Hazardous Substances in Wastewater Management

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Doctoral thesis

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vilka tror vi att vi är
vi ljuger blint för dom vi älskar
och glömmer bort dom få vi känner
och skryter om hur bra vi mår
tror att nån bryr sig hur det går

vilka tror vi att vi är?
vilka tror vi att vi är?
här famlar vi omkring
och fattar ingenting
vilka tror vi att vi är?

Bo Kaspers Orkester

Preface

To me, creating a doctoral thesis has been one of my more extraordinary projects so far. To practice research with all it involves has left various impressions: it is interesting, creative, educational, enjoyable, and (more or less painfully) progressive. It has certainly given me insights into a variety of subjects as well as into myself; it is challenging, frustrating, stressful, complicated, and sometimes very eccentric. I have been writing, rewriting and re-rewriting – new texts and old texts. And some calculations. Sometimes digging deep into details, but more often a search for context – a sheer necessity when you are completely lost in complexity. And last, but not least – the few, but golden seconds of aha-experiences.

However, none of this work would have seen the daylight without the fantastic network of people who surrounds and supports me – colleagues, family, friends, and dogs(!).

Financial funding was manly provided by the Foundation for Strategic Environmental Research (Mistra), but also by Luleå University of Technology, the Swedish Research Council for Environment, Agricultural Sciences and Spatial Planning (Formas), the Swedish Water and Wastewater Association (VA-Forsk), and the Stockholm Water Company (Stockholm Vatten). Their support is gratefully acknowledged.

The work has been carried out at the Division of Sanitary Engineering at Luleå University of Technology between the years 1999 and 2004. I would like to express my special gratitude to my supervisor, Prof. Jörgen Hanæus, for his support and advice throughout my doctoral period. With a profound presence, Jörgen reads, listens, and reflects on the subject matter, and he reflects and focuses on the personal progress in large ('the process'), which has been very encouraging for me as a PhD-student. I have had the pleasure to cooperate with my assisting supervisor, Ass. Prof. Per-Arne Malmqvist, who also acted as co-author in paper VII included in this thesis. Per-Arne has in particular cared for the progress of making the results operational, sometimes a difficult task, but which has also been the excellent driving force in inspiring me to search for new approaches. His experience and personal qualities have improved and enriched my struggles with this thesis.

I have had the great opportunity to attend the Swedish research program Sustainable Urban Water Management (Urban Water), supported by Mistra, from 1999 to 2005. Besides their financial support, which is gratefully acknowledged, participation in Urban Water provided me a unique network of researchers and professionals in the urban water and wastewater sector, not only in Sweden, but also internationally. I would like to especially acknowledge PhD Henriette Söderberg, programme coordinator of Urban Water, not only for being an excellent guide in the Urban Water 'Circus', but also for her personal and professional qualities with which she serves as a terrific role model in the academic world. Furthermore, it has been an exclusive and exciting experience to be one of the seventeen doctoral students in the Urban Water research school. The common aims and the arena for cooperation and discussions within the Urban Water programme have provided me much motivation and inspiration and not the least of which, many new friends.

Sincere gratitude is expressed to Ass. Prof. Håkan Jönsson and PhD Björn Vinnerås at the Swedish University of Agricultural Sciences, for their cooperation and valuable knowledge in the agricultural branch of wastewater engineering. Without Håkan's enthusiasm and driving force, some of this work would have been unachievable (the Gebers measurements were quite an extraordinary experience!). Håkan also acted as co-author in two of the papers included in this thesis. My collaboration with Björn (co-author in paper III) not only made me sharpen my arguing skills, but also inspired me to dig deeper into the wastewater fractions. I would also like to express gratitude to Professor Marcus Boller and PhD Wouter Pronk, at the Swiss Federal Research Institute for Environmental Science and Technology (EAWAG) in Zürich, who welcomed and introduced me to the friendly and inspiring atmosphere at the EAWAG during several months in 2003. Wouter coached me in my lab work and contributed to this work with his wide knowledge about membrane technology (co-author in paper VIII).

I am grateful to all my dear colleagues at the Division of Sanitary Engineering for the great spirit of community and for the many interesting discussions – in great as in little things. You made my stay at the division a joyful and memorable time. I am also appreciative to the staff at the Department of Civil and Environmental Engineering for all academic support and the nice fika room chitchat.

Roland Palmquist (my father) is very much appreciated for his nice illustration on the cover. Wayne Chan is sincerely acknowledged for making the English language more fluent and correct.

Many thanks to my parents, Karin and Roland Palmquist, who equipped me with enough confidence and courage, as well as to the rest of my lovely ‘old’ and ‘new’ family members for their great support throughout this work. I am also grateful to all my dear friends who have seen to it I have not neglected the other needs in life, such as skiing…

Finally I would like to thank my dear Johan and our Hannibal for being my greatest support! You gave me distance and rest from research, which helped me in regaining energy, motivation, and to find a sound balance in life and work.

Luleå, December 2004

Helena Palmquist

Abstract

The extensive use of materials and substances in society causes diffuse source emissions that lead to uncontrolled spreading of hazardous substances, largely channelled via wastewater systems, to the surrounding nature. Complex mixtures of substances appear in wastewater as a result of use, wear, and corrosion of goods (e.g. pipes, taps, carpets, furniture) as well as the use of household chemicals from doing the laundry and dishwashing and the use of pharmaceuticals and personal care products. As many as 30,000 substances regarded as everyday chemicals are regularly used in households, implying that the flows of hazardous substances in wastewater systems are not only a complex issue for wastewater management, but for society as a whole.

As a part in analysing the flows of hazardous substances in wastewater systems, domestic wastewater fractions (greywater, urine, and faeces) were chemically characterised through full-scale field samplings at the source separating domestic wastewater systems Vibyåsen and Gebers with respect to a selected number of hazardous substances. Data on the characteristics of wastewater fractions were essential to improve the prerequisites for performing substance flow analysis and chemical risk assessment of the wastewater systems.

The mass flows of hazardous metals from households emerged in similar quantities in the greywater and toilet fractions. However, ratios of hazardous metals to phosphorus and nitrogen were significantly lower in the urine than in the faecal matter and greywater. The mass flows of organic hazardous substances from households were mainly searched for in the greywater, resulting in 50-60% of the 81 measured substances being found, with representatives from all of the substance groups investigated. Of the 72 measured organic hazardous substances, 36% were found in the blackwater at Vibyåsen.

However, it was not possible to exactly identify their specific sources as the mass flows of organic hazardous substances derive from diffuse household sources like everyday activities (laundry, cleaning, etc.), the wear of things such as pipe material and interior fittings, and from airborne deposition. The input of organic hazardous substances to urine and faeces occurs mainly via the excretion of, for instance, pharmaceuticals, pesticides, and food additives.

Other examples of relevant pathways are when emptying a scouring pail and throwing in cigarette butts, snuff, etc., into the blackwater via the water closet.

Based on a number of recent measurements (including Vibyåsen and Gebers) a proposal for new Swedish design values for nutrients (e.g. phosphorus and nitrogen) and seven heavy metals (e.g. copper and cadmium) in household wastewater fractions was put forward. However, the consumption patterns of society changes over time, and with it, so do the wastewater characteristics. Therefore, design values should be used with good judgement and require regular updating, assumingly each 5th to 10th year.

A possible management approach was suggested to interpret and compare different wastewater systems, and to serve to find out if and how much the flow of hazardous substances can be stopped, diverged, or transformed at the source or during transport throughout the wastewater system. The barriers approach was proposed as a tool on a conceptual level (a way of thinking) as an attempt to support a shift in perspectives by combining a traditional end-of-pipe perspective with more systems-oriented perspectives, thereby linking the use of resources and the spreading of hazardous substances to their underlying causes and driving forces (i.e. consumption and lifestyle) rather than only focusing on the emissions. Organisational and behavioural barriers, system design, process barriers, and optional recipients were suggested, implying that various kinds of measures are needed in the management of hazardous substances to achieve a change in direction towards sustainability.

Sammanfattning

Målet med avhandlingen är att analysera flödet av kemiska substanser i samhället och dess gränssnitt till avloppssystemen. Idag existerar ca 100 000 ämnen varav 30 000 ämnen är betraktade som vardagligt använda. Vissa av dessa har konstaterad negativ påverkan på människa och miljö. De allmänt tillgängliga kunskaperna om de existerande ämnenas egenskaper och användning är dock överlag bristfälliga, men man kan konstatera att avloppssystemens funktion medför en koncentrering och transport av samhällets ämnesflöden till omgivande miljö. Samhällets ämnesomsättning (metabolism) beror framförallt på konsumtion av varor och produkter, vittring från byggmaterial och infrastruktur, trafik, industriella processer samt nedfall av luftburna föroreningar.

Under de senaste decennierna har fokus alltmer flyttats från punktkällor (t.ex. industrier) till diffusa källor (t.ex. trafik och hushåll) för att finna och åtgärda miljöstörande utsläpp till miljön. Det har visat sig att hushållen bidrar med en betydande del av farliga metalliska och organiska föreningar till avloppsvatten. Med syfte att spåra källor till kemiska substanser från hushåll gjordes mätningar på fraktionerat hushållsspillvatten – gråvatten, urin, fekalier och i förekommande fall äv. klosettvatten (s.k. svartvatten) – i de källsorterande avloppssystemen i Vibyåsen och Gebers.

Mätningarna visade att metallflödena från hushåll var i samma storleksordning till både gråvatten och toalettavlopp med undantag för urin. Kvoten mellan metall och närsalterna fosfor och kväve var betydligt lägre i urin än i fekalier och gråvatten. Massflödena av organiska substanser undersöktes främst i gråvatten, där 50-60% av de 81 substanserna detekterades med representanter från samtliga analyserade ämnesgrupper. 36% av de 72 analyserade organiska substanserna hittades i svartvatten. Utifrån mätningarna förslogs dessutom s.k. schablonvärden för innehållet av närsalter och tungmetaller (mängder per person och dygn) i avloppsfraktionerna gråvatten, urin och fekalier.

Mätningarna bekräftar att ett stort antal farliga ämnen faktiskt används i hushållen och att de kanaliseras via avloppen. Att definiera de exakta källorna till de farliga ämnesflödena var däremot svårare eftersom de kommer från en mängd olika vardagsaktiviteter såsom städning, tvätt, användning av kosmetika och hygienprodukter samt från konsumtion av läkemedel.

Avloppsfraktionernas innehåll av både närsalter och farliga ämnen är viktig kunskap för att kunna jämföra olika typer av avloppslösningar. Utifrån egna mätningar och litteraturdata beräknades substansflöden av närsalter och farliga ämnen i olika typer av avloppssystem (scenarier) vilket låg till grund för systemanalys. Substansflödesanalys (SFA) är ett managementverktyg som skapar förutsättningar för att ta beslut om design och driftsstrategier i avloppssystem.

Som förslag på hur man systematiskt kan hantera flöden av farliga ämnen i avloppssystem introducerades begreppet barriärer. Barriärer är en tankemetafor som syftar till olika metoder och strategier för att förhindra farliga flöden. Organisatoriska barriärer kan vara lagar och regler och handlar främst om källkontroll. De organisatoriska barriärerna genomsyrar de övriga barriärerna eftersom man (i princip) kan lagstifta kring och reglera dessa. Brukarbarriärer handlar om hur brukarna använder VA-systemen. Finns vissa barriärer redan i hushållen? Vilka produkter konsumeras? Systembarriärer gäller systemlösningen (VA-systemets design) som styr var de farliga flödena hamnar – ytterligheterna är källsorterande system kontra kombinerade system (blandande flöden). Teknik- eller processbarriärer är kanske den allra tydligaste barriärmetaforen och innefattar behandlingsprocesser i avloppsreningsverket. Val av recipient – ytvatten, grundvatten, mark – var ytterligare en barriär som kan tillämpas för att skydda särskilt känsliga recipienter. Som ett praktiskt exempel på en processbarriär testades membranfiltrering (nanofiltrering) i bänkskala (i laboratorium) för att skilja närsalter från läkemedel i källsorterad humanurin. Studien visade att typ av membran och urinens pH påverkade resultaten samt att man kunde separera kväve men inte fosfor från läkemedelsresterna.

En övergripande slutsats var att flödet av farliga ämnen i avloppssystem är ett komplext problem, inte bara för VA-sektorn, utan för samhället i stort och som kommer att kräva många olika typer av åtgärder för att vända trenden emot uthållig utveckling.

List of papers

Paper I
Palmquist, H. & Hanæus, J. (2004). A Swedish Overview of Selecting Hazardous Substances as Pollution Indicators in Wastewater. *Management of Environmental Quality: An International Journal,* **15**(2), 186-203.

Paper II
Palmquist, H. & Hanæus, J., Hazardous Substances in Separately Collected Grey- and Blackwater from Ordinary Swedish Households. Submitted to *Science of the Total Environment*, *July 2004.*

Paper III
Vinnerås, B., Palmquist, H., Balmér, P. & Jönsson, H. The Characteristics of Household Wastewater and Biodegradable Solid Waste – a Proposal for New Swedish Design Values. Presented at the *IWA Leading-Edge Conference on Sustainability in Water-Limited Environments,* November 8-10, 2004, Sydney, Australia.

