent Removed
15
20
25
400
600
800
TOC Added (ppb)

Figure 37.

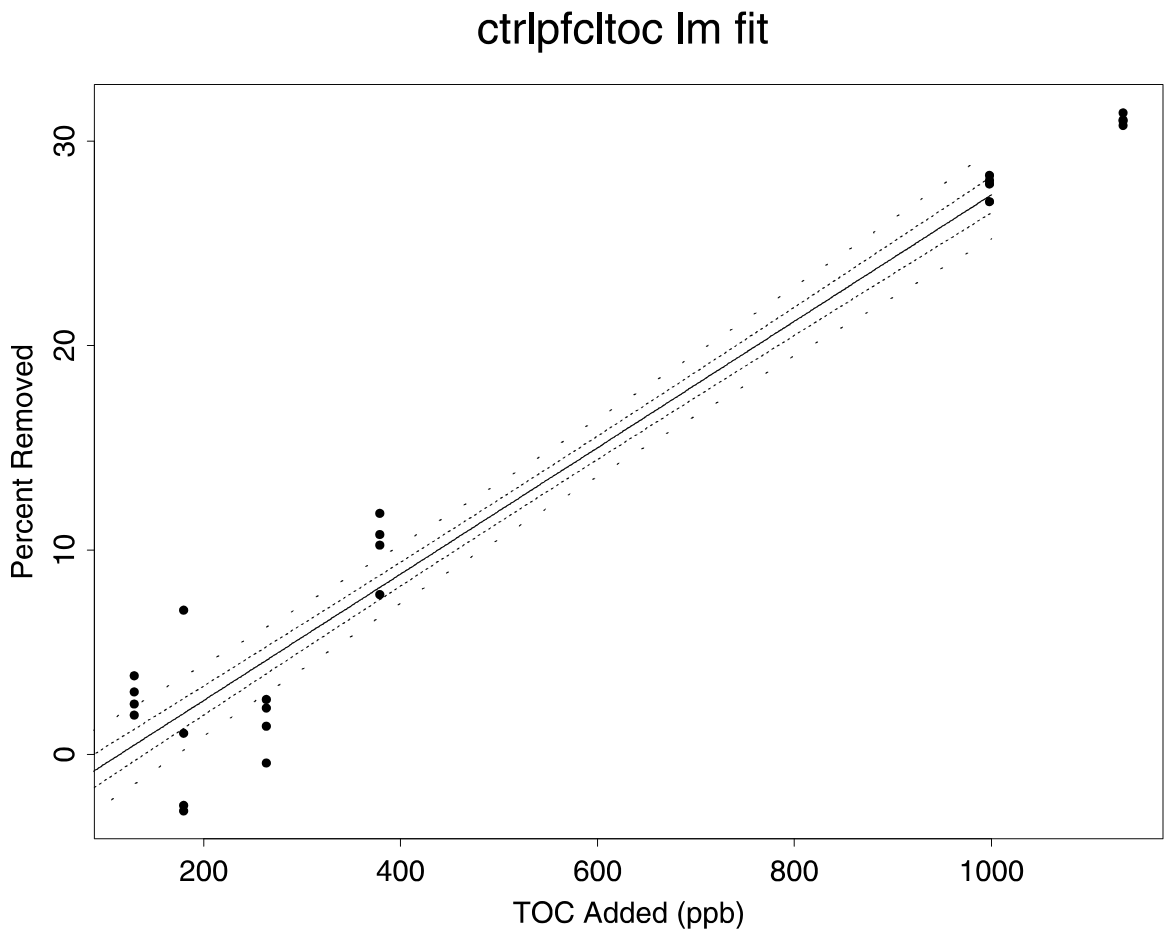

ctrlpfcltoc lm fit
Percent Removed
0
10
20
30
200
400
600
800
1000
TOC Added (ppb)

Figure 38.

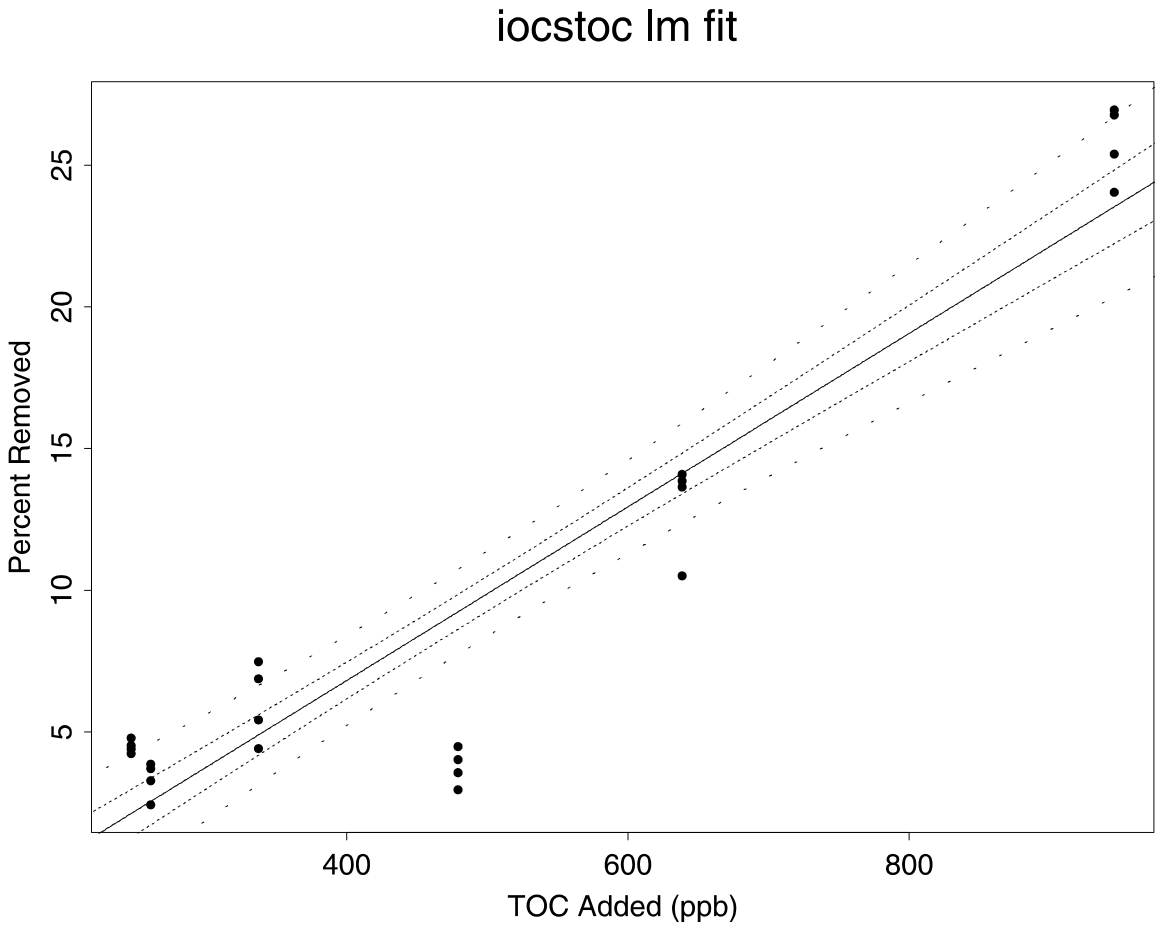

iocstoc lm fit
Percent Removed
5
10
15
20
25
400
600
800
TOC Added (ppb)

Figure 39.

