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Effects of Conventional Treatment, Tertiary Treatment and Disinfection Processes on Hygienic and Physico-Chemical Quality of Municipal Wastewaters

Doctoral dissertation

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ABSTRACT

Conventional wastewater treatment, primary and secondary treatment processes with simultaneous phosphorus precipitation, is the most common process combination used in Finnish wastewater treatment plants (WWTPs). It typically eliminates most of the organic load and phosphorus, as well as part of the enteric microorganisms present in raw wastewater. Municipal secondary effluents still contain some organic matter and nutrients, which cause eutrophication and increase the oxygen demand on the natural waters. The presence of enteric microorganisms in the wastewater discharges decreases the hygienic quality of natural waters. Over-loading situations of WWTP can significantly decrease the efficiency of wastewater treatment and even force WWTP to by-pass untreated wastewaters directly into natural waters, causing adverse environmental effects. Conventionally treated wastewater may not meet the authority requirements set for wastewater discharges or wastewater reuse, especially in the future, as the regulation of wastewater discharges may become stricter in many locations. The efficiency of wastewater treatment can be improved by tertiary treatment and disinfection processes.

The aim of this study was to evaluate the efficiency of different wastewater treatment processes on the removal of enteric microorganisms, phosphorus and organic matter from municipal wastewaters. The treatment efficiency of conventional biological-chemical wastewater treatment processes was studied in four Finnish municipal WWTPs. The effect of tertiary rapid sand filtration (RSF) and dissolved air flotation (DAF) processes, as well as chemical and biological-chemical contact filtration processes, on wastewater quality was studied in pilot-scale experiments. The applicability of the DAF process for treatment of primary wastewater effluents was also studied in pilot-scale experiments to assess the applicability of the process for treatment of WWTP by-passes. Some experiments were carried out in two full-scale tertiary DAF plants. The disinfection efficiencies of peracetic acid (PAA), hydrogen peroxide (H_2O_2), sodium hypochlorite (NaClO) and ultraviolet (UV) disinfection treatments as well as the synergistic effects of combined use of chemical disinfectant and UV were investigated in laboratory-scale experiments, followed by pilot-scale PAA disinfection experiments of municipal primary, secondary and tertiary effluents.

Primary and secondary wastewater treatment with simultaneous phosphorus precipitation achieved around 95 % reductions of organic matter and phosphorus from the municipal wastewaters. The numbers of enteric microorganisms were typically reduced by between 90 and 99.9 %, but the secondary effluents still contained high microbial numbers, including pathogenic salmonellae. The tertiary RSF or DAF processes efficiently removed residual organic matter and phosphorus, and removed 90-99 % of enteric microorganisms from the secondary effluents. Increasing the coagulant dose (from 2 to 10 gAl^{3+}/m^3) and the dispersion water recycle ratio (from 11 to 22 %) improved the purification results, whereas changing the flocculation conditions (G-value, retention time) or increasing the hydraulic surface load (from 5 m/h to 10 m/h) did not clearly affect the tertiary DAF process efficiency. The DAF process achieved significant reductions of enteric microorganisms, phosphorus and organic matter in the treatment of primary wastewater effluents, demonstrating that the process can tolerate high loads of suspended solids and could be used for the treatment of WWTP by-pass wastewaters during the WWTP over-loading situations.

Peracetic acid was demonstrated to be an efficient disinfectant against enteric microorganisms in municipal primary, secondary and tertiary wastewater effluents. The combined PAA/UV treatments showed high disinfection efficiency and synergy benefits, while hydrogen peroxide and sodium hypochlorite showed low efficiencies in laboratory-scale disinfection experiments with organic matter rich synthetic wastewater. The results of the present study suggest that the combination of PAA and UV disinfection could increase the efficiency and reliability of wastewater disinfection processes.

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CAB Thesaurus: waste water; waste water treatment; biological treatment; chemical treatment; disinfection; peracetic acid; sodium hypochlorite; hydrogen peroxide; ultraviolet radiation; synergism; filtration; flotation; coagulation; flocculation; faecal coliforms; Enterococcaceae; bacteriophages; Salmonella; phosphorus; quality

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Kuopio, August 2007

Jari Koivunen

ABBREVIATIONS

AOP	Advanced oxidation process
ATCC	American Type Culture Collection
BOD	Biochemical oxygen demand
C	Concentration
CFU	Colony forming unit
ClO_2	Chlorine dioxide
Cl_2	Chlorine gas
COD	Chemical oxygen demand
Cp	Coliphage
$C \times t$	$C \times t$ product; C is the disinfectant dose and t is the contact time
DAF	Dissolved air flotation
DBP	Disinfection by-product
DNA	Deoxyribonucleic acid
EC	Enterococci
FC	Faecal coliform
FS	Faecal streptococci
G	Mean velocity gradient
HPC	Heterotrophic plate count
H_2O_2	Hydrogen peroxide
MF	Microfiltration
MPN	Most probable number
NaOCl	Sodium hypochlorite
NF	Nanofiltration
NTU	Nephelometric turbidity unit
$OH\cdot$	Hydroxyl radical
O_3	Ozone
PAA	Peracetic acid
PACl	Polyaluminium chloride
PFU	Plaque forming unit
pK_a	Negative logarithm of acidity constant
P_{tot}	Total phosphorus
PW	Peptone water
R	Dispersion water recycle ratio
RNA	Ribonucleic acid
RO	Reverse osmosis
RSF	Rapid sand filtration
SFS	Finnish Standards Association
S_h	Hydraulic surface load
SS	Suspended solids
SSF	Slow sand filtration
t	Contact time
TC	Total coliform
THM	Trihalomethane
TOC	Total organic carbon
UF	Ultrafiltration
U.S.EPA	United States Environmental Protection Agency
UV	Ultraviolet
WWTP	Wastewater treatment plant

LIST OF ORIGINAL PUBLICATIONS

The thesis is based on the following articles, referred in the text by the Roman numerals I-IV.

I	Koivunen, J., Siitonen, A., Heinonen-Tanski, H. 2003. Elimination of enteric bacteria in biological-chemical wastewater treatment and tertiary filtration units. Water Research, 37, 690-698.
II	Koivunen, J., Sutinen, P., Heinonen-Tanski, H. 2005. Inactivation of enteric microorganisms with chemical disinfectants, UV irradiation and combined chemical/UV treatments. Water Research, 39, 1519-1526.
III	Koivunen, J., Heinonen-Tanski, H. 2005. Peracetic acid (PAA) disinfection of primary, secondary and tertiary treated municipal wastewaters. Water Research, 39, 4445-4453.
IV	Koivunen, J., Heinonen-Tanski, H. Dissolved air flotation (DAF) for primary and tertiary treatment of municipal wastewaters. (in press)

CONTENTS

1 INTRODUCTION

Conventional wastewater treatment processes, primary and secondary treatment with simultaneous phosphorus precipitation, the process combination most commonly used in Finnish wastewater treatment plants, typically eliminate around 95 % of the organic load and phosphorus present in raw wastewater. Even though typical microbial reductions are usually at the level of 90-99.9 %, secondary treated wastewaters may still contain high numbers of enteric microorganisms, including pathogenic species.

Conventionally treated wastewater may not meet the requirements set in many locations for wastewater discharges or wastewater reuse. Municipal secondary effluents still contain organic matter and nutrients, which cause eutrophication and elevate the oxygen demand on the natural waters. Sewage discharges also increase pathogen contamination of natural waters. This may also result in the appearance of waterborne infections, if the polluted surface waters are used as raw water for drinking water production, for recreational purposes, for seafood harvesting or for agricultural uses such as irrigation or drinking water for animals. In situations where the wastewater treatment plant (WWTP) becomes overloaded, the efficiency of wastewater treatment may decline dramatically, and in some situations the WWTP may be forced to discharge untreated wastewaters directly into natural waters, which can have adverse environmental consequences.

The quality of wastewater effluents can be improved by tertiary treatment processes, such as rapid sand filtration (RSF) or dissolved air flotation (DAF) processes. Elimination of pathogenic microorganisms can be further improved by disinfection of the wastewater effluent. These processes could also be used for treatment of wastewater discharges during WWTP by-pass situations to reduce their harmful effects on the natural waters.

The aim of this study was to evaluate the efficacy of different wastewater treatment processes on microbiological and physico-chemical wastewater quality. The treatment efficiency of conventional biological-chemical wastewater treatment processes (primary and secondary treatment with simultaneous phosphorus precipitation) was studied in four Finnish municipal WWTPs. The effect of tertiary RSF and DAF processes on wastewater quality was studied in pilot-scale experiments. The applicability of the DAF process for treatment of primary effluents was also studied in pilot-scale experiments to assess the applicability of the process for treatment of WWTP by-passes. In addition, DAF process was studied as tertiary treatment process in two full-scale WWTPs. The disinfection efficiencies of peracetic acid (PAA), hydrogen peroxide (H_2O_2), sodium hypochlorite (NaOCl) and ultraviolet (UV) disinfection treatments as well as the synergistic effects of combined use of chemical disinfectant and UV were investigated in laboratory-scale experiments, followed by pilot-scale PAA disinfection experiments in a municipal WWTP.

2 REVIEW OF THE LITERATURE

2.1 Wastewater quality

Wastewater can be characterized in terms of its physical, chemical and biological composition. The principal contaminants found in wastewaters are summarized in Table 1. The quality and quantity of wastewater entering the wastewater treatment plant typically varies widely and is affected by many factors, e.g. the size of the population, the extent of industrial wastewater discharges, groundwater infiltration into sewer lines and inflow of storm waters.

Table 1. Typical wastewater constituents and their concentrations in municipal wastewaters (Tchobanoglous and Schroeder, 1985; Metcalf and Eddy, 1991)

Contaminant	Typical concentrations in municipal wastewater
Biodegradable organics - Biochemical oxygen demand (BOD_7) - Chemical oxygen demand (COD_{Cr})	 220 mg/l (100-400 mg/l) 500 mg/l (250-1000 mg/l)
Suspended solids	220 mg/l (100-350 mg/l)
Nutrients - Nitrogen - Phosphorus	 40 mg/l (20-85 mg/l) 8 mg/l (4-15 mg/l)
Refractory organics	variable compounds and concentrations
Dissolved inorganic solids	variable concentrations
Heavy metals	variable concentrations
Enteric microorganisms	variable species and numbers

The biodegradable organic matter in municipal wastewaters is composed mainly of carbohydrates, proteins, fats and oils, and these lead to consumption of oxygen resources when they are being degraded in natural waters. The main nutrients of municipal wastewaters include phosphorus and nitrogen, which, along with carbon compounds, are essential nutrients for growth and cause eutrophication of natural waters. Nitrogen loads may cause direct oxygen consumption in natural waters when ammonium-nitrogen is oxidized into nitrate-form in nitrification. Wastewaters also typically contain heavy metals and other inorganic ions, such as sodium and sulphate, as well as some refractory organic compounds (e.g. phenols, pesticides and surfactants).

Municipal wastewaters always contain different enteric microorganisms, including bacteria, viruses and protozoa, a part of which are pathogenic (disease causing) for humans and/or animals (Yaziz and Lloyd, 1979; Zutter and Hoof, 1984; Kayser et al., 1987; Emparanza-Knörr and Torrella, 1995; Scott et al., 2002). Wastewaters may also contain antibiotic resistant enteric microbes (Mach and Grimes, 1982; Alcaide and Garay, 1984; Iwane et al., 2001). Some of the pathogenic microorganisms that are commonly present in municipal wastewaters are listed in Table 2. The presence and numbers of these microbes in wastewaters are variable and mainly depend on the prevalence of the organisms in the population connected to the sewage network and the ability of the microbes to survive in wastewaters. Wastewaters also contain high numbers of faecal bacteria and viruses that are non-pathogenic. Some of these bacteria and viruses, or microbial groups, are used as indicators of faecal contamination, e.g. for the assessment of hygienic quality of different waters (Scott et al., 2002).

Table 2. Pathogenic microorganisms that are commonly present in municipal wastewaters (Jones and Watkins 1985; EPA 1999a; Scott et al., 2002).

Bacteria	Viruses	Intestinal parasites
Salmonella sp.	Enteroviruses (e.g. poliovirus, echovirus and coxsackie viruses)	*Giardia lamblia*
Campylobacter sp.	Hepatitis type A	*Cryptosporidium parvum*
Mycobacterium tuberculosis	Norwalk virus	*Entamoeba histolytica*
Listeria monocytogenes	Rotavirus	*Ascaris lumbricoides*
Yersinia enterocolitica	Reovirus	
Enteropathogenic *E. coli*	Adenovirus	
Staphylococcus aureus	Parvovirus	
Clostridium sp.		
Shigella sp.		
Vibrio cholerae		
Brucella sp.		

2.2 Conventional wastewater treatment

Wastewater treatment typically consists of various unit operations and processes that are selected on the basis of the raw wastewater characteristics, wastewater flow rate, goal of treatment etc. The unit operations and processes are typically grouped together into several stages with different levels of treatment, called primary, secondary and tertiary (or advanced) wastewater treatment.

Conventional wastewater treatment typically consists of primary and secondary unit operations and processes. Primary treatment consists mainly of physical unit operations, including screening, grit removal and primary sedimentation, which are used to remove solid wastewater constituents and part of the suspended solids from the wastewater. The removal of suspended solids also decreases the numbers of enteric microorganisms as many of the microorganisms are attached to solid particles in wastewater (Tanji et al., 2002). Primary treatment efficiency can be improved by addition of coagulant chemicals (Ødegaard, 2001).

Secondary treatment contains biological unit processes, for instance activated sludge process, which are principally used to reduce organic matter and nutrients in the wastewater. Biological wastewater treatment is based on the action of an active biomass that degrades the organic matter present in the wastewater. The process is affected by a number of factors, including the composition of wastewater (quality of organic matter, toxic compounds), temperature, pH and oxygen concentration and retention time of the treatment process. Biological treatment processes include aerobic and anaerobic processes, as well as suspended growth and biofilm processes.

Aerobic biological wastewater treatment alone does not efficiently remove nutrients from the wastewater, but around 20-30 % total phosphorus and total nitrogen reductions are typically achieved (Nieuwstad et al., 1988). The traditional method to improve phosphorus removal is to add a precipitation chemical into the activated sludge process, the process being called simultaneous precipitation. The most common precipitation chemicals include ferrous and ferric salts (e.g. ferrous sulphate, ferric chloride), as well as aluminium salts (e.g. aluminium sulphate). Biological phosphorus removal can be enhanced by rearranging the biological treatment stage (addition of anaerobic and anoxic stages) to favour the growth of specific phosphorus accumulating microorganisms. Nitrogen removal from wastewaters is typically achieved by a biological nitrification-denitrification process. This process transforms the typical nitrogen compounds in wastewater, organic-N and ammonium-N, through the nitrite-

N and nitrate-N into gaseous nitrogen compounds that escape from the wastewater into the atmosphere.

Conventional biological-chemical wastewater treatment by primary and secondary treatment processes can eliminate 90-99 % of the organic load and phosphorus and 90-99.9 % of the enteric microorganisms present in raw wastewater (Yaziz and Lloyd, 1979, 1982; Nieuwstad et al., 1988; Suwa and Suzuki, 2003). The mechanism of microbial removal in wastewater treatment processes is a combination of biological and physical processes. The extent of microbial reductions is affected by the treatment process used, pH, temperature and natural die-off of the microbes, as well as retention time, oxygen concentration, predation and antagonistic forces of the biological flora in the biological treatment stage (Popp, 1973; Drift et al., 1977; Yaziz and Lloyd, 1979, 1982; Schüsseler et al., 1986; Kayser et al., 1987; Morozzi et al. 1988; Scott et al., 2002; Tawfik et al., 2004). Microorganisms mainly are attached to sludge particles or a biofilm in the biological treatment stage, and thus the efficiency of separating suspended solids from the effluent water is of major importance (Drift et al., 1977; Yaziz and Lloyd, 1979, 1982; Teitge et al., 1986; Tanji et al., 2002; Nakajima et al., 2003; Tawfik et al., 2004). In some cases, pathogenic microorganisms may be able to survive for a prolonged period of time or even grow in wastewaters (Popp 1973; Kampelmacher et al. 1976; Zutter and Hoof, 1984; Kayser et al., 1987; Emparanza-Knörr and Torrella, 1995).

Secondary treated wastewaters still contain residual phosphorus and organic matter causing increased eutrophication and elevated oxygen demand on the natural waters. Secondary effluents also contain high numbers of enteric microorganisms, including pathogenic species and antibiotic resistant microorganisms (Kampelmacher et al. 1976; Yaziz and Lloyd 1979; Langeland 1982; Sobotta et al. 1986; Iwane et al., 2001; Scott et al., 2002). The sewage discharges increase pathogen contamination of surface waters and increase the risks of waterborne infections, if the polluted surface waters are used as raw water for drinking water production, for recreational purposes, for seafood farming or for agricultural uses (Popp, 1973; Kampelmacher et al., 1976; Suwa and Suzuki, 2003). Viruses are of particular concern, since even low levels in the environment can pose a risk to human health as many viruses have a very low infective dose (<10 virus particles). Enteric microorganisms can survive well in natural waters and they can be transported long distances downstream from the wastewater discharge area (Kampelmacher et al., 1976; Merch-Sundermann and Wundt 1987; Rajala and Heinonen-Tanski 1998). The survival of enteric microbes in natural waters is affected by several factors, including temperature, solar radiation, oxygen concentration, pH, nutrient concentrations, predation by protozoa, etc. (Roszak and Colwell 1987).

In many cities, the sewer systems are combined or partly combined, causing large variations in the hydraulic load on the wastewater treatment plants. During storm water conditions, the hydraulic load of the WWTP may exceed the treatment capacity, and some primary treated or untreated wastewater may be discharged into the receiving waters without any secondary and/or tertiary treatment. In these events, as well as during the disturbance of treatment processes, the loads of enteric microorganisms and other pollutants may significantly increase (Hanner et al., 2004; Rechenburg et al. 2006). In order to avoid those low quality wastewater discharges and to improve the treatment efficiency, several solutions are possible: A separation of the combined sewer system decreases the load of the WWTP and thus prevents the overflow discharges. However, this requires large investment and such activities require much time to be implemented. Building of storage tanks near the WWTP for handling peak flows or the expanding of the WWTP to increase the capacity are also possible solutions, but they may incur major expenses and require large areas. The peak flow can also be treated separately in a high rate chemical treatment process. Hanner et al. (2004) reported around 90 % reductions of total phosphorus and suspended solids when treating screened raw wastewater by direct precipitation and settling (S_h of 3.75 m/h) in the existing primary settling tank in municipal WWTP. BOD reductions of 50-60 %, 70-90 % reductions of SS and over 85 % reductions of total phosphorus were achieved by using alternative treatment method, containing chemical/mechanical treatment with precipitation and lamella separation.

2.3 Tertiary wastewater treatment

After conventional wastewater treatment, the quality of the wastewater effluent can further be improved by tertiary (or advanced) treatment processes. Tertiary treatment processes are typically used to remove organic matter, suspended solids, synthetic organic compounds, enteric microorganisms and inorganic ions, such as sulphate and phosphate, from the secondary effluents.

There are a number of different processes, including post-precipitation, rapid sand filtration (RSF), slow sand filtration (SSF), dissolved air flotation (DAF), microfiltration, ultrafiltration, ion exchange, reverse osmosis, chemical oxidation and carbon adsorption, which have been used for tertiary treatment of wastewaters in different applications (Nieuwstad et al. 1988; Metcalf and Eddy, 1991; Jolis et al., 1996; Al-Mogrin 1999; Ødegaard, 2001; Pinto Filho and Brandão, 2001; Hamoda et al., 2002; Rajala et al. 2003). The applicability and choice of treatment process is affected by several factors, e.g. the goal of treatment, wastewater quality and flow rate, the compatibility of the various operations and processes, the ease of operating the process, the space requirements and the environmental and economical feasibility of the system.

2.3.1 Filtration

Granular medium filtration is currently a widely applied unit operation in different water and wastewater treatment applications. Although granular media filtration has been practiced in water treatment for a long time, the filtration of wastewater effluents is a relatively recent practice. Nowadays filtration is used extensively for tertiary treatment of wastewater effluents. Filtration processes are principally used to remove suspended solids and organic matter prior to a final disinfection treatment or to remove phosphorus and organic matter from wastewater effluents prior to their discharge into natural waters.

In terms of operation, filters can be classified as either semicontinuous or continuous. Within each classification, there are a number of types e.g. depending on the filter-bed depth, types of filter medium, stratification of filter media, direction of flow and flow rate (Metcalf and Eddy, 1991; Jolis et al., 1996; Kuo et al., 1997; Cikurel et al., 1999; Hijnen et al., 2004; Lau et al., 2004). Different filter media (e.g. sand, anthracite, activated carbon) with different separation characteristics have been developed and used for filtration of water and wastewater. The principal types of filter bed configurations include mono-medium, dual-medium and multi-medium filter beds. These systems vary in the number of different filtering media layers in the filter unit. Dual- and multimedium filters, as well as deep bed mono-medium filters have been developed to improve the solids-storage capacity of filters and to achieve longer filter runs between the backwashing phases. The filters also differ in the direction of wastewater flow in the filter (downflow and upflow filters). The filters can further be classified by the driving force (i.e. gravitation or pressure).

The filtration process typically involves coagulation and a rapid rate filtration, either in conventional plants preceded by flocculation and clarification (by sedimentation or flotation) processes, or in direct filtration plants, in which the clarification phase is omitted (Nieuwstad et al., 1988; Hall et al., 1995; Offringa, 1995; Kuo et al., 1997; Logsdon, 2000). In the rapid sand filtration (RSF) process, a coagulant chemical is typically dosed before the sand filter, and the flocculation and separation of solids will take place inside the filter-bed (Kuo et al., 1997; Rajala et al., 2003). RSF processes have also been operated successfully as mechanical filters, without coagulant dosing (Hamoda et al., 2002).

2.3.1.1 Description of the filtration process

The filtration process is composed of two phases: filtration and cleaning of the filter medium (called as backwashing). The mechanisms of filtration phase are essentially the same for all the various types of filters used for wastewater filtration, but there are differences in the cleaning operations. In semicontinuous filtration, water is filtered through the filter bed until the filter headloss becomes excessive or the effluent quality starts to deteriorate due to detachment of particulate matter from the filter medium. At that point, the filtration is ceased and the filter bed is backwashed by air scour and/or water wash to remove the accumulated solids. In continuous filtration, the filtering and cleaning phases occur simultaneously. After passing through the filter media, filtered water is collected and removed from the process.