Paper IV
Palmquist, H. & Jönsson, H. Urine, Faeces, Greywater, and Biodegradable Solid Waste as Potential Fertilisers. In the Proceedings of *"Ecosan – closing the loop". The 2nd Int. Symposium on Ecological Sanitation,* April 7–11, 2003, Lübeck, Germany, 587-594.

Paper V
Palmquist, H. & Hanæus, J., Organic Hazardous Substances in Greywater and Human Urine from Swedish Households. Submitted to *Environmental Management,* November 2004.

Paper VI
Palmquist, H., Substance Flow Analysis of Hazardous Substances in a Swedish Municipal Wastewater System. Accepted for publication in *Vatten*.

Paper VII
Malmqvist, P-A. & Palmquist, H., Decision Support Tools for Urban Water and Wastewater Systems - Focussing on Hazardous Flows Assessment. In the Proceedings of *the 14th Stockholm Water Symposium*, August 16-24, 2004, Stockholm, Sweden (IWA Publishing: Water Science & Technology).

Paper VIII
Pronk, W., Palmquist, H., Biebow, M. & Boller, M., The Separation of Pharmaceuticals from Nutrients in Source-separated Urine. Submitted to *Environmental Science & Technology,* September 2004.

Contents

Introduction

Until recently, the main emphasis of wastewater treatment was on traditionally observed problems in receiving waters, such as oxygen depletion and eutrophication. Wastewater treatment plants (WWTP) were developed and built to prevent this kind of pollution by implementing different treatment technologies as well as control procedures to fulfil the treatment requirements. As a consequence, the current European effluent standards for urban wastewater treatments plants only include the parameters organic matter (BOD and COD), suspended solids (SS), nitrogen (N), and phosphorus (P) which are regulated by, e.g. the Urban Wastewater Treatment Directive 91/271/EEC (EU, 1991). For hazardous substances such as heavy metals and xenobiotic organic compounds, no effluent standards in the WWTP exist, though they are regulated via water quality objectives (e.g. the EU WFD, 2000) to be met in the receiving waters after dilution (Jacobsen et al., 2004).

In this thesis *substances* were used in terms of “chemical elements and their compounds in the natural state or obtained by any production process” while *hazardous properties* were used as the “inherent capacity that a substance has to cause adverse effects” (Commission of the European Communities, 2001). Moreover, an important distinction is the difference between the terms *hazard* and *risk; Hazardous* refers to the inherent properties of a substance while the *risk* indicates the probability that adverse effects may occur in a human population or environmental compartment due to exposure by a substance. Accordingly, there will be no risk without any exposure, or vice-versa: a very toxic (hazardous) substance may already pose a risk at low exposure while a larger exposure is required for a less toxic substance to cause a risk.

In addition to conventional wastewater parameters, the last few years have seen an increased emphasis on wastewater hazardous substances like triclosan, brominated flame retarding agents, endocrine disruptors, pharmaceutically active compounds, and personal care products. If these compounds are not completely mineralised within a WWTP, it is possible for some fractions of the substance involved to be released into the environment as part of the final effluent discharge, as a component of the sludges produced, or, indeed, volatilised directly into the atmosphere (Byrns, 2001). The effluents of wastewater treatment plants have shown to be significant pathways for hazardous substances to enter the aquatic environment (Daughton and Ternes, 1999; Heberer, 2002).

A complete mineralisation of xenobiotic compounds in treatment systems is rare, with the term biotransformation more accurately describing the potential changes of the composition and molecular structure of such compounds (Byrns, 2001). Some compounds are biotransformed into harmless products possibly entering a particular metabolic pathway and become degraded. Other compounds form daughter products that may be more or less toxic than the parent, whilst others may prove to be generally recalcitrant under the existing conditions and persist in one or more phases within the treatment plant (Byrns, 2001; Heberer, 2002). Several investigations have shown substances of pharmaceutical origin to be seldomly eliminated during wastewater treatment and are not biodegraded in the environment (Ternes, 1998; Daughton and Ternes, 1999; Heberer, 2002).

While water quantity is not of prime concern in most parts of the EU, the availability of water of sufficient quality is (van der Voet et al., 2004). Hazardous substances that resist degradation in WWTPs end up in the effluent and are emitted to the receiving water. In areas where surface water partly originating from the effluent wastewater is used as a raw water source for drinking water production, concentrations of these substances may build up over time and pose risks to human health (van der Voet et al., 2004). In the conclusions from the 14th Stockholm Water Symposium 2004, Professor Malin Falkenmark (SIWI, 2004) reported that a crucial question addressed was the future sustainability of water provision systems, thereby emphasizing the importance of avoiding pollution of future raw water sources. Although sanitation is progressing, hazardous substances that escape water and wastewater treatment continue to raise increasing concerns, especially regarding their potential effects in water ecosystems and on human fertility (SIWI, 2004).

The transport of hazardous substances with wastewater is not only a critical issue for receiving waters, but also for managing wastewater sludge. The content of hazardous elements and organic substances in wastewater sludge reflects a society where large amounts of different chemicals are used for a variety of purposes. Sludge quality changes over time and is dependent on the lifestyle and behaviour of all system users and customers (Kroiss, 2004). Compounds with a strong hydrophobic character are, in general, not significantly degraded by biochemical reactions in the WWTP (Byrns, 2001).

The principal transfer mechanism for these compounds in the WWTP is through sorption to sludge particles i.e. a major pathway for the persistent and hydrophobic compounds is the sludge produced during wastewater treatment. Hydrophobic compounds may to a limited, but sometimes significant extent, also be transported to the receiving water associated with suspended solids in the effluent (Byrns, 2001). This feature makes the reuse of wastewater sludge on arable land a controversial issue in many countries – a widely discussed and debated topic (Kroiss, 2004; Rulkens, 2004). In brief, the disagreements are represented by two main parties – one promoting the reuse of sewage sludge, thereby simultaneously resolving a waste problem and managing the recycling of biosolids and nutrients to arable land; if not recycled, these substances must be replaced by mineral fertilisers. The other party often agrees with the benefits of nutrient recycling, but is concerned about the associated recycling of hazardous substances to arable land, possibly endangering the long term soil fertility and jeopardizing the trust in crops and other products grown on the fields. According to Lee et al. (2002), the management of wastewater sludge – economically and environmentally – has become a critical issue for modern society. Currently, the cost of sludge management often represents more than fifty percent of the total wastewater treatments cost (Rulkens, 2004). Furthermore, there has been a dramatic increase in sludge production in many countries resulting from extended sewerage, new network installations, and the upgrading of existing facilities (Lee et al., 2002). At the same time, outlet routes for wastewater sludge are becoming increasingly restricted due to public health and environmental concerns that leave sludge managers to deal with more complex questions and less technical options (Lee et al., 2002; Kroiss, 2004).

As described by Kroiss (2004), the public has accepted very well the water protection strategy, which has been successfully applied during the last few decades. Politicians like to demonstrate the positive effects of wastewater treatment or receiving water quality, but are much less sensitive regarding resolving sludge disposal because of its controversy. Sewage sludge containing valuable compounds is commonly accepted, perhaps since we are reminded of the long tradition of nutrient recycling by wastewater and manure application in agriculture over centuries, maybe millennia (Kroiss, 2004). The actual challenge, in this respect, is a completely different metabolism of the society that is today strongly influenced by not previously existing material flows from past centuries (fossil fuel energy use, traffic, chemical and pharmaceutical industry products, food industry, the use of heavy metals in buildings, cars, etc.) (Kroiss, 2004). The mentioned complex metabolism of materials and hazardous substances in society is an obvious challenge for wastewater management demanding new approaches for the future. This is the starting point of this thesis.

Background and aim

Urban water and wastewater systems

All urban citizens have personal contact to the drinking water supply and wastewater systems in their everyday life. However, it seems like people have generally lost interest in urban supply systems (water, electricity, solid waste removal, etc.) and prefer an ‘out-of-sight-out-of-mind’ approach. In an overview Tchobanoglous et al. (2003) briefly describe how wastewater systems are designed and managed in urban areas (in the industrialised North): Liquid and solid wastes and air emissions are produced in every community. The liquid waste – i.e. wastewater – is essentially the community’s water supply after being used in a variety of applications. From the standpoint of sources of generation, wastewater may be defined as a combination of the liquid and water-carried waste removed from households, institutions, and commercial and industrial establishments, together with, e.g. stormwater. When untreated wastewater accumulates, the decomposition of the organic matter that it contains will lead to nuisance conditions, including the production of malodorous gases. In addition, untreated wastewater contains numerous pathogenic microorganisms that have dwelled in the human intestinal tract. Wastewater also contains nutrients that may stimulate the growth of aquatic plants, as well as hazardous compounds that may potentially be, e.g. toxic, mutagenic, and carcinogenic. For these reasons, the immediate and nuisance-free removal of wastewater from its sources of generation, followed by treatment, reuse, or disposal into the environment, is necessary to protect public health and the environment (Tchobanoglous et al., 2003).

The wastewater system in this thesis was regarded from a management perspective. Apart from the main task of wastewater management, i.e. the operation and maintenance of existing systems, wastewater management also includes planning for future wastewater strategies and investments. Aiming for an open planning perspective necessitates a systems transparency to facilitate the development of various scenarios for future wastewater systems. Figure 1 visualises how the wastewater system and the substance flows thereof are conceived in this thesis. The wastewater and its constituents originate in numerous sources, emerging in the (main) wastewater fractions of urine, faeces, greywater, stormwater, and industrial wastewater. From their sources these wastewater fractions are further transported and treated in

WWTPs where the end products later reach various destinations in the surrounding natural environment or are further managed, e.g. fertilisers on arable land, treated at landfills, or incinerated.

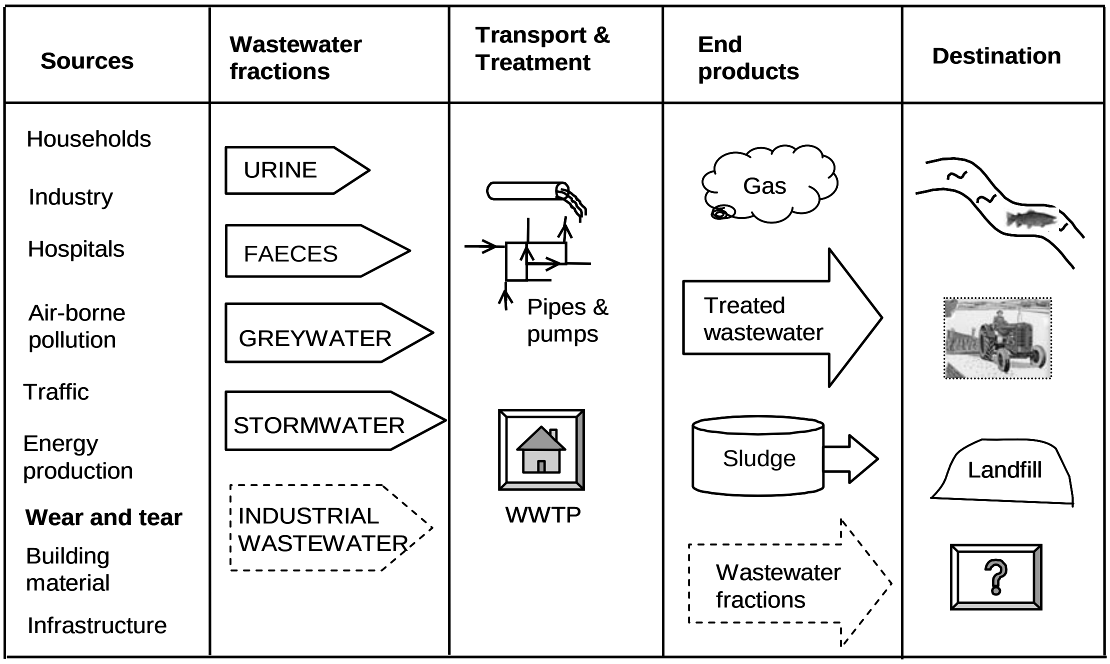


Figure 1 An illustration of how the wastewater system and the substance flows thereof are conceived in this thesis.

