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TWO-STEP GRANULAR ACTIVATED CARBON FILTRATION
in Drinking Water Treatment

Riku Vahala

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Abstract

In the treatment of Finnish surface waters, the main objective is to reduce natural organic matter (NOM) before the water is disinfected. In many treatment plants, granular activated carbon (GAC) filtration is applied for the enhanced removal of NOM after conventional treatment. The aim of this study was to assess the performance of two-step GAC filtration in NOM removal from cold humic lake water and to determine its effects on the water quality in the distribution system. Furthermore, the optimal operation conditions of the existing process scheme at Helsinki Water's treatment plant after installing two-step GAC filtration and UV disinfection was evaluated. The study revealed that the major factors influencing the effective NOM removal in GAC filtration are the choice of GAC type and the frequency of regeneration. Other factors, such as empty bed contact time and biological degradation rate, were of minor importance. However, even the fully exhausted "old" GAC effectively decreased the assimilable organic carbon (AOC) and the regrowth of heterotrophic bacteria in the distribution system. The installation of two GAC filters in series did not result in the more effective use of GAC when a long service time was used. The most interesting finding of the study was that introducing GAC filtration and UV disinfection increased the nitrification in the chloraminated distribution system. The increased growth of nitrifying autotrophs can be explained by the fact that they compete with heterotrophs for biofilm space. In contrast to some other published studies, an excessive release of GAC fines was observed into the treated water during the filter run and filter ripening sequence. The air-scour backwash increased the release of carbon fines as a result of the grinding of GAC. Furthermore, the rapid changes in hydraulic loading were accompanied with the deterioration of GAC effluent water quality. Introducing GAC filtration is a challenge for the more efficient use of the existing treatment process. The preceding conventional treatment may be optimized for the efficient NOM removal, which was shown to increase the GAC service time. The optimal NOM removal was achieved with the use of ferric chloride sulphate instead of aluminium sulphate. GAC filtration also provided additional removal of residual metals resulting from enhanced coagulation. The transformation of NOM into a more biodegradable form was most efficient when a short ozone contact time was provided. The water pH and alkalinity did not affect AOC formation. Although optimal ozonation probably provided the highest biological activity in the following exhausted GAC filters, alone it does not justify the use of ozone. After installing GAC filtration and UV disinfection, the role of ozone is rather to provide an additional barrier against unexpected changes in the source water quality.

Keywords Adsorption; natural organic matter; optimization; biological processes

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Tiivistelmä

Aktiivihiilisuodatusta käytetään yleisesti yhtenä vaiheena pintavedestä valmistettavan juomaveden käsittelyssä. Suomessa pintaveden käsittelyn pääasiallinen tehtävä on vähentää raakaveden sisältämää orgaanista ainetta ennen desinfiointia. Tämän tutkimuksen tavoitteena oli selvittää kaksivaiheisen aktiivihiilisuodatuksen soveltuvuutta orgaanisen aineen poistoon kemiallisesti esikäsitellyistä kylmistä, pehmeistä ja humuspitoisista vesistä. Lisäksi pyrittiin optimoimaan Helsingin Veden tuleva puhdistusprosessi sen loppuvaiheeseen sijoitettavien aktiivihiilisuodatuksen ja UV-desinfioinnin tuomien mahdollisuuksien mukaan. Tutkimuksessa havaittiin, että aktiivihiilisuodatuksessa merkittävin orgaanisen aineen vähenemään vaikuttava tekijä on käytettävän hiililaadun ja sen regenerointitiheyden valinta. Viipymän ja biologisen hajoamisen vaikutus orgaanisen aineen kokonaisvähenemään oli pieni. Sen sijaan orgaanisella aineella kyllästynytkin ”vanha” aktiivihiili poisti tehokkaasti mikrobeille käyttökelpoista orgaanista hiiltä ja heterotrofisten bakteerien jälkikasvua vesijohtoverkostossa. Käytettäessä harvoja eli yli kahden vuoden regenerointivälejä, peräkkäisten aktiivihiilisuodattimen käyttö ei parantanut hiilen käytön tehokkuutta yksivaiheiseen ratkaisuun verrattuna. Tutkimuksen uusi kansainvälisestikin merkittävä havainto oli, että aktiivihiilisuodatuksen käyttöönotto lisää klooriamiinisuojatun vesijohtoverkoston biofilmissä tapahtuvaa nitrifikaatiota. Todennäköisesti tämä johtuu siitä, että heterotrofisten bakteerien vähentyessä biofilmissä kilpailu tilasta hidaskasvuisten autotrofisten nitrifikaatiobakteerien kanssa vähenee. Päinvastoin kuin monissa muissa tutkimuksissa, aktiivihiilisuodattimelta havaittiin vapautuvan suuria määriä hiilipölyä, joka kulkeutui käsiteltyyn veteen. Ilman käyttö vastavirtahuuhtelun yhteydessä rikkoi hiiltä ja lisäsi hiilipölyn määrää. Myös äkilliset muutokset suodatusvirtaamassa heikensivät merkittävästi veden laatua. Aktiivihiilisuodatuksen käyttöönotto antaa mahdollisuuksia ympäröivän prosessikokonaisuuden tehokkaampaan hyödyntämiseen. Suodatusta edeltävä kemiallinen saostus voidaan optimoida orgaanisen aineen vähentämiseksi, minkä osoitettiin pidentävän hiilen käyttöikää. Tutkitulla raakavedellä ferrikloridisulfaatti oli alumiinisulfaattia tehokkaampi orgaanisen aineen poistossa. Aktiivihiilisuodatin vähensi myös tehokkaasti saostuksessa veteen jääneiden metallien pitoisuuksia. Otsonoinnin havaittiin pilkkovan veden orgaanista ainetta mikrobeille käyttökelpoiseen muotoon tehokkaimmin silloin, kun käytettiin lyhyttä kontaktiaikaa. Veden pH:lla ja alkaliniteetilla ei havaittu olevan siihen vaikutusta. Otsonoinnin aktiivihiilisuodatusta tehostava vaikutus oli kuitenkin vähäinen ollakseen yksinään riittävä peruste sen käytölle. Aktiivihiilisuodatuksen ja UV-desinfioinnin käyttöönoton jälkeen otsonointi toimiikin tarkastellussa prosessissa lähinnä ylimääräisenä turvamarginaalina veden ennalta odottamattomia laatumuutoksia vastaan.

Asiasanat Adsorptio; orgaaninen aine; optimointi; biologiset prosessit

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PREFACE

Research work reported in this thesis has been carried out during the years 1995-1998 in the Laboratory of Environmental Engineering at the Helsinki University of Technology. Financial support given by Helsinki Water and Maa- ja vesitekniikan tuki ry is gratefully acknowledged.

I thank my supervisor professors Heikki Kiuru, Jukka Rintala and Risto Laukkanen for motivation and encouragement. Especially, Jukka Rintala's advice on scientific writing is highly appreciated. I am also grateful to the pre-examiners Professor Michele Prévost from École Polytechnique Montreal and Dr.ir. Jan Peter van der Hoek from Amsterdam Water Supply for reviewing this thesis and for their valuable suggestions.

All my co-workers and friends at the Finnish Water and Wastewater Works Association, HUT Laboratory of Water and Wastewater Engineering, and Helsinki Water deserve thanks for their company and assistance.

My deepest gratitude goes to my wife, Hanna-Maria, for her patience and love, and to my children, Johannes and Anniina, for giving me joy and support during past years.

Helsinki, October 2002

Riku Vahala

LIST OF ABBREVIATIONS

AOB	ammonia-oxidizing bacteria
AOC	assimilable organic carbon
AOX	adsorbable organic halogen
ATP	adenosine triphosphate
AWWA	American Water Works Association
BDOC	biodegradable organic carbon
DBP	disinfection by-product
DO	dissolved oxygen
EBCT	empty bed contact time
FP	formation potential
FRS	filter-ripening sequence
GAC	granular activated carbon
HGR	heterotrophic growth response
HPC	heterotrophic plate count
HPSEC	high-performance size-exclusion chromatography
MAP	microbially available phosphorus
MX	Z-3-chloro-4-(dichloromethyl)-5-hydroxy-2(5H)-furanone
NOM	natural organic matter
RSCCT	rapid small scale column test
SDS	simulated distribution system
SPC	standard plate count (agar)
THM	trihalomethane
TOC	total organic carbon
TSS	total suspended solids
UVA_{254}	ultraviolet absorbance at 254 nm
WTP	water treatment plant

LIST OF ORIGINAL PUBLICATIONS

This thesis is based on the following five original journal and four proceeding articles, which are referred to in the text in Roman numerals. The contribution of the author in each article is explained separately.

ARTICLES IN REFERRED JOURNALS

I. **Vahala R.**, Ala-Peijari T., Rintala J., Laukkanen R. (1998) Evaluating ozone dose for AOC removal in two-step GAC filters. Water Science and Technology 37(9), 113-120. *The author was responsible for the design and implementation of the pilot study. The other authors supervised the work and commented on the author's manuscript.*

II. **Vahala R.**, Långvik V.-A., R. Laukkanen R. (1999) Controlling AOX and THM formation by ozonation and two-step GAC filtration. Water Science and Technology 40(9), 249-256. *Author's contribution was the same as in I, except that V.-A. Långvik analyzed the samples for mutagenicity and MX.*

III. **Vahala R.**, Niemi R.M., Kiuru H., Laukkanen R. (1999) The effect of GAC filtration on bacterial regrowth and nitrification in a simulated water main. Journal of Applied Microbiology Symposium Supplement, 85, 178-185. *The author's contribution was the same as in I.*

IV. **Vahala R.**, Moramarco V., Niemi R.M., Rintala J., Laukkanen R. (1998) The effects of nutrients on NOM removal in BAC filtration. Acta hydrochimica et hydrobiologica, 26(3), 196-199. *The author was responsible for the design and instructions of the pilot study. V. Moramarco operated the pilot plant, took the samples and later prepared his MSc thesis from the material. The other authors supervised the work and commented on the manuscript, which was written by the author in co-operation with V. Moramarco.*

V. Vuorio E., **Vahala R.**, Rintala J., Laukkanen R. (1998) The evaluation of drinking-water treatment performed with HPSEC. Environment International 24(5/6), 617-623. *The author was responsible for the design and* *implementation of the pilot study. E. Vuorio analyzed the samples and prepared the manuscript based on her MSc thesis under instruction of the author. The other authors supervised the work and commented on the manuscript.*

CONFERENCE PROCEEDINGS

VI. **Vahala R.**, Rintala J., Järvinen A. (1996) Removal of TOC by two-step GAC filtration in cold humic waters. Proceedings of NOM workshop, September 18-19, Poitiers, France. *The author's contribution was the same as in I.*

VII. **Vahala R.**, Miettinen I.T., Laukkanen R. (1998) The effect of contact time, pH and ozone dose on the formation of biodegradable NOM during ozonation. Proceedings of IOA Conference on Ozonation and AOPs in Water Treatment, September 23-25, Poitiers, France, 31-1-8. *The author was responsible for the design and implementation of the pilot study. I.T. Miettinen analyzed the samples for AOC and HGR, and commented on the author's manuscript.*

VIII. **Vahala R.**, Miettinen I.T., Korhonen S., Laukkanen R. (1998) Evaluation of operational conditions for intermediate ozonation. Proceedings of AWWA Water Quality Technology Conference, November 1-4, San Diego, California. *The author was responsible for the design and performance of the pilot study. I.T. Miettinen analysed the samples for AOC. S. Korhonen analysed the samples with HPSEC. The principal author's manuscript was commented on by other authors.*

IX. Mikola A., **Vahala R.**, Laukkanen R. (1999) The response of two-step GAC filters to backwashing. Proceedings of AWWA Water Quality Technology Conference, October 31-November 4, Tampa, Florida, CD-ROM. *The author was responsible for instructing the study and writing the manuscript together with A. Mikola, who performed the study, analyzed the samples and prepared her MSc thesis from the results. Professor Laukkanen supervised the work and commented on the manuscript.*

TABLE OF CONTENT

1 INTRODUCTION

1.1 Background

Granular activated carbon (GAC) is a porous filter medium, that has been used in drinking water treatment for decades. Its large surface area adsorbs small compounds from water and provides sheltered conditions for the biological degradation of several different organic compounds (Wilcox *et al.* 1983). Frequent regeneration of GAC is necessary if effective adsorption is to be maintained.

Helsinki Water started to study GAC filtration with its present raw water in 1993 by launching a pilot-scale study, in which anthracite and GAC were compared as filter media. In the following year, the author published a literature survey, in which the pilot study was reported and the experiences from major cities using GAC in Finland were summarized (Vahala 1994).

The driving force for the interest in GAC was the deterioration of water quality in the distribution system, especially in Helsinki city centre, where the usage rate of water had decreased. The deterioration of water quality was seen as an increased biofilm formation and harmful deposits resulting in the increased replacement rate of water meters. The other driving force was the European drinking water legislation. When Finland joined the European Union in 1994, a target level of 2.0 mg/l total organic carbon (TOC) was introduced in the Finnish Drinking Water Degree. At that time, the TOC in the finished water was around 2.8 mg/l.

Helsinki Water has two treatment plants, the Pitkäkoski and Vanhakaupunki Water Treatment Plants (WTP), each of almost similar size and process configurations. Their raw water is supplied from Lake Päijänne. During the study, both plants consisted of chemical coagulation with aluminium sulphate, sedimentation, sand-filtration, ozonation and chloramination. Lime and carbon dioxide were used for pH control in three different phases.

Soon after the author's literature survey, the general layout plan of the GAC plant was finalized by the consultant (Ala-Peijari *et al.* 1994). The plan proposed introducing two-step GAC filtration and ultraviolet (UV) disinfection between ozonation and chloramination. The layout plan provided a framework for the pilot-scale studies at Pitkäkoski WTP (1995-1998) which are described in this thesis and in Annexes I-VIII. The results of this study were used in the process design of the full-scale treatment plants.

The two-step GAC process was selected to combine biological and adsorptive filtration in two sequential steps. The first filter would contain saturated GAC and was expected to eliminate the biodegradable compounds, while the fresh GAC in the second step adsorbs part of the non-biodegradable compounds. When saturated GAC is replaced with fresh GAC in the first filter, the flow is redirected so that it goes through the fresh GAC last. Thus the first filter always contains microbe-colonized GAC, and the carbon usage rate should be lower with a longer service life than can be expected with single-stage contactors.

In January 1998, Vanhakaupunki WTP launched two-step GAC filtration, which was followed by the world's biggest UV disinfection unit at that time. Three parallel online low-pressure UV units with a minimum UV dose of 25 J/m^2 were installed to eliminate the bacteria released from the GAC filters. The major changes in the final design compared to the general layout plan were that the first GAC filter was an upflow filter (instead of downflow) and that the bed depth of the single filter was 2.8 m (instead of 2.0 m). Later in December 1998, Pitkäkoski WTP was launched with a similar process modification. In the same year, the backwashing of GAC filters was studied in Vanhakaupunki WTP's full-scale GAC filters (IX).

1.2 Objectives

The aims of the study were as follows:

1. To assess the performance of two-step GAC filtration in natural organic matter (NOM) removal in cold, low alkalinity lake water, which was pre-treated with chemical coagulation, sedimentation, sand filtration and ozonation.
2. To evaluate the optimal operation conditions of the process scheme at Helsinki Water's treatment plants after installing two-step GAC filtration and UV disinfection.
3. To determine the effects of two-step GAC filtration on the water quality in the distribution system.

The results of each part are discussed in different Chapters (3-5) of this thesis. Chapter 6 discusses some different approaches in the development of Helsinki Water's treatment processes.

1.3 Scope and other supporting studies

The selection of the process scheme and operational parameters in the pilot plant study was based on the general layout plan of the full-scale plants. Owing to different process configuration (flow direction, bed depth, GAC type) in full-scale application, the results from the pilot-study are not directly applicable to the full-scale.

Since UV irradiation does not significantly alter the amount or composition of biodegradable NOM in the conditions normally applied in water treatment (Shaw *et al.* 2000), the UV disinfection had a limited role in the research framework.

Backwashing studies in the pilot-scale filter columns are difficult to interpret into full-scale. In order to obtain more reliable strategy for backwashing of GAC filters, the backwashing research was prolonged until the full-scale application was available. Annex IX describes the most interesting and unexpected results in the tests performed under author's instruction. A more detailed description of the study can be found elsewhere (Mikola 1999).

During the study, Helsinki Water tested several GAC types for NOM adsorption in small pilot filters (diameter 50 mm). The importance of GAC type selection for NOM adsorption is demonstrated by a selection of breakthrough curves from this test (Figure 2). The choice of GAC type (Filtrasorb F400) for the pilot study and full-scale application was based on these tests, but owing to commercial confidentiality the full report with manufacturer details has not been published. The full-scale filters were filled with a slightly modified Filtrasorb SF400.