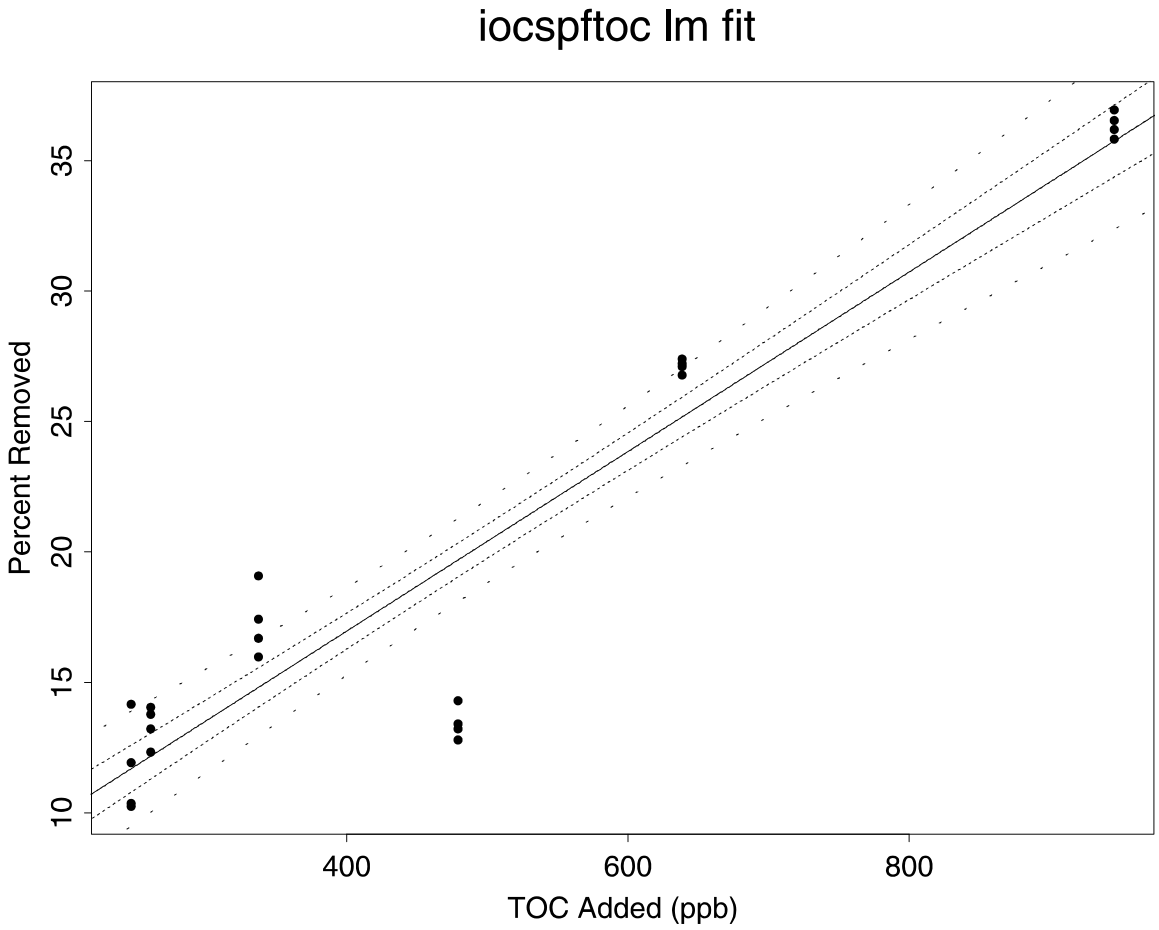

iocspftoc lm fit
Percent Removed
10
15
20
25
30
35
400
600
800
TOC Added (ppb)

Figure 40.

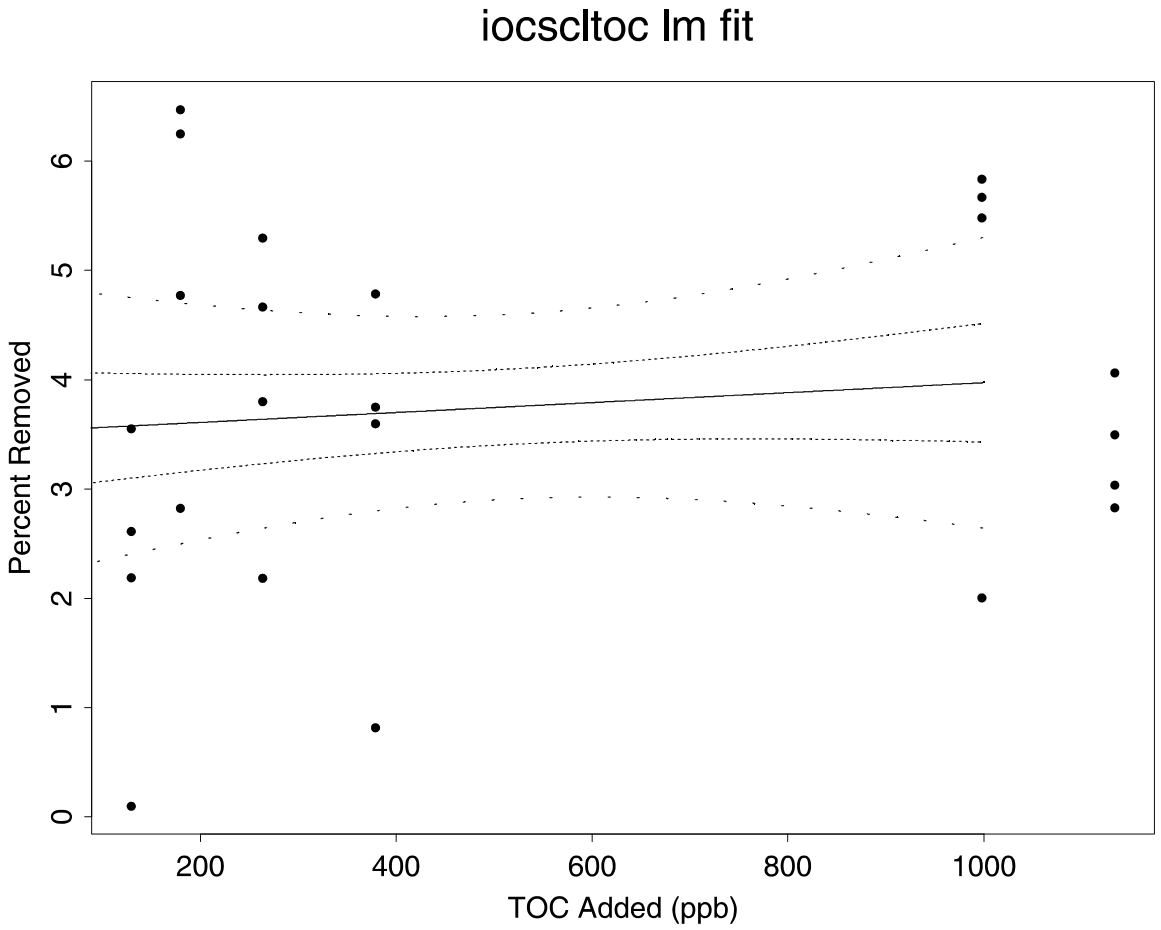

iocscltoc lm fit
Percent Removed
0
1
2
3
4
5
6
200
400
600
800
1000
TOC Added (ppb)

Figure 41.

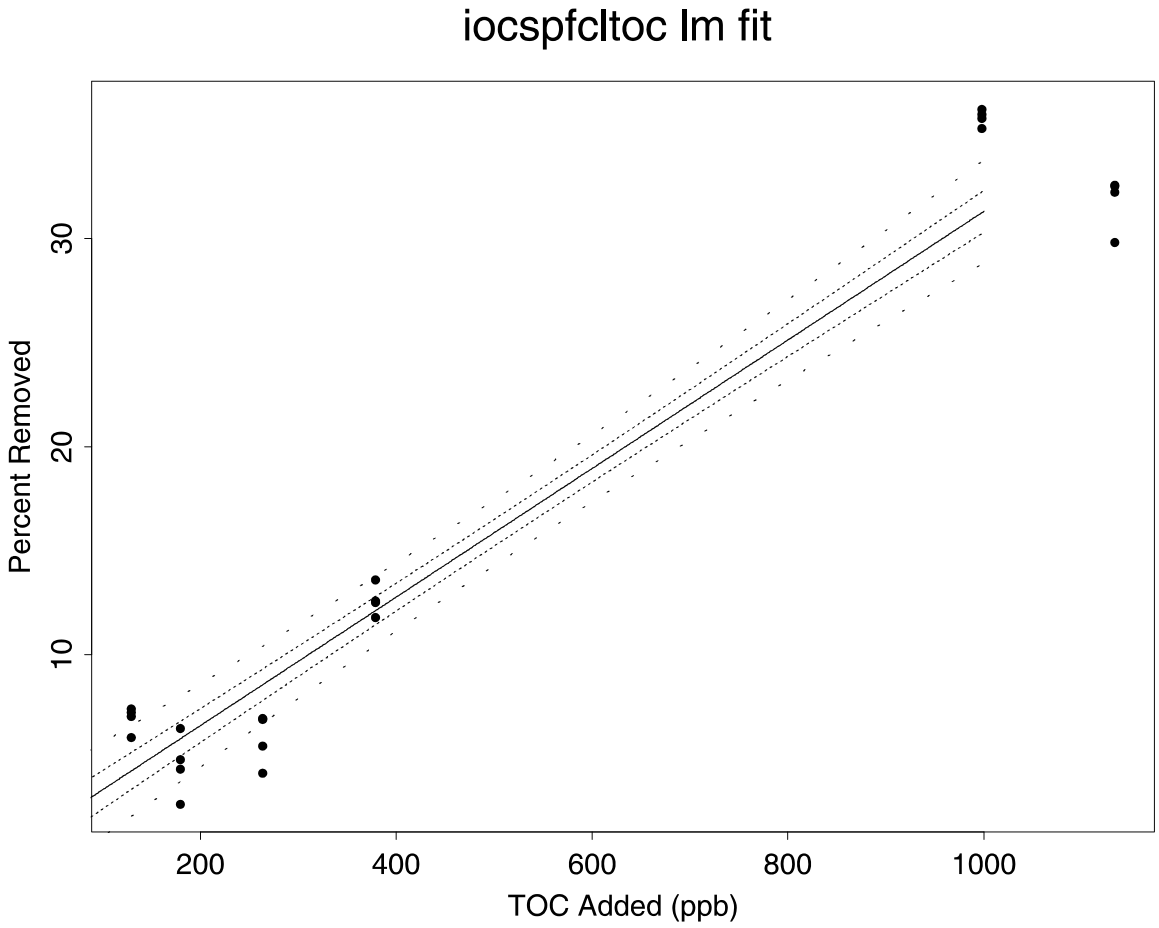

iocspfcltoc lm fit
Percent Removed
10
20
30
200
400
600
800
1000
TOC Added (ppb)

Figure 42.