The different mechanisms that contribute to the removal of materials within a filter bed include straining, sedimentation, impaction, interception, adhesion and adsorption (Metcalf and Eddy, 1991; Stevik et al., 1999, 2004). Straining is the principal mechanism affecting removal of suspended particles during the filtration process. It is based on the mechanical straining of particles larger than the pore size of the filtering medium. Straining is mainly controlled by grain size and the amount of filter clogging (e.g. due to the accumulation of suspended solids or growth of microflora in the filter medium), as well as by size of the particles to be separated. The mechanism of sedimentation is based on the settling of particles on the filtering medium within the filter bed. Removal of particulate matter by impaction mechanism occurs when heavy particles will not follow the flow streamlines and impact on the filter grains. The adhesion mechanism is based on the attachment of particles to the surface of the filtering medium as the water passes through the filter. Once the particle has been brought in contact with the surface of the filtering medium or with other particles on the filtering medium, chemical adsorption or physical adsorption may take place and hold the particles. The extent of adsorption is influenced by several physical and chemical factors including the grain size and the surface characteristics of the filter medium, water flow velocity, ionic strength and species in the water, pH and surface characteristics of the adsorbed material (Stevik et al., 1999, 2004). Soluble compounds may also be adsorbed on to the surface of the filter medium. Some material can be sheared away through the action of fluid shear forces.

2.3.1.2 Factors affecting the filtration process efficiency

The principal process variables in filter design include filtration rate, influent particle characteristics, filter medium characteristics, allowable headloss, filter-bed porosity, filter-bed depth, pre-treatment means and filter backwash control (Metcalf and Eddy, 1991; Kuo et al., 1997).

The grain size, specific surface area and surface characteristics (such as surface charge) of the filter medium can affect the removal efficiency of particles and microorganisms from the water, with a smaller grain size typically improving the efficiency of removal (Stevik et al., 1999, 2004; Logan et al., 2001; Manios et al., 2002). The positive effect of the increased specific surface area may be explained by the increased availability of adsorption sites, resulting in improved removal efficiency. The increase of grain size of the filter medium typically increases the penetration of small particles through the filter bed. Decreasing the grain size may increase the headloss development and lead to clogging problems during the filter run.

The filtration rate is an important process variable, because it determines the required filter surface area. The increase in the filtration rate may reduce the removal efficiency of particles from water and shorten the filter run lengths (Kuo et al., 1997; Adin, 1999; Cikurel et al., 1999; Stevik et al., 1999, 2004; Graaf et al., 2001). With some filter types, the difference in particle removal over typical filtration rates of 5-14 m/h may become insignificant, while some other filter types are more affected by an increase in the hydraulic loading (Kuo et al., 1997).

The influent characteristics affecting the filtration process include the concentration, particle size, size distribution, charge and strength of solid particles or flocs (Jolis et al., 1996; Kuo et al., 1997; Adin, 1999; Stevik et al., 2004). The size distribution and surface characteristics, such as surface charge, of particulate matter influence their removal mechanisms in the filter bed. Colloidal and particulate matter in wastewater typically has a negative charge, thus repelling each other and becoming stabilized. With regard to wastewater effluent suspensions, the particle size typically ranges from several nanometers up to more than 100 micrometers. Particles of 1-2 µm size and smaller have a minimal opportunity for removal in the filter unit, since the transport mechanisms of these particles within the filter bed are less efficient. Particles smaller than about 1 µm are transported by diffusion, whereas larger particles are transported by gravity. The transport of larger particles may also be dominated by interception, or they may be retained by straining. A high concentration of suspended solids and turbidity in the influent of filter unit may decrease the process efficiency and cause clogging problems, increasing the need for filter backwashing (Nieuwstad et al., 1988; Jolis et al., 1996; Kuo et al., 1997; Hamoda et al., 2002). Chemical factors, such as pH and ionic composition and strength of the wastewater, typically have a smaller influence on the filtration efficiency (Stevik et al., 1999, 2004).

Inorganic coagulant chemicals and organic polymers are commonly used in the filtration process to improve the efficiency of treatment (Diamadopoulos and Vlachos, 1996; Cikurel et al., 1996, 1999; Heinonen-Tanski et al., 2002; Rajala et al., 2003). Chemical addition may cause destabilization and coagulation of particulate and colloidal matter (via a decrease in the electrostatic repulsion between the particles), followed by particle aggregation and separation of particulate matter in the filter bed (Adin, 1999). Addition of an inorganic coagulant chemical may also precipitate soluble substances, such as inorganic phosphorus. The organic polymeric flocculants may work through bridging suspended materials into larger aggregates and increasing the strength of particle aggregates and chemical flocs. The efficiency of coagulation-flocculation process is affected by the coagulant dose and the pH of the water, each coagulant type having its own optimum process conditions. Overdosing of coagulant may cause operational problems, such as filter clogging or breakthrough of turbidity through the filter medium (Jolis et al., 1996; Kuo et al., 1997; Cikurel et al., 1999).

2.3.1.3 Reductions of pollutants in the filtration process

Rajala et al. (2003) reported that tertiary rapid sand filtration (RSF) as a contact filtration reduced the numbers of enteric microorganisms by 90-99 %, suspended solids by 56-93 % (residual 1-4 mg/l), turbidity by 65-87 % (residual 1-2 NTU), total phosphorus by 75-89 % (residual 0.04-0.1 mg/l) and COD by 34-53 % (residual 18-39 mg/l), and achieved >70 % UV transmittance values in pilot-scale experiments. The tertiary RSF process when used as a mechanical filtration was reported to achieve lower reductions.

Kuo et al. (1997) reported 78-80 % average reductions of turbidity (residual 1.2-1.4 NTU), 81-87 % reductions of SS (residual 1.6-2.0 mg/l) and 17 % reductions of COD (residual 53-54 mg/l) in three different pilot-scale tertiary filtration processes treating domestic secondary effluent. Jolis et al. (1996) have reported comparable results in pilot-scale tertiary filtration of municipal secondary effluents, by using two different filters. They also reported 1.1 log average reductions of coliform bacteria and 0.6 log reductions of MS2 coliphage in the tertiary filtration processes. Nieuwstad et al. (1988) achieved about 30-50 % reductions of organic matter (BOD, COD), 70-75 % reductions of P_{tot} and SS, as well as 20-70 % reductions of enteric microbes (*E. coli*, faecal streptococci, F-specific phages and spores of sulphite reducing clostridia) in the tertiary mechanical filtration process. Tertiary filtration with iron coagulant chemical addition before the filter unit (direct filtration) improved the filtration process performance, achieving about 40-60 % reductions of organic matter (BOD, COD), 90 % reductions of P_{tot} and 80-99.4 % reductions of enteric microbes.

Suwa and Suzuki (2003) achieved about 0.5 log reductions of *Cryptosporidium* oocysts in a tertiary mechanical filtration process, while a tertiary direct filtration process with polyaluminium chloride coagulant addition achieved 2.6 log reductions of *Cryptosporidium* oocysts. Scott et al. (2002) reported around 1 log reductions of viruses and 1-1.5 log reductions of *Giardia* spp. and *Cryptosporidium* spp. in tertiary filtration units (shallow bed anthracite filter + polymer addition; deep bed sand/anthracite filter, no coagulant addition).

2.3.2 Flotation

Flotation is a unit operation that is used to separate solids or liquid particles (e.g. oil suspension) from the liquid phase. Separation of solids is achieved by introducing fine air bubbles into the water to be treated. The bubbles become attached to the solids, and the buoyant force of the particle-bubble aggregate causes the particle to rise to the surface of water. The separated solids floating on the surface of water can then be collected mechanically or hydraulically.

There are a number of different flotation technologies. Flotation is typically described in terms of the method of bubble formation, e.g. dissolved air flotation (DAF), dispersed air flotation and electroflotation (Edzwald, 1995; Rubio et al., 2002). Sometimes, flotation is described in terms of material being removed or separated, e.g. mineral flotation, precipitate flotation and colloid flotation. Processes combining flotation and filtration processes into one unit, flotation filters, have also been developed for the treatment of water and wastewater (Arnold et al., 1995; Eades and Brignall, 1995; Krofta et al., 1995a, 1996; Kiuru, 2001).

Flotation technology has its origin in the mineral or ore processing industry. It has been used since the early 1900’s to separate different mineral ores from each other (Edzwald, 1995; Rubio et al., 2002). Flotation technology was first introduced into water treatment in the

1920's, but its use in different water and wastewater treatment applications significantly increased in the 1960's and 1970's. Nowadays, flotation processes are widely used for the treatment of water and wastewater, for sludge thickening and in different industrial applications (wastewater and process water treatment, recovery of valuable materials, etc.) e.g. in the mining, metal, wood processing, textile and food industries (Arnold et al., 1995; Edzwald, 1995; Heinänen et al., 1995; Offringa, 1995; Viitasaari et al., 1995; Schofield, 2001; Rubio et al., 2002). Flotation processes have been used for many applications, e.g. for the separation of suspended solids, colloids, oils, ions, macromolecules, pigments, fibers, minerals and algae from different waters (Ferguson et al., 1995; Rubio et al., 2002; Buisine and Oemcke, 2003).

The most common flotation technology in water and wastewater treatment applications is dissolved air flotation (DAF), which has been used for water and wastewater clarification since the 1960's (Edzwald, 1995; Offringa, 1995; Pinto Filho and Brandão, 2001). In the following sections, the emphasis in this review will be on pressurized dissolved air flotation.

The principal advantage of flotation when compared to the more traditional clarification process, sedimentation, is that even very small and light particles (such as algae and chemical flocs) with poor settling ability can be separated more efficiently and with much higher overflow rates (typically 5-15 m/h), also in cold waters (1-4ºC). Flotation process may require only around 10 % of the surface area and around 5 % of the volume of a comparable sedimentation process. The advantages of flotation process also include lower chemical consumption, rapid start-up and ability to withstand periodic stoppages to the process, its relative robustness to hydraulic and quality variations in the water to be treated and the positive control over separation process (Schofield, 2001). One other advantage of flotation in wastewater treatment is that the effluent from flotation is aerobic, and the process may thus have a beneficial effect on the oxygen balance of the recipient water body, at least when compared with sedimentation process. Flotation process is, however, a more complex and mechanically intensive process, which requires electrical power and includes a large number of process control variables, when compared to a sedimentation-based technique. Flotation typically has lower capital costs and higher operating costs than the conventional clarifiers. Based on the practical experiences, flotation system can be a technically suitable and economically applicable clarification process (Teerikangas, 2000; Huhtamäki, 2007).

2.3.2.1 Description of the dissolved air flotation (DAF) process

The DAF facilities are typically composed of the following steps: 1) coagulation and flocculation prior to flotation, 2) bubble generation, 3) bubble-floc collision and attachment, and 4) rising and separation of bubble-floc agglomerates in a flotation tank.

The raw water to be treated typically enters the DAF unit through rapid mixing and flocculation processes, where a selected chemical coagulant is dosed and mechanical or hydraulic agitation of water is done in a stirred tank, baffled channel or in pipe and the formation of chemical flocs starts. The flocculated water then flows to the entrance part of the flotation tank, called the reaction zone.

In the DAF process, a portion of clarified water is recycled into the pressure vessel (saturator) by recycle pumps and is pressurized by air in a pressure of 400 to 600 kPa (Edzwald, 1995; Amato et al., 2001; Schofield, 2001). The pressurized recycling water, called dispersion water, is then recycled through specially designed injection nozzles or needle valves into the reaction zone of the DAF tank, where it is mixed with the unpressurized wastewater stream. The decrease of the recycle flow pressure to atmospheric pressure leads to the formation of microbubbles (typical diameter 10-100 µm, average 40 µm), the bubble size depending on the saturator pressure, injection flow rate and nozzle design (Rykaart and Haarhoff, 1995). The injection flow must provide a rapid pressure drop and be sufficient to prevent backflow and bubble growth on pipe surfaces in the vicinity of the injection system. The process of bubble formation is assumed to include two steps: nucleation and growth (Edzwald, 1995; Rykaart and Haarhoff, 1995). In a supersaturated system of clear water, the large pressure difference across the nozzle produces bubble nuclei spontaneously (homogeneous nucleation), while in heterogeneous system, bubble formation occurs on a particle nuclei or other surfaces. Then the nuclei growth into bubbles occurs and the bubble size increases due to coalescence.

The released microbubbles collide and attach to flocculated particles and these form bubble-solid aggregates. The bubble-particle interactions take place mainly in the reaction zone by turbulent transportation, and also in the final stage of reaction zone and in the clarification zone of the flotation tank by an interception mechanism (Fukushi et al., 1995). The formation of bubble-particle aggregates reduces the density of particles, causing them to rise to the surface of the DAF tank in the clarification zone of the flotation tank. The best contact and attachment is typically achieved with microbubbles (diameter <100 µm). Three different mechanisms for bubble-particle interactions have been described (Edzwald, 1995; Rubio et al., 2002): In the DAF process, part of the dissolved air in water, which does not convert into bubbles in the nozzles, remains in solution and can “nucleate” at the particle surface, followed by bubble growth. This mechanism occurs to varying degrees in most DAF applications. Another bubble-particle interaction mechanism includes bubble entrapment into flocs or aggregates; this mechanism is important in applications where large particles or flocs exist (e.g. thickening, treatment of wastewaters). The third mechanism includes particle collision and adhesion with preformed bubbles, which is believed to be the most important mechanism. The most important parameters that affect collision efficiency are zeta potential and sizes of particles and bubbles (Han et al., 2001). Coagulant chemicals and/or polymeric compounds are commonly used to aid the process by creating a particle surface or a structure that can easily adsorb or entrap air bubbles, as described below.

The concentration of supplied air bubbles is the key design and operating parameter affecting DAF process performance, as it affects particle-bubble collisions, particle separation and removal. The three fundamental parameters of the supply air include the mass concentration, the air bubble volume concentration and the bubble number concentration (Edzwald, 1995). The recycle ratio is used as a surrogate measure of the supplied air, and is defined as:

$$R=Q_r/Q_0 \qquad (1)$$

where R is the recycle ratio, Q_r is the recycle flow and Q_0 is the unpressurized influent flow. The amount of air can be increased by increasing the recycle ratio, the saturator efficiency or the saturator pressure. Saturator efficiencies may vary from 60 to 70 % in unpacked saturators, rising to around 90 % in packed saturators (Edzwald, 1995; Amato et al., 2001).

The separated sludge can be removed from the surface of the DAF tank mechanically by a sludge skimmer or hydraulically. The dry matter content of the mechanically removed sludge is typically 3-8 % and is affected mainly by the raw water quality and the type and concentration of coagulant or polymer (Arnold et al., 1995; Amato et al., 2001; Mels et al., 2001). When hydraulic sludge removal is used, the dry solids content of the sludge may be around 0.5 %.

The clarified water is removed from the bottom of the flotation tank. Equipments with different designs have been developed for clarified water removal, including single submerged end-wall outlets, multi-distributed outlets and perforated pipes. The outlet design has an effect on DAF tank hydraulics and process performance, as described below.

The development of DAF technology has been very rapid in the last decades and many of its earlier limitations have been resolved. The first generation DAF was developed in the beginning of 1900's (Kiuru, 2001). It was characterized by a rather long, narrow and shallow DAF tank and was designed for hydraulic loading rates of 2-3 m/h. The second generation DAF was developed in the 1960's. These so-called conventional DAF processes have a shorter, broader and deeper DAF tank. The process is typically designed for S_h of 5-7 m/h, and they can be operated even at 10-15 m/h hydraulic loading rates. Flotation filters, combining dissolved air flotation and rapid sand filtration in the same tank, were developed in the 1960's. The process brought significant improvements to the hydraulics of the DAF process. The third generation DAF was developed in 1990's, with the target being to further increase the flow rate in the DAF units. The third generation DAF unit replaced the sand filter of the flotation filter with another mechanical structure (such as a thin, stiff, horizontal plate with round orifices) capable of controlling hydraulic behaviour of the DAF tank. The third generation DAF unit is characterized by deep flotation tank, which can be operated at turbulent flow conditions, even with S_h of 25-40 m/h (Amato et al., 2001; Kiuru, 2001).

2.3.2.2 Factors affecting the DAF process efficiency

The DAF processes include liquid, solid and gaseous phases, where physical, chemical and electrical forces affect at both the micro and macro levels. A number of variables, including water quality, temperature, process hydraulic loading rate and volume of air to mass of solids (A/S) –ratio, as well as addition of coagulant or polymer, their dosage and flocculation conditions affect the DAF process efficiency (Krofta et al., 1995b; Ødegaard 1995; Haarhoff and Edzwald, 2001; Reali et al., 2001a, 2001b; Pinto Filho and Brandão, 2001; Jokela and Immonen, 2002). The interactions between water, solid particles and air bubbles within the DAF tank are complex phenomena and are not fully understood (Haarhoff and Edzwald, 2001). For this reason, the technological development of the DAF process is largely the result of empirical observations and experimentation during the last decades. Recently, however, mathematical analysis and modelling of different elements in the DAF process have improved our fundamental understandings of the technique (Edzwald, 1995; Fukushi et al., 1995; Krofta et al., 1995b; Haarhoff and Edzwald, 2001; Han et al., 2001). To optimise the DAF process performance under different process conditions, it is necessary to understand the interactive role of the parameters that influence the process.

Particle destabilization via charge neutralization and the production of hydrophobic particles (hydrophobic solids or solids with hydrophobic spots) are necessary to achieve favourable flotation (Edzwald, 1995; Han et al., 2001). Particulate matter, as well as microbial cells, in municipal wastewater have a typical size range of 0.01 to 100 μm and generally have a negative surface charge (negative zeta-potential) and hydrophilic surface properties, all of these features not being favourable for flotation (Bustamante et al., 2001; Mels et al., 2001; Ødegaard, 2001; Dockko and Han, 2004). The air bubbles also have negative charge (Fukushi et al., 1995; Han et al., 2001). The efficient flotation is favoured by particle size bigger than the bubble size. The bubbles also attach only to hydrophobic (or positively charged) surfaces. For those reasons, coagulation-flocculation prior to a flotation unit is typically needed to enlarge the particles to be removed and to convert their surface properties to being more hydrophobic (Bunker et al. 1995; Cáceres and Contreras, 1995; Fukushi et al., 1995; Klute et al. 1995; Edzwald, 1995; Han et al., 2001; Mels et al., 2001).

Inorganic coagulant chemicals and various organic polymers are typically used in the DAF process to improve the efficiency of the process. These chemicals are used to generate floc, to bind the particulate and colloidal matter together and to create a surface or a structure that can adsorb or entrap air bubbles. Inorganic coagulant chemicals also precipitate dissolved phosphorus and organic matter, while such an effect cannot be achieved by using organic polymers (Mels et al., 2001; Reali et al., 2001a, 2001b). Hydrolyzing coagulants, such as aluminium and iron salts, produce a range of hydrolysis products in water. Some of those hydrolysis products are positively charged and can effectively neutralize the charge of negatively charged colloids or particles and hence promote coagulation. At around neutral pH, they form amorphous hydroxide precipitates, which can enmesh particles in water (sweep flocculation) and result in better clarification (Gregory and Dupont, 2001). Recently, the use of pre-hydrolyzed coagulants, such as polymeric hydrolysis products of aluminium, has increased. Polyaluminium chloride (PACl) products contain highly charged cationic species (such as $Al_{13}O_4(OH)_{24}^{+7}$) that strongly adsorb onto negative colloids or particles causing charge neutralization. PACl products also precipitate amorphous aluminium hydroxide. There are several benefits associated with PACl products over hydrolysing coagulant, such as aluminium sulphate (“alum”). These include improved performance at low temperatures, lower aluminium residual concentrations in treated water and lower sludge volumes produced in the process (Gregory and Dupont, 2001). In addition, the flocs produced by PACl products are generally larger, stronger and more easily separated than those produced with alum. Inorganic aluminium salts and PACl coagulants have been reported to remove viruses from water, causing also some inactivation of viruses (Matsushita et al., 2004).

Typically the increase of coagulant dose improves the flocculation-DAF performance, but overdosing of coagulant may lead to charge restabilization of the particles (positively charged particles and bubbles) and poor flocculation or production of too large and too heavy flocs, resulting in less efficient clarification process (Edzwald, 1995; Ødegaard, 1995; Pinto Filho and Brandão, 2001). The flocculation-DAF process performance has been reported to increase with increasing flocculation time, probably due to more chances for particle collisions and floc formation (Ødegaard, 1995; Pinto Filho and Brandão, 2001). Coagulation-flocculation should produce robust and dense flocs that resist fracture in the turbulent conditions of flotation tank, with a floc size of <100 μm being favourable (Ødegaard 1995; Edzwald, 1995; Penetra et al., 1999; Reali et al., 2001b). There is, however, some disagreement on the preferable floc size for successful clarification in the DAF process. Fukushi et al. (1995) assessed that a larger floc size (up to 10^3 μm), also typically present in the water treatment plants, may be favourable in the DAF process. Ljunggren et al. (2004) reported increased particle separation efficiency with increasing particle size (>100 μm), when tertiary DAF process was used for separating biological flocs from the biofilm process effluent. A larger particle size was reported to increase the number of bubbles attached to a particle, increasing the rising rate and the separation efficiency of the particle-bubble aggregates. The flocculation G-values (mean velocity gradient) of 20-100 s^{-1} and retention time of 5-25 min have been reported to be favourable in the DAF process (Bunker et al., 1995; Ødegaard, 1995). There is, however, a wide variation in the applied flocculation intensities and flocculation times in different practical DAF applications (Edzwald et al., 1995; Haarhoff and van Vuuren, 1995;

Amato et al., 2001). The optimum coagulation-flocculation conditions are process specific and the number, size, density, structure and strength of the flocs must be optimised case-specifically.

Proper design of the air dispersion system is an important factor for successful DAF clarification, since the concentration and the size of the air bubbles clearly influence the performance of the DAF process. The bubble size is of importance, with a bubble size of <100 μm being typically favourable for efficient clarification. The optimum bubble size and number in the DAF process may vary, however, according to the size and number of the particles or flocs to be removed (Haarhoff and Ezdwald, 2001; Han et al., 2001; Dockko and Han, 2004). With respect to the removal of small flocs, the air efficiency, the bubble size and the air volume are critical, while larger flocs are less sensitive to these parameters. Large air bubbles result in high rise rates, which may cause poor clarification due to floc breakup and interference of the rapidly rising macrobubbles with the slowly rising particle-bubble aggregates (Rykaart and Haarhoff, 1995). Large air bubbles also have less surface area per unit volume, which decreases the number of bubbles and the chance for bubble-particle collisions, thus impairing clarification efficiency. The bubble size is affected by the saturator pressure and injection flow rate (Edzwald, 1995; Dockko and Han, 2004). The recycle ratio is typically used as a surrogate measure of the supplied air. The optimum recycle ratio in the treatment of wastewater effluents typically varies in the range of 10-20 % (Ødegaard 1995, 2001; Pouet and Grasmick, 1995; Pinto Filho and Brandão, 2001). However, wide variations in the applied recycle rates (from 5 to over 40 %) have been reported in different DAF applications (Arnold et al., 1995; Edzwald et al., 1995; Haarhoff and van Vuuren, 1995; Amato et al., 2001).