Enhanced coagulation was studied using jar tests and pilot-studies under the author's instruction and are reported elsewhere (Pyrhönen 1997). The study compared different coagulants and coagulation conditions for optimum NOM removal at Pitkäkoski WTP. This thesis refers to some of the most important conclusions.

Parallel to this study, the Finnish Environment Institute carried out a microbiological study by taking water and biofilm samples at different treatment steps at the WTP and in the distribution system. The aim of the study was mainly to develop new microbiological methods, but also to assess the existence of potentially harmful microorganisms in the GAC filter bed and distribution system. The results are reported elsewhere and are not discussed here (Niemi *et al.* 1996, 1998; Niemi and Heiskanen 1997; Heikkilä 1999).

2 MATERIALS AND METHODS

2.1 Outline of experimental design

The study consisted of several pilot-scale runs at Pitkäkoski WTP (I-VIII) and a full-scale study at Vanhakaupunki WTP (IX). The rapid small-scale column tests (RSCCT) were not performed, because the short empty bed contact time (EBCT) and duration in the RSCCT complicate the prediction of the role of biodegradation in GAC filters (Summers *et al.* 1992; DiGiano *et al.* 1998).

In the beginning of the study Helsinki Water already had a pilot-scale WTP, that was able to simulate the existing treatment process in two parallel treatment trains (~ 0.2 m^3/h each). This pilot WTP was later updated to reflect the new process combination with two-step GAC filtration and UV disinfection. The study described in Annexes VII and VIII utilized the updated pilot WTP. This thesis not only summarizes the published studies, but also briefly describes some of the most interesting unpublished results obtained from the pilot WTP.

In order to obtain comparable results from different operational conditions, a pilot GAC plant was constructed in the beginning of this study. Most of the results in this thesis were obtained from four parallel pilot-scale filter pairs (diameter 200 mm), which were supplied from the full-scale process either before or after ozonation (I-VI). As discussed before, the pilot GAC plant was constructed to reflect the general layout plan of the full-scale GAC plant. All of the filter pairs were operated in downflow mode with a bed depth of 1.7-2.0 m. A pilot-scale ozonator was used to assess the effect of ozone on the GAC performance (I, V)

The first phase of the pilot test (29.5.1995- 12.3.1996) compared the effect of EBCT, ozone dose and GAC age on the filter performance (I, II, V, VI). In the second phase (15.4-10.9.1996), the non-ozonated train was subjected to a variation in ozone doses (I, V). In one train, Picabiol "biological" activated carbon was tested (V). Two trains were operated continuously for 473 days (I, II). The third phase (18.10.1996-29.1.1997) tested the effect of nutrient addition (IV; Moramarco 1997) and advanced oxidation techniques (Bicelli 1997) for 103 days.

The water quality changes of GAC filtered water in the distribution system were tested by supplying water from one train to the pilot water main after UV disinfection, pH control and chloramination. The reference water main was supplied with the effluent from the full-scale treatment process. Annex III gives a detailed description of the study.

The full-scale backwashing study was conducted at Vanhakaupunki WTP. The effects of operational parameters on the quality of the filter effluent were studied in a filter pair with fairly fresh GAC (IX).

2.2 Water source and quality

The raw water was taken from Lake Päijänne, which has typically low alkalinity and a moderate concentration of NOM. Although the specific UV absorbance was below 4.0, it was amenable to coagulation. During the study, chemical coagulation with aluminium sulphate at Pitkäkoski WTP removed some 50 % of the raw water TOC (Table 1). The raw water quality was subjected to minor seasonal changes.

Table 1. The average (min-max) water quality characteristics in the raw water and pilot GAC plant influent.

		Raw water	Sand-filtered	Ozonated
Temperature	^{o}C	6.3 (1-14)	8.7 (4.3-14.1)	7.5 (3.8-13.7)
pH		7.1	7.3 (7.1-7.5)	7.1 (6.5-7.6)
Alkalinity	mg $CaCO_3$	26	54 (34-70)	52 (28-69)
UVA_{254}	1/m	15.2	5.0 (4.3-6.2)	2.6 (2.1-3.7)
TOC	mg/l	5.7 (5.5-6.1)	2.8 (2.6-2.9)	2.7 (2.5-2.8)
Oxidizability	O_2 mg/l	5.1 (4.8-5.8)	1.7 (1.6-1.9)	1.5 (1.4-1.6)
Turbidity	FTU	0.18	0.06 (0.04-0.09)	0.05 (0.03-0.10)
Bromide	µg/l	< 20*	n.a.	n.a.

* Nissinen *et al.* (2002); n.a.= not analyzed

2.3 Analyses

UV absorbance (UVA_{254}) was measured at 254 nm with a Perkin-Elmer Lambda UV/VIS spectrophotometer using 1-cm quartz cells and distilled water as a reference, except in VII and VIII, where a Dr. Lange Cadas 100 UV/VIS spectrophotometer was used, and in IX where a Shimatsu UV-1601 spectrophotometer was used. The TOC analyses were performed in duplicate with an Astro 2001 TOC analyzer incorporating wet-oxidation catalyzed by UV light (precision ±0.1 mg/l). The total suspended solids (TSS) was determined using GF-C glass fiber filters and the optical particle counting was performed with a Pamas SVSS Analyser, in which the size of the particle was measured according to the area of its shadow (IX).

Chlorine demand samples were collected in 1 000-ml brown glass bottles and buffered to pH 7.0 with a phosphate buffer (II). The samples were then dosed at a 2:1 Cl2:TOC ratio with sodium hypochlorite and stored in the dark at room temperature. These samples were analyzed for residual chlorine by the iodometric method (Standard Methods 1985) following the 4 h, 1 d, 2 d, 3 d, 7 d and 9 d incubation period.

In the simulated distribution system (SDS) tests (II), samples were collected to determine the disinfection by-product (DBP) levels formed under potential chlorination conditions. Therefore, the pH of the sample was adjusted to 8.4 using a phosphate buffer, 0.6 mg/l of sodium hypochlorite was added and after half an hour of contact time, 0.2 mg/l ammonia was added to produce chloramines. After seven days' incubation period in the dark at room temperature, the samples were dechlorinated with tiosulphate and were then analyzed for adsorbable organic halogens (AOX) and trihalomethanes (THM). The THM were determined according to ISO/DIS 10301 with minor modifications, such as smaller sample volume (5 ml) and pentane extraction in the ampoule. AOX was determined according to ISO 9562, with 2-chlorobezoic acid as a standard, and nitrate wash of the column.

The biological stability of the ozonated water was determined using assimilable organic carbon (AOC) analyses and heterotrophic growth response (HGR). In the AOC assay (I, III, IV, VI, VIII), the growth of two inoculated bacterial strains (*Pseudomonas fluorescens* P17 and *Spirillum* NOX) in a sterilized water sample was monitored for nine days (Standard Methods 1995). The results for strain P17 were expressed as μg acetate C eq/l and for strain NOX as μg oxalate C eq/l. To ensure that nitrogen, phosphorus and other mineral salts were present in excess, inorganic nutrients were added to pasteurized samples, as presented by Miettinen *et al.* (1996b).

In the HGR assay (VII), the growth of indigenous heterotrophic bacteria in the water sample was followed. The glass flasks for HGR samples were acid-washed and baked at 250 °C for 8 h. HGR was defined as the maximum heterotrophic plate count (HPC) obtained during a three-week incubation period at 15 °C. The HPC were obtained three times per week using either the spread plate technique, with R2A agar incubated at 20±2 °C for 7 days.

The water samples for high-performance size-exclusion chromatography (HPSEC) were stored at -18°C (V, VIII). The samples were first filtrated through a 0.45-μm cellulose membrane filter (Millipore HA). Size-exclusion fractionation was performed using High-Pressure Liquid Chromatography with a Hewlett-Packard Series 1050 pump and a Hewlett-

Packard 4010 Series II Diode Array UV-detection system. The analytical column was a TSKgel G3000SW 7.5 mm (i.d.) × 30 cm and the guard column a TSKgel 7.5 mm (i.d.) × 7.5 cm. The injection volume was 25 μl, column temperature 30°C and UV detection wavelength 254 nm. A 0.01 M sodium acetate solution eluent was used with a flow rate of 1 ml/min. The data was analyzed using an HP ChemStation. In order to find the effect of ozonation on the molecular size distribution of NOM, the height of each peak was compared to the height of the same peak from the non-ozonated sample. Repeated analyses of the same water sample were conducted to control the precision.

Heterotrophic bacteria were enumerated by the spread plate (III) or pour plate technique, with R_2A incubated at 20 ^oC for 7 days, and with standard plate count (SPC) agar incubated at 20 ^oC for 3 days. The biomass of active bacteria was estimated with the adenosine triphosphate (ATP) analyses of extracts from the duplicate GAC samples, extracted in a buffer with TCA by applying ultrasonication for 20 minutes to enhance the release of ATP (IV). All the other analyses were performed according to the relevant Finnish Standards.

3 THE PERFORMANCE OF TWO-STEP GRANULAR ACTIVATED CARBON FILTRATION

3.1 Adsorption of natural organic matter

3.1.1 Adverse effects of natural organic matter in drinking water

NOM in water is a complex mixture of various compounds (such as humic substances, amino acids and carboxylic acids) generally present in such low concentrations that qualitative and quantitative analyses of specific molecules are difficult (Thurman and Malcolm 1981). The high amount of NOM (median 12 mgTOC/l) in Finnish lake waters results from the large proportion of the catchment that is covered by peatlands (Kortelainen 1993). Effective NOM removal in drinking water treatment is needed, because of its several adverse effects. NOM produces harmful disinfection-by-products and deteriorates the organoleptic quality, when reacting with chlorine (Symons *et al.* 1975; O'Connor *et al.* 1975). Furthermore, NOM increases the chlorine demand and may affect the corrosion rate of metals in the distribution system (Lee *et al.* 1980). The biodegradable part of NOM promotes microbiological growth in the biofilms of the piping system (Van der Kooij and Hijnen 1984).

In some countries, acceptable NOM levels in tap water are set in drinking water legislation. In the recent European drinking water directive (98/83/EY), NOM is no longer directly controlled by limit values for TOC or oxidizability. Instead, new standards for disinfection-by-products (trihalomethanes and bromate) and microbiological indicators (*Clostridium perfringens* and *Escherichia coli*) were introduced as indirect controls for NOM.

In the conventional drinking water treatment process, a large fraction of NOM is removed in chemical coagulation, flocculation, sedimentation and sand filtration. If better control of NOM is required, enhanced methods such as biological filtration, GAC adsorption or membrane technology needs to be addressed. In recent years, nanofiltration has gained a lot of interest owing to decreasing costs and superior water quality. However, even nanofiltered water with TOC levels below the detection limit can support microbiological growth in the distribution system without disinfectant residual (Noble *et al.* 1996; Sibille *et al.* 1997; Escobar and Randall 1999).

3.1.2 Two granular activated carbon filters in series

Conceptually, two-step GAC filtration is a modification of the Mülheim process (Sontheimer *et al.* 1978), where ozonation is followed by rapid sand filtration and GAC adsorption.

Bouwer and Crowe (1988) suggested using two GAC adsorbers in series, the first adsorber without regeneration and the second one with regeneration. In such an application, biodegradation of AOC is expected to reduce the TOC load for the second GAC adsorber, thus extending the service time *i.e.* regeneration frequency (Bouwer and Crowe 1988). In addition, Snoeyink (1990) expected lower GAC usage rates (the mass of activated carbon required per unit volume of water treated), when the GAC filters are arranged in series. He suggested that when GAC is replaced in the first column, the flow is redirected so that it goes through the freshest GAC last. Oxendorf and Lykins (1991) referred to an investigation, that estimated the in-series operation would treat about 40 % more water than a single GAC filter. Finally, Uhl *et al.* (1994, 1995) showed that a two-step GAC process with separate biodegradation and adsorption results in longer service time of filter beds than the Mülheim process alone. Besides Helsinki Water, *e.g.* Leiduin Plant in the Netherlands has recently introduced two-step GAC filtration with a total EBCT of 40 minutes (Van der Hoek *et al.* 2000).

In our first phase study, we wanted to make sure that these expectations would come true in Helsinki Water WTPs. Two-step GAC filtration was applied in four parallel pilot trains after conventional treatment with and without preceding ozonation (VI). The first filter was filled with exhausted GAC, while the second step contained fresh GAC (F400). In one reference train, both steps were filled with fresh GAC (F400). A comparison between the reference train and similar train with exhausted GAC revealed that unlike Bouwer and Crowe (1988) predicted and Uhl *et al.* (1994, 1995) demonstrated, the first step filtration with exhausted GAC did not significantly extend the service time of the following adsorber (2-step) (VI). The fresh GAC filter (Run 4) as a first step adsorbed 42 % of the influent TOC during 291 days, while at the same time the similar 2-step process with the fresh GAC filter in the second step (Run 3) adsorbed 40 % of the influent TOC (VI). Table 2 in Annex VI has some calculation errors. For the corrections, see Table 2 below.

The benefits gained from installing two adsorbers in series are apparently related to the service time of GAC. If the second GAC step has adsorption capacity left in the middle of the service time when the flow is redirected, the two-step process uses GAC more efficiently (lower usage rate) than single-stage adsorbers. On the other hand, if a long service time is applied and GAC is fully exhausted already in the middle of the service time, the benefits gained from using two-step GAC process are marginal. Our study showed that fully exhausted first-step GAC filter could not improve the performance of a second-step adsorber and, therefore the expectations of Bouwer and Crowe (1988) did not come true in this water. As can be seen from Figure 1, the point where GAC was fully exhausted with NOM seemed to occur after filtration of 15 000 bed volumes, which represented approximately 290 days with an EBCT of 15 minutes. Thus, if service time of more than 2 years is used, applying two-step GAC process cannot be justified by the lower GAC usage rate.

Although the operation of GAC columns in series did not result in improved performance compared to single stage design, the simultaneous parallel operation of multiple columns will minimize the GAC usage rate owing to blending. Dvorak and Maher (1999) demonstrated that when multiple parallel columns are operated in a staggered fashion (where the columns are placed on-line at different times), a system with 2-4 parallel GAC columns will give a dramatic improvement in the GAC usage rate compared to a single column system. They also observed that larger EBCTs utilize the GAC more efficiently.

Table 2. Removal of TOC by two-step GAC filtration during the pilot study (VI).

		Run 1	Run 2	Run 3	Run 4
TOC applied to contactors	Kg	**4.39**	**6.39**	4.26	4.26
	kg/m^{3}GAC	41.9	**60.9**	40.6	37.6
TOC removed in first filter	Kg	**0.20 (6.0 %)**	**0.39 (6.0 %)**	0.35 **(8.1 %)**	**1.78 (42 %)**
	kg/m^{3}GAC	**5.47**	**8.00**	**7.17**	**31.5**
TOC removed in second filter	Kg	**1.75 (40 %)**	**1.80 (28 %)**	**1.35 (32 %)**	**1.01** (24 %)
	kg/m^{3}GAC	**30.8**	**31.8**	**23.8**	**17.8**
TOC removed in two-step GAC filtration	Kg	**2.01 (46 %)**	**2.19 (34 %)**	**1.70 (40 %)**	**2.79 (66 %)**
	kg/m^{3}GAC	**19.2**	**20.9**	**16.2**	**24.6**

In addition to Figure 1, Annex VI shows the TOC breakthrough curves for the first 291 days in each run separately. The breakthrough of oxidizability and ultraviolet absorbance at 254 nm (UVA$_{254}$) were consistent with TOC (data not shown). The breakthrough of long-term chlorine demand and UVA$_{254}$ for the reference train (both filters filled with fresh F400) is illustrated in Annex II. As expected, the adsorption of DBP precursors strongly correlated with the decrease in TOC and UVA$_{254}$ (II).

Also, the effect of EBCT on NOM adsorption in the studied range (10-15 min.) was minimal. The breakthrough curve of the second-step GAC filter with a velocity of 8 m/h is almost identical to a filter with 12 m/h, which indicates the same GAC usage rate. For the utility with the on-site regeneration facility, this would allow smaller contact basins with shorter EBCT in order to achieve the same NOM removal efficiency as with longer EBCT. However, the decreased construction costs increase the operational costs, because more frequent regeneration is needed.

