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Pharmaceuticals in the aquatic environment

The role of decentralised wastewater treatment in risk management

Case study: Recirculating Biofilter (RBF)

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Abstract

There are more than 80 pharmaceuticals and pharmaceutical metabolites detected in the aquatic environment. Regarding negative impacts of these compounds, the main concern lays in sensitive aquatic environment and long-term effects regarding human health and the environment. Although we have procedures to assess the risk that pharmaceutical residues represent to the environment, these procedures include various assumptions and data gaps. Therefore current knowledge is not enough to say how serious the threat is that we are dealing with.

Many concerned governments and organisations apply the precautionary principle (PP) regarding the potential risks that pharmaceuticals in the aquatic environment present. This means that while there is no full proof about such risks, risk management alternatives are nevertheless developed and assessed. In this thesis preventative alternatives and treatment alternatives of risk management are discussed. Preventative alternatives include source control and source separation as well as increasing awareness. Treatment alternatives include wastewater treatment both in centralised systems and decentralised systems.

Current knowledge about how effective decentralised systems are in removing pharmaceuticals is not sufficient. To be able to assess the role that decentralised wastewater systems can have in risk management, pharmaceutical removal effectiveness of a bench scale recirculating biofilter (RBF) was observed in laboratory experiments. The objective was to test if a nitrifying biofilter was able to remove four selected pharmaceuticals (ibuprofen, gemfibrozil, naproxen and diclofenac) from pre-treated wastewater. According to the laboratory results, ibuprofen, gemfibrozil and naproxen were removed more than 90 percent and diclofenac was removed with approximately 70 percent efficiency. These results show that there is potential in decentralised systems regarding pharmaceutical removal. However more research is needed.

Local conditions regarding pharmaceutical use patterns, water consumption, and treatment technology have an effect on determining the most efficient way to manage this environmental risk. Therefore efficient risk management includes both preventative and treatment approaches.

Executive summary

There is scientific evidence that there are traces of pharmaceuticals in our groundwater, drinking water, watercourses and wastewater. Wastewater treatment plants are the main route for human pharmaceuticals to the environment. So far it is known that the present concentrations of pharmaceutical residues do not represent an acute threat to human health but there is a reason to believe that other organisms in the aquatic environment suffer from this pollution. There are many uncertainties regarding the environmental impact of pharmaceuticals and therefore, some governments and organisations have adapted the precautionary principle as a guiding approach when dealing with this issue. Even though there is no full scientific proof about the level of the risk, they are looking for ways to reduce and prevent these pollutants reaching our watercourses.

This thesis looks into different current possibilities regarding risk management of pharmaceutical residues in wastewater. For this thesis two research questions were composed:

1) How can we manage environmental risks regarding pharmaceutical residues in wastewater?

    - What role can decentralised wastewater treatment have as a risk management alternative?

2) How effective is nitrifying Recirculating Biofilter (RBF) for reducing four selected pharmaceuticals from wastewater?

Risk management alternatives can be divided into prevention measures and treatment measures. In this study, preventative measures have been discussed based on a literature review and include for example source control, source separation and raising awareness of stakeholders about the environmental effects of pharmaceuticals.

Even if we would implement all reasonable preventative measures, pharmaceuticals that are consumed usually are excreted and so disposed of with wastewater. Therefore in addition to the preventative approach, treatment possibilities have been studied. According to literature, conventional centralised wastewater treatment plants can remove or reduce the amount of certain pharmaceuticals. The chemical structure of pharmaceuticals and certain treatment parameters affect how effective the removal is. For example, in this study, sludge retention time (SRT) and nitrification have been discussed and it seems that long SRT and a good nutrient removal increases degradation chances of certain pharmaceuticals.

Despite the potential of centralised solutions, there is an inevitable fact that a considerable share of the population is not, and cannot, be connected to centralised systems. In these situations wastewater can be treated onsite in decentralised wastewater systems. Decentralised systems have several benefits when compared to centralised systems including for example reduced investments regarding drainage, possibility to recycle nutrients and water reuse, flexibility regarding population growth and in the case of operational failure, environmental problems are usually smaller when compared to centralised systems. When well maintained, treatment results can reach very high standards regarding for example BOD and nutrients removal. However there was not information available about how decentralised systems can treat pharmaceuticals.

To be able to evaluate the role that decentralised wastewater treatment systems can have regarding risk management and pharmaceutical residues in the aquatic environment,

laboratory experiments were implemented. Testing was done with a bench scale recirculating biofilter (RBF). Regarding conventional centralised wastewater treatment systems (WWTS), it seems that if the treatment process includes nitrification (conversion of ammonia to nitrate) the process can also remove certain pharmaceuticals. Therefore during the experiments both ammonia removal and pharmaceutical removal were observed by analysing concentrations in system influent and system effluent.

Based on the laboratory analyses RBF bench scale system was able to remove both ammonia and four selected pharmaceuticals. According to the laboratory analyses the average removal of gemfibrozil, naproxen and ibuprofen was more than 90 percent and diclofenac was removed by average little more than 70 percent.

It is clear that based on these experiments, it cannot be assessed if the removal efficiency is similar regarding field systems and more research is needed to determine this. Results do however indicate that it might be possible to remove pharmaceuticals from wastewater with field RBF and in addition removal could be possible also with other decentralised systems.

Today there are no requirements for wastewater treatment regarding pharmaceutical residues but this might change in the future when we have a more complete picture about the impact of these substances. However it should be noted that wastewater treatment currently, but also most probably in the future is focused on removing organic matter and nutrients and it would be extremely expensive to design and construct treatment systems that target certain micro pollutants. Therefore it is important to know the capacity of current treatment systems regarding pharmaceutical removal. Acknowledging the current capacity it is possible to target for example preventative measures more efficiently. This means that when we know what we can treat we can target our preventative measures to those pollutants that we cannot treat.

If field studies will show that decentralised systems can have a similar ability to remove pharmaceuticals in wastewater than centralised systems, it can mean for example that regarding the treatment of these compounds, it is not necessary to prefer centralised wastewater systems. This is important for communities with low investment abilities, with special needs regarding water reuse and requirements for system flexibility.

It seems that both in the European Union area and in North America research currently is focused to fate research, developing analytical methods and risk assessment measures as well as to studying effects of different pharmaceutical residues to the environment. This includes the development of different risk management measures. When research covers all levels including the risk assessment process and risk management measures, the results benefit the whole research sector. For example when we know that naproxen is effectively removed from wastewater using conventional treatment methods regarding both centralised and decentralised systems, it might not then be a main priority to study how this substance affects aquatic organisms. When we know that diclofenac is highly persistent regarding wastewater treatment it is possible to consider for example preventative alternatives to deal with this specific pharmaceutical. When we learn more about the effects of different pharmaceuticals, their fate in the environment and possibilities to prevent or treat this pollution, we can select the most efficient risk management measures. This means that in the future we will have integrated solutions for risk management including both preventative and treatment management approaches.

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1 Introduction

1.1 Background

Clean water is essential for living organisms and globally it is a scarce resource. Only three percent of earth's total water volume is fresh water and more than half of this is tied to glaciers and icecaps (Gleick, 1996). Currently approximately one of six people worldwide do not have regular access to a safe and affordable drinking water source (Joint Monitoring Programme for Water and sanitation (JMP), 2004). There are various reasons for this, for example water resources can be exploited more than fresh water is generated and because of this communities need to start using lower quality water sources. Problems can be also economic where people cannot afford safe water and are forced to use unsafe sources. Making the problem more severe is the fact that these limited amounts of more accessible fresh water resources can be subject to pollution.

All human activities cause pollution which partly ends up in ground and surface waters. According to Miller (2005) *"water pollution is related to air pollution, land use, practices, climate change, energy use, solid and hazardous waste, and number of people, farms, and industries producing sewage and other waste."* Water pollution is considered to be any change (biological, chemical or physical) in the water quality that is harmful for living organisms. It should be noted here that harmfulness affects both humans and other forms of life. Water pollution has diverse effects and it can cause for example different health problems like diarrhoea and environmental changes like eutrophication and alteration of food chains. During the last decade new concerns about water pollution have developed.

Developments in analytical methods have made it possible to measure lower and lower concentrations of pollutants in water. One new group of pollutants that is now possible to track is Pharmaceuticals and Personal Care Products (PPCPs). Low concentrations of these have been found in Europe and in North America in drinking water sources, in rivers, groundwater and in wastewater effluents. Even though concentrations have been low, and might not have an acute effect to human health when quantities remain at the present level, we do not know the long term effects these substances may have. Also because unlike other pollutants, pharmaceuticals are designed to have a reaction in human body, we should ask: is a reaction possible in other organisms. Aquatic environments are sensitive. It is a possibility that bacteria exposed to pharmaceuticals may become resistant to disinfection and the increasing use of pharmaceuticals and personal care products have raised concern about the fate and effects of these compounds. Therefore we can state that these compounds represent a possible environmental risk and implementing the precautionary principle (PP) can be justified.

According to the precautionary principle, even when there is no full scientific proof of the level of the risk that these substances represent; the lack of scientific evidence should not forbid us to act and try to prevent and reduce this risk, when there is a sufficient reason to believe that these substances indeed are harmful to the environment (Rio Declaration, 1992). Measures that we can take to prevent or reduce the risks are called risk management measures. There are many different approaches that can be used to manage risk and these can include preventative and treatment alternatives. Preventative approaches can include for example preventing pharmaceuticals and personal care products from reaching water systems by using source control and waste separation. In addition we can also use treatment as a risk management alternative and in this case wastewater treatment would be the first option to look into since one of the main routes of entry for pharmaceuticals to the environment goes through wastewater treatment plants. Very often wastewater treatment is performed in conventional centralised systems, which means that a large facility collects the community wastewater through sewage collection system and wastewater is then treated using a series of processes. However it is important to notice that centralised wastewater treatment service is not available for everybody. There is a large population in countries like Sweden, Finland, the United States and Canada as well as in many other countries that are living in rural areas and are not connected to centralised sewerage. In such instances onsite wastewater treatment can be used to replace the common centralised system. The use of these decentralised systems has been considered to be an environmentally preferred approach to the wastewater treatment since water is treated near the place it has been generated and so there is the possibility to save resources.

It is known that conventional centralised wastewater treatment plants can reduce the amount of some pharmaceuticals in wastewater, but at the moment there are no studies available demonstrating the effectiveness of decentralised systems in dealing with such substances. Therefore it is important to assess the role of decentralised wastewater systems to have a more complete picture about choices we have regarding risk management.

There are various different decentralised wastewater treatment applications that use several different treatment technologies. Recirculating Biofilter (RBF) is one of these. It is scientifically researched, easy to use and highly effective regarding BOD and pharmaceutical removal (Farzana, 2008; Hu, 2006). Due to such characteristics RBF forms a good candidate to investigate the pharmaceutical removal potentials of decentralised wastewater treatment plant units.

This thesis is targeted for professionals in municipalities that are making decisions about wastewater treatment systems as well as for those who develop and produce decentralised wastewater treatment systems. It also presents information for policy developers as well as other parties interested about pharmaceuticals in the aquatic environment. This thesis has been written for International Institute for Industrial Environmental Economics (IIIEE) in Lund University, Sweden for the Master of Science program in Environmental Management and Policy.

1.2 Research objectives

The goal of this thesis is to contribute to the general understanding regarding risk management alternatives for pharmaceuticals in the aquatic environment.

To reach this objective, the research concentrates on the following questions:

1) How can we manage environmental risks regarding pharmaceutical residues in wastewater?

    - What role can decentralised wastewater treatment have as a risk management alternative?

2) How effective is the nitrifying Recirculating Biofilter (RBF) for reducing selected pharmaceutical substances from wastewater?

1.3 Methodology

1.3.1 Literature review

In the first stage of the research a desktop study is performed in order to achieve a general understanding about the relationship regarding pharmaceutical residues and the environment. Information about environmental risk assessment regarding pharmaceuticals, the policy directions in the European Union (EU) and North America about pharmaceuticals and the environment, and descriptions about wastewater treatment technologies were gathered. Studies about occurrence and fate of different pharmaceuticals and potential risks that pharmaceuticals represent were searched. Literature review was carried out focusing to studies that were done in the European Union area and in North America.

An assessment was completed on the current state of policy and perceived risk level in the European Union and North American regulatory agencies. Also research topics supported by government were screened and compared. This information was gathered using publicly available internet sources and published sources from Environment Canada, the United States Environmental Protection Agency (USEPA) and the European Union.

1.3.2 Assessment of centralised and decentralised wastewater treatment

Wastewater treatment technologies used both in centralised and decentralised systems were studied, and the benefits and shortcomings of each system were assessed. Descriptions of

different treatment technologies were extracted mainly from engineering study books. Legislation concerning wastewater treatment requirements in general in the EU and Canada were examined, and in addition requirements for decentralised systems were studied using Finish and Swedish sources. Legislation issues as well as regulations and requirements concerning pharmaceuticals and wastewater were obtained from environmental administrations with publicly available internet pages. Scientific articles about conventional wastewater treatment plants' ability to reduce pharmaceuticals in wastewater were reviewed to get an understanding about the potential and limitations of these technologies regarding certain pharmaceuticals. Scientific articles were searched from the European Union area, Canada and the United States.

The objective for the second step was to study if it is possible to reduce pharmaceuticals from wastewater using RBF. Experiments were done with a bench scale RBF in a laboratory conditions. First the bench scale system was observed regarding nitrification (ammonia removal) since it was assumed that this has an effect on pharmaceutical removal. This assumption was based on research done with centralised systems that has shown that systems that are nitrifying are also able to reduce the amount of certain pharmaceuticals (Servos & al., 2005; Assessment of Technologies for the Removal of Pharmaceuticals and Personal Care Products in Sewage and Drinking Water Facilities to Improve the Indirect Potable Water Reuse (POSEIDON), 2004). During the first experimental step the treatment system used different filter media to observe its affect to the ammonia removal. Nitrification efficiency was studied by analysing ammonia concentration from influent and effluent. During the second experimental step four different pharmaceuticals were fed to the biofilter system. Then the influent and effluent were analysed regarding both ammonia concentrations and pharmaceutical concentrations. A more detailed description about the experiments is presented in the Chapter 3.

The pharmaceutical agents that were selected for this research represent are commonly used drugs and have been studied in centralised systems. The results for laboratory analyses were compared with the results presented in scientific articles from centralised systems. Comparison was done to see if there were differences in the ability to remove pharmaceuticals when comparing centralised and decentralised systems.

Using the results of RBF experiments on the system's ability to remove ammonia and pharmaceuticals from wastewater, a statistical analysis was performed. To evaluate the significance of the results a "paired t-test" was used. This statistical analysis was selected because the samples that were taken from the system could be paired accordingly as they were taken at the same time (samples from the influent and effluent were collected within a short time interval). Even though it is not possible to sample exactly the same parcel of water in RBF system (i.e., the pairing is not exact), according to Berthouex & Brown (2002) *"any variation caused by this will be reflected as a component of the random measurement error"*. In the paired t-test the analysis is done by averaging the difference of each pair of sample results and to calculate if the average of these differences is different from zero. Pharmaceutical removal efficiency was analysed and the 95 percent confidence- intervals were calculated for each tested drug.

1.4 Scope

The focus of this thesis is in human pharmaceutical residues in wastewater. Even though there are also other micro pollutants found in wastewater, for example personal care products, pharmaceuticals were chosen as a main topic. This was for two reasons: firstly, it is impossible to prevent all of these substances from ending up in the wastewater even when preventative measures are included and secondly, they are targeted to react in a human body and there is reason to believe that the reaction is possible also in other organisms.

Current occurrences of pharmaceutical residues in the aquatic environment is presented from the European Union and North American points of view. Policies and the legal requirements for wastewater treatment and removing pharmaceuticals from wastewater are discussed following Canadian and North European (Sweden and Finland) regulations and visions.

The focus of this thesis is in decentralised wastewater treatment as a risk management alternative and examines the application of decentralised RBF system for pharmaceutical removal. The ability for decentralised systems to remove pharmaceuticals from wastewater is assessed by implementing laboratory experiments with a bench scale RBF. Pharmaceuticals that were selected for the experiments were gemfibrozil, naproxen, diclofenac and ibuprofen. These pharmaceuticals were selected since they are commonly used, their degradability, that is different for each compound, has been studied regarding centralised wastewater treatment, and analytical methods for the selection of these pharmaceuticals are available.