Hydraulic loading rate (S_h) and fluid dynamics in the DAF tank have a central role in the implementation of an effective DAF process (Haarhoff and van Vuuren, 1995; Lundh et al., 2000, 2001; Kiuru, 2001). Through improvements of hydraulic characteristics, the process capacity and performance from the first generation DAF units to the third generation DAF units have been significantly improved. The inlet and outlet arrangements (such as the gross-flow velocity from the contact zone to the clarification zone) and the geometry of the DAF tank affect the hydraulic behaviour of the process. The flow patterns within the separation zone are complex, due to the high density gradients induced by the air suspension that is unevenly distributed in the DAF tank. The changes of hydraulic loading and recycle rates can affect the bubble-bed formation and flow patterns (flow directions, recirculation, short-circuiting) in the DAF tank, and thus influence process performance (Lundh et al., 2000, 2001). An evenly tight bubble bed has a clear filtration impact, increasing the rate of attachment of bubbles onto solids, as most of the water to be treated is forced to flow through the bubble-bed. One of the major factors in hydraulic control of the process is the way in which the clarified water is drawn from the flotation tank (Schofield, 2001). Single submerged end-wall outlets with low resistance and a low headloss design have been successfully replaced by higher resistance multi-distributed outlets, perforated pipes, sand in combined DAF/filters and lately by perforated plates in the third generation DAF processes, which produce more uniform down-flow conditions in the separation zone of the flotation tank (Kiuru, 2001; Schofield, 2001). The DAF process is typically rather insensitive to changes in the hydraulic loading rate (Ødegaard, 1995).

The solids loading rate is an important design parameter of the DAF process. The amount of bubbles must be increased with increasing concentration of suspended solids. The size and weight of the particles to be separated, as well as the bubble size and the number of bubbles attached to a particle all affect the bubble-particle aggregate rising speed and the process capacity (Haarhoff and Edzwald, 2001; Reali et al., 2001; Rubio et al., 2002; Ljunggren and Jönsson, 2003).

The sludge removal technique may have an effect on process performance. The separated sludge is removed from the surface of the tank mechanically or hydraulically. The float removal from the DAF unit by continuous or intermittent mechanical scraping could result in turbulent conditions at the float/water interface and may lead to floc break-up and solids re-entering the clarification zone and the clarified water.

2.3.2.3 Reduction of pollutants in the DAF process

Tertiary DAF has shown good efficiency for removal of suspended solids, organic matter and phosphorus from wastewater effluents. The tertiary DAF process has been reported to achieve around 90 % reductions of COD and around 95 % P_{tot} reductions in pilot-scale experiments and in full-scale WWTPs (Ødegaard, 2001). In laboratory-scale DAF experiments, around 80-95 % reductions of SS, COD and P_{tot} have been reported when treating different domestic sewage effluents (Penetra et al., 1999; Pinto Filho and Brandão, 2001; Reali et al., 2001a, 2001b). The DAF process has also been used for treatment of raw sewage. In the direct chemical treatment of raw sewage and DAF clarification, treatment efficiencies of 96 % on total phosphorus and 78 % on COD have been reported (Ødegaard, 2001).

There is no literature data on the elimination of enteric viruses, bacteria or protozoan parasites in the DAF treatment of municipal wastewaters. In pilot-scale drinking water treatment experiments, the DAF process has been reported to achieve 1.7 and 2.5 log (Edzwald et al., 2001) average reductions of *Cryptosporidium* during the winter (2-3 ºC) and spring (13-14 ºC) time, respectively. Combinations of DAF and filtration processes have been reported to achieve 3-4 log (Hall et al., 1995) and above 5.4 log (Edzwald et al., 2001) reductions of spiked *Cryptosporidium* oocysts in pilot-scale drinking water treatment experiments.

2.4 Disinfection of wastewater

Although secondary and tertiary wastewater treatment processes cause significant reduction in the numbers of enteric microorganisms, normally they cannot guarantee microbiologically safe effluents to be discharged into natural waters or to be reused. To achieve more efficient elimination of enteric microorganisms, some kind of disinfection treatment must be adopted.

Water disinfection methods can be divided into chemical and physical treatments. Chemical disinfectants include for instance the use of chlorine gas (Cl_2), hypochlorite (sodium hypochlorite, NaOCl; calcium hypochlorite, $Ca(OCl)_2$), chloramines, chlorine dioxide (ClO_2), ozone (O_3) and peracetic acid (PAA). Ultraviolet (UV) irradiation is the most important physical disinfection method for water and wastewater.

2.4.1 Factors affecting disinfection process efficiency

The most important factors affecting the efficiency of wastewater disinfection include the type of disinfectant, disinfectant concentration, contact time and the type and number of microorganisms in the water, as well as environmental variables, such as the quality of the water, pH and temperature (Tchobanoglous and Schroeder, 1985; Bitton, 1994; U.S.EPA.

1999c; Kuo et al., 1997; Lazarova et al., 1998; Liberti and Notarnicola, 1999; Collivignarelli et al., 2000; Stampi et al., 2001; Azzellino et al., 2002; Nasser et al. 2006).

The efficiency of disinfection depends most strongly on the characteristics of the disinfectant being used. The different disinfectants have their own distinct disinfection mechanisms and oxidizing powers. Chemical disinfectants typically disrupt the integrity of bacterial cell membranes, resulting in the loss of selective membrane permeability. Chemical disinfectants may also damage the nucleic acids and enzymes of the microbes, thus preventing cell replication and metabolic functions. Chemical disinfectants may damage nucleic acids and the protein coat of viruses (Bitton, 1994). The efficiency of disinfection is typically improved with increasing concentration of disinfectant (C) and/or the contact time (t) between disinfectant and microorganism.

The microbial resistance against disinfectants varies according to the microbial species, microbial age and the physiological status of the microorganism. Generally the resistance increases in the following order: vegetative bacteria < enteric viruses < spore forming bacteria < protozoan cysts (Tchobanoglous and Schroeder, 1985; Bitton, 1994; Kuo et al., 1997; Jacangelo et al., 2003; Nasser et al. 2006). The microbial resistance also varies between different microbial species and between different strains of the same species as well as being dependent of other physiological features, such as nutrition status, age, etc. (Nasser et al. 2006).

The disinfection process is also affected by temperature and pH (Tchobanoglous and Schroeder, 1985). The pH of the water affects the form of some chemical disinfectants in water, and in this way can affect the disinfection efficiency. Microbial inactivation usually increases with increasing temperature (Stampi et al., 2001), which may be due to temperature effects on the reaction kinetics. The metabolism of the microorganisms may also respond more slowly to disinfectants at lower temperatures.

Water quality typically has a strong effect on disinfection process efficiency. Chemical compounds, including inorganic and organic nitrogenous compounds, iron and organic matter interfere with the disinfectants. Organic compounds can react with the disinfectant, elevating disinfectant demand, and this leads to decreased elimination of microbial agents and the production of harmful DBPs. The presence of suspended solids and organic matter can protect the microbes by preventing contact of microbes with the disinfectant or UV radiation (Kuo et al., 1997; Lazarova et al., 1998; Liberti and Notarnicola, 1999; Collivignarelli et al., 2000; Stampi et al., 2001).

There are various microbial inactivation pathways and kinetics with different disinfectants, but usually the microbial inactivation can be described by analytical expressions which take into account the main process parameters, such as disinfectant concentration, contact time, microbial number and environmental variables, such as temperature and pH. Microbial inactivation in a disinfection process can be generally described by a first-order function:

$$N_t/N_0 = e^{-kt} \qquad (2)$$

where N_0 and N_t are numbers of microorganisms present at the start and end of the time t and k is the microbial decay constant. Field observations have, however, revealed deviations from that ideal behaviour, which are usually explained by the survival of resistant microorganisms during the disinfection treatment or by the protection of microorganisms for instance inside the particles or microbial aggregates present in water to be treated (Bitton, 1994; Wagner et al., 2002; Nasser et al. 2006). Various mathematical models, such as Chick-Watson's, Hom's, Selleck's and a new S-model, have been developed to better describe the inactivation of microorganisms by disinfectants (Azzellino et al., 2002).

2.4.2 Disinfection process applicability

An ideal disinfection system should efficiently and reliably destroy infectious agents under normal operational conditions. As the disinfection efficiencies of different disinfection methods differ from each other, the target level of disinfection may be the determining factor in which method is chosen. Efficient wastewater disinfection should be achieved without producing toxic, mutagenic or carcinogenic disinfection by-products (DBPs) or persistent disinfectant residues that can have adverse effects on environment or human health. The production of DBPs is related to the disinfectant characteristics and dose, as well as to the characteristics and amount of organic matter present in the water (Monarca et al., 2000). Simple and flexible operation and control of the process, ease of maintenance of devices as well as occupational and environmental safety of handling, storage and transport of disinfectant are important factors which must be taken into consideration when evaluating disinfection process applicability. The disinfection process should also be inexpensive in terms of capital, operational and maintenance costs.

2.4.3 Disinfection methods

2.4.3.1 Chlorination

Chlorination is the most common wastewater disinfection method. Chlorine disinfection has been practised from the beginning of the 1900's and it has played a major role in the prevention of waterborne infections all over the world (Bitton, 1994). Different chlorine compounds are used for disinfection, including chlorine gas (Cl_2), sodium hypochlorite (NaOCl), calcium hypochlorite ($Ca(OCl)_2$) and chloramines. NaOCl and $Ca(OCl)_2$ are more expensive disinfectants than gaseous chlorine, but their use is preferred, especially in smaller treatment plants, because of their safer use (Tchobanoglous and Schroeder, 1985; Lazarova et al., 1999).

Gaseous chlorine and hypochlorite salts hydrolyse in water to form the hypochlorous acid:

$$Cl_2 + H_2O \leftrightarrow HOCl + H^+ + Cl^- \qquad (3)$$

$$NaOCl + H_2O \leftrightarrow HOCl + Na^+ + OH^- \qquad (4)$$

Hypochlorous acid further dissociates in water to form hypochlorite ion:

$$HOCl \leftrightarrow H^+ + OCl^- \qquad (5)$$

At acidic and neutral pH, hypochlorous acid is the predominant form in the water, while at alkaline pH the hypochlorite ion dominates. Hypochlorous acid is a stronger disinfectant than hypochlorite ion and thus the chlorine disinfection is more efficient at acidic pH values (Bitton, 1994).

Chlorine in the form of HOCl and OCl^- is called free available chlorine. HOCl reacts readily with ammonia or organic nitrogen compounds in the water. These non-specific side reactions result in the formation of combined chlorine (chloramines), chloro-organics and chloride. Chloro-organics and chloride are not effective as disinfectants. Chloramines are called combined available chlorine. The species distribution of chloramines is dependent on the pH, temperature, contact time and the initial ratio of chlorine to ammonia in water (Tchobanoglous and Schroeder, 1985). Free chlorine is about 50-100 times more efficient and much more reactive disinfectant than combined chlorine. The residual free chlorine concentration is typically used as a measure of chlorination efficiency (Tchobanoglous and Schroeder, 1985; Bitton, 1994).

The production of free chlorine residues in water can be achieved by breakpoint chlorination, which requires the addition of a sufficiently high chlorine dose. Breakpoint chlorination is not usually applied in wastewater treatment, because high chlorine doses would be needed. Therefore chlorine exists in wastewaters primarily in the combined form, monochloramine being the predominant compound (Tyrrell et al., 1995).

Chlorine is a quite efficient disinfectant against many enteric bacteria, but it has much lower efficiency against viruses, bacterial spores and protozoan cysts (Tyrrell et al. 1995; Hassen et al., 2000; Scott et al., 2002; Veschetti et al. 2003). The typical chlorine dosage for the treatment of wastewater effluents ranges from 5 to 20 mg/l (Lazarova et al., 1999; Mujeriego and Asano, 1999; U.S.EPA, 1999a). Higher chlorine doses (20-60 mg/l) may be needed to achieve the inactivation of more resistant microorganisms, such as viruses (Tchobanoglous and Schroeder, 1985; Bitton, 1994). Wastewater impurities may cause significant chlorine demand, thus increasing the needed chlorine dose (Baldry et al. 1991). Typical contact times needed in chlorine disinfection range from 30 min up to 2 hours (Lazarova et al., 1999; Mujeriego and Asano, 1999).

Chlorination is a well-established technology and it has long traditions in the field of water and wastewater disinfection. Chlorination may also be used for the oxidation of certain organic and inorganic compounds and to eliminate certain noxious odours. Although the chlorination is technically relatively simple and flexible process, the use of chlorination for wastewater disinfection has been declining, mainly due to toxic, mutagenic and/or carcinogenic disinfection by-products (DBPs), as well as due to chlorine residuals and the increase in the salinity of the effluents (Oppenheimer et al. 1997; Lazarova et al., 1999; Koukouraki and Diamadopoulos, 2002; Mezzanotte et al., 2002; Veschetti et al. 2003). An increase in the chlorine dose, contact time, pH and temperature have been reported to increase the DBP formation, while a decrease in the organic carbon content of wastewater and an increase in the ammonia-nitrogen reduce the DBP formation (Koukouraki and Diamadopoulos, 2002). The chlorine residues must usually be neutralized before the wastewater effluent is discharged into natural waters, which causes an additional load of chemicals into environment, consumes dissolved oxygen in the water and increases the costs of disinfection (Tyrrell et al., 1995; Lazarova et al., 1999; U.S.EPA, 1999a). Chlorine is a highly corrosive and toxic compound, and there are safety concerns and environmental risks during the transport, handling and storage of the chemical.

2.4.3.2 Chlorine dioxide (ClO_2)

Chlorine dioxide (ClO_2) is a strong oxidizing agent and an efficient disinfectant in water and wastewater (U.S.EPA, 1999d; Collivignarelli et al., 2000; Salgot et al., 2002). Chlorine dioxide is equal or superior to chlorine in inactivating bacteria and viruses and it is also effective against protozoan cysts (Collivignarelli et al. 2000; Salgot et al. 2002).

Chlorine dioxide must be generated on-site from sodium chlorite and gaseous or aqueous chlorine, from sodium chlorite and hydrochloric acid or by irradiation of sodium chlorite by UV radiation (U.S.EPA, 1999d). The production of ClO_2 is relatively easy and economical, but the costs of chlorine dioxide disinfection are higher than those of chlorination (Collivignarelli et al., 2000).

Chlorine dioxide does not hydrolyse in water, but exists as a dissolved gas. ClO_2 does not have a residual effect. Chlorine dioxide reacts weakly with nitrogen compounds and organic material in water, reducing the formation of DBPs (Collivignarelli et al. 2000). The inorganic by-products of ClO_2 are chlorite ClO_2^- and chlorate ClO_3^-, which are potentially toxic (Collivignarelli et al., 2000). Monarca at al. (2000) found that ClO_2 disinfection of secondary treated sewage evoked some bacterial mutagenicity and toxicity but not genotoxicity.

2.4.3.3 Peracetic acid

Peracetic acid (PAA), CH_3COOOH, is an organic peroxy compound, which has strong oxidizing properties. PAA has been widely used as a disinfectant or sterilant in laboratories and in the food processing, medical and pharmaceutical industries. It has also been used as a decolouring or disinfectant agent in textile, pulp and paper industries, for disinfection of ion exchangers and cooling towers as well as for pathogen reduction in biosolids, sludge debulking and for eliminating unpleasant odors (Kitis, 2004).

PAA is available as a quaternary equilibrium mixture, containing peracetic acid, acetic acid, hydrogen peroxide and water, as well as some stabilizers:

$$CH_3COOH + H_2O_2 \leftrightarrow CH_3COOOH + H_2O \qquad (6)$$

PAA breaks down in water according to the following equations (Lefevre et al., 1992; Lubello et al., 2002):

$$CH_3COOOH \rightarrow CH_3COOH + \tfrac{1}{2} O_2 \qquad (7)$$
$$CH_3COOH \rightarrow CH_4 + CO_2 \qquad (8)$$
$$H_2O_2 \rightarrow H_2O + \tfrac{1}{2} O_2 \qquad (9)$$

PAA may be consumed by the following reactions in aqueous solution (Wagner et al., 2002):

$$2\ CH_3COOOH \rightarrow 2\ CH_3COOH + O_2 \qquad (10)$$
$$CH_3COOOH + H_2O \rightarrow CH_3COOH + H_2O_2 \qquad (11)$$
$$CH_3COOOH + \text{Metal} \rightarrow O_2 + \text{other decomposition products} \qquad (12)$$

Hydrolysis is negligible in the pH range from 5.5 to 8.2. The rate of spontaneous decomposition follows second order kinetics with respect to the PAA concentration. The biocidal active form of PAA is considered to be the undissociated acid, which is predominant at pH values below 8.2 (Wagner et al., 2002). Above pH 8.2, the dissociation of PAA increases and the efficiency of PAA disinfection declines.

The proposed disinfection mechanisms of PAA include the release of active oxygen (Lefevre et al. 1992; Liberti and Notarnicola 1999) and the production of reactive hydroxyl radicals (Lubello et al. 2002) that inactivate microorganisms. PAA disinfection does not produce significant amounts of toxic or mutagenic DBPs or chemical residues into effluents (Booth and Lester 1995; Liberti and Notarnicola 1999; Collivignarelli et al. 2000; Monarca et al. 2000; Liberti at al., 2002; Veschetti et al. 2003; Kitis 2004; Crebelli et al., 2005). However, all the chemical transformations undertaken by PAA on the wastewater matrix do not appear to be fully known (Stampi *et al.* 2001; Veschetti *et al.* 2003).

Some recent studies have shown that peracetic acid treatment is an attractive method for wastewater disinfection. PAA has shown good disinfection efficiency against many enteric bacteria in different wastewaters, but it has lower efficiency against viruses, bacterial spores and protozoan cysts (Lefevre et al. 1992; Baldry et al. 1995; Liberti and Notarnicola 1999; Stampi et al. 2001, 2002; Salgot et al. 2002; Wagner et al. 2002; Heinonen-Tanski and Savolainen, 2003). PAA treatment is quite simple to install and operate.

The potential disadvantages of PAA treatment include the increasing of BOD/TOC content of water and the relatively high costs (Lazarova et al. 1998; Liberti and Notarnicola 1999; Collivignarelli et al. 2000; Wagner et al. 2002; Kitis 2004). Regrowth or recovery of the microbial population which has undergone PAA treatment may occur under certain conditions (Sánchez-Ruiz et al., 1995; Lazarova et al., 1998; Lefevre et al., 1992). Antonelli et al. (2006) and Mezzanotte at al. (2002) have not observed any significant bacterial regrowth after PAA disinfection treatments.

2.4.3.4 Ultraviolet (UV) irradiation

UV irradiation is the most important physical disinfection procedure and it is practised in many wastewater treatment plants (Kuo et al., 1997; Lazarova et al., 1999; Sakamoto et al., 2001). The mechanism of UV disinfection is based on the UV absorption of nucleic acids of microorganisms, causing thymine dimerization. This blocks nucleic acid replication, prevents the multiplication of the organism and results in death of the cell. The maximum absorption of UV radiation and thus the optimum wavelength for microbial inactivation, is in the spectral region of 250-260 nm (White et al., 1986; U.S.EPA, 1999b). The inactivation of microorganisms is proportional to the absorbed UV dose (mWs/cm^2), which is the product of UV intensity (mW/cm^2) and exposure time (s).

The most common UV disinfection technology uses low-pressure mercury arc lamps that emit UV radiation predominantly at a germicidal wavelength of 253.7 nm (White et al., 1986; Bitton, 1994; U.S.EPA, 1999d). The output efficiency of a low-pressure UV lamp is about 30-40 % and the output energy is quite low (10-50W). Another UV disinfection technology uses medium-pressure UV lamps that emit polychromatic radiation. Their output efficiency is lower but their output energy (200 W) is high. UV reactors using medium-pressure UV lamps require less space, but the medium-pressure UV lamps have higher running costs due to the higher energy demand. There are two main types of UV disinfection reactors: in contact type reactors UV lamps are encased in a quartz sleeve that is submerged in the flowing water in a tank or canal, while in the non-contact type reactor, the UV lamps and sleeves are not in contact with water.

UV disinfection typically efficiently eliminates enteric bacteria, viruses, bacterial spores and parasite cysts, but the sensitivities of different microorganisms to UV radiation vary strongly, however (Meng and Gerba, 1996; Oppenheimer et al. 1997; Lazarova et al. 1998; Liberti and Notarnicola 1999; Collivignarelli et al. 2000; Liberti et al. 2000; Bourrouet et al., 2001; Craik et al., 2001; Jacangelo et al., 2003; Rajala et al. 2003). Typically 20-45 mWs/cm^2 UV doses achieve 2-5 log reductions of indicator bacteria TC, FC and FS in secondary or tertiary treated wastewaters, while higher doses may be needed for efficient elimination of viruses, bacterial spores and parasite cysts or to achieve very low microbial numbers, such as the State of

California limit value of ≤2.2 MPN/100 ml TC for non-restricted effluent reuse (Jolis et al., 1996; Kuo et al., 1997; Lazarova et al., 1999; Sakamoto et al. 2001; Jacangelo et al., 2003; Nasser et al. 2006).

UV disinfection efficiency is influenced by several factors, including the hydraulic properties of the reactor and the wastewater characteristics, such as initial microbial numbers, UV absorbance and the SS concentration (White et al., 1986; Jolis et al., 1996; Kuo et al., 1997; Salgot et al. 2002). To achieve the best performance from a UV unit, it is desirable to maximise mixing in the direction perpendicular to the flow and to minimise mixing in the same direction as the flow. Under these conditions, all the microorganisms in the water to be treated experience approximately the same irradiation dose in the UV reactor and short-circuiting is prevented. Configuration of the lamps and UV lamp aging are two factors which can influence UV intensity in a reactor. Water quality (e.g. iron, humic substances, dissolved organic matter and SS) has a strong effect on lamp fouling and disinfection efficiency. Water impurities may scatter or absorb UV radiation, thus protecting microorganisms and increasing the needed UV dose to achieve efficient disinfection. The UV lamps must also be regularly cleaned by mechanical, chemical or ultrasonic cleaning methods (Jolis et al., 1996; Bitton, 1994; Andreadakis et al., 1999; U.S.EPA, 1999b). Thus the upstream treatment processes should continuously produce high quality water for UV disinfection.

UV technology has many advantages that have made it an attractive alternative disinfection method. The main benefit of a UV system is its relatively high disinfection efficiency without the production of harmful or toxic DBPs and chemical residues (Oppenheimer et al., 1997; Liberti and Notarnicola, 1999; Collivignarelli et al., 2000; Liberti et al., 2002). The use of UV technology eliminates the need for transport, handling and storage of (hazardous) disinfection chemicals. UV treatment units have small space requirements and a short contact time compared to chemical disinfection. UV disinfection is safe and relatively simple to use.

The disadvantage of UV disinfection is the lack of a bacteriostatic effect and the possibility for regrowth of the microbial population under certain conditions (Lazarova et al. 1998; Collivignarelli et al., 2000). Microorganisms treated by UV irradiation may only lose their culturability, but stay alive (Blatchley et al., 2001). Some UV-damaged microorganisms may undergo enzymatic photoreactivation or dark repair, which can repair the UV-damaged microorganisms and enable them to reproduce again (Bitton, 1994; Baron and Bourbigot, 1996; Baron, 1997; Tosa and Hirata, 1999; Oguma et al., 2001; Zimmer and Slawson, 2002). In photoreactivation, enzyme activities can repair the damage to the DNA, when UV-damaged microorganisms are exposed to visible light (wavelength 300-500 nm). Dark repair can occur by enzyme activities occurring in the dark.