Tchobanoglous et al. (2003) claim: "The ultimate goal of wastewater management is the protection of public health in a manner commensurate with environmental, economic, social, and political concerns". Such a general goal or vision is trouble-free for most stakeholders in wastewater management to agree upon, as was done by, e.g. the European Union of National Associations of Water Suppliers and Wastewater Services (EUREAU, 2004). However, this vision of wastewater management is conceived and approached differently as the achievements of the vision vary. Nevertheless, formulating this goal expresses the multi-dimensional nature of wastewater management very well, as is also underlined by the EUREAU (2004) who stated: "Our vision for water supply and waste water services in Europe can only be realised by an active partnership of many different actors i.e. the European Union, national governments, regional and local authorities, representative organisations, water service providers, citizens, industry, and agriculture". Figure 2 shows a framework of an integrated urban water and wastewater system having been divided into three equally important subsystems (***paper VII***):

The *organisation* owns, plans, finances, and manages the urban water system, and may be public or private, central, or local

The *users* use the water and need to get rid of the waste products

The *technical system* (pipes, pumps, treatment plants, etc.) supply the water and take care of the wastewater

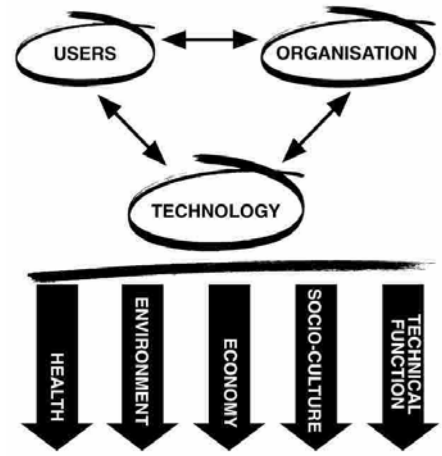


Figure 2 A framework for the integrated sustainability assessment of urban water and wastewater systems as suggested by the Swedish research programme Urban Water (see ***paper VII***).

As illustrated in Figure 2, the wastewater system in this thesis is perceived not as one system, but as a conglomerate of three subsystems – the technological, the organisational, and the users. Since these subsystems are interrelated, changes in one of the systems will affect the performances of the other subsystems.

Implementing changes in urban water and wastewater systems are complex issues demanding strategic decisions support, in which the long term perspectives seem to be a main challenge. For example, the urban drainage infrastructure has a very long life, though political, institutional, and professional practice frameworks and associated values unfortunately have a much shorter life (Ashley et al., 2004). Hence, a short-term bad decision now – about what form of infrastructure we should provide – will have repercussions for decades to come (Ashley et al., 2004). As the best decisions made in the past now present us with a possibly unsustainable future maintenance of our inherited infrastructure (Ashley et al., 2004), implying that current generations will have to pass on some of these cost burdens to future generations (Ashley et al., 2004), which do not meet the goals of sustainable urban water supply and wastewater services. Therefore, strategic decisions support is generally important in wastewater management and becomes critical if inviting public participation into the process (***paper VII***).

Material flows in society

In modern society, great amounts of materials are imported to maintain, support, and develop its metabolism and infrastructure. The downside is the vast amounts of residues and waste produced, e.g. ‘molecular waste’, wastewater, municipal solid waste, industry wastes, and degenerated infrastructure (Andrén et al., 2004). Various hazardous substances enter society through either imports or industrial production. The production of goods and extraction of primary materials may result in point source emissions (Sörme, 2003). The term ‘good’ has been defined as “an economic entity with a positive or a negative value, made from one or several substances” (Brunner, 2002). Examples of goods are roofs, pipes, taps, carpets, furniture, household chemicals, and personal care products.

Goods containing heavy metals, both accumulating in society are called stock (Sörme, 2003). Goods enter society in an annual inflow. Depending on the type of goods, large amounts may remain for a long time, such as water and wastewater pipes that are in use over many decades. Some parts of goods are recycled after use, others become waste. Diffuse emission is generated by, e.g. use, wear, and corrosion of goods (Sörme, 2003). Moreover, consumption emissions like the use of household chemicals from doing the laundry and dishwashing, as well as the use of pharmaceutical drugs and personal care products were regarded as diffuse emissions in this thesis.

As described in the “White Paper – Strategy for Future Chemicals Policy” (Commission of the European Communities, 2001) – chemicals reap benefits that modern society is entirely dependent upon, such as in food production, medicines, textiles, and cars. They also contribute to the economic and social well-being of citizens regarding trade and employment (Commission of the European Communities, 2001). Then again, certain chemicals have caused serious damage to human health resulting in suffering and premature death, and to the environment. Well-known examples are asbestos, known to cause lung cancer and mesothelioma, or benzene, which leads to leukaemia (Commission of the European Communities, 2001). An abundant use of DDT led to reproductive disorders in birds. Although these substances have been totally banned or subjected to other controls, measures were not taken until after the damage was done because knowledge about the adverse effects of these chemicals was not available before they were extensively used (Commission of the European Communities, 2001).

The global production of chemicals has increased from 1 million tonnes in 1930 to 400 million tonnes in 2001. In Europe, about 100,000 different substances are registered in the EU market, of which 10,000 are marketed in volumes of more than 10 tonnes, and a further 20,000 are marketed at 1-10 tonnes (Commission of the European Communities, 2001). The chemical industry is also Europe's third largest manufacturing industry, employing 1.7 million people directly and up to 3 million people indirectly (Commission of the European Communities, 2001).

The use of hazardous chemicals (fuels excluded) is about 8 kg per capita per day in Sweden (Azar et al., 2002), equivalent to around 2.8 tonnes per capita per year. The use of hazardous chemicals per capita in Sweden has been roughly constant in recent years. However, the increasing production and import of hazardous chemicals in the EU-15 countries is about the same as or higher than economic growth (Azar et al., 2002), see Figure 3.

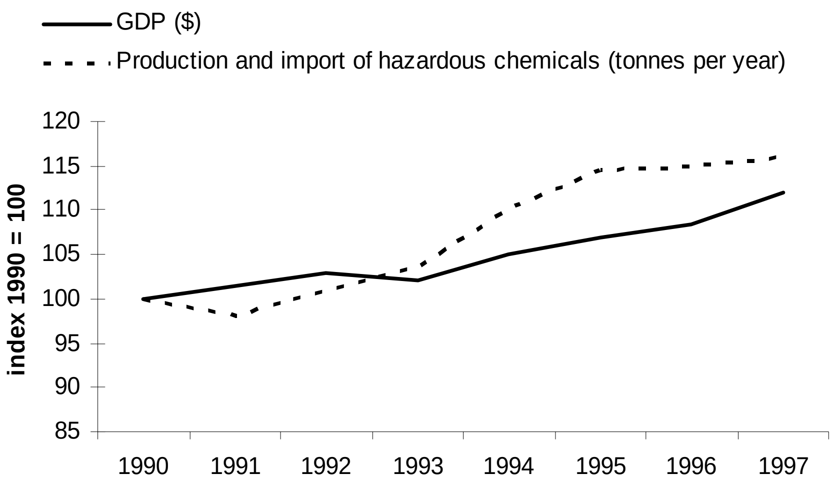


Figure 3 Trends in production and import of hazardous chemicals within EU-15 countries, 1990-1997 (Azar et al., 2002). GDP: Gross Domestic Product.

The production and use of metals are mainly found in infrastructure, buildings, businesses, vehicles, and households (Sörme, 2003). In a case study from Stockholm, Sörme (2003) shows lead (Pb), with about 70% of the total stock, to be a large part of infrastructure (Table 1). The main goods are power cables, telephone cables, and sewer system pipe joints. Principally, copper (Cu) is used mainly in heavy electrical equipment such as generators and transformers, followed by power cables and water pipes, the cadmium (Cd) stock by stabilizers in plastics and batteries, chromium (Cr) and nickel (Ni) in stainless steel in households, such as kitchen sinks and various apparatuses, mercury (Hg) in amalgam (households), and zinc (Zn) in brass and galvanized steel, both used in buildings. The stock of metals within households (per capita) in Stockholm 1995 was about 4 kg Cu, 1 kg Pb, and 2 kg Zn (Sörme, 2003), represented by, e.g. electrical appliances, electronics, personal computers, TV sets, video, and crystal glass. Sörme (2003) reports that, historically, the total metal stock has grown from less than 10 kg per household in 1820 to over 500 kg per household in 1995.

Table 1 The inflow and stock of heavy metals (Cd, Cr, Cu, Hg, Ni, Pb, and Zn) in Stockholm in 1995, and the sector where most of the stock is used (Sörme, 2003).

Metal	Inflow/capita *(kg year^{-1})*	Stock/capita *(kg)*	Dominating sector for the metal stock
Cd	0.01	0.2	Enterprises
Cr	0.5	8	Households
Cu	3	170	Infrastructure
Hg	0.0007	0.01	Households
Ni	0.3	4	Households
Pb	2	73	Infrastructure
Zn	3	40	Buildings

When trying to increase the sustainability of material usage, it is important to note that certain materials are mined as by-products, i.e. an increased use of a certain product might render certain scarce and potentially harmful elements more available, thereby making them less expensive and used in wasteful forms. For this reason, the aim should not be to phase out specific elements, but rather their use in specific applications (Azar et al., 2002).

Furthermore, when analysing the materials flows in society, Azar et al. (2002) states that the focus is increasingly on the intake (or extraction) of materials. However, if the intake leads to an increase in the valuable stock or materials in society, this is not necessarily a problem. It is not the stock of resources in society *per se* that is a problem, it is the leakage of materials to the environment from specific uses that is the key cause for concern, e.g. copper emissions from brake linings (Azar et al., 2002). Andrén et al. (2004) stress that the recycling of material and substances can never be performed with

100% precision but will always be accompanied by leakage, mistakes, accidents, and quantitative degradation of the resources due to thermodynamics, ecological interactions, and results of societal and human factors (Andrén et al., 2004). However, the kretslopp principle i.e. recycling is a powerful model for the societal metabolism of materials and substances, though we need to be aware of its practical shortcomings. Therefore, the use of safety margins and the precaution principle become essential. One example is the necessity of reverse burden of proof regarding hazardous substances on the market to ensure the safety of chemicals, thereby preventing their release until facts can prove their harmlessness (proposed in the new EU Chemicals Policy REACH) (Commission of the European Communities, 2001).

Consumption

In view of the diffuse emissions from the societal metabolism of materials and substances, consumption is one important factor. Consumption was recognised as one of the most central and also one of the most neglected elements in the global search for sustainable development by the Worldwatch Institute (2004). Consumption is essential to human well-being, but over consuming or consuming inappropriately undermines both our personal health and the health of the natural environment on which we depend (Worldwatch Institute, 2004). New consumption patterns will be required to lift billions of people out of poverty in a manner that is consistent with global sustainability, which implies consumption being in part a societal challenge that will require effective use of government regulation and financial policy to achieve the common good (Worldwatch Institute, 2004). Nevertheless, we all make individual important daily decisions that affect not only our own communities, but also the world as a whole – both its current and its future inhabitants (Worldwatch Institute, 2004). Andrén et al. (2004) presented a rather pessimistic analysis on the driving forces for and the future prospect of consumption:

> "In consideration of the many different factors that influences modern people's choices of lifestyle and consumption patterns, it is hard to believe that information and rational arguments alone noticeably will affect these choices. Modern society offers each individual an existential vacuum and the imperative to fill it with meanings; as long as consumption remains the primary strategy to do this, purchasing power and resource depletion will go hand in hand."

In view of consumption and the subsequent diffuse emissions – what visions are realistic for the management of urban water and wastewater regarding the flow of hazardous substances in wastewater systems? Seemingly, the arena is partly driven by players outside the urban water and wastewater sector. Accordingly, the flows of hazardous substances in wastewater systems are not only a complex issue for wastewater management, but for society as a whole.

A systems approach

We live in a systems society – technology, economics, politics, humans, and the environment – all interwoven in a system of connections and dependencies (Andrén et al., 2004). To understand the societal problems of today, such as the issues regarding sustainable development, it may be fruitful to consider the foundations for systems theory and systems analysis. According to O R Young (in General Systems Yearbook, 1964), "A system is a set of objects together with relationships between the objects and between their attributes" (Ingelstam, 2002). The fundamental ambition of the systems theory is to adopt a comprehensive perspective to understand phenomenon and processes in nature and society. According to Ingelstam (2002), there is a need to try and understand systems in an overall view – as seen from above and from without, rather than from beneath and from within. Ingelstam (2002) stresses, furthermore, that the use of interdisciplinary methodologies is important in this attempt with theories and methods from different disciplines complementing and stimulating each other.

Systems and sub-systems

"Everything is interconnected" is not just a popular expression in ecology, but reflects upon a basic principle within systems ecology. Societies with their technical and economical systems depend and have an influence on the surrounding ecosystems (Andrén et al., 2004). Günter and Folke (1993) describe the interdependence between subsystems and how these are self-organising parts in whole:

> We argue that cells, organisms, and ecosystems, even the entire ecosphere, are autopoietic, i.e. far-from-equilibrium dissipative systems […] They are nested within each other, and from this view inseparable, since they, though clearly individual, consist of each other. The nested system consists of identifiable, self-organising parts or "holons". […] It is itself a whole composed of parts, but at the same time a part of some greater whole. Holons are open sub-systems of systems of higher order, with a continuum from the cell to the ecosphere. The hierarchy of holons we prefer to call "holarchy".

From a systems perspective, it is easier to discuss mankind and society's systems in relation to the ecosystems. Andrén et al. (2004) stress that we may perceive "everything is interconnected", but question if we truly understand the societal system's fundamental dependence on the ecosystems, denoted "life-support ecosystem" by the ecologist Eugene Odum:

> "The life-support environment has been defined as that part of the earth that provides the physiological necessities of life, namely food and other energies, mineral, nutrients, air, and water. The life-support ecosystem is the functional term for the environment, organisms, processes, and resources interacting to provide these physical necessities."