1.5 Limitations

This thesis focuses on domestic sources of pharmaceutical residues even though there are different pharmaceuticals used also for veterinary purposes. Veterinary products are excluded from this study since the main routes of entry are different than human pharmaceutical residues. Also personal care products are not included because they have different use patterns and purposes than pharmaceutical agents.

Risk assessment can be divided into 1) environmental risk assessment and 2) risk assessment for human health. In this study only environmental risk assessment is briefly described to give the reader an overview about the process. Regarding environmental risk management both treatment and preventative measures are included. However the former, not the latter is the primary focus. Treatment is discussed both from centralised wastewater treatment systems and decentralised wastewater treatment systems point of view, the main focus being in decentralised systems. Because of this focus, advanced treatment methods for centralised systems (ozonation and membrane technology) are only briefly discussed. Wastewater treatment process produces sludge that can contain pharmaceutical residues. However sludge treatment has been excluded from this thesis.

Different use patterns of pharmaceuticals and water alongside wastewater treatment technologies affect pharmaceutical removal. This might have an effect to the results of this thesis

Experiments done with RBF cannot be used to predict how pharmaceuticals can be removed with decentralised systems in general, as other systems may use different removal mechanism other than biofilters. Results of this study describe how RBF can decompose these chemicals in laboratory conditions. As such, results can only give indications how pharmaceuticals can be removed with field applications. Also it should be noted that the raw wastewater used in experiments was gathered from a centralised wastewater treatment plant so there might be a difference in characteristics of wastewater that was used for the experiments and wastewater that is usually emitted to the decentralised systems. Experiments were done by using selected pharmaceuticals so the results should not be applied as a generic model for all pharmaceutical agents.

1.6 Thesis outline

Chapter 1 Introduction contains background information for this thesis providing information about the topic, stating research objectives and discussing the scope and limitations of this study.

In **Chapter 2 Pharmaceutical residues in the aquatic environment**, the reader will be given an overview about how and why pharmaceuticals end up in our watercourses and how different characteristics of pharmaceuticals affect their occurrence in the aquatic environment. The risk that these substances represent, what is seen by the scientific community, and the description of environmental risk assessment for pharmaceuticals is discussed. In this chapter the implementation of the precautionary principle regarding environmental impact of pharmaceuticals is discussed. Different risk management approaches, preventative and treatment measures, are presented and their strengths and weaknesses are weighted. This chapter includes introduction to wastewater treatment and both centralised and decentralised wastewater treatment systems are assessed. Also design parameters and operational characteristics that have an effect to the pharmaceutical removal efficiency are explained.

In the **Chapter 3 Case study: Recirculating Biofilter (RBF)**, the reader will find information about RBF treatment application and benefits and weaknesses of this system. This chapter includes a description about the laboratory experiments that were implemented with a bench scale RBF system. The results of the experiments are presented. This chapter is targeted to those who are interested about the details of laboratory experiments.

In the **Chapter 4 Discussion** the results of the laboratory experiment are presented and compared to other studies. Policy direction and the relationship to the precautionary principle are discussed and efficient risk management measures weighed.

Chapter 5 Conclusions and recommendations provide a summary of this thesis and recommendations for future research.

2 Pharmaceutical residues in the aquatic environment

Drugs or pharmaceuticals in this study are defined following Gunnarssons & Wennmalms, (2006) definition that states that they are products that are designed to diagnose, cure or alleviate disease. These products are usually made from active substances, excipients and packaging. The active substance is the main element of the product and excipients can be designed to give the drug a proper volume, sufficient shelf life or make it for example easier to take.

There are more than 80 different compounds, pharmaceuticals and pharmaceutical metabolites, detected in the aquatic environment in the Europe, U.S, Canada and Latin America. Pharmaceuticals like analgesics and anti-inflammatory drugs that are generally used as pain killers, antibiotics, antiepilectic drugs, beta-blockers, blood lipid regulators, iodinated X-ray media, cytostatic drugs, contraceptives etc. have been detected in sewage, drinking water, groundwater and watercourses. Since the human body does not completely metabolize pharmaceuticals they are excreted slightly transformed or even unchanged, and end up in water systems. Measured concentrations have been low, for example in sewage influents and effluent samples have been up to the μg/l –level, so detecting these compounds from water has been possible only in the past ten years since developments in analytical methods (National Water Research Institute NWRI, 2007; Heberer, 2002).

The main routes of entry for the pharmaceutical residues to the aquatic environment are wastewater and wastes and the biggest points of pollution are wastewater treatment plants. Septic fields, landfills that leach to the groundwater and storm water overflow from residential sources can also be sources for these chemicals (Backus, 2007). Agriculture may contribute to the occurrence of pharmaceuticals in the environment because of the use of veterinary drugs and feed additives for livestock. Also manure use as fertilizer can lead to a leakage of these compounds to the surface waters (Herberer, 2002).

Pharmaceuticals represent a versatile group of chemical compounds. In general chemicals can be harmful and toxic because of many different mechanisms. For example chemicals can bind to molecules such as hormones, DNA and RNA, lipid membranes and proteins. This interaction can destroy these molecules or modify their structures potentially leading to changes in physiological functions. Pharmaceuticals can bind to transport proteins or receptors that are designed for hormones and block or enhance a natural function. They can also block the function of enzymes. Pharmaceuticals however differ from other chemicals in a way that they are designed to have an effect in the human body, and they can for example be designed to target enzyme and hormone systems of humans. These hormonal systems are in all probability also present in lower trophic level organisms, hence pharmaceuticals can affect other organisms, such as mammals, fish and lower order species and even plants (Breitholtz & Bengtsson, 2006; Gunnarsson & Wennmalm, 2006).

Since most pharmaceuticals are designed to be swallowed, the active substance needs to survive through the stomach acidic environment and must be resistant. Pharmaceuticals contain a wide range of compounds that have very different chemical and physical properties. The harmfulness of pharmaceuticals to the environment and/or human health is dependent on various properties such as: degradability (describes how fast substances are degraded), volatility (tells how easily a substance is passed into the gas phase and so determine how much of a certain substance can be transported to the atmosphere and react there), and water solubility (describes how easily a substance can be dissolved in to water). According to Tysklind & Fick (2006) pharmaceuticals can be divided in three different categories regarding solubility. First those that break down easily, second those that are water-soluble and third those that are fat-soluble. Water-soluble substances are found naturally in water and they are transported by the water cycle. Fat-soluble substances can be found in sediments and soil and they have a tendency to accumulate in food chains. Following this categorisation, pharmaceuticals like acetylsalicylic acid that breaks up easily can be found only near to the sources when there are large amounts of it released. Some lipid-lowering drugs that are water-soluble and stable can pass through the wastewater treatment plant and disperse into the receiving water system more or less unchanged. Some antibiotics (e.g. fluoroquinolones) are fat-soluble and stable and are absorbed to sludge particles in wastewater treatment process and can be released back to the environment when sludge is disposed.

The potential for a pharmaceutical to enter the environment and the level of concentration in watercourses is dependent for example on the volumes of that drugs that are used, how these pharmaceuticals behave in the human body (meaning if they metabolise well or if they are extracted unchanged to the wastewater) and also and how well wastewater treatment plant and drinking water facility or natural water systems can eliminate the pharmaceutical compound (Metcalfe & al., 2003). Concentration of pharmaceuticals in wastewater varies for different pharmaceuticals in different countries. This can be explained with different drug use patterns, differences in water consumption and differences in wastewater treatment technology (Lishman & al., 2006; Metcalfe & al., 2003).

Various factors can affect the use patterns and consumption of pharmaceuticals. One is that there are demographic differences between countries. The proportion of elderly in the population and shares of men and women can have affects upon use patterns and consumed volumes. Variation in therapeutic traditions, prescribing habits, availability of pharmaceuticals as well as the variation in over-the-counter drugs influences which and how much of pharmaceuticals are consumed (Nordic Medico- Statistical Committee (NOMESCO), 2004).

At the moment there seems to be a consensus that present concentrations of pharmaceuticals in the environment are not an immediate threat for human health (e.g. Christensen, 1998; Schwab & al. 2005; Breitholtz & Bengtsson, 2006). Consumption of pharmaceuticals is however, increasing for example in the Nordic Countries (NOMESCO, 2004). Also in Canada during the past 10 years the number of prescriptions filled has increased significantly (Paris & Docteur, 2006). According to Redshaw & al. (2008) there is a

potential for accumulation of certain pharmaceuticals. Accumulation can occur for example when sewage sludge is used as soil fertilizer or disposed. This raises concern that concentration levels in the environment could be higher in the future and that threshold limits regarding human health might be passed.

Since species share similar hormone and enzyme systems, pharmaceuticals that are designed to have an effect in these systems in humans can have similar but also totally different effects in other species and organisms. Therefore pharmaceutical residues in the aquatic environment are considered to be a potential risk to other species. According to Breitholtz & Bengtsson (2006), unintended intake of pharmaceuticals by humans is limited usually to drinking and cooking water. On the other hand organisms living in the aquatic environment are exposed to these chemicals via their gills during oxygen replenishment. Organisms that are small or at young stage, can have very thin wall separating them from the aquatic environment and so osmotic transport can become an important route of entry. Some organisms live in sediments and/or feed by filtering particles suspended in the water, hence they expose themselves to chemicals that are not easily soluble in water and tend to bind to these particles. According to Gross-Sorokin, Roast, & Brighty (2006) it seems that estrogen emissions could be causing for example increasing number of hermaphrodite fish in English rivers. Gagné, (2007) states that *" there are indications that some aquatic species are likely to accumulate some drugs and that they are likely to produce harmful effects on fish"*. Cleuvers (2002) points out that even though acute effects in the aquatic environment seem unlikely "*considerable combination effects can occur*".

2.1 Environmental risk assessment of pharmaceuticals

Risk assessment is a way to evaluate how big a threat pharmaceutical residues are in the environment. Risk is considered by the United States Environmental Protection Agency (USEPA, 2008b) considered to be:

> *"the chance of harmful effects to human health or to ecological systems resulting from exposure to an environmental stressor. A stressor is any physical, chemical, or biological entity that can induce an adverse response. Stressors may adversely affect specific natural resources or entire ecosystems, including plants and animals, as well as the environment with which they interact."*

According to this definition pharmaceutical residues in the aquatic environment can be seen as chemical stressors that can have harmful effects on human health or to the ecological system. To be able to estimate if this potential risk is something that must be dealt with, one can perform risk assessments. A risk assessment can be determined in various ways. A risk analysis can be defined as:

> *"an assessment of the probability that adverse effects will occur and of their possible extent"* (Statens offentliga utredningar (SOU), 1984).

Risk analysis is a demanding procedure where quantitative estimates of the probability of adverse effects occurring are included. A precise assessment is seldom done, and a more common way to assess risks is by risk characterisation, which is:

> *"An estimation of the incidence and severity of the adverse effects likely to occur in a human population or environmental compartments (e.g. soil, air or water) due to actual or predicted exposure to a substance, and may include a "risk estimation", i.e. the quantification of this likelihood* (Technical Guidance Document (TGD), 2003).

One component in risk assessment is the hazard analysis that assesses the properties of chemicals that cause effects on the environment (SOU, 1984).

The USEPA (2008b) divides risk assessment into two main categories 1) Human Health Risk Assessment and 2) Ecological Risk Assessment (in this thesis Environmental Risk Assessment (ERA)). Risk assessment discussed in this study as a procedure for environmental risk assessment. ERA is a combination of different sub-analysis and it is usually implemented as step-by-step process (see Figure 2-1) According to TGD (2003) the main elements (steps) of a risk assessment are:

- hazard identification;
- dose (concentration) - response (effect) assessment;
- exposure assessment;
- risk characterisation.

Risk assessment is based on gathered data that can vary within very wide limits concerning both data quality and extent. Therefore risk assessment contains varying degrees of uncertainty, compensated for by using different application factors. These factors are discussed more in detail below (Gunnarsson & Wennmalm, 2006; Lundgren, 2006).

The objective of the first step, **hazard identification,** is to identify substances relevant to environmental effects and their dose-response relationships. This is done based on the information of substances and their inherent chemical and physical properties and information about the substance's ecotoxicological properties (Gunnarsson & Wennmalm, 2006; Lundgren, 2006).

During the second step, **effect assessment**, the Predicted No Effect Concentration (PNEC), is determined. PNEC can be calculated for different environmental compartments - for example for sewage treatment plants (micro-organisms), inland water (organisms living in water and sediment), terrestrial ecosystems and groundwater etc. - separately. PNEC is formed using data about the concentration of active substance that does not have effect to three defined aquatic organisms. If one organism is more sensitive to the substance than the

other the lower concentration is chosen in the evaluation. Here an assessment factor is included to compensate various uncertainties of the data (for example that there might be more sensitive organisms that has not been tested or that tests are done to show acute effects and not chronic exposure) (Gunnarsson & Wennmalm, 2006; Lundgren, 2006).

During the third step, **exposure assessment,** Predicted Environmental Concentration (PEC) is calculated. When calculating PEC it is possible to estimate the concentration of a certain substance in the environment. This can be done for example by calculating the quantity of the active substance in the product that is sold, consumed (assumption that everything that is sold is also consumed) and excreted and dividing this by the volume of used water (dilution of the substance). PEC is also corrected with assessment factor (Gunnarsson & Wennmalm, 2006).

The last step in risk assessment is to **combine earlier steps** by comparing PEC and PNEC. If PEC is greater that PNEC active substance can have adverse effects in the aquatic environment. However if the value of PEC is smaller from these two the active substance should in theory, not have an adverse effect on the aquatic environment. It should be noted that this type of an assessment can give only an approximate idea of the risk of environmental effects and this method includes many approximations that can over- and underestimate the risk (Gunnarsson & Wennmalm, 2006).

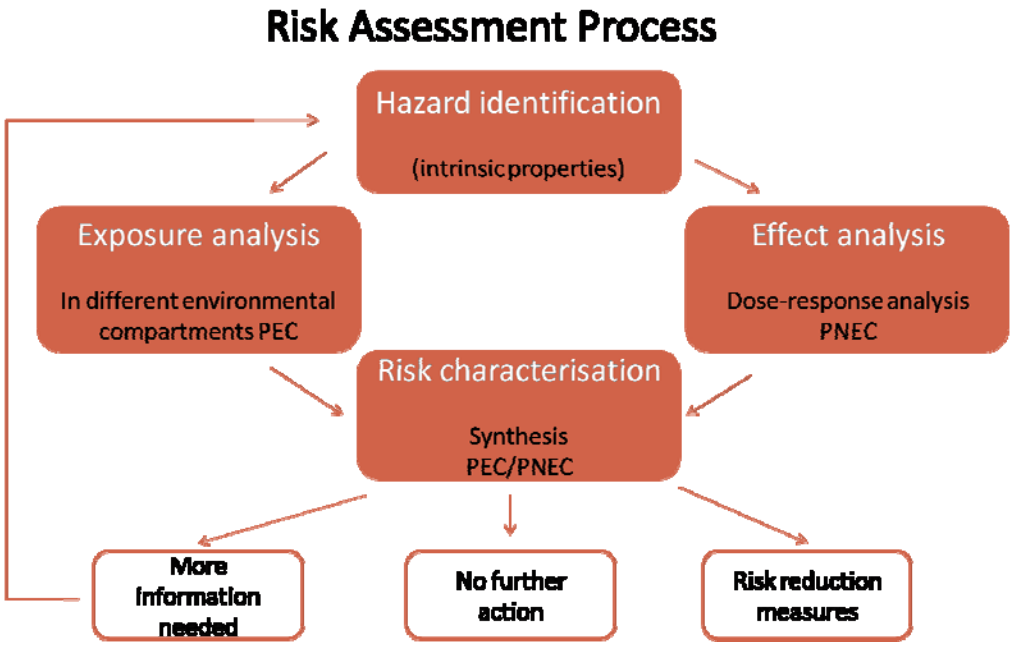

Figure 2-1. Risk assessment process for pharmaceuticals.(Source: Lundgren, 2006:108)

There is a need to have standardised methods for the implementation of different steps within environmental risk assessments. This has been an objective for the European Union financed project Environmental Risk Assessment of Pharmaceuticals (ERAPharm). This project follows risk assessment steps and it is divided in three different stages: 1) fate and exposure assessment; 2) effect assessment; and 3) environmental risk assessment. Fate and exposure studies are looking into environmental variables and how they affect the fate of pharmaceuticals in water. Also models for predicting the concentrations of pharmaceuticals in water and other environments are developed. During the effect assessment different tests are evaluated based on how helpful for risk assessment they can be. This stage also includes studies about effects that certain pharmaceuticals have in different organisms. The objective of environmental risk assessment stages are to evaluate action limits and trigger values of environmental risk assessment procedures. The overall goal is to gain a more complete picture about risk assessment of pharmaceuticals and to develop improved guidance on the environmental risk assessment of pharmaceuticals (ERAPharm, 2008). Similar studies are currently implemented in North America (USEPA 2008a; NWRI, 2007).