2.4.3.5 Combined disinfection treatments

Different disinfection methods can be combined in order to achieve better disinfection efficiencies or synergistic benefits. The synergy means that the efficiency of combined disinfection method is higher than the efficiency achieved when summing all the single effects. The combined use of chloramine and copper was reported to achieve synergistic inactivation of MS2 coliphage and *E. coli* in disinfection of the well water (Straub et al., 1995). Biswas et al. (2003) reported synergistic inactivation of *Cryptosporidium parvum* using ozone followed by free chlorine in the disinfection of natural waters. Butkus et al. (2004) observed a synergistic inactivation of MS2 coliphage with the combined use of UV irradiation and silver in water.

Recently, advanced oxidation processes (AOPs) for disinfection of water and wastewater have gained increased attention (Lubello et al., 2002; Caretti and Lubello 2003; Cho et al., 2004; Sommer et al., 2004). AOPs have proven to be very effective in treating a wide variety of organic pollutants in contaminated groundwaters and industrial effluents (Hirvonen, 1999; Sommer et al., 2004). AOPs are based on radical type reactions using powerful oxidising intermediates, mainly hydroxyl radicals (OH·), to oxidise the organic pollutants. The hydroxyl radicals can be generated in several ways, e.g. by the combined application of ozone/hydrogen peroxide, UV/ozone and UV/hydrogen peroxide (Hirvonen, 1999; Sommer et al., 2004). Increased disinfection efficiency and synergistic benefits in combined PAA/UV disinfection of wastewater effluents have been reported (Rajala-Mustonen et al., 1997; Lubello et al., 2002; Caretti and Lubello, 2003). Cho et al. (2004) demonstrated a linear correlation between the amount of hydroxyl radicals and the extent of *E. coli* inactivation in TiO_2 photocatalytic disinfection. They assessed that the hydroxyl radical is one thousand to ten thousand times as effective as other chemical disinfectants such as chlorine and ozone in *E. coli* inactivation. Sommer et al. (2004) reported a good disinfection efficiency of the ozone/hydrogen peroxide process with respect to vegetative bacteria and bacteriophages.

2.4.3.6 Membrane filtration processes

Membrane separation processes, including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO), are promising technologies for removal of microorganisms from waters and have recently received considerable attention for the advanced treatment of municipal wastewaters (Jolis et al., 1996; Lazarova et al., 1999; Mujeriego and Asano, 1999; Collivignarelli et al., 2000; Hu et al., 2003; Farahbakhsh and Smith, 2004; Sano et al., 2006). The principal advantages of these processes include efficient microbial removal without producing harmful DBPs. The membrane processes separate microorganisms that are larger than the cut-off size ("pore size") of the membrane, the separation efficiency being affected also by membrane fouling, operational pressure and permeate flux (Hu et al., 2003; Sano et al., 2006). Jolis et al. (1996) reported that a pilot-scale MF system produced an effluent which was virtually free of suspended solids, turbidity and coliform bacteria (around 4 log reductions of TC), and achieved 1.9 log average removal of MS2 phage. Farahbakhsh and Smith (2004) achieved complete removal of TC and FC (4-6 log reductions) when treating secondary effluent by a MF process, though the removal of coliphages was more variable (0.2-3.4 log). Hu et al. (2003) reported 0.37-1.83 log, 1.93-3.05 log and 3.52-4.40 log reductions of MS2 coliphage in UF, NF and RO processes, respectively. The applicability of membrane technology is typically dependent on the appropriate pre-treatment. The useful life of the membrane depends on the fouling and scaling characteristics of water to be treated. At the moment, membrane technology is relatively expensive.

2.5 Wastewater discharge, reclamation and reuse

The most common method for wastewater effluent disposal is by discharge into natural waters. In recent years, the recycling and reuse of wastewaters has been increasing in many countries, as wastewater reuse reduces the needs for consumption of natural water resources, and also improves the quality of surface waters due to decreased wastewater discharges. There are many wastewater reuse applications, e.g. irrigation of agricultural lands, urban reuse, industrial reuse, environmental reuse and groundwater recharge (Asano and Levine, 1996; Bontoux and Courtois, 1996; Krofta et al., 1996; Bonomo et al., 1999; Mujeriego and Asano, 1999; Barbagallo et al., 2001; Sakamoto et al., 2001; Anderson, 2002; Angelakis et al., 2002; Nurizzo, 2002; Nasser et al., 2006).

3 AIMS OF THE STUDY

The aim of this study was to evaluate the efficiency of different wastewater treatment processes on the removal of enteric microorganisms, phosphorus and organic matter from municipal wastewaters. The specific aims were:

- To study the effect of conventional wastewater treatment (primary and secondary treatment with simultaneous phosphorus precipitation) on microbiological and physico-chemical wastewater quality in full-scale municipal WWTPs and to assess the elimination of enteric microorganisms in tertiary rapid sand filtration (RSF), biological contact filtration and biological-chemical contact filtration processes. (I)
- To evaluate in laboratory-scale disinfection experiments the relative disinfection efficiencies of PAA, H_2O_2, NaOCl and UV disinfection treatments, and to investigate if combined chemical-UV disinfection treatments would provide any synergy benefits. (II)
- To investigate at the pilot-scale the PAA disinfection of primary, secondary and tertiary treated municipal wastewaters and to evaluate the effect of process conditions on disinfection efficiency. (III)
- To assess in pilot-scale experiments the efficiency of DAF process for the tertiary treatment of wastewaters. The DAF treatment of primary wastewater effluents was investigated to evaluate the applicability of the process for treatment of WWTP by-pass wastewaters. (IV)

4 MATERIALS AND METHODS

4.1 Conventional wastewater treatment

In substudy I, the occurrence and elimination of enteric microbes in Finnish full-scale biological-chemical wastewater treatment plants (Helsinki, Espoo, Turku and Kuopio WWTPs) was studied. The flow diagrams of the WWTPs are presented in the Figure 1.

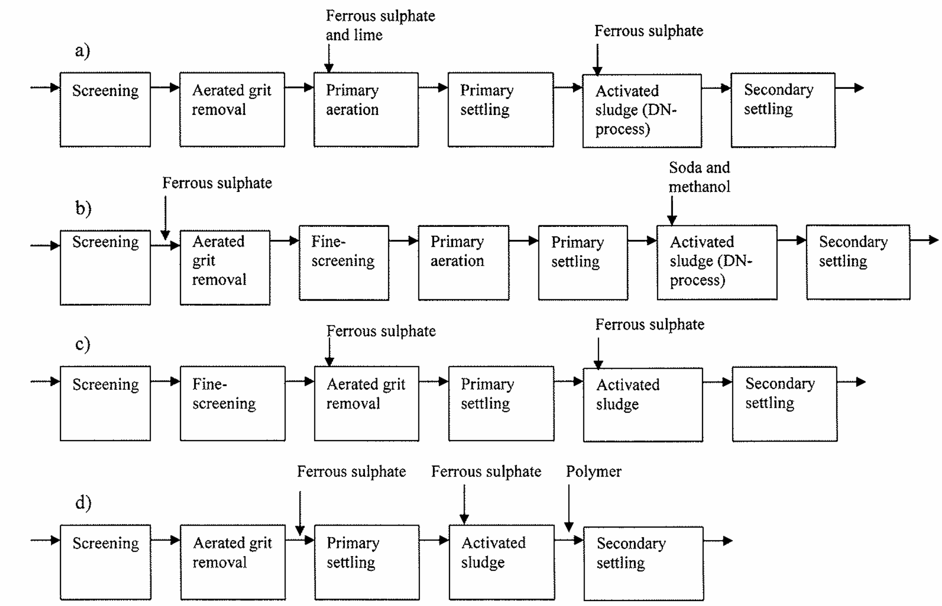

Figure 1. Flow diagrams of a) Helsinki, b) Espoo, c) Turku and d) Kuopio WWTPs

Average wastewater flow into the Helsinki WWTP (population equivalent, pe, 1 000 000) is approximately 270 000 m^3/d. The treatment process (Figure 1a) typically removes BOD_7 and total phosphorus by over 90 % and total nitrogen by over 60 %. In the Espoo WWTP (pe. 250 000), the average wastewater flow is 75 000 m^3/d. Around 95 % reductions of BOD_7 and P_{tot}, and approximately 65 % reductions of N_{tot} are typically achieved (Figure 1b). The Turku WWTP (pe. 140 000) receives approximately 65 000 m^3/d daily wastewater flow. The treatment process (Figure 1c) typically removes BOD_7 and P_{tot} by over 90 %. Average wastewater flow into the Kuopio WWTP (pe. 105 000) is approximately 22 000 m^3/d and over 90 % reductions of BOD_7 and P_{tot} are typically achieved (Figure 1d).

To assess microbial reductions in the WWTPs, influent and final effluent samples (24 h composite samples) collected from the WWTPs were analysed for enteric microbial numbers.

4.2 Tertiary wastewater treatment

4.2.1 Rapid sand filtration (RSF)

In substudy I, the efficiency of tertiary RSF at removal of enteric microorganisms was studied in a pilot-scale experiment. The RSF unit (Figure 2) was installed in the Kuopio WWTP and used to treat the secondary effluent of WWTP during a 3 month period (I, Table 1).

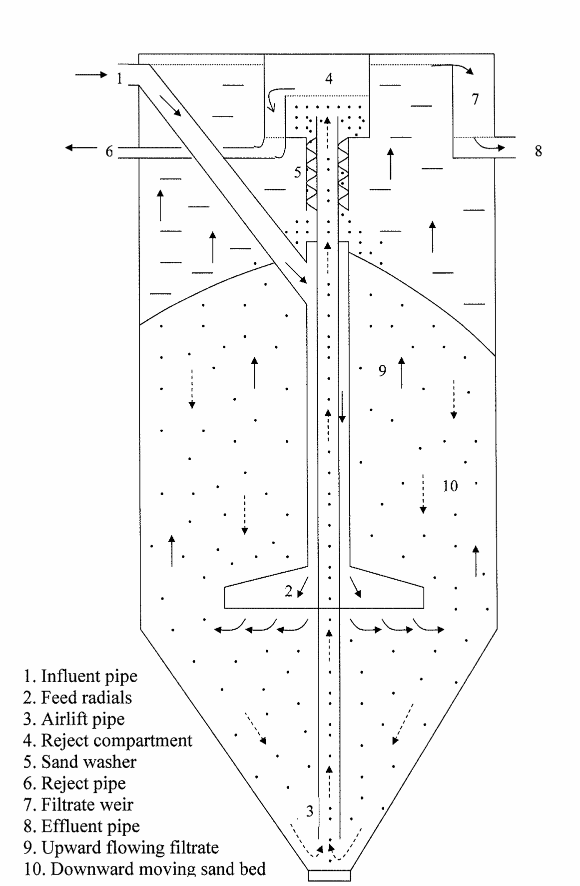

Figure 2. Schematic of the rapid sand filtration (RSF) pilot-plant (DynaSand)

The RSF unit was a deep-bed, up-flow sand filter with continuous backwash. It had a cross-section of 0.7 m^2 and it contained 3.5 m of 0.9-1.2 mm particle size sand. Influent feed was introduced at the top of the filter and flowed downward through an annular section between the influent feed pipe and airlift housing. The feed was introduced into the bottom of the sand bed through a series of feed radials. The influent wastewater flowed upwards through the downward-moving sand bed. The filtrate then exited the filter from an effluent weir at the top of the filter unit and leaved the filter unit through the effluent pipe.

The RSF filter medium was continuously backwashed. The sand bed was drawn downward into the suction of an air-lift pipe. Compressed air was introduced into the bottom of the pipe, where a fluid mixture of air, sand and water was formed. This mixture was transported hydrostatically up through an air-lift pipe and during the transport, the sand grains were cleaned of accumulated material by abrasion and fluid shear forces. The slurry was pushed to the top of the airlift and into the reject compartment. From the reject compartment, the sand fell into the sand washer and the lighter reject solids were carried over the reject weir and out of the reject pipe. In the sand washer, the accumulated material was removed from the sand over a weir and washing of sand took place. After passing through the sand washer, the clean sand was distributed to the top of the filter bed. A portion of the filtrate flowed up through the sand washer and carried the debris that had been sheared from the sand over the reject weir. The quantity of filter washing water was typically around 5-10 % of the influent flow rate.

The RSF was operated as a contact filter by dosing polyaluminium chloride coagulant into the wastewater (dose <1.0-2.2 g Al/m^3, PAX-18, 9 % Al, Kemira Chemicals Ltd Kemwater) before the sand filter. The RSF operation as a mechanical filter without coagulant dosing was also investigated. The hydraulic load of the RSF was 7-8 m/h.

To assess microbial reductions in the processes, RSF influent and effluent samples (24 h composite samples) were collected at four different times and analysed for enteric microbial numbers. The effect of RSF on physico-chemical wastewater quality was also assessed (Rajala et al. 2003).

4.2.2 Chemical contact filtration and biological-chemical contact filtration

In substudy I, the efficiencies of tertiary biological-chemical contact filtration and chemical contact filtration processes for removal of enteric microorganisms were studied in pilot-scale experiments in the Espoo WWTP. The filtration pilot plants were used to treat effluents from a technical-scale activated sludge process with biological nitrogen and phosphorus removal (Figure 3). The activated sludge process was fed with the WWTP influent wastewater (100 m^3/d) after the screening phase.

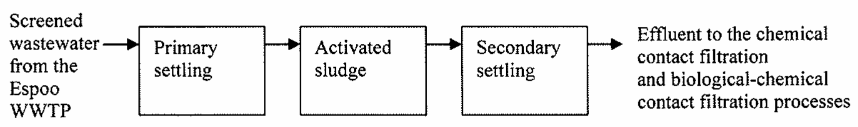

Figure 3. Flow diagram of the technical-scale biological nutrient removal process

The chemical contact filter was designed for the removal of phosphorus from the wastewater. It was a gravitation filter, containing from top to bottom a 60 cm layer of 1.25-2.5 mm grain size anthracite, 60 cm of 0.8-1.5 mm grain size sand, 20 cm of 3-5 mm grain size sand and 20 cm of 5-9.5 mm grain size rubble (Figure 4a). Polyaluminium chloride coagulant (dose 3.2 g Al/m^3, PAX-18) or an iron coagulant (dose 0.7 g Fe/m^3, PIX-115, 11 % of Fe, Kemira Chemicals Ltd Kemwater) was dosed into the wastewater before the filter. Wastewater flow to the filter was approximately 15 m^3/d. The hydraulic load on the filter was 5-7.9 m/h.

The biological-chemical contact filter was designed for the removal of nitrogen and phosphorus from wastewater. It was an upflow filter, containing from top to bottom a 2.25 m layer of 3-6 mm particle size gravel and a 25 cm layer of 5-9.5 mm particle size rubble (Figure 4b). Polyaluminium chloride coagulant (dose 6 g Al/m^3, PAX-18) was dosed into the wastewater before the filter. Methanol was dosed into the wastewater to improve the biological nitrogen removal by denitrification. Wastewater flow to the filter was approximately 10 m^3/d. The filter was operated at 8 and 16 m/h hydraulic loads, corresponding to 20 and 10 minute contact times of wastewater with the filter. The washing of filter was by counter current washing. The quantity of filter washing water was at maximum 12 % of the influent flow rate.

To assess microbial reductions in the filtration processes, influent and effluent samples (24 h composite samples) were collected and analysed for enteric microbial numbers.

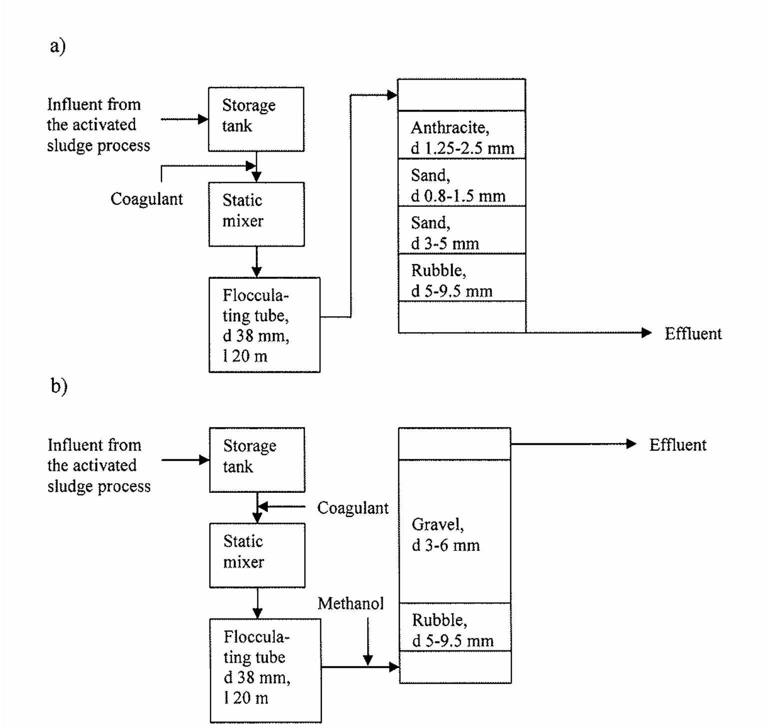

Figure 4. Flow schemes of the pilot-scale a) chemical contact filter and b) biological-chemical contact filter units

4.2.3 Dissolved air flotation (DAF)

In substudy IV, tertiary treatment of wastewater by a DAF process was studied in a pilot-scale experiment. The DAF pilot-plant (Figure 5) was installed in the Kuopio WWTP.

The DAF process was fed with the secondary effluent of the Kuopio WWTP. Coagulant chemical (polyaluminium chloride PAX XL-60, Kemira Oyj, Kemwater) and organic flocculation aid (anionic Fennopol A305 or cationic Fennopol K5060, Kemira Oyj, Kemwater) were dosed into the wastewater before it entered the flocculation unit of the pilot-plant through rapid-mixing (static mixer). After the flocculation of wastewater in two flocculation chambers, each chamber having one horizontal shaft mechanical mixer, wastewater entered the inlet shaft of the DAF tank. In the inlet shaft, wastewater was mixed with the pressure relieved dispersion water that was produced by pumping the DAF process effluent and compressed air into the pressure vessel (unpacked saturator) operating at 500-600 kPa pressure. The microbubble-solid aggregates were transferred onto the surface of the DAF tank, while the purified wastewater left the DAF tank through an underflow wall on the bottom of the tank. The separated sludge was removed mechanically (sludge scraper) from the surface of the DAF tank to a storage container and further pumped by submersible pump into the sludge treatment process of the WWTP.

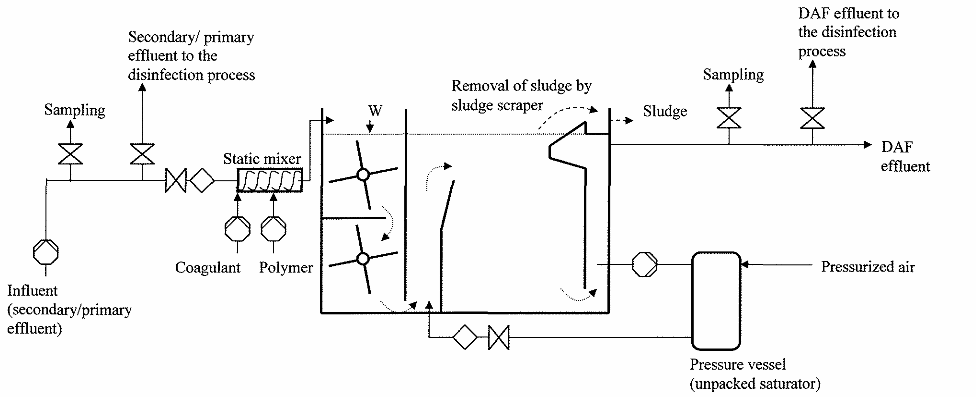

Figure 5. Simplified flow scheme of the DAF pilot-plant and connections to the PAA disinfection pilot-plant

During the experiments, the effects of changing the coagulant and polymer dosages, flocculation conditions, dispersion water recycle ratios (R= Q_r/Q_{flot} ×100 %, where Q_r is the dispersion water flow and Q_{flot} is the wastewater flow into the DAF process) and hydraulic loading rates were tested. The dispersion water recycle ratio into the DAF tank was adjusted by changing the number of injector nozzles (max. 13 nozzles).

To assess the efficiency of the DAF processes, influent and effluent samples were collected and analysed for enteric microbial numbers and physico-chemical wastewater quality parameters. During the adjustment of the process operating parameters, grab samples were collected from the process. When an optimal DAF process performance had been achieved, 24 h composite samples were collected daily from the process.

4.3 Primary wastewater treatment

4.3.1 Dissolved air flotation (DAF)

In substudy IV, the pilot-scale DAF process (Figure 5) was used for treatment of Kuopio WWTP primary effluents. The aim was to assess the efficiency of the DAF process for the treatment of WWTP by-passes. During the study, the quality of primary wastewater effluent entering the DAF process differed from the typical composition of by-pass water, as will be discussed in the later chapters. The effects of different coagulant dosages, flocculation conditions and dispersion water recycle ratios were tested.

To assess the efficiency of the processes, influent (WWTP primary effluent) and effluent samples from the DAF unit were collected and analysed for enteric microbial numbers and physico-chemical wastewater quality parameters. During the adjustment of the process operating parameters, grab samples were collected from the process. When an optimal DAF process performance had been achieved, 24 h composite samples were collected daily from the process. During the experiments, samples from the raw sewage entering the WWTP were also collected to investigate the effect of primary wastewater treatment processes on the wastewater quality.

4.4 Full-scale tertiary DAF experiments

The performance of full-scale tertiary DAF processes was studied in two municipal wastewater treatment plants, Heinävesi WWTP (Figure 6a) and Pieksämäki WWTP (Figure 6b). The aim was to obtain information about the DAF processes performance in different full-scale treatment plants, and to compare the treatment results of the plants to those achieved in the pilot-scale experiments.

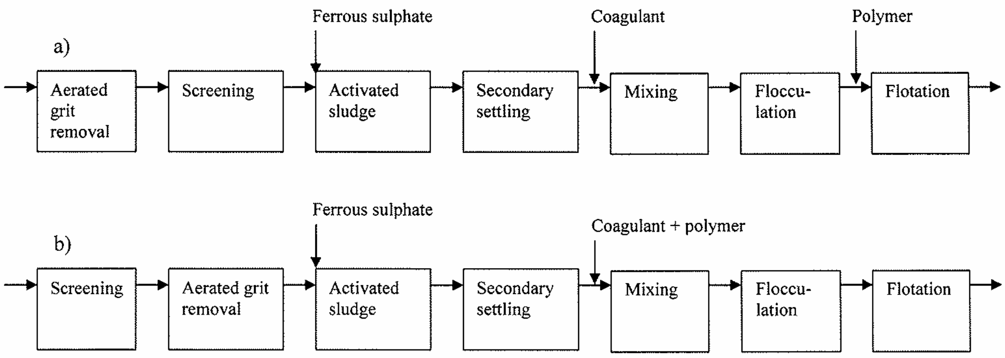

Figure 6. Flow diagrams of a) Heinävesi and b) Pieksämäki WWTPs

Heinävesi WWTP: Average wastewater flow into the Heinävesi WWTP is approximately 600 m^3/d and about 1000 people are connected to the sewer system. The wastewater treatment process contains primary treatment with aerated grit removal and screening, followed by an activated sludge process with simultaneous phosphorus precipitation and secondary settling. The final treatment of wastewater is by a tertiary DAF process. The treatment plant is clearly affected by a nearby laundry, whose wastewaters at times contain high amounts of detergents, which can disturb the secondary settling process of the WWTP and this then leads to a high load of suspended solids entering into the DAF unit.