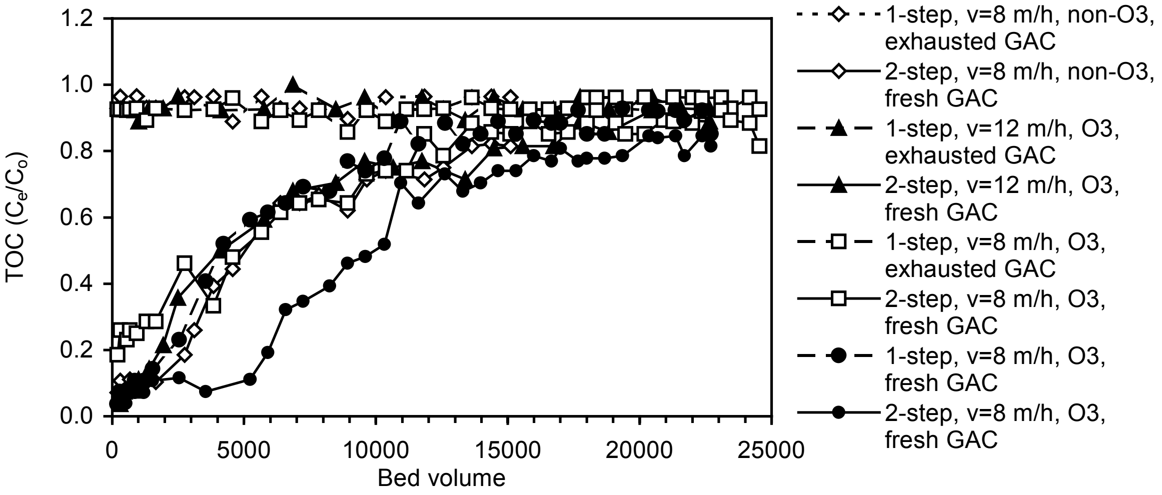


Figure 1. TOC breakthrough curves for two-step GAC filters.

For an unknown reason, the y-intercept in the breakthrough curve of the ozonated train (v=8 m/h) suggested that 20 % of TOC was non-adsorbable. However this is not true, but the effect is rather due to the wall-effect and short-cuts in the fresh GAC filter column, which both decrease the actual contact time. During the backwashing (once a week) the number of blocking GAC fines were decreased in the media and thus the filter trains were more comparable after the first few weeks of operation. It is possible that a larger column diameter than 200 mm could have prevented the wall-effects.

3.1.3 The type of granular activated carbon

The choice of GAC type is essential in efficient NOM adsorption. For example, Carlson *et al.* (1994) reported seven times higher removal of TOC with GAC manufactured for adsorption than with GAC for biological support. Before our pilot GAC study was conducted, several different GAC types were compared in 50 mm diameter filter columns with 10 minutes of EBCT. Figure 2 illustrates the importance of the choice of the GAC type by showing selected UVA_{254} breakthrough curves of six different GAC types.

Type F (Picabiol) is GAC manufactured for biological support and owing to its rapid exhaustion it was used to demonstrate the effect of GAC age on the removal of different molecular fractions in Annex V. The breakthrough curve of large and intermediate molecules was similar to that of TOC. However, the larger molecules seemed be less adsorbable than the smaller ones and the smallest molecules were occasionally released into the filter effluent.

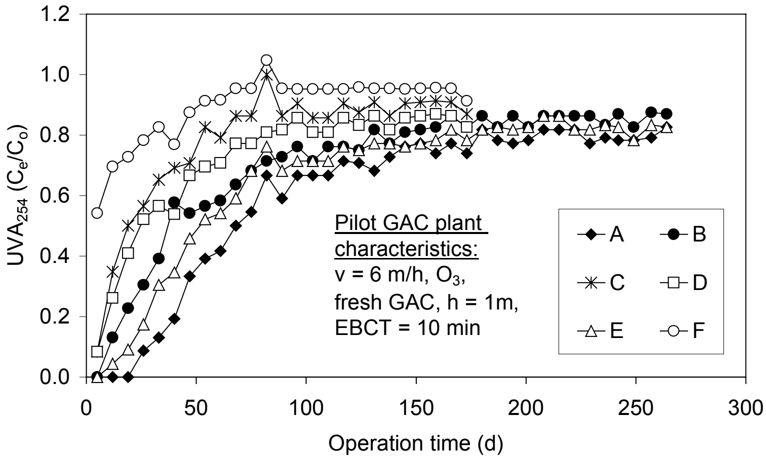


Figure 2. TOC breakthrough curves for different GAC types (A-F).

In this study, the choice of GAC type was based on its adsorption properties for NOM. Thus, the Type A (Filtrasorb F400), which showed the highest adsorption capacity for UV-adsorbing compounds and TOC, was selected for most of the pilot-studies (I-VI).

3.1.4 Service time

The cost of the GAC process depends on its service time for removing target compounds and the need to replace or regenerate it once it loses its adsorption capacity. If the service time is dictated by the operational criteria for the average finished water TOC, it is essential that the start-up of a new GAC installation and the schedule of regeneration be carefully managed. The following calculation based on the breakthrough curves in Figure 1 illustrates how the selected operational criteria affects the service time of the single GAC filter pair.

The calculation is based on the following assumptions:

1. The TOC removal in the exhausted GAC is gradually increased from 0.3 to 0.6 mg/l, when the EBCT is increased from 10 to 15 minutes, respectively. The influent TOC (2.8 mg/l) is not subjected to seasonal changes.
2. The plant consists of 14 parallel two-step filter pairs, which are placed on-line in a staggered fashion. This ensures that the plant consists of 28 GAC filters of different age.

3. Accordingly, the replacement of filters with fresh GAC takes place at even intervals. For example, if the calculation gives a service life of two years, the replacement of a single filter with fresh GAC will take place every month. Before the replacement in a filter pair, the age of the first filter and the second filter is two years and one year, respectively. During the replacement the flow is redirected so that the age of the first GAC filter is one year and fresh GAC is in the second step.
4. The adsorption capacity of GAC is not significantly deteriorated during regeneration.

Figure 3 illustrates how the small changes in the targeted NOM removal rate significantly affect the expected GAC service time. For the given operational criteria of 2.0 mgTOC/l, the service life of 1 170 days (3.2 years) can be expected using the design parameters of the pilot GAC plant. At Helsinki Water's Pitkäkoski and Vanhakaupunki full-scale WTPs, a service life of four years was expected for the total number of 28 filters in each plant. For comparison, at Amsterdam Water Supply's Leiduin WTP, the service life of the two-step GAC filter is two years. Consequently, with a total of 40 filters, every 2-3 weeks a filter bed is sent for regeneration (Van der Hoek *et al.* 2000). The impact of decreasing influent NOM loading into GAC filters is discussed later in Section 4.1.1.

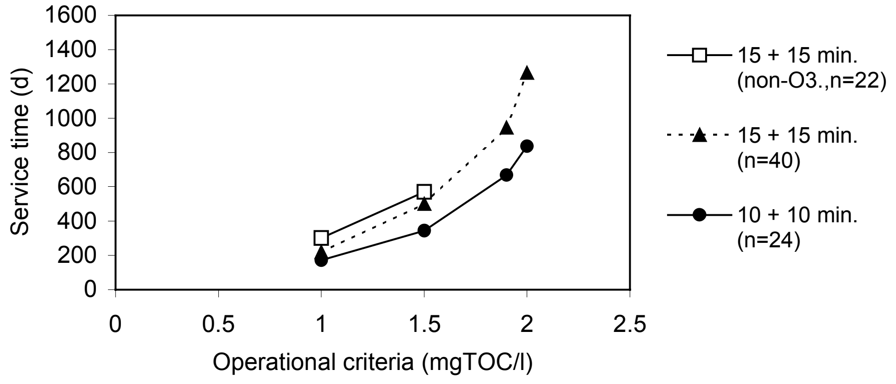


Figure 3. Service life of GAC at different TOC targets.

3.1.5 Conclusions

The major factor that affects the effective NOM adsorption is the choice of GAC type and the frequency of regeneration. In order to obtain treated water with constant NOM levels, the replacement of GAC needs to be carefully managed. Other studied process parameters were of minor importance in NOM removal. Neither the installation of two GAC filters in series, nor the longer EBCT in the studied range (10-15 min.) resulted in more effective use of GAC, *i.e.* a lower GAC usage rate when a long service time is used.

In order to obtain a clear picture of the operational criteria in GAC adsorption, the economical benefits of effective NOM removal should be evaluated. This would allow optimization of the service time of GAC beds and minimization of the overall costs that are derived from frequent regeneration and NOM adverse effects in the distribution system. At Helsinki Water, public health protection and customer satisfaction were already at a high level before process modifications and thus were not the driving force for the investment.

3.2 Biological stabilization of drinking water

3.2.1 Introduction

It is well established that ozonation transforms NOM in a more available form for the bacteria (Van der Kooij *et al.* 1989; Miltner *et al.* 1992; Kruithof *et al.* 1996). Therefore, many post-ozonation plants have introduced biological filtration to limit bacterial regrowth in the distribution system. In biological filtration, a large part of the biodegradable NOM is eliminated by the heterotrophic bacteria attached to the filter media (Weber *et al.* 1978; Van der Kooij 1979; Cairo *et al.* 1979; Servais *et al.* 1991; Cauchi *et al*. 1993; Cipparone *et al.* 1997). Compared to sand-anthracite filter media, GAC has several advantages. GAC provides better NOM removal owing to adsorption and biodegradation especially at colder temperatures. GAC establishes a biodegradable NOM-removing biofilm more rapidly, provides increased protection against oxidant residual and permits faster establishment of NOM removal after periods out of service (Urfer *et al.* 1997). Several authors have reported that bacterial activity in GAC filters contributes to an increase in the GAC service life (Werner *et al.* 1979; De Laat *et al.* 1985; Bouwer and Crowe 1988; LeChevallier *et al.* 1992; Kim *et al.* 1997).

Biodegradable NOM is generally quantified with the analyses of biodegradable organic carbon (BDOC) or assimilable organic carbon (AOC). Our study measured the amount of organic carbon available for microbial growth by AOC assay (I). AOC was determined with salt addition to describe the part of AOC, which the test bacteria (*P. fluorescens P17* and *Aquaspirillum NOX*) can potentially use as their energy and carbon source without growth limitations by inorganic nutrients (Miettinen *et al.* 1999). During our study, other researchers showed that in many waters, phosphorus rather than organic carbon is the limiting nutrient for the microbial growth in AOC assay (Miettinen *et al.* 1996a, 1997; Sathasivan *et al.* 1997;

Charnock and Kjønnø 2000). Unfortunately, the amount of biologically available phosphorus was not determined in our study.

According to Rittmann (1985), the kinetics of the biodegradation is controlled at least by three factors: the mass transfer resistance, the amount of active biomass and the substrate concentration. Carlson and Amy (1998) showed that the removal of biodegradable NOM was controlled by the biomass concentration, only. Whichever the case, the amount of active biomass is the only factor that can be controlled during the design and operation of GAC contactors. Above a minimum biomass level, the temperature rather than the amount of biomass is the rate-limiting factor for biodegradation (Urfer *et al.* 1997). In the following chapters, different aspects of biological GAC filtration, *i.e.* filtration with exhausted GAC are discussed separately.

3.2.2 The flux of substrates into the filter

The characteristics of NOM are the major factors that determine the extent of biodegradation in the GAC media. NOM with greater diffusivity and faster degradation kinetics is removed to a greater extent. Recently, it has been demonstrated that the presence of readily biodegradable substrate significantly enhances the removal of slowly degradable NOM owing to its ability to maintain higher amount of biomass in the filter media (Bouwer and Hozalski 2001).

In our study, the first-step filtration with exhausted GAC decreased 51 % of AOC_{P17} and 72 % of AOC_{NOX} from ozonated water (I), which was slightly lower than generally observed (Van der Kooij 1990; Huck *et al.* 1994; Hu *et al.* 1999). Higher removal rates were attributed to higher influent AOC concentrations. However, increased influent AOC also resulted in increased effluent AOC (I). For example, if 500 µg-C/l of AOC_{NOX} was introduced in the exhausted GAC filter, 370 µg-C/l was removed and if 300 µg-C/l of AOC_{NOX} was introduced, only 200 µg-C/l was removed. The percentage reduction is 74 % in the first case and 67 % in the second one, but the effluent AOC is still higher in the first case (130 µg-C/l) than in the second one (100 µg-C/l). Similar findings have been observed with BDOC, but with AOC a constant percentage removal has been proposed (Billen *et al.* 1992; Huck *et al.* 1994; Zhang and Huck 1996).

The AOC levels were generally higher than reported elsewhere (Charnock and Kjønnø 2000). Volk and LeChevallier (2000) examined 95 treatment plants in United States, where the mean AOC in finished water ranged from 18 to 214 µg Ac-C/l. In our study, the finished water

AOC was 179±12 µg-C/l and 580±59 µg-C/l with and without GAC filtration, respectively (III). It must be noted that the AOC assay was performed with salt addition, and that the AOC for strain NOX was expressed as µg/l oxalate C eq/l, which gives four times higher values than if expressed as µg/l acetate C eq/l. In order to obtain comparable results with the other published studies, we analyzed a single set of samples for AOC in the KIWA N.V. laboratory, in the Netherlands, where the AOC assay was invented. In Table 3, all the values are expressed as µg/l acetate C eq/l.

Although, the AOC levels that were determined in KIWA laboratory were much lower than in our study, the AOC in finished water was still higher than recommended for biological stability (10 µg/l acetate C eq/l). Interestingly in this single sampling campaign, the sand-filtered water could be considered as biologically stable. The variation between two laboratories could be attributable to several reasons, which demonstrates the need for careful consideration when comparing absolute AOC values in different studies. Accordingly, the definition of biologically stable water is quite arbitrary and the criteria must be established based on the applied assay for determination of biological stability and the characteristics of the distribution system.

Table 3. Comparison of AOC results (µg/l acetate C eq/l) between two laboratories (22.4.1996).

	AOC, with salt addition $_{(1)}$	AOC $_{(2)}$
Sand-filtered water	40	4.5 ± 1.1
Ozonated water	185	84 ± 32
GAC filtered water	50	28 ± 6.8*
Finished water	70	28 ± 7.4

1) National Public Health Institute, Kuopio, Finland (I, III, IV, VI, VIII, Miettinen *et al.* 1996b)
2) KIWA N.V., Netherlands, KIWA standard LMB-014 (* One of the duplicate bottles had a variation coefficient of the maximum colony count of 59 %)

3.2.3 The structure of carbon for bacterial attachment

The type of filter media may have significant influence on NOM biodegradation. Generally, GAC provides a better NOM removal than sand or anthracite media (Li and DiGiano 1983; Bablon *et al.* 1988; Servais *et al.* 1991; LeChevallier *et al*. 1993). However, Liu *et al.* (2001)

concluded that easily biodegradable NOM can be effectively removed even at low temperatures using either GAC or anthracite if there is no chlorine in the backwash water.

Also, the type of GAC may affect biodegradation. For example, Wang *et al.* (1995) observed a better performance of microporous coal-based GAC at steady-state compared to sand, anthracite-sand and macroporous wood-based GAC, which they attributed to differences in GAC surface, adsorption of substrates early in the bed life, residual adsorption capacity, and the differences in the microbial communities. On the other hand, Carlson *et al.* (1994) found no difference in TOC or DBP precursor removal between micro- and macroporous GAC types. However, chlorine demand was more effectively reduced with macroporous GAC and was less deteriorated by the low water temperature. Similarly, Prévost *et al.* (1991) found that the amount of attached biomass was greater in macroporous GAC and suggested that shorter contact times could be used with macroporous GAC in cold waters.

In our first pilot study (VI), the TOC removal in exhausted GAC filters varied between 6-8 % (EBCT 8.5-13 min. v = 8-12 m/h, T = 4-12 ^{o}C), while the fully exhausted GAC filters (EBCT 15 min., v = 8 m/h, T = 4-10 ^{o}C) in the second study (IV) contributed to 12-14 % reduction in TOC. In a pilot-scale WTP (EBCT 13-14 min., v = 6 m/h, T = 4-11 ^{o}C), the TOC reduction of 11-12 % was achieved in a study where two coagulants were compared as a pretreatment for GAC (data not shown). The most significant difference between the conditions of these studies was the different batch of Filtrasorb 400 that was used in the exhausted GAC filters of the first study. In all our studies, the NOM removal efficiency was quite low in comparison to those generally observed (Bouwer and Crowe 1988; Dussert and Tramposch 1997; Urfer *et al*. 1997).

Our other, unpublished study compared the performance of macroporous wood-based Picabiol and microporous charcoal-based Filtrasorb 400 in NOM removal. As already illustrated in Figure 2, Picabiol had a low adsorption capacity for NOM. After the short adsorptive phase, the removal of TOC, UVA_{254}, AOC and chlorine demand did not significantly differ from the reductions in the Filtrasorb 400 GAC filter (Table 4). However, the HPC of the Picabiol filter effluent significantly increased during the first two months of operation after which it remained stable (Figure 5). The higher HPC levels in the wood-based GAC filter effluent might be explained by the macroporous structure of Picabiol, which offers more suitable sites for bacterial attachment than the microporous structure of the reference filter, or simply by the higher initial phosphate concentration in wood-based GAC, which stimulated the growth of free-living bacteria (IV). The latter explanation is supported by Wang *et al.* (1995), who could not find a difference in biomass accumulation between F400 and Picabiol, but observed slightly better performance of F400 in TOC and AOC removal. Although they did not report the phosphorus levels, it is possible that in their water phosphorus was not limiting the microbial growth and thus the wood-based GAC media did not enhance the biodegradable NOM removal. It should also be noted that the increase in effluent HPC number does not necessary indicate increased bacterial biomass in the GAC media.