Appendix 3 - Data Record

Table 9. Master summary table.
trtmnt: treatment train
flux: membrane flux at 120 hours - ml/(min*mm^2)
cell: membrane fouling layer log(cells/mm^2)
sem: fouling layer thickness measured with scanning electron microscope (microns)
dapi: fouling layer thickness measured with thin section and DAPI stain (microns)
beadassay: biofilm cells on downstream glass beads log(cells/cm^2)

trtmnt	flux	cell	sem	dapi	toc	beadassay
bac	0.000416	7.4298	6.589	6.34	20.49697	7.4257
bac	0.000501	7.6303	10.514	11.238	22.51709	7.6957
bac	0.000604	7.8308	14.439	16.138	24.5372	7.9657
bacpf	0.001718	6.8475	6.063	3.68	36.2204	5.8197
bacpf	0.002201	7.048	9.988	8.577	37.93932	6.0897
bacpf	0.002819	7.2485	13.913	13.477	39.65825	6.3597
iocspf	0.000872	6.7075	4.602	3.48	28.13967	5.3097
iocspf	0.001045	6.908	8.527	8.376	30.72282	5.5797
iocspf	0.001252	7.1085	12.452	13.276	33.30597	5.8497
iocs	0.000341	7.4038	6.063	7.57	16.6208	6.7407
iocs	0.000443	7.6043	9.988	12.473	19.05596	7.0107
iocs	0.000575	7.8048	13.913	17.373	21.49112	7.2807
ctrl	0.000388	7.7585	17.87	38.82	0	7.7957
ctrl	0.000461	7.959	21.795	43.724	0	8.0657
ctrl	0.000548	8.1595	25.72	48.624	0	8.3357
ctrlpf	0.000705	7.1215	5.262	14.68	21.63263	6.521
ctrlpf	0.00087	7.322	9.187	19.583	22.71544	6.791
ctrlpf	0.001074	7.5225	13.112	24.483	23.79824	7.061
iocscl	0.000526	7.4102	7.672	5.79	2.842262	5.8923
iocscl	0.000698	7.6107	11.597	10.685	3.882449	6.1623
iocscl	0.000927	7.8112	15.522	15.585	4.922636	6.4323
iocspfcl	0.000777	6.8525	1.711	7.19	23.18836	5.5017
iocspfcl	0.001068	7.053	5.636	12.092	25.1215	5.7717
iocspfcl	0.001468	7.2535	9.561	16.992	27.05463	6.0417
bacpfcl	0.001396	6.8558	3.044	6.43	34.41073	5.597
bacpfcl	0.00188	7.0563	6.969	11.332	36.16253	5.867
bacpfcl	0.002532	7.2568	10.894	16.232	37.91432	6.137
baccl	0.000583	7.3305	15.015	8.72	14.30745	6.2513
baccl	0.000728	7.531	18.94	13.622	16.47266	6.5213
baccl	0.000908	7.7315	22.865	18.522	18.63788	6.7913
ctrlcl	0.000419	7.8802	27.307	33.98	0	6.8563
ctrlcl	0.000584	8.0807	31.232	38.882	0	7.1263
ctrlcl	0.000814	8.2812	35.157	43.782	0	7.3963
ctrlpfcl	0.000878	7.1605	3.598	8.51	19.49634	6.5583
ctrlpfcl	0.00111	7.361	7.523	13.411	21.1862	6.8283
ctrlpfcl	0.001403	7.5615	11.448	18.311	22.87624	7.0983

Table 10. BAC TOC removal.
Non-purgeable organic carbon (NPOC) removal
ppb - amount of NPOC added to the feed water
rem - percent of NPOC removed by the treatment
bac1 - BAC experiment one
bac2 - BAC experiment two
bac3 - BAC experiment three

trtmnt	ppb	rem
bac1	412.86	7.4794
bac1	412.86	8.249
bac1	412.86	8.3351
bac1	412.86	8.609
bac1	269.15	8.17
bac1	313.03	7.675
bac1	313.03	9.2371
bac1	313.03	9.6046
bac1	313.03	6.8149
bac2	337.36	12.7943
bac2	337.36	7.8123
bac2	337.36	9.2162
bac2	337.36	15.5611
bac2	246.69	4.3342
bac2	246.69	6.9707
bac2	246.69	7.9912
bac2	246.69	8.4132
bac3	945.91	26.3923
bac3	945.91	26.0985
bac3	945.91	25.7919
bac3	945.91	25.7504
bac3	638.38	19.0638
bac3	638.38	20.5635
bac3	638.38	20.3211
bac3	638.38	20.5446

Table 11. BACPF TOC removal.
bacpf1 - BACPF experiment 1
bacpf2 - BACPF experiment 2
bacpf3 - BACPF experiment 3

trtmnt	ppb	rem
bacpf1	260.6	19.6624
bacpf1	260.6	19.9859
bacpf1	260.6	20.2572
bacpf1	260.6	20.7685
bacpf1	479.08	22.8771
bacpf1	479.08	23.6198
bacpf1	479.08	22.9996
bacpf1	479.08	24.3115
bacpf2	337.35	19.8016
bacpf2	337.35	16.8954
bacpf2	337.35	16.7179
bacpf2	337.35	17.5387
bacpf2	246.69	16.2703
bacpf2	246.69	18.076
bacpf2	246.69	15.4133
bacpf2	246.69	18.5699
bacpf3	945.91	44.0158
bacpf3	945.91	44.7822
bacpf3	945.91	44.2745
bacpf3	945.91	45.0281
bacpf3	638.38	30.6029
bacpf3	638.38	31.7163
bacpf3	638.38	32.0117
bacpf3	638.38	32.4017

Table 12. BACPFCL TOC removal.
bacpfcl1 - BACPFCL experiment 1
bacpfcl2 - BACPFCL experiment 2
bacpfcl3 - BACPFCL experiment 3

trtmnt	ppb	rem
bacpfcl1	129.49	15.7595
bacpfcl1	129.49	14.3402
bacpfcl1	129.49	16.3781
bacpfcl1	129.49	15.1658
bacpfcl1	379	19.0405
bacpfcl1	379	21.0958
bacpfcl1	379	21.6948
bacpfcl1	379	22.2565
bacpfcl2	179.91	15.1263
bacpfcl2	179.91	21.9656
bacpfcl2	179.91	25.0853
bacpfcl2	179.91	15.8847
bacpfcl2	263.83	17.1875
bacpfcl2	263.83	18.4388
bacpfcl2	263.83	18.536
bacpfcl2	263.83	17.985
bacpfcl3	998.2	43.7673
bacpfcl3	998.2	45.0577
bacpfcl3	998.2	45.5116
bacpfcl3	998.2	45.9625
bacpfcl3	1133.86	44.3944
bacpfcl3	1133.86	44.6763
bacpfcl3	1133.86	44.717
bacpfcl3	1133.86	45.0599

Table 13. BACCL TOC removal.
baccl1 - BACCL experiment one
baccl2 - BACCL experiment two
baccl3 - BACCL experiment three

trtmnt	ppb	rem
baccl1	129.49	5.2026
baccl1	129.49	7.9833
baccl1	129.49	7.42
baccl1	129.49	7.605
baccl1	379	10.5426
baccl1	379	13.1968
baccl1	379	13.0904
baccl1	379	14.0062
baccl2	179.91	15.2815
baccl2	179.91	16.4776
baccl2	179.91	15.6641
baccl2	179.91	16.943
baccl2	263.83	5.2224
baccl2	263.83	6.6844
baccl2	263.83	7.1123
baccl2	263.83	7.2549
baccl3	998.2	17.26
baccl3	998.2	19.6573
baccl3	998.2	19.6978
baccl3	998.2	19.5205
baccl3	1133.86	18.3483
baccl3	1133.86	20.5306
baccl3	1133.86	19.127
baccl3	1133.86	20.4725