From this definition, it is obvious that human and societal systems never can be entirely self-supporting (Andrén et al., 2004). Most human dominated systems such as cities, settlements, and industries depend on areas of natural and domesticated systems – often of a larger area than the system supported. Societal systems must therefore always be seen as sub-systems of the life-supporting ecosystems (Andrén et al., 2004). Like the strong interdependence between subsystems in the biosphere there are also interdependencies between subsystems in society. Many processes and interactions in nature are so complex and unpredictable that we can neither fully understand, nor control the state of conditions. This feature is an inherent property of self-organising systems, which to a high degree is also valid for societal systems (Andrén et al., 2004). Accordingly, urban water and wastewater systems are sub-systems in the urban society – influencing and being influenced by societal activities to varying degrees.

Objectives

The objective of this thesis is to analyse the interface between the use of chemical substances in society and wastewater systems.
As a part of this objective the aim was to chemically characterise domestic wastewater fractions (greywater, urine, and faeces) with respect to a selected number of hazardous substances.
Furthermore, the aim was to suggest a management approach regarding hazardous substances in wastewater systems.

Methods

The interface between the use of chemical substances in society and wastewater systems was mainly analysed by literature studies, presented to varying details in this thesis and ***papers I-VIII***. The integration of knowledge was also made by attending conferences, workshops, and Internet searches.

The chemical characterisation of domestic wastewater fractions – greywater, urine, and faeces – was done through full-scale field samplings at the source separating domestic wastewater systems Vibyåsen and Gebers. Detailed information about the field measurements is presented in ***papers II, III, IV, and V***. Most of the analytical work was made by Swedish accredited contract laboratories. Data on the characteristics of wastewater fractions for a selected number of hazardous substances, attained by the field measurements and from the literature, were essential to improve the prerequisites for performing substance flow analysis and chemical risk assessment of the wastewater systems.

Assessing substances within a system is called substance flow analysis, SFA. In general, the basis of SFA methodology is to obtain knowledge and understanding of the regional metabolism of a certain (group of) substance(s) within a given system. SFA was applied in ***papers VI and VII***. According to Kleijn (2000), the basic principle of SFA is that mass is never lost due to physical or chemical processes, but only transferred to a new medium, product, or good. The core of the SFA method is therefore also to be found in the mass balances of substances within a system. From a perspective of municipal environmental management, Burström (1999) claims that SFA can provide important quantitative and qualitative knowledge on the regional metabolism of different substances that support municipal environmental planning and management. SFA in municipal environmental management may also help policy-makers in learning about structural inter-relationships between different socio-economic activities and the surrounding nature. For example, it is not the wastewater treatment plants themselves that cause eutrophicating emissions of nitrogen and phosphorus to lakes and coastal waters, but the anthropogenic consumption of animal food and other products in households (Lindqvist and von Malmborg, 2004).

The basic ideas for the management approach of barriers against hazardous substances in wastewater systems originate from the area of drinking-water safety where the concept of microbial safety barriers is crucial for drinking-water quality. Raw water sources for drinking-water production are protected, and treatment methods at waterworks are designed to function as safety barriers against hazardous microorganisms (i.e. bacteria, viruses, and parasites). This is regulated by, e.g. the EU Council Directive 98/83/EC and World Health Organization (WHO) guidelines, which state that, "Drinking water should contain pathogenic microorganisms only in such low numbers that the risk for acquiring waterborne infections is below an accepted limit".

From these basic ideas the barrier approach evolved by combining knowledge from associate areas, mainly via literature studies, dialogues at workshops, and conferences.

Selecting indicator substances in wastewater

Controlling wastewater quality is difficult due to the complex mixture of substances. The key problem for selecting proper indicator substances is the large number of chemicals used in society. Moreover, the limited knowledge of many of these substances provides a weak base for assessing the chemical risks in wastewater systems. The process of selecting hazardous substances as pollution indicators in wastewater systems was analysed in ***paper I***. The reviewed methodologies from the literature showed one clear commonality: the grouping of substances with various characteristics representing hazardous properties. From each group, one or several indicator substances were selected to represent the hazardous property of that specific group. The selected set of indicator substances was meant to represent the chemical risk as a whole **(*paper I*)**.

The process of selecting hazardous substances as pollution indicators turned out to be complex. Creating a comprehensive list of indicator substances for the measurement or monitoring of chemical risks in wastewater and residues meant accepting many simplifications. Several approaches for hazard classification and selection processes for indicator substances were found in the literature, though, the sets of indicators, or priority pollutants, are still not identical on the lists reviewed (***paper I***).

It also becomes clear in ***paper I*** that much work is needed for the selection and classification of hazardous substances. For example, the COMMPS-method, elaborated by the Commission of the European Communities (2000), used 752,000 analytical data concerning 330 substances from environmental control programmes all over Europe. After the data was compiled, resulting in four lists of substances, a group of experts finished the work by selecting 32 priority pollutants, while thoroughly motivating their choice for each substance. Similar work has also been performed in developing other priority pollutants lists, indicating that much work is needed for these issues.

Currently, the European effluent standards for WWTPs only include 5 parameters – BOD, COD, SS, N, and P (EU, 1991), though another 32 groups of substances are still to come, as having been suggested for the control of wastewater quality according to the European Water Framework Directive (2000). Measuring all potentially present substances is not possible due to the cost and practicality involved as well as the creation of monitoring programs based on selected indicator substances being a delicate issue. Uncertainties exist as to if the selected substances represent the true chemical risk to which we are exposed, while in fact possibly consisting of other substances than those measured or combinations of substances. This means that there will be large, misspent expenditures for chemical analyses if the results render little about the true chemical risks. Monitoring programs for substances being spread to the surrounding environment also means costs and work for the organisation responsible, i.e. the more extensive the monitoring programs, the greater the expenditures and workload. Since urban wastewater systems and treatment plants are nodes for the large number of chemicals used and emitted by society, one may question if it is realistic for the urban water and wastewater sector to meet these expenses alone – in costs and workload – for controlling wastewater pollution.

Characteristics of wastewater fractions

Complex mixtures of substances appear in domestic wastewater as a result of the consumption of numerous goods and products, e.g. household chemicals and personal care products, and the wear of interior fittings, e.g. carpets, kitchen sinks, and furniture. As many as 30,000 substances regarded as everyday chemicals are regularly used in households (Commission of the European Communities, 2001).

To increase knowledge about hazardous flows from households to wastewater systems, the characteristics of domestic wastewater fractions urine, faeces, and greywater were investigated. The flows, sources, and the fate of the water and its major constituents, such as nutrients, pathogens, and harmful chemicals, are essential knowledge when assessing different wastewater strategies. Qualitative and quantitative data on the characteristics of wastewater fractions may be applied for substance flow analysis, thereby serving as a base for risk assessment, improvements of the system, and information about hazardous substances in wastewater systems.

The two Swedish housing areas Vibyåsen and Gebers were selected for the field measurements, since their wastewater systems have separate flows of wastewater fractions. Wastewater systems with separate flows are generally quite rare. The sites and methods are thoroughly described in ***papers II, III, IV, and V***. Greywater is generally defined as household wastewater without any input from toilets, i.e. wastewater produced from bathing, showering, hand washing, laundry, and the kitchen sink. At Vibyåsen, urine and faeces were collected in water closets and thus combined into blackwater. At Gebers, urine and faeces were collected separately in dry toilets.

Ordinary wastewater parameters

The distributions of ordinary wastewater constituents in the wastewater fractions at Vibyåsen and Gebers are presented in Figure 4. A comparative evaluation revealed that the distribution of flow was different in the two systems and is likely related to the use of water closets (thus flush water) at Vibyåsen, but dry toilets at Gebers. However, the total water consumption was less at Vibyåsen with 66 l p^{-1} d^{-1} of greywater and 28.5 l p^{-1} d^{-1} of blackwater (***paper II***) than at Gebers with 110 l p^{-1} d^{-1} of greywater use (***paper IV***).

The distribution of total solids (TS) and macronutrients – nitrogen (N), phosphorus (P), potassium (K), and sulphur (S) – between the fractions appeared to be quite similar in both systems. At Gebers, the N:P:K:S relationships of the urine mixture was 15:1:3:1 and 3:1:1:0.3 for the faecal matter, corresponding well to the crop uptake of macronutrients. According to Hammar et al. (1993), most crops take up 4 to10 times as much nitrogen, 1 to 8 times as much potassium, and 0.3 to 1 times as much sulphur as the phosphorus crop uptake. The contents of macronutrients make the toilet fractions, i.e. the blackwater at Vibyåsen as well as the urine mixture and the faecal matter at Gebers, potential fertilisers from a plant nutrients point of view (***paper IV***).

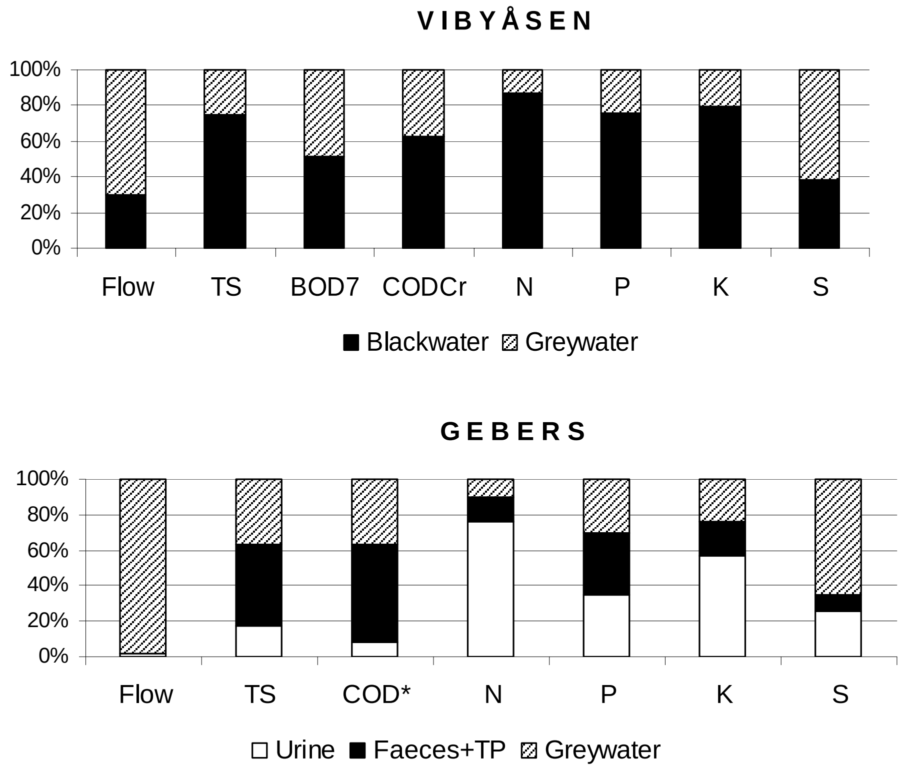


Figure 4 The distribution of ordinary wastewater parameters between the wastewater fractions at Vibyåsen and Gebers. COD* (Gebers) show calculated values for urine and faeces, but measured (COD_{Cr}) for the greywater.

The analytical procedures of biochemical oxygen demand (BOD_7), and chemical oxygen demand (COD_{Cr}) for the separate fractions of urine and faeces (from Gebers) were not reliable. Therefore, COD values were instead calculated from the measured values of the volatile suspended solids (VSS) of those fractions (denoted COD* in Figure 4). The average VSS_{urine} was 2.6 kg p^{-1} $year^{-1}$ and $VSS_{faeces+TP}$ was 16.2 kg p^{-1} $year^{-1}$, which was converted by the COD/VSS ratio 1.6 (Henze et al., 2002) into 4 kg COD* p^{-1} $year^{-1}$ for urine and 26 kg COD* p^{-1} $year^{-1}$ for faecal matter (***paper V***). In the blackwater at Vibyåsen the (measured) COD_{Cr} averaged 23.5 kg p^{-1} $year^{-1}$ (***paper II***), see Figure 4.

In greywater, the BOD_7 and COD_{Cr} were measured and the mass flows compared, see Figure 5, showing the BOD_7 in greywater to be higher at Vibyåsen than at Gebers, though the COD_{Cr} value showed the opposite pattern. This pattern was most likely related to normal variations of the greywater composition, as was shown to be very variable for oxygen demanding substances (***papers II and V***).

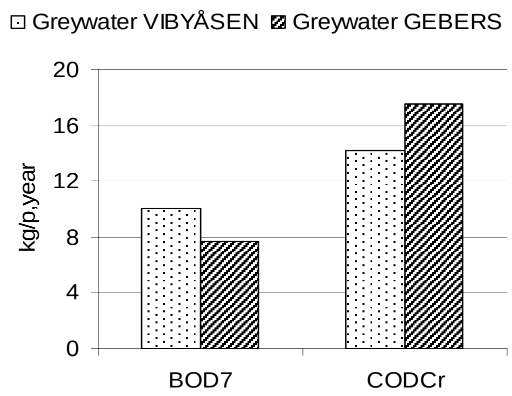


Figure 5 The mass flows of BOD and COD in greywater at Vibyåsen and Gebers.