Table 4. The average HPC in the exhausted GAC filter effluent, and the removal of UVA_{254}, TOC, AOC and long-term chlorine demand.

	Picabiol	Filtrasorb 400
Effluent HPC	3000 CFU/ml (n=6)	170 CFU/ml (n=4)
UVA_{254}	7 % (n=12)	9 % (n=11)
TOC	10 % (n=9)	10 % (n=8)
AOC	83 % (n=2)	79 % (n=1)
Chlorine demand (7 days)	19 % (n=3)	24 % (n=3)

3.2.4 Empty bed contact time

Contact time, usually expressed as EBCT, is one of the most important variables for biological filtration (Urfer *et al.* 1996; Huck *et al.* 1998). Increasing the EBCT usually increases the biodegradable NOM removal (Sontheimer and Hubele 1987; Merlet *et al.* 1991; Servais *et al.* 1992; LeChevallier *et al.* 1992; Prévost *et al.* 1992), although effective reduction of biodegradable NOM may be achieved with a relatively short 5-9 minutes of EBCT (Hozalski *et al.* 1995; Koffskey and Lykins 1999; Carlson and Amy 2001). Since the external mass transfer plays a minor role in biodegradable NOM removal, hydraulic loading, *i.e.* velocity seems to be a less important design parameter (Servais *et al.* 1992; Urfer *et al.* 1996). Instead, the utilization of the substrate, which is a function of EBCT and biomass concentration, appears to limit biodegradable NOM removal (Wang *et al.* 1995). To sum up, EBCT is an acceptable design parameter throughout the range of hydraulic loading rates and filter bed depths (Carlson and Amy 1998).

In our study, the effect of EBCT was compared in two parallel ozonated pilot plant trains, which were operated with an EBCTs of 8.5 and 13 minutes, respectively (VI). Table 5 summarizes the percentage removal of NOM in the exhausted GAC filters.

Table 5. NOM removal in the exhausted GAC filters with different EBCTs.

EBCT	8.5 min.	13 min.	n
UVA_{254}	7 %	8 %	42
TOC	7 %	8 %	23
Oxidability	14 %	13 %	10
AOC	68 %	69 %	5
Chlorine demand (7 days)	13 %	15 %	3

It can be seen that the increase in NOM removal is minimal when the EBCT is increased from 8.5 to 13 minutes. The curves for TOC removal efficiency are shown in Figure 1 and VI, from which it is concluded that the studied EBCT range did not affect the biodegradation of TOC. The results show that the provided time was enough for efficient AOC removal, but the decrease in slowly biodegradable NOM degradation was limited. Others have suggested that most of the rapidly biodegradable compounds (like AOC) are removed in the upper layers, while the contact time needed for the removal of chlorine demand and slowly biodegradable compounds (like BDOC) is longer (Prévost *et al.* 1992).

The effect of longer EBCT is illustrated in Table 6, where the NOM removal in two exhausted serial GAC filters is summarized. The limited data describes the NOM removal of Run 4 in Annex VI during operation days 389-473. It must be noted that the values cannot be compared with Table 5, because the GAC properties were different in this GAC batch. The substantially better response to an increase in EBCT is probably due to remaining adsorption capacity and better performance of two separate serial filters, where the mass transfer zone is not mixed during the backwashing.

Table 6. The effect of EBCT on NOM removal in the exhausted two-step GAC filters.

EBCT	15 min. (1-step)	30 min. (2-step)	n
UVA_{254}	9 %	21 %	42
TOC	10 %	18 %	23
Oxidability	14 %	21 %	10
Chlorine demand (7 days)	24 %	32 %	3

The decrease in long term chlorine demand (7d) was higher than the TOC or UVA_{254} removed in exhausted GAC filters, suggesting that GAC preferentially remove compounds susceptible to reactions with chlorine (II). Merlet *et al.* (1991) observed similar results and suggested that the part of NOM that can be biologically removed is more reactive to chlorine than refractory carbon.

3.2.5 Operating at low temperatures

Decreasing temperature increases the acclimatization period needed to achieve biological activity in the GAC filter and the contact time for removing the same amount of biodegradable NOM (Servais *et al.* 1992; Krasner *et al.* 1993; Moll and Summers 1999; Hozalski and Bouwer 2001). The simulations by Hozalski and Bouwer (2001) indicated that, when operating at temperatures below 9 ^{o}C, the accumulation of biomass and removal of biodegradable NOM are severely inhibited. Moll and Summers (1999) found that operating at 5 ^{o}C was detrimental to biomass growth and substrate removal. They also showed that low temperature not only decreases the rate of substrate metabolism, but also changes the microbial community structure (Moll *et al.* 1999). On the other hand, Fonseca *et al.* (2001) demonstrated that operating temperature did not affect total amount of biomass in biofilters. However, the microbial (dehydrogenase) activity was 70 % higher at 12 ^{o}C than at 3 ^{o}C.

In our studies, there was no obvious correspondence between the TOC removal with exhausted GAC and temperature, which varied between 4-14 ^{o}C (VI); the decrease in UVA_{254} and oxidizability showed no correlation with the temperature either (data not shown). However, as illustrated in Figure 4, the HPC in the GAC filter effluent decreased as temperature decreased. The figure shows the effluent HPC in the filter pair, where both the filters were filled with fresh GAC at the beginning of the study.

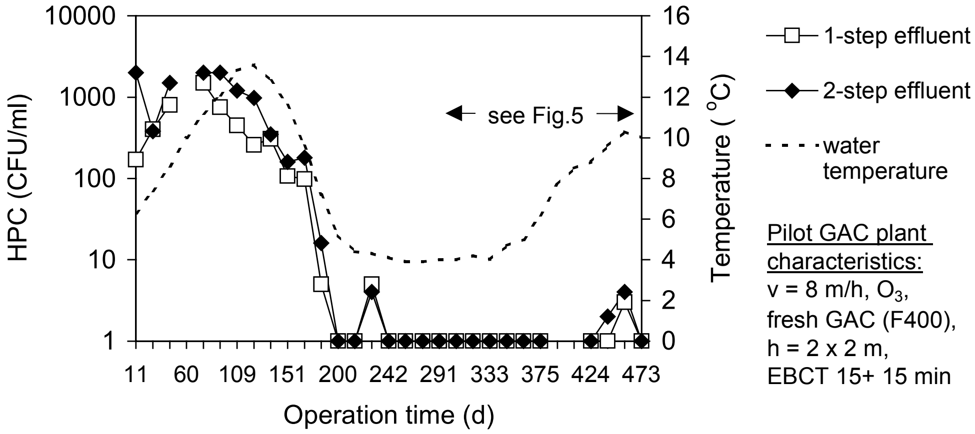


Figure 4. Heterotrophic plate count (HPC) and temperature in the two-step GAC effluent.

Koffskey and Lykins (1999) found no indication of a reduction in biological activity during the low temperature period (6-10 °C), when monitoring effluent HPC, AOC and DOC. Similar results were reported by Carlson *et al.* (1994), who found that in the temperature range of 5-30 °C, TOC removal was not affected. By monitoring the reduction of dissolved oxygen (DO), they observed an increased degree of biological degradation during the summer months. However, AOC was not measured. In our study, the decrease in DO remained typically in the range of 0.3-0.4 mg/l and 0.4-0.8 mg/l in exhausted and adsorptive GAC filters, respectively (data not shown). The DO consumption was much lower than observed by Carlson *et al.* (1994) and did not appear to be a good surrogate parameter for the low degree of biological activity. In order to assess the expected impact of temperature fluctuations on AOC removal in our study, a broader database would be required (I).

3.2.6 Nutrient limitation

The amount of assimilable organic carbon is often regarded as the limiting factor for the growth of heterotrophic microorganisms in drinking water (Van der Kooij 1982). Nevertheless, there is evidence that in some drinking waters the growth of bacteria in the bulk water can be stimulated by the addition of microbially available phosphorus (MAP), such as ortophosphates (Miettinen *et al.* 1996a, 1997; Sathasivan *et al.* 1997; Sathasivan and Ohgaki 1999). In our waters, the nutrient limitation in AOC assay (data not shown) suggested that part of the biodegradable NOM could not be utilized in the GAC filter owing to nutrient limitation. Although we were not aware of any previous studies showing improved biodegradation in the GAC filter owing to nutrient addition, we were encouraged to study whether it could stimulate bacterial growth in our exhausted GAC filters (IV).

To our disappointment, the addition of nutrients had no statistically significant influence on the NOM removal when monitored by TOC, UVA_{254} and AOC (IV). However, in the limited number of samples, the nutrient addition increased the HPC of the filter effluent, while the amount of active biomass on GAC measured by the adenosine triphosphate (ATP) analyses did not show any increase. The limited information suggested that phosphorus could be a limiting nutrient, but the increased bacteria could not attach themselves during the relatively short acclimatization period in the GAC media (IV).

During our study, the assay for MAP was not available (Lehtola *et al.* 1999) and therefore unfortunately not performed. Based on other more recent investigations, it seems that the limiting nutrient in all the targeted waters after chemical treatment was MAP (Lehtola *et al.* 2002b). It was also shown, that the MAP concentrations in the studied raw water and drinking water were 5.55 µg/l and 0.48 µg/l, respectively (Lehtola *et al.* 2002b). It must be noted, that there is a typing error in Annex IV: the total phosphorus concentration in raw water is 6-8 µg/l, instead of 6-8 mg/l.

It has been demonstrated that chemical coagulation effectively decreases the concentration of biologically available phosphorus (Sathasivan and Ohgaki 1999; Lehtola *et al*. 2002b), thus making it possible that MAP is partly the limiting nutrient in many GAC applications following coagulation. This assumption is supported by the general observation from several WTPs where the concentration of total phosphorus after GAC filtration declined below the detection limit of 2 µg/l and the MAP was decreased by 47 % (Lehtola *et al.* 2002b). Nishijima *et al.* (1997) showed that in some cases the bacterial colonization in GAC media is more rapid with phosphorus addition. Recently, Lehtola *et al.* (2001) revealed that similar to AOC, ozonation increases the amount of MAP. Thus, it is possible that part of the increased NOM degradation in ozonated biofilters could be due to increased amount of MAP.

It remained unclear whether the phosphorus limitation occurred only in the bulk water. It is possible that phosphorus did not limit the biomass accumulation, because of the phosphorus turnover from the lysed cells, or because of the accumulation of phosphate within the biofilm matrix, which is in accordance with the observation that the amount of active biomass was not increased owing to phosphorus addition (IV). Chandy and Angles (2001) have recently obtained similar results in the annular reactor, where the addition of phosphorus did not result in additional biofilm development compared to reactor without phosphorus addition. Furthermore, Appenzeller *et al.* (2001) observed that phosphate addition limited the proliferation of heterotrophic bacteria in corroded pipes by modifying the the properties of corrosion products. Meanwhile, Keinänen *et al.* (2002) observed changes in the microbial community structure in drinking water biofilms due to the addition of phosphate. Recently, Lehtola *et al.* (2002a) revealed that the addition of phosphate can increase the amount of active biomass and the total number of bacteria in laboratory biofilm experiments. It appears that in most of these studies, biologically available carbon is present in excess (Rompré *et al.* 2000; Appenzeller *et al.* 2001; Chandy and Angles 2001), while in the Finnish experiments (Lehtola *et al.* 2002a; Keinänen *et al.* 2002) MAP is the limiting nutrient for the bacterial growth.Thus, the latest research shows that increased phosphorus levels in the plant effluent may also stimulate the bacterial regrowth in the distribution system. . Therefore, until the role of phosphorus recycling and accumulation in the distribution system biofilm is carefully examined, the addition of phosphorus in excess should be avoided in those systems where it could be the limiting nutrient in some parts of the network. This issue is further discussed in section 5.1.

3.2.7 Colonization of biomass onto GAC media

The biomass on the GAC filter accumulates owing to deposition and growth as the water flows through the filter bed. At the same time, the biomass is removed from the filter media by decay, fluid shear and backwashing. Mostly, the biomass losses occur from backwashing, as discussed later in Section 3.3.2. Hozalski and Bouwer (2001) have recently illustrated in their modelling work that, if the biomass lost during backwashing does not exceed 60 %, it has a minor impact on biodegradable NOM removal. In our full-scale study, the reduction in active biomass was increased by increasing the duration of backwashing and by introducing air scour (Mikola 1999). However, the loss in biomass always remained below 60 %.

It has been estimated that the colonization of the GAC filter with biomass into the steady-state condition takes approximately three months (Cauchi *et al.* 1993). At decreased temperature, colonization can take much longer. In our study, the colonization was indirectly monitored by the effluent HPC, which started to increase soon after the beginning of the experiment (Figure 4). The type of GAC strongly affected the effluent HPC. In the filter pair with fresh Picabiol GAC, the effluent HPC was significantly higher than in other runs and showed a slight increase during the first two months of operation, which could be partly attributed to the increase in temperature (from 4 to 10 ^{o}C) at the same time (Figure 5.). The minor changes in the effluent HPC in most runs can be explained by bacterial detachment in the beginning of the filter run. It is suggested that the detachment of bacteria is related to the phase of bacterial growth. Once the colonization on the GAC media has reached a steady-state, the effluent HPC decreases (Cairo *et al.* 1979).

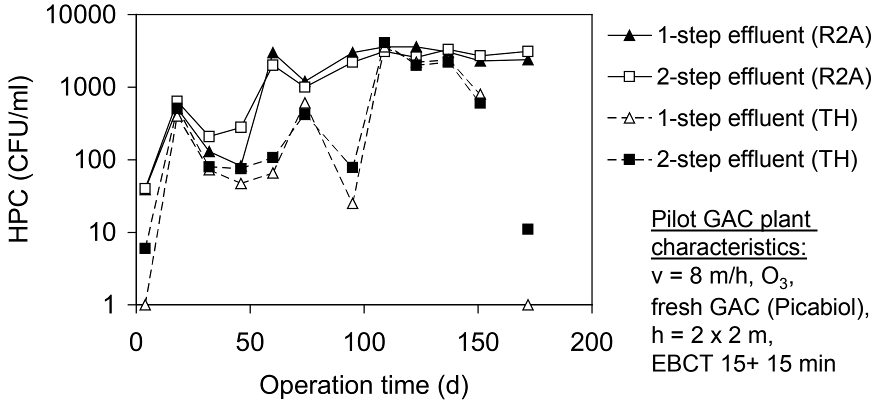


Figure 5.Increase in effluent HPC (R2A or TH, 20 °C, 7 d, pour plate) as a function of time (fresh Picabiol in both columns).

In order to obtain more reliable results of the biological activity in the GAC filter, the amount of the attached biomass needs to be estimated in addition to effluent HPC. During our study, ATP analyses were adapted for the measurement of active biomass on the GAC media, which proved to be a more suitable indicator than effluent HPC for assessing the biological activity in the GAC filter. Unfortunately, the number of samples in this study was too small to give any conclusive results.

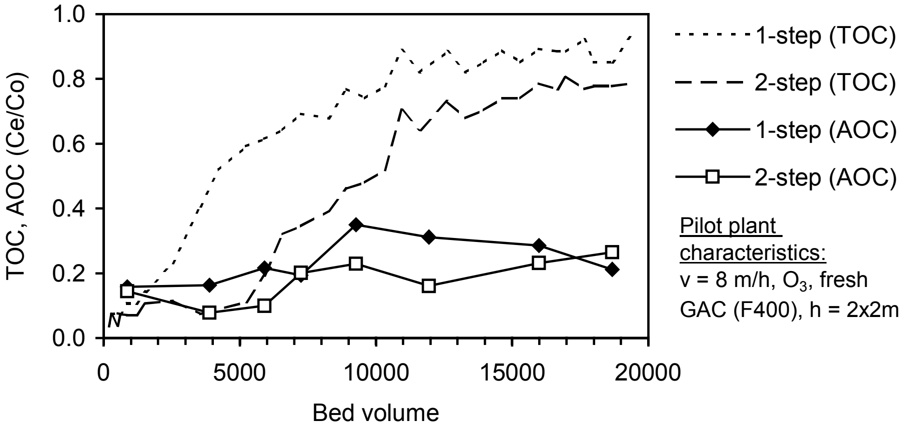


Figure 6. AOC breakthrough curves for two-step GAC filters (fresh Filtrasorb 400).

