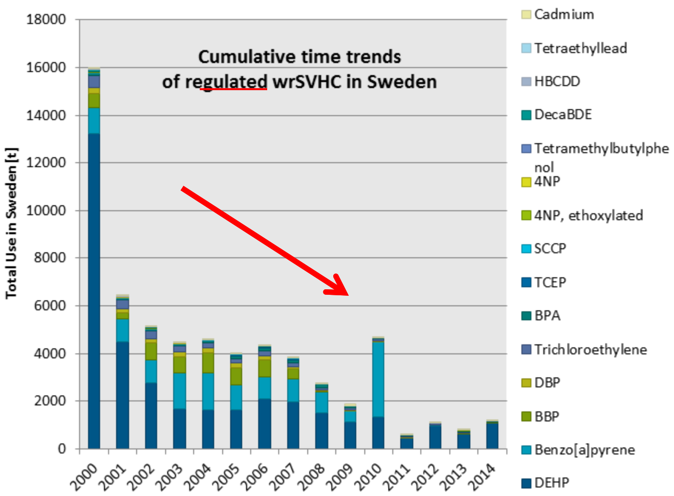
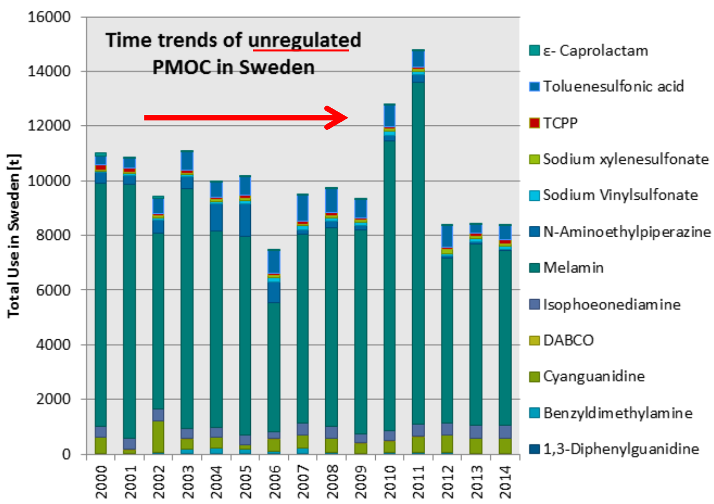
Professur für Hydrologie

der Albert-Ludwigs-Universität Freiburg i. Br.

Kathrin Sackmann

Impact of regulation on emerging pollutants and surface water pollution of tomorrow





Masterarbeit unter Leitung von Prof. Dr. Markus Weiler

Freiburg i. Br., Oktober 2017

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Using the SPIN database for time trends of problematic substances in Sweden

Referent: Prof. Dr. Markus Weiler
Korreferent: Prof. Dr. Dirk Bunke

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Acknowledgements

This thesis contributed to the SOLUTIONS project was funded by the European Union's 7th Framework Programme for research; technological development and demonstration (grant agreement no. 603437). Firstly, I would like to state that this thesis would not have been possible without the great and unconditional support of my secondary supervisor Dirk Bunke. I am very grateful for all possibilities and ideas that Dirk shared with me during the time I spent on the thesis. I am especially grateful for the opportunity to participate and present my thesis results at the SOLUTIONS general assembly. Moreover, I am very glad for the friendly and cheerful communication and unreserved option to or pose questions or ask for feedback.

Also I would like to thank Hildis Strigl for her initial commitment to me during the application procedure and her support during the incorporation phase. Moreover, I would like to express my gratitude for the numerous supportive and friendly colleagues and especially to Carola, who cooked delicious meals for everyone during the time I spent at Öko- Institut e.V Öko- Institut e.V.

I would also like to thank experts who shared their data and knowledge with me. Special thanks go to Prof. Dr. Thorsten Reemtsma who provided me with a list of PMOCs. Further, I would like to thank Magnus Rahmberg for the policy framework analysis of the PMOCS and the wrSVHC. Without their participation and input, thesis in this form could not have been written.

Moreover, I would also like to acknowledge Prof. Dr. Markus Weiler of the Hydrology department at Albert- Ludwigs Universität Freiburg as the supervisor of this thesis, and I am grateful for his guidance on the process during the course of the thesis.

Last, I would like to thank my mother and aunt and especially my close friend Nathalie Wössner who always supported me in any endeavor undertaken.

Abstract

REACH, the European chemicals regulation, aims to substitute substances of very high concern (SVHC). Substances identified as SVHC are placed on the so-called REACH "Candidate List". The intention of REACH is to enforce the substitution of SVHC eventually. However, numerous substitutes are not adequately researched but are still industrially used. Given these substances are emitted to the environment they can constitute a future threat to humans and the environment. The scientific community accepts that humans and the environment will face a new class of emerging pollutants (EPs). The goals of the thesis are an understanding of the impact of regulation on the use of SVHC, the patterns of substitution of chemicals (which chemicals are substituted and by what) and the identification of substances that could constitute future EPs in Swedish surface waters.

To address these goals, the Nordic SPIN database was used to retrieve time trends of water relevant substances of very high concern (wrSVHC). Moreover, SPIN was used to identify substitute chemicals in Sweden. It was shown that the Swedish industry decreased the total use of SVHC over time in accordance with the regulation exerted by the candidate list under REACH. As control group a group of less regulated water relevant chemicals (persistent and mobile organic contaminants, PMOC) was analyzed, showing no decrease in total use over time. To analyze the pattern of substitution for SVHC, the technical use categories "plasticizers", "flame retardants", "surfactants" were introduced and analyzed in depth. In order to identify potential future EPs in Swedish surface waters, all identified substitutes were compared to the NORMAN list of emerging substances in the environment. This resulted in various substances with increasing use trend that were already found in European surface waters or other environmental matrixes. Monitoring data for the identified emerging substances is scarce in Sweden. Future investigation needs to be done on the emission of the identified substances that are used increasingly and are classified as emerging substances. Moreover, the individual physio-chemical properties of the substances and their behavior in the environment need to be researched and coupled with monitoring programs in Sweden.