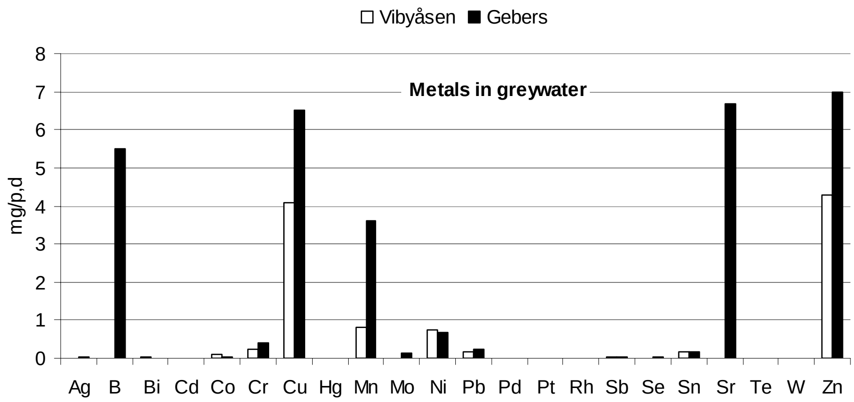
Selected hazardous substances in wastewater fractions

Metals

The content of metals in wastewater is a reflection, or an inevitable effect, of the flow of metals in society. During recent years, the content of some rare metals such as gold, silver, and wolfram, have been observed in various environmental compartments and wastewater sludge (Eriksson, 2001). The presence of these rare and potentially hazardous elements in wastewater and sludge poses chemical risks to arable land and the receiving waters.

A total of 24 elements were measured in the grey- and blackwater at Vibyåsen, of which 22 (92%) and 23 (96%) were detected in each of the fractions (***paper II***). Of the 22 metals measured at Gebers, 19 were detected in both the greywater and in the faecal matter (86%) (***paper IV***), while 15 were detected in the urine (68%) (***paper IV***). In a comparative evaluation of the two systems, hazardous metals in wastewater fractions showed the metal flows to be generally higher at Gebers, see Figure 6 and 7. However, in the toilet fractions, the flows of some metals (e.g. Ag, Cu, Hg, Ni, and Pb) were higher at Vibyåsen than at Gebers, see Figure 7.

(a)



(b)

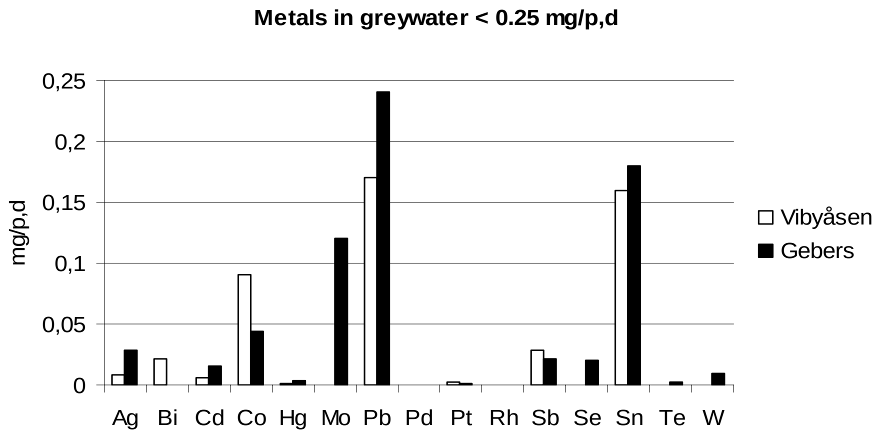


Figure 6 The metals flows in greywater at Vibyåsen and Gebers. Diagram (a) shows all the metals investigated, diagram (b) is an extract of (a) showing the metals with <0.25 mg p^{-1} d^{-1}.

To analyse the fertilising potential for urine, faeces, and greywater, the ratios of 12 hazardous (non-essential) metals to phosphorus and nitrogen were presented in ***paper IV***. The hazard/nutrient ratios were lower for urine than for faeces, greywater, and municipal wastewater sludge, which were also included in the study (***paper IV***). Basically, the levels of hazardous metals in food equal the amount of metals removed from the fields by crop uptake. Metals may also be added to food during processing and refinement of food products as well as through food packaging and containers. However, to reach a mass balance of the metals in the field, the fertiliser should not contain higher hazard/nutrient ratios than the food. Since the main contribution of hazardous metals to urine and faeces is provided via food, those fractions may be potential fertilisers, provided that external metal sources can be restricted. The fertilising potential of the wastewater fractions greywater and municipal wastewater sludge was questioned in a long term perspective due to higher hazard/nutrient ratios than what the plant uptake can counter balance, implying a possible accumulation of hazardous metals in the fields (***paper IV***).

Organic hazardous substances

More than 900 organic hazardous compounds may potentially appear in greywater, though literature data on organic hazardous substances in greywater is entirely missing (Eriksson et al., 2002).

A selected number of organic hazardous substances were searched for in both the grey- and blackwater at Vibyåsen, but only in the greywater at Gebers. The expected very low concentrations of organic hazardous substances in separately collected urine and faeces convinced us to use the restricted budget for chemical analyses on mainly the greywater, thereby ensuring distinct data achievements (***paper V***). At both sites, around 80 selected hazardous organic substances were measured, namely nonylphenol- and octylphenol ethoxylates, brominated flame-retardants, organotin compounds, PAH, PCB, phthalates, and triclosan. Moreover, monocyclic aromatics were investigated at Vibyåsen and linear alkyl benzene sulfonate (LAS) at Gebers.

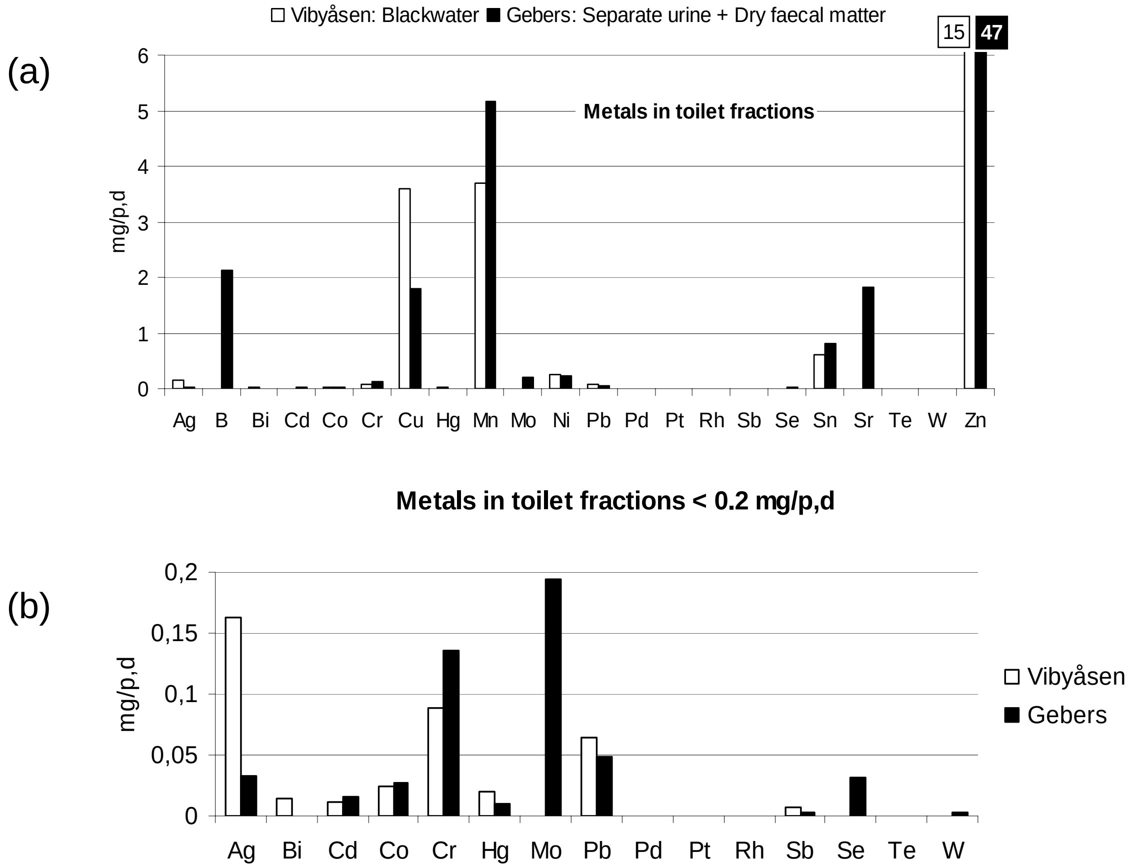


Figure 7 The metals flows of the toilet fractions at Vibyåsen and Gebers. Diagram (a) shows all the metals investigated and diagram (b) is an extract of (a) showing the metals with <0.2 mg p^{-1} d^{-1}.

At Vibyåsen, 46 of the 81 measured substances were found in greywater (57%) and 26 of the 72 measured substances were found in the blackwater (36%) (***paper II***). At Vibyåsen, none of the PCBs was detected in either of the fractions. At Gebers, 41 of the 81 selected organic hazardous substances were found in the greywater, representing all of the investigated substance groups (***paper V***). In Figures 8a to 8e the mass flows of the selected organic hazardous substances in greywater at Vibyåsen and Gebers are presented.

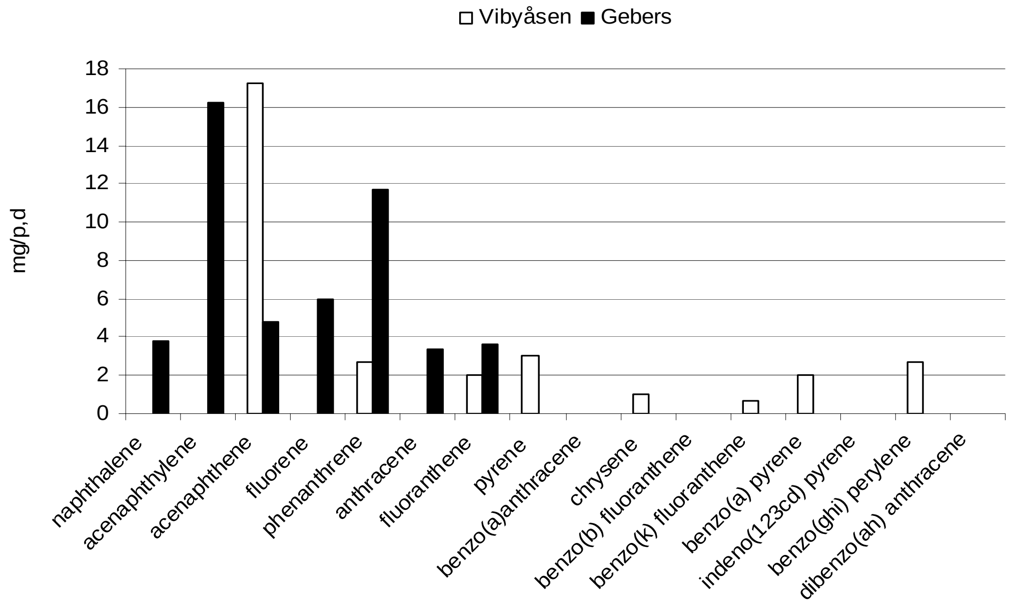


Figure 8a The mass flows of PAHs in the greywater at Vibyåsen and Gebers.