The minimal impact of GAC age on AOC removal suggests that adsorption has a minor role in the biological stabilization of drinking water (I). Figure 6 shows the unpublished AOC breakthrough curves for two GAC adsorbers in series. The TOC, UVA_{254} and chlorine demand breakthrough curves for this filter pair are shown in Annexes II and V.

3.2.8 Distribution of active biomass

According to the literature, it seems that a minimum biomass is needed for biodegradation, but above the minimum, the amount of biomass is not critical for the rate of biological NOM removal. Consequently, the increase in biomass does not necessarily result in increased NOM removal and it cannot be used as a sole indicator of the biological GAC filter performance (Wang *et al.* 1995, Moll *et al.* 1999, Fonseca *et al.* 2001). Better correlation could be achieved by using microbiological activity assays.

Several studies have shown that the inlet part of the GAC filter bed is biologically more active than the rest of the filter (Servais *et al*. 1991, 1992; Wang *et al.* 1995; Moll *et al.* 1999; Van der Hoek *et al.* 2000). This can be seen as a decrease of biomass and as a difference in microbial community structure as a function of filter depth due to a decrease in labile substrates through the filter bed (LeChevallier *et al.* 1992; Moll *et al.* 1998, 1999). It seems that most of the AOC is removed during the first two minutes, while a longer contact time is needed for the removal of chlorine demand and BDOC (Prévost *et al.* 1991, 1992). Thus, selecting EBCT based on BDOC removal efficiency results in more conservative design values than the use of AOC alone. In order to obtain more accurate design criteria and a more complementary view of the biodegradable NOM removal (Kaplan *et al.* 1994; Volk and LeChevallier 2000; Escobar and Randall 2001; Sharp *et al.* 2001), a combination of bio-film based methods, such as biofilm annular reactor method, and different standard NOM measurements, such as AOC and BDOC, should have been used in our study.

If AOC removal mainly appears in the top layers of the GAC bed, it could be expected that it be accompanied with a higher amount of the active biomass. However, we could not observe any vertical distribution of active biomass in the full-scale adsorptive filter beds (IX), which could have resulted from the low temperature, deep GAC bed or backwashing with air. Servais *et al.* (1991, 1992) observed a vertical distribution of bacterial biomass in GAC beds at 20 ^{o}C, but not at 9 ^{o}C. Liu *et al.* (2001) observed biodegradable organics removal throughout the entire filter bed at lower temperatures (5^{o}C), whereas it occurred only in the top layers of the filter at higher temperatures (20^{o}C). Billen *et al.* (1992) showed a decreased stratification of bacteria, when the GAC bed depth was increased, *i.e.* filtration velocity was increased. In the studies of Beaudet *et al.* (1996), the use of air scour in backwashing broke the bacterial stratification in the GAC bed.

Since the flow of substrates is higher in the first GAC filter step, it is expected that also the amount of biomass is higher in the first filter. Our full-scale study indicated that the amount of active biomass did not differ between the first and second step filters, when both of the filters were still in the adsorptive phase (IX). A single sampling experiment in our exhausted two-step pilot filters showed significantly higher ATP and detached HPC in the first step GAC media than in the second step (Niemi and Heiskanen 1997). However, the effluent HPC was lower in the first step effluent than in the second step.

The observed bacterial activity in both of the filter steps contradicts the expectations of Bouwer and Crowe (1988). They suggested that the bacterial activity in the second GAC step would be minimal, because nearly all the biodegradable NOM is removed in the first step. This could have been attributable to overestimating the capability of heterotrophic bacteria to degrade slowly biodegradable NOM. Similar to our study, the case study in Jefferson Parish, LA, USA showed that during the passage through the GAC filter, the HPC increased by 3 to 4 logs and remained at these higher levels with subsequent serial passage through the following GAC filters (Geldreich 1996).

3.2.9 Conclusions

Although exhausted GAC contributed only to a minor decrease in NOM concentration, it effectively stabilized NOM. It seems that exhausted GAC filtration is a feasible method for decreasing AOC even when treating cold humic waters. The frequent regeneration of GAC is not necessary for the biological stabilization of NOM.

Improving the performance of NOM biodegradation in a GAC filter by increasing the EBCT does not appear to be an economically feasible solution. A major portion of rapidly biodegradable compounds is removed with a relatively short EBCT of less than 10 minutes. However, it must be noted that NOM consists of both rapidly biodegradable compounds, which bacteria can directly use for metabolism, and slowly biodegradable compounds such as humic substances, which can partially be degraded after exoenzymatic action. Optimizing the GAC type for biodegradation or adding inorganic nutrients did not provide additional NOM removal. Instead, they increased the HPC in the GAC effluent and could possibly increase the risk of bacterial contamination in the distribution system.

The primary limitation for effective biodegradation was the nature of NOM. Although, the relative proportion of the biodegradable NOM was small, it yielded a high AOC level in ozonated water and was effectively reduced in exhausted GAC. The AOC reduction in the GAC filter was more effective with higher influent AOC values, which suggests that the flux of substrates limited effective biodegradation in the exhausted GAC filter.

Although the biodegradation rate did not depend on water temperature, it seemed to effect the effluent HPC. Since we could not observe any correlation between low temperature and performance for NOM removal in GAC filters, it is possible that the attached biomass was all the time below the level, at which biodegradation is limited by temperature. Neither, we were able to show any relationship between active biomass in GAC media, effluent HPC and biodegradation of NOM. It seems that the increase in effluent HPC does not necessarily indicate an increase in attached biomass on GAC, which in turn does not correlate with the NOM biodegradation. More comprehensive research and a database are required before concluding what is the most feasible monitoring parameter for the effectiveness of GAC biodegradation.

3.3 Release of particles

3.3.1 During the filter run

In biological processes, a small amount of bacteria that colonize the surface of the GAC is always released into the treated water (AWWA 1981; Wilcox *et al.* 1983; LeChevallier *et al.* 1992). Mostly, these free living bacteria are harmless and easily disinfected. Sometimes, the GAC filter may colonize potentially pathogenic micro-organisms and micro-organisms of faecal origin (Camper *et al.* 1986; Stewart *et al.* 1990), thus increasing the risk of bacterial contamination of the drinking water. However, the pathogens do not usually succeed in the competition with the natural heterotrophic bacteria in an oligotrophic environment (Camper *et al.* 1985; Rollinger and Dott 1987). It also seems that the bacteria released from the biologically active filters are unlikely to colonize in the distribution system owing to high level of competition with the previously established biofilm organisms (Norton and LeChevallier 2000)

Several researchers have shown that during the operation of the GAC filters, a small number of GAC fines is released into the finished water (Amirtharajah and Wetstein 1980; Camper *et al.* 1986, 1987; Stewart *et al.* 1990, Stringfellow *et al.* 1993). These fines provide a shelter for bacteria against the oxidative disinfectant (Ridgway and Olson 1981; LeChevallier *et al.*

1984, 1988; Stewart *et al.* 1990) and contribute to the formation of loose deposits, which are re-suspended into the water during changes of the hydraulic conditions in the distribution system (Gauthier *et al.* 1999). This may generate not only complaints from customers, but also pose a threat to the public health if the deposits are colonized with the opportunistic pathogens (Camper *et al.* 1986; LeChevallier 1990; Stewart *et al*. 1990). On the other hand, more recent findings suggest that the small number of released GAC fines, colonized by a small number of organisms, may not be a threat if normal disinfection is applied (Morin *et al*. 1996; Morin and Camper 1997; Pernitsky *et al.* 1997). It must be noted that the studies refer to disinfection practice in the United States, where much higher chlorine doses are maintained in the distribution system than in Europe.

The concern of an excessive release of GAC fines may be especially important if the underdrain system of the GAC filter is damaged or is generally in such a poor condition that the GAC media can escape into the treated water (Stringfellow *et al.* 1993). A layer of sand (0.3-0.5 m) as a support for GAC may prevent the escape of GAC fines in the finished water (Dussert and Van Stone 1994).

Our full-scale study (IX) showed that the hydraulic step *i.e* sudden increase in hydraulic loading was accompanied with rapid increase in GAC effluent turbidity and total suspended solids (TSS). The microscopic analyses of the glass-fibre filters in the TSS analyses revealed that a significant number of GAC fines were released from the filter bed into the finished water. The hydraulic step was due to backwashing in other filter pairs and the recycling of filtrates from the GAC and sand filters. However, when the water flow rate was slowly increased under controlled conditions, no significant increase in turbidity occurred.

Similarly, Cleasby *et al.* (1963) demonstrated that the hydraulic step resulted in particle detachment into the filtered water. Ahmad *et al*. (1998) observed that a 30 % increase in water flow rate caused the previously deposited material to be flushed into the effluent. However, the HPC was not affected suggesting that the shear forces needed for the detachment of hydrophobic bacteria are higher than those needed to dislodge hydrophilic non-biological particles (Ahmad and Amirtharajah 1998). Recently, Huck *et al.* (2002) showed that hydraulic step (25 % increase in filtration rate) had little effect on *Cryptosporidium* oocyst concentration, but did increase turbidity and particle counts in some experiments.

Owing to the sensitivity of the GAC filter, it is critical to carefully consider the flow conditions in the process design. The plant must be designed to avoid hydraulic steps, *e.g.*

when one filter pair is taken into the backwashing procedure. The contradictory results in different studies indicate that not all the variants affecting the release of GAC fines are established. Our results are different from those obtained *e.g.* by Stringfellow *et al*. (1993), who concluded that the mass of carbon fines entering the treated water is small. They could not find any difference in the release of carbon fines between old, reactivated and new GAC. With the chlorine doses usually involved in the USA, the low concentration of colonized particles was not thought to penetrate the chlorine barrier. Apparently, more research is needed to assess the effects and quantity of GAC fines in the distribution system biofilms and deposits, and to create effective measures in controlling the release of GAC fines during the operation and backwashing of GAC filters.

3.3.2 During the filter-ripening sequence

Frequent backwashing of biological GAC filters is needed to remove biomass and non-biological particles, and to prevent the growth of protozoa in the filter media. Backwashing controls the development of filter head-loss and limits the accumulation of particles and flocs, which may inhibit the bacterial activity and decrease the actual contact time in the filter (Niquette *et al.* 1998). Unless backwash water contains high levels of chlorine, the bacterial biomass attached on the GAC surface is not sufficiently removed and the biofilter performance is impaired during backwashing with water alone (Servais *et al.* 1991, 1992; Miltner *et al.* 1995; Hozalski and Bouwer 1998). In an adsorber, backwashing should be minimized in order to prevent mixing of the bed (Snoeyink 1990). However, it is quite theoretical to separate biological and adsorptive GAC filtration, because in all the GAC filters both mechanisms take place simultaneously.

Backwashing two-step GAC filters involves a different strategy for both steps. Before the study, it was assumed that the particle release from the first-step filter is not critical, because the following step would prevent particles escaping from the filter pair. Accordingly, it was expected that the filtrate recycling during the filter-ripening sequence (FRS) of the first GAC step was not necessary. However, the study revealed that the second-step filter could not retain particles released during FRS of the first-step GAC filter (IX). This suggests that the tolerance of the GAC bed against rapid changes in the loading was low and there is a need for filtrate recycling during FRS of the first GAC step as well.

Insufficient removal of biomass and non-biological particles may be a problem for unchlorinated water backwash at low bed expansions (Hozalski and Bouwer 1998). Since chlorinated water backwash may be detrimental to biological activity in the filters, enhanced backwash with air-scour has been introduced. Air-scour generally cleans GAC media much better than water-only backwash and impairs neither AOC removal nor biological activity (Ahmad *et al.* 1998). However, controversial results exist about the effects of air-scour on GAC characteristics. Some authors state it is possible that during air-scour backwash, the GAC breaks apart and results in a greater pressure drop over time, a greater loss of carbon during backwash and a release of carbon fines during filtration (Boller and Kavanaugh 1995; Humby and Fitzpatrick 1996). On the other hand, Grens and Werth (2001) found only a marginal effect on the particle characteristics during 500 air-scour backwash cycles in pilot filters. Our full-scale study demonstrated that air-scour backwash increased the release of carbon fines due to grinding of GAC (IX). As already noted by Hozalski and Bouwer (1998), more research is needed to define an optimum backwash strategy for GAC filters and the effects of air scour backwash.

3.3.3 Conclusions

In order to obtain effective removal of biomass and non-biological particles from GAC filter bed, many utilities apply backwash with air-scour. To limit the accumulation of colonized GAC fines in the distribution system, careful attention should be paid to the factors that affect the detachment of particles during FRS and filter run, unless it can be shown that the bacteria attached in the GAC fines are efficiently disinfected and that they do not provide any harm in the tap water, and in the biofilm and deposits of the distribution system. A site-specific backwashing strategy needs to be established in the varying operational conditions, which reflect the changes in temperature, hydraulic loading, GAC age and influent water quality.

4 THE CHALLENGES IN THE OPTIMIZATION OF THE UPDATED TREATMENT PROCESS

4.1 Enhanced coagulation

4.1.1 Increase in GAC service life

Effective NOM removal in coagulation increases the GAC filter's useful service life by reducing the TOC load for GAC filters and by increasing the adsorptive capacity of GAC for the remaining NOM (Semmens and Field 1980; Randtke and Jepsen 1981; Hooper *et al.* 1996; Semmens *et al.* 1986b, Hozalski *et al.* 1995; Nowack *et al.* 1999). Although the relatively low specific UV absorbance in our raw water (2.7 m^2/g) suggested that the water is not amenable to coagulation (Edzwald 1993), alum coagulation and sand-filtration efficiently removed the largest molecular fractions of NOM (V). The fraction of the largest molecules disappeared completely and the next two fractions were reduced by 92 % and 85 %, respectively. At the same time, the TOC reduction averaged 52 % (V). The seasonal variation of NOM size distribution was minimal (data not shown).

In enhanced coagulation higher coagulant doses are used, the pH of the raw water is optimized and/or the coagulant type is selected to achieve maximum NOM removal in addition to the decrease in suspended particles (Cheng *et al.* 1995; Crozes *et al.* 1995). In some cases, iron salts appear to remove NOM more efficiently than alum; in other cases NOM has a greater affinity for iron than for alum (Kavanaugh 1978; Randtke and Jepsen 1981; Randtke 1988; Julien *et al.* 1994; Crozes *et al.* 1995; Bell-Ajy *et al*. 2000). The studies performed at Pitkäkoski WTP showed that, in this water, ferric chloride sulphate was far more efficient in NOM removal than aluminium sulphate. In optimal conditions for NOM removal, a TOC of 2.4 mg/l and 1.7 mg/l was achieved with aluminium sulphate and ferric chloride sulphate, respectively (Pyrhönen 1997). Owing to low raw water alkalinity (28 mg/l $CaCO_3$), coagulant dosage alone was efficient in achieving the optimal pH for NOM removal.

Semmens *et al.* (1986b) has demonstrated that the coagulant dosages required to optimize the cost of water treatment by GAC are typically higher than those for normal turbidity removal are. It is obvious that to minimize the overall cost of the NOM removal, enhanced coagulation is required. Figure 7 illustrates the increased service life of GAC columns resulting in enhanced coagulation with iron. This unpublished study was performed in the pilot-WTP at Pitkäkoski WTP. The breakthrough curves indicate a better performance of

GAC adsorber when it is preceded by iron coagulation. Nowack *et al.* (1999) expected higher adsorbability of NOM after iron coagulation, which could result from iron neutralizing the charge of TOC, from the multivalent cation causing individual organic molecules to take a more compact shape, from iron cations forming a bridge between the negatively charged sites of the GAC surface and the NOM, or from all three of these effects. In our study, the mass of adsorbed TOC into GAC was 0.64 kg and 0.50 kg in the treatment trains with alum and iron coagulation, respectively. Since the filters were not fully exhausted during the study period (126 days), the expected increase in adsorbability was not observed.

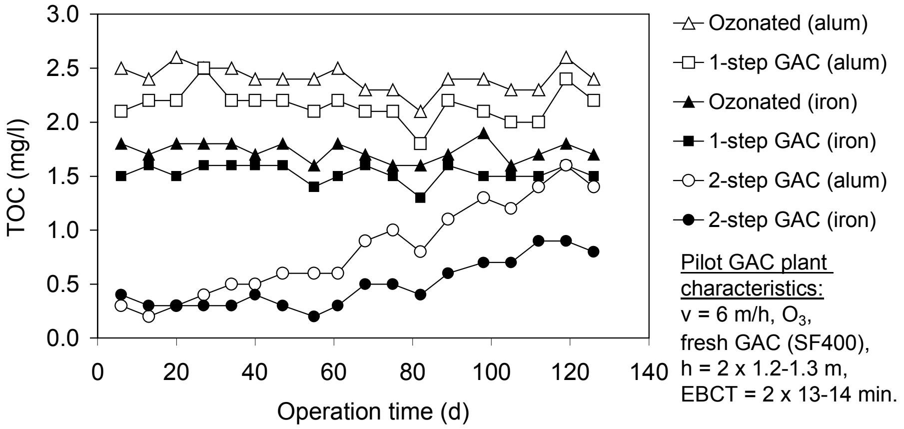


Figure 7. Effect of pretreatment coagulant type on breakthrough of TOC in two-step GAC filters.