According to Lundgren (2006), there are three possible conclusions based on risk assessment: 1) more information and testing is needed; 2) there is no need for further information gathering and testing and there is no need for risk reduction measures; and 3) there is a need for limiting (managing) the risks (Lundgren, 2006).

However, as we understand the ERA procedure presented earlier, risk assessment currently contains many uncertainties. Data gaps and various assumptions need to be included to the assessment process for example in a form of assessment factors. It is important to consider the procedure, how it is implemented currently, the missing data, and these subsequent effects on the interpretation of the results of these assessments.

One way to acknowledge data gaps and uncertainty included to the ERA process is to apply the precautionary principle (PP). The PP was introduced in 1992 in the Rio Declaration in Principle 15 in a following way:

> *"In order to protect the environment, the precautionary approach shall be widely applied by States according to their capabilities. Where there are threats of serious or irreversible damage, lack of full scientific certainty shall not be used as a reason for postponing cost-effective measures to prevent environmental degradation."* (The Rio Declaration, 1992)

According to Grandjean (2005) there are two key elements regarding PP. First it justifies our acts (e.g. policy guidance) when we are uncertain of the risks from environmental exposures and when there is *"limited but plausible and credible"* scientific evidence of *"likely and substantial"* harm. Second, the burden of proof should be shifted *"from demonstrating the presence of risk toward demonstrating the absence of risk."* (Enick & Moore,2007; Grandjean, 2005)

Enick & Moore (2007) state that regarding pharmaceuticals and the environment, we should not forget the context of the ERA process which is "*as critical to the output of the risk assessment*

as is the numerical data that is used for risk assessment derivation". They continue by stating that scientific objectivity is necessary but not sufficient to direct policy development and that it should be noted that *"The recognition that ERA* [Environmental Risk Assessment] *is a scientific process dependent on its current social context has the potential to increase the accuracy and transparency of the risk evaluation system."* They point out that when we have acknowledged inherent uncertainty the ERA precautionary principle needs to be applied regarding policy development. This raises a question: in which stage should we integrate the PP into risk assessment process? Enick & Moore (2007) state that it should be integrated to the risk assessment process already in the early stage as in if the PP is included when risk reduction measures are considered, it is already too late. They support this statement by stating that the outcomes of risk assessment are often irreversible, for example, *"if a substance is initially assessed as being non-toxic, but further use/research proves it to be toxic ... , some of the effects may be mitigated but the damage to the affected individuals cannot easily be eliminated."*

The aquatic environment in general is a sensitive ecosystem that includes various different species that have developed for example complicated mutual dependencies and specialisations (Breitholtz & Bengtsson, 2006). We know that there are indications that pharmaceuticals might be harmful for some aquatic organisms already when concentrations are at the current level (Gagné, 2007; Gross-Sorokin, Roast, & Brighty, 2006). In addition the consumption of pharmaceuticals is increasing (NOMESCO, 2004) and we know that some of these pollutants can potentially accumulate (Redshaw & al., 2008). We have also acknowledged that we have data gaps and therefore, risk assessments include various assumptions. Considering this one can interpret that conditions to apply the PP are fulfilled. Even though there is no full scientific proof of the level of the risk, various organisations and governments have adapted the precautionary principle and they are already taking steps to assess and develop risk management measures (NWRI, 2007; Assessment of Technologies for the Removal of Pharmaceuticals and Personal Care Products in Sewage and Drinking Water Facilities to Improve the Indirect Potable Water Reuse (POSEIDON), 2004).

2.2 Preventative risk management approaches

We have wide selection of measures that we can use to prevent or reduce environmental risks. In this study risk management approaches are divided to preventative alternatives and treatment alternatives.

The pollution prevention approach has been successful considering many other pollutants. According to USEPA (2008c);

> *"Pollution prevention (P2) is reducing or eliminating waste at the source by modifying production processes, promoting the use of non-toxic or less-toxic substances, implementing conservation techniques, and re-using materials rather than putting them into the waste stream".*

Regarding pharmaceuticals it is possible for the pharmaceutical industry to modify

manufacturing processes so that less residues are produced – during production or in the use - or to select less environmentally harmful substances for the pharmaceuticals. However, domestic use is an important issue in this matter and solutions that are good for the industry are not valid. Therefore, it might be beneficial to understand pollution prevention as any act that is taken to reduce waste before its disposal and treatment. In this study two preventative approaches are discussed 1) source control and 2) waste separation.

2.2.1 Source control

Source control and life cycle management approaches are considered to be beneficial ways to reduce pharmaceuticals in the environment. Implementation would require **raising awareness** of consumers about the effects that pharmaceuticals have in the environment and secondly, educating public how to prevent negative impacts when using and disposing of pharmaceuticals. Collaboration by all stakeholders, government, pharmaceutical industry, pharmacist associations, the media and the education system is considered to be important (NWRI, 2007). There are different ways to raise awareness and activate stakeholders and one interesting suggestion would be to use **ecolabelling**. Ecolabels are used already globally and for example ecolabel Swan that is used in European Nordic Countries have schemes to assess product groups within personal care products (Joutsenmerkki, 2008). Ecolabelling provides information for consumers about ecological characteristics of a product so that consumers can make decisions about what products to buy or not to buy. The mechanism to include pharmaceuticals to existing ecolabel schemes is not however known, and currently there are no official international standards about how to classify pharmaceuticals regarding their environmental impacts.

One option could be to use **classification models for pharmaceuticals** that are developed in Sweden. There are two models. "The Stockholm model" was developed by the Stockholm County Council, Apoteket AB, Swedish Chemicals Inspectorate and other ecototoxicology experts. It aims to provide environmental information about pharmaceuticals for patients and prescribers. The objective is to encourage pharmaceutical companies to consider environmental issues when developing new pharmaceuticals and reduce the impact that these products have. According to this model pharmaceuticals are assessed for their biodegradability, potential bioaccumulability and toxicity to aquatic organisms. Pharmaceuticals are assigned a value based on these characteristics, which describe the harmfulness to the environment. The higher the value, the more harmful is the pharmaceutical. The second model, "the Swedish model", is an initiative of the Swedish Association of the Pharmaceutical Industry (LIF), the Swedish Medical Products Agency, Apoteket AB, the Swedish Association of Local Authorities and Regions and Stockholm County Council. While the first model is based on assessment of inherent environmental hazards the second model is based on a combination of hazard and risk assessment. The results of this combined assessment are presented to three different target groups: patients, prescribers and specialists. The content of the information for these groups varies so that it is most specific regarding specialists (Gunnarsson & Wennmalm, 2006).

Even though it is important to have information available about environmental impacts of pharmaceuticals, it does not necessarily mean that consumers are making environmentally

sound decisions. Keeping in mind that pharmaceuticals are meant to heal or prevent illnesses, it is not likely that medication would not be taken because it is or can be harmful to the environment. On the other hand, it might be that if there are two alternative pharmaceuticals that have a same effect, it is possible for customers to choose the one with less environmental impact.

In addition to the schemes that are targeted to give information for consumers it is possible to control the amount of pharmaceuticals ending to watercourses by **reducing medical consumption**. Even though medical treatment obviously should not be reduced just because of the environmental impact, we can include to our preventative risk management alternatives measures that prevent illnesses and so minimise the need to use pharmaceuticals. These measures are versatile selection of policies and approaches from general health education to tax policies. Also, it would be beneficial not to medicate "just in case". Since the use of pharmaceuticals is increasing it might be that we sometimes take medication unnecessarily. Reducing unnecessary pharmaceuticals consumption can also have benefits regarding public health and public costs.

Regarding attempts to reduce pharmaceutical consumption it should be noted that different countries have different mechanism to support pharmaceutical use. For example governments have different schemes to subsidise these products for consumers. Also it might be that a doctor's salary is dependent of the amount of prescriptions he or she gives. Different countries can have very different health care cultures and there might be mechanisms that prevent actions to reduce the use of pharmaceuticals. It is important to be aware of these kinds of structures when designing risk management alternatives.

In addition to information one reasonable approach could be **product take-back programs** for pharmaceutical residues. For example in Canada campaigns for a consumer to return residual drugs to the pharmacist at no extra charge have been implemented. However according to NWRI (2007) in Canada these take-back programs need to be further studied to analyse their cost-effectiveness. In Finland residual drugs are classified as hazardous waste and consumers are allowed and encouraged to return them to any pharmacy without an extra fee. Medical waste is gathered from pharmacists and it is then treated in a special waste treatment plant. In Sweden a similar system is used and there it has been estimated that 65 percent of residual drugs are returned to the pharmacy (Castensson & Gunnarsson, 2006). Take-back programs do not only benefit the environment but also they prevent pharmaceuticals from getting into the hands of people that should not be using them since unused pharmaceuticals are not disposed with conventional domestic waste.

One interesting scheme would be to implement **extended producer responsibility** and involve producers to participate in residual pharmaceutical disposal. Take-back programs financed by manufactures could be a good way to involve pharmaceutical industry. Usually customers need to get pharmaceuticals from special shops (pharmacist) and this makes collection for this kind of a waste quite simple as the Swedish and Finish examples show.

2.2.2 Source separation

Source control is however not the only way to deal with pharmaceuticals ending up in the aquatic environment and source separation is worth consideration. Source separation means that **pharmaceutical waste streams are separated from other wastes and then treated**. This could mean that we collect and treat for example wastewaters that we know are more concentrated before discharging it to the sewage system. This could be implemented in various ways and for example industrial, hospital and nursing home wastewater could be treated onsite before discharging it to the common sewage. Regarding the definition above we could include take-back programs to this category.

Since high amounts of pharmaceuticals are excreted via urine, it is possible to use **urine separation** to separate more concentrated waste stream from less concentrated stream. Separated urine is then possible to treat in terms of both pharmaceuticals and nutrients. Since urine constitutes less than 1 percent of the wastewater volume but contains most of the nutrients that end up in wastewater, this approach would also give benefit in nutrient removal and it can offer an option for nutrient recycling. However this approach would require adjustment in toilet seats and possibly in drainage and so would need investments from home owners as well as operators of the sewage collection systems. Also for example Larsen & al., (2004) states that removal mechanisms in source separated urine are not yet fully developed and technology lags behind when it is compared to research in conventional wastewater treatment plants.

The preventative approach has been successful both in industry and households concerning many different waste streams. However, even though all preventative actions should be taken into consideration when management measures are developed it might be impossible to prevent all pollution. First of all if there is no source separation, pharmaceuticals that are used will end up in wastewater. Secondly this work discusses only pharmaceuticals, but if we take into consideration personal care products that have very different use patterns, then for example urine separation is not an answer. To assure effectiveness, risk management approaches have to include treatment as well.

2.3 Treatment approach

One promising approach to reduce the amount of pharmaceuticals in receiving water systems is wastewater treatment. Even though current conventional wastewater treatment plants are not designed to reduce pharmaceuticals but rather to treat pollutants like organic matter and nutrients, they have (according to recent studies), an ability to reduce the amount of certain pharmaceuticals. There are also more advanced technologies that can remove these pollutants effectively. The effectiveness of the removal of certain pharmaceuticals depends on the treatment process and design and operating parameters of the plant. Therefore, it is important to understand how the general treatment process works.

2.3.1 Introduction to the wastewater treatment

Wastewater treatment is done to reduce the pollutants from wastewater before discharging it

to natural water bodies. The constituents of wastewater can create health and environmental problems. Wastewater can be evaluated by its characteristics that are three-fold: physical; chemical; and biological. The first group includes for example water temperature and different solid materials that are flushed with wastewater. The second group includes for example total dissolved solids (TDS) that are the solids (residue) that are left after wastewater has been filtrated and evaporated. It also includes heavy metals like cadmium, copper, lead, mercury, organic material, nutrients (nitrogen N and phosphorous P) and pharmaceuticals. The biological characteristics include for example bacteria that can be either beneficial or harmful for the treatment process as well as viruses. By analysing these characteristics it is possible to evaluate the quality of the wastewater and estimate how harmful it is when discharged to the receiving water body. It is also possible to determine which harmful substances need to be treated. This is dependant on the characteristics of the receiving water body, since some natural water systems can be more sensitive to for example organic matter or nutrients than other water systems. This means that usually the treatment requirements are stated according to the sensitiveness of the receiving water system (Drinan, 2001).

Pollutants in the wastewater can have several negative effects when discharged to the receiving water body. When organic matter is released to natural water systems, micro-organisms start to use it to create energy (micro-organisms use organic matter for example for growth and reproduction). They use organic matter by decomposing it, which requires oxygen that they take from the receiving water body. If there are large amounts of organic matter that is decomposed, the oxygen level of the water (the amount of oxygen that is dissolved in water DO) can decrease. Oxygen depletion affects the fauna and flora of the water system and it can lead to the situation where large amounts of fish die due to lack of oxygen. Heavy metals are important to remove from wastewater since they are dangerous to health in general. Nutrients (N and P) cause eutrophication of surface waters when algae and water plants use them and overgrow. Eutrophication causes for example massive algae blooms and it can create anoxic areas in bottom waters. It can also lead to alteration of food chains and this has an effect to the fish stocks (Boesch & al. 2006). It should be noted that nutrients in surface waters can have also other sources than wastewater and for example fertilizer run-off from agriculture can be a major source. Ammonia is also a source for eutrophication and can be toxic for fish and other aquatic organisms. When it is discharged to the watercourse it needs a lot of oxygen to oxidise and therefore ammonia oxidizing can lead to oxygen depletion in the waterbed.

Problems that untreated wastewater causes in natural water systems are eliminated or reduced by wastewater treatment. Wastewater treatment can be implemented in centralised systems in which one plant collects the community's wastewater through a sewage system. After, the treatment wastewater effluent is discharged to receiving watercourse that can be for example a river, coastline or a lake. Treatment can be also done onsite where the water is used and after treatment water can be released near the place where water has been

generated. Onsite treatment, also called decentralised wastewater treatment, has some similarities with centralised systems.

Technology used in conventional wastewater treatment plants has developed from physical treatment (when only solid objects were removed from the water) to modern multi-stage operation that is currently common in the western world. The wastewater treatment (WWT) legislation has been evolving together with technology development. At the moment wastewater treatment in Europe is legislated by the European Union and by the States themselves. EU is regulating urban wastewater treatment by Council Directive 91/271/EEC that was adopted on 21.5.1991. This Directive "*concerns the collection, treatment and discharge of urban waste water and the treatment and discharge of waste water from certain industrial sectors*". Its objective is "*to protect the environment from the adverse effects of ... waste water discharges*". The Directive regulates, depending on the location of the outflow, wastewater discharges that exceed 2,000 population equivalent (p.e.)[1]. At the moment there is no EU regulation for small-scale treatment, but for example both Sweden and Finland have their national treatment requirements also for small-scale systems (Council Directive 91/271/EEC).

In Canada wastewater treatment is regulated by Canadian Environmental Protection Act. 1999 and the Fisheries Act. According to Canadian Council of Ministers of the Environment (CCME, 2008) wastewater is managed at federal, provincial and municipal levels and a variety of policies, bylaws and legislation are regulating wastewater treatment. Treatment technology used in Canada varies greatly from no treatment (only screening) to state-of-the-art treatment. Effluent quality and the necessary degree of treatment in federal establishments are stated in Guidelines for Effluent Quality and Wastewater Treatment at Federal Establishments (EPS 1-EC-76-1, April 1976). The policy is that these regulations are always stricter or equal to the standards or requirements of any provincial regulatory agency. This means that there can be establishments that are following lower treatment standards, if they are not federal establishments. These guidelines state that they do not regulate small installations. However the size of a small installation is not defined.

2.3.2 Conventional centralised wastewater treatment process

Conventional wastewater treatment usually consists of physical, chemical and biological treatment steps. The process includes preliminary treatment and the objective is to remove different waste solids from the water stream. This is done to prevent these solids from disturbing the equipment and the purification process during later steps. Preliminary treatment can include various different processes like: screening, grit removal, flow equalisations, shredding etc. (Drinan, 2001).