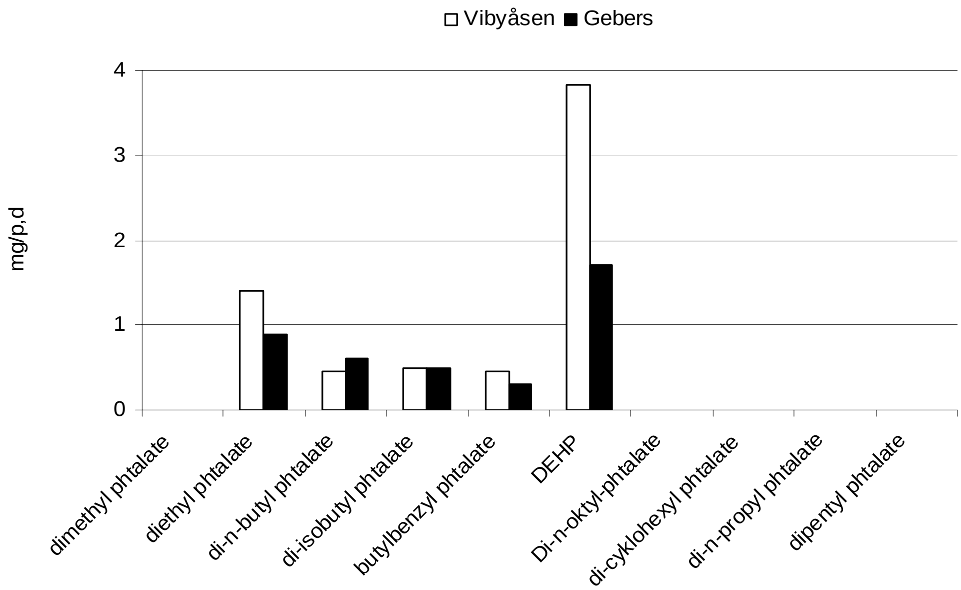
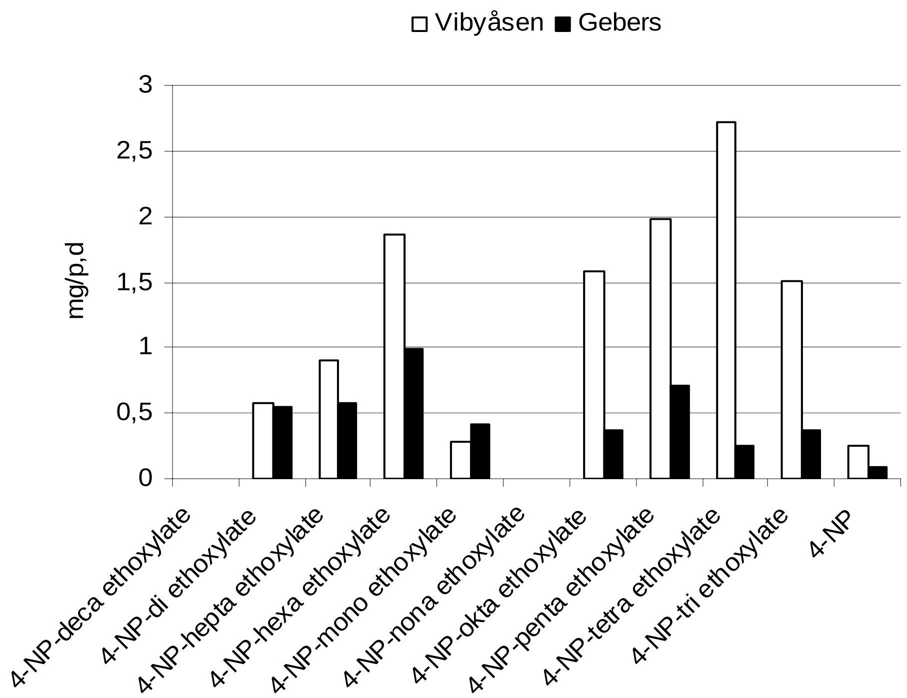


Figure 8b The flows of phthalates in the greywater at Vibyåsen and Gebers.



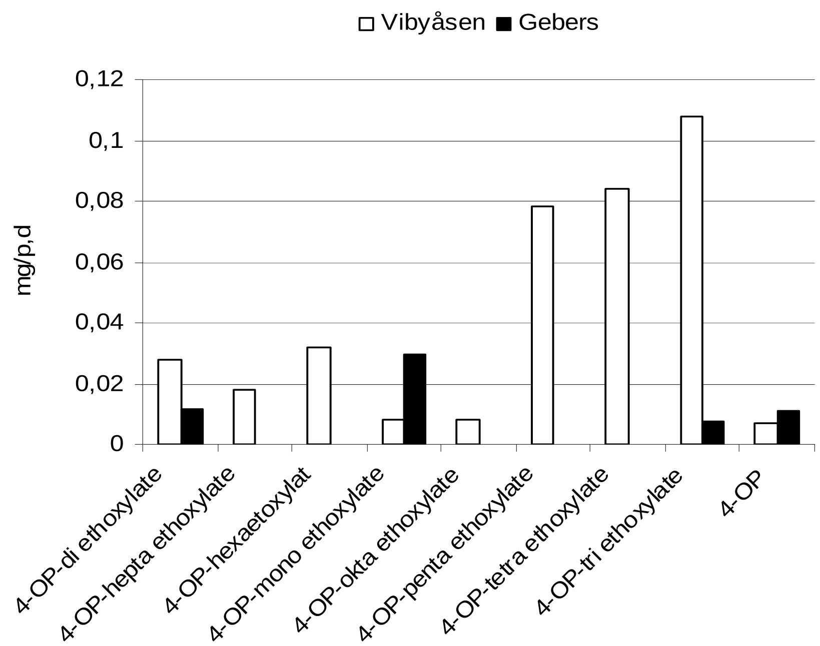


Figure 8c The flows of nonylphenol- and octylphenol ethoxylates in the greywater at Vibyåsen and Gebers. Note the different scales in the diagrams.

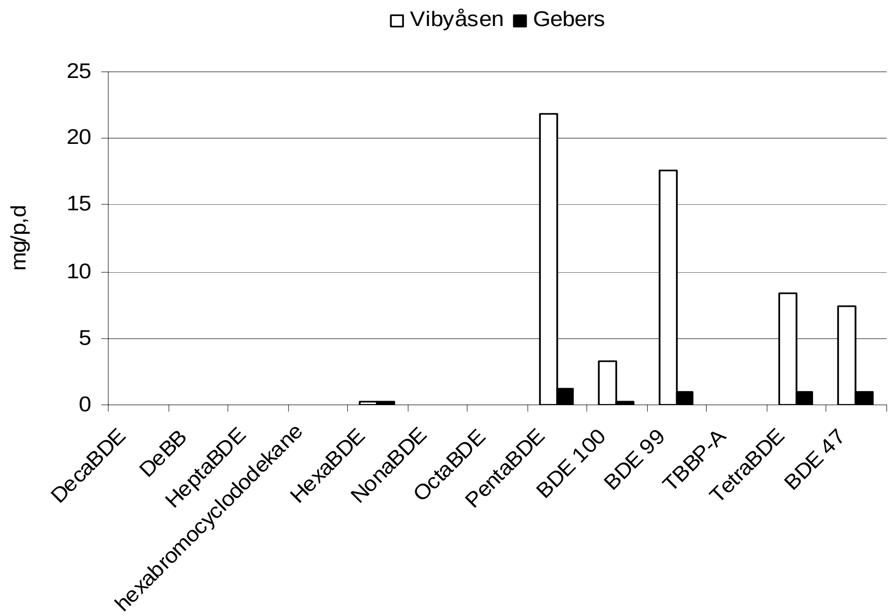
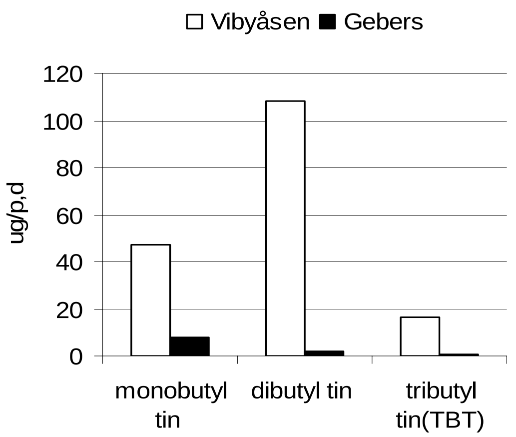


Figure 8d The flows of brominated flame-retarding agents in the greywater at Vibyåsen and Gebers.



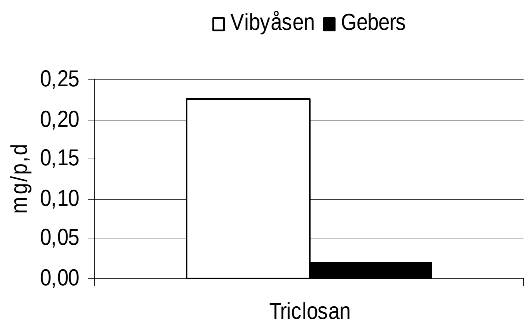


Figure 8e The flows of organotin compounds (to the left) and triclosan (to the right) in the greywater at Vibyåsen and Gebers.

A comparative evaluation of organic hazardous substance flows in the greywater between the two systems revealed generally larger mass flows at Vibyåsen (Figures 8a to 8e).

The contribution of organic hazardous substances with toilet fractions occurs mainly via excretions of, e.g. pharmaceuticals, pesticides, and food additives into urine and faeces. Such compounds are not completely eliminated in the human body but they are often excreted only slightly transformed or even unchanged as conjugated polar molecules e.g. as glucoronides (Heberer, 2002). For example, 3.5 µg p^{-1} d^{-1} of the antibacterial agent triclosan was found in the urine at Gebers. Regarding the blackwater at Vibyåsen, the possibilities of adding polluted residues into the blackwater via the water closet, e.g. when emptying a scouring pail and throwing in cigarette butts, snuff, etc., are other relevant pathways (see ***paper II***).

Sources of hazardous substances in wastewater fractions

Both two sites revealed differences regarding mass flows of the selected hazardous substances in wastewater fractions, while the flows of some substances were higher at Vibyåsen, others were higher at Gebers. This random pattern is most likely related to which household activities occurred during the actual sampling occasions.

The different designs of the systems, i.e. the use of water closets at Vibyåsen and source separating dry toilets at Gebers, may account for some of the differences. Repeating these investigations would probably lead to comparable, though still unique data sets since the use of consumer goods alters over time.

The input flows of hazardous metals from households to wastewater were rather similar in the greywater and toilet fractions (see Figures 6 and 7). The flows of organic hazardous substances from households to wastewater, however, seem mainly to emerge in the greywater and derive from many diffuse household sources, including everyday activities, the wear of, for example, pipe material and interior fittings, and airborne deposition.

Due to the complex mixture of substances found in separate wastewater fractions, it is not possible to exactly distinguish their specific sources; many of these substances are used as chemical additives in a wide array of consumer goods, e.g. food and drinking containers, household cleaning products, furniture, shampoos, cosmetics, and household and agricultural pesticides. An inventory of household chemicals (presented in ***paper V***) revealed on average 10 different product types being used in each household at Gebers, with each product type representing several brands, e.g. 20 brands of washing powder, 11 brands of hair styling products, and 31 brands of shampoo. Since each brand of the product types has its unique formula, the total number of substances became substantial.

Design values for wastewater fractions

Present Swedish design values (DV) for the quantities and composition of domestic wastewater fractions urine, faeces, and greywater were presented by the Swedish Environmental Protection Agency (Swedish EPA, 1995). As discussed in ***paper III***, there was a need for this data to be scrutinized, as substance flows in society change over time, e.g. some heavy metals in society are decreasing (Lohm et al., 1997) and food consumption patterns are changing. An important weakness of the existing Swedish design values was the often inappropriate origins of the data.

The DVs for urine and faeces were based upon Swedish food consumption data from the early 1990s combined with literature data on human excretion of urine and faeces (Swedish EPA, 1995). The excretion studies were based on a small number of people, almost always fewer than 20 and often fewer than 10, over a short period of time – normally 1 to 10 days. The DVs for greywater were mainly based on samples from 16 apartments in an eco-village during 7 days of sampling. Therefore, a number of recent measurements (including Vibyåsen and Gebers) for the characteristics of household wastewater fractions were combined in ***paper III*** and a proposal for new design values was put forward, see Table 2.

Applying design values, such as the ones proposed in Table 2, in substance flow analysis may serve as a base for systems analysis and decision support in wastewater management. However, design values are a delicate issue, as they always represent a selection of substances, people, time, geographical context, and technical systems. A huge number of measurements may overcome these problems. Once published, the DVs will be very useful in various kinds of studies. However, society's consumption patterns and lifestyles change over time, along with wastewater characteristics changing with it. Therefore, design values should be used with good judgement and need regular updating, assumingly each 5th to 10th year.

Table 2 Proposed DV for the compositions of different household fractions of wastewater per person and year (***paper III***).

	Unit	Urine	Faeces	Greywater
Wet mass	kg	550	51	36500
Dry mass	kg	21	11	20
BOD_7	kg	-	-	9.5
COD_{Cr}	kg	-	-	19
N	g	4000	550	500
P	g	365	183	190
K	g	1000	365	365
Cu	mg	37	400	2500
Cr	mg	3.7	7.3	365
Ni	mg	2.6	27	450
Zn	mg	16.4	3900	3650
Pb	mg	0.73	7.3	350
Cd	mg	0.25	3.7	12
Hg	mg	0.30	3.0	1.5

Hazardous substances in wastewater management

Systems for sustainable urban water services require tools to assess and control various risks related to urban water and wastewater flows. Throughout the development of centralised wastewater systems, great trust has been appointed to wastewater treatment plants (Kroiss, 2004) to remove environmentally hazardous substances and make the effluent harmless to the receiving waters. However, complex mixtures of substances appear in the wastewater due to diffuse emissions from the societal metabolism of materials and substances. Today's operating WWTPs indeed provide different types of technical treatment processes; however, their capacity cannot manage the entire flow of hazardous substances through the wastewater system alone (Byrns, 2001; Palmquist, 2001).

The flow of hazardous substances from society to the surrounding nature is a consequence of industrialisation, urbanisation, and welfare, built into society's physical infrastructure as well as our social behaviour. Since wastewater systems are sub-systems of urban infrastructure, hazardous substances are channelled via wastewater flows. Existing waterborne sanitary systems signal to their users the removal of their (unwanted) waste by just opening the tap or flushing the toilet. Therefore, it seems relevant to search for wastewater management tools that support a shift in perspectives by combining a traditional end-of-pipe perspective with more systems-oriented perspectives, thereby linking the use of resources and the spreading of hazardous substances to their underlying causes and driving forces (i.e. consumption and lifestyle) rather than only focusing on the emissions.