4.1.2 Turbidity and soluble metal control

Optimizing coagulation for turbidity removal effectively reduces the density of bacteria and viruses in raw water. However, increasing the coagulant dose without consideration given to pH may result in increased bacteria, turbidity and soluble metal levels following sedimentation and sand filtration. A significant amount of dissolved manganese may release from iron salts unless it is specified as highly pure. An increased soluble metal concentration may cause a non-compliance with the drinking water standards (98/83/EY). On the other hand, optimum coagulation conditions for NOM removal may sometimes decrease turbidity, particle counts and residual metal concentrations (Vrijenhoek *et al.* 1998; Bell-Ajy *et al.* 2000).

The presence of flocs and particles on the GAC may disturb the substrate and oxygen diffusion to the biomass thus inhibiting the biological activity and NOM removal, which however is restored during filter backwash (Niquette *et al.* 1998, 1999). Conversely, in the more recent study Liu *et al.* (2001) could not observe any deterioration in biodegradable

NOM removal due to addition of nonbiological particles (kaolinite clay, dosage 1.5 mg/l) or coagulants (aluminium sulphate, dosage 3 mg/l).

If iron coagulation precedes GAC filtration, iron may accumulate onto the GAC through precipitation, ion exchange or chelation with organic compounds. During the thermal regeneration, iron may catalyze the oxidation of carbonaceous materials and thus change the pore structure of GAC and deteriorate the adsorption capacity. However, Cannon *et al.* (1997) showed that the presence of naturally occurring sulphur in GAC inhibits the catalyses of coal-based GAC gasification. Therefore, in practical applications, neither aluminium, which is not catalytic, nor iron accumulation do not seem to have a significant influence on the quality of GAC during regeneration. What is more important, also calcium aggressively catalyses gasification and dramatically alters the GAC pore structure despite the presence of sulphur (Knappe *et al.* 1992; Cannon *et al.* 1993). If re-alkalization is performed, it appears to be more important to minimize the calcium content in the spent GAC by postponing the lime addition after GAC filtration than to avoid the accumulation of iron onto GAC.

In our study, the two-step GAC filtration effectively reduced the concentrations of Fe and Al in the filter influent in the pH range of 6.5-7.5. The average Al reduction in five different pilot two-step GAC filter runs varied between 61-86 % and the average effluent Al concentration always remained below 0.02 mg/l (data not shown). The average Fe reductions in the pilot WTP study were 34 % and 69 % and the average effluent Fe concentration were 0.03 mg/l and 0.02 mg/l, at pH 7.0 and 6.5, respectively (data not shown). However, the differences resulting from the pH change were not statistically significant. Further research should determine the Fe removal rate at a lower pH range, the impact of precipitated metals on the bacterial activity, and the role of backwashing in the accumulation of metals on the GAC bed.

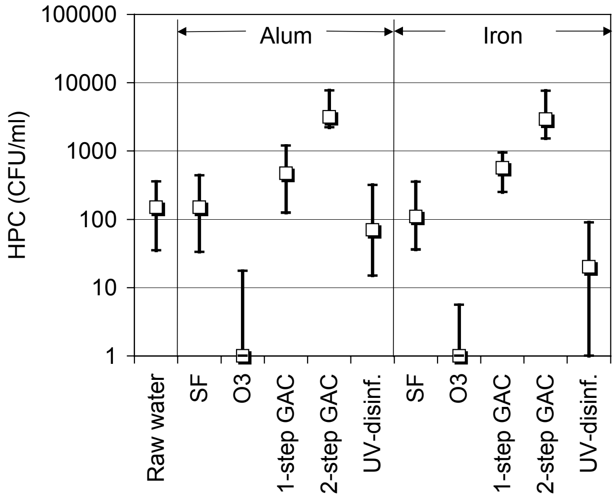
4.1.3 Conclusions

The results clearly reveal that in this type of process configuration NOM removal can be used as a primary criterion for the optimization of coagulation conditions, because the following filtration steps effectively remove the residual iron and aluminium, and they provide additional barriers against particles, residual metals and turbidity. In the studied water, ferric chloride sulphate proved to be far more effective in NOM removal than aluminium sulphate. Therefore, further investigations should focus on the site-specific practical aspects, such as flocculation time and sand-filter performance, which need to be taken into consideration when replacing alum with iron.

4.2 The role of ozonation

4.2.1 Primary disinfection

Although the proliferation of pathogens in the GAC bed is unlikely (AWWA 1981; Camper *et al.* 1986), a substantial number of coliforms and other organisms of public health concern may release from the GAC filter media unless appropriate disinfection is provided prior to biological filtration (Wilcox *et al.* 1983; Camper *et al.* 1986; Stewart *et al.* 1990; Morin *et al.* 1996). Recently, Carlson and Amy (2001) proposed that when utilities apply ozone for optimal disinfection, the following biological filtration would be operating close to optimum. Owing to the high source-water quality and the multiple disinfection barrier approach, disinfection was not considered a major criterion in optimizing the operational conditions for intermediate ozonation in our study. During the pilot study, in 59 % of the ozonated samples, no bacteria could be detected with the HPC analyses (n=17). Enhanced coagulation, *i.e.* a lower NOM level, was expected to improve the disinfection capacity of the following ozonation, because of a decrease in the rate of ozone decay (Urfer *et al.* 1999). Figure 8 illustrates the bacterial profile during the treatment (enhanced coagulation) and the effective destruction of cultivable bacteria in ozonation. This unpublished data was obtained from the two parallel pilot-WTP runs with ferric chloride sulphate (iron) and aluminium sulphate (alum) coagulation. A more detailed microbiological study is reported elsewhere (Niemi and Heiskanen 1997; Niemi *et al.* 1996, 1998; Heikkilä 1999).



SF = coagulated, sedimentated and sand-filtered water
O3 = ozonated water
Alum = aluminium sulphate
Iron = ferric chloride sulphate

Pilot GAC plant characteristics:
v = 6 m/h, O_3,
fresh GAC (SF400),
h = 2 x 1.2-1.3 m,
EBCT = 2 x 13-14 min.

Figure 8. The median HPC (20 ^{o}C, R2A, 7d, spread plate) and quartiles in pilot WTP.

4.2.2 Improving biodegradation

It is widely accepted that ozonation transforms NOM into a more biodegradable form, thus increasing the regrowth of bacteria in the distribution system if ozone is applied without subsequent filtration (Van der Kooij *et al.* 1989; Miltner *et al.* 1992; Kruithof *et al.* 1996). On the other hand, intermediate ozonation in optimal conditions may enhance the performance of the following biological filtration by producing a biologically available substrate for bacteria (Maloney *et al.* 1985; Sontheimer and Hubele 1987; DeWaters and DiGiano 1990).

In our study, the transformation of NOM into a more biodegradable form in ozonation was evidently seen as an increase in AOC and heterotrophic growth response (HGR) of coagulated and sand-filtered water (I, VII, VIII). Since *Aquaspirillum* strain NOX, which was used in the AOC assay, was more suited to utilizing oxygenated organic compounds (like carboxylic acids) and competes better for the available substrate in simultaneous cultivation than *Pseudomonas fluorescens* strain P17, the increase in AOC_{NOX} was more pronounced than the increase in AOC_{P17}. The increase in AOC_{NOX} was also associated with the increase in heterotrophic growth response (HGR), which suggests that AOC is related to the potential of this specific water to maintain the growth of bacteria (VIII). Similar results were obtained by Miettinen *et al.* (1998), who found a positive correlation between maximum HPC counts and AOC in ozonated waters. They suggested that the differences in microbial growth between the samples could be explained by the changes in the AOC level.

It was expected that optimizing the ozone dose would increase the biodegradation in the following biological filter and that the transformation of NOM could be optimized using an AOC assay (Van der Kooij *et al.* 1989; Miltner *et al.* 1992; Volk *et al.* 1993a, 1993b; Paode *et al.* 1997). In our first study, it seemed that the maximum AOC formation could be obtained with an ozone dose of 0.4-0.5 $mgO_3/mgTOC$ (I), which was similar to the observation of Orlandini *et al.* (1997b), who could not find any further increase in AOC when the ozone dose was increased from 0.6 to 1.2 $mgO_3/mgTOC$. In our more detailed study, comparing the effects of operational conditions on AOC formation, the optimum ozone dose was not that apparent. It was demonstrated that other factors than ozone dose alone were at least equally important operational variants in NOM transformation (VII, VIII).

The second pilot ozonation study (VII, VIII) confirmed our preliminary experimental results (I) and the findings of Volk *et al.* (1993), where a maximum AOC formation was obtained with a shorter contact time than is normally used for disinfection. The higher AOC formation was explained by the rapid reactions with ozone through direct and selective action. As the contact time is further increased, also part of the biodegradable NOM is mineralized. These results suggest that a short ozone contact time is favourable in the pretreatment of biological filtration if the operational criterion is maximum AOC formation.

The effects of ozonation pH and alkalinity are discussed later in Section 4.3.3.

Despite the massive increase in AOC during ozonation, the biodegradation of persistent NOM in the following GAC filter did not significantly improve compared to the non-ozonated GAC filter. The average TOC removal in the exhausted GAC filter increased only from 6 to 8 %, when ozone was applied (VI). Although most studies indicate enhanced NOM removal in biofilters following ozonation, similar minor or negligible improvement has been reported, *e.g.* by Wilcox *et al.* (1983), Fiessinger *et al.* (1983), Järvinen *et al.* (1990) and Pietari (1996). Our results suggest that the proportion of AOC in NOM was rather small even after ozonation, and that most of the NOM was composed of slowly biodegradable or recalcitrant organic compounds. In order to obtain a more reliable and complete picture of the changes in biodegradable NOM characteristics in different treatment steps and distribution system, BDOC analyses should be performed by separating rapidly and slowly biodegradable fractions during the assay.

The major parameter that affects the NOM transformation in ozonation is the characteristics of the influent NOM. As the chemical treatment changes the NOM characteristics, we wanted to evaluate whether the choice of coagulant has any effect on AOC formation in ozonation. Although a lower TOC level was achieved with ferric chloride sulphate than with aluminium sulphate, the AOC formation was not affected (VIII).

4.2.3 Reduction in reactivity

Ozone destroyed the UV-absorbing sites of NOM molecules, which was seen as a decline in UVA_{254}. The UV destruction was dependent on the applied ozone dose (I, VIII) being on average 48 % in the full-scale ozonation. Although ozone did not significantly mineralize NOM, the reduction in UVA_{254} does reduce the operation costs of the secondary disinfection; namely, UV irradiation benefits from the effective NOM removal in the preceding treatment train. Water that adsorbs a significant amount of UV light will need a higher UV irradiance or longer exposure time to achieve the same level of inactivation as water with lower UVA_{254} (Cotton *et al.* 2001).

As illustrated in Annex II, ozonation alone yielded an average 35 %, 37 % and 76 % reduction in AOX, SDSAOX and SDSTHM, respectively. Although ozonation transformed the THM and AOX precursors into a less reactive form, ozonation did not decrease either short-term (4 hours) or long-term chlorine demand (7 days).

In ozonated and GAC-filtered waters, UVA_{254} seemed to be a slightly better surrogate for predicting DBP formation than TOC (II). Similarly, Najm *et al.* (1994) showed that UVA_{254} is a better indicator for SDSTHM formation than TOC in chemically coagulated waters. On the other hand, Bell-Ajy *et al.* (2000) recently stated that in coagulation studies TOC would be a better surrogate for DBP precursor removal than UVA_{254}. Clearly, the prediction of DBP formation depends on the given process and NOM type. If the DBP formation is critical for the process design, the formation potential should not be estimated with surrogates only.

4.2.4 Decrease in molecular size

Although the average decomposition of TOC varied between 2 to 7 % depending on the pH and applied ozone dose (VIII), ozonation shifted the molecular size distribution slightly towards the smaller molecules (V, VI). However, the observed increase in the smallest molecular fraction was minimal compared to the increase in AOC.

In the studied water, the relative proportion of AOC in NOM was generally small, although it might seem high if only the AOC/TOC ratio is considered. However, it must be noted that AOC is not an absolute measure, but rather an indirect measure of biologically available energy that the NOM contain in water (Zhang and Huck 1996). Merely for convenience, the energy is expressed as acetate and oxalate carbon equivalents. If the decrease in AOC in exhausted GAC filters (or in the distribution system as shown later in 5.1) is compared to the corresponding TOC removal, the proportion seems much smaller.

It seems that, depending on the water characteristics, the relative amount of rapidly and slowly biodegradable NOM in different molecular fractions varies. LeChevallier (1999) ultrafiltered Mississippi River water samples and found AOC from both the filtrate and retentate, but in Putnam reservoir water the retentate practically did not contain any AOC. Merlet *et al.* (1991) suggest that the BDOC is composed of all sizes of molecules. Volk *et al.* (2000) found that the AOC was mainly composed of small molecules, while BDOC included larger molecules. Our HPSEC study could not relate the changes in AOC to the certain molecular fraction of NOM (V). This may have been a result of the inability of the size exclusion chromatography to detect the smallest fraction of NOM (Nissinen *et al.* 2001), or that the AOC is not formed from the smallest molecules only. Rather than simply saying that AOC is formed of small molecules, it seems that the complex and varying mixture of NOM macromolecules has sites in all sizes, which are more amenable to biodegradation than the other parts of different nature.

4.2.5 Effect on adsorbability

Several studies suggest that low molecular weight fractions of NOM are more adsorbable than larger, heavier fractions (Lee *et al.* 1981; Ødegaard *et al.* 1986; Semmens and Staples 1986; Sontheimer *et al.* 1988). In addition, in our study the adsorption of large molecules was slightly slower than with smaller ones (V). This effect is generally attributed to the inability of high-molecular weight compounds to enter the smaller GAC pores (McCreary and Snoeyink 1980). On the other hand, ozonation transforms NOM to more polar and hydrophilic, which result in a decrease in adsorbability of NOM in GAC filtration (Delaat *et al.* 1991; Orlandini *et al.* 1997a). Thus, the net effect of ozone on adsorbability may be either positive or negative. In our study, the slower adsorption kinetics of the ozonated NOM was seen in the lower amount of TOC that was adsorbed onto the GAC media in the ozonated run compared to the non-ozonated run (Table 2, VI).

4.2.6 Conclusions

After installing GAC, ozonation has a new role in the treatment process. When GAC is expected to improve the organoleptic quality of water and to reduce NOM that serves as DBP precursors, the justification of the ozone process is not a matter of course. Furthermore, if an effective disinfection barrier, such as UV irradiation, is introduced after GAC filtration, the operational criteria for the ozone process can be reconsidered.

Optimizing the operational conditions in intermediate ozonation increases the biodegradation rate in the following exhausted GAC filters. In our study, a short ozone contact time was sufficient for NOM transformation into a more biodegradable and less aromatic form, which provided the highest AOC values and probably the highest biological activity in the following exhausted GAC filters. On the other hand, ozonation reduced the adsorbability of NOM, which was not compensated by the minor increase in biodegradation of NOM.

If the objective is to minimize the release of AOC into the distribution system, this would result in ozone not being applied at all, because the effluent AOC in the non-ozonated run was higher than the effluent AOC in all the other runs. If the objective is the maximum formation of AOC in ozonation, this would result in short ozone contact time in a concentration normally used for primary disinfection. High influent AOC is followed by a maximum NOM degradation rate in GAC media, but also by higher effluent AOC level.

Since little is known about the characteristics of the remaining biodegradable NOM, the role of other nutrients and the possible risks of inadequate disinfection prior to GAC media, further studies should be conducted before the ozone contact time can be reduced from those normally used for disinfection.

4.3 Alkalinity and pH control

4.3.1 Coagulation

Controlling pH during the treatment process is needed to obtain optimal coagulation, ozonation, adsorption and chlorination conditions. Before the water is pumped into the distribution system, corrosion control is needed. A part of corrosion control in Helsinki Water is pH adjustment with carbon dioxide and lime addition. Alkalinity and pH also play a major role in optimal NOM and particle removal during coagulation. Sometimes, adding supplemental hydroxide ions and calcium from lime may be needed in low alkalinity cold waters to achieve optimal coagulation and flocculation conditions.