Keywords: REACH regulation, SVHC, chemical substitution, regrettable substitution, emerging substances, emerging pollutants in surface water, SOLUTIONS project

Zusammenfassung

Durch die Regulierung von lang genutzten sogenannten besonders besorgniserregenden Chemikalien (SVHC) wurde der Ersatz dieser Chemikalien andere Stoffe unumgänglich. Häufig sind diese Substitute ungenügend erforscht und werden trotzdem industriell genutzt. Vorausgesetzt diese Stoffe gelangen in die Umwelt, können sie eine zukünftige Bedrohung für Mensch und Umwelt darstellen. Experten erwarten, dass Mensch und Umwelt durch die Nutzung neuartiger Chemikalien zukünftig mit einer Vielzahl neu aufkommender Schadstoffe (EPs) belastet werden. Das Ziel dieser Masterarbeit ist es, den Einfluss der REACH Kandidatenliste auf die Nutzung bestimmter SVHC zu quantifizieren. Außerdem sollen Muster der Substitution (welche Chemikalien werden substituiert und womit) aufgedeckt werden. Weiterführend sollen Substanzen identifiziert werden, die eine zukünftige Gefährdung von schwedischen Oberflächengewässern darstellen könnten.

Hierzu wurde die nordische SPIN Datenbank genutzt, um Zeitreihen der wasserrelevanten besonders besorgnis erregenden Substanzen (wrSVHC) für Schweden abzurufen. Die schwedischen SPIN Daten wurden weiter genutzt um Substitutionschemikalien zu identifizieren. Es wurde gezeigt, dass die schwedische Industrie die Regulierung der Nutzung von wrSVHC unter der REACH Kandidatenliste für die wrSVHC über die Jahre umsetze. Als Kontrolle für nicht regulierte wassergefährdende Stoffe, wurde eine Gruppe von wasserrelevanten, persistenten und mobilen organischen Schadstoffen (PMOC) ausgewählt. Bei ihnen zeigt die Analyse, dass die verwendeten Mengen im Gegensatz zu den wrSVHC nicht zurückgehen. Um das Substitutionsmuster der wrSVHC zu analysieren, wurden die technischen Nutzungsklassen „Weichmacher", „Flammschutzmittel" und „Tenside" eingeführt. Um zukünftig aufkommende Schadstoffe in schwedischen Oberflächengewässern zu identifizieren, wurden alle identifizierten Substitutionschemikalien mit der europäischen NORMAN Liste der in der Umwelt aufkommenden Substanzen verglichen. Dies ergab einige Substanzen, die bereits in europäischen Oberflächengewässern oder angrenzenden Umweltmatrizen gefunden wurden. Monitoring Daten der Oberflächengewässer in Schweden sind unzureichend für die identifizierten Stoffe, die jedoch in Schweden zunehmend genutzt werden und als aufkommende Substanzen in der EU registriert wurden. Daher wird Schweden verstärkte Forschung zum Verhalten dieser Chemikalien in der Umwelt, gekoppelt an ein Monitoring Programm für diese Substanzen empfohlen.

Keywords: REACH Regulierung, SVHC, chemische Substitution, aufkommende Schadstoffe, Chemikalien in Oberflächengewässern, SOLUTIONS Projekt

List of Abbreviation and terms

Abbreviation/ Terms	Stands for
REACH	Registration, Evaluation, Authorisation and Restriction of Chemical Substances (EU legislation)
Regulated substance	Chemicals which are under strict regulatory control (REACH authorization process incl. REACH Candidate List, restrictions). These regulatory steps go beyond registration.
REACH candidate List	List holding Substances of very high concern
REACH restriction list (Annex XVII)	List holding substances that are restricted for certain uses/ applications
REACH authorization list (Annex XIV)	List holding substances of very high concern that cannot be used any longer, unless authorized for specific uses/applications
SPIN	Substances in Preparations in Nordic countries (Database)
SVHC	Substance of high concern according to REACH
wrSVHC	Water relevant substance of high concern
STP	Sewage treatment plant
ES	Emerging substance
EP	Emerging pollutants
HFR	Halogenated flame retardant
HFFR	Halogen- free flame retardant
OP	Organophosphates
TCCP	Tris(2,3-dichloropropyl) phosphate
PVC	flexible polyvinyl chloride
DEHP	Bis(2-ethylhexyl) phthalate
DINCH	1,2-Cyclohexane dicarboxylic acid diisononyl ester
QA	Quaternary ammonium
LAS	linear alkylbenzene sulfonate
APE	Alkylphenol ethoxylate
AE	Alcohol ethoxylate

1. Introduction

Chemical pollution of European surface waters has been on the agenda of the European Commission since the 70ties and resulted in the regulation of chemical pollutants in water with Directive 76/464/EEC in 1976 (European Commission, 2000). However, despite significant achievements regarding European surface water quality, chemical pollution poses a risk to almost half of the water bodies monitored in Europe (Malaj et al., 2014). At present, 33 substances or groups of substances on the list of priority substances which are frequently monitored (European Commission, 2007). Yet, there are numerous unmonitored substances in the aquatic environment which are expected to be of ecotoxicological and potentially human health concern (Petrie et al., 2014). These emerging pollutants (EPs) are the product of various industrial, societal, economical and climatic changes and can originate from various sources such as industries, personal care products, pharmaceuticals or household chemicals. Currently in the EU, man-made products contain 30,000 to 70,000 (Loos, 2012). Many of these substances finish their life cycle as unidentified EPs in the environment in multiple environmental matrixes one of them being surface waters. Plenty of research has been conducted on specific classes of EPs, such as pharmaceuticals or pesticides which are regularly detected in many European rivers and lakes and have an established rank in the discussions of the scientific community. However, little research has been conducted on a huge number of industrial chemicals and their faith and potential influence as EPs in surface waters. In order to prevent future pollution of aquatic environments emerging industrial chemicals need to be identified, reduced and/or replaced at their source. However, a great difficulty is to identify and prioritize chemicals, as the whole cycle of these chemicals from the production and use over the emission up to their hazard profiles need to be evaluated. The problem starts with the use of novel chemicals that are insufficiently studied with respect to their impact for the environment and human health. Given those chemicals are emitted to the environment and certain exhibit certain physio-chemical properties such as slow biodegradability or specific mode of actions (e.g. photosynthesis inhibition or cholinesterase inhibition), these chemicals can constitute EPs. However, needs to be emphasized that industrial use does not directly translate to emission to the environment. Emission can occur however, via various routes such as leaching from chemical products or transport and discharge of chemicals, illegal dumping, insufficiently treated effluents from sewage treatment plants (STPs), or wet dry deposition (Holt, 2000). The amount of chemical that is found in the environment is strongly dependent on the use category of the chemical. A fertilizer for example is very likely to be a dangerous non-point source for import of problematic substances to various environmental matrixes, whereas an industrial chemical that is used in a closed system as an intermediate in a production process under strictly controlled conditions is unlikely to be emitted to the environment. Nonetheless, emission of industrial chemicals to the environment is commonly observed, also via leaching from articles.