The flows and sources of wastewater fractions and their constituents, such as nutrients and hazardous substances, are essential knowledge when assessing alternative wastewater strategies that render the SFA methodology a useful tool. To compare two different wastewater management scenarios in the Swedish town of Surahammar regarding hazardous flows, 16 selected hazardous substances were assessed in a comparative SFA – a conventional scenario vs. a separating scenario, see Figure 9 (***paper VI***).

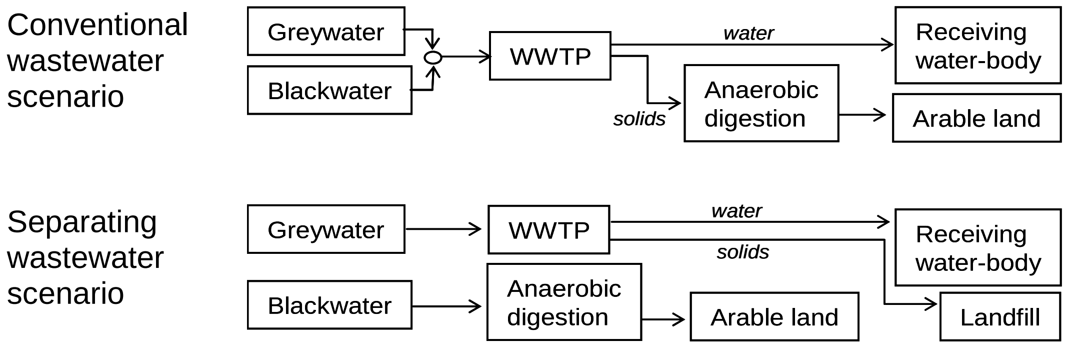


Figure 9 System boundaries for and the design of the two wastewater management scenarios studied in comparative SFAs in both the ***papers VI and VII***.

The comparative SFA in ***paper VI*** revealed the conventional scenario to cause an overall higher flow of the selected hazardous substances to the receiving environment, i.e. the receiving water and the arable land, than the separating scenario. In the separating scenario parts of the hazardous flow were directed to the landfill. In the separating scenario the studied substances predominantly emerged in the greywater with the exceptions of Ag and Sn that subsisted to about 80% in blackwater, and Hg, Zn, and 4-NP that occurred between 40-60% in the blackwater. The remaining 12 substances subsisted 0-20% in the blackwater (***paper VI***).

A barriers approach

A barriers approach for the assessment of hazardous flows in municipal wastewater systems was suggested. The Oxford English Dictionary (2004) defines a barrier as "a fence or material obstruction of any kind erected (or serving) to bar the advance of persons or things, or to prevent access to a place". In wastewater management, the barriers approach was intended to be used to interpret and compare different wastewater systems, and to serve to find out if and how much the flow of hazardous substances can be stopped, diverged, or transformed at the source or during transport throughout the system (***papers VI and VII***). Five kinds of barriers were suggested (see Figure 10):

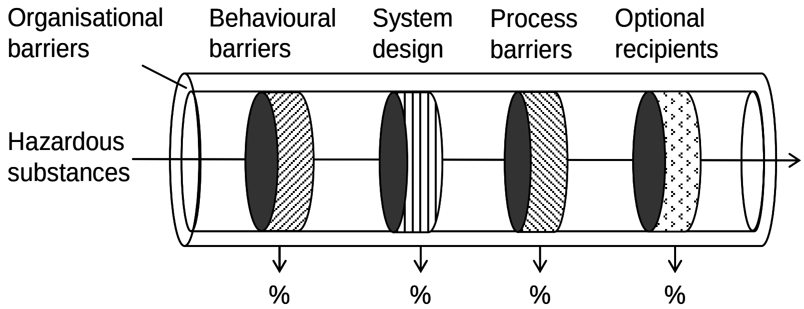


Figure 10 A schematic outline of the barriers concept illustrating four of the suggested barriers embraced by the organisational barrier (the tube), which by legislative and administrative measures directly or indirect can affect all other barriers (***paper VII***).

Organisational Barriers

Organisational barriers with the aim to prevent risks from the flow of hazardous substances in society represent a wide spectrum of legislative and administrative measures at global, national, and local levels. In Europe, general large scale regulatory and organisational changes are preferably governed by the EU; the field of water policy by, e.g. the Water Framework Directive (WFD), that specifies objectives of achieving a ’good status’ for all European waters by 2015 with sustainable water use throughout Europe (EU WFD, 2000). The WFD will affect the institutional arrangements and incentives of the urban water and wastewater sector within all EU Member States, including the monitoring and management of hazardous substances in wastewater. Nationally, central governments may establish various kinds of organisational barriers for hazardous flows in society as part of the ambitions to change course towards sustainable development. In these ambitions, municipalities must be supported by carefully prepared incentive structures to act as the prime movers in the transformation towards sustainable development, since leaving the responsibility for such comprehensive changes to the municipalities has shown to imply only minor changes in practice (Söderberg, 1999).

Potential organisational barriers for hazardous flows in society are:

- Chemicals policy, e.g. REACH based on the "White Paper – Strategy for a future chemicals policy" (Commission of the European Community, 2001)
- Emission regulation, e.g. IPPC directive (Integrated Pollution Prevention and Control) (Council Directive 96/91/EC)
- Technical regulations, regarding the technical design and function of wastewater systems
- Fertilising policies, e.g. the approach of the farming and food industries to the use of wastewater residues (e.g. sewage sludge) on arable land
- Eco-labelling regulation (Regulations European Commission No. 1980/2000)

Phasing out or substituting hazardous substances is a possible legal measure and an example of implementing an organisational barrier, e.g. the current objective is to heavily regulate or even phase out the use of cadmium (Azar et al., 2002). But cadmium is mined as a by-product of zinc, and if OECD (Organisation for Economic Co-operation and Development) countries phase out cadmium, the price will drop, possibly resulting in dissipative uses in non-OECD countries. However, if cadmium is extensively used in solar cells made of cadmium telluride or in large nickel-cadmium batteries or both, its value would increase and more wasteful uses would decrease (Azar et al., 2002). Referred to as a soak-up strategy, this could also be applied to other flows. It could be beneficial from an environmental perspective, since it could provide an incentive to reduce the leakage to nature of toxic metals (Azar et al., 2002).

Bans on detergents containing phosphate are another example on the phase-out strategy. Banning was successful in reducing the phosphate flows to the receiving water bodies, but as the tenside compounds that often replaced the phosphate turned out to be persistent (and thus relatively resistant to degradation in the WWTPs), the phasing out of phosphate (done legally, i.e. an organisational barrier) replaced one environmental hazard with another. These examples highlight the importance of studying how material flows are nested. For this reason, the aim should not always be to phase out specific substances, but to phase out their use in specific applications (Azar et al., 2002) and thoroughly assess which scenarios may arise by substituting substances.

Behavioural Barriers

Behavioural barriers include the users' perspective. Which barriers for hazardous flows are to be set up in the households? Which products are consumed? What effects on hazardous flows can be expected of information campaigns? A straightforward example is the information campaign about cadmium in artist paint, performed by the Stockholm Water Company. Artist paint may contain up to 45% Cd, which is the pigment in these paints. According to the Stockholm Water Company, their municipal WWTPs receive more than 30 kg of Cd per year originating from artist paints (Stockholm Vatten, 2004). They recommend the use of alternative paints, and instruct how to handle the cleaning of brushes and waste. The barrier effect of such measures is very difficult to assess and one should probably not be overconfident in the response. As a consequence, it becomes essential to phase out or replace hazardous substances in consumer goods and products (***paper VII***).

System Barriers

System barriers relate to the infrastructural and technical design of urban water and wastewater systems. The extremes vary with separation of urine, faeces, greywater, and stormwater occurring at the source, while combined flows that mix wastewater occur from numerous other sources. For example, one-fourth of domestic wastewater phosphorus emerged in the greywater and three-fourths in the blackwater (***paper VII***). For cadmium and triclosan the result was almost the opposite, i.e. 80% in the greywater and 20% in the blackwater (***paper VII***). This relevant information about the system barrier is needed to decide on the design of the system.

Technical or Process Barriers

Technical or process barriers include process units in wastewater treatment plants. These are concrete physical barriers, based on mechanical, biological, and chemical treatment processes supplying separation or decomposition or both of the constituents in the wastewater. Complete mineralization of xenobiotic compounds in treatment systems is rare; the term biotransformation more accurately describes the potential changes to the composition and molecular structure of such a compound (Byrns, 2001). As presented in Figure 11 the biodegradation of xenobiotic substances vary with the operating conditions in the WWTP and between substances (from ***paper VI***).

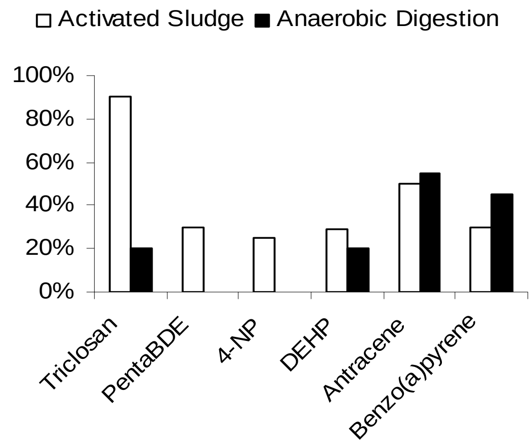


Figure 11 The biodegradation of organic hazardous substances in the aerobic activated sludge step, and in the anaerobic digestion step in the WWTP. For PentaBDE and 4-NP, no biodegradation was expected under anaerobic conditions (see ***paper VI***).

The fate and distribution of hydrophobic chemicals in treatment systems is largely controlled by the physico-chemical properties and, according to Byrns (2001), biodegradation mostly influences compounds with moderate hydrophobic properties (log K_{ow} values in the range 1.5-4). The long solids retention times (SRT) in biological treatment processes that are typically practiced with biological nutrient removal seem to favour the biodegradation of many, but not all potentially hazardous organic substances (Byrns, 2001; Clara et al., 2004; Jacobsen et al., 2004; Kroiss, 2004).

Sedimentation, flocculation, chemical precipitation, sand filtration, and membrane processes are all separation processes in wastewater treatment. Membrane processes (micro-, ultra-, and nanofiltration, as well as reverse osmosis) are wide applied in the treatment of aqueous based systems involving material recovery, reuse, and pollution prevention. As a stand-alone process, a membrane will separate wastewater into two streams, a purified stream and a concentrated stream that set requirements for further management of the concentrated residues.

A nanofiltration (NF) membrane was modelled as an additional process barrier in the WWTP in ***paper VII***. This additional process barrier considerably reduced the substance flow to the receiving water body in both the conventional and separating scenarios, see Table 3. Data for this modelling was gathered from Visvanathan and Roy (1997), who reported a phosphorus separation efficiency for NF as tertiary treatment of wastewater to > 95%, resulting in an effluent concentration of less than 0.1 mg L^{-1} P. For cadmium the NF of synthetic wastewater reduced Cd^{2+} from 500 ppm to 15 ppm, corresponding to 97% separation efficiency, as reported by Qdais and Moussa (2004). The NF separation of triclosan was assumed to be 80% (from experiences in ***paper VIII***). The principal disadvantages in membrane filtration are higher costs and the operation and maintenance requirements compared to conventional treatment methods (Clara et al., 2004). Operating membrane filtration directly on wastewater may often be problematic due to the variable composition and high fouling potential of most wastewater (Côté and Thompson, 2000).

Table 3 A NF membrane as an additional process barrier in the WWTP (in ***paper VII***) would considerably reduce the substance flow to the receiving water body. The figures in brackets represent the substance flow without nanofiltration modelling.

		Conventional scenario	**Separating scenario**
P	kg $year^{-1}$	11 (221)	3 (57)
Cd	g $year^{-1}$	2.6 (76)	2.2 (60)
Triclosan	g $year^{-1}$	7 (37)	6 (30)

The separation of pharmaceuticals from nutrients in human urine by nanofiltration was investigated in ***paper VIII*** as an example of a process barrier. Several nanofiltration membranes were tested for the separation of pharmaceutical and estrogenic compounds from urine to generate a micropollutant-free nutrient solution to be used as fertiliser. A fresh urine solution containing most nitrogen in the form of urea and a synthetic urine solution with a similar inorganic composition were tested at different pH values to investigate the separation behaviour. These solutions were spiked with the pharmaceutical and estrogenic compounds propranolol, ethinylestradiol, ibuprofen, diclofenac, and in some cases, carbamazepine. The retention of both pharmaceuticals and inorganic ions was influenced by pH. In general, with increasing pH the acidic compounds (ibuprofen and diclofenac) had an increased retention, while the basic (propranolol) and neutral (ethinylestradiol) compounds had a decreased retention (***paper VIII***).