Wastewater from the secondary settling entered through a rapid mixing unit and two flocculation chambers to the DAF basin. Coagulant application was done prior to entering the mechanical rapid mixer, which was located before the inlet of the flocculation tank. The first flocculation tank had one vertical shaft mechanical mixer and an average flocculation time of approximately 1.6 h. The second flocculation tank had two horizontal shaft mechanical mixers and an average flocculation time of approximately 0.8 h. The flocculation time used in the process was much longer than typically used prior to DAF process, as discussed later. The rotational speed of the mixer blades could not be adjusted, but they were constant and had not been determined. No coagulant chemical was regularly used. Coagulant was dosed only in the cases when increasing amounts of suspended solids escaped from the secondary settling process. The dosing of coagulant was based on secondary effluent turbidity measurement: when the turbidity was below 6 NTU, no coagulant was added, when turbidity exceeded 6 NTU, then 120 g/m^3 PAX-14 polyaluminium coagulant was dosed (9 gAl^{3+}/m^3), and when turbidity was above 50 NTU, 220 g/m^3 PAX-14 was used (16 gAl^{3+}/m^3).

After the flocculation process, the wastewater entered the DAF tank (area 33 m^2). Wastewater was mixed with the dispersion water in the reaction zone of the DAF tank. Dispersion water was produced by recirculating DAF treated wastewater into a pressure vessel that operated at 7 bar pressure. The dispersion water flow-rate to the flotation process was constant and had not been measured. The separation of solids was improved by the constant dosing of an organic polymer (3.75 g/m^3 anionic polymer). The treated wastewater then left the process from the bottom of the DAF tank. Separated sludge was removed mechanically from the surface of the DAF tank and transferred into the sludge thickener. The hydraulic loading rate of the tertiary DAF process was below 1 m/h during the experiments, which is the normal range for that particular process. The hydraulic loading of the process was much lower than typically used for DAF process, which is due to fact that the process has been built to an old sedimentation basin, and as a result it is over-dimensioned.

Pieksämäki WWTP: Wastewater flow-rate into the Pieksämäki WWTP is typically between 3500-6000 m^3/d and approximately 14 000 people are connected to the sewer system. The wastewater treatment process contains primary treatment with screening and aerated grit removal, followed by an activated sludge process with simultaneous phosphorus precipitation and secondary settling. Final treatment of wastewater is by the tertiary DAF process.

Wastewater from the secondary settling entered the rapid mixing unit (mechanical mixing, average retention time of 0.1-0.2 h). The coagulant chemical, ALF-30 (mixture of aluminium and ferric sulphate) and a cationic organic polymer (Fennopol K504), was added into the wastewater before the rapid mixing unit. After the rapid mixing, wastewater entered two parallel flocculation basins, both having one vertical shaft mechanical mixer and an average flocculation time of approximately 1.0-1.7h. The flocculation time was longer than those typically used prior to DAF process, as discussed later. The rotational speed of the mixer blades could not be adjusted, but they were constant and had not been determined.

After the flocculation process, the wastewater entered the DAF tank (total area of 100 m^2). At the inlet of the DAF tank, the wastewater was mixed with the dispersion water, produced by recycling the DAF process effluent into a pressure vessel that operated at 4 bar pressure. The dispersion water recycle ratio varied between 20-29 % (mean 26 %) during the experiments. The purified wastewater left the process from the bottom of the DAF tank, while separated sludge was removed mechanically from the surface of the DAF tank and transferred into the sludge thickener. The hydraulic loading rate of the tertiary DAF process was 1.6-2.5 m/h during the experiments, which is the normal range for that particular process. The hydraulic loading of the process was lower than typically used for DAF process, which is due to over-dimensioning the process.

Sampling of the DAF processes was done at 15 different times in both wastewater treatment plants. 24 h composite samples were collected from the influent and effluent of the DAF processes and analysed for enteric microbial numbers and physico-chemical wastewater quality parameters.

4.5 Wastewater disinfection

4.5.1 Laboratory-scale disinfection experiments

In substudy II, disinfection efficiencies of peracetic acid (PAA), hydrogen peroxide (H_2O_2), sodium hypochlorite (NaOCl) and ultraviolet (UV) irradiation disinfection treatments as well as the synergistic effects of the combined use of chemical disinfectant and UV irradiation were studied in laboratory-scale experiments. UV disinfection treatments were carried out with a collimator device (Blatchley III, 1997) and a low pressure mercury arc lamp as a UV source. In a collimator device, UV lamp is enclosed in a horizontal tube with a vertical collimating tube (Figure 7). The collimating tube directs the UV radiation directly to the sample.

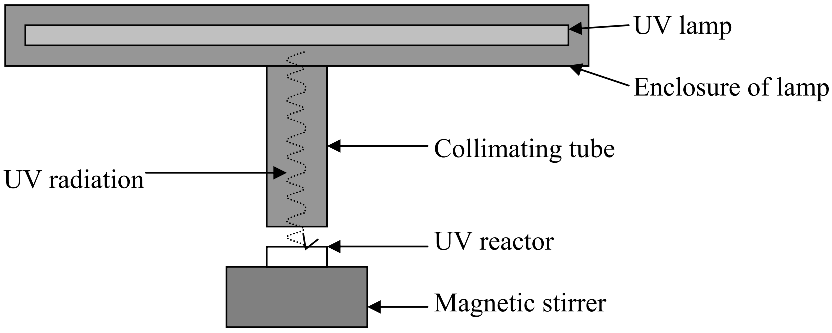

Figure 7. Schematic diagram representing the structure of a collimator device

Peptone water (PW) test medium was prepared to be used in the experiments (II). Culture collection strains of *Escherichia coli* (ATCC 15597), *Enterococcus faecalis* (ATCC 19433), *Salmonella enteritidis* (B678/95/1, obtained from the National Veterinary and Food Research Institute) and coliphage MS2 virus (ATCC 15597-B1) were used as test microorganisms. Prior to each disinfection experiment, test microorganisms were cultured and added into PW to yield a microbial density of approximately 10^5-10^7 CFU or PFU/ml in the test medium. A 10 ml volume of test medium was transferred onto a glass petri dish and mixed with a magnetic stirrer.

In the chemical disinfection treatments, the disinfectant chemical was added into the test medium and a 10 minute contact time was allowed, after which the residual concentration of disinfectant was measured by commercially available analytical test. The residual PAA residues were quenched by the addition of sodium thiosulphate and catalase, while chlorine residues were quenched by sodium thiosulphate and H_2O_2 residues by catalase. In the UV disinfection treatments, the applied UV doses (mWs/cm^2) were calculated as the product of average UV intensity in the reactor (mW/cm^2) and irradiation time (s). In the combined PAA/UV and H_2O_2/UV treatments, the UV irradiation was always started 30 seconds after the addition of disinfectant chemical and was stopped by shielding of the collimating tube after the desired UV irradiation time.

Four replicate tests were carried out for each disinfection treatment. During each set of experiments, two untreated samples were taken to determine the initial microbial numbers.

4.5.2 Pilot-scale disinfection experiments

In substudy III, the peracetic acid disinfection of primary, secondary and tertiary treated wastewaters was studied at a pilot-scale. The disinfection pilot-plant was a plug-flow reactor (made of polyethylene plastic) and a wastewater volume of approximately 0.4 m^3 (Picture 1).

The disinfection pilot plant was fed with the primary or secondary effluent of the Kuopio WWTP, or tertiary effluent from the pilot-scale DAF unit treating WWTP secondary effluent (Figure 5). The wastewater effluent was fed into the disinfection reactor at a flow rate of 1

m^3/h. Wastewater first entered the mixing chamber of the reactor, was mixed hydraulically with the PAA disinfectant chemical which was pumped from a storage container, and then entered the disinfection reactor through an overflow wall. PAA doses of 2-7 mg/l (as pure PAA) were used for the disinfection of secondary and tertiary effluents, while 5-15 mg/l PAA was used for the disinfection of primary effluents.

Samples were collected from the influent and from different stages of the disinfection reactor, corresponding to contact times of 4-27 min. The residual PAA concentration was measured by commercially available analytical test and PAA residues in samples were quenched by the addition of sodium thiosulphate and bovine catalase. A total of 2-9 replicate tests were carried out for each disinfection treatment, the numbers being affected by the variability of the disinfection efficiency between parallel tests and the aim being to perform each disinfection treatment at various wastewater qualities.

Picture 1. PAA disinfection pilot-plant

4.6 Analytical methods

4.6.1 Enumeration of microorganisms

In substudy I, enumeration of salmonella was by the MPN-method by using an ISO two-step enrichment method (ISO 6579, 1993) and culturing on selective agar plates. The preliminary confirmation of Salmonella isolates was by the slide agglutination test, by using antisera (Salmonella O Antiserum Poly A-1 and Vi, Difco laboratories Detroit, Michigan, USA). The final serotyping and phage typing of 197 isolates was carried out in the National Salmonella Centre, Laboratory of Enteric Pathogens, National Public Health Institute, Helsinki. The sensitivity of the Salmonella isolates to 12 antimicrobial agents was determined by the disc diffusion method according to the National Committee for Clinical Laboratory Standards (NCCLS, 1997).

In substudy II, enumeration of *Escherichia coli*, *Enterococcus faecalis* and *Salmonella enteritidis* was by culturing on TYG-agar by using a spread-plate-technique. Enumeration of

Coliphage MS2 was by using a double-agar-layer-method on phage TYG-agar, with *E. coli* ATCC 15597 as the host bacterium. After the incubation period, bacterial colonies and virus plaques were counted and the results calculated as CFU or PFU/ml.

In substudies I, III and IV, faecal indicator microorganisms were analysed from wastewater samples. Faecal coliforms (FC), total coliforms (TC) and enterococci (EC) were enumerated by using a spread-plate-technique from appropriately diluted samples and the membrane filtration method using 0.45 µm pore size nitrocellulose filters. FC, TC and EC were cultured on mFC agar (Difco), Les Endo agar (Merck KGaA, Darmstadt, Germany) and Slanetz & Bartley agar (Lab M, IDG plc, Bury, Lancashire, UK), respectively. After incubation, bacterial colonies were counted and microbial numbers calculated as CFU/100 ml. F-RNA coliphages (Cp) were determined on phage TYG-agar by using a double-agar-layer-method (Adams, 1959), with *E. coli* ATCC 15597 as the host bacterium. After the incubation period, virus plaques were counted and the results calculated as PFU/100 ml.

4.6.2 Physico-chemical analysis

In substudy I, the laboratories of the wastewater treatment plants carried out the process condition measurements and the physico-chemical wastewater analyses.

In substudy II, turbidity (Hach Ratio X/R Turbidimeter), UV-absorbance at 253.7 nm wavelength (Shimadzu UV-1201 UV-Vis Spectrophotometer), pH (Knick Portamess 751 pH-meter), chemical oxygen demand (COD_{Cr}, Hach DR/2010 spectrophotometer, standard SFS 5504) and total organic carbon (TOC, APHA, AWWA, WPCF, 1998) values of the test medium were measured.

In substudies III and IV, the following physico-chemical parameters were measured from the wastewater samples: pH (Knick Portamess 751 pH-meter), temperature, chemical oxygen demand (COD_{Cr}, Hach DR/2010 spectrophotometer, according to standard SFS 5504), suspended solids (SS), total phosphorus (P_{tot}), turbidity (HACH Ratio X/R turbidimeter), apparent colour (HACH DR 2000 spectrophotometer), UV transmittance (253.7 nm, Shimazu UV-1201 UV-Vis spectrophotometer) and total aluminium residue (HACH DR 2000 spectrophotometer). Analyses were conducted by using the recommended standard methods (APHA, AWWA, WPCF, 1998).

4.7 Data analysis and presentation of results

In substudies I and IV, the efficiency of wastewater treatment processes was assessed by determining the reductions in the microbial numbers and the improvement of the physico-chemical quality of wastewater in the processes. In substudy I, linear regression analysis after log-transformation of microbial numbers was used to assess correlations between wastewater enteric bacterial numbers and physico-chemical wastewater quality parameters or process parameters.

In substudies II and III, the efficiency of disinfection was assessed by determining microbial reductions with different disinfection treatments. Microbial reductions as a function of $C \times t$ products were determined, where C is the applied disinfectant dose (mg/l) and t is the contact time (min). The synergy values of combined chemical/UV disinfection treatments were calculated by the following equation:

Synergy (log units) = log reduction by combined chemical/UV disinfection –
(log reduction by UV disinfection + log reduction by chemical disinfection) (13)

Wilcoxon signed ranks test was used to test if the synergy values achieved in combined disinfection treatments were statistically significant. A simple correlation test was used to evaluate the effect of wastewater quality and temperature on PAA disinfection efficiency.

5 RESULTS

5.1 Conventional wastewater treatment

Conventional biological-chemical wastewater treatment typically removed 90-99 % of biological oxygen demand (BOD), suspended solids (SS) and total phosphorus (P_{tot}) from the wastewaters (I, Table 1). The mean reductions of indicator bacteria varied from 96 to 99.8 % (I, Table 3), while the reductions of Salmonella bacteria were from 95 to >99.9 % (I, Table 2). The numbers of indicator bacteria in raw wastewater and secondaty effluent exhibited quite high variability. Salmonellae were detected from all the influent samples (93-11000 MPN/100 ml) and most of the effluent samples (<3-240 MPN/100 ml) of municipal WWTPs.

A total of 32 Salmonella serovars were found from the wastewaters in four WWTPs (I, Table 5). Almost half of the isolates (87 strains) belonging to 13 different serovars displayed resistance to at least one of the 12 antimicrobial agents tested (I, Table 6). In all, 62 isolates (32 %) of seven serovars were resistant to nalidixic acid and 36 isolates (18 %) of eight serovars were multiresistant, showing resistance against three to six antimicrobials.

Microbial numbers in secondary treated effluents exhibited some correlations with effluent residual BOD, COD, suspended solids and total phosphorus concentrations (I, Table 4). Weaker correlations were shown with residual total nitrogen concentration, wastewater flow rate and pH as well as with the activated sludge process retention time and oxygen concentration.

5.2 Tertiary wastewater treatment

5.2.1 Rapid sand filtration (RSF)

The use of tertiary RSF as contact filtration reduced FC numbers on average by 97 % (I, Table 3). The numbers of Salmonellae were always less than the detection limit (<3 MPN/100 ml) after the RSF treatment, while their numbers in the RSF influent (secondary effluent) varied between <3-240 MPN/100 ml (I, Table 2). When the RSF process was operated as a mechanical filter (without polyaluminium chloride coagulant chemical) a 25 % reduction in FC numbers was achieved. The tertiary RSF unit efficiently reduced SS, COD and phosphorus concentrations in wastewater effluents (Rajala et al. 2003).

5.2.2 Chemical contact filtration and biological-chemical contact filtration

Pilot-scale chemical contact filter and biological-chemical contact filter for tertiary treatment of wastewater reduced FC numbers on average by 39 and 71 %, respectively (I, Table 3). Salmonella numbers in the influents of filters varied between <3-3.6 MPN/100 ml and they were never detected from wastewaters after the filtration units (<3 MPN/100 ml) (I, Table 2). The filtration units also affected the physico-chemical quality of wastewater (Rantanen, 2001).

5.2.3 Dissolved air flotation (DAF)

The increase in the coagulant dose improved the reductions of TC and P_{tot} in the tertiary DAF process (IV, Figure 1). When using 5-10 gAl^{3+}/m^3 (70-137 g/m^3 PAX) coagulant dose, 95-98 % reductions of TC and 70-81 % reductions of P_{tot} were achieved. With lower doses of 2-4

gAl^{3+}/m^3 (30-55 g/m^3 PAX), slightly lower reductions were achieved. The mean reductions of TC (95-97 %) and P_{tot} (70-76 %) remained relatively constant, when the G-values of flocculation ranged from the slow mixing conditions (G 10 and 10 s^{-1}) to higher mixing intensities (G 55 and 26 s^{-1}) (IV, Figure 2). The increase of dispersion water recycle ratio from 11 to 22 % slightly improved the reductions of TC and P_{tot}, with their mean reductions with different recycle ratios varying between 95-97 % and 56-65 %, respectively (IV, Figure 3). Similar reduction patterns with changing coagulant dose, flocculation pattern and recycle ratio were observed also for the other water quality parameters, with the typical reductions of colour and COD_{Cr} being 40-50 % and 20-45 %, respectively.

During an experimental period in winter time (wastewater temperature 11-14ºC), when the DAF process was operated with a coagulant dose of 7 gAl^{3+}/m^3 and 5 m/h hydraulic surface load, 80-99 % reductions of enteric microorganisms were achieved (IV, Table 1). The average P_{tot} reduction in DAF process was 67 % (range 54-81 %), the average influent and effluent concentrations being 0.48 mg/l (range 0.28-0.97 mg/l) and 0.14 mg/l (range 0.08-0.21 mg/l), respectively.

During an experimental period in summer time (wastewater temperature 17-19ºC), when operating the DAF process with 3-6 gAl^{3+}/m^3 (40-85 g/m^3 PAX XL-60) coagulant dose and 5 or 10 m/h hydraulic surface load, the reductions of enteric microorganisms were typically between 85-99 %. The P_{tot} reduction in DAF process were typically between 48-86 % and the average influent and effluent total phosphorus concentrations between 0.20-0.51 and 0.04-0.17 mg/l, respectively. The increase in the coagulant dose from 3 to 6 gAl^{3+}/m^3 improved the removal of enteric microorganisms and total phosphorus, while the increase of the hydraulic surface load from 5 to 10 m/h did not clearly affect the efficiency of the DAF process.

The reductions of all of the water quality parameters were typically higher during the summer than during the winter experimental period. Wastewater turbidity was typically reduced to the level of 1-2 NTU, and the average reductions of COD_{Cr} in DAF process were 28-39 %. The concentration of suspended solids remained relatively constant. During the winter experimental period, the average influent and effluent SS concentrations were 6.0 and 12.4 mg/l, respectively. During the summer experimental period, the corresponding values were 4.1-8.5 and 5.1-9.0 mg/l. The UV-transmittance of wastewater increased in the DAF process from the average influent values of 44-53 % to average effluent values of 54-63 %.

5.3 Dissolved air flotation (DAF) treatment of primary wastewater effluents

Primary treatment of wastewater in Kuopio WWTP, preceding the pilot-scale DAF process, achieved approximately 46-99 % reductions of enteric microbial numbers and 30-80 % reductions of P_{tot}, COD and other measured water quality parameters (IV, Table 2). The DAF treatment of primary effluents, operated with a 12 gAl^{3+}/m^3 coagulant dose and 5 m/h hydraulic surface load, achieved 94-99.98 % reductions of enteric microbial numbers, 85-94 % reductions of P_{tot} (residual concentrations 0.13-0.97 mg/l), 36-60 % reductions of COD (77-180 mgO_2/l) and 66-95 % reductions of suspended solids (5-42 mg/l) (IV, Table 2).

5.4 Full-scale DAF experiments

The results of the full-scale DAF experiment and their comparison with the results obtained in the pilot-scale DAF experiments are shown in the Table 3.

Heinävesi WWTP: The tertiary DAF process of Heinävesi WWTP was operated in two different ways during the experimental period. In the first phase, the dosing of the polyaluminium coagulant (PAX-14) was not continuous, but was only started if the turbidity of secondary effluent increased above the level of 6 NTU. In the second phase, coagulant chemical was continuously dosed (see Table 3). In both phases of the experiments, cationic polymer was dosed into the wastewater.

In the first phase of the experiments, the average wastewater flow rate to the DAF process was 505 m^3/d (range 428-591 m^3/d) and the hydraulic surface load of the DAF process was 0.5-0.7 m/h (Table 3). The DAF process achieved 93 % and 96 % average reductions of TC and EC, respectively. The coliphage virus numbers in the secondary effluents were typically low, and their numbers were reduced near to or even below the detection limit of 100 PFU/100 ml in the DAF process. The mean reduction of total phosphorus was 63 % (range 26-84 %) and the mean P_{tot} concentration of the DAF effluent 0.19 mg P_{tot}/l (0.07-0.40 mg P_{tot}/l).

In the second phase of the experiments, the average wastewater flow rate to the DAF process was 581 m^3/d (range 490-672 m^3/d) and the hydraulic loading rate of the DAF process was 0.6-0.8 m/h (Table 3). The DAF process achieved 98.8 % and 99.6 % average reductions of TC and EC, respectively. Coliphage virus numbers in secondary effluents were lower than in the first experimental phase, and they were always reduced below the detection limit of 100 PFU/100 ml. The concentrations of P_{tot}, COD_{Cr} and SS in secondary effluents exhibited high variability during the experimental period. The mean reduction of P_{tot} was 95 % (range 92-98 %) and the mean effluent concentration was 0.04 mg P_{tot}/l (0.01-0.06 mg P_{tot}/l).

Pieksämäki WWTP: The average wastewater flow rate to the DAF process was 5066 m^3/d (range 3911-5936 m^3/d) and the hydraulic loading rate of the DAF process was 0.6-0.8 m/h (Table 3). The DAF process achieved 91 % average reductions of TC and EC (Table 3). The mean reduction of coliphage viruses was 97 % and they were typically reduced to near to or even below the detection limit of 100 PFU/100 ml. The mean reduction of P_{tot} was 69 % (range 42-78 %) and the mean DAF effluent concentration 0.09 mg P_{tot}/l (0.07-0.14 mg P_{tot}/l).

Table 3. The removal of enteric microorganisms (geometric mean and range) and improvements of physico-chemical parameters (mean and range) in the pilot-scale and full-scale tertiary DAF processes.