With the establishment of the network of reference laboratories, research centers and related organizations for monitoring of emerging environmental substances (NORMAN network) in 2005 the first institution concerned with EPs was founded. Since then, over 600 EPs have been identified and categorized into various technical use classes, such as “flame retardants” “plasticizers” or “surfactants”. Given the need for a platform that combines information on chemicals and their faith in the environment, the European Commission launched IPCHEM - the Information Platform for Chemical Monitoring. IPCHEM holds all information of the NORMAN network plus data from other databases on occurrences of chemical substances in the environment or the human body. Apart from the existing databases and networks, specific EU projects like SOLUTIONS were designed to model the aquatic pollution of tomorrow.

The European Union research project SOLUTIONS[1] has the objective to deliver tools and models to identify substances and mixtures that endanger aquatic ecosystems and human health. This is done I) analyzing the complex contamination present in the environment and II) analyzing the multitude of chemicals produced and used in Europe (Brack et al., 2015). SOLUTIONS is divided into various sub-projects (SPs) concerned with different tasks, such as a monitoring based approaches for chemicals that are already in the environment, exposure and effect modelling for compounds that are produced, applied and probably emitted to the environment, and scenarios to identify future trends in pollution (Brack et al., 2015). The SOLUTIONS project includes predictions of time trends for industrial chemicals and which are registered under REACH (Moritz et al., 2017). They are produced, used and are likely to be emitted via one or multiple possible routes to surface waters. Data sources for use trend analysis are market and use volumes for chemicals registered under REACH (given as tonnage bands) and information from the Nordic SPIN database (given as precise “total- use tonnages” for individual substances). Moreover, SOLUTION operates on multiple scales such as EU-level or national level. Of the multiple databases used in and for SOLUTIONS, this thesis uses the Nordic SPIN database, the REACH candidate list, and databases from the NORMAN network to predict future EPs for Sweden. The contributions to SOLUTIONS are the evaluation of utility and reliability of the different functions and datasets found in the Nordic SPIN database. As described above, use categories of chemicals are important to estimate the likelihood of emission of surface waters. To this end it was planned to use SPINs Use Category data, holding tonnage data for individual chemicals within 62 specific use

[1] SOLUTIONS: EU 7th Framework program: http://www.solutions-project.eu/

categories and an ExposureTool, estimating risk to various environmental matrixes. However, in the course of the thesis the usefulness and reliability of the ExposureTool and the Use Categories were questioned and rated as insufficient as can be seen in the appendix chapter 8.2. Nonetheless, the TotalUse data was found to be operative, holding specific use tonnage data. These specific tonnage data allow modeling of the time trends of surface water relevant SVHC in Sweden. Surface water relevant SVHC are defined as SVHC that are either: 1) listed on the Water Framework Directive (2000/60/EC) (WFD) substance list or 2) on the NORMAN list of emerging substances.

1.1. Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) for substances of very high concern (SVHC) leads to declining SVHC use

With the REACH Regulation (Registration, Evaluation, Authorization and Restriction of Chemicals) in 2007 legislation was adopted to ensure – beyond other objectives - the safe use of chemicals. To surveil safe use, chemicals have to be registered before they can be placed on the European market. For substances with an annual production volume of 10 tons and more (per producer or importer), a chemical safety assessment has to be made in order to demonstrate that the substances can be used safely. Beyond registration, REACH has two other processes for substances which even after registration can pose a risk to humans and the environment, namely restriction and authorization. Restriction aims at prohibition of specific uses of particular chemicals, such as for example Toluene which is restricted as adhesive or spray paints intended for supply to the general public. Even though certain uses are restricted, all other uses of the chemical are still allowed. Currently there are 65 valid entries on REACH restriction list (Annex XVII) (ChemSafetyPro, 2017). Contrary, Authorization aims to replace very problematic substances by less problematic substances or processes, whenever technically and economically feasible. Authorization applies to the most problematic substances. Under REACH, these substances are called “substances of very high concern (SVHC)”. They are placed on the so-called candidate list (CL). The list is called this way because placing on this list is a first step of imposing an obligation of authorization on them (Grunwald and Hennig, 2014). The REACH candidate list currently holds 173 entries of substances of very high concern (SVHC) (ECHA, 2017a). Most of these entries refer to a distinct chemical substance. However, quite a relevant number of entries refer to more than one substance or to a complete group of substances. As described below in the methodology chapter, in total 250 distinct substances have been identified which refer to the 173 entries of the REACH candidate list. Once the substances are assessed as SVHC and placed on the Candidate List, these chemicals can be forwarded to the Annex XIV authorization list. At present, this list includes more than 43 substances that after a given sunset date, which is a binding fade out requirement, cannot be used any longer (ECHA, 2017b). Solely in rare cases companies can advocate that there is no other chemical which could substitute the chemical of concern – in a specific use. An example here is the plasticizer DEHP, which is authorized for blood transfusion bags to counteract blood coagulation. It is expected that industries react to the placement of chemicals on the candidate list by replacing the SVHC with substitutive chemicals as the SVHC hold the risk of undergoing strict regulation in the foreseeable future. Currently there are no indicators for policy performance of REACH in place. Within this thesis not only potential emerging pollutants are identified, but also the impact of regulation of the REACH Candidate list is analyzed. This is done with the temporal use developments for SVHC in the SPIN database, where scenarios before and with REACH are compared. This is a sound indicator for the policy performance of the REACH Candidate List in Sweden.