Table 14. CTRLPF TOC removal.
ctrlpf1 - CTRLPF experiment 1
ctrlpf2 - CTRLPF experiment 2
ctrlpf3 - CTRLPF experiment 3

trtmnt	ppb	rem
ctrlpf1	260.61	13.9205
ctrlpf1	260.61	14.5283
ctrlpf1	260.61	14.9797
ctrlpf1	260.61	15.6501
ctrlpf1	479.08	17.9918
ctrlpf1	479.08	19.6023
ctrlpf1	479.08	19.6457
ctrlpf1	479.08	18.3644
ctrlpf2	337.35	15.5801
ctrlpf2	337.35	14.6769
ctrlpf2	337.35	16.8447
ctrlpf2	337.35	16.0048
ctrlpf2	246.69	10.8698
ctrlpf2	246.69	13.7974
ctrlpf2	246.69	13.2904
ctrlpf2	246.69	13.4768
ctrlpf3	945.91	22.5125
ctrlpf3	945.91	24.8978
ctrlpf3	945.91	25.4055
ctrlpf3	945.91	25.7823
ctrlpf3	638.38	17.6702
ctrlpf3	638.38	20.5559
ctrlpf3	638.38	20.9081
ctrlpf3	638.38	21.0331

Table 15. CTRLPFCL TOC removal.
ctrlpfcl1 - CTRLPFCL experiment 1
ctrlpfcl2 - CTRLPFCL experiment 2
ctrlpfcl3 - CTRLPFCL experiment 3

trtmnt	ppb	rem
ctrlpfcl1	129.49	2.4742
ctrlpfcl1	129.49	3.0569
ctrlpfcl1	129.49	3.8522
ctrlpfcl1	129.49	1.9275
ctrlpfcl1	379	7.8271
ctrlpfcl1	379	10.2284
ctrlpfcl1	379	10.7476
ctrlpfcl1	379	11.8071
ctrlpfcl2	179.91	-2.7541
ctrlpfcl2	179.91	-2.4815
ctrlpfcl2	179.91	7.058
ctrlpfcl2	179.91	1.0395
ctrlpfcl2	263.83	-0.4182
ctrlpfcl2	263.83	2.2724
ctrlpfcl2	263.83	1.3842
ctrlpfcl2	263.83	2.6971
ctrlpfcl3	998.2	27.0483
ctrlpfcl3	998.2	27.8971
ctrlpfcl3	998.2	28.0744
ctrlpfcl3	998.2	28.3356
ctrlpfcl3	1133.86	30.7625
ctrlpfcl3	1133.86	30.9834
ctrlpfcl3	1133.86	31.0473
ctrlpfcl3	1133.86	31.3931

Table 16. IOCSCL TOC removal.
iocscl1 - IOCSCL experiment 1
iocscl2 - IOCSCL experiment 2
iocscl3 - IOCSCL experiment 3

trtmnt	ppb	rem
iocscl1	129.49	2.6114
iocscl1	129.49	0.0991
iocscl1	129.49	3.5506
iocscl1	129.49	2.1862
iocscl1	379	0.8142
iocscl1	379	3.7495
iocscl1	379	3.5963
iocscl1	379	4.7832
iocscl2	179.91	2.8238
iocscl2	179.91	4.7698
iocscl2	179.91	6.2498
iocscl2	179.91	6.4717
iocscl2	263.83	2.1817
iocscl2	263.83	4.6647
iocscl2	263.83	3.7975
iocscl2	263.83	5.2976
iocscl3	998.2	2.0022
iocscl3	998.2	5.668
iocscl3	998.2	5.4828
iocscl3	998.2	5.8373
iocscl3	1133.86	2.8298
iocscl3	1133.86	3.4962
iocscl3	1133.86	4.0622
iocscl3	1133.86	3.0368

Table 17. IOCSPFCL TOC removal.
iocspfcl1 - IOCSPFCL experiment 1
iocspfcl2 - IOCSPFCL experiment 2
iocspfcl3 - IOCSPFCL experiment 3

trtmnt	ppb	rem
iocspfcl1	129.49	6.0117
iocspfcl1	129.49	7.2294
iocspfcl1	129.49	7.3869
iocspfcl1	129.49	7.0196
iocspfcl1	379	11.7779
iocspfcl1	379	12.51
iocspfcl1	379	12.5819
iocspfcl1	379	13.5962
iocspfcl2	179.91	4.4917
iocspfcl2	179.91	2.8095
iocspfcl2	179.91	6.4394
iocspfcl2	179.91	4.9467
iocspfcl2	263.83	4.2888
iocspfcl2	263.83	6.8756
iocspfcl2	263.83	6.9178
iocspfcl2	263.83	5.5952
iocspfcl3	998.2	35.2881
iocspfcl3	998.2	35.7669
iocspfcl3	998.2	36.2178
iocspfcl3	998.2	35.9597
iocspfcl3	1133.86	29.8065
iocspfcl3	1133.86	32.5148
iocspfcl3	1133.86	32.2388
iocspfcl3	1133.86	32.5758

Table 18. IOCS TOC removal.
iocs1 - IOCS experiment 1
iocs2 - IOCS experiment 2
iocs3 - IOCS experiment 3

trtmnt	ppb	rem
iocs1	260.61	2.4418
iocs1	260.61	3.2792
iocs1	260.61	3.7175
iocs1	260.61	3.8767
iocs1	479.08	2.9582
iocs1	479.08	3.5606
iocs1	479.08	4.0353
iocs1	479.08	4.4922
iocs2	337.35	4.4211
iocs2	337.35	5.4321
iocs2	337.35	6.871
iocs2	337.35	7.4795
iocs2	246.69	4.4029
iocs2	246.69	4.2393
iocs2	246.69	4.7954
iocs2	246.69	4.5239
iocs3	945.91	24.0452
iocs3	945.91	25.3832
iocs3	945.91	26.7691
iocs3	945.91	26.9447
iocs3	638.38	10.5052
iocs3	638.38	13.6408
iocs3	638.38	14.0801
iocs3	638.38	13.8567

Table 19. IOCSPF TOC removal.
iocspf1 - IOCSPF experiment 1
iocspf2 - IOCSPF experiment 2
iocspf3 - IOCSPF experiment 3

trtmnt	ppb	rem
iocspf1	260.61	12.3317
iocspf1	260.61	13.2239
iocspf1	260.61	14.0405
iocspf1	260.61	13.7744
iocspf1	479.08	13.4179
iocspf1	479.08	12.7977
iocspf1	479.08	14.3036
iocspf1	479.08	13.2163
iocspf2	337.35	16.6925
iocspf2	337.35	19.0758
iocspf2	337.35	17.4278
iocspf2	337.35	15.9668
iocspf2	246.69	14.1703
iocspf2	246.69	10.2548
iocspf2	246.69	11.9296
iocspf2	246.69	10.3693
iocspf3	945.91	36.1892
iocspf3	945.91	35.8283
iocspf3	945.91	36.9428
iocspf3	945.91	36.5404
iocspf3	638.38	26.7818
iocspf3	638.38	27.3915
iocspf3	638.38	27.0961
iocspf3	638.38	27.2249

Table 20. Membrane cell counts.
Membrane fouling layer cell counts. The numbers are the log(cells/mm^2).