The first actual treatment step after preliminary treatment is primary sedimentation or the primary clarifier. During this step suspended organic solids settle to the bottom of the

[1] According to the Council Directive 91/271/EEC, 1 p.e. (population equivalent) stands for "*the organic biodegradable load having a five-day biochemical oxygen demand (BOD_5) of 60 g of oxygen per day*".

primary sedimentation tank. Solids that are lighter than water are floating on the surface of the water and are skimmed and removed. The second treatment step is biological treatment that can be performed in various different ways but the most used is an activated sludge system. The alternative for the activated sludge process is the attached growth process. During this step, the organic matter and some of the nutrients are decomposed by biological processes: nitrification, denitrification and biological mineralisation. Activated sludge treatment requires aeration to operate sufficiently (Metcalf & Eddy, 2003; Drinan, 2001).

After the aeration step, wastewater is settled by the secondary sedimentation process wherein the bacteria absorb the pollutants and are removed from the water. This is done so that advanced wastewater treatment processes can occur. Advanced treatment is implemented so that the purification process can meet the special requirements regarding wastewater discharge. There are different processes that are used during advance treatment steps and they are selected according to the required purification results. During this step it is possible to remove for example nitrogen, phosphorous, soluble chemical oxygen demand (COD) and heavy metals. Nitrogen is usually removed by biological nitrification/denitrification process. Phosphorous removal is usually done by chemical precipitation. Toxins and heavy metals can be treated by membrane processes or by different land application (Schröder & al., 2007; Drinan, 2001).

The concentration of pharmaceuticals is reduced during all of these stages depending for example on the type of the pharmaceutical, sludge age which means the time that the bacteria is present in aeration process, substrate availability, concentration of oxygen in different stages of the process and so on (Larsen & al. 2004). Parameters that influence the degradation efficiency of pharmaceuticals are discussed more in detail in the section 2.3.6.

2.3.3 Strengths and weaknesses of centralised systems

Treatment in centralised facilities is effective for example because of the more stable wastewater flow rate that keeps the biological process more stable. Also it is easier to monitor and adjust one big facility than several small ones. For a homeowner, connecting property to the common sewage is a care free way to handle wastewaters. Property owners do not need to maintain the system or take responsibility for the treatment results.

In urban areas where there is a lack of space, centralised systems offer an efficient way to treat wastewater. Well maintained conventional plants operated by professionals can reach good results. Centralised systems usually need to observe the treatment results and report them to the authorities hence treatment plants are under the supervision of authorities.

Even though treatment results are good regarding well-maintained centralised systems, overflows, for example, can be a problem. This problem occurs with combined sewer systems where both storm water and wastewater are collected to the treatment plant. During the heavy rains it might be that the capacity of a treatment plant is not enough and part of the storm water and wastewater mix needs to be directed untreated pass the treatment plant

strait to the receiving waster system (USEPA, 2008d).

Guterstam (1997) points out that it is necessary to "*create feedback loops of resources from wastewater to society*" and he claims that ecological demands for wastewater treatment are not achieved by using conventional treatment when we consider toxic substances, recycling of nutrients and the use of natural resources in general. This means that for example nutrients and heat of the wastewater should be used as well. Also the treatment process itself should need as less energy as possible. Schröder & al. (2007) claim that:

> "*The main aim of applied environmental sciences in the field of wastewater treatment has to be the amelioration of the effluent quality from WWTPs* (Waste Water Treatment Plants) *and the enforcement of reliable standards of regenerated waters in contact with ground water resources. Only hereby will Europe be able to increase the sustainability of drinking water resources and contribute in a modest way to decrease effects of global change by lowering energy usage, CO_2 emission and waste production during wastewater treatment."*

To answer this criticism, an ecological engineering approach has been recommended. This would mean that when new technologies are developed, ecological aspects and social awareness of different environmental problems should be taken into account (Guterstam, 1997). According to UNEP (2002) Environmentally Sound Technologies (ESTs);

> "*..are technologies that have the potential for significantly improved environmental performance relative to other technologies. ESTs protect the environment, are less polluting, use resources in a sustainable manner, recycle more of their wastes and products, and handle all residual wastes in a more environmentally acceptable way than the technologies for which they are substitutes."*

The conclusion of Schröder & al. (2007) is that environmentally sound water treatment will include the reduction of greenhouse gasses, recycling materials and development of affordable technologies and these should be prioritised. Jenssen & Vant (1997) define ecologically sound wastewater treatment as "*treatment concepts that promote a high degree of recycling as well as minimizing environmental stress*". According to them the two main principles of ecologically sound wastewater treatment are 1. recycling and 2. decentralised and onsite treatment.

2.3.4 Decentralised wastewater treatment systems

Concerning risk management and pharmaceuticals in the aquatic environment we have first looked into a preventive approach. It was concluded that preventing this pollution reaching our watercourses completely may be impossible. Therefore a treatment approach needs to be included. Centralised wastewater systems can be an efficient way to treat wastewater but it

should be noted that there are large communities for example in Finland, Sweden, Canada and the United States that are not connected to centralised wastewater treatment systems and that are treating their wastewater onsite. In Finland there are approximately one million people, about 20 percent of the Finnish population, living in households that are not connected to centralised sewage systems and because of this wastewater has to be treated onsite in 350,000 permanent residences and in 450,000 holiday houses (Finnish Environmental Institute, 2008). In Sweden there are approximately 500,000 permanent residents and same amount of holiday homes that are not connected to centralised wastewater systems. In the near future 50-60 percent of these households need to make improvements to their systems to be able to meet the requirements for wastewater treatment (NUTEK, 2003). In Canada, approximately 25 percent of the residents, 7.5 million people, are dependent on decentralised wastewater treatment system (Environment Canada, 1999) and in the U.S. approximately 23 percent of the estimated 115 million occupied homes are connected to onsite systems (U.S. Census Bureau, 1999). These communities are also sources of pharmaceutical pollution. However the role that decentralised systems have or can have in risk management is not clear.

The treatment process in decentralised systems follows the same steps that are used in centralised treatment plants. However, usually the advanced treatment steps are excluded. The main difference between centralised and decentralised treatment processes is the scale meaning that centralised systems can be large facilities treating wastewater from large communities when decentralised systems can treat wastewater produced by a single home or by a small community. The principle of treatment however is the same. In decentralised systems the treatment steps can include settling that is followed by biological treatment. According to Venhuizen (1998) decentralised systems are based on the idea that *"wastewater should be treated (and reused, if possible) as close to where it is generated as is practical."*

While centralised wastewater treatment plants are operated by a municipality or a company, decentralised systems can be used and operated by a single household or a small community. Finland's Environmental Administration recommends that the first step for property owner should be to investigate if there is a possibility to connect the property to a centralised wastewater sewage system. However if there is no such system available, the second option would be to connect several households together and treat wastewater in a small community plant. These choices are supported since it is convenient and easy when the property owner does not have to worry about the treatment facility or treatment results alone and costs can be shared between participants (Suomen ympäristökeskus, 2005). If these two options are not available or the costs are too high, the household needs to take care of the wastewater onsite. One option is to collect wastewater into a septic tank and when it is filled up, transport wastewater to the nearest treatment plant. This however can be quite costly if the treatment plant is far away and a lot of wastewater is produced. The second option is to treat wastewater onsite. There are various different applications that can be used for onsite treatment regarding both single households and small communities.

The technology used today in decentralised treatment is based on wastewater's biological treatment that can be combined with chemical treatment. The most used biological applications can be divided in two different treatment processes:

1) Suspended growth treatment processes (activated sludge process)

2) Attached growth treatment processes

Constructed wetlands and lagoons are also used for wastewater treatment but for example in Finland, because of the cold climate, these applications are not common. However where the climate is warmer, these applications can be sufficient and cost-efficient choices for WWT. Treatment process in lagoons and wetlands follows the natural water purification process and is not discussed here in detail (UNEP, 2002).

There are various models available for decentralised wastewater treatment. The treatment process is usually depended on micro-organisms biological processes. In this process micro-organisms reduce the amount of organic matter and nutrients in the wastewater. Usually the first step for wastewater treatment, no matter what model is used, is primary sedimentation in a septic tank, where solids and floating scum is separated from the wastewater.

In an activated sludge reactor the micro-organisms develop sludge where the decomposing of the organic matter happens. The reactor is always aerated. Depending on the retention time, also ammonia can be removed by biologically converting it to nitrate nitrogen. Phosphorous can be removed by using precipitation chemicals. Sludge reactors can have a continuous flow or can treat one batch of wastewater at a time (Sequencing Batch Reactors, SBR). According to USEPA (1999) "*The difference between the two technologies* [conventional activated sludge reactor and SBR] *is that the SBR performs equalization, biological treatment, and secondary clarification in a single tank using a timed control sequence."*

Attached growth treatment processes differ from activated sludge process so that the micro-organisms are attached on the surface of different filling material where they develop a biofilm[2]. In a trickling filter the filling material can be for example rocks, plastic or sand and gravel. For example in sand filter the filter media is a bed of sand and gravel that is buried to the ground. When wastewater flows through the sand bed, micro-organisms develop a biofilm on the surface of soils particles. Micro-organisms of the biofilm start to decompose wastewaters organic matter and depending on the retention time ammonia removal occurs. Phosphorous is usually precipitated either so that phosphorous adsorbing media is mixed in sand or that purified water is collected though drainage system from the sand bed and gathered into a precipitation tank where precipitation chemicals are added.

There are many different factors that have an effect on what kind of a system is chosen for

[2] *"Biofilm: A colony of bacteria and other microorganisms that adheres to a substrate and is enclosed and protected by secreted slime. Biofilms readily form on virtually any surface, whether nonliving or living, where there is moisture and a supply of nutrients. They are important components of aquatic and terrestrial ecosystems, typically providing nutrients for small organisms at the base of food chains."* (Martin & Hine, 2008)

the property. For example it is necessary to take into consideration purification requirements, the area available for wastewater treatment plant, the slope of the site (is it necessary to pump water or can gravity be used), the level of ground water, wastewater treatment systems distance to the fresh water source and natural waters like rivers and lakes. Also maintenance requirements and costs including both investment and use phase, needs to be assessed.

Even though at the European Union level or in Canada, there are no federal established treatment goals for small-scale treatment, Finland and Sweden are regulating decentralised treatment plants. In Finland new legislation for decentralised wastewater treatment, the Government Decree on Treating Domestic Wastewater in Areas Outside Sewer Networks (542/2003), came into force on 1.1.2004. This Decree "*sets minimum standards for wastewater treatment and the planning, construction, use and maintenance of treatment systems*" (Government Decree on Treating Domestic Wastewater in Areas Outside Sewer Networks (542/2003), Valtioneuvoston asetus 542/2003). The Swedish Environmental Protection Agency has stated requirements for small-scale onsite treatment plants and they are very similar with Finnish ones. Requirements for treatment are divided in two categories; normal level and high level. The basic necessity is that treatment has to be implemented in a way that it does not pose a threat to health, living comfort or the environment. Requirement levels for Finnish and Swedish small-scale systems are presented in a Table 2-1. (Naturvårdsverkets allmänna råd om små avloppsanordningar för hushållsspillvatten (NFS) 2006:7).

Table 2-1. Treatment requirements for small scale systems in Finland and in Sweden

Parameter	Finland % removal requirement	Sweden % removal requirement	
		Normal level	High level
organic material	≥ 90	90	90
total phosphorus (P_{tot})	> 85	70	90
total nitrogen (N_{tot})	> 40		50

Finnish reductions are calculated for person-equivalent load that is defined as "*the average load of untreated wastewater generated by one resident measured as grams per day (g/d), where a person-equivalent load of one means the daily load in which organic matter expressed as biological oxygen demand over seven*

days (BOD_7) amounts to 50 g/d, total phosphorus to 2.2 g/d and total nitrogen to 14 g/d." Swedish reductions are calculated for person equivalent load and it is defined almost in the same way that in the Finnish Degree. In Sweden the reductions are calculated using following p.e. BOD_7 amounts to 48 g/d, total phosphorus to 2 g/d and total nitrogen to 14 g/d and daily water use is calculated to be 170 l/day (NFS 2006:7).

Also in Finland when it is justifiable, for example in sensitive areas, the local conditions can be taken into consideration and municipalities can have higher or lower requirements for reductions. However the main requirement that is stated in Finland´s Environmental Protection Act (86/2000) is that "*wastewater in areas not connected to any centralized sewerage system must be treated so that it does not pollute the environment and there is no risk of pollution*". In both countries regulations state only the reduction target for small-scale treatment but it does not set requirements for certain technologies. This means that as long as the reduction targets are met, households can make their own choices about the technology and way to treat their domestic wastewaters.

According to Environment Canada's federal discharge guidelines wastewater that is discharged to freshwater lakes and low-flow streams, rivers and estuaries, and open coastline should have BOD_5 less than 5, 20, or 30 mg/l, respectively and ammonia concentration should be less than 1.0 mg/l. Total phosphorous should less than 1.0 mg/l. Also in Canada municipalities can have stricter discharge requirements (Hu, 2006).

According to the research done in Finland and in Sweden single family treatment plants are able to approach required purification results and there does not seem to be great differences between different models. The Finnish research concluded that phosphorous reduction was the most difficult to achieve. The best technologies for phosphorous removal were advanced sand filter (sand filter including phosphorous precipitation) and Sequencing Batch Reactors (SBR). Conventional sand filter (sand filter without phosphorous precipitation) did not achieve as good results in phosphorous removal. On the other hand both conventional and advanced sand filters removed ammonia and organic matter more effectively than SBRs. Both in Finnish and in Swedish studies proper maintenance was highlighted as an important factor regarding good treatment result (Vilpas & Santala, 2007; Hu, NUTEK, 2003).

2.3.5 Strengths and weaknesses of decentralised systems

Venhuizen (1998) points out several communities in the U.S. where it has been concluded that decentralised systems are a far more cost-effective choice regarding local conditions. He states that in case of a failure for example in pump units or pipelines the environmental harm is smaller than it would be with large centralised systems. Decentralised systems are usually flexible in a way that treatment facilities can be added to the systems when there is growth in population. There is in contrast to centralised systems which must be designed usually with extra capacity to be able to answer to the future treatment needs. One benefit for decentralised systems, especially in the areas suffering from water scarcity is that reusing treated water for example for irrigation, toilet flushing, car washing etc., is easier since treatment is done near water generation and it is not transported out site community. One of the benefits when implementing decentralised systems is that there is no need to invest in

large collection systems that have only one objective, to transport wastewater.

However if decentralised systems are not well maintained it can be a source of environmental pollution as well as a health hazard. In rural areas where communities are dependent on their own water sources, leaking and non-functional treatment applications can for example pollute wells. Because of the number of the onsite treatment plants, it might be difficult to supervise all the systems. Biological treatment needs stable conditions to operate and it can be difficult for example in a single household treatment plant to have a stable wastewater flow. This is even more challenging in summerhouses and properties that are used only part time. Biological treatment needs time before the treatment efficiency reaches the best possible level and this might be a problem if the system is used only short periods of time. These challenges highlight the importance of choosing a suitable application to a property.

In conclusion it can be stated that conventional small-scale wastewater treatment plants, when well maintained, are sufficient for achieving currently required treatment results. However there is no information available about how these applications are able to remove organic micro pollutants like pharmaceutical residues. Regulations at the moment do not set any requirements on reductions for these substances, but for example in Lund's commune environmental authorities do not exclude the possibility that these requirements can be introduced in the future (U. Hedberg-Henriksson, personal communication, 5.3.2008).

2.3.6 Pharmaceutical residue removal in conventional centralised wastewater treatment plants

Centralised wastewater treatment systems have been criticised because not only are they seen as a wasteful way to control water pollution but also because they are not capable of removing all PPCPs from the wastewater. For example Schröder & al. (2007) state that performance of the state-of-the-art wastewater treatment plants is nowadays not effective enough regarding pharmaceuticals.

This statement is based on the parameters that are considered as relevant by the legislation when designing treatment plants. Plants are usually designed to remove organic matter and nutrients. Schröder & al. (2007) claim that even though a reduction of organic matter and nutrients has had a significant effect in water quality in general, organic micro pollutants like PPCPs and endocrine disrupting substances are not targeted at the moment when treatment plants are designed. It is true that currently legislation does not include micro pollutants like pharmaceutical residues into the treatment requirements. But even though conventional treatment plants are not designed to remove for example pharmaceuticals from wastewater and they do not have the ability to remove all PPCPs, according to resent studies (e.g. Lishman, L. & al. 2006; Servos & al. 2005; PODEIDON, 2004; Metcalfe & al. 2003) conventional treatment plants can remove or reduce the amount of certain pharmaceuticals from wastewater.