SPIN, even though holding merely data from the Nordic countries is of great interest. SPIN holds information of about 27 000 chemical substances in mixtures, recorded annually from the year 2000 onwards in Denmark, Sweden, Finland and Norway. SPIN data is derived from the NPG (Nordic product register) where the chemical composition and volume of all chemical products in Sweden is registered. However, it is important to mention that SPIN does not include articles that are imported to Sweden, which can also contribute to the environmental pollution burden in Sweden. For example an imported plastic toy using DEHP as plasticizer, which is discarded in the environment, can contribute to DEHP occurrence in the Swedish environment, most likely by leaching from the article. The same holds for PVC floorings, facades and other imported construction or consumer articles. Swedish officials are aware of this and are setting up a database for the inclusion of articles to the chemical burden currently. For this thesis only the use and potential emissions from chemical products (substances as such and mixtures) in Sweden are considered. Given the regulatory impact of REACH on the Swedish industry, SVHC use is expected to decrease, most likely resulting in decreased emissions of SVHC to the environment. Understandably, the decreased use of SVHC in the industry needs to be compensated for by substitution of the strictly regulated chemical.

In the following, the term "**regulated chemical**" is used for chemicals which are under **strict regulatory control** – with the aim to replace these substances. Strict regulatory control goes beyond registration. In this thesis this term is used for two groups of chemicals: chemicals of very high concern placed on the REACH Candidate list, and priority substances of the Water Framework Directive. For these priority substances use and emissions have to be reduced as far as possible in Europe.

1.2. The need for substitution of SVHC: Substitutes as emerging pollutants?

Substitution is “the replacement or reduction of hazardous substances in products and processes by less hazardous or non-hazardous substances, or achieving an equivalent functionality via technological or organizational measures” (Lißer et al., 2003). However, chemical-by-chemical substitution as currently the most widely applied option (Fantke et al., 2015). In many cases these chosen substitutes are of minor change in chemical composition which results in a similar hazard profile. This incremental change hampers the successful fade- out of hazardous chemicals and thereby the necessary shift towards green chemistry / sustainable chemistry (Blum et al. 2017) and cleaner surface waters. When a chemical substitution does not mitigate the negative environmental or human health impact and merely shifts the burden to another sector the substitution is termed "regrettable" (Fantke et al. 2015). Substitution of hazardous chemicals is strongly reinforced and promoted by chemical agencies such as the ECHA. Ideally, these substitutions lead to a functional substitution, meaning that the efficiency and cost are not substantially higher and the environment and human safety is improved. However, numerous substitutes of hazardous chemicals are termed “regrettable” such as the substitution of bisphenol A for bisphenol S. Regrettable substitution can impact many environment and health aspects. However, in this thesis the term regrettable is exclusively used for substitutes that are detrimental to the environment, especially surface waters, not taking into account possible detrimental implications for human health or other matrixes of the environment. Given the evidence of occurrence of substitute chemicals in surface water, stated by the NORMAN list of emerging substances, these substances can be considered as emerging substances. In order to determine if the emerging substance could also be an emerging pollutant, a risk assessment can be performed also taking the exposure level into account.

The term emerging pollutant suggests that the pollutant has not been considered as threat to humans or the environment before and was discovered just recently. However, this is not the case as many pollutants, such as lead, have been around for centuries but only recently came into public discussions. Sauvé and Desrosiers (2014) suggests that EPs should be termed “contaminants of emerging concern”. The emerging concern regarding specific substances is shaped and expressed by the NORMAN network. The NORMAN network was formed in 2005 to collect data on emerging pollutants and to encourage validation and harmonization of measurement methods and monitoring tools for an improved risk assessment (Brack et al., 2012). According to the network emerging pollutants are “substances currently not included in routine environmental monitoring programs and which may be candidate for future legislation due to its adverse effects and / or persistency” (NORMAN network, 2005). Moreover, the NORMAN network has a list of Emerging substances that “can be defined as substances that have been detected in the environment, but which are currently not included in routine monitoring programs at EU level and whose fate, behavior and (eco)toxicological effects are not well understood.” Chemical substances can be considered of scientific importance once the NORMAN network has put the substance on the NORMAN list, which expresses that the substance is of emerging concern for the environment according to NORMAN network specialists. Until today, the NORMAN network has classified EPs into 20 substance classes, of which “flame retardants”, “surfactants” and “plasticizers” are extensively discussed and researched with regard to their detrimental effect on surface waters in the scientific community. In the following section a short introduction to these three water relevant substance classes is given. Within this thesis those substance classes are called “technical use categories”. With accordance to the NORMAN network, they are named “flame retardants”, “surfactants” and “plasticizers”. The introduction of technical use categories is essential to the analysis of substitution patterns as one chemical is usually not replaced by solely one other substitute. Industry wide, regulated chemicals are likely to be replaced by numerous substitutes, depending on the industrial context and process the original chemical was used in. Most likely, the substitute fulfills the same technical function as the chemical it replaces. For example a plasticizer is likely to be replaced by another plasticizer. However, which exact plasticizer the substitute is going to be, is dependent on the process and industrial context of the chemical it replaces. Therefore, it is necessary to look at a list of plasticizers that could be used as substitutes.

Often, chemical classes within a specific technical function were used in the past and then some of them were regulated, such as for example phthalate plasticizers. One typical example is that DEHP as a highly used phthalate plasticizer entered the REACH Candidate List and was forwarded to the Authorization List. Many other phthalates were then further discussed and the whole chemical class of phthalates came into critical focus. Ultimately not only DEHP is expected to be substituted but most of the chemicals belonging to the class of phthalates. They are expected to be substituted by non-phthalates. This shift is called pattern of substitution in this thesis and applies to all technical use categories (flame retardants and surfactants alike) with different chemicals classes that are known to be problematic. Each of the technical use categories holds substance groups that have been used in the past and are now considered hazardous. In plasticizers phthalates for plasticizers are known to be hazardous, in flame retardants, halogenated flame retardants are commonly addressed as problematic, and surfactants alkylphenol ethoxylates are subject of scientific concern. Interestingly, also substitutes that are not in these criticizes chemical classes can pose problems and lead to regrettable substitution. An introduction to all the technical use categories and the known emerging substances from the respective chemical classes is given below for all technical use categories.

1.2.1. Flame retardants

Flame retardants (FRs) are necessary additives in electronics, textiles and plastics, to inhibit the flammability of products and have contributed greatly to fire safety over the last decades. The occurrence of FRs in the water ecosystems depends on their **production volumes, usage,** disposal, their persistence and their fate in the environment (their distribution between the environmental compartments is often characterized in a first step by the octanol water partition coefficient (Iqbal et al., 2017). Emission, water solubility and octanol water partition coefficient are the key elements for prediction of behaviors of flame retardants in fresh water ecosystems. A more detailed characterization of these elements is beyond the scope of this thesis as it addresses industrial use time trends, not substance properties.