trtmnt	cellsmm2	trtmnt	cellsmm2	trtmnt	cellsmm2	trtmnt	cellsmm2
bac	7.83	IocsCl	8.05	CtrlPf	7.3	BacPfCl	6.8
bac	7.8	IocsCl	8.36	CtrlPf	7.3	BacPfCl	6.8
bac	7.62	IocsCl	7.94	CtrlPf	7.15	BacPfCl	6.6
bac	7.92	IocsCl	8.32	CtrlPf	7.48	BacPfCl	6.91
bac	8.11	IocsCl	7.81	CtrlPf	7.68	BacPfCl	7.04
bac	8.18	IocsCl	8.18	CtrlPf	7.53	BacPfCl	6.84
bac	7.97	IocsCl	7.61	CtrlPf	7.45	BacPfCl	7.24
bac	7.72	IocsCl	8.35	CtrlPf	7.08	BacPfCl	6.76
bac	7.89	IocsCl	7.88	CtrlPf	7.65	BacPfCl	6.71
bac	7.62	IocsCl	8.07	CtrlPf	7.63	BacPfCl	6.91
baccl	7.59	IocsPf	6.93	CtrlPfCl	7.35	Ctrl	7.6
baccl	7.73	IocsPf	6.97	CtrlPfCl	7.35	Ctrl	7.6
baccl	7.7	IocsPf	6.97	CtrlPfCl	7.35	Ctrl	7.62
baccl	7.87	IocsPf	6.89	CtrlPfCl	7.41	Ctrl	7.74
baccl	7.96	IocsPf	7.03	CtrlPfCl	7.52	Ctrl	7.76
baccl	7.75	IocsPf	7.23	CtrlPfCl	7.38	Ctrl	7.64
baccl	7.63	IocsPf	7.17	CtrlPfCl	7.19	Ctrl	7.73
baccl	7.29	IocsPf	7	CtrlPfCl	7.41	Ctrl	7.59
baccl	7.72	IocsPf	7	CtrlPfCl	7.52	Ctrl	7.7
baccl	7.63	IocsPf	6.84	CtrlPfCl	7.41	Ctrl	7.85
BacPf	7.11	IocsPfCl	7.31	Iocs	7.6	CtrlCl	7.83
BacPf	6.99	IocsPfCl	7.24	Iocs	7.6	CtrlCl	7.83
BacPf	7.07	IocsPfCl	7.43	Iocs	7.6	CtrlCl	7.96
BacPf	7.42	IocsPfCl	7.61	Iocs	7.82	CtrlCl	7.72
BacPf	7.44	IocsPfCl	7.01	Iocs	7.55	CtrlCl	7.46
BacPf	7.22	IocsPfCl	7.13	Iocs	7.66	CtrlCl	7.8
BacPf	7.03	IocsPfCl	7.09	Iocs	7.85	CtrlCl	7.8
BacPf	7.2	IocsPfCl	7.31	Iocs	8.06	CtrlCl	7.76
BacPf	7.2	IocsPfCl	7.13	Iocs	7.96	CtrlCl	7.38
BacPf	7.29	IocsPfCl	6.79	Iocs	7.55	CtrlCl	7.94
BacPfCl	7.18	bac	7.63	IocsCl	7.45	CtrlPf	7.11
BacPfCl	7.12	bac	7.63	IocsCl	7.45	CtrlPf	7.11
BacPfCl	7.09	bac	7.54	IocsCl	7.38	CtrlPf	7.08
BacPfCl	6.88	bac	7.68	IocsCl	7.45	CtrlPf	6.71
BacPfCl	7.18	bac	7.41	IocsCl	7.56	CtrlPf	7.14
BacPfCl	7.27	bac	7.96	IocsCl	7.38	CtrlPf	7.11
BacPfCl	7.33	bac	7.68	IocsCl	7.72	CtrlPf	7.01
BacPfCl	7.01	bac	7.75	IocsCl	7.51	CtrlPf	6.84
BacPfCl	7.15	bac	7.71	IocsCl	7.93	CtrlPf	6.84
BacPfCl	7.2	bac	7.71	IocsCl	7.38	CtrlPf	7.28
Ctrl	8.21	baccl	7.67	IocsPf	7.21	CtrlPfCl	7.27
Ctrl	7.91	baccl	7.67	IocsPf	7.21	CtrlPfCl	7.27
Ctrl	7.96	baccl	7.34	IocsPf	7.32	CtrlPfCl	7.2

Ctrl	8.18	baccl	7.36	IocsPf	7.26	CtrlPfCl	7.11
Ctrl	8.09	baccl	7.64	IocsPf	7.36	CtrlPfCl	7.5
Ctrl	8.01	baccl	7.78	IocsPf	7.32	CtrlPfCl	7.42
Ctrl	8.12	baccl	7.67	IocsPf	7.21	CtrlPfCl	6.86
Ctrl	8.33	baccl	7.5	IocsPf	7.29	CtrlPfCl	7.52
Ctrl	7.96	baccl	7.46	IocsPf	7.06	CtrlPfCl	7.24
Ctrl	8.09	baccl	7.12	IocsPf	7.21	CtrlPfCl	6.86
CtrlCl	8.22	BacPf	7.12	IocsPfCl	7.2	Iocs	7.1
CtrlCl	8.59	BacPf	7.12	IocsPfCl	7.2	Iocs	7.1
CtrlCl	8.8	BacPf	7.08	IocsPfCl	7.34	Iocs	7.21
CtrlCl	8.48	BacPf	7.15	IocsPfCl	7.29	Iocs	7.42
CtrlCl	8.22	BacPf	7.08	IocsPfCl	7.23	Iocs	7.06
CtrlCl	8.09	BacPf	6.78	IocsPfCl	6.98	Iocs	7.69
CtrlCl	8.29	BacPf	7.51	IocsPfCl	7.16	Iocs	7.42
CtrlCl	8.22	BacPf	7.3	IocsPfCl	7.2	Iocs	7.19
CtrlCl	8.4	BacPf	6.85	IocsPfCl	7.12	Iocs	7.29
CtrlCl	8.67	BacPf	7.12	IocsPfCl	7.34	Iocs	7.29
CtrlPf	7.36	BacPfCl	7.14	bac	7.51	IocsCl	7.39
CtrlPf	7.44	BacPfCl	7.14	bac	7.51	IocsCl	7.39
CtrlPf	7.74	BacPfCl	7.25	bac	7.27	IocsCl	7.69
CtrlPf	7.77	BacPfCl	7.11	bac	7.43	IocsCl	6.84
CtrlPf	7.64	BacPfCl	7.05	bac	7.27	IocsCl	7.12
CtrlPf	7.56	BacPfCl	7.4	bac	7.33	IocsCl	7.53
CtrlPf	7.31	BacPfCl	7.08	bac	7.27	IocsCl	7.29
CtrlPf	7.26	BacPfCl	7.23	bac	7.03	IocsCl	6.88
CtrlPf	7.4	BacPfCl	7.11	bac	7.6	IocsCl	7.09
CtrlPf	7.7	BacPfCl	7.16	bac	7.33	IocsCl	7.32
CtrlPfCl	7.4	Ctrl	7.91	baccl	7.49	IocsPf	6.3
CtrlPfCl	7.37	Ctrl	7.91	baccl	7.49	IocsPf	6.3
CtrlPfCl	7.33	Ctrl	8.05	baccl	7.43	IocsPf	6.51
CtrlPfCl	7.4	Ctrl	8.51	baccl	7.48	IocsPf	6.88
CtrlPfCl	7.33	Ctrl	8.21	baccl	7.62	IocsPf	6.51
CtrlPfCl	7.47	Ctrl	8.16	baccl	7.42	IocsPf	6.38
CtrlPfCl	7.59	Ctrl	8.29	baccl	7.26	IocsPf	6.08
CtrlPfCl	7.54	Ctrl	7.98	baccl	7.32	IocsPf	6.56
CtrlPfCl	7.69	Ctrl	8.25	baccl	7.34	IocsPf	6.86
CtrlPfCl	7.57	Ctrl	7.81	baccl	7	IocsPf	6.38
Iocs	7.66	CtrlCl	8.27	BacPf	6.86	IocsPfCl	6.81
Iocs	7.84	CtrlCl	8.27	BacPf	6.86	IocsPfCl	6.81
Iocs	7.8	CtrlCl	8.05	BacPf	6.81	IocsPfCl	6.35
Iocs	7.76	CtrlCl	7.87	BacPf	6.68	IocsPfCl	6.98
Iocs	7.94	CtrlCl	7.75	BacPf	6.98	IocsPfCl	6.85
Iocs	7.76	CtrlCl	7.87	BacPf	6.68	IocsPfCl	6.65
Iocs	7.76	CtrlCl	8.22	BacPf	6.89	IocsPfCl	6.89
Iocs	8.12	CtrlCl	7.97	BacPf	7.15	IocsPfCl	6.55
Iocs	7.76	CtrlCl	8.38	BacPf	6.59	IocsPfCl	6.71
Iocs	7.71	CtrlCl	8.31	BacPf	6.86	IocsPfCl	6.88

Table 21. BAC Flux.

dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
bac	23	0.000878
bac	38.5	0.000707
bac	64.5	0.000569
bac	90.5	0.000435
bac	111.5	0.000404
bac	160.5	0.000335
bac	183.5	0.000316
bac	16.28	0.001406
bac	42.43	0.000951
bac	65.12	0.00083
bac	91.95	0.000808
bac	111.8	0.000667
bac	139.42	0.000612
bac	17.4	0.001196
bac	40	0.000957
bac	63.2	0.000731
bac	88.2	0.00055
bac	111.75	0.000645

Table 22. BACPF Flux.
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
bacpf	23	0.007066
bacpf	38.5	0.003201
bacpf	64.5	0.002708
bacpf	90.5	0.001852
bacpf	111.5	0.001616
bacpf	160.5	0.001343
bacpf	183.5	0.001551
bacpf	16.28	0.01312
bacpf	42.43	0.004662
bacpf	65.12	0.003758
bacpf	91.95	0.002286
bacpf	111.8	0.002828
bacpf	139.42	0.002563
bacpf	17.4	0.007446
bacpf	40	0.006146
bacpf	63.2	0.004649
bacpf	88.2	0.003532
bacpf	111.75	0.004032

Table 23. BACCL Flux.

dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
baccl	23	0.001268
baccl	38.5	0.001129
baccl	64.5	0.000877
baccl	90.5	0.000679
baccl	111.5	0.000602
baccl	160.5	0.000577
baccl	183.5	0.000543
baccl	16.28	0.002455
baccl	42.43	0.00155
baccl	65.12	0.001238
baccl	91.95	0.001412
baccl	111.8	0.001048
baccl	139.42	0.000856
baccl	17.4	0.001533
baccl	40	0.001004
baccl	63.2	0.000743
baccl	88.2	0.000577
baccl	111.75	0.000738

Table 24. BACPFCL Flux.
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
bacpfcl	23	0.002821
bacpfcl	38.5	0.00229
bacpfcl	64.5	0.001969
bacpfcl	90.5	0.001449
bacpfcl	111.5	0.001265
bacpfcl	160.5	0.001142
bacpfcl	183.5	0.001094
bacpfcl	16.28	0.005689
bacpfcl	42.43	0.004362
bacpfcl	65.12	0.003495
bacpfcl	91.95	0.003435
bacpfcl	111.8	0.002833
bacpfcl	139.42	0.002379
bacpfcl	17.4	0.007317
bacpfcl	40	0.004195
bacpfcl	63.2	0.003252
bacpfcl	88.2	0.002187
bacpfcl	111.75	0.002941

Table 25. CTRL Flux.
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
ctrl	23	0.000854
ctrl	38.5	0.000628
ctrl	64.5	0.000503
ctrl	90.5	0.000418
ctrl	111.5	0.000377
ctrl	160.5	0.000301
ctrl	183.5	0.000302
ctrl	16.28	0.001024
ctrl	42.43	0.000739
ctrl	65.12	0.000645
ctrl	91.95	0.000707
ctrl	111.8	0.000583
ctrl	139.42	0.0005
ctrl	17.4	0.000839
ctrl	40	0.000767
ctrl	63.2	0.000639
ctrl	88.2	0.000551
ctrl	111.75	0.000705

Table 26. CTRLCL Flux.
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
ctrlcl	23	0.001084
ctrlcl	38.5	0.000831
ctrlcl	64.5	0.000629
ctrlcl	90.5	0.000473
ctrlcl	111.5	0.000432
ctrlcl	160.5	0.000424
ctrlcl	183.5	0.00039
ctrlcl	16.28	0.002926
ctrlcl	42.43	0.001662
ctrlcl	65.12	0.001298
ctrlcl	91.95	0.001496
ctrlcl	111.8	0.000977
ctrlcl	139.42	0.000755
ctrlcl	17.4	0.001354
ctrlcl	40	0.000886
ctrlcl	63.2	0.000657
ctrlcl	88.2	0.000453
ctrlcl	111.75	0.000579

Table 27. CTRLPF Flux
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
ctrlpf	23	0.001866
ctrlpf	38.5	0.001192
ctrlpf	64.5	0.000949
ctrlpf	90.5	0.000755
ctrlpf	111.5	0.000668
ctrlpf	160.5	0.000548
ctrlpf	183.5	0.000543
ctrlpf	16.28	0.002696
ctrlpf	42.43	0.001683
ctrlpf	65.12	0.001437
ctrlpf	91.95	0.001312
ctrlpf	111.8	0.001082
ctrlpf	139.42	0.000903
ctrlpf	17.4	0.001826
ctrlpf	40	0.001655
ctrlpf	63.2	0.001379
ctrlpf	88.2	0.001175
ctrlpf	111.75	0.001504

Table 28. CTRLPFCL Flux.
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
ctrlpfcl	23	0.003096
ctrlpfcl	38.5	0.002235
ctrlpfcl	64.5	0.001674
ctrlpfcl	90.5	0.001152
ctrlpfcl	111.5	0.001054
ctrlpfcl	160.5	0.000917
ctrlpfcl	183.5	0.000869
ctrlpfcl	16.28	0.007288
ctrlpfcl	42.43	0.003099
ctrlpfcl	65.12	0.002034
ctrlpfcl	91.95	0.002035
ctrlpfcl	111.8	0.001382
ctrlpfcl	139.42	0.001161
ctrlpfcl	17.4	0.003124
ctrlpfcl	40	0.00159
ctrlpfcl	63.2	0.001199
ctrlpfcl	88.2	0.00087
ctrlpfcl	111.75	0.001225

Table 29. IOCS Flux.
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
iocs	23	0.001291
iocs	38.5	0.000861
iocs	64.5	0.000727
iocs	90.5	0.000458
iocs	111.5	0.000448
iocs	160.5	0.000396
iocs	183.5	0.000404
iocs	16.28	0.00247
iocs	42.43	0.001013
iocs	65.12	0.000708
iocs	91.95	0.000775
iocs	111.8	0.000639
iocs	139.42	0.000626
iocs	17.4	0.001753
iocs	40	0.000748
iocs	63.2	0.000545
iocs	88.2	0.000269
iocs	111.75	0.000275

Table 30. IOCSCL Flux.
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
iocscl	23	0.001494
iocscl	38.5	0.001085
iocscl	64.5	0.000896
iocscl	90.5	0.000687
iocscl	111.5	0.000615
iocscl	160.5	0.000577
iocscl	183.5	0.000541
iocscl	16.28	0.003067
iocscl	42.43	0.001702
iocscl	65.12	0.001338
iocscl	91.95	0.001492
iocscl	111.8	0.001007
iocscl	139.42	0.000822
iocscl	17.4	0.001343
iocscl	40	0.000879
iocscl	63.2	0.000651
iocscl	88.2	0.000449
iocscl	111.75	0.000647

Table 31. IOCSPF Flux.
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
iocspf	23	0.005917
iocspf	38.5	0.002
iocspf	64.5	0.001772
iocspf	90.5	0.001108
iocspf	111.5	0.001
iocspf	160.5	0.000872
iocspf	183.5	0.00089
iocspf	16.28	0.004278
iocspf	42.43	0.001816
iocspf	65.12	0.001506
iocspf	91.95	0.001447
iocspf	111.8	0.001313
iocspf	139.42	0.00114
iocspf	17.4	0.003076
iocspf	40	0.002041
iocspf	63.2	0.001701
iocspf	88.2	0.000966
iocspf	111.75	0.000883

Table 32. IOCSPFCL Flux.
dtime - elapsed time (hours)
flux - measured flux through the membrane (ml/(min*mm^2))

trtmnt	dtime	flux
iocspfcl	23	0.001831
iocspfcl	38.5	0.001431
iocspfcl	64.5	0.001157
iocspfcl	90.5	0.000845
iocspfcl	111.5	0.000795
iocspfcl	160.5	0.00071
iocspfcl	183.5	0.000696
iocspfcl	16.28	0.006841
iocspfcl	42.43	0.003085
iocspfcl	65.12	0.002351
iocspfcl	91.95	0.002265
iocspfcl	111.8	0.001573
iocspfcl	139.42	0.0013
iocspfcl	17.4	0.003152
iocspfcl	40	0.001801
iocspfcl	63.2	0.001501
iocspfcl	88.2	0.001035
iocspfcl	111.75	0.001523

Table 33. DAPI Thickness.
Fouling layer thickness in microns measured with epifluorescent microscopy and DAPI stain.