Degradation of pharmaceuticals during WWT process is dependent for example on the process used on the treatment plant and chemical properties of the pharmaceutical. According to Lishman & al. (2006) depending on the chemical properties the pharmaceuticals entering the WWTP can be subject to:

1) mineralisation to CO^2 and water,

2) retention by the solids portion (sludge/biosolids) if the compound is fat soluble,

3) release to the receiving water either as the original compound or as a degradation product.

Although several studies both from North America and in Europe (e.g. Lishman, L. & al. 2006; Servos & al. 2005; POSEIDON, 2004; Metcalfe & al. 2003), provides information how different parameters of WWT process affect the degradation of pharmaceuticals, the actual influence of such parameters on the efficiency of degradation is not yet fully understood. So far, sludge age (solids retention time and hydraulic retention time), substrate availability (substrate inhibition), redox conditions (aerobic, anoxic or anaerobic environment), sorption and reactor configuration (for example number of cascaded compartments, biofilm growth surface and sand filtration) have been discovered to have an affect on pharmaceutical concentration reduction (Larsen & al., 2004). In this study sludge age and nutrient removal that is one indication of biofilm growth are discussed more in detail.

One of the most important parameters for pharmaceutical reduction is sludge age. This parameter is used to design treatment plants that are based on activated sludge process. **Sludge retention time (SRT)** tells how old the sludge is in the aeration process and in addition how long time micro-organisms have time to grow in the system. SRT affects the performance of the plant, volume of the aeration tank, how much sludge is produced during the treatment and oxygen demand of the process. Different bacteria need different lengths of time to grow and for example heterotrophic bacteria that remove organic matter grow faster than bacteria that are responsible for nitrogen removal. SRT is dependant for example on temperature so that the lower the temperature, the longer the SRT should be to achieve the same results. Usual SRT, when just organic matter is the target, is from 5 to 6 days in 10°C (Metcalf & Eddy, 2003). For example Servos & al. (2005) found out that those plants and treatment lagoons with high SRT were very effective in reducing the level of hormones. Lishman & al. (2006) reported that removal of diclofenac seemed more efficient when SRT was more than 30 day. There was no change in removal efficiency of this drug when SRT was less than 15 days.

However not all treatment plants use activated sludge process. According to POSEIDON (2004) also biofilters seem to reduce certain pharmaceuticals from wastewater if the system reaches the same nutrient removal as activated sludge system. Observations of Servos & al.

(2005) support this by stating that it appears that centralised plants that are well operated and which also achieve nitrification tends to have a higher removal of estrogens. Therefore in this study it is assumed that if a treatment system has an ability to remove ammonia from wastewater (nitrification), it can also remove certain pharmaceuticals. However linkage between pharmaceutical removal and nitrification has not been yet fully studied or understood.

Ammonia is removed from wastewater usually by using biological nitrification. During this process certain bacteria are used to convert ammonia to less harmful compounds. Nitrification is a two step microbiological process. During the first step ammonia (NH_4-N) is oxidised to nitrite (NO_2-N) and during the second step nitrite is oxidised to nitrate (NO_3-N). Oxidation is done by nitrifying bacteria so that during the first step it is done by ammonia-oxidising bacteria (AOB) and during the second step nitrite-oxidising bacteria (NOB) (Siripong & Rittmann, 2007).

1. step:

$$2NH_4^+ + 3O_2 \rightarrow 2NO_2^- + 4H_2O \quad (2.1)$$

2. step:

$$2NO_2^- + O_2 \rightarrow 2\,NO_3^- \quad (2.2)$$

Total oxidation reaction:

$$NH_4^+ + 2\,O_2 \rightarrow N\,O_3^- + 2H^+ + H_2O \quad (2.3)$$

As we can see from the equations above, nitrification process needs the presence of dissolved oxygen. Nitrifying bacteria need also carbon source, phosphorous and trace elements like calcium, copper, zinc etc. for growth. The nitrification process is sensitive to various factors like low temperature, extreme pH and different chemical inhibitors. Low temperature can be a problem of course in Nordic parts of the world but also heavy rains can lower the process temperature if combined sewage system is used. Optimal pH according to Metcalf & Eddy (2003) is in the range from 7.5 to 8.0. As we can see from the nitrification equation, during the nitrification process hydrogen ions H^+ are released. This means that the nitrification process itself makes the environment more acidic. Therefore the process might need adjustment, for example addition of lime. Nitrifying organisms are sensitive also to a wide range of organic and inorganic compounds hence they are good indicators of the presence of toxics already at low concentrations. Toxic compounds include for example solvent organic chemicals, amines, tannins, proteins, alcohols etc. These toxins

can inhibit nitrification or even kill the nitrifying bacteria (Metcalf & Eddy, 2003).

In some wastewater plants nitrification is followed by denitrification, a process where bacteria convert nitrate to nitrogen gas (N_2) in an anaerobic environment. However denitrification is not within the scope of this study.

Even though conventional wastewater treatment systems cannot remove all micro pollutants from wastewater, for example novel membrane techniques and ozonation with or without urine separation have shown potential. According to POSEIDON (2002) ozonation is a good post treatment step and it can be recommended if wastewater is reused. Ozonation is considered to be rather cheap but highly energy consuming process. This might limit possibilities to apply this technology. Reif & al., (2007) reported a high reduction for ibuprofen and naxproxen by using a membrane bioreactor (MBR). Diclofenac was removed less than 10 percent. This technology is used in activated sludge process so that membrane separates active biomass and treated wastewater. Similar reduction results have been reported also using conventional treatment processes (Lishman & al., 2006; POSEIDON, 2004). In addition there are highly effective membrane technologies for example reverse osmosis, but the costs for this kind of a treatment are high (Metcalf & Eddy, 2003). Schröder & al. (2007) criticise high costs and maintenance requirements of these advanced technologies and he points out that there is a possibility that these techniques are adopted only by wealthier countries and even then they are used only in large scale wastewater treatment plants.

3 Case study: Recirculating Biofilter (RBF)

In order to assess the viability of decentralised wastewater treatment as a risk management tool, a recirculating biofilter (RBF) (decentralised wastewater system), was selected for a case study. Since there was no information available on pharmaceutical degradation and decentralised wastewater systems it was important to test if it was possible to remove pharmaceuticals from wastewater and thereby contribute to the understanding regarding the role of decentralised systems in risk management. Since this was the first study to look into this issue using a RBF it was necessary to do the experiments in laboratory conditions. As mentioned earlier, regarding centralised systems there are different parameters and operational characteristics that affect the pharmaceutical removal efficiency. One of these characteristics is nutrient removal. In this case study both RBF nitrification efficiency and the ability of pharmaceutical removal were tested.

RBF has been scientifically studied before regarding for example BOD removal (Hu, 2006) and nitrification and denitrification efficiency (Farzana, 2008). These studies offer a background for experiments performed with pharmaceuticals and they provide the possibility to compare results of this study with previous results.

3.1 Recirculating Biofilter as a treatment application

Recirculating biofilters use sand, gravel or other media to treat settled wastewater. Treatment application includes a septic tank and a filter bed that can be lined and excavated to the ground. During the treatment settled wastewater flows from the septic tank to recirculation tank. From the recirculation tank wastewater is dosed though distribution network onto the surface of a filter bed. Wastewater percolates through the filter bed and it is gathered through under drain collection systems. Part of this water is then re-circulated to the recirculation tank and part of it is discharged from the treatment system (treated wastewater effluent). Feeding frequency onto the filter bed is usually 1 to 3 times per hour and typical recirculating ratios are between 3:1 and 5:1 (United Sates Environmental Protection Agency (USEPA), 2008e). A schematic flow diagram for recirculating biofilter is presented in Figure 3-1.

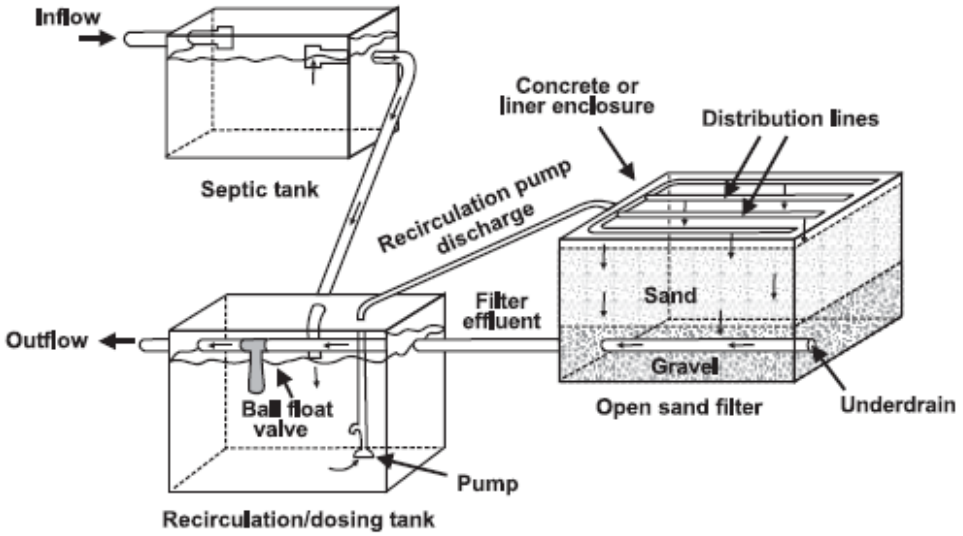

Figure 3-1. Schematic flow diagram for a recirculating biofilter (RBF). (Source USEPA, 2008c)

RBF treatment is based on microbial processes. Microorganisms form biofilm on the surface of the filter media and they absorb soluble and colloidal waste material (organic material and nutrient) in the wastewater. Absorbed materials are then either used to build new cell mass or they are degraded to CO_2 and water. Most of the treatment occurs on the surface of the filter media bed (approximately 15 cm of the filter depth). In this top layer suspended solids (SS) and BOD are removed. It is stated that BOD is possible to remove almost completely with sufficiently long retention time. Nitrifying bacteria are located deeper in the filter media (USEPA, 2008e).

Recirculation of wastewater provides some advantages compared to the single-pass filter. It minimises odours and improves BOD and total suspended solids (TSS) treatment (Hu, 2006). It is also possible to use higher hydraulic loading rate (HLR) when recirculation is included. Hydraulic loading rate determines wastewater retention time in filter media. According to USEPA (2008e) depending of the filtering medium size, HLR ranges from 0.12 to 0.24 $m^3/m^2/day$.

RBF has shown good ability to remove BOD, TSS and total nitrogen (TN). Usually BOD and TSS effluent concentrations are less than 10 mg/l and nitrification is usually complete (except in severely cold conditions). Recirculation tank promotes natural denitrification which can result 40 to 60 percent removal of TN. Chemical absorption of phosphorous is limited by the characteristics of the filter media and chemical precipitation might be necessary if phosphorous removal is included to the treatment requirements (USEPA, 2008e).

Recirculating biofilter is a flexible application. It can be used for both single households and small communities. According to USEPA (2008e) RBFs are "*Extremely reliable treatment*

devices." and they are resistant to flow variations. Since RBFs can use higher hydraulic loading rates than for example a systems including septic tank and conventional percolation area, the footprint required for the treatment can be reduced. RBFs can use various different filter media materials depended on what is locally available and studies have shown that crushed glass is a good alternative for silica sand and that geotextile has a potential (Farzana, 2008; Hu & Gagnon, 2006).

Hu (2006) points out that crushed glass has several advantages when it is compared to silica sand. He states that crushed glass is less expensive and that is has an environmental advantage since crushed glass is recycled product. Also since crushed glass can be pulverised into different sizes for specific design requirement, when sand is sieved for specific sizes, it is considered to have an advantage. These advantages however might be depended on local conditions. It might be for example that there is no glass recycling scheme implemented, or that there is sand available locally with a low price. There are several factors that can affect to the environmental performance of a product and without further research; it is hard to state that crushed glass has an environmental advantage. According to Hu (2006) by Loomis & al. (2004) textile filters can also have an advantage when they are compared to natural media filters. This is because of the smaller foot print required for the treatment (higher HLR can be used) and lightweight compact design.

Like any other treatment application also RBF needs to be constructed carefully according to design specifications and it needs to be maintained sufficiently. Regular maintenance on RBF includes inspection of the pressure head in the end of the distribution systems. Also lines need to be cleaned (drained and brushed) at least once per year. Recirculation pumps and controls need to be inspected and calibrated and the surface of the filter media unit needs to be scraped and cleared from vegetation. Recirculation tank needs to be observed for sludge accumulation and sludge is good to remove one to three times per year (USEPA, 2008).

3.2 RBF bench scale setup

The RBF system setup has been presented in the Figure 3-2. Raw wastewater (system influent) was first put into to the septic tank from where it was pumped to the recirculating tank. From recirculating tank wastewater was pumped by the second pump to the biofilter columns where wastewater travelled through the columns. Biofilter effluent was gathered to the biofilter effluent tank where the water was divided into two parts. Four units of the water volume were circulated to the recirculation tank and one unit of the biofilter effluent was discharged to the RBF system effluent tank (treated wastewater). Wastewater was fed into the system at a constant frequency and feeding was controlled by a computer based pumping and timing system. Based on the previous studies by Hu (2006) and Farzana (2008) the feeding frequency was selected to be 96 times per day. The pumping frequency was structured in cycles that were 15 minutes long. During one cycle pumps were operating first 4 minutes after which there was 11 minutes long recess.

During the experiment time two trials were done. During the first trial there were five parallel treatment lines as described in Figure 3-2 and during the second trial six parallel treatment lines. During the second trial bench scale system was modified so that the pump that was pumping system effluent from biofilter tank to the effluent tank was removed and the system effluent was gathered to the biofilter tank. Pumps that were used in the system were not designed to operate in such a low flow (minimum 1 ml/4 minutes) and this caused problems during the trial. Eliminating one pump from the system helped to guarantee effluent for sampling.

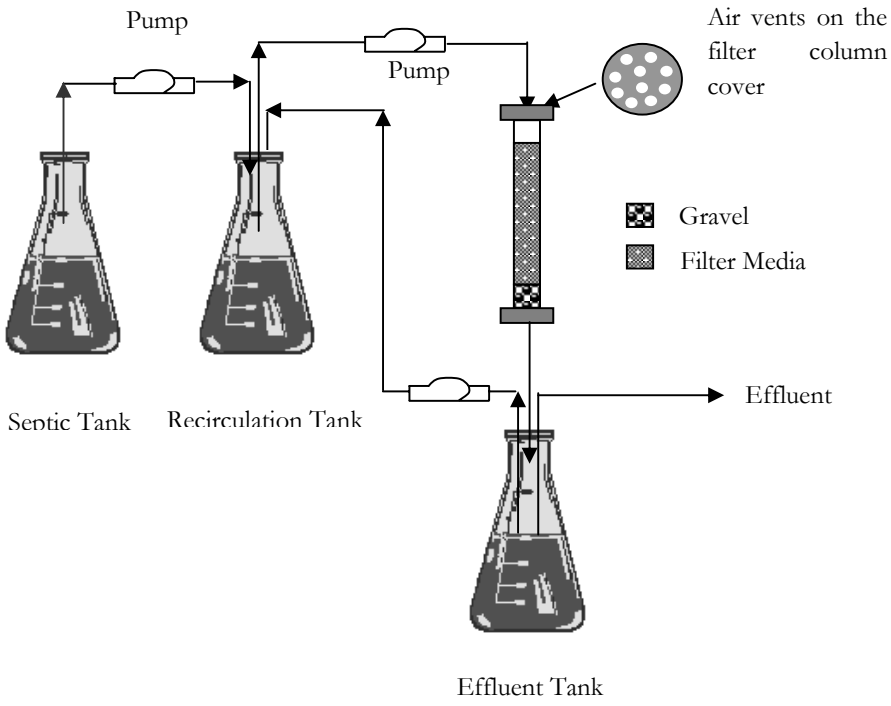

Figure 3-2. RBF system setup. (Source Hu, 2006)

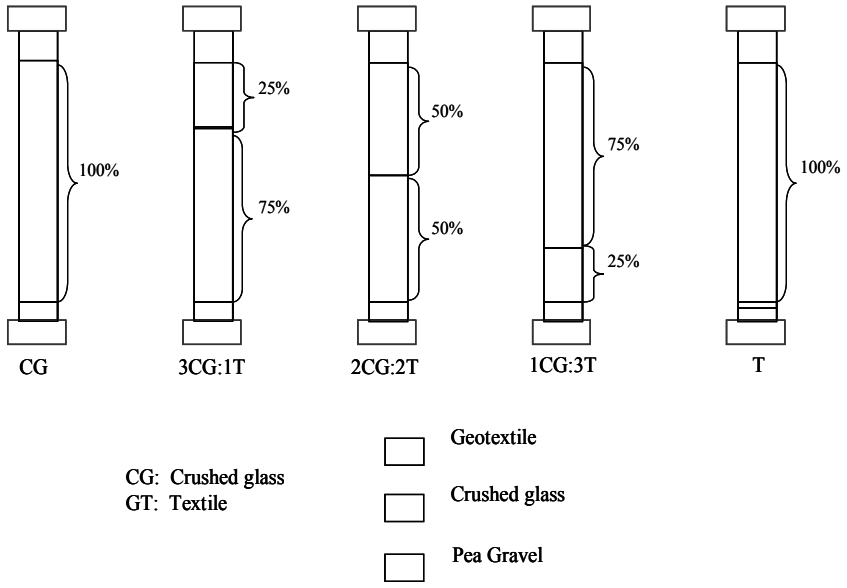

Figure 3-3. Biofilter columns different media combinations. (Source Hu, 2006)

Biofilter columns (Ø 2.5 cm, total length 24 cm) were filled with filtermedia (15 cm of the total length). During the first trial both crushed glass (CG) and geotextile (GT) were used as single filter media but also as combinations (dual filter media) in a following way:

- 1. column: 100 % crushed glass (CG:GT 4:0),
- 2. column: 75% crushed glass and 25 % geotextile (CG:GT 3:1),
- 3. column: 50% crushed glass and 50 % geotextile (CG:GT 2:2),
- 4. column: 25% crushed glass and 75 % geotextile (CG:GT 1:3), and
- 5. column :100 % geotextile (CG:GT 0:4).