Among the membranes tested, the NF270 membrane showed the best performance in retaining the pharmaceutical compounds (Figure 12). Optimum retention of the pharmaceutical compounds was obtained at pH values around 5. At this point, the retention of all the pharmaceuticals in human urine was above 92%, while the retention in the synthetic urine solution was above 75%. The differences in retention behaviour could be partly explained by the influence of organic matrix substances in human urine. These substances (such as oxalic acid, uric acid, amino acids, and the like) can form complexes with Ca^{2+} and Mg^{2+}. Organic compounds or their complexes can essentially adsorb to the membrane by electrostatic or unspecific interactions (van der Waals), perhaps functioning as a secondary membrane, and thereby increasing the retention of the organic compounds (***paper VIII***).

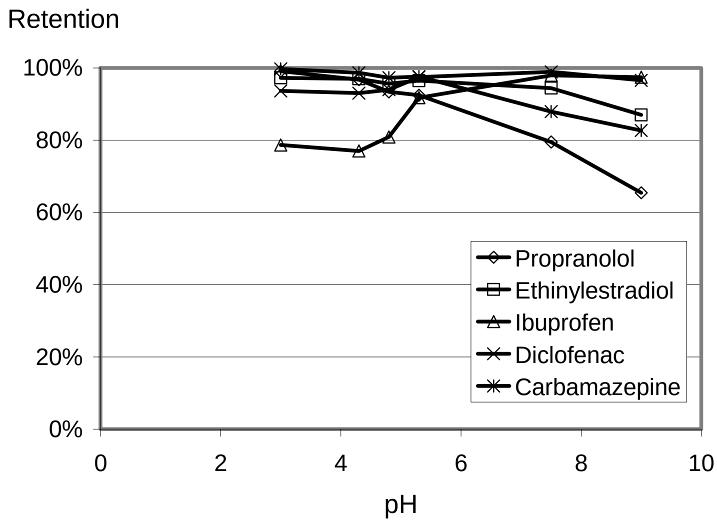


Figure 12 Retention of pharmaceuticals as a function of pH in fresh urine using a NF 270 nanofiltration membrane (***paper VIII***).

The NF separation of ions is largely based on electrostatic interactions, as displayed by the much higher retention of multivalent ions (phosphate and sulphate) than single valent ions (Na^+, K^+, Cl^-, NH_4^+) (Figure 13). Furthermore, non-charged compounds such as urea had a much smaller rejection, meaning that the multivalent ions (phosphate and sulphate) were retained together with the pharmaceutical compounds in the concentrate while urea was permeated, thus forming a nitrogen rich solution largely free from pharmaceuticals.

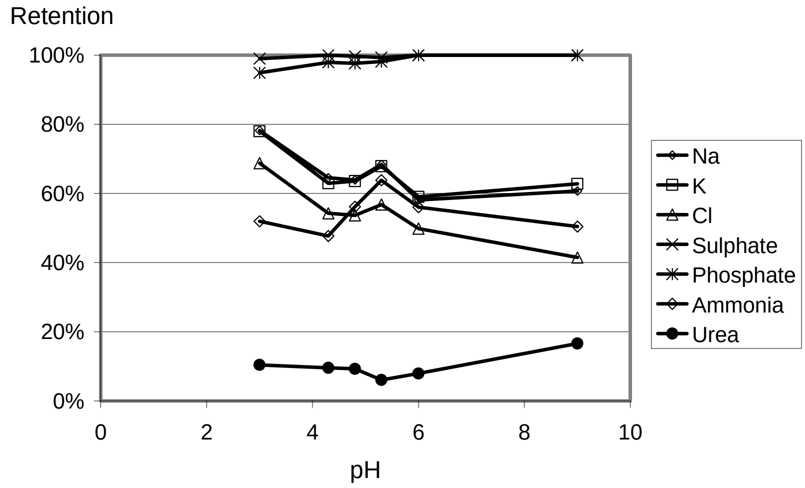


Figure 13 Retention of inorganic ions as a function of pH in fresh urine using a NF270 nanofiltration membrane (***paper VIII***).

The results in ***paper VIII*** show that membrane filtration is a potentially promising process barrier, though membrane processes need thorough optimization, i.e. inaccurate operating conditions and operating failures result in poor process performance, thereby implying weak barrier function.

Optional Recipients

Assessing optional recipients is a way to protect sensitive recipients. By selecting another recipient for wastewater residues, sensitive recipients may be protected by redirecting the hazardous flow elsewhere to a less sensitive recipient. Optional recipients are highly dependent on the geographical context, which could be lakes, rivers, seas, or soils. The SFA in ***paper VII*** shows the barrier effect for Cd to be moderate in both tested wastewater management scenarios. An additional NF membrane would protect the receiving water body, but to protect the arable land, additional measures are required. Here, optional recipients could be a matter of discussion. For example, the wastewater sludge from the combined wastewater system might be applied in soil applications other than as fertiliser for food production, to safeguard clean food production. As claimed in ***paper IV*** the fertilising potential of wastewater sludge must be questioned in a long term perspective, since the metal/nitrogen ratios of 12 hazardous metals (including Cd) showed higher ratios in sludge than what the plant uptake can counter balance, thus implying metal accumulation in the soils. LeBlanc et al., (2004) propose various ways to use wastewater sludge – composting, mine site reclamation, landfill cover, tree farming, sod farm base as soil enrichment, and topsoil manufacturing.

Combined barrier effect

It is not only important to identify each barrier within a defined system, but also assess the combined effects from all existing barriers. Evaluating the combined barrier effect implies considering the whole chain of barriers backwards from the actual receivers of the hazardous substances (the end-points), e.g. the receiving water, arable land, and landfill. To obtain the combined barrier effect, the reduced amounts of each substance at each barrier are multiplied at the end-points of the system. Figure 14 shows the combined barrier effects (counting systems and process barriers) for the case of Surahammar, issued from the end-points (A) emission to water and (B) emission to arable land (***paper VI***).

An evaluation of the combined barrier effect implied that a change from a conventional wastewater system to a source separating system in Surahammar would have a greater impact for the management of solid residues (i.e. the emissions to arable land) than for the effects in the receiving waters, see Figure 14 (***paper VI***). The flow of hazardous substances to the receiving water would not be greatly affected by such a systems change, shown by the small difference in the combined barrier effects for emissions to water (Figure 14). However, high levels of barrier protection do not guarantee chemical safety. Substances passing through the barriers, even in very small amounts, but which are very toxic to the receiving environment, may cause more severe ecotoxicological effects than high volume substances with low toxicity.

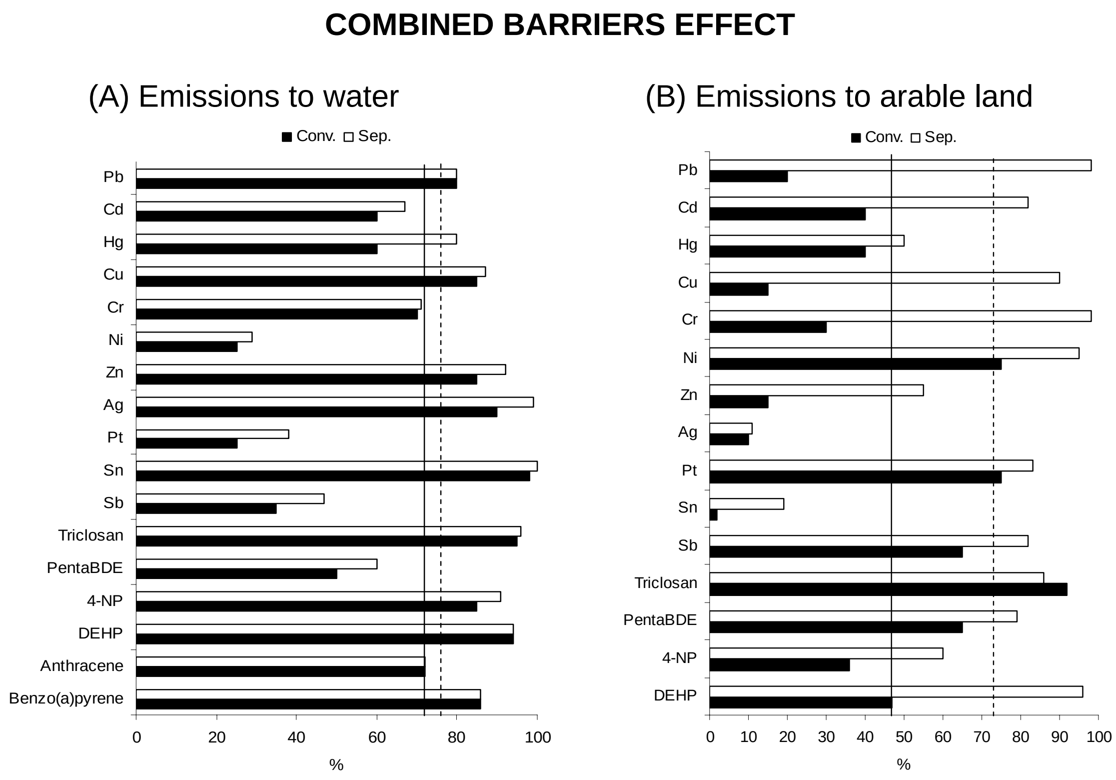


Figure 14 With regards to the system barrier and process barriers, the combined barriers effects were presented for the end-points: (A) the receiving water and (B) the arable land. The average combined barrier effects for these substances are marked in the diagrams by a vertical solid black line for the conventional scenario (——) and a vertical dotted line for the separating scenario (- - - -). The combined barrier effects for emissions to arable land could not be evaluated for the two PAHs due to a lack of data (***paper VI***).

The barriers approach was proposed as a tool on a conceptual level (a way of thinking) as an attempt to develop a basis for systems analysis, risk assessment, improvements of the system, and for communication regarding hazardous substances in the wastewater systems. Barriers depend on the context, implying that a barriers model developed for the wastewater system in one municipality may not be fully applicable on the wastewater system in another municipality due to differences in physical structures, the inherited infrastructure, the number of citizens, environmental ambitions, etc. Furthermore, the different barriers need to be developed and managed on different levels. For instance, organisational barriers require measures from, e.g. national and international legislation, while system barriers are rather decided upon and managed by urban planners and engineers at the regional and municipal levels. Behavioural barriers are, however, influenced by individual choices (consumption) as well as by the market for goods and products. Accordingly, the flows of hazardous substances in wastewater systems is a complex issue, not only for wastewater management, but for society as a whole, i.e. various kinds of measures are needed to achieve a change in direction towards sustainable development.

Conclusions

The extensive use of materials and substances in society causes diffuse source emissions that lead to uncontrolled spreading of hazardous substances within the technosphere and to the surrounding nature. Hazardous substances are largely channelled from society via wastewater flows to surrounding nature. The complex and unpredictable flow of hazardous substances in wastewater systems raises increasing concern, especially in view of the negative effects in the water and soil ecosystems as well as for the potential risks to human health. Since urban wastewater systems are nodes for the numerous substances used and emitted by society, one may question if it is realistic for wastewater management to meet these expenses alone – in costs and workload – to control wastewater pollution.

Two Swedish housing areas, Vibyåsen and Gebers, were selected for field measurements, since their wastewater systems have separate flows of wastewater fractions. The mass flows of TS, oxygen demanding substances, and macronutrients in greywater and toilet fractions appeared to be quite similar in the two systems, and summarised, those values corresponded well to the mass flows in ordinary combined domestic wastewater.

The mass flows of hazardous metals from households emerged in similar quantities in the greywater and toilet fractions. However, ratios of hazardous metals to phosphorus and nitrogen were significantly lower in the urine than in the faecal matter and greywater.

The mass flows of organic hazardous substances from households were mainly searched for in the greywater, resulting in 50-60% of the 81 measured substances being found, with representatives from all of the substance groups investigated. However, it was not possible to exactly identify their specific sources as the mass flows of organic hazardous substances derive from diffuse household sources like everyday activities (laundry, cleaning, etc.), the wear of things such as pipe material and interior fittings, and from airborne deposition.

The input of organic hazardous substances to urine and faeces occurs mainly via the excretion of, for instance, pharmaceuticals, pesticides, and food additives. For example, 3.5 µg p^{-1} d^{-1} of triclosan was found in the urine at

Gebers. Other examples of relevant pathways are when emptying a scouring pail and throwing in cigarette butts, snuff, etc., into the blackwater via the water closet. Of the 72 measured organic hazardous substances, 36% were found in the blackwater at Vibyåsen.

Based on a number of recent measurements (including Vibyåsen and Gebers) a proposal for new Swedish design values for the characteristics of household wastewater fractions (including ordinary wastewater parameters and seven metals) was put forward. However, the consumption patterns of society changes over time, and with it, so do the wastewater characteristics. Therefore, design values should be used with good judgement and require regular updating, assumingly each 5^{th} to 10^{th} year.

A possible management approach was suggested to interpret and compare different wastewater systems, and to serve to find out if and how much the flow of hazardous substances can be stopped, diverged, or transformed at the source or during transport throughout the wastewater system. Organisational and behavioural barriers, system design, process barriers, and optional recipients were suggested.

Accordingly, the flows of hazardous substances in wastewater systems is a complex issue, not only for wastewater management, but for society as a whole, i.e. various kinds of measures are needed to achieve a change in direction towards sustainable development.

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