trtmnt	thickness	trtmnt	thickness	trtmnt	thickness	trtmnt	thickness
bac	10.85	bacpfcl	7.35	ctrlpf	7.72	iocscl	9.01
bac	10.58	bacpfcl	8.78	ctrlpf	6.88	iocscl	8.78
bac	13.34	bacpfcl	8.07	ctrlpf	11.39	iocscl	9.26
bac	9.34	bacpfcl	15.54	ctrlpf	5.24	iocscl	8.34
bac	11.11	bacpfcl	15.49	ctrlpf	7.12	iocscl	10.99
bac	8.08	bacpfcl	14	ctrlpf	6.79	iocscl	10.92
bac	7.83	bacpfcl	14.01	ctrlpf	22.3	iocscl	12.3
bac	13.89	bacpfcl	9.15	ctrlpf	21.5	iocscl	11.21
bac	9.56	bacpfcl	10.97	ctrlpf	20.7	iocscl	15.13
bac	10.72	bacpfcl	9.82	ctrlpf	21.4	iocscl	10.65
bac	12.01	bacpfcl	10.5	ctrlpf	20.8	iocscl	9.09
bac	14	bacpfcl	10.7	ctrlpfcl	13.69	iocscl	10.6
bac	11.21	bacpfcl	13	ctrlpfcl	10.24	iocscl	12.1
bac	11	bacpfcl	12.3	ctrlpfcl	17.87	iocscl	11.6
bac	13.1	bacpfcl	10.3	ctrlpfcl	13.52	iocscl	11.2
bac	10.4	ctrl	39.4	ctrlpfcl	18.32	iocscl	10.9
bac	11.1	ctrl	38.53	ctrlpfcl	19.36	iocspf	13.54
bac	12.5	ctrl	37.91	ctrlpfcl	12.39	iocspf	9.06
bac	12.9	ctrl	40.11	ctrlpfcl	14.86	iocspf	9.97
baccl	9.98	ctrl	38.67	ctrlpfcl	10.89	iocspf	11.35
baccl	9.34	ctrl	39.16	ctrlpfcl	10.86	iocspf	12.15
baccl	13.8	ctrl	53.55	ctrlpfcl	9.04	iocspf	12.15
baccl	9.28	ctrl	51.79	ctrlpfcl	6.75	iocspf	3.4
baccl	8.28	ctrl	49.59	ctrlpfcl	13.7	iocspf	4.14
baccl	10.57	ctrl	42.75	ctrlpfcl	13.5	iocspf	3.23
baccl	11.26	ctrl	48.6	ctrlpfcl	14.1	iocspf	6.68
baccl	18.98	ctrl	44.5	ctrlpfcl	14.6	iocspf	4.73
baccl	17.6	ctrl	43.7	ctrlpfcl	14.3	iocspf	5.64
baccl	20.19	ctrl	42	iocs	13.75	iocspf	4.84
baccl	17.88	ctrl	45.6	iocs	26.72	iocspf	6.98
baccl	15.1	ctrlcl	37.26	iocs	14.8	iocspf	7.77
baccl	13.7	ctrlcl	33.92	iocs	20.88	iocspf	8.9
baccl	13.9	ctrlcl	28.13	iocs	9.25	iocspf	9.6
baccl	14.3	ctrlcl	37.08	iocs	10.68	iocspf	12.1
baccl	13.8	ctrlcl	45.83	iocs	7.21	iocspf	10.9
bacpf	12.36	ctrlcl	39.08	iocs	8.56	iocspf	10.4
bacpf	5.93	ctrlcl	40.7	iocs	12.33	iocspfcl	4.03
bacpf	14.48	ctrlcl	40.4	iocs	8.71	iocspfcl	5.46
bacpf	9.73	ctrlcl	43.3	iocs	7.65	iocspfcl	7.83
bacpf	10.94	ctrlcl	42.1	iocs	10.15	iocspfcl	7.61
bacpf	3.09	ctrlcl	39.9	iocs	11.99	iocspfcl	24.7
bacpf	4.15	ctrlpf	28.98	iocs	9.91	iocspfcl	11.86
bacpf	5.97	ctrlpf	29.37	iocs	9.5	iocspfcl	18.86

bacpf	7.61	ctrlpf	31.66	iocs	9.75	iocspfcl	16.61
bacpf	9.4	ctrlpf	39.12	iocs	14.4	iocspfcl	11.63
bacpf	8.3	ctrlpf	41.73	iocs	13.5	iocspfcl	13.1
bacpf	8.5	ctrlpf	38.24	iocs	12.9	iocspfcl	13.5
bacpf	9.4	ctrlpf	11.86	iocs	15.1	iocspfcl	10.9
bacpf	10.1	ctrlpf	10.76	iocs	14.2	iocspfcl	11.3
bacpf	8.7	ctrlpf	8.11	iocscl	9.56	iocspfcl	11.9

Table 34. SEM Thickness.
Fouling layer thickness in microns measured with scanning electron microscopy.

trtmnt	thickness	trtmnt	thickness	trtmnt	thickness	trtmnt	thickness
bac	11.92	bacpf	3.95	ctrlcl	26.46	iocs	8.95
bac	11.87	bacpf	13.81	ctrlcl	34.36	iocs	9.04
bac	11.97	bacpf	14.12	ctrlcl	19.03	iocs	10.71
bac	10.71	bacpf	16.39	ctrlcl	34.1	iocscl	11.05
bac	10.5	bacpf	15.74	ctrlcl	39.44	iocscl	9.26
bac	11.14	bacpf	10.25	ctrlcl	34.39	iocscl	12.09
bac	12.11	bacpf	11.08	ctrlcl	40.92	iocscl	8.93
bac	12.43	bacpf	11.17	ctrlpf	4.43	iocscl	8.96
bac	11.14	bacpfcl	5.13	ctrlpf	4.3	iocscl	9.89
bac	11.46	bacpfcl	5.05	ctrlpf	4.82	iocscl	14.59
bac	8.13	bacpfcl	4.11	ctrlpf	10.4	iocscl	14.89
bac	8.02	bacpfcl	10.72	ctrlpf	11.48	iocscl	14.71
bac	7.92	bacpfcl	10.32	ctrlpf	12.92	iocspf	15.07
bac	9.78	bacpfcl	10.27	ctrlpf	10.47	iocspf	14.36
bac	9.12	bacpfcl	10.78	ctrlpf	11.62	iocspf	12.69
bac	10.33	bacpfcl	10.55	ctrlpf	12.24	iocspf	4.03
bac	10.66	bacpfcl	3.63	ctrlpfcl	6.34	iocspf	4.4
bac	8.44	bacpfcl	2.76	ctrlpfcl	6.33	iocspf	3.53
bac	12.81	bacpfcl	3.34	ctrlpfcl	6.91	iocspf	8.66
bac	9.81	ctrl	21.8	ctrlpfcl	5.92	iocspf	7.42
baccl	21.82	ctrl	22.56	ctrlpfcl	10.8	iocspf	6.58
baccl	23.7	ctrl	24.95	ctrlpfcl	10	iocspfcl	4.62
baccl	23.7	ctrl	17.65	ctrlpfcl	9.82	iocspfcl	4.8
baccl	19.6	ctrl	16.92	ctrlpfcl	6.23	iocspfcl	4.89
baccl	20.71	ctrl	19.32	ctrlpfcl	6.79	iocspfcl	3.68
baccl	20.08	ctrl	22.91	ctrlpfcl	6.09	iocspfcl	4.01
baccl	12.95	ctrl	23.68	iocs	17.92	iocspfcl	3.72
baccl	13.67	ctrl	23.81	iocs	15.39	iocspfcl	8.32
baccl	14.23	ctrl	24.35	iocs	19.89	iocspfcl	7.79
bacpf	4.94	ctrlcl	27.96	iocs	12.23	iocspfcl	8.89
bacpf	4.39	ctrlcl	27.86	iocs	12.81		
bacpf	4.03	ctrlcl	27.8	iocs	12.29		

Table 35. Bead Assay Cell Counts.
Bead assay biofilm cell counts log(cells/cm^2).

trtmnt	exp	cellsmm2	trtmnt	exp	cellsmm2	trtmnt	exp	cellsmm2
Bac	1	7.64	Ctrl	1	8.79	Iocs	1	6.42
Bac	1	7.6	Ctrl	1	8.34	Iocs	1	6.68
Bac	1	7.42	Ctrl	1	8.68	Iocs	1	6.24
Bac	1	7.96	Ctrl	1	8.55	Iocs	1	6.12
Bac	1	7.76	Ctrl	1	8.49	Iocs	1	6.49
Bac	1	7.64	Ctrl	1	8.6	Iocs	1	6.12
Bac	1	8	Ctrl	1	8.24	Iocs	1	6.12
Bac	1	7.55	Ctrl	1	8.64	Iocs	1	6.42
Bac	1	7.72	Ctrl	1	8.64	Iocs	1	6.24
Bac	1	7.76	Ctrl	1	8.72	Iocs	1	6.49
Bac	2	7.82	Ctrl	2	7.49	Iocs	2	7.42
Bac	2	7.96	Ctrl	2	7.64	Iocs	2	7.49
Bac	2	7.34	Ctrl	2	7.12	Iocs	2	7.24
Bac	2	7.72	Ctrl	2	7.64	Iocs	2	7.9
Bac	2	7.87	Ctrl	2	7.68	Iocs	2	7.72
Bac	2	7.76	Ctrl	2	7.49	Iocs	2	7.42
Bac	2	7.68	Ctrl	2	7.96	Iocs	2	7.6
Bac	2	7.12	Ctrl	2	7.6	Iocs	2	7.12
Bac	2	7.6	Ctrl	2	7.42	Iocs	2	7.34
Bac	2	7.72	Ctrl	2	7.64	Iocs	2	7.68
Bac	3	7.68	Ctrl	3	8.02	Iocs	3	7.15
Bac	3	7.82	Ctrl	3	7.98	Iocs	3	7.22
Bac	3	7.72	Ctrl	3	8.24	Iocs	3	7.17
Bac	3	7.76	Ctrl	3	8.07	Iocs	3	7.3
Bac	3	7.34	Ctrl	3	7.94	Iocs	3	7.2
Bac	3	8.12	Ctrl	3	8.06	Iocs	3	7.13
Bac	3	7.87	Ctrl	3	8.25	Iocs	3	7.24
Bac	3	7.6	Ctrl	3	8	Iocs	3	7.25
Bac	3	7.68	Ctrl	3	8.4	Iocs	3	7.09
Bac	3	7.64	Ctrl	3	7.64	Iocs	3	7.3
BacCl	1	5.92	CtrlCl	1	6.79	IocsCl	1	5.6
BacCl	1	5.98	CtrlCl	1	6.9	IocsCl	1	5.82
BacCl	1	6.28	CtrlCl	1	6.72	IocsCl	1	5.6
BacCl	1	6.02	CtrlCl	1	6.85	IocsCl	1	5.79
BacCl	1	6.04	CtrlCl	1	6.87	IocsCl	1	5.72
BacCl	1	6.34	CtrlCl	1	6.64	IocsCl	1	5.72
BacCl	1	5.76	CtrlCl	1	6.85	IocsCl	1	5.79
BacCl	1	6.04	CtrlCl	1	6.82	IocsCl	1	5.6
BacCl	1	5.85	CtrlCl	1	6.64	IocsCl	1	5.87
BacCl	1	5.87	CtrlCl	1	6.96	IocsCl	1	5.82
BacCl	2	6.94	CtrlCl	2	6.68	IocsCl	2	6.34
BacCl	2	6.64	CtrlCl	2	6.79	IocsCl	2	6.12
BacCl	2	6.6	CtrlCl	2	6.42	IocsCl	2	6.64
BacCl	2	6.55	CtrlCl	2	6.55	IocsCl	2	6.55