Volume ratios for different columns are presented in Figure 3-3. Different filtermedia combinations were selected based on the previous studies by Farzana (2008). Filter media was supported in the column by bottom gravel bed which helped to avoid clogging of the columns. Air circulations and aerobic conditions within the filter beds were secured by 0.5 cm-diameter holes on the caps of the columns. Columns were covered with aluminium foil to avoid the contact with sunlight and so to prevent photosynthesis process if any algae should grow in the system. Calculations regarding geotextile filter volume are presented in Appendix II.

Like the filter columns all the tanks were also covered with aluminium foil and both the septic tanks and recirculating tanks were sealed with parafilm and foil to simulate the conditions of the field plant. Water balance and loading rates for the system were calculated using experiences from previous studies and following USEPA (2002e) recommendation. Calculations about water balance in detail are presented in Appendix II. RBF system was operated in a room temperature which was measured to be between 21 and 23ºC during the experiments.

As mentioned RBF system was examined during two different trials. During the first trial the systems (all five parallel treatment lines) were fed with raw wastewater[3]. All treatment lines were installed with different biofilter column as they are presented in Figure 3-3. The purpose of the first trial was to first study biofilm growth and observe the time for the biofilters to reach a steady stage (ammonia removal is steady). Second purpose was to study nitrogen removal efficiency of different biofilter media. Biofilm growth was monitored by analysing the concentration of ammonia both from system influent and systems effluent. When according to the laboratory analysis, ammonia concentration of the effluent was less than 10 mg/l**,** it was assumed that biofilm had developed into the filter media and that the filter was nitrifying.

Based on the results of the first trial one filter media (100 percent of crushed glass, (CG:GT 4:0)) was selected for the second trial. During the second trial similar filter media was used in all six biofilter columns. Crushed glass filter was selected since it appeared that it removed ammonia most efficient during the first trial.

During the second trial first all the six treatment lines were fed with raw wastewater to grow biofilm to the filter media. When biofilters reached the steady stage raw wastewater was changed to treated wastewater[4] and pharmaceutical feeding to the system started. Treated wastewater had a lower amount of solids which made it easier to run the bench scale system (less clogging). However according to the laboratory analyses, ammonia level of the influent stayed on the same level during both trials (see Figure 3-6). The first two treatment lines (biofilter columns 1. and 2.) were fed with mixture of treated wastewater and ethanol that included gemfibrozil. The second two treatment lines (columns 3. and 4.) were fed with treated wastewater and ethanol that included a mixture of four different pharmaceuticals: gemfibrozil, naproxen, diclofenac and ibuprofen. The last two treatment lines (columns 5. and 6.) were used as control filters and these lines were fed only with treated wastewater.

Pharmaceutical solutions were made in methanol since the solutions were above solubility of these drugs. The final concentration of pharmaceuticals in the wastewater was 200 µg/l. It should be noted that concentration used in this research is higher than concentrations that have been observed in field studies. Higher concentration was used to make sure that it was possible to track pharmaceuticals using available pharmaceutical analyses.

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[3] Wastewater was gathered from Mill Cove Wastewater Treatment Plant Bedford, Nova Scotia, Canada.

[4] Treated wastewater was gathered from Mill Cove Wastewater Treatment Plant Bedford, Nova Scotia, Canada.

The raw wastewater used in the experiments was gathered from Mill Cove Wastewater Treatment plant during three separate times and kept refrigerated. Samples were taken during the first trial from influent, recirculating tanks and effluent tanks and they were analysed for pH, ammonia (NH_4^+-N), nitrate nitrogen (NO_3^--N), total nitrogen (TN) (which is the summation of ammonia nitrate and organic nitrogen). The first trial lasted 16 days and during that time samples were taken six times. The second trial lasted 24 days and during that time samples were taken nine times.

Samples were analysed using standard methods. Ammonia was measured with 8038, HACH DR/2500 Spectrophometer. Nitrate nitrogen was measured with method 8039, HACH DR/2500 Spectrophometer. Total nitrogen was measured with 10071, HACH DR/2500 Spectrophometer (HACH DR/2010 Spectrophometer Handbook). The pH was measured by the Orion Model 230A pH meter.

In addition to the analyses that were done during the first trial, during the second trial the concentrations of four pharmaceuticals were analysed both from the influent and the effluent regarding all six parallel treatment lines. Analyses of pharmaceuticals were done for five samples sets. Pharmaceuticals were analysed using Solid Phase Extraction Procedure (Krkosek, 2006). All laboratory analyses were performed at the Dalhousie University Centre for water Resources Laboratory.

3.3 The results of RBF bench scale system experiments

3.3.1 Ammonia removal efficiency using different biofilter media's

During the first trial it was examined how different biofilter media could remove ammonia. According to the laboratory analyses RBF bench scale system reduced ammonia effectively. During the experimentation the ammonia concentration was higher in raw wastewater and in recirculation tank than in system effluent. After 16 days, ammonia concentration in RBF effluent in the Column CG:GT 4:0 was close to 0,0 mg/l and the biofilter had reached a steady stage. Other columns showed similar results except the Column CG:GT 0:4 that removed ammonia only slightly.

Figure 3-4 shows ammonia concentration variation in time for the raw wastewater (system influent) and for the system effluent from the biofilter column CG:GT 4:0.

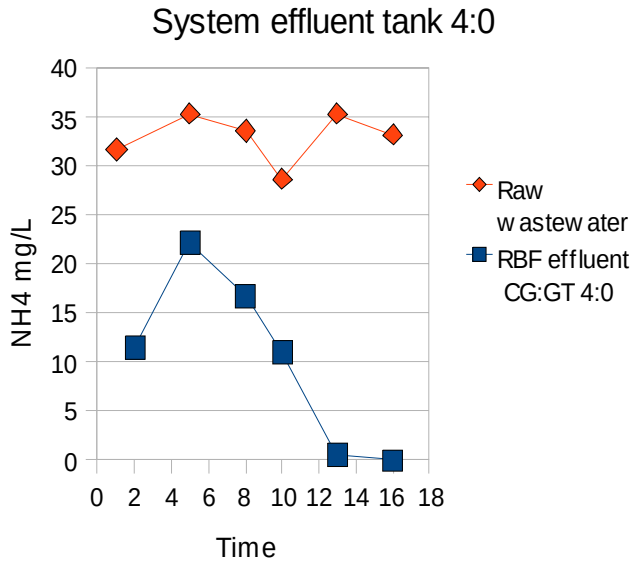

Figure 3-4. Ammonia concentration for the system influent and for the systems effluent from the biofilter column 100 % crushed glass.

Concentration of nitrate increased in time in all column systems except in the Column CG:GT 0:4. This indicates that when biofilters started nitrification and as ammonia was converted to nitrate the concentration of ammonia reduced and concentration of nitrate increased. Since the Column CG:GT 0:4 was not nitrifying, the concentration of nitrate stayed low.

Total nitrogen (TN) concentration varied during the first trial both in raw wastewater and in all samples taken from recirculating- and RBF system effluent tanks. The total amount of nitrogen compounds should remain the same before and after nitrification reaction. This means that the concentrations of TN should be similar in both effluent and influent. However results shows that TN concentration reduces when wastewater has been treated in the system. This indicates denitrification; nitrate was converted to nitrogen gas (N_2) that escaped the system through ventilation holes of the column caps. Denitrification is a benefit for the RBF systems since even though ammonia removal has been considered to be most important, reduction in nitrate reduces eutrophication in the receiving water system. Figure 3-5 presents TN concentrations of influent and effluent for the column CG:GT 4:0.

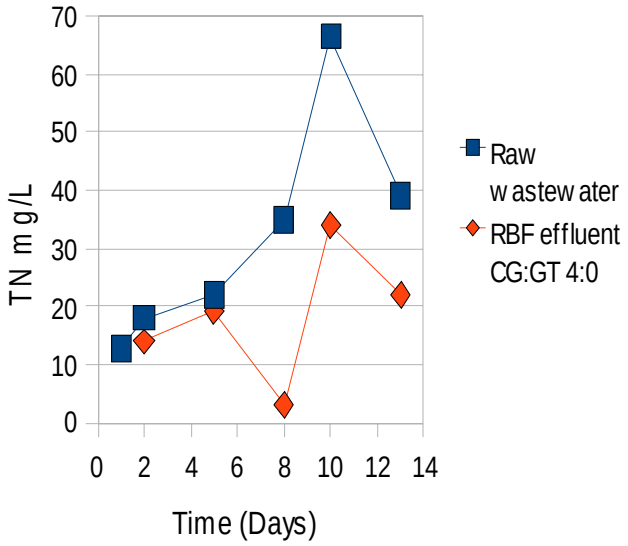

Figure 3-5. Total nitrogen concentration of the system influent and effluent from the biofilter column 100 % crushed glass

pH varied during the first trial in raw wastewater, recirculating tank effluent and in RBF system effluent. All the columns showed similar results. After couple of days pH was slightly lower in RBF system effluent tanks than in recirculating tanks. This also indicates nitrification since when ammonia is converted to nitrate H^+ ions are released and these ions make water more acidic.

3.3.2 Pharmaceutical removal efficiency

During the second trial pharmaceuticals were added to treated wastewater and the mixture was fed to the system. Gemfibrozil was added to the first two Columns (1. and 2.), and a mixture of gemfibrozil, naproxen, diclofenac and ibuprofen was added to the Columns 3. and 4. Columns 5. and 6. were feed with treated wastewater without additional pharmaceuticals.

Ammonia concentration was analysed as it was during the first trial. Figure 3-6 presents ammonia removal of the first column that was filled with 100 percent cursed glass during both trials. The red arrow shows the time when second trial started and pharmaceuticals were started to add to the system. To this column only gemfibrozil was added. As it shows in the Figure 3-6 the concentration of ammonia in the influent seems to be higher that it is in the system effluent. The same figure shows that the concentration of ammonia in the influent varied during the experiment time. However we cannot see a great difference between concentrations of ammonia during the first trial when raw wastewater was used and during the second trial when treated wastewater was used.

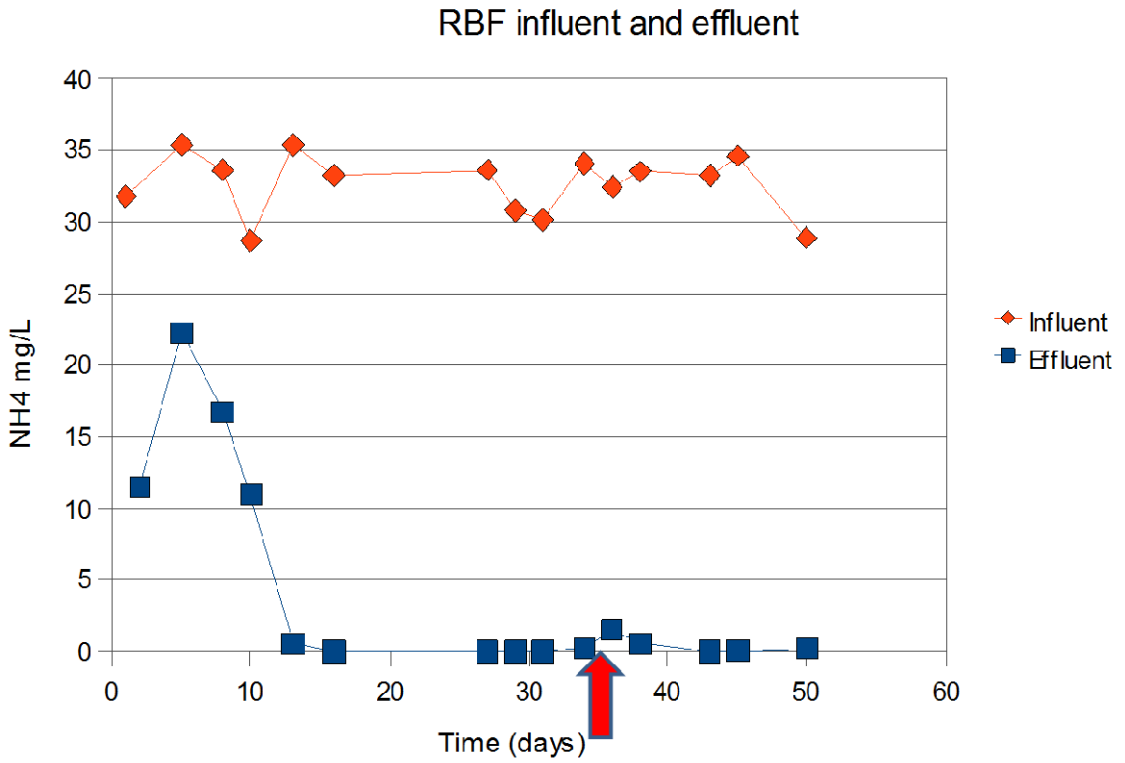

Figure 3-6. Ammonia concentration of influent and effluent in the biofilter that was filled with only crushed glass and the time line for first and second trial

The other biofilters showed similar behaviour regarding ammonia removal. This gives indications that nitrification was in process in all biofilter columns.

According to the laboratory analyses the average removal of gemfibrozil, naproxen and ibuprofen was more than 90 percent. Diclofenac was removed by average approximately 70 percent.

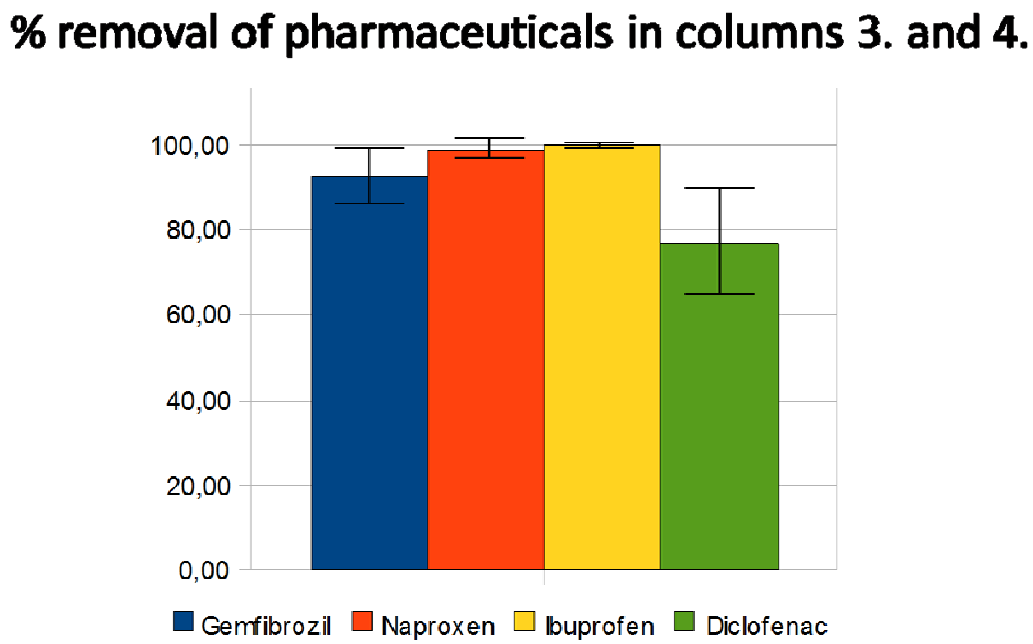

Figure 3-7. % removal of gemfibrozil, naproxen, ibuprofen and diclofenac in the biofilter columns 3. and 4.