As discussed earlier, more efficient NOM removal in coagulation may be achieved by using ferric chloride sulphate. As an added benefit, calsium loading onto following GAC filter bed will be reduced (Frederick and Cannon 2001). The optimal coagulation pH for iron is lower than for alum and may be achieved without acid dosing in low alkalinity waters. This gives an opportunity for the WTPs, which normally involve lime addition before coagulation: the dosing point may be postponed to the later phase in the treatment train. However, careful consideration must be given to whether the changes in chloride-to-sulphate ratio affect the corrosivity of water.

4.3.2 Sand filtration

As summarized by the AWWA expert workshop (Carlson *et al.* 2000), the changes in flocculation conditions, such as pH, can affect the floc density and integrity by making it less dense and more fragile. This may result in floc carryover from the sedimentation basin to the sand-filters, which in turn results in shorter filter runs or premature filter breakthrough. In some Finnish WTPs, the pH is increased before sand filtration by adding lime in order to improve manganese removal efficiency (Wahlroos 1991). However, a change in pH can increase particle breakthrough as well as alum or iron carryover. Although we did not study the optimization of sand filtration, the evaluation of possible benefits and disadvantages of pH increase and optimum backwashing strategy could help optimize particle and soluble metal removal prior to ozone and GAC.

4.3.3 Ozonation

A reduction in pH reduces the ozone decay rate by decreasing the formation of hydroxyl radicals that can initiate the self-decomposition process (Langlais *et al.* 1991). This reduction results in a lower ozone dose required to achieve equivalent disinfection efficiency. On the other hand, bicarbonates and carbonates act as scavengers of the highly reactive hydroxyl radicals, and thus the increased alkalinity also has a stabilizing effect on molecular ozone. The destruction of DBP precursors is generally more effective at lower pH, but poorer performance in taste and odour control may be expected (Langlais *et al.* 1991).

In our study, the variation in ozonation pH and alkalinity in the studied range did not show any changes in NOM transformation (I, VII, VIII). The effect of alkalinity on AOC formation was studied by the addition of sodium bicarbonate into chemically pretreated (ferric chloride sulphate) and sand-filtered water (VIII). Increasing the alkalinity from 32 to 152 mg$CaCO_3$ did not affect AOC formation, nor did any other parameter show differences in the NOM reaction pathway. However, as expected, a lower ozone decomposition at lower pH was observed.

The results of other similar studies are somewhat confounding. Zoungrana *et al.* (1998) observed more intense formation of BDOC when the alkalinity was increased from 25 to 100 and finally to 250 mg$CaCO_3$/l. On the other hand, Paode *et al.* (1997) observed that the formation of AOC_{NOX} in ozonation increased as alkalinity decreased, but AOC_{P17} was not affected suggesting that a free radical mechanism may be more strongly influencing the formation of AOC. Higher UVA_{254} reductions have been observed at higher alkalinity (Legube *et al.* 1985; Paillard *et al.* 1989; Zoungrana *et al.* 1998). On the other hand, in high alkalinity waters (144 mg$CaCO_3$), a further increase in alkalinity did not increase UVA_{254} reduction in ozonation (Andrews and Huck 1994). Finally, Nawrocki and Kalkowska (1998) observed no differences in aldehyde formation with sodium bicarbonate addition before ozonation.

If the NOM transformation is affected by the ozone reaction pathway, as suggested by Legube *et al.* (1985), Paillard *et al.* (1989) and Zoungrana *et al.* (1998), adding bicarbonates or lowering pH should have further stabilized ozone. The results shown in Annex VIII indicate that ozone did primarily react through a molecular pathway and thus the increase in radical scavengers did not have any influence on the ozone reaction kinetics and NOM transformation. Although in our other study the addition of hydrogen peroxide after five minutes' ozone contact time did not show any changes in NOM transformation (Bicelli 1997), it would be interesting to establish the possible benefits of promoting rapid ozone decomposition through radical pathway. Recently, Speitel *et al.* (2000) found neither an improvement nor an inhibition in BDOC formation, when hydrogen peroxide was used together with ozone.

4.3.4 Biological filtration

In the studies of Moll and Summers (1999), varying the source water pH did not affect biomass concentration or the removal of NOM in biological filtration, but it did slightly change the microbial community structure. They suggested that enhanced coagulation followed by biological filtration at lower pH would have little impact on biofilter performance. In our study the filtration pH was always above 6.5 and thus effects of lower pH on biological activity were not assessed.

4.3.5 Adsorption

Decreasing pH generally improves NOM removal in GAC adsorption (McCreary and Snoeyink 1980; Weber *et al.* 1983). Semmens *et al*. (1986) found that an increase in the service life of the GAC adsorber was 2- to 4-fold depending on the target effluent value of TOC when the pH was decreased from 8.7 to 5.0. Nowack *et al.* (1999) found only a slight increase in service life when the pH was decreased from 7.1 to 6.0. Some of their explanations for the increased NOM adsorption capacity at lower pH were changes in the carbon surface charge and the degree of NOM dissociation. They suggested that it might be economically beneficial even to consider pH reduction prior to GAC adsorption.

In our study, the pH effect was not intentionally studied at the pilot plant. However, the effect can be seen from the breakthrough curves in Annex V, where the adsorption capacity of ozonated runs rapidly declines after 220 days. At that time, the influent pH was increased from 6.6 to 7.4. This indicates that at Helsinki Water's WTPs the pH reduction could increase the adsorption capacity of GAC adsorbers. The low adsorption pH is easily achieved by postponing the lime addition to the end of the treatment train.

It must be noted that the lime addition in re-alkalization may accumulate calcium in the GAC filter bed. Interestingly, it has been reported that the presence of calcium ions in solution substantially increases the adsorption capacity of GAC for humic substances (Weber *et al.*

1983) and fulvic acids (Randte and Jepsen 1982), which are both typical representatives of NOM. However, calcium present in the pore structure of GAC can accelerate the destruction of micropores during thermal regeneration (Cannon *et al.* 1993). If lime is added prior to GAC filtration, the utility may need to monitor the calcium build-up in the GAC filters.

4.3.6 Conclusions

Implementing GAC filtration together with enhanced coagulation using ferric chloride sulphate in the treatment train gives an opportunity for the utility to postpone the pH adjustment until the final stage, where corrosion control is performed. Before a lower pH is introduced step-by-step, careful attention must be paid to the expected changes in the treatment parameters, such as water corrosivity, soluble metals removal and metal accumulation onto GAC bed.

4.4 Ultraviolet irradiation and chloramination

4.4.1 Disinfection by-product control

One factor determining the extent of DBP formation is the chlorine dose required to achieve effective disinfection and to maintain chlorine residual in the distribution system. In order to assess the effect of ozonation and two-step GAC filtration on the required chlorine dose, we studied the chlorine decay kinetics. As shown in Annex II, two-step GAC filtration significantly reduced the short-term (4h) and long-term (7d) chlorine demand. Similarly, others have observed that biologically treated water had a lower chlorine demand than water that was ozonated alone (Bablon *et al.* 1988; Ventresque *et al.* 1990; Prévost *et al.* 1991; Cipparone *et al.* 1997). Cipparone *et al.* (1997) suggested that the chlorine demand is related to the TOC concentration in the water. However, we observed that the chlorine demand was less affected by the age of the GAC filter than the reduction in NOM (II), which suggests that GAC preferentially removes compounds susceptible to reactions with chlorine, such as amino acids (Prévost *et al.* 1998a). In practical applications, the required chlorine dose for the equivalent residual seems to be significantly lower, when GAC filtration is applied after ozonation. Accordingly, a lower disinfectant dose results in the lower DBP formation during treatment and in the distribution system.

Besides decreasing chlorine demand, the exhausted GAC filter contributed to a minor reduction in SDSTHM and SDSAOX (II). In the adsorptive phase, the removal was strongly dependent on the GAC age (II). Since the SDS tests were performed with a low chlorine dose

(0.6 mg/l) and short chlorine contact time before ammonia addition (30 minutes), the measured AOX and THM values in all the samples were extremely low. Even in the experiments without ammonia addition, the THM levels remained well below the European drinking water standard of 100 µg/l (98/83/EY). Thus, it is expected that, regardless of the selected the chlorination practice, after installation of GAC filtration, the THM formation rate will meet the goals set for the protection of public health.

4.4.2 Disinfection of the filter effluent

Biological filtration takes advantage of the bacteria that colonize the filter media. Thus, it also provides opportunities for the growth of indicator organisms and opportunistic pathogens. Some of them may be more resistant to disinfection, and GAC fines released from the filter media provide additional protection against disinfectant. The accumulation of pathogens poses a risk for public health, unless the biological filtration is followed by adequate disinfection, which eliminates the bacteria released from the filter media (Bouwer and Crowe 1988). Effective disinfection can be provided by UV irradiation or using relatively high doses of chlorine. UV disinfection seems to be a fascinating alternative, since even at low doses it provides high inactivation levels of the bacteria, viruses and protozoa being almost independent of the water pH and temperature (Parrotta and Bekdash 1998; Clancy *et al.* 1998; Bukhari *et al.* 1999; Craik *et al.* 2001; Modifi *et al.* 2001). Furthermore, the formation of known DBPs is minimal compared to other disinfectants. However, turbidity and TSS can inhibit UV inactivation by shielding the micro-organisms from the UV light. Little is known about the effectiveness of UV irradiation against the bacteria sheltered inside the GAC fines.

In our study with the pilot water main, the HPC of the GAC-filtered and UV-disinfected water was zero in 29 % of the samples (data not shown). In an other study, a pilot UV irradiation unit provided a 2-log reduction in HPC of the GAC effluent (Figure 8). Although, the effectiveness of UV irradiation has been addressed elsewhere in more detail (Niemi *et al.* 1998), it seems obvious that the effectiveness of secondary disinfection is largely dependent on the extent of particle release from the GAC media.

Although UV irradiation is expected to effectively disinfect water and contribute to a minor or negligible increase in biodegradable NOM (Shaw *et al.* 2000), we observed that the combination of UV disinfection, pH adjustment and chloramination contributed to a 41 % increase in AOC_{NOX}, but the AOC_{P17} was not significantly affected. In two-step GAC filter effluent (I) the AOC_{P17} and AOC_{NOX} averaged (± standard deviation) 46±33 μg-C/l and 99±14 μg-C/l, respectively. In the four samples taken at the same time (29.1, 25.3, 13.5, 9.9) from UV-disinfected, pH-adjusted and chloraminated water, the AOC_{P17} and AOC_{NOX} averaged 39±25 μg-C/l and 140±16 μg-C/l, respectively (III). Thus, it is possible that the regrowth of heterotrophic bacteria in UV-disinfected and chloraminated water is slightly higher than in the chloraminated water alone.

4.4.3 Conclusions

Introducing GAC filtration after ozonation provides additional removal of DBP precursors and decreases the chlorine demand required for the equivalent disinfection efficiency. The removal efficiency is limited with the exhausted GAC, and thus the DBP formation can be controlled by shortening the GAC service time. However, if chloramination is practiced *i.e.* the DBP formation is retarded by a ammonia addition, the resulting DBP levels in the studied treatment process remain well below the guidelines.

Effective disinfection is necessary to prevent the escape of harmful bacteria from the GAC media into the finished water. If low doses of free chlorine or chloramines were normally applied as a secondary disinfectant, UV irradiation would provide an additional barrier against the micro-organisms of health concern. However, little is known about the effectiveness of UV irradiation against bacteria sheltered inside the carbon fines. More research is needed also to determine the impacts of UV irradiation on the bacterial regrowth in the distribution system.

5 WATER QUALITY CHANGES IN THE DISTRIBUTION SYSTEM

5.1 Proliferation of heterotrophic bacteria and AOC degradation rate

The biofilm accumulation in drinking water systems is stimulated by the biodegradable compounds present in drinking water or dissolved from the construction materials of the piping system. These compounds serve as a source of nutrients and energy for the growth of bacteria. If the nutrients are effectively removed in the treatment train, it is expected that the biofilm formation in the pipe walls be reduced and the increase in the HPC with respect to residence time be limited. It has been shown that extremely low concentrations of biodegradable compounds can maintain biofilm activity in the distribution system (Sibille *et al.* 1997, 1998; Van der Kooij *et al.* 1995, 1999). However, in many cases other factors, such as water temperature, disinfectant type and residuals, corrosion control, hydraulic conditions and selection of pipe material may be more important factors in maintaining biofilm activity (Volk and LeChevallier 1999). Especially, unlined ferrous metal pipes and its corrosion products in the distribution system dramatically influence the composition, activity and disinfection resistance of biofilm bacteria (Norton and LeChevallier 2000, Butterfield *et al.* 2002).

In Section 3.2.2, it was demonstrated that two-step GAC filtration efficiently decreased AOC in ozonated water, which was seen as low levels of AOC entering the simulated water main (I, III). In the simulated water main, a significant decrease in AOC degradation rate and HPC was achieved when two-step GAC filtration and UV disinfection were employed, suggesting that GAC filtration increases the biological stability of distributed water (III). A decrease in biodegradable NOM degradation rate in the distribution system has been attributed to the degree of biological stability of drinking water in other studies, too (LeChevallier *et al.* 1987, 1991; Van der Kooij 1990, 1992; Laurent *et al.* 1993; Gatel *et al.* 1995; Miettinen *et al.* 1997). However, TOC was not sensitive enough to detect the decreased NOM degradation rate. No significant differences due to GAC filtration and UV disinfection were detected for decreases in TOC in the simulated water main. It is probable that increasing the residence time in the water main would have increased the TOC degradation. Nissinen *et al.* (2002) took samples from the full-scale distribution system (10 km from waterworks) and observed a 0.2 - 0.3 mgTOC/l reduction. This is twice as high as observed in pilot water main with similar post-ozonated water (III). These results indicate that the biodegradable proportion of

NOM is rather small and persistent in its nature. It remains questionable whether BDOC analyses could have shown any changes in the distribution system.

The measurement of AOC is based on the general observation that in most waters, carbon limits the growth of heterotrophic bacteria. However, a few authors have shown that in some waters, phosphorus may be a minimum factor for growth (Miettinen *et al.* 1997b; Sathasivan *et al.* 1997). Since all our samples analyzed for AOC were supplemented with phosphorus and other inorganic nutrients, this study did not address the issue of whether phosphorus was the limiting nutrient, but the AOC described only the effect of bio-available carbon on the potential growth of *Ps. fluorescens* strain P-17 and *Spirillum* strain NOX pure cultures. Interestingly, Miettinen *et al.* (1997a, 1997b) observed that the AOC in distributed water did not correlate with HPC and that the addition of phosphorus to a finished water sample of Pitkäkoski WTP increased bacterial growth. The nutrient limitation has already been discussed in Section 3.2.6, and our results are clearly not conclusive. It is possible that both the decrease in microbially available phosphorus and organic carbon resulted in the decreased HPC in the distribution system. It is also possible that the turnover rate of phosphorus is much higher than the biodegradation rate of carbon-containing substrates and thus phosphorus has a limited role in the biofilms of the distribution system. Also, it must be noted that an efficient corrosion control program will result in increased disinfection efficacy and lower biofilm densities (Abernathy and Camper 1998; Appenzeller *et al.* 2001). Therefore, it is possible even in the phosphorus-limited distribution systems, that corrosion control with phophate addition would result in lower biofilm densities.

Because of the complexities inherent of distribution systems, water quality models are essential tools needed to describe fundamental processes. The more recently developed multi-species models describe water quality reactions using sets of interdependent mass-balance equations capable of tracking multiple species. The first multi-species model designed for drinking water system is the SANCHO model (Servais *et al.* 1995). It contains mass-balance equations describing microbial synthesis, biodegradable NOM utilisation, chlorine reactivity with NOM and disinfection processes. It calculates the biomass concentrations in the water phase and attached to pipe surfaces, but is limited to the analyses of straight pipes of decreasing diameter. Recently, SANCHO was applied to full-scale distribution systems (Laurent *et al.* 1997). Other multi-species models are described in Dukan *et al.* (1996) and Piriou *et al.* (1998). In the future studies, fundamental processes should be examined at the laboratory and distribution system scale using multi-species models as a tool for developing water quality improvement plans.

5.2 Decay of monochloramine

The decay of chloramine in distribution system can be attributed to the reactions of chloramine with organic matter, biofilm and construction material. Several recent studies have highlighted the importance of maintaining a disinfectant residual in the distribution system (Escobar *et al.* 2001; Zhang and DiGiano 2002; Butterfield *et al.* 2002) It seems that in most cases the presence of disinfectant residual is the most effective measure in minimising biofilm development and HPC levels. Prévost *et al.* (1998b) suggests that in cold waters, maintaining a high concentration of disinfectant residual may be sufficient alone to compensate higher concentration of biodegradable compounds.