		DAF pilot plant (n=17)	DAF pilot plant (n=7)	DAF pilot plant (n=9)	Heinävesi wwtp (n=10)	Heinävesi wwtp (n=5)	Pieksämäki wwtp (n=15)
Sh (m/h)		5	5	5	0.6 (0.5-0.7)	0.7 (0.6-0.8)	2.1 (1.6-2.5)
Recycle ratio (%)		15-18	15-18	15-18	N.D.	N.D.	20-29
Coagulant dose		[a]	[b]	[c]	[d]	[e]	[f]
Total coliforms (CFU/100 ml)	Influent	6.1×10^5	8.2×10^6	8.7×10^6	4.4×10^5	7.9×10^5	3.4×10^5
	Effluent	3.0×10^4	1.1×10^6	1.6×10^5	2.1×10^4	1.9×10^3	2.4×10^4
	Reduction %	93.1 (79.7-98.8)	84.8 (70.0-92.6)	97.6 (94.4-99.5)	93.0 (85.1-99.4)	98.8 (96.5-99.99)	90.6 (71.8-98.3)
Enterococci (CFU/100 ml)	Influent	1.7×10^4	1.4×10^5	1.4×10^5	4.0×10^4	1.2×10^5	2.3×10^4
	Effluent	9.5×10^2	1.3×10^4	1.5×10^3	7.4×10^2	2.9×10^2	1.7×10^3
	Reduction %	92.7 (82.0-98.6)	89.9 (87.5-95.1)	98.7 (97.6-99.8)	95.8 (87.7-99.94)	99.6 (98.9-99.96)	91.3 (83.9-97.5)
F-RNA coliphages (PFU/100 ml)	Influent	6.2×10^4	3.0×10^4	3.3×10^4	9.1×10^2	4.9×10^2	7.5×10^3
	Effluent	4.6×10^3	8.8×10^3	1.6×10^3	8.1×10^1	$<1.0\times10^2$	1.3×10^2
	Reduction %	91.9 (86.2-97.6)	68.1 (43.8-84.1)	94.5 (89.6-98.2)	86.5 (63.0-99.2)	>80.0	96.9 (90.5-99.7)
Total phosphorus (mg/l)	Influent	0.48 (0.28-0.97)	0.41 (0.31-0.51)	0.31 (0.24-0.41)	0.51 (0.36-0.68)	1.14 (0.14-3.39)	0.31 (0.24-0.40)
	Effluent	0.14 (0.08-0.21)	0.14 (0.10-0.17)	0.06 (0.04-0.11)	0.19 (0.07-0.40)	0.04 (0.01-0.06)	0.09 (0.07-0.14)
	Reduction %	67.2 (54.1-80.5)	65.1 (58.1-72.1)	80.9 (66.7-86.2)	62.6 (25.9-83.7)	94.9 (92.3-98.2)	69.4 (41.7-77.5)
COD (mg O_2/l)	Influent	56 (26-70)	60 (28-102)	57 (41-67)	60 (33-77)	101 (48-204)	51 (30-71)
	Effluent	36 (17-60)	43 (23-69)	35 (10-60)	45 (25-70)	48 (31-78)	33 (8-55)
	Reduction %	32.0 (7.7-62.2)	27.9 (10.7-50.0)	38.5 (9.1-75.6)	25.7 (7.9-41.9)	47.3 (35.4-61.8)	36.3 (2.9-73.3)
Suspended solids (mg/l)	Influent	6.0 (4.0-9.0)	8.5 (6.5-13.0)	5.4 (3.0-8.0)	12.1 (5.5-22.0)	50.9 (2.0-138.5)	8.8 (2.5-19.0)
	Effluent	12.4 (6.5-19.5)	9.0 (7.5-10.5)	5.1 (3.5-7.0)	7.5 (3.5-15.5)	8.1 (5.0-17.0)	10.9 (2.5-18.0)
Turbidity (NTU)	Influent	2.5 (1.7-3.5)	4.5 (2.8-6.0)	2.8 (2.2-3.6)	2.9 (1.5-5.5)	8.7 (1.0-23.0)	4.1 (1.5-5.2)
	Effluent	2.2 (1.1-3.5)	2.3 (1.7-3.0)	1.2 (0.8-1.7)	2.0 (1.2-3.3)	2.9 (1.5-6.0)	5.6 (3.6-7.0)
Transmittance, 253.7 nm (%)	Influent	45.9 (40.3-50.2)	43.7 (39.3-49.1)	48.5 (42.4-68.9)	44.3 (29.8-56.2)	26.1 (5.3-47.4)	53.7 (48.6-50.2)
	Effluent	53.5 (47.0-62.5)	53.6 (50.8-56.4)	63.1 (55.0-67.0)	55.7 (43.7-62.2)	58.2 (51.3-65.5)	53.4 (48.0-59.4)
Temperature (°C)	Influent	11-14	17-19	17-19	11-13	11-12	9-13

[a] Coagulant PAX-XL60, 7 mg^{Al3+}/l; [b] Coagulant PAX-XL60, 3 mg^{Al3+}/l; [c] Coagulant PAX-XL60, 6 mg^{Al3+}/l; [d] Polymer (Praestol), 3.0-3.75 mg/l + non-continuous PAX-dosing (<6 NTU: 0 mg/l PAX-14; >6<50 NTU: 100 mg/l PAX-14 (7 mg^{Al3+}/l); >50 NTU: 220 mg/l PAX-14 (16 mg^{Al3+}/l)); [e] Polymer (Praestol), 1.9-3.0 mg/l + continuous PAX-dosing (<100 NTU: 50 mg/l PAX-14 (4 mg^{Al3+}/l); >100 NTU: 220 mg/l PAX-14 (16 mg^{Al3+}/l)); [f] polymer Fennopol K504, 0.4-0.71 mg/l + coagulant ALF-30 52-94 mg/l (4-7 mg^{Al3+}/l and 1.5-3 $mgFe^{3+}$/l)

5.5 Wastewater disinfection

5.5.1 Laboratory-scale disinfection experiments

The effect of different disinfection treatments on enteric microorganisms in a PW medium was studied. Approximately 2-3 log (99-99.9 %) reductions of *E. coli*, *S. enteritidis* and *E. faecalis* were achieved by using 3 mg/l PAA dose and 10 min contact time, whereas 7-15 mg/l PAA was needed to achieve approximately 1-1.5 log (90-97 %) coliphage MS2 reductions (II, Table 2). Disinfection treatments with 3-150 mg/l H_2O_2 achieved below 0.2 log (37 %) microbial reductions. Sodium hypochlorite disinfection with 18 mg/l chlorine dose caused below 1 log reductions of *E. coli*, *S. enteritidis* and coliphage MS2, whereas 2.6 log reductions of *E. faecalis* were achieved with 12 mg/l chlorine dose. UV doses of 6-18 mWs/cm^2 achieved 1-3 log enteric bacterial reductions with UV doses of 22-38 mWs/cm^2 exhibiting 1-1.5 log reductions of coliphage MS2.

Chemical and UV disinfection treatments were combined to investigate if this provides synergy benefits. Synergy means that the efficiency of combined disinfection method is higher than the efficiency achieved when summing all the single effects. Combined PAA/UV disinfection treatments improved enteric bacterial reductions and achieved synergy benefits, the highest synergy values (see definition in chapter 4.7) exceeded 2 log units, when 1.5-3 mg/l PAA in combination with UV irradiation were used (II, Tables 2 and 3). The increase of UV or PAA dose typically increased the disinfection efficiency and synergy values. The synergies in combined PAA/UV treatments were statistically significant for *E. faecalis* and *E. coli*, but not for *S. enteritidis* and coliphage MS2 (II, Table 3). H_2O_2/UV treatments showed small but statistically significant antagonistic effects with *E. faecalis* as the test microorganism, and statistically non-significant synergies with coliphage MS2 as the test microorganism.

5.5.2 Pilot-scale disinfection experiments

5.5.2.1 Disinfection of secondary and tertiary effluents

Disinfection of secondary effluents of Kuopio WWTP and tertiary effluents from the DAF pilot-plant with 2-7 mg/l PAA doses and 27 min contact time achieved around 3 log reductions of total coliforms and enterococci, and below 1 log reductions of F-RNA coliphages (III, Tables 2-4). The numbers of TC in disinfected secondary and tertiary effluents ranged between 8.2×10^1-2.9×10^3 CFU/100ml and <10-3.4×10^2 CFU/100ml, respectively. The corresponding enterococcal numbers were <10-1.1×10^3 CFU/100ml and <10-1.3×10^2 CFU/100ml, and those of F-RNA coliphages 1.0×10^2-1.5×10^4 PFU/100ml and <1.0×10^2-6.8×10^3 PFU/100ml, respectively.

With 5-7 mg/l PAA, most of the TC and EC reductions were achieved during the first 4 and 13 min of contact time, after which microbial numbers were low and further microbial inactivation took longer (Figure 8). With PAA doses of 2-3 mg/l, most of the TC reductions occurred during the first 13 minutes of contact time, while the reductions of EC numbers were slower and approximately followed first order kinetics during the contact time of 27 min. With 2-7 mg/l PAA doses, the F-RNA coliphage inactivation was slow and their reductions were lower than those of TC and EC.

The physico-chemical quality of secondary and tertiary wastewater effluents correlated poorly with the microbial reductions in different disinfection treatments (linear regression analysis, R^2 values typically <0.3). PAA residues were typically 1-2 mg/l lower than the applied PAA dose.

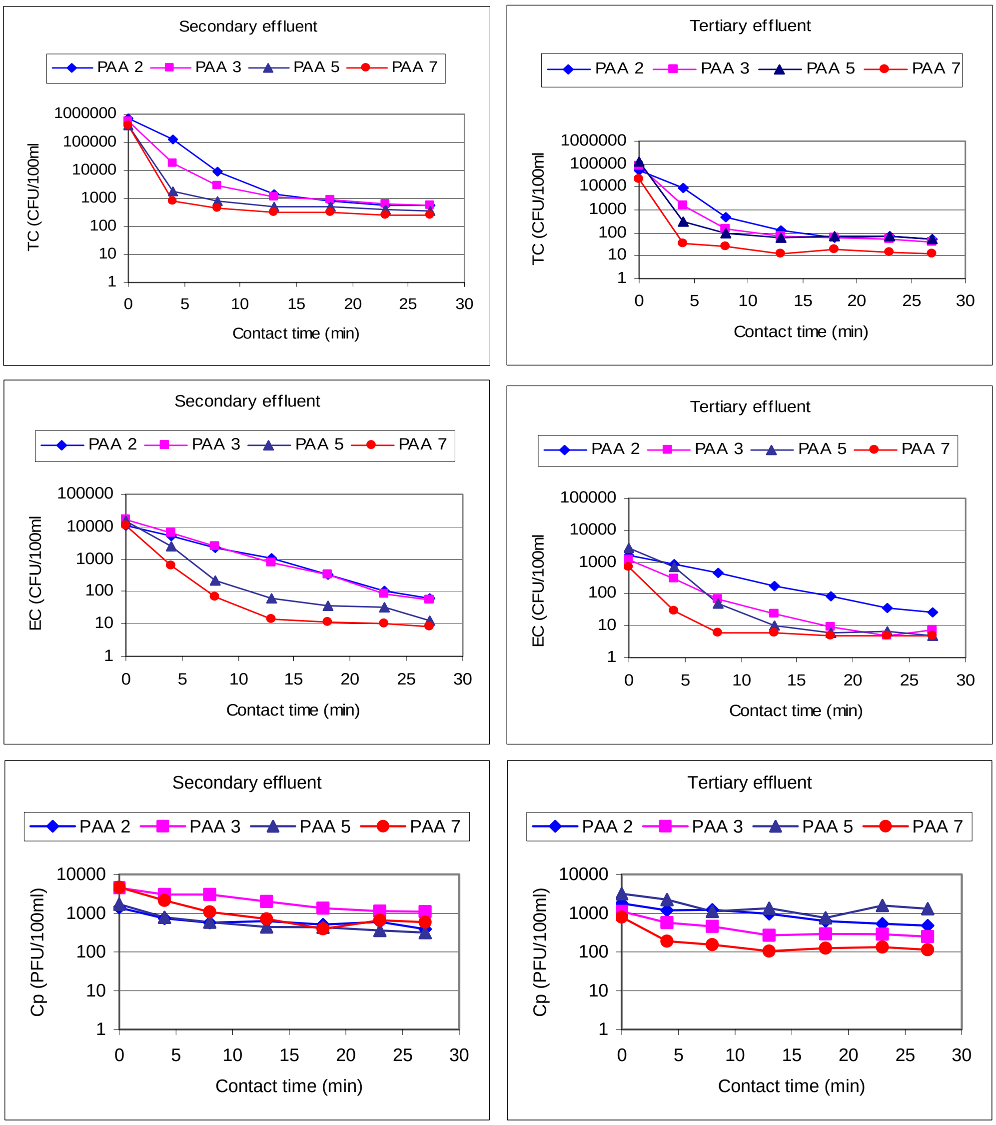

Figure 8. The numbers (geometric means) of total coliforms (TC), enterococci (EC) and F-RNA coliphages (Cp) as a function of contact time in the PAA disinfection treatments of secondary and tertiary (DAF) effluents. Results are means of 4-9 experiments.

5.5.2.2 Disinfection of primary effluents

Disinfection of WWTP primary effluents with 10-15 mg/l PAA doses and 27 min contact time achieved approximately 3-4 log (99.9-99.99 %) reductions of TC and EC, and below 1 log (90 %) reductions of F-RNA coliphages (Figure 9; III, Tables 2-4). With PAA dose of 5 mg/l, average microbial reductions were 1 log unit or below. The best TC and EC reductions were achieved during the first 8 and 18 min of contact time, respectively. The inactivation of F-RNA coliphages was slower than those of TC and EC.

The physico-chemical wastewater quality had an effect on disinfection efficiency in the primary effluents. The disinfection treatments with 10-15 mg/l PAA resulted in 0-2.5 mg/l PAA residues, while disinfection with 5 mg/l PAA did not produce any detectable PAA residues in the effluents.

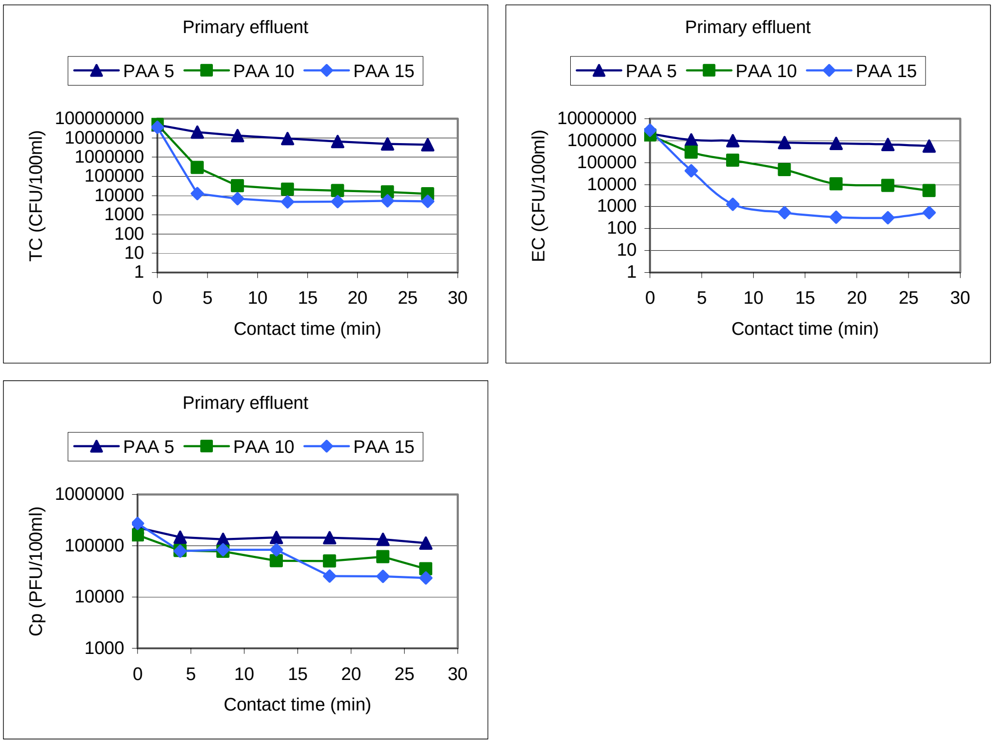

Figure 9. The numbers (geometric means) of total coliforms (TC), enterococci (EC) and F-RNA coliphages (Cp) as a function of contact time in the PAA disinfection treatments of primary effluents. Results are means of 3 experiments.

6 DISCUSSION

The aim of this study was to evaluate the efficiency of different wastewater treatment processes on removal of enteric microorganisms, phosphorus and organic matter from municipal wastewaters. Conventional biological-chemical wastewater treatment typically achieved 90->99 % reductions of enteric microorganisms and around 95 % reductions of organic matter and phosphorus. Even though there seems to be high percent reductions, secondary effluents always contained substantial numbers of enteric microorganisms. Tertiary rapid sand filtration (RSF) and dissolved air flotation (DAF) processes efficiently removed residual organic matter and phosphorus, and removed 90-99 % of enteric microorganisms from the secondary effluents. The DAF process also showed good performance in the treatment of primary wastewater effluents, demonstrating that the DAF process can tolerate high loads of suspended solids and could be used for treatment of WWTP by-pass wastewaters during the treatment plant over-loading situations. Peracetic acid (PAA) proved to be an efficient disinfectant against enteric microorganisms in municipal primary, secondary and tertiary wastewater effluents. The combined PAA/UV treatments showed high disinfection efficiency and synergistic benefits in the laboratory-scale disinfection experiments.

6.1 Conventional wastewater treatment

Municipal biological-chemical wastewater treatment plants efficiently removed organic load (BOD_{7atu}, COD_{Cr}), suspended solids and phosphorus from wastewaters, their typical reductions after the primary and secondary treatment phases being 30-80 % and 90-99 %, respectively. Total phosphorus concentrations of secondary effluents typically ranged between 0.2-0.7 mg/l, but during short periods of time, total phosphorus concentrations even higher than 1 mg/l were measured.

The wastewater enteric microbial numbers (salmonellae, faecal coliforms and total coliforms) were typically reduced by 46-99 % and 94->99 % in the primary and secondary wastewater treatment processes, respectively. The efficiency of primary treatment on microbial removal displayed quite high variability, and might depend on the extent to which the microbes were associated with the solid matter in wastewater and on the efficiency of the precipitation-sedimentation process (Tanji et al., 2002). The microbial reductions in full-scale WWTPs with simultaneous phosphorus precipitation processes were at the same level as those achieved in the technical-scale biological nutrient removal process, indicating that those two processes exhibit no significant differences in their efficiencies at removing enteric microorganisms. Some earlier studies in activated sludge wastewater treatment plants have shown approximately 90-99.9 % enteric bacterial reductions, i.e. similar to the values achieved in the present study (Yaziz and Lloyd, 1979; Teitge et al., 1986; Kayser et al., 1987; Nieuwstad et al. 1988; Zaiss and Hennies, 1988; Jacangelo et al., 2003).

Even though there were high percent reductions of enteric microorganisms, some pathogenic salmonellae and high numbers of faecal indicator bacteria were found to survive the conventional wastewater treatment processes and were discharged into receiving natural waters. The numbers of total coliforms (TC), enterococci (EC) and F-RNA coliphages (Cp) in secondary effluents typically ranged by 2-3 orders of magnitude, and contained microbial numbers around 10^4-10^7 CFU/100ml TC, 10^3-10^6 CFU/100ml EC and 10^3-10^5 PFU/100ml Cp. The highest detected number of culturable salmonellae in effluent waters was 240 MPN/100 ml. The microbial numbers in the secondary effluents were significantly higher than the limit values for wastewater reuse in many countries (e.g. Italian limit-value of 2 CFU TC/100 ml and 20 CFU TC/100 ml for unrestricted and restricted wastewater reuse in agriculture, respectively; The State of Florida, USA, limit value for wastewater reuse in restricted public access areas and industrial uses 200 CFU FC/100 ml and limit value of no detectable FC/100 ml for the irrigation of parks, golf courses and food crops) (Crook and Surampalli, 1996; Barbagallo et al., 2001; Lubello et al., 2002). The microbial numbers of WWTP secondary effluents were also higher than the regulatory limit values for wastewater discharge into surface waters in Italy (5 000 MPN/100 ml *E. coli*, 20 000 MPN /100 ml TC, 12 000 MPN/100 ml FC and 2 000 MPN/100 ml faecal streptococci) or limit values for bathing waters in the European Union region (Collivignarelli et al., 2000; Stampi et al., 2001; EC, 2006).

The elimination of enteric microorganisms, as well as the elimination of other pollutants in conventional wastewater treatment based on activated sludge, is clearly dependent on the efficiency of the final clarification process (Teitge et al., 1986; Kayser et al., 1987). In the present study, some correlations of microbial numbers with the residual BOD, COD, suspended solids and total phosphorus concentrations in secondary effluents were observed. In previous studies, enteric microorganisms have been reported to concentrate to the sludge flocs in the activated sludge process (Drift et al., 1977; Yaziz and Lloyd, 1979, 1982; Nakajima et al., 2003). Adsorption of enteric microbes onto sludge flocs is through extra-cellular polymeric substances that form “bridges” between the surfaces of microorganisms and the sludge flocs (Tawfik et al., 2004). High numbers of enteric microbes are also in the free-swimming -state in the activated sludge (Yaziz and Lloyd, 1982) and cannot be removed from the wastewater during a final clarification stage. Microorganisms may also be detached from the sludge in the final clarifier.

6.2 Tertiary wastewater treatment

The present study showed that tertiary RSF and DAF processes, following conventional wastewater treatment, can significantly improve wastewater treatment efficiency and achieve decreasing of pollution loads into receiving natural waters.

6.2.1 Rapid sand filtration (RSF)

The tertiary rapid sand filtration when utilized as contact filtration efficiently removed faecal coliforms and Salmonellae from wastewaters, with typical reductions being 90-99 %. The reductions of F-RNA coliphage viruses were typically slightly lower than those seen with the faecal indicator bacteria (Rajala et at. 2003). Despite of high percent reductions, tertiary RSF effluents still contained relatively high enteric microbial numbers. The lower reductions of coliphage viruses were probably due to their smaller size and different surface characteristics when compared to those of enteric bacteria. The size of the bacterial cells and virus particles is typically in the size range of 1-10 µm and 0.01-0.1 µm, respectively, but a considerable proportion of microorganisms in wastewater is, however, adsorbed to particles which means that they have a larger size range (Ødegaard, 2001). Particles of 1-2 µm size and smaller have minimal opportunity for removal in the filter unit, since transport mechanisms for these particles within the filter bed are less efficient (Adin, 1999). The microbial reductions achieved in the present study are comparable to those reported in some other tertiary filtration experiments (Nieuwstad et al., 1988; Jolis et al., 1996; Heinonen-Tanski et al., 2002; Scott et al., 2002).

The tertiary RSF treatment efficiently reduced phosphorus, turbidity, SS and COD, as well as improved the UV transmittance of wastewater effluents (Rajala et al. 2003). Similar improvements in the physico-chemical wastewater quality in tertiary filtration processes have been previously reported (Nieuwstad et al. 1988; Jolis et al., 1996; Kuo et al., 1997; Hamoda et al., 2002; Heinonen-Tanski et al., 2002). Comparable results have also been achieved by alternative final clarification processes, post-precipitation and tertiary DAF (Teitge et al. 1986; Nieuwstad et al. 1988; Ødegaard, 2001; Pinto Filho and Brandão, 2001).