Regarding both ammonia removal and pharmaceutical removal, statistical analyses were performed. Paired t-test was used to compare the average influent and effluent concentrations. Statistical analysis demonstrated (with 95 percent confidence) that there was a difference in the influent and effluent ammonia concentrations regarding all six parallel treatment lines. The paired t-test regarding pharmaceutical reduction demonstrated (with 95 percent confidence) that there was a difference in the influent and effluent pharmaceutical concentrations regarding all five examined pharmaceuticals. Also a 95 percent confidence interval was constructed and results are presented in the Figure 3-7. The observed reductions in pharmaceuticals are statistically significant.

4 Discussion

Several studies prove that there are more than 80 different pharmaceuticals and pharmaceutical metabolites found in water sources and wastewater influents and effluents (Herberer, 2002). Even though current understanding among the scientific community seems to be that pharmaceuticals do not have an acute affect on human health (Breitholtz & Bengtsson, 2006; Schwab & al. 2005; Christensen, 1998), there are indications that these pollutants are harmful for other organisms. Studies have found that these chemicals have an effect on some aquatic organisms for example fish (Gross-Sorokin, Roast, & Brighty, 2006) and that some aquatic species are likely to accumulate pharmaceuticals (Redshaw & al., 2008; Gagné, 2007). There seems to be consensus that there is uncertainty regarding the fate of pharmaceuticals in the environment, the effects of these substances on lower trophic level organisms, the long term effects regarding both humans and the environment, and what are the effects of mixtures of different Pharmaceuticals and Personal Care Products (PPCPs) (Enick & Moore, 2007; National Water Research Institute (NWRI), 2007; Cleuvers, 2002).

We have procedures like Environmental Risk Assessment (ERA) to assess environmental risks of pharmaceuticals, however they need to be further developed (Environmental Risk Assessment of Pharmaceuticals (ERAPharm), 2008) since at the moment ERAs include various assumptions. For example the ERA procedure looks into acute effects of substances; however we should also be interested in chronic effects. To compensate for this, different assessment factors are used during the ERA process. Here the question arises: Do these assessment factors represent our values? How should we interpret these assessments? Bodansky (1994) has stated:

> *"Risk assessment, unlike the precautionary principle, generally assumes that we can quantify and compare risks. It is information intensive and rational. Moreover, it can and often does take a neutral attitude towards uncertainty. (..) In contrast, the precautionary principle is not neutral towards uncertainty—it is biased in favor of safety."*

Even though risk assessment is "*information intensive and rational*" we have to deal with data gaps mentioned before. Tallacchini (2005) points out that since there are so many uncertainties regarding ERA:

> *"The prospect inherent in the precautionary principle tends to reduce as much as possible the mistakes that produce risks for people, considering that it is better to make a mistake harmful to the economy – a mistake that limits development not risky in itself – but not harmful to people".*

It seems that even though we do not have full scientific proof about the risk level that pharmaceutical residues represent in the aquatic environment, governments are implementing the precautionary principle and they are already looking into different risk

management alternatives (NWRI, 2007; Assessment of Technologies for the Removal of Pharmaceuticals and Personal Care Products in Sewage and Drinking Water Facilities to Improve the Indirect Potable Water Reuse (POSEIDON), 2004). In Canada a list of priorities has been developed that guides of research regarding PPCPs and an environment. This list contains 1) suggestions of what issues should be considered when effects of PPCPs are examined, 2) proposals for developing a monitoring network and 3) proposals for developing approaches for risk management.

Efficient risk management includes both preventative and treatment alternatives. Further, these two alternatives include various different measures. In this study preventative measures like source control, source separation and raising awareness have been discussed. These measures can be implemented using different management tools. For example, source control can be implemented by reducing pharmaceutical consumption or providing take-back programs for pharmaceutical residues. Since it is impossible to prevent pollution fully, efficient risk management includes also treatment alternatives. Regarding pharmaceuticals in wastewater it is quite natural to look into our current infrastructure and to examine how we can use our already existing wastewater treatment facilities when dealing with these compounds. Various different studies has addressed how effective our conventional centralised wastewater treatment plants are reducing pharmaceuticals (e.g. Lishman, L. & al. 2006; Servos & al. 2005; PODEIDON, 2004; Metcalfe & al. 2003). The results have been encouraging since some pharmaceuticals can be removed or the amount can be reduced with conventional treatment.

Wastewater can be treated in centralised units or in smaller scale onsite treatment plants. Figure 4-1 presents different risk management approaches. To be able to gain deeper understanding about the risk management choices that we have we need to also look into decentralised wastewater treatment. It seems however that regarding PPCPs this subject has been less researched than centralised systems.

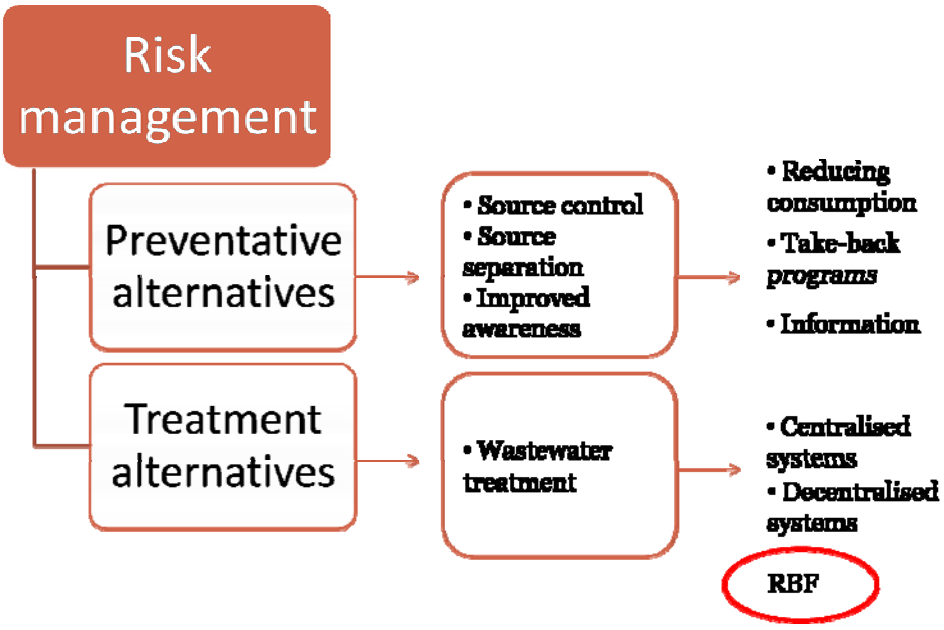

Figure 4-1. Different risk management approaches.

Because there was no information available about decentralised system's capacity to remove pharmaceuticals, it was necessary to research this area by experiments. To be able to determine the role that decentralised wastewater systems can have regarding risk management, the pharmaceutical removal effectives of Recirculating Biofilter (RBF) was evaluated. The main objective of laboratory tests was to study if a bench scale nitrifying biofilter is capable of removing four selected pharmaceuticals (gemfibrozil, naproxen, ibuprofen and diclofenac). During the experiments both ammonia removal and pharmaceutical removal were observed.

During the first trial ammonia, removal of five different filter materials was compared. According to laboratory analyses the average concentration of ammonia in the influent was 32 mg/l. After the system reached the steady stage (16 days from the start up), the average concentration of the ammonia in the effluent for the filter column that was 100 percent crushed glass CG:GT 4:0 was 0,28 mg/l achieving 99 percent removal efficiency. This result is consistent with Farzana (2008), Hu & Gagnon, (2006) and USEPA (2002e). The biofilter column CG:GT 4:0 showed slightly better ammonia removal results during the first trial than the other filters. The filter column that had only geotextile (CG:GT=0:4) as filter material did not reach the steady stage as fast as others since after 16 days there were no indications of nitrification.

Based on the results of the first trial, crushed glass was used as the biofilter media during the second trial. Six biofilters were dosed with the mixture of treated wastewater and pharmaceuticals. According to laboratory analyses, ibuprofen, gemfibrozil and naproxen were removed more than 90 percent. Diclofenac was removed approximately with 70 percent efficiency.

It seems that pharmaceuticals studied in this thesis are not studied in RBF systems before. However there are studies that have been looking into how centralised treatment plants are capable of reducing these substances. Also some other laboratory studies have been done.

According to research performed by the EU project: POSEIDON (2004), in nutrient removing plants it was possible to remove more than 95 percent of ibuprofen. Lishman & al.(2006) reported 91-98 percent removal efficiency in centralised treatment plants for ibuprofen and 79-98 percent removal efficiency for naproxen and in a number of cases it was not possible to measure concentration of these two from effluent indicating 100 percent removal. Reif & al. (2007) reported similar removal results regarding ibuprofen and naproxen (more than 90 percent removal) by using pilot scale membrane bioreactor (MBR).

According to Reif & al. (2007) they reached less than 10 percent removal of diclofenac by using pilot scale MBR. Lishman & al.(2006) reported that gemfibrozil median reduction was 66 present and data for diclofenac showed several negative reduction values the median reduction being – 34 percent regarding centralised WWTP. These negative results were not explained in the study but it might be that since it seems that diclofenac is very persistent it can bind to sediment particles and might then be released again to water when for example sludge is removed from the treatment system. Also there might be some problems when measuring influent and effluent since it can be difficult to be able to measure exactly the same particles that go through the systems. According to Heberer (2002) gemfibrozil had shown 17 percent removal efficiency in the municipal sludge treatment plant (STP) but he also states that Ternes (1998) has reported a removal of 69 percent for the same pharmaceutical in the STP.

Laboratory results of this thesis cannot be compared as such with results from previous studies and one must be careful when interpreting results. First of all the scale of the treatment systems can have an effect on how effective the removal of compounds is. Also the treatment technology can be different. However it can be said that it seems that reduction of ibuprofen and naproxen seems to follow the same scale in this research as it has been presented regarding centralised treatment systems. But regarding gemfibrozil and diclofenac there are differences between literature and laboratory results of this research. The removal of gemfibrozil seems to vary in the literature and laboratory results of this study are more close to results of Ternes (1998). Also the difference in diclofenac removal between previous studies and laboratory experiments for this research seems substantial and cannot be explained based on this research.

As stated, there are limitations when interpreting the results of the laboratory experiments. In laboratory environment conditions can be controlled and this can affect the results. For example experiments were performed in steady room temperature (21-23 °C) and it is known that nitrification process is sensitive to temperature (Metcalf & Eddy, 2003). This means that for example RBF nitrification performance on field conditions can be different and this might affect the pharmaceutical removal. Even though regarding literature

(POSEIDON, 2004; Servos & al., 2005) there are indications that treatment plants that remove nutrients also remove pharmaceuticals, based on this research it cannot be stated if there is a connection between ammonia removal and pharmaceutical removal. This means that it is not known if the biofilter would have removed pharmaceuticals also if nitrification would had been inhibited.

For laboratory experiments influent was gathered from a centralised treatment plant. It might be that wastewater characteristics are different in decentralised systems and so they can effect to the treatment results. Also because of the analysing methods it was necessary to use higher concentrations of pharmaceuticals in the influent than are reported in the literature. Therefore it might be that the removal efficiency is different with lower concentrations.

Even though, based on this research, it cannot be stated that decentralised systems have a similar ability to remove pharmaceuticals than centralised systems have, it seems that there is potential and further research is needed. Even though at the moment there are no treatment requirements for pharmaceuticals, it is possible that in the future treatment requirements will also include certain micro pollutants. If these requirements are going to be part of a policy it is important to know if we have technical possibilities to achieve requirements in a cost-efficient way.

If future studies show that decentralised systems have the same ability regarding pharmaceutical removal as centralised systems, it will be important for communities where decentralised systems are the cost-efficient option. It would mean, for example, that we could enhance the use of decentralised systems since the difference regarding pharmaceutical removal would not be substantial when compared to centralised systems. If however, future research shows that decentralised systems do not reach the same level in pharmaceutical removal and risk management is needed, preventative alternatives would have an important role regarding rural areas. This would mean that for example urine separation could be used as one of the management measures since this kind of a technology might be easier to apply in rural areas than in cities where we can have old and inflexible infrastructure. Regarding urine separation it should be noted that biological treatment needs nutrients to operate well and this should be considered when applying separation

If in the future, when we have a more specific picture about the risk level, it is necessary to apply risk management measures, it seems that in general, the most efficient way to use risk management tools is to combine preventative and treatment measures. Local conditions, treatment needs, costs, available technology etc., needs to be considered when risk management choices are selected.

When we have more knowledge about which pharmaceuticals we can treat from our wastewater, we can focus our prevention measures to most harmful substances that are not treatable. The pharmaceutical industry can look for alternative compounds or we can inform consumers about the impacts of the drug and highlight proper disposal.

It is also important to recognise the need to operate and manage wastewater treatment processes in a sufficient way regarding both centralised and decentralised systems. This seems to benefit nutrient and BOD as well as pharmaceutical removal. Since there seems to be a connection between pharmaceutical removal and nitrification, it might be possible to reduce the amount of pharmaceuticals in wastewater without specific reduction requirements targeted to pharmaceuticals. Instead we could still focus on nutrients. For example regarding wastewater and treated wastewater used for laboratory experiments for this thesis, there was no difference in ammonia concentrations. Knowing this we can state that there are problems regarding plants activated sludge process and the main focus for this plant leys somewhere else than in micro pollutants.

There are many factors that affect which pharmaceuticals and how much of them are found in wastewater. These include for example the volumes of that drugs that are used, use patterns, how well they metabolise, what kind of treatment technology is used and how much water is used. These factors are very different between different countries and so concentrations of pharmaceuticals in the wastewater can differ in different regions (Lishman & al., 2006; Metcalfe & al., 2003). This should be recognised when management alternatives and policy directions are assessed. One tool that works in Scandinavia might not be useful in Canada and so efficient risk management measures should be selected considering these differences and local conditions.

5 Conclusions and recommendations

5.1 Conclusions

We have scientific evidence that there are traces of pharmaceuticals in our groundwater, drinking water and watercourses. Since this discovery, it has been necessary to study if these substances are representing a threat to human health and to other organisms. Evaluation of the risk level needs to be done in a systematic and scientifically sufficient way understanding also the economic limitations and the context of the risk assessment. Risk assessment tools have been developed both in Europe and in North America and the objective of these tools is to help us to focus research and also risk management measures to the most critical environments, organisms and chemicals.

So far it is known that the present levels of pharmaceutical residues do not represent an acute threat to human health but there is a reason to believe that other organisms in the aquatic environment suffer from this pollution. What is not yet known is how these chemicals affect humans and the environment in the long run and what affects different combinations of different substances might have. Since there are many uncertainties regarding pharmaceuticals in the environment it is necessary to consider implementing the precautionary principle (PP). Adapting this approach means that when we interpret environmental risk assessments, we take into account the assumptions that are done when performing the assessment. When evaluating results we have to consider if the context of the assessment has been integrated into the assessment and does it represent the values like the PP. If the assessment does not include these, we should be aware of this when concluding which actions should be taken.

Applying the precautionary principle means that we cannot wait for full scientific proof on the level of the risk. It is important to first assess how we are able to prevent these compounds from reaching the environment, and second to assess measures to eliminate or treat these pollutants. Based on different choices we have, it is possible to act and to reduce the potential adverse effects and thereby to reduce the risk.

This study looks into different possibilities that we have regarding risk management of pharmaceutical residues in the wastewater and researching what role treatment and more specific decentralised wastewater treatment could have as a risk management tool. The overall goal for this research was to contribute to the general understanding regarding risk management measures for pharmaceuticals in the aquatic environment. To reach this objective, two research questions were tested:

1) How can we manage environmental risks regarding pharmaceutical residues in wastewater?

- What role can decentralised wastewater treatment have as a risk management alternative?

2) How effective is nitrifying Recirculating Biofilter (RBF) for reducing four selected pharmaceuticals from wastewater?