The first EPs of public concern were halogenated flame retardants (HFRs). HFRs were found in the environment due to a strong industrial use between 1929 and 1977 (Sauvé and Desrosiers, 2014), where it became apparent that industrial activity is strongly connected to the emergence of new pollutants in the environment. Concerning surface water, the levels of HFRs in Swedish river systems seemingly indicate that the environmental release of these compounds is declining, yet high levels in sperm whales demonstrate that lower HFRs already have reached the deeper regions of the Atlantic Ocean (R.W.P.M. Laane, 2000). Knowing that most HFRs are lipophilic (expressed in high log Kow value) they bind well to particles and are therefore below detection limit in the water but accumulate in river sediments and fish (de Wit, 2002). These finding underline the importance of industrial use data analysis to quantify the potential emerge of novel pollutants right on the production site not only with respect to surface waters but other environmental matrixes. In order to understand the nature of novel flame retardants that are released in the environment and can potentially contribute to adverse effects, flame retardants are grouped into

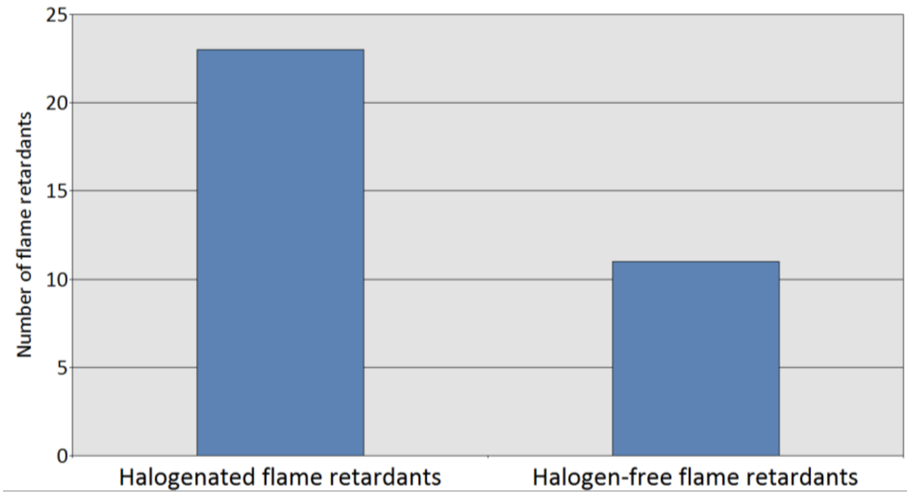

Figure 1: Number of emerging flame retardants by class

two classes in accordance with ENFIRO (EU Project: Environment-Compatible Flame Retardants) classifications i) Halogenated Flame Retardants (HFRs) and ii) halogen-free flame retardants (HFFRs). As described above, HFRs are widely under critique due to their known impact on the environment. Novel halogenated flame retardants that substituted the initial HFRs, have been proven slightly less, or even equally harmful to the environment (Iqbal et al., 2017), giving a typical example of regrettable substitution. During recent years, industrial substitution efforts were mostly directed towards replacing HFRs by HFFRs assuming that these are less harmful to the environment and human health. However, HFRs are not the only flame retardants that are industrially used and can be classified found in the European environment. When comparing the self- compiled flame retardant list with the NORMAN list of emerging substances it becomes apparent that 23 HFRs and also 11 HFFRs that are used in the Swedish industry were found in the European environment as shown in Figure 1. Therefore, substitution efforts do not need to be only directed at replacing HFRs but rather at replacing EPs as classified by the NORMAN network. Novel flame retardants should equally be included in future policies and regulations irrespective of chemical class but rather based on their classification as EP conducted by the NORMAN expert team.

1.2.2. Plasticizers

Plasticizers are a substance group used in polymer production to facilitate processing and to increase the flexibility of the final plastic product. An estimated 80- 90 % of the plasticizer use is in flexible polyvinyl chloride (PVC). Over the last 60 years more than 30,000 different substances have been evaluated for their plasticizing properties. Of these, approximately 50 are today in commercial use. The plasticizer list compiled for the thesis covers all high volume plasticizers, holding 80 plasticizers (Malveda, 2015) Plasticizers are grouped into phthalates, terephthalates, epoxy, aliphatics. trimellitates, polymerics and phosphates of which phthalates accounted for 70% of the world consumption of plasticizers in 2014. In 2005 the phthalate marked share accounted for 88% and in 2019 a further decrease to 65% of world consumption is expected (Malveda, 2015). The decrease in market share can be explained by fading out of the main phthalate plasticizer

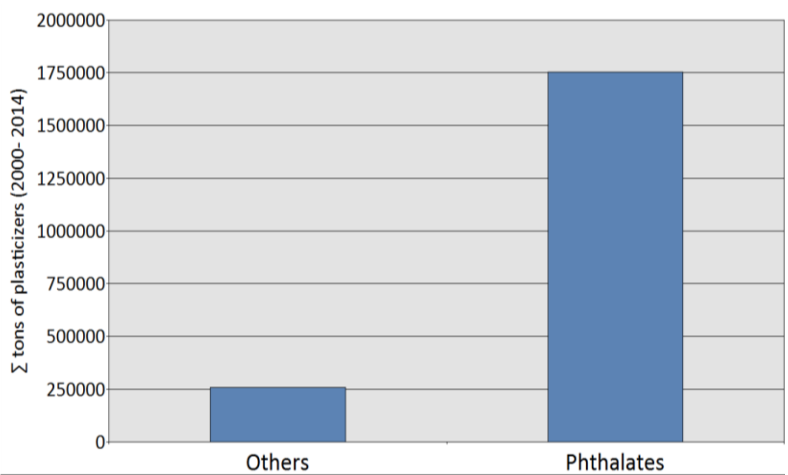

Figure 2: Quantitative dominance of phthalate plasticizer use in Sweden

DEHP. As mentioned DEHP is strictly regulated in many regions due to its human health and environmental threats and therefore replaced. For Sweden it was corroborated that phthalates were quantitatively far more important than other plasticizers as shown in Figure 2.

During the last decades multiple efforts were put into substituting DEHP and other phthalates by other non-plasticizers. However, a comparison of the self- compiled plasticizers list with the NORMAN list of emerging substances shows that a total of 13 non-phthalate plasticizers are considered by the NORMAN list of emerging substances and were registered on the Swedish SPIN database as substances that are industrially used in Sweden. There should be awareness in the industry, that there are non-phthalate plasticizers that are also considered by the NORMAN list of emerging substances and can potentially threaten the environment. Uninformed substitution of a phthalate for an apparently less hazardous non- phthalate plasticizer is the basis for regrettable substitution.

Again, the occurrence of plasticizers in the water ecosystems depends not only on their production volumes and usage, disposal, their persistence and partition behavior. Most plasticizers are characterized by a high log K_{ow}, indicating poor water solubility, pointing to accumulation of plasticizers in fatty tissue. Due to the high fat solubility and structural similarity to estrogen numerous plasticizers such as BPA and phthalates are characterized as endocrine disrupting chemicals (EDCs) (Fromme et al., 2002). Plasticizers are often not covalently bound to the polymer matrix of the plastic and can therefore leach from plastics into the environment. Therefore, the major source of plasticizer contamination in the environment is landfill leachate of disposal sites where plastics are discarded (Zheng et al., 2007). Again, the analysis and modelling of the behavior of plasticizers in water is beyond the scope of this thesis as it exclusively addresses industrial use data time trends to identify potentially emerging pollutants.

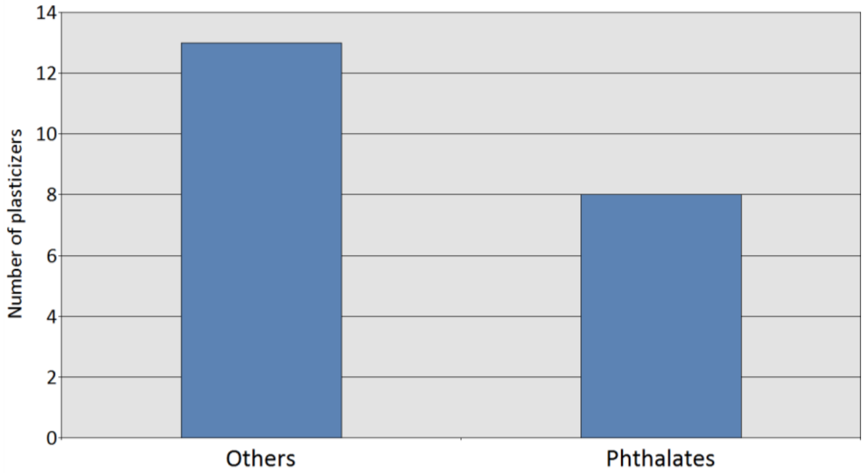

Figure 3: Number of emerging plasticizers by class

1.2.3. Surfactants

Surfactants are compounds that lower surface tension between molecules and constitute the main components in detergents, wetting agents, emulsifiers and dispersants, and foaming controls. There are various classes of surfactants of which the most widely used are anionic surfactants, such as linear alkylbenzene sulfonate (LAS) used for cloth and dish washing, detergents and shampoos with global market share of 53% (Ceresana, 2015). Another high volume surfactant is the nonionic surfactant, are alcohol ethoxylates (AE) (Cowan-Ellsberry et al., 2014). Most laundry detergents contain both nonionic and anionic surfactants because nonionic surfactants contribute to making the surfactant system less sensitive to water hardness. The end use of these high volume surfactants is in laundry detergents, dishwashing detergents, household cleaners, and personal care products both in the home, industrial, and institutional applications. These applications will result in release to the environment, primarily in wastewater discharges, which makes surfactants especially interesting when considering EP in surface waters. Surfactants are hazardous to aquatic organisms due to their surface active properties and numerous detergents exhibit estrogenic activity, especially alkylphenols like nonylphenols (NPs) and octylphenols (OPs), which are degradation products of the widely used alkylphenol ethoxylate surfactants (APEs). Due to their low biodegradability, the toxicity of NPs and endocrine disruptive characteristic APEs are priority hazardous substances in listed in the Water Framework Directive 2000/60/EC and the final European Union decision No. 2455/2001/EC (Lopez de Alda et al., 2003). The European Parliament approved market and use restrictions in 2003 so APEs cannot be used at 0.1% or greater in a wide variety of applications. Common substitutes are linear alcohol ethoxylates (AEs) (Burlington Chemical Co., L. L.C and Leuk, 2011), a class of surfactants that has no compound listed on the NORMAN list of emerging substances due to rapid biodegradability. Yet, there are other classes of surfactants that hold substances considered as ES. Especially, quaternary ammoniums that are used for softening purposes in hair rinsing and in fabric softeners and the high

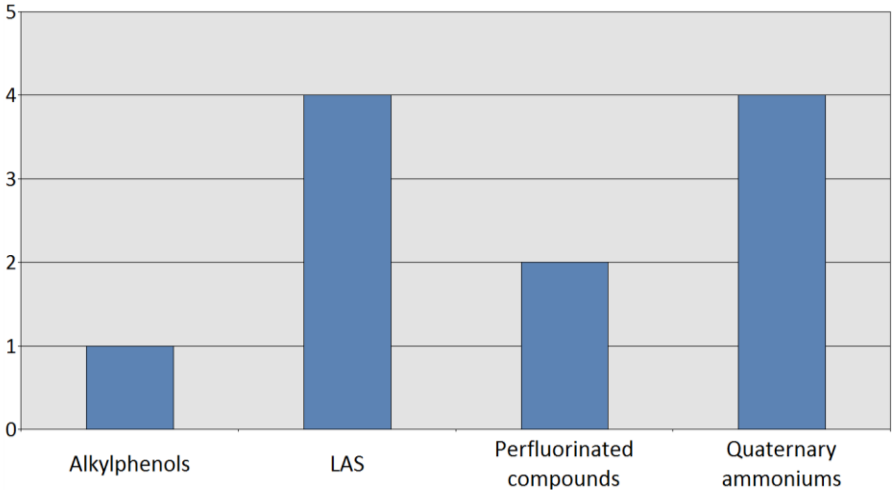

Figure 4: Number of emerging surfactants by class

volume calls LAS holding cloth and dishwashing detergents both registered 4 substances in Sweden that are listed on the NORMAN list.

1.3. Other substances of emerging concern: Persistent and mobile organic chemicals (PMOC)

Persistent and mobile organic chemicals (PMOC) are found to be very water relevant by an expert team (Stefanie Schulze, Daniel Zahn, Rosa Montes, Rosario Rodil, José Benito Quintana, Thomas P. Knepper, Thorsten Reemtsma, Urs Berger) as they are highly mobile and a persistent thus fulfilling important criteria to become an a pollutant of emerging concern. PMOC depict a threat to the quality of our water resources as they are highly polar can pass through wastewater treatment plants, subsurface environments and potentially also drinking water treatment processes. Very few compounds are known in terms of analysis, monitoring, water treatment and regulation (Reemtsma et al., 2016). This was corroborated comparing the list of 43 PMOC to the NORMAN list of EPs on which only 3 PMOC are already listed. Even though PMOC are not found on the candidate list, this thesis takes PMOC into consideration for the lack of regulative legislation and the evident detrimental impact on surface waters, which makes them very potent EPs. Moreover, they can be used as a control group for the regulated wrSVHC as they are widely unregulated.

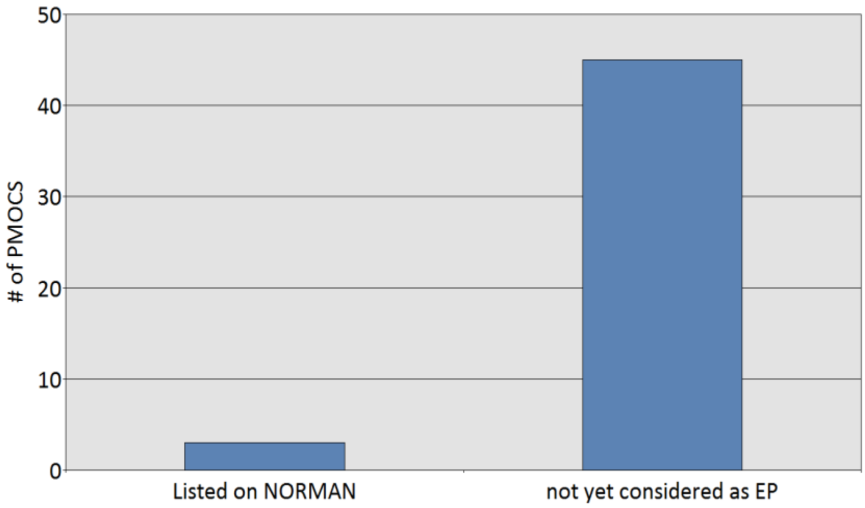

Figure 5: Number of PMOC already considered by the NOMAN list

In the previous sections all substance groups and potential pollutants of emerging concern have been introduced and the relevance of regulation in the emergence of novel substance and/or pollutants in the environment has been explained. It became clear that there are various knowledge gaps: 1) currently there is no indicator to evaluate the impact of (REACH) regulation on the use of problematic chemicals. 2) There might be patterns of substitution could lead to emerging substances from chemical classes that were thought to be safe alternatives.

To fill these knowledge gaps the SPIN database was used as analytical tool of choice. The following section describes the SPIN database including opportunities and limitations.

1.4. Introduction to NPG & SPIN

The Nordic product registers (NPG) are worldwide under the most comprehensive databases with respect to completeness of information and the number of registered products and substances. NPG data have already been used for risk assessments, substance flow analyses and many more purposes (NPG, 2007). The data in the NPG is based on information from four individual countries: Denmark, Sweden, Finland and Norway. The respective national legislation requires manufacturers and importers of chemical products to inform their respective product register about function, industrial category, classification, composition and quantity of the substance/product. However, there are differences in reporting and the type of data required by the respective countries, depending on the country where the substance/product is manufactured. For the purpose of the thesis the data from the Swedish product register is used. In the following section, opportunities and shortcomings of this data are described and explained. The detailed data analysis can be found in the appendix 8.2. The Swedish product register is represented by KEMI (Swedish Chemicals Agency) and contains information on the largest numbers of chemical products and the highest proportion of those on the market. All chemical products that are produced or imported into Sweden with more than 100 kg/year have to be registered annually with respective quantities, classification; codes for areas of use and the codes for product types. The declaration requirements apply to all chemical products, substances and mixtures alike according to the custom tariff codes. The NPG passes data on to the SPIN database, given the data is not confidential.

1.4.1. SPIN Data: Opportunities and Limitations

SPIN (**S**ubstances in **P**reparations **i**n the **N**ordic countries) holds “non- confidential” data on substances as such and in mixtures derived from the confidential data in the NPG. SPIN publishes as much data as possible to without threatening individual company’s confidential data by aggregating data. SPIN can be used by the public. Research institutes, governments or individual researchers are encouraged to use SPIN data for their purposes. The information included in the SPIN database is the number of products containing the substance, the annual tonnage of a given substance used in mixture and the industrial categories and use categories of the mixture the substance is part of. Moreover, the annual tonnage within these categories and the presence or absence of the substance in consumer products is registered in SPIN (NPG, 2007). This enables to analyze time trends for substances and the number of the substances is used in. As compared to other databases of “in-use” data, such as ProdCom from Eurostat, SPIN holds substances with individual and unambiguous CAS number as compared to substance groups, which are used in ProdCom. In addition SPIN covers polymers, a group of substances not registered elsewhere in Europe and also exempted from REACH (Ahrens and Reihlen, 2007). There are various features of interest in SPIN that can be taken into account for analysis and risk assessment of EPs, such as the exposure toolbox, which describes the risk of chemical substances to humans and the environment by various indices in numerical form but was evaluated to be insufficiently consistent in the course of this thesis as shown on 8.2 in the appendix.

SPIN offers data on chemical products as defined above (substances as such and mixtures), thus neither ***non-chemical products nor articles are included***. Foodstuffs, cosmetics, medicinal products, biocides, pesticides, pure heavy metals and quantities of chemical products less than 100 kg/year per company are not obligatorily included but can still be mentioned. There are no data about substances in articles only about substances in chemical products (KEMI, 2017). The saddle of a bicycle, for instance is an article, and often contains softeners and it may be painted. The saddle would not be listed in SPIN. However, substances in the paint and the softener are expected to be accounted for in SPIN if the saddle was produced in Sweden as they were once in a mixture (paint or polymer granulate). These substances are not accounted in SPIN if the article was imported. This mainly hampers the evaluation of emerging pollutants from leaking substances from imported articles, such as plasticizers that are mainly imported from Asia.

Another shortcoming of SPIN is that some substances are subject to ***confidentiality*** and are only mentioned in by their name but not as total quantities. Substances do not have to be reported in SPIN if the substance is reported to the NPG by less than 3 companies or in less than 4 preparations. This confidentiality issue can depict a major source of data exclusion especially for SVHC that are used in very specific processes by one company in one or two products but could still be used in large quantity. The impact of confidential data in Swedish SPIN data was analyzed and specified in the appendix 8.2.2. The analysis shows that there are many confidential data; however, the total volume of these confidential data is not expected to be of major importance.

SPINs information on areas of use (industry sector or function of chemical) is only available for 25% of the substances (Ahrens and Reihlen, 2007), making the ***use categories unreliable*** for analysis, which was confirmed by the data analysis in 8.2.2 the appendix. SPIN furthermore describes potential sources of error with regard to total quantities such as ***double registration of chemical products***. This risk is estimated to be between 20% and 50%, being more prevalent in the categories raw materials (11 July 2017; 18 August 2017). However, the double counting always overestimates the actual quantity, therefore adding a buffer zone, which is not detrimental with regard to risk assessment.

Having described all important shortcomings and pitfalls of SPIN, 144 of the 173 entries of the REACH Candidate list can be found in the Swedish SPIN total use database. It is important to emphasize that one SVHC entry can stand for one chemical with unambiguous CAS, but some entries do hold a group of chemicals. The one entry one substance scenario is true for 132 SVHC. The remaining entries hold more than one chemical with at least 2 CAS numbers. These entries hold groups of chemicals because there are closely related chemicals like isomers that are easier classified with one entry but several CAS numbers. In total 12 of the 144 SVHC entries hold more than one CAS as illustrated in Table 1. For 22 entries (13%) no data was found. It has been analyzed whether this fraction could be relevant for the total amount used. The analysis showed that the missing substances probably only have a negligible impact on the total volume (see appendix 8.2.1). Concluding the opportunities and limitations of SPIN to retrieve time trends for individual substances, the table shows the data richness of SPIN with respect to the SVHC entries from the REACH candidate list. From the **173** entries (substances or substance groups) listed on the Candidate list, data for **144** entries were found. This means: the SPIN database covers 84% of all entries of the REACH Candidate list.

Table 1: Number of SVHC entries on the REACH Candidate list for which data are found in SPIN, and related number of substances per entry.

	Total	1 CAS #	2 CAS #	3 CAS #	4 CAS #	5 CAS #	8 CAS #	24 CAS #
Identification of SVHC entries								
# of entries	**173**	157	5	6	1	1	1	2
% of entries	100%	90,8%	2,9%	3,5%	0,6%	0,6%	0,6%	1,2%
SVHC entries found in SPIN								
# of SVHC entries in SPIN	**144**	132	4	4	0	1	1	2
% of SVHC entries in SPIN	83%	84%	80%	67%	0%	100%	100%	100%

No other database in Europe is known which has a similar amount of publicly available data. Therefore, SPIN can be considered as a valuable tool for identification and prioritization of industrially used chemicals. It represents a data base for the majority of chemicals. The ones not registered on SPIN have been demonstrated to be of low quantitative importance (Appendix Table 10: Quantitative relevance of SVHC absent on SPIN) or exempted from SPIN but available on other databases (pharmaceuticals, heavy metals). Despite the huge amount of data, SPIN has not been used before to calculate and compare time trends for water relevant SVHC and their substitutes. It has been used to show time trends for specific groups of substances, e.g. plasticizers (KEMISTAT, 2012). The figures from these earlier assessments have been used to validate the methodology applied in this thesis.

2. Goal & Research Questions

The goals of this thesis are an understanding of the impact of regulation on the use of SVHC, the patterns of substitution of chemicals (which chemicals are substituted and by what) and the identification of substances that could constitute future EPs in Swedish surface waters.

To address these goals, the Nordic SPIN database was used to retrieve time trends of water relevant substances of very high concern (wrSVHC) to develop a quantitative picture of time trends for the use of problematic substances and their substitutes. The analysis is based on industrial use data for chemicals. A methodology has been developed which allows answering the following research questions: I) Is the SPIN database a workable and data-rich database for time trends of industrial chemicals? II) Does strict regulatory action (such as placing on the REACH Candidate list) cause a reduction in the amount of water relevant chemicals used? III) Does strict regulatory action cause increased use of substitutes and can patterns of substitution be detected? IV) Can substitutes be identified which have the potential to be future emerging substances and possible pollutants? V) Does the identification of substitutes allow for future monitoring recommendations?

These research questions are described in more detail in the following sections.

1) *Is the SPIN database a workable and data-rich database to develop time trends for industrial chemicals?*

The starting point of this thesis was to find a way to gather industrially used tonnage data from national chemical registers to develop time trends for selected chemicals. To this end the SPIN database needed to analyzed in terms of reliability and utility for time trend analysis and other features of the database that promise risk assessment, such as the ExposureTool. These results are found in the appendix on 8.2.

2) *Regulative impact of REACH candidate list on water relevant SVHC use in Sweden*

The first hypothesis was that chemicals under regulatory pressure will be used in smaller quantities. Therefore, the regulative impact on wrSVHC under the REACH candidate list was quantified for individual wrSVHC used in Sweden. To compare the impact of strict regulation with less regulated problematic substances with water relevance, time trends for a non-regulated class of persist mobile chemicals (PMOC) are analyzed and compared to the under REACH regulated wrSVHC. The assumption is that wrSVHC decrease drastically due to the regulation under REACH as compared to less strictly regulated PMOC.

3) *Identification of substitute chemicals and patterns of substitution*

In accordance with the expected decrease of regulated wrSVHC use, an increase in substitutes is expected. The substitutes are grouped into three technical use groups “plasticizers”, “flame retardants", "surfactants". As one regulated problematic substance can lead to numerous different substitutes as described in chapter 1.2., technical use categories are implemented to follow the patterns of substitution.

4) *Identification of substitutes which are already