If there was no addition of coagulant, then the process efficiency decreased, this finding being in agreement with the literature data (Diamadopoulos and Vlachos, 1996; Cikurel et al., 1999; Suwa and Suzuki, 2003). This may be explained by the particle size remaining too small without chemical modification, and the small particles could penetrate through the filter. The filtration process efficiency typically decreases with decreasing particle size (Kuo et al., 1997; Adin, 1999; Graaf et al., 2001). Removal of contaminants during contact filtration is based on the processes of flocculation, straining and adsorption of pollutants onto the surfaces of floc and sand particles, i.e. processes which take place inside the sand filter bed. Coagulant chemical addition improves the separation of soluble compounds, such as phosphorus, through formation of precipitates that can be separated in the filter unit. The coagulation process also reduces the numbers of dispersed microorganisms in the wastewater through adsorption of microbial cells to the flocs, followed by floc separation in the filter bed.

6.2.2 Chemical contact filtration and biological-chemical contact filtration

The chemical contact filter reduced microbial numbers slightly better than the biological-chemical contact filter. That could be related to the smaller grain size and the lower hydraulic loading rate of the chemical contact filter. The decrease of grain size and increase of specific surface area of the filter medium, as well as the decrease of hydraulic loading rate, have been reported to reduce the transport of enteric microorganisms through the filter unit (Stevik et al., 1999; Logan et al., 2001; Manios et al., 2002). The microbial reductions in the chemical and biological-chemical contact filters were lower than those achieved in the RSF process. That may be attributable to the smaller sand particle size, efficient coagulation-flocculation and continuous backwash in the RSF unit. The chemical contact filter and biological-chemical contact filter also improved the physico-chemical quality of wastewater effluents (Rantanen, 2001).

6.2.3 Dissolved air flotation (DAF)

The present study showed that the tertiary DAF process improves the hygienic quality of wastewater effluent, reducing microbial loads into the recipient water body. The typical reductions of enteric microbial numbers in the process were 90-99 %. The reductions of F-RNA coliphage viruses were typically slightly lower than those of total coliforms and enterococci, probably due to the smaller size and different surface characteristics of viruses when compared to those of bacteria, affecting their attachment onto chemical flocs and subsequent removal. Similar reduction patterns of enteric bacteria and viruses were observed also in the tertiary RSF unit, as observed in the present study and also reported by Rajala et al. (2003). The less effective removal of the coliphages in the treatment process agrees with the literature data (Nieuwstad et al., 1988).

Despite the high percent reductions, tertiary DAF effluents still contained relatively high enteric microbial numbers. No previous literature data exist on enteric microbial reductions in the tertiary DAF process. Alternative tertiary treatment methods, RSF as contact filtration and precipitation-sedimentation processes, have been reported to achieve comparable 90-99 % enteric microbial reductions in municipal wastewater treatment (Nieuwstad et al., 1988; Rajala et al., 2003). In drinking water treatment, the DAF process has been reported to achieve 1.7 and 2.5 log (Edzwald et al., 2001) average reductions of *Cryptosporidium* during the winter (2-3ºC) and spring (13-14ºC) time, respectively. A combination of DAF and filtration processes has achieved 3-4 log (Hall et al., 1995) and more than 5.4 log (Edzwald et al., 2001) reductions of *Cryptosporidium*. Cryptosporidia are much larger size microorganisms (size range 2-60 µm) than are enteric bacteria and viruses (size range 1-10 µm and 0.01-0.1 µm, respectively), which may account for the better removal efficiency of Cryptosporidia in the treatment processes.

The tertiary DAF treatment efficiently removed phosphorus from wastewaters, the typical total phosphorus reductions being 50-80 % and the residual concentrations around 0.10 mg P_{tot}/l. The DAF process also reduced COD and turbidity, as well as improved the UV transmittance of wastewater effluents. The SS concentration of the DAF effluent was typically around 5-10 mg/l, which was slightly higher that that typically achieved in the tertiary RSF process. The higher SS concentration was probably due to escape of small amounts of floc from the tertiary DAF process, the phenomenon observed in both the pilot-scale and full-scale DAF processes. Comparative purification efficiencies for phosphorus, organic matter and suspended solids have been previously achieved when using tertiary DAF (Penetra et al., 1999; Ødegaard, 2001; Pinto Filho and Brandão, 2001; Reali et al., 2001), tertiary RSF (Jolis et al., 1996; Kuo et al., 1997; Rajala et al., 2003) or tertiary precipitation-sedimentation (Nieuwstad et al., 1988) processes.

The full-scale tertiary DAF processes in municipal WWTPs of Heinävesi and Pieksämäki achieved comparable purification efficiencies than was achieved in the present study by using pilot-scale DAF process. Although there were significant differences in operating the processes (differences in hydraulic loading, coagulants, flocculation conditions, recycle ratio etc.), the reductions of enteric microorganisms, phosphorus and organic matter were comparable. The hydraulic loadings of both the investigated full-scale DAF processes (0.5-2.5 m/h) were much lower than those used in the pilot-scale experiments and those normally used in full-scale DAF processes (5-10 m/h). The results of the present study show that low hydraulic loading does not necessarily improve the DAF process effluent quality. Over-dimensioning of the process, however, increases the land area needed and the construction and operational costs. DAF process typically achieves steady purification results in variable process loading conditions, as discussed later. It is suggested that the investigated full-scale DAF processes could be operated by 5-10 m/h hydraulic loads without significant reduction of treatment efficiency.

The increase of the coagulant dose from 2 to 10 gAl^{3+}/m^3 improved the DAF process performance, which is in agreement with the results of the previous studies (Ødegaard, 1995; Penetra et al., 1999; Pinto Filho and Brandão, 2001). An increase in the coagulant dose may improve the efficiency of coagulation-flocculation, favouring efficient particle-bubble aggregate formation and separation in the DAF basin. At low coagulant doses, small flocs with poorer separation characteristics may be formed (Adin, 1999). The use of cationic polymer, in addition coagulant chemical, slightly improved the reductions of enteric microorganisms, total phosphorus and other water quality parameters. That was probably due to the more efficient flocculation and capture of small particles, as well as to the formation of stronger flocs that could resist fracture, as also reported by Reali et al. (2001b). The polymer may also improve particle-bubble aggregate formation through increasing the particle hydrophobicity and through direct attachment of the polymer chain to the microbubbles.

The DAF process performance remained relatively constant when the G-values of flocculation were changed from the slow mixing (G-values 10 s^{-1} in both the 1st and 2nd flocculation chambers) to a process with higher mixing intensities (G-values 55 s^{-1} and 26 s^{-1}) or when the flocculation time was decreased from 8 to 4 minutes with increasing hydraulic load. The results of the present study are in agreement with the literature data (Bunker et al., 1995; Amato et al., 2001; Ødegaard, 1995, 2001; Pinto Filho and Brandão, 2001; Reali et al., 2001). Some previous studies have shown, however, that slightly higher G-values of flocculation (60-80 s^{-1}) and flocculation retention times (15-30 min) are favourable prior to the DAF process (Ødegaard, 1995; Reali et al., 2001). The improvement of process performance with increasing flocculation time may be because there are more opportunities for particle collisions and floc formation (Ødegaard, 1995; Pinto Filho and Brandão, 2001). Ødegaard (1995) also reported that the optimal flocculation unit should be divided into at least two compartments, which is in agreement with the results of the present study (results not shown). The increase of the stages of flocculation causes the process to approach plug-flow conditions, thus achieving a more constant flocculation retention time and decreasing the possibility for short circuiting. The optimal flocculation conditions are, however, always process-specific and affected by many factors, e.g. by the flocculation unit configuration, coagulant type, water quality and temperature.

The appropriate conditions of flocculation seems to be wider in the DAF systems than in the flocculation-sedimentation systems, where higher coagulant doses, lower mixing intensities (G-values 20-40 s^{-1}) and longer retention times (>30 min) are typically used to generate large (>100 µm) and heavy floc particles with adequate settling properties (Edzwald, 1995; Ødegaard, 1995). Smaller size and strong structure of flocs, produced by the higher intensities of flocculation and shorter flocculation times, are typically favourable in the flocculation-DAF process. There is, however, a critical minimum floc size, which may be about 3-10 µm, that must be reached to achieve adequate bubble/particle number ratio and efficient clarification by DAF (Haarhoff and Edzwald, 2001). In practise, there is a wide variation on the applied flocculation intensities (20-100 s^{-1}) and flocculation times (4-130 min) in different DAF applications (Edzwald et al., 1995; Amato et al., 2001). Generally, the use of shorter flocculation times permits the use of smaller flocculation basins, thus resulting in savings in the land area needed for the basin and its construction costs.

The increase of the recycle ratio from 11 to 22 % slightly improved the purification results in the DAF process. The optimum recycle ratio value was assessed to be 15-20 %, when the operation pressure of the dispersion water system was 5-6 bars. Increasing the recycle ratio above 20 % did not significantly improve the process performance, while at lower recycle ratios, the process efficiency exhibited a slight decline. With lower recycle ratios, the sensitivity of process for increased loading of suspended solids was also observed to increase. The decrease of process performance with decreasing recycle ratio is probably due to a decrease in the numbers of microbubbles, which may lead to insufficient bubble-particle interactions, the formation of a coarse bubble-bed in the DAF tank and penetration of particulate matter through the process. A decrease of recycle rate may also affect the flow patterns in the DAF tank and increase the possibility of break-through flow development (Lundh et al., 2000, 2001), increasing penetration of suspended solids through the process. The findings in the present study are in agreement with the literature data (Ødegaard, 1995; Pinto Filho and Brandão, 2001). The DAF processes are typically operated at a constant dispersion water flow rate, chosen to cope with different process loads. That was the case also in the Heinävesi and Pieksämäki WWTPs, where the full-scale DAF experiments were carried out in the present study. A higher recycle ratio may increase the capacity of the process during high loading conditions. The adjustment of dispersion water flow proportionally to wastewater flow rate and quality would allow for better optimizing the process operation and reduce operating costs (costs of producing dispersion water). In practise, there is a wide variation in the applied recycle ratios (5-40 %) in the different DAF applications (Edzwald et al., 1995; Amato et al., 2001).

The performance of the pilot-scale DAF process remained constant when the hydraulic loading of process increased from 5 to 10 m/h, which is in agreement with the results of previous studies (Ødegaard, 1991, 1995; Pinto Filho and Brandão, 2001). The results indicate that DAF is a very area efficient process, and it can cope with significant variations in flow rate without suffering any drastic reduction in effluent quality. The DAF process used in this study can be classified as conventional DAF that typically operates efficiently at overflow rates as high as 10-15 m/h. Currently there exist a newer DAF technology that can be successfully operated with hydraulic surface loads as high as 25-40 m/h (Amato et al., 2001; Kiuru, 2001). By comparison, RSF and precipitation-sedimentation processes are typically more sensitive than DAF to changes of process loading and can only cope with overflow rates of less than 10 m/h and 1-2 m/h, respectively.

The results of the tertiary DAF experiments indicate that DAF process can be used to decrease the loads of organic matter and phosphorus into receiving natural waters. It could be used as an efficient preliminary treatment process preceding the final disinfection stage, improving the disinfection process efficiency and reducing the potential for DBP formation.

6.3 Dissolved air flotation (DAF) treatment of primary wastewater effluents

At times, wastewater treatment plants face overloading situations, e.g. increased hydraulic loading during storm events. During these events, WWTPs may be forced to by-pass untreated or only primary treated wastewaters into receiving natural waters. This means that significant loads of organic matter, nutrients and enteric microorganisms can pass into receiving natural waters, as shown in the present study and reported by Rechenburg et al. (2006). Increased hydraulic loading of WWTP, changes of wastewater quality, etc. may also cause disturbances in the treatment processes, impairing wastewater treatment efficiency. There can be many consequences to increased wastewater loads into the natural waters, e.g. restrictions on use of water for swimming or other recreational purposes, increased eutrophication and disturbances to fish populations. Treatment of by-pass wastewaters and elimination of peak pollution loads would significantly decrease these pollution loads and reduce the risks they pose to the recipient water bodies (Hanner et al., 2004).

Any evaluation of the treatment process for treatment of WWTP overflows must be based on several criteria (Hanner et al., 2004): a) sufficient removal of the desired substances must be achieved, b) the process should be easy to start at short notice, and c) the process should also fit into existing structures considering land area, weight and transport of water, chemicals and sludge. Since the process may only be operated during the high flow situations, higher operational costs may be acceptable.

In the present study, the DAF treatment efficiently removed enteric microorganisms, phosphorus and organic matter from primary treated wastewaters, when operated with a coagulant dose of 12 g Al^{3+}/m^3 and 5 m/h hydraulic surface load. The DAF treatment achieved 94-99.98 % reductions of enteric microbial numbers. The microbial reduction percentages were higher than those achieved in the tertiary treatment of wastewaters, probably due to the high numbers of microorganisms present in the suspended and colloidal matter in the primary effluents and their efficient removal in the DAF process. The average reductions of total phosphorus, COD_{Cr} and suspended solids were 90 %, 47 % and 77 %, respectively, demonstrating significant reductions of organic load and nutrients. Comparable improvements of physico-chemical effluent quality have been previously reported in the full-scale and pilot-scale DAF treatment of raw and primary treated wastewaters (Ødegaard 1995, 2001; Mels et al., 2001), and in laboratory-scale DAF treatment of other low quality effluents (Reali et al., 2001a, 2001b; Pinto Filho and Brandão, 2001). Some Finnish municipal WWTPs have practised DAF treatment of by-pass wastewaters, achieving around 90 % reductions of total phosphorus, BOD and SS. Hanner et al. (2004) has reported around 90 % reductions of P_{tot} and SS when treating screened raw wastewater by direct precipitation and settling (3.75 m/h). They also reported 50-60 % reductions of BOD, 70-90 % reductions of SS and over 85 % reductions of P_{tot} by using chemical/mechanical treatment with direct precipitation and lamella separation. Suwa and Suzuki (2003) have previously reported 1-3 log reductions of *Cryptosporidium* oocysts in chemical precipitation of raw wastewater.

The DAF process achieved significant improvement of wastewater quality in treatment of primary wastewater effluents. The results demonstrate that the DAF process can tolerate high loads of suspended solids and could be used for treatment of by-pass wastewaters during the WWTP over-loading situations. In this study, the quality of primary wastewater effluent entering the DAF process was slightly different than the typical by-pass water: by-pass wastewater typically is more diluted due to diluting effect of high storm water flow. It is suggested, that the efficiency of DAF process for treatment of more diluted by-pass wastewaters could be even better than the efficiency that was achieved in the present pilot-scale study when treating primary wastewater effluents.

Treatment of by-pass wastewaters significantly improves the hygienic quality of effluents and reduces the effect of those wastewater discharges on receiving water body. Treatment of by-pass wastewaters could also help in the achieving of target purification levels of phosphorus and other pollutants in municipal WWTPs. Although the WWTP overloading situations may constitute only a small portion of the flow on an annual basis, wastewater by-passes typically correspond to a few percent, but occasionally an even larger proportion, of the annual loads of phosphorus and BOD into the environment, and they can result in exceeding the regulatory limit values for wastewater discharges (Hanner et al., 2004).

The treatment of by-pass wastewaters by the DAF process poses some specific demands for the design of the process, with respect to dispersion water system and the removal of bottom sludge. The pressure vessel and dispersion water injection nozzles must be specially designed to avoid clogging of the system and problems in the process operation and maintenance. Efficient removal of the accumulating bottom sludge must be arranged to avoid maintenance problems and deterioration of process performance.

6.4 Wastewater disinfection

After secondary or tertiary wastewater treatment, the removal of enteric microorganisms from the wastewater effluents can be further improved by subjecting the water to a separate disinfection process. The present study showed that PAA disinfection can significantly decrease enteric microbial numbers in the primary, secondary or tertiary wastewater effluents, thus decreasing microbial loads into receiving natural waters. The results also suggest that a combined PAA/UV disinfection treatment may achieve synergistic benefits and improved disinfection efficiency.

6.4.1 Peracetic acid (PAA)

In laboratory-scale disinfection experiments with a synthetic test medium, peracetic acid was demonstrated to be an efficient disinfectant against enteric bacteria. PAA doses of 1.5-3 mg/l and a 10 min contact time resulted in approximately 2-3 log (99-99.9 %) *Escherichia coli*, *Enterococcus faecalis* and *Salmonella enteritidis* reductions. In pilot-scale disinfection of municipal secondary and tertiary effluents, PAA showed high disinfection efficiency against total coliforms and enterococci. PAA doses of 2-7 mg/l and 27 min contact time achieved around 3 log reductions of these indicator bacteria, and their numbers were typically reduced below 500 CFU/100 ml TC and 100 CFU/100 ml EC, in many cases to <10-100 CFU/100 ml. Approximately similar PAA disinfection efficiencies against enteric bacteria have been previously reported (Collivignarelli et al., 2000; Stampi et al., 2001; Wagner et al., 2002; Caretti and Lubello, 2003), but in some studies slightly higher PAA doses (around 10 mg/l) have been used to achieve similar inactivation of enteric bacteria (Lazarova et al., 1998; Liberti and Notarnicola, 1999). The needs for higher PAA doses could be explained by the differences in water quality, reactor design and operational parameters in the different experiments.

PAA disinfection efficiently reduced microbial numbers also in the primary effluents, 10-15 mg/l PAA doses achieving approximately 3-4 log enteric bacterial reductions. Sánchez-Ruiz et al. (1995) have previously reported 2.0-6.5 log TC reductions in the disinfection of raw sewage by using 10-20 mg/l PAA and 15 min contact time.

In pilot-scale disinfection of municipal secondary and tertiary effluents, F-RNA coliphages were more resistant than the enteric bacteria against PAA disinfection: around 1 log coliphage reductions were achieved with 5-7 mg/l PAA in the disinfection of secondary and tertiary effluents, and with 10-15 mg/l PAA in the disinfection of primary effluents. In laboratory-scale disinfection experiments, 7-15 mg/l PAA dose was needed to achieve 1-1.5 log reductions of coliphage MS2. The elevated resistance of viruses against disinfection has been demonstrated also in previous studies (Lefevre et al., 1992; Salgot et al., 2002; Veschetti et al., 2003; Nasser et al. 2006). The results of the present study highlight the variability of microbial resistance against the disinfection treatments, emphasizing the importance of assessing disinfection efficiency by using more than one indicator organism, as proposed also by Gehr et al. (2003) and Lucena et al. (2004).

The disinfection action of PAA is thought to be due to release of active oxygen or the production of reactive hydroxyl radicals that attack the bacterial cell, causing disruption of the cell wall and membranes as well as damaging some enzymes and nucleic acids (Liberti and Notarnicola, 1999; Lefevre et al., 1992; Lubello et al., 2002). Inactivation of coliphages may be related to damage to their surface structures, the protein coat or the attachment sites needed for infection of host cells, or to effects on their nucleic acids. Viruses are typically more resistant than enteric bacteria to other disinfection methods, such as UV, chlorine and ozone disinfection (Tyrrell et al., 1995; Rajala et al., 2003; Nasser et al. 2006).

PAA disinfection of the primary, secondary and tertiary effluents typically achieved fast microbial inactivation. Most of the TC and EC reductions were achieved during the first 4-18 min of contact time, the increase of PAA dose resulting in higher microbial inactivation rates. Also in the previous studies, the disinfection action of PAA has been reported to be rapid, with the most significant microbial inactivation occurring during the first 5-10 min of treatment (Sanchez-Ruiz et al., 1995; Rajala-Mustonen et al., 1997; Liberti and Notarnicola, 1999; Liberti et al., 2000; Gehr et al., 2003).

The inactivation of TC occurred in two phases, first there was a steep decline in TC numbers at the beginning of process followed by a phase of decreasing inactivation rate (tailing of the inactivation curve). This tailing phenomenon is typical of many disinfection methods (Tyrrell et al., 1995; Liberti et al., 2000; Nasser et al. 2006), and is characterized by the slowing of microbial inactivation with increasing disinfectant dose or contact time, i.e. the last viable microorganisms being more difficult to destroy. The phenomenon can be explained by the fact that some microorganisms are attached to suspended solids or are present in microbial aggregates which need a higher dose of the disinfectant or a longer contact time to achieve penetration of the disinfectant into particles and subsequent microbial inactivation. The free-swimming portion of the microorganisms is more easily inactivated than the microorganisms inside the particles or microbial aggregates. The physiological state of the microorganisms affect their inactivation, e.g. through changes in the cell composition.

The inactivation of EC with higher PAA doses (5-7 mg/l PAA in the secondary and tertiary effluents; 15 mg/l PAA in the primary effluent) occurred in two phases, similarly to that of TC, while with lower PAA doses (2-3 mg/l PAA in the secondary and tertiary effluents; 5-10 mg/l PAA in the primary effluent) microbial inactivation seemed to approximately follow first-order kinetics. The inactivation of coliphages was always slower than that of TC and EC.

The optimum contact time for PAA disinfection was assessed to be between 10 and 15 minutes, but a further increase of contact time may still increase microbial inactivation, if a sufficient PAA dose is applied to produce PAA residues. The PAA dose was found to be an important factor affecting microbial elimination with short contact times, since increase of disinfectant dose increases the inactivation rate. The PAA dose was an important factor also in disinfection of primary effluents, as higher disinfectant doses are needed in organic matter rich wastewaters due to higher disinfectant demand of the wastewater matrix. Wagner et al. (2002) and Azzellino et al. (2002) have reported that PAA dose is a more important factor than contact time in determining disinfection efficiency. In the present study, the effect of contact time was found to be significant at the lower PAA doses, which is in agreement with the results reported by Mezzanotte et al. (2002). With low PAA doses, longer contact times will be needed due to slower microbial inactivation rate. Sufficient PAA dose is needed, however, to produce sufficient PAA residues.

The physico-chemical quality of secondary and tertiary effluents correlated poorly with the microbial reductions in the disinfection treatments. The lack of correlations may be explained by the low and relatively constant concentrations of SS, COD and other water quality parameters in the secondary and tertiary effluents throughout the disinfection experiments. Disinfected secondary effluents, however, typically showed slightly higher residual microbial numbers than the disinfected tertiary effluents, which may be due to higher initial microbial numbers and the organic matter concentrations in the former wastewaters. Similar observations have been reported in previous studies, with small changes of effluent quality having no significant effects on disinfection efficiency (Baldry et al., 1995; Stampi et al., 2001; Nasser et al., 2006). Efficient disinfection of primary effluents required considerably higher PAA doses than the secondary and tertiary effluents. That was due to the high microbial numbers and higher COD and suspended solids concentrations in the primary effluents, providing protection for microorganisms within large particles and increasing the consumption of the PAA disinfectant in reactions with the wastewater matrix. Our results are in agreement with the results of previous studies (Baldry et al., 1991, 1995; Sánchez-Ruiz et al., 1995; Stampi et al., 2001, 2002; Gehr et al., 2003).