Based on the laboratory tests, where the reduction of long-term chlorine demand (seven days, Cl_2:TOC = 2:1) was about 30 % in two-step GAC filtration (II), it was expected that introducing GAC would decrease the chloramine decay in the distribution system. The simulated distribution system study showed similar results: GAC filtration slightly increased the stability of chloramine in the simulated water main (III). Thus, the equivalent chloramine residual can be achieved in the consumer tap by applying a lower chloramine dose. These results conflict with the observations of Volk and LeChevallier (1999), who observed in annular rectors that biological filtration had little effect on chloramine consumption, although it improved the stability of free chlorine.

5.3 Incomplete nitrification

The use of chloramines in the secondary disinfection may result in the regrowth of ammonia-oxidizing bacteria (AOB) in the distribution systems. AOB obtain the energy for their growth from the oxidation of ammonia to nitrite, which is available from the decay of chloramine. The nitrite may exert to increased chloramine demand thus releasing more ammonia. This may allow the rapid destruction of disinfectant residual and possible pathogen accumulation in the distribution system (Valentine 1985). In addition, AOB may enhance the growth of heterotrophs by the production of biodegradable soluble microbial products (Watson *et al.* 1981, Regan *et al.* 2002). It has been estimated that, in the United States, two-thirds of medium and large chloraminating utilities experience nitrification (Wilczak *et al.* 1996). Lately, it has been demonstrated that the AOB are widely present in the deposits of the Finnish drinking water distribution systems (Lipponen *et al.* 2002). Recommended approaches to prevent and control nitrification include decreasing detention time, increasing the pH, decreasing the temperature, decreasing TOC concentrations, increasing chloramine residual, increasing chlorine-to-ammonia ratio and decreasing the excess ammonia concentration. Recently, it has been shown that maintaining the finished water pH at 9.3 (Skadsen 2002) and the addition of chlorite to the treatment plant effluent (McGuire *et al.* 1999) have a significant potential for controlling nitrification. However, both techniques have serious drawbacks, which need to be carefully considered, but are not discussed here.

In our study, GAC filtration contributed to increased bacterial nitrification in the simulated water main (III). After a three-month acclimatization period, AOB started to convert ammonia to nitrites more intensively. The decreased nitrite concentration towards the end of the water main together with increased nitrate concentrations indicates that nitrite was further converted to nitrate. Interestingly, the oxidizing of nitrite to nitrate occurred during the normal residence time in a large distribution system.

Nitrification occurred despite the constant low water temperature (7-9 ^{o}C). Generally, a high temperature and long residence time favour the proliferation of slowly growing nitrifying bacteria (Larson 1939; Ike *et al.* 1988; Wolfe *et al.* 1988; Lieu *et al.* 1993), although nitrification episodes in temperature conditions below 15 ^{o}C have also been reported (Wolfe *et al.* 1990; Cunliffe 1991; Odell *et al.* 1996; Pintar and Slawson 2002). The results obtained confirm that nitrification episodes may occur even under cold water conditions in the decreased water flow sections.

After installing full-scale GAC filtration at Vanhakaupunki WTP the nitrite levels started to increase in its distribution system, too. Statistical analysis demonstrated that, after three months of operation, nitrite levels were significantly higher than in the previous years (Vahala and Laukkanen 1998). At the same time in Pitkäkoski WTP distribution system, which was supplied with postozonated water, the nitrite levels did not differ significantly from the previous years.

The mechanism contributing to increased nitrification after GAC filtration needs further investigation. Previously, Skadsen (1993) observed that GAC contributed to nitrification in the Ann Arbor Plant. However, they supplied the GAC filter with excess ammonia in prechloraminated water, thus enabling nitrifying bacteria to proliferate already in the GAC filters. On the other hand, Odell *et al.* (1996) proposed that removing NOM in the treatment plant could control nitrification. Conversely, Strauss and Lamberti (2000) observed that organic carbon addition decreased the nitrification rate but increased total microbial activity in the stream sediment. In our study, exhausted GAC did not remove NOM efficiently and probably the low difference in TOC between the mains had no significant influence on nitrification. Rather, decreased competition between nitrifying and heterotrophic bacteria for space, slightly lower pH (optimal growth conditions for AOB varies between 7.2-8.5), the introduction of a shelter inside the carbon fines and a slightly higher excess ammonia concentration (lower chlorine-to-ammonia ratio) may have caused a higher proliferation of nitrifying bacteria in the GAC-filtered water main. It appears that the GAC filtration shifted the increase in heterotrophic bacteria to the treatment plant and thus reduced the interference for the growth of autotrophic nitrifying bacteria in the water main. In a bench-scale study, Pintar and Slawson (2002) could not observe any specific relationship between heterotrophs and AOB, but noted that the competition for space may affect the growth opportunities of these organisms. This was observed in the ammonia-seeded BAC column, where the nitrifiers increased as the heterotrophs decreased (Pintar 2002).

Recently, the role of protozoa as a predator in controlling biomass growth and accumulation in the distribution system has been discussed. Sibille *et al.* (1998) observed in pilot-scale pipe loops that *E.coli* was lost from GAC-filtered water more rapidly than from the nanofiltered water perhaps because of the grazing activity. The role of protozoa in AOB accumulation needs further research.

The increases in nitrite and nitrate together with the consumption of ammonia indicated the occurrence of nitrification in a simulated water main. In addition to increased oxidized nitrogen concentrations, nitrification has been associated with increased HPC, the accelerated decay of chloramines and decreased pH, alkalinity and DO concentration (Feben 1935; Larson 1939; Hill 1946; Ike *et al.* 1988; Odell *et al.* 1996). However, in our study the non-specific measurements such as pH, alkalinity and DO proved unsatisfactory for detection of the nitrification. For example, in post-ozonated water main the average DO concentration decreased from 13.2 mg/l to 11.8 mg/l during the residence time, while in GAC-filtered water main the average DO concentration decreased from 12.2 mg/l to 10.7 mg/l, respectively. On the other hand, the use of HPC as an indicator in this study failed as GAC filtration decreased the HPC by removing nutrients for the growth of heterotrophic bacteria. Furthermore, in the presence of low disinfectant residues in the distribution main, the accelerated decay of chloramines was not observed.

5.4 Conclusions

The application of ammonia in chloramination to provide more persistent chlorine residual and to control the production of DBPs stimulates nitrification in the distribution system. It was expected that the presence of nitrites would decrease the chloramine residual and thus increase the heterotrophic bacterial population in the distribution system. The most interesting finding in our study was that, in GAC-filtered and UV-irradiated water, the nitrification effect seemed to be more pronounced. In this water, the increased growth of nitrifying autotrophs can be explained by the fact that they compete with heterotrophs for biofilm space. Owing to their higher growth rate and lower sensitivity to shear forces, heterotrophs generally win the competition resulting in minimal nitrification. The competition was probably not for the dissolved oxygen as it was present in excess in the aqueous phase.

Although the removal of nutrients alone does not inhibit the biofilm growth, introducing GAC filtration into the treatment scheme transfers most of the AOC reduction from the distribution system into the treatment plant, where biological processes are better controlled. In contrast to the published literature, we could not observe that the growth and metabolism of autotrophs would significantly stimulate the growth of heterotrophic biomass by forming organic products. Neither could be observe any decrease in chloramine concentration owing to nitrification, which could have been due to low chloramine levels in our study. Rather, the removal of reactive NOM species aided in the maintenance of chloramine residual and increased the availability of chloramines to inhibit the biofilm development in the system. However, it can be expected that by minimizing the nitrification, the growth of heterotrophic bacteria is further reduced. More research is needed to determine the interactions between heterotrophs, autotrophs, protozoa and NOM in the proliferation of nitrifying bacteria.

6 FUTURE OPTIONS

6.1 Process alternatives

This chapter discusses the different approaches in the development of Helsinki Water's treatment processes. Since the number of possible process combinations is almost unlimited, only the following viable alternatives are selected for the discussion (Table 7):

0. Conventional treatment + ozone alone (existing process before modification)
1. Conventional treatment + ozone + exhausted GAC + UV irradiation
2. Conventional treatment + ozone + adsorptive GAC + UV irradiation
3. Conventional treatment + ozone + two-step GAC + UV irradiation (selected process combination)
4. Conventional treatment + adsorptive GAC + UV irradiation
5. Conventional treatment + UV irradiation

Table 7. Summary of benefits and adverse effects of selected process combinations after conventional treatment (O_3 = ozonation, GAC = adsorption, BAC = exhausted GAC).

	O_3 (0)	O_3+BAC+UV (1)	O_3+GAC+UV (2)	O_3+BAC+GAC+UV (3)	GAC+UV (4)	UV (5)
AOC in finished water	>>	0	0	0	<	<<
Heterotrophic regrowth in distribution system	>>	<	<	<	n.a.	n.a.
Nitrification in distribution system	>	>>	>>	>>	n.a.	n.a.
AOX in finished water	<	<	<<	<<	<	0
THMs in finished water	<<	<<	<<	<<	<	0
Decay of chloramines in distribution system	<	<<	<<	<<	<<	<
Disinfection efficiency	>	>>	>>	>>	>	>
Risk of pathogen intrusion	<<	<	<	<	0	<<
Taste and odour in finished water	<<	<<	<<	<<	<<	0

<< very low, < low, 0 moderate, > high, >> very high, n.a. not analysed

Every treatment alternative is followed by corrosion control and secondary disinfection. In corrosion control, lime and carbon dioxide are added to provide optimal pH and alkalinity. A low dose of monochloramine (< 1 mg/l) is used for secondary disinfection to provide a more persistent chlorine residual and to control the production of DBPs in the finished water. Conventional treatment consists of coagulation, flocculation, sedimentation and sand-filtration with the necessary pH adjustment to obtain optimal operation conditions.

6.2 Existing treatment process

During the study ozonated water was supplied into the distribution system without subsequent filtration. As indicated earlier, the driving force for the interest in GAC was the deterioration of postozonated water quality in the distribution system, especially in Helsinki city centre, where the use of water has decreased. The deterioration of water quality was seen as an increased biofilm formation and harmful deposits, which increased the replacement rate of water meters.

It is well known that ozonation increases the biodegradable portion of NOM and thus increases the regrowth of bacteria in the distribution system. Nowadays, it is widely accepted that ozonation should always be followed by a filtration step. However, conventional treatment followed by ozonation alone is effective in improving the organoleptic quality and transforming DBP precursors into a less harmful form. Furthermore, it provides effective disinfection barrier against bacteria, viruses and protozoa.

6.3 Ozone followed by biological filtration

When exhausted GAC is used for the biological stabilization of drinking water it is often referred to as biological activated filtration. However, in this thesis, this term is intentionally not used, because the biological activity had a limited role in a process with low temperature and low influent flux of nutrients. Despite extreme conditions for GAC performance, the AOC was significantly reduced in the exhausted GAC filter. Although the relationship between decreased AOC and decreased HPC in the simulated distribution main is not conclusive, it can be concluded that exhausted GAC decreased the regrowth of heterotrophic bacteria in the distribution system. Although, the average AOC reduction in the exhausted GAC filter was more than 50 % of AOC, the water was not biologically stable and needed a disinfectant residual to limit bacterial growth.

Exhausted GAC contributed to increased nitrification in the distribution system. In nitrification, AOB may produce soluble microbial products, which increase the growth of heterotrophs. Thus, the benefit from introducing GAC is partly lost, if nitrification cannot be controlled. One key factor that may affect the nitrification occurrence is the release of GAC fines, which needs further investigation. Although the low nitrite levels is not an issue of public health protection, the factors that contribute to increased AOB proliferation need to be established.

Exhausted GAC does not significantly decrease the DBP precursors from ozonated water. However, it does decrease the chloramine decay rate and thus decreases the dose needed for the same residual.

Although ozone is a powerful oxidant, an effective disinfection after GAC provides additional safety. The efficiency of UV irradiation against micro-organisms sheltered inside the GAC fines is poorly known and therefore the theoretical risk of pathogen intrusion exists. Both the ozone and GAC improve the organoleptic quality of water and the high level of customer satisfaction can be achieved unless a large amount of GAC fines reach the customer tap.

6.4 Ozone followed by adsorptive filtration

Compared to exhausted GAC, fresh GAC efficiently removes NOM, *i.e.* DBP precursors. The efficiency of adsorptive filters is strongly dependant on the selected GAC type and regeneration frequency. Fresh GAC efficiently removes TOC, UVA254, chlorine demand and DBP precursors, but the rapid deterioration of adsorptive capacity takes place soon after the installation. Since the adverse effects of NOM in a distribution system are rather difficult to assess, an arbitrary operation criterion is often set for the utility. However, the high regeneration cost of GAC would justify more accurate research on the expected adverse effects.

Except for the better performance in NOM removal, the adsorptive GAC has limited advantages over exhausted GAC. Fresh GAC provides an additional barrier against accidental pollution and rapid changes in influent quality, but on the other hand, it may release more GAC fines into the finished water than exhausted GAC.

6.5 Selected process modification

This thesis studied the ozonated two-step GAC process, where optimal process performance for NOM removal was expected. In contrast to the published literature, the first step filter with exhausted GAC did not extend the bed life of the following adsorber when the first-step GAC filter was operated with fully exhausted GAC. If shorter regeneration frequency were applied, also the first step GAC filter would have some adsorption capacity left, and the concept of two sequential filters would probably decrease the GAC usage rate.

Despite the long contact time in two sequential GAC filters, the AOC in the effluent remained higher than before ozonation. From the standpoint of efficient AOC removal, the long EBCT in the GAC filtration cannot be justified. However, it is possible that slowly biodegradable NOM necessitates a longer contact time. Unfortunately, this was not studied and therefore no conclusive recommendations about the optimal EBCT can be given. However, the latest research and constructed GAC applications seem to favour a short contact time of less than 10 minutes.

6.6 Switching off the ozonation

The use of ozonation in water treatment must be judged as a trade-off between its beneficial effects and its potential adverse effects. In the history of Helsinki Water's treatment processes, ozone was first used for primary disinfection and for improving organoleptic quality. The raw water quality has gradually improved, and nowadays ozone is mainly used for primary disinfection.

When UV irradiation was introduced in the existing treatment train, effective disinfection was provided against bacteria, viruses and protozoa. Therefore, the role of ozone can be reconsidered. However, at the same time, the supporter of biological activity, namely GAC, was introduced. Preceding ozonation provides a barrier against the colonization of filter media with micro-organisms of public health concern. Although conventional treatment effectively reduces the micro-organisms from the raw water, it is possible, although unlikely, that a GAC filter would colonize opportunistic pathogens. If the pathogens are attached to the surface of GAC fines and released into the finished water, it is possible that UV light cannot deactivate them and thus they pose a risk for public health.

When GAC follows conventional treatment without intermediate ozonation, the biological activity in the GAC media is not intentionally encouraged, although it always exists. In a non-ozonated GAC filter, NOM is as efficiently removed as in ozonated filter media, but the AOC is slightly increased. The impact of increased AOC on the HPC in the distribution system was not studied, but it could be expected that the regrowth of heterotrophs is at least smaller than in postozonated water.

If the ozone is switched off, the total decrease in DBP precursors and UVA_{254} in the GAC is smaller than with the combination of ozone and GAC. When chloramines are applied, the

DBP formation is relatively low in all the process alternatives. However, the higher UVA_{254} increases the UV dose needed for the equivalent disinfection efficiency. The chlorine demand tests suggested that the chloramine decay rate in the distribution system is not affected.

6.7 Replacing ozone with UV irradiation

Changing the primary disinfectant provides a low-cost alternative to the previously discussed process modifications. Although, chloramination was considered as a matter of course in the secondary disinfection, the primary disinfection can be performed effectively with UV irradiation alone. After conventional treatment alone, all the parameters remain below the drinking water standards, and it would be possible to deliver water without ozone and GAC treatment. However, this approach reduces the multiple barriers in the treatment and thus the risk of customer complaints increases and the level of public health protection decreases.

6.8 Closing words

Helsinki Water has upgraded its treatment process by introducing two-step GAC filtration and UV disinfection after conventional treatment and ozonation. This process configuration provides additional barriers against unexpected conditions. An example of such a situation is the change in raw water source, which took place during autumn 2001, when the 120 km-long raw water tunnel from Lake Päijänne was under rehabilitation for four months. During this time, raw water was taken from River Vantaa, which has higher turbidity, NOM concentration and coliforms counts. In such circumstances, the additional barriers in the treatment train can guarantee customer satisfaction, although in a normal situation good water quality could be achieved with a conventional treatment process.

The decision of upgrading the treatment process is always partly political, and it depends on the willingness to pay for the extra safety and comfort. In recent years, the deterioration of water quality in distribution systems has gained more interest among water professionals. Partly this is because of the ageing infrastructure, but also owing to efficient new treatment methods, such as nanofiltration, which produce ultrapure water. It would be interesting to know how far it is economically reasonable to further improve finished water quality, and when it is time to direct the efforts and investments towards the maintenance of the drinking water distribution system.

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