In this study different risk management measures have been assessed and both preventative and treatment approaches have been look into. Pollution prevention measures include for example raising awareness of all stakeholders about the environmental effects of pharmaceuticals. Different stakeholder groups need different types of information. For example in Sweden this has been acknowledged when models about drug categorisation have been developed. Targeting information to different groups, it is possible to activate them to act inside their own sector in a way that the risk that pharmaceutical residues represent is reduced. This means that the pharmaceutical industry is aware about the need to design pharmaceuticals that are less harmful to the environment and also to develop manufacturing processes so that less waste is produced. When it is reasonable and possible from the healthcare point of view, pharmacies and prescribers can consider choices that are less risky to the environment regarding prescriptions. Information that is focused for consumers points out the necessity of safe and sound pharmaceutical disposal. To support safe disposal, take-back programs for pharmaceutical residues have been implemented for example in Canada, Sweden and in Finland. Information about the harmful effects of pharmaceuticals and possibility to return pharmaceutical residues without extra fee can prevent consumers to dispose these chemicals by flushing them or mixing them with other domestic wastes.

Even if we prevent or, more realistically, reduce the unused pharmaceuticals ending up in our wastewater by controlling their disposal, those drugs that are consumed usually are excreted and so disposed with wastewater. Wastewater is a main entry for these pollutants to the environment. As a part of the prevention approach it is possible to separate highly concentrated wastewater produced for example by pharmaceutical industry, hospitals and nursing homes. Then we can treat this wastewater regarding pharmaceuticals before discharging it to the centralised drainage and wastewater treatment plant. Since urine contains most of the drugs that are excreted we can separate it and treat it before it is discharged to the treatment plant. To be able to treat more concentrated wastewaters, onsite facility is needed. Urine separation needs special toilets and new drainage structures. These might require high investments. On the other hand since urine contains also most of the nutrients ending up to wastewater, urine separation would benefit also nutrient removal.

In addition to the preventative approach treatment possibilities have been studied. Results from conventional centralised wastewater treatment plants show that certain pharmaceuticals can be removed or at least it is possible to reduce the amount of some pharmaceuticals with treatment. Chemical structure and certain treatment parameters effect how effective the removal is. In this study sludge retention time (SRT) and nitrification process have been discussed. Also some advanced technologies like ozonation and membrane technology have

been acknowledged. According to studies it seems that long SRT and a good nutrient removal increases degradation chances of certain pharmaceuticals. Also ozonation and membranes have been discovered to have a potential to reduce or eliminate such compounds. However it has been noted that ozonation and membrane technology can be expensive way to treat wastewater.

Despite the potential of centralised solutions, there is an inevitable fact that a considerable share of the population is not, and cannot, be connected to centralised systems. In these cases wastewater can be treated onsite in decentralised wastewater systems. Decentralised systems have various benefits when compared to centralised systems. These include, for example, reduced investments regarding drainage, possibility to recycle nutrients and water reuse, flexibility regarding population growth and in the case of operational failure, environmental problems are usually smaller when compared to centralised systems. Well maintained systems can reach high quality treatment regarding BOD and nutrients. However there was not information available about how decentralised systems can treat pharmaceuticals.

To be able to evaluate the role that decentralised wastewater treatment systems can have regarding risk management and pharmaceutical residues in aquatic environment, laboratory experiments were implemented. Research was implemented with a bench scale recirculating biofilter (RBF). Regarding conventional centralised waste water treatment systems (WWTS), it seems that if the treatment process includes nitrification (conversion of ammonia to nitrate) the process can also remove certain pharmaceuticals. Therefore during the experiments both ammonia removal and pharmaceutical removal were observed by analysing concentrations in system influent and system effluent.

Based on the results of laboratory experiments and statistical analysis it can be stated that RBF bench scale systems were able to remove both ammonia and four selected pharmaceuticals. Regarding the results it seems that small scale systems can also reduce pharmaceuticals from wastewater as centralised WWTSs. Also ammonia removal was highly effective. It seems that by using crushed glass as a filtermedia it is possible to remove almost all ammonia from wastewater. It also seems that geotextile as a filter media is not as effective as crushed glass. Analysed samples indicated that biofilter was also able to denitrify (convent nitrate to nitrogen gas). According to the laboratory analyses the average removal of gemfibrozil, naproxen and ibuprofen was more than 90 percent and diclofenac was removed by average little more than 70 percent.

Based on these experiments it is clear that it cannot be assessed if the removal efficiency is similar regarding field systems. More research is needed to determine this. Experiments show some similarities with the previous results from centralised systems which stated that if the system removes nutrients, also pharmaceutical degradation is possible. This indicates that it might also be possible to remove pharmaceuticals from wastewater with field RBF and in addition removal could be possible with other decentralised systems that remove ammonia. When interpreting experiment results it should be noted that there are limitations. The pharmaceutical concentrations that were used during experiment were higher than the

measured concentrations in wastewater usually are. It might be that if there are low concentrations of pharmaceuticals in the influent, the removal effectiveness will differ. Also, because during the experiments, treated wastewater and not raw wastewater was used (even though the ammonia level was the same), it might be that when there is more organic material in the water, the removal effectiveness differs.

At the moment there are no requirements for wastewater treatment regarding pharmaceutical residues but this might change in the future. Even though there can be requirements regarding pharmaceutical removal in the future, it should be noted that wastewater treatment currently, but also most probably in the future is focused on removing organic matter and nutrients and it would be extremely expensive to design and construct treatment systems that are targeted on certain micro pollutants. Therefore it is important to know the capacity of current treatment systems regarding pharmaceutical removal. Acknowledging the current capacity it is possible to target for example preventative measures more efficiently. This means that when we know what we can treat we can try to prevent those pollutants that we cannot treat, reaching our watercourses.

If field studies show that decentralised systems can have a similar ability to remove pharmaceuticals in wastewater than centralised systems, it means for example that it is not necessary to prefer centralised wastewater systems regarding pharmaceutical removal. This is important for communities with low investment abilities, special needs regarding water reuse and system flexibility. It is also important to note that linkage between nutrient removal and pharmaceutical removal highlights the importance of maintenance of the treatment system. This means that removal of both pollutants requires stable operation of the treatment system.

It seems that both in the European Union area and in North America research currently is focused on fate research, developing analytical methods and risk assessment measures as well as studying effects of different pharmaceuticals on the environment and human health. Also, development of different risk management measures is included. When research covers all the levels including risk assessment and risk management measures, the results benefit the whole sector. For example when we know that naproxen is effectively removed from wastewater using conventional treatment methods regarding both centralised and decentralised systems, it might not be then our main priority to study how this substance effect to aquatic organisms. When we learn more about the effects of different pharmaceuticals, their fate in the environment and possibilities to prevent or treat this pollution, we can select the most efficient risk management measures. This means that in the future we will have integrated solution for risk management including both preventative and treatment management approaches.

5.2 Recommendations

This research looked into the possibility to remove pharmaceuticals from wastewater using a nitrifying biofilter. This was done because there are indications that wastewater treatment

systems that are capable of ammonia removal can also reduce pharmaceuticals in the wastewater. Based on this research it cannot be stated if ammonia removal and pharmaceutical removal have a connection. The next step regarding laboratory experiments would be to study if removal of pharmaceuticals appears also when nitrification is inhibited in the biofilter. This way it might be possible to gain a deeper understanding about the pharmaceutical removal process. When we have a fuller understanding about the mechanisms and parameters that influence the reduction process it is possible to design more efficient treatment systems or develop the current systems so that we use them with maximum capacity regarding micro pollutants.

To be able to evaluate further the role that decentralised systems can have as risk management measures regarding pharmaceuticals and the environment, it is necessary to implement field studies. These studies should include analysing pharmaceutical removal effectives of different decentralised applications. This is important since it might be that different technologies can have a different capability to remove pharmaceuticals. Also when we have more knowledge about which systems represent the best practice in this field, it is easier for both authorities and property owners to choose a treatment system that is needed depending of the location of the treatment plant and the sensitivity of the environment.

Phosphorous removal is included for example in Sweden and in Finland to the treatment requirements for decentralised small scale systems. This is because phosphorus is one source for eutrophication in natural water systems. Therefore, in addition, it would be important to study the RBFs phosphorous removal efficiency since it seems that it has not been researched.

From a policy development point of view in current situation, it would be important to clarify how to interpret risk assessment results. It is important to develop systematic procedures for risk assessment but as important it is to have a commonly agreed system how to include values like the precautionary principle to the environmental risk assessment. Policy developers should be provided training and information about the risk management procedures and weaknesses that they include. On the other hand harmonised and further developed methods for risk assessment (as they are developed in the EU for example) will help the interpretation in the future.

Cost-effectives assessment regarding different risk management measures would offer a good base for policy directions in the future. In the future when we have more complete picture about the level of the risks that pharmaceutical residues in the environment represent, we can choose the most cost-effective measures to manage these risks. It is also important to study what is the capacity of our current infrastructure regarding micro pollutant removal. It might be that the most cost effective manner to reduce the risk is to first of all use our present treatment systems in a way that we reach the best level regarding treatment results in nutrient removal and second to select the most cost-effective ways to prevent pharmaceutical residues reaching wastewater.

Identification of the different structures like reducing pharmaceutical consumption (the use of a preventative approach), would offer better understanding how to focus risk management. Research should touch all the stakeholders and structures that can affect this issue. This means that stakeholders from the pharmaceutical industry to the single consumer as well as political and economical structures that support pharmaceutical used should be indentified and assessed. Identification and assessment should be implemented considering local and national conditions.

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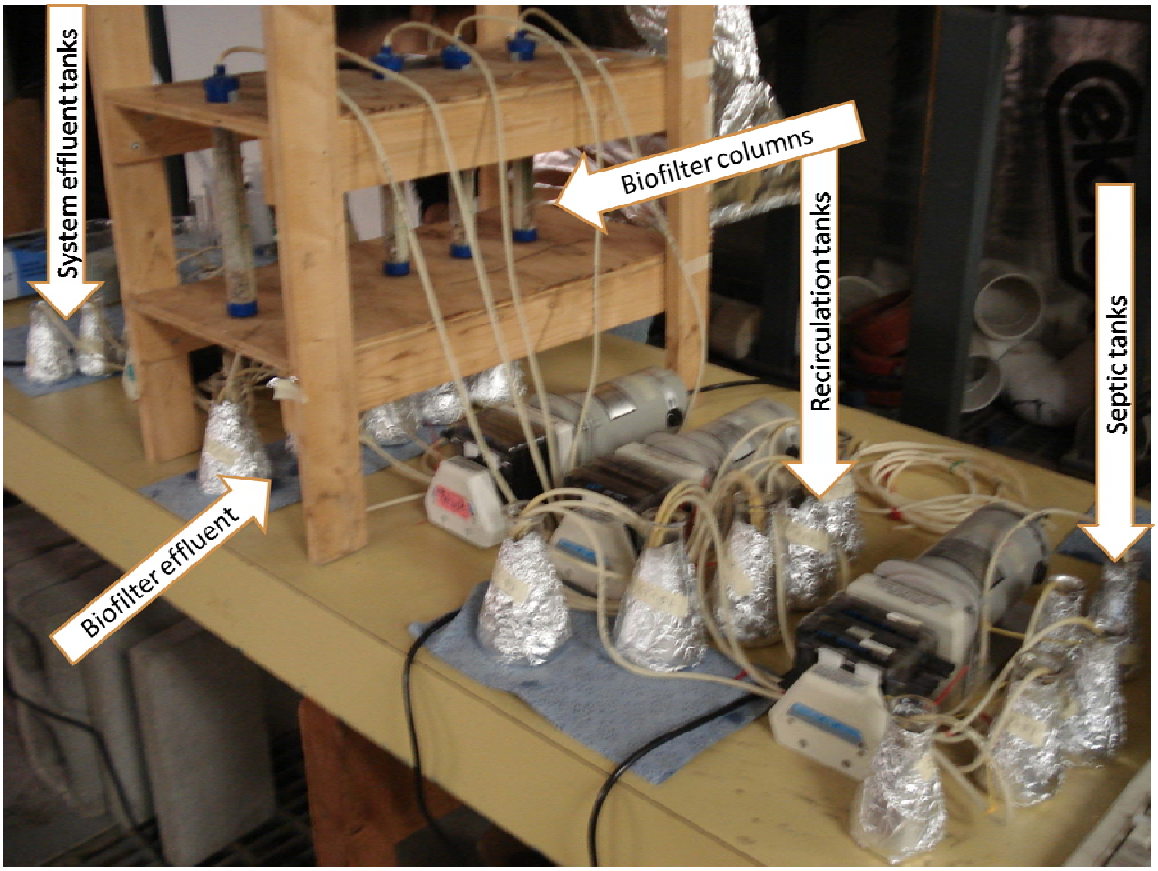
Appendix I

Bench scale setup



Appendix II

Experimental set-up for bench scale RBF

1 Water Balance and Loading Rates Determination

The flow rate for the RBFs was determined by:

$$Q = HLR * A = (0.20\ m^3/m^2/d*(\pi*0.01252) = 0.000098174\ m^3/d \rightarrow 98.2\ ml/d$$

Where,

Q is the flow rate of septic tank effluent, ml/day,

HLR is selected hydraulic loading rate 0.20 $m^3/m^2/d$ * and

A is the filter column cross sectional area, m^2.

Q Septic Tank = 98 ml/d

According to USEPA (2002) recirculation tank is sized equal to 1.5 times the Q Septic Tank

Q Recirculation Tank = 1.5 * [Q Septic Tank]

= 1.5 * 98 ml/d

= 147 ml/d

Specifications

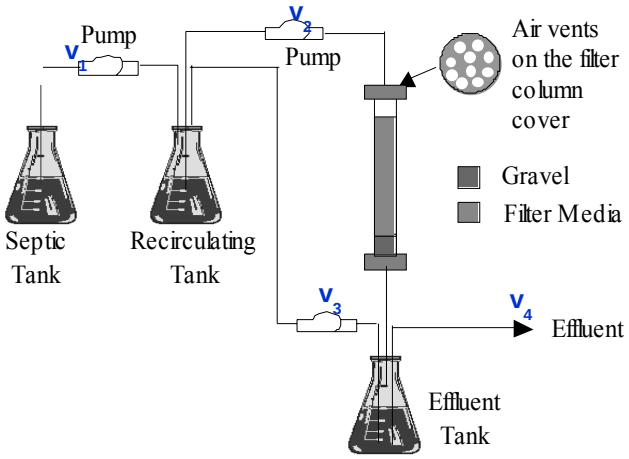
HLR = 0.20 $m^3/m^2/d$

Column Diameter = 25 mm

Column Length = 23 cm

Recycle Ratio = 4:1

Dosing Frequency = 96 times/day



* HLR follows USEPA recommendations

Volume per dose into the filter bed from the recirculation tank, mL/dose

$$V\ Dose = \frac{Q*(RecycleRatio+1)}{DosingFrequency}$$

=5.1 ml/dose ≈ 5 ml/dose

Volume dose for Septic Tank effluent, $m^3/m^2/day$: v_1 = Design HLR/Dosing Frequency

Volume dose for Recirculating Tank effluent, $m^3/m^2/day$: v_2 = (Recycle Ratio + 1)*v_1

Volume dose for Recycled Filter effluent, $m^3/m^2/day$: v_3 = (Recycle Ratio)*v_1

Volume dose for RBF effluent, $m^3/m^2/day$: $v_4 = v_1$

v_1= Designed HLR

= 98 $m^3/m^2/d$ /96 dose/day

= 1,0 $m^3/m^2/d$

= 1 ml/dose

v_2 = (Recycle Ratio + 1) * v_1

= (4+1) * 1 ml/dose

= 5 ml/dose

v_3 = Recycle ratio * v_1

= (4/1) * 1ml/dose

= 4 ml/dose

$v_4 = v_1$ = 1 ml/dose

2 Steps to fill the biofilter media columns with geotextile

A) Determine the volume filled by geotextile

Geotextile volume = Column total volume * (1-determined porosity)

B) Determine the mass of geotextile

Mass of geotextile = geotextile * geotextile density

C) Fill column with determined mass of geotextile evenly

Mass of the geotextile for the column filled 100 % with geotextile =

Column total volume * (1-determined porosity) * geotextile density

= 0,15 m * (π*0.01252 m^2)* (1-0.90) * 100 kg/m^3= 0.0007362 kg ≈ 736 g

Mass of the geotextile for column 3:1 = 25 % * 736 g = 184 g

Mass of the geotextile for column 2:2 = 50 % * 736 g = 368 g

Mass of the geotextile for column 1:3 = 75 % * 736 g = 552 g

Specifications

Column Diameter = 2.5 cm

Total Filter Length = 15 cm

Geotextile porosity = 0.90

Geotextile density = 100 kg/m^3