Peracetic acid residues were typically present in the disinfected secondary and tertiary effluents, the concentrations being 1-2 mg/l lower than the applied PAA dose. This reflects the low reactivity of PAA for the water matrix of relatively high quality effluents, which improves the efficiency for disinfection action. PAA residues were typically low or absent in disinfected primary effluents, indicating that high COD and SS concentrations of wastewater effluents may significantly increase PAA consumption and thus decrease disinfection efficiency. Moderate PAA residual concentrations are not thought to cause any harmful ecological effects, since they are diluted and rapidly dissociate in the recipient water body, producing non-toxic dissociation products such as acetic acid (Liberti et al., 2002). In fact, a low residual concentration of disinfectant could be considered as a benefit in some wastewater disinfection applications. If there are long effluent discharge pipelines then these residues could be used as disinfection contact reactors. Disinfectant residues could also prevent microbial regrowth in pipelines or storage tanks, for instance if disinfected wastewaters have to be transferred over longer distances or are stored prior to reuse. Recent studies have shown that no bacterial regrowth takes place after PAA disinfection treatments, suggesting that bacteria are unable to repair the damage caused by even low doses of PAA disinfectant (Antonelli et al. 2006). PAA and acetic acid residues do increase the organic carbon content of wastewater effluent, but the increase may not be significant at economically applicable PAA doses. The increase of TOC concentration correspond the amount of carbon that is added into wastewater with the PAA product, and is approximately 1 mg/l of TOC for each mg/l of PAA product added.

The applicability of PAA treatment is affected by the target level of disinfection. PAA treatment could be a competitive disinfection method for secondary and tertiary effluents if the microbiological limit values would be at the level of 100-1000 CFU/100 ml TC or EC, and if strict limit values for virus and protozoan parasite removal would not be set. Under these conditions, PAA doses as low as 2 mg/l could be adequate. Although the disinfection of secondary and tertiary effluents by 2-7 mg/l PAA doses may achieve even <10-100 CFU/100ml TC or EC numbers, our pilot-scale disinfection experiments and previous studies have demonstrated that high PAA doses may be needed to achieve significant reductions of viruses, spore forming bacteria and protozoan parasites, and if one strives to meet the most strict microbiological limit values (Baldry et al., 1991; Liberti and Notarnicola, 1999; Liberti et al., 2000; Veschetti et al., 2003).

PAA treatment could also be used for the disinfection of primary effluents or WWTP by-pass wastewaters to reduce microbial loads passing into natural waters. Approximately 10-20 mg/l PAA dose could achieve adequate disinfection of primary effluents, depending on the influent quality and effluent quality criteria. The U.S.EPA has classified PAA as one of the five disinfectants for use on combined sewer overflows (U.S.EPA 1999e). The efficiency of PAA in treatment of low quality wastewaters may be better than that of chlorine. Chlorination forms harmful DBPs and may require very high disinfectant doses for efficient elimination of enteric microorganisms in organic matter rich wastewaters (Baldry et al., 1991). UV disinfection exhibits highly variable treatment efficiencies in the treatment of low quality wastewaters (Kuo et al., 1997; Al-Mogrin, 1999; Gehr et al., 2003), and cannot be used for disinfection of primary effluents or WWTP by-passes.

PAA treatment could represent a good alternative method for the disinfection of secondary and tertiary wastewater effluents. PAA has several advantages associated with its use in the treatment of different wastewaters i.e. relatively high disinfection efficiency (also in the organic matter rich wastewater), short contact time requirement and the absence of persistent residuals or DBPs. A PAA disinfection process is also relatively simple to install and operate. Recent studies have demonstrated that PAA disinfection could be a viable alternative, when compared to chlorination-dechlorination, UV, ozone or chlorine dioxide disinfection processes (Collivignarelli et al., 2000; Nurizzo et al., 2001). Some Italian WWTPs already use PAA disinfection, instead of chlorination, to achieve elimination of enteric microorganisms and to avoid formation of harmful DBPs, which are formed in chlorine-based disinfection processes (Mezzanotte et al., 2002).

6.4.2 Hydrogen peroxide (H_2O_2)

The disinfection efficiency of H_2O_2 was demonstrated to be low, which is in agreement with the previous studies (Liberti et al. 2000; Lubello et al. 2002; Wagner et al. 2002). The results suggest that the disinfection action of PAA product is mainly due to PAA itself, with the role of hydrogen peroxide being less important. H_2O_2 and PAA are both peroxides, but only PAA proved to be an efficient microbicide. The difference in disinfection efficiencies of PAA and H_2O_2 may be explained by the differences in their disinfection mechanisms or by the higher reactivity of PAA. PAA is organic peroxide and it may penetrate more efficiently into microbial cells than the hydrogen peroxide molecule, and this could account for its better disinfection properties. Some microorganisms may also be protected against hydrogen peroxide since they possess catalase enzyme activity. Peracetic acid can inactivate or inhibit catalase activity, and furthermore the catalase enzyme may not have any protecting effect against PAA (Wagner et al. 2002).

6.4.3 Sodium hypochlorite

The efficiency of sodium hypochlorite disinfection against *E. coli*, *S. enteritidis* and coliphage MS2 in organic matter rich PW was low. In the present study, sodium hypochlorite disinfection exhibited higher microbial reductions than H_2O_2 treatment, but lower reductions than PAA and UV disinfection treatments. These results differ from those obtained by Morris (1993) and Veschetti et al. (2003), who have reported similar microbial reductions in PAA and hypochlorite disinfection treatments of secondary effluents. The differences in experimental results may due to differences in the qualities of water that were treated. The synthetic PW used in this study was probably more susceptible to chemical oxidation by disinfectants than the typical secondary effluents.

6.4.4 Combined chemical/UV disinfection treatments

Relatively low UV doses (<18 mWs/cm^2) were needed to achieve 1-3 log (90-99.9%) enteric bacterial reductions in PW. Coliphage MS2 viruses were more resistant against UV treatment: their 1-1.5 log inactivation required higher UV doses of 22-38 mWs/cm^2, which was consistent with the values reported in the literature (Havelaar et al., 1990). In secondary and tertiary treated wastewaters, slightly higher UV doses (30-100 mWs/cm^2) are typically needed to achieve similar microbial reductions than those achieved in this study (Oppenheimer et al. 1997; Lazarova et al., 1998, 1999; Andreadakis et al. 1999; Rajala et al. 2003). Suspended solids in municipal wastewater effluents protect microorganisms against UV radiation, thus increasing the UV dose needed for microbial inactivation. In a collimator device, the microbial suspension is almost ideally mixed and the contact time is relatively long due to the low UV intensity in the equipment. Thus the efficiency of microbial exposure to radiation may be higher than in actual UV disinfection systems.

The combination of PAA and UV disinfection increased disinfection efficiency and showed synergistic benefits, with the highest synergy values reaching 2 log units for enteric bacteria. An increase in either the UV or the PAA dose increased the synergy values, probably due to increased radical formation. The combination of H_2O_2 and UV treatments exhibited no synergies. These findings are in agreement with the results obtained in previous studies (Lubello et al. 2002; Caretti and Lubello 2003). The present study revealed the high resistance of coliphage MS2 virus against combined disinfection treatments, i.e. the combined disinfection treatments produced no significant synergistic benefits.

The synergies achieved in PAA/UV treatments may be due to interaction between PAA and UV, producing reactive and microbicidal hydroxyl radicals due to the direct photolysis of PAA (Lubello et al. 2002; Caretti and Lubello 2003). The formation of the hydroxyl radicals could also be due to an indirect photolysis process (Lubello et al. 2002). In this case, the UV radiation affect other substances in the water (such as dissolved oxygen or hydroxyl ions) that act as initiator radicals: they attack the PAA molecule, generating hydroxyl radicals. The lack of synergies in H_2O_2/UV treatments suggests that there are differences in the radical formation potential between PAA and H_2O_2, the potential and the resulting disinfection efficiency being higher for PAA. Cho et al. (2004) has previously demonstrated a linear correlation between the amount of hydroxyl radicals and the extent of *E. coli* inactivation in TiO_2 photocatalytic disinfection.

The mechanism of synergy in combined PAA/UV disinfection could also be explained by a multiple damage mechanism: two different disinfection techniques may cause different types of injuries to the microorganisms. The principal targets of UV radiation are the nucleic acids, while chemical disinfectants, such as PAA and H_2O_2, are thought first to attack microbial cell walls, membranes and enzymatic or transport systems. As a result, the microbial repair mechanisms, which are required to repair minor damage, may become overwhelmed and be unable to repair the injuries leading to the subsequent death of the microorganism. In the case of a single disinfectant, the damage may be less extensive and susceptible to repair, but this may not be possible in the case of two disinfectants causing a greater spectrum of damage. The synergism achieved by using PAA/UV treatment compared to H_2O_2/UV treatment could be related to the greater disinfection efficiency of PAA compared to H_2O_2. PAA may also inactivate the catalase enzyme which is one defence mechanism involved in detoxifying free hydroxyl radicals. This would allow free radicals to more efficiently attack and inactivate microbial cells in combined PAA/UV treatment. Hydrogen peroxide does not inhibit catalase enzyme activity, and thus the enzyme may detoxify free hydroxyl radicals in combined H_2O_2/UV treatment.

The combination of PAA and UV disinfection methods in wastewater treatment plants could provide some advantages. It could allow lower disinfectant doses or lower contact times and thus allow smaller size of UV units and chemical disinfection basins. The efficiency and reliability of disinfection in existing UV disinfection units could be improved by the addition of a low PAA dose before the UV unit. This method could also improve the bacteriostatic effect and decrease the reactivation potential of microorganisms which had undergone the disinfection process. Lazarova et al. (1998) observed viable but non-culturable bacteria and some bacterial regrowth after PAA or UV treatments of wastewater effluent. Recently Antonelli et al. (2006) reported no bacterial regrowth after PAA disinfection of wastewater effluents. The combination of two disinfection methods could also destroy a wider range of microorganisms than a single disinfection method, since there are differences in the resistance of microbes against different disinfection treatments. The magnitude of the synergistic effect may be a function of the specific characteristics of the water to be treated (Biswas et al., 2003). In municipal wastewater effluents, the wastewater quality, as well as the characteristics and physiological state of microorganisms, may affect the disinfection efficiency and the synergies achievable by combined disinfection methods.

6.5 The practical implications of the results

6.5.1 Needs for tertiary treatment and disinfection of wastewaters

Wastewater discharges are regulated rather strictly in Finland. Efficient organic matter and phosphorus removal are the primary goals of wastewater treatment. Nitrification and nitrogen removal are also required in many WWTPs. However, in Finland, there are typically no limit values for the microbiological quality of wastewater effluents.

In recent years, the efficiency of wastewater treatment has significantly improved in many Finnish WWTPs as a result of changes in the operating the processes. Optimizing coagulant choice, its application point and dosing, as well as improvement of the final settling performance have significantly improved the efficiency of many WWTPs. Currently many conventional WWTPs achieve around 95 % total phosphorus and BOD reductions and with the final wastewater effluent having residual phosphorus concentrations even below 0.4 mg/l.

Conventional wastewater treatment processes without tertiary treatment cannot, however, continuously achieve optimal process performance, since at times changes in the wastewater quality, flow rate and temperature, as well as disturbances in treatment processes decrease the treatment efficiency. To achieve optimal performance of conventional WWTP, the final settling process must be very efficient, since already low concentrations of suspended solids in the final effluent increase loads of phosphorus, organic matter, microorganisms and other pollutants gaining access to receiving natural waters. Hydraulic overloading situations may also force WWTPs to by-pass untreated or only primary treated wastewaters into recipient water bodies.

There are few wastewater treatment plants in Finland that have tertiary wastewater treatment processes. The DAF process is the most common tertiary treatment process, while there are no RFS units in use in Finnish municipal WWTPs. A process combining flotation and filtration, called flotation filter, is used for tertiary wastewater treatment in at least one Finnish municipal WWTP (Savonlinna WWTP). In Finland, there are no full-scale municipal WWTPs with continuous disinfection of wastewater effluents. Many wastewater treatment plants have, however, as a precautionary measure, readiness to start disinfection of wastewater with chlorine under special circumstances, such as during a serious disease epidemic in the population. In other countries, such as in South and Central Europe, disinfection of wastewater is a common practise preceding wastewater reuse or effluent discharge into recipient water body.

In the future, the permit requirements for the discharge of wastewater effluents will become stricter, which will increase the needs to improve the treatment results in many Finnish WWTPs. As a result, adoption of tertiary wastewater treatment processes will probably be necessary in many Finnish WWTPs in the future. Efficient treatment of by-pass wastewaters and wastewater disinfection may also be needed in the future, at least under certain circumstances (e.g. discharge to small streams and lakes and other environmentally sensitive water bodies), to decrease wastewater pollution of natural waters and to meet the stricter treatment requirements.

6.5.2 Comparison of tertiary treatment processes

The present study showed that tertiary wastewater treatment by dissolved air flotation (DAF) or rapid sand filtration (RSF) can significantly improve the efficiency of wastewater treatment. Tertiary DAF or RSF processes efficiently remove enteric microorganisms, phosphorus and residual organic matter from secondary effluents, thus reducing pollution loads into recipient natural waters. Tertiary RSF or DAF processes can help improve the stability of the WWTP effluent quality as they lessen variations in the quality of the secondary effluent. Tertiary wastewater treatment also improves the efficiency of the final disinfection process by decreasing the amount of SS and organic matter that interfere with the chemical disinfectants or UV irradiation.

The tertiary RSF process typically achieved slightly lower residual concentrations of SS, turbidity and phosphorus and achieved higher UV transmittance values than was achieved in the DAF process, as shown by the present study and as reported by Rajala et al. (2003). However, by optimising the DAF process operation, a comparable effluent quality may be achieved.

The application of coagulant chemical is important in the RSF and DAF processes to achieve extensive removal of enteric microorganisms, phosphorus, SS, and organic matter residues. In some previous studies, RSF processes used as mechanical filtration have also been reported to achieve efficient removal of SS and turbidity (Kuo et al., 1997; Hamoda et al., 2002). In the present study, slightly lower coagulant doses were needed in RSF than in the DAF process to achieve comparable treatment results, which may be due to differences in mechanisms of flocculation and separation of solids. The optimum coagulant dose and effectiveness of coagulation are related to the characteristics of the influent water, and thus the optimising of process performance could be achieved by continuous detection of influent quality and adjustment of coagulant dose (Jolis et al., 1996; Kuo et al., 1997). The increased coagulant needs in the tertiary treatment could be compensated for by decreasing the amount of coagulant in the preceding treatment stages.

The RSF process is probably more sensitive than the DAF process to disturbing factors, such as changes of wastewater quality and increased hydraulic loading. A high concentration of suspended solids and turbidity in the tertiary filter influent may decrease the process efficiency and cause filter clogging problems (Kuo et al., 1997; Hamoda et al., 2002). A high hydraulic loading rate may cause an increase of the filter head loss and lead to possible wash-out of filter material in the RSF unit. There are, however, differences between different filter types in their abilities to tolerate increased loading situations, some filter types being capable of treating relatively high solids and hydraulic loadings without suffering any operational problems (Kuo et al., 1997). The RSF process used in this study, a continuous backwash filter, can typically better tolerate higher loads of suspended solids without operational problems, such as filter clogging and the generation of anaerobic conditions. In the RSF processes with intermittent backwashing those operational problems may cause large problems. To avoid the clogging problems and the generation of anaerobic conditions during the long-term usage of the RSF process, filter backwashing must be carried out so that sufficient removal of solids from the filter bed is achieved. The characteristics of the filter bed, such as the particle size of the filter medium and the filter bed configuration, must also be optimised. The DAF process can typically tolerate quite high concentrations of suspended solids in the influent water, as shown here in the pilot-scale and full-scale DAF experiments and as reported in previous studies (Ødegaard 1995, 2001; Mels et al., 2001; Reali et al., 2001a, 2001b; Pinto Filho and Brandão, 2001). The DAF process removes SS until the maximum separation capacity for SS or the maximum tolerable hydraulic load is achieved, after which part of the SS starts to escape from the process. The process efficiency may remain relatively high also under these circumstances. As a result of high tolerance for increased hydraulic and solids loading, the DAF process could be used for the treatment of WWTP overflows or by-passes during WWTP overloading situations.

The DAF process produces a thick sludge (dry solids content up to 3-10 %) that can be treated along with the primary and surplus sludges in the WWTPs (Arnold et al., 1995; Amato et al., 2001; Mels et al., 2001). In the RSF process, the separated pollutants are removed from the filter by backwashing and then circulated back to the wastewater treatment process. As the quantity of filter backwash water is typically around 5-10 % of the influent flow rate, the RSF process elevates the hydraulic loading of the wastewater process.

Another advantage of tertiary RSF and DAF processes is that both can be quickly started and stopped. This permits rapid start up of the process when the secondary effluent quality decreases and by-passing of the process if the secondary effluent quality fulfils the limit values of wastewater discharge. The DAF process can also be used for the treatment of primary effluents or by-pass wastewaters, as discussed above. It could also be used to lighten the load on the biological treatment process during high-loading situations.

The investment, operation and maintenance cost of DAF, RSF and sedimentation processes differ from each other, and they depend on many factors, e.g. the plant capacity, quality of the water to be treated, goal of the treatment and local conditions (Viitasaari et al., 1995; Teerikangas, 2000; Schofield, 2001; Heinonen-Tanski et al., 2002).

6.5.3 PAA disinfection processes

Conventional wastewater treatment and tertiary wastewater treatment processes typically cannot achieve microbiologically safe wastewater effluents to be discharged into natural waters or to be reused. Depending on the environmental conditions and the limit values for wastewater discharges, disinfection of wastewaters may be needed in WWTPs to regularly achieve efficient elimination of enteric microorganisms.

PAA offers many advantages in the disinfection of municipal wastewaters. PAA disinfection processes can efficiently destroy enteric bacteria. However, the PAA disinfection efficiency against enteric viruses and spore forming bacteria is lower. PAA disinfection has been shown to be more effective than other disinfectants also in the presence of higher levels of suspended solids and organic matter concentrations. Thus PAA does not necessarily require a tertiary treatment process before the disinfection process. PAA treatment typically requires at least a

10 minute contact time to be efficient, and a further increase in contact time can improve the destruction of enteric microorganisms.

One of the significant advantages of PAA disinfection is that it does not lead to the production of toxic, mutagenic or carcinogenic disinfection by-products (DBPs) or harmful disinfectant residues which then gain access to effluent waters (Booth and Lester 1995; Liberti and Notarnicola 1999; Collivignarelli et al. 2000; Monarca et al. 2000; Veschetti et al. 2003; Kitis 2004). The advantages of the PAA disinfection process also include the relatively simple and flexible operation and control of process, as well as relatively easy maintenance of equipment. When properly operated, PAA disinfection is occupationally and environmentally safe. The PAA disinfection system can quite easily substitute for sodium hypochlorite, e.g. it can use existing disinfection reactors. The only significant modification is related to the storage and dosing system, due to corrosion risks associated with PAA. The potential disadvantages of PAA treatment include the elevation of the BOD/TOC content of effluent water.

The investment, operation and maintenance cost of PAA disinfection process, as well as those of other disinfection processes such as chlorination, ozone and UV disinfection, depend on many factors, e.g. the plant capacity, quality of the water to be treated, goal of the treatment and local conditions, such as availability of the PAA product (Collivignarelli et al., 2000; Nurizzo et al., 2001; Heinonen-Tanski et al., 2002).

6.6 Contributions to knowledge

Primary and secondary wastewater treatment with simultaneous phosphorus precipitation was shown to eliminate around 95 % of the organic load and phosphorus and 90-99.9 % of enteric microorganisms present in raw wastewater. Even though there were high percent reductions, secondary effluents still contained some residual phosphorus and organic matter, and high numbers of enteric microorganisms, including pathogenic and antibiotic resistant salmonellae.

Tertiary RSF and DAF processes achieved 90-99 % reductions in enteric microbial numbers and efficient removal of residual phosphorus and organic matter in the wastewater effluents. The tertiary RSF and DAF processes could be potentially used to improve the efficiency of wastewater treatment, as the regulatory limit values for wastewater treatment are getting stricter in the future. They could also be used as efficient preliminary treatment processes preceding the final disinfection stage, improving the disinfection process efficiency.

The DAF process efficiently removed organic matter, phosphorus and enteric microorganisms from the primary treated wastewater effluents. The results demonstrate that the DAF process could be potentially used for the treatment of WWTP by-pass wastewaters during the treatment plant over-loading situations to avoid the harmful effects of those pollution discharges on natural waters.

The results of the present study point to the applicability of using PAA disinfection process for reducing the microbiological risks of very different wastewater effluents, including the municipal secondary and tertiary wastewater effluents, as well as organic matter rich primary wastewater effluent or WWTP by-passes. The results of the present study also demonstrate that the combined PAA/UV disinfection treatment can achieve high disinfection efficiency and synergy benefits. Both the PAA and PAA/UV disinfection treatments showed high efficiency against enteric bacteria, but coliphage viruses showed to be more resistant against the disinfection treatments.

7 CONCLUSIONS

1) Conventional biological-chemical wastewater treatment, including primary and secondary treatment, could typically reduce organic matter and phosphorus by around 95 %, and their residual concentrations in the secondary effluents were relatively low. The numbers of enteric microorganisms were usually reduced by 90-99.9 %, but the secondary effluents still contained high microbial numbers, including pathogenic, antibiotic resistant salmonellae. At times, the efficiency of secondary treatment process decreased and the effluent quality deteriorated, causing increased pollution load passing into the receiving water body.

2) The tertiary RSF process efficiently removed enteric microorganisms, phosphorus, suspended solids and organic matter residues from the secondary effluents. Their removal clearly improved when the coagulant chemical was dosed into the wastewater effluent before the filter unit (contact filtration). Reductions of enteric microbial numbers in the RSF process used as contact filtration were typically between 90-99 %.

3) The tertiary DAF process efficiently removed enteric microorganisms, phosphorus and organic matter from the secondary effluents. Approximately 90-99 % reductions of enteric microbial numbers, 50-90 % total phosphorus reductions and 10-50 % reductions of COD_{Cr} were achieved. The increase of coagulant dose and dispersion water recycle ratio improved the purification results, while changing the flocculation conditions (G-value, retention time) or increasing the hydraulic surface loading rate (from 5 m/h to 10 m/h) did not clearly affect the efficiency of the DAF process.

4) The tertiary RSF or DAF processes were capable of consistently achieving a high quality of wastewater effluent, in spite of variations in the efficiency of secondary treatment process. The tertiary wastewater treatment improved the efficiency of the final disinfection process, through reducing the SS, organic matter and initial microbial numbers in the wastewater effluent prior to the disinfection unit.

5) The DAF process efficiently removed enteric microorganisms, phosphorus and organic matter from the primary wastewater effluents. The results of the present study demonstrate that the DAF process can tolerate high loads of suspended solids and could be potentially used for treatment of primary effluents or WWTP by-pass wastewaters during the treatment plant over-loading situations.

6) Peracetic acid disinfection significantly improved the hygienic quality of primary, secondary and tertiary wastewater effluents. These results of the present study highlight the applicability of the PAA disinfection process for reducing the microbiological risks of very different wastewater effluents, including municipal secondary and tertiary effluents, as well as organic matter rich wastewaters, such as primary effluents or WWTP by-passes.

7) Combined PAA/UV disinfection treatment exhibited synergistic benefits, suggesting that the combination of PAA and UV disinfection could increase the efficiency and reliability of wastewater disinfection.

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