BacCl	2	6.24	CtrlCl	2	6.55	IocsCl	2	6.34
BacCl	2	6.6	CtrlCl	2	6.87	IocsCl	2	6.68
BacCl	2	6.79	CtrlCl	2	6.85	IocsCl	2	6.55
BacCl	2	6.64	CtrlCl	2	6.64	IocsCl	2	6.49
BacCl	2	6.68	CtrlCl	2	6.34	IocsCl	2	6.76
BacCl	2	6.55	CtrlCl	2	6.64	IocsCl	2	6.76
BacCl	3	6.55	CtrlCl	3	7.76	IocsCl	3	6.38
BacCl	3	6.94	CtrlCl	3	8	IocsCl	3	6.56
BacCl	3	7.1	CtrlCl	3	7.82	IocsCl	3	6.04
BacCl	3	7.06	CtrlCl	3	8.19	IocsCl	3	6.32
BacCl	3	7.24	CtrlCl	3	8.13	IocsCl	3	6.22
BacCl	3	7.04	CtrlCl	3	7.82	IocsCl	3	6.39
BacCl	3	6.6	CtrlCl	3	8.06	IocsCl	3	6.09
BacCl	3	6.87	CtrlCl	3	7.9	IocsCl	3	5.6
BacCl	3	6.64	CtrlCl	3	7.68	IocsCl	3	6.28
BacCl	3	7.27	CtrlCl	3	8.06	IocsCl	3	6.43
BacPf	1	5.68	CtrlPf	1	6.87	IocsPf	1	5.49
BacPf	1	5.64	CtrlPf	1	6.68	IocsPf	1	5.79
BacPf	1	5.55	CtrlPf	1	7.06	IocsPf	1	5.49
BacPf	1	5.79	CtrlPf	1	6.92	IocsPf	1	5.12
BacPf	1	6.12	CtrlPf	1	6.64	IocsPf	1	5.6
BacPf	1	5.55	CtrlPf	1	6.82	IocsPf	1	5.24
BacPf	1	5.76	CtrlPf	1	6.64	IocsPf	1	5.79
BacPf	1	5.64	CtrlPf	1	6.92	IocsPf	1	5.55
BacPf	1	5.9	CtrlPf	1	6.68	IocsPf	1	5.42
BacPf	1	5.92	CtrlPf	1	7.02	IocsPf	1	5.64
BacPf	2	6.42	CtrlPf	2	6.55	IocsPf	2	5.55
BacPf	2	6.6	CtrlPf	2	6.68	IocsPf	2	5.24
BacPf	2	6.49	CtrlPf	2	6.72	IocsPf	2	5.92
BacPf	2	6.79	CtrlPf	2	6.6	IocsPf	2	5.64
BacPf	2	6.64	CtrlPf	2	7.09	IocsPf	2	5.76
BacPf	2	6.24	CtrlPf	2	6.76	IocsPf	2	4.94
BacPf	2	6.34	CtrlPf	2	6.64	IocsPf	2	5.72
BacPf	2	6.85	CtrlPf	2	6.72	IocsPf	2	5.12
BacPf	2	6.34	CtrlPf	2	6.34	IocsPf	2	5.49
BacPf	2	6.55	CtrlPf	2	6.79	IocsPf	2	5.68
BacPf	3	5.96	CtrlPf	3	7.02	IocsPf	3	5.9
BacPf	3	6.12	CtrlPf	3	6.55	IocsPf	3	5.64
BacPf	3	6.06	CtrlPf	3	6.85	IocsPf	3	5.85
BacPf	3	5.92	CtrlPf	3	7.04	IocsPf	3	5.55
BacPf	3	6.1	CtrlPf	3	7.27	IocsPf	3	5.64
BacPf	3	5.9	CtrlPf	3	6.79	IocsPf	3	5.49
BacPf	3	6.24	CtrlPf	3	6.49	IocsPf	3	5.72
BacPf	3	5.72	CtrlPf	3	6.87	IocsPf	3	5.87
BacPf	3	5.82	CtrlPf	3	6.92	IocsPf	3	5.82
BacPf	3	6.04	CtrlPf	3	6.79	IocsPf	3	5.72
BacPfCl	1	5.82	CtrlPfCl	1	6.72	IocsPfCl	1	5.68
BacPfCl	1	5.87	CtrlPfCl	1	6.87	IocsPfCl	1	5.6

BacPfCl	1	5.64	CtrlPfCl	1	6.87	IocsPfCl	1	5.82
BacPfCl	1	5.85	CtrlPfCl	1	7.23	IocsPfCl	1	5.34
BacPfCl	1	6	CtrlPfCl	1	6.94	IocsPfCl	1	5.42
BacPfCl	1	5.87	CtrlPfCl	1	7.1	IocsPfCl	1	5.64
BacPfCl	1	5.82	CtrlPfCl	1	7.06	IocsPfCl	1	5.6
BacPfCl	1	5.72	CtrlPfCl	1	6.6	IocsPfCl	1	5.82
BacPfCl	1	5.9	CtrlPfCl	1	7.12	IocsPfCl	1	5.6
BacPfCl	1	5.72	CtrlPfCl	1	7.2	IocsPfCl	1	5.85
BacPfCl	2	5.72	CtrlPfCl	2	6.34	IocsPfCl	2	5.72
BacPfCl	2	5.55	CtrlPfCl	2	6.34	IocsPfCl	2	5.6
BacPfCl	2	5.76	CtrlPfCl	2	6.12	IocsPfCl	2	5.6
BacPfCl	2	5.6	CtrlPfCl	2	6.34	IocsPfCl	2	5.55
BacPfCl	2	5.68	CtrlPfCl	2	6.42	IocsPfCl	2	5.6
BacPfCl	2	6.07	CtrlPfCl	2	6.34	IocsPfCl	2	5.68
BacPfCl	2	5.64	CtrlPfCl	2	6.6	IocsPfCl	2	5.64
BacPfCl	2	5.85	CtrlPfCl	2	6.24	IocsPfCl	2	5.85
BacPfCl	2	5.64	CtrlPfCl	2	6.34	IocsPfCl	2	5.68
BacPfCl	2	5.55	CtrlPfCl	2	6.24	IocsPfCl	2	5.79
BacPfCl	3	6.06	CtrlPfCl	3	7.41	IocsPfCl	3	5.85
BacPfCl	3	5.92	CtrlPfCl	3	7.09	IocsPfCl	3	6.2
BacPfCl	3	6.13	CtrlPfCl	3	7.42	IocsPfCl	3	5.96
BacPfCl	3	5.94	CtrlPfCl	3	7.48	IocsPfCl	3	6.15
BacPfCl	3	6.21	CtrlPfCl	3	7.1	IocsPfCl	3	6.23
BacPfCl	3	6.25	CtrlPfCl	3	7.04	IocsPfCl	3	5.87
BacPfCl	3	6.07	CtrlPfCl	3	7.39	IocsPfCl	3	5.68
BacPfCl	3	5.92	CtrlPfCl	3	6.55	IocsPfCl	3	5.87
BacPfCl	3	6.15	CtrlPfCl	3	7.27	IocsPfCl	3	6.24
BacPfCl	3	6.09	CtrlPfCl	3	7.07	IocsPfCl	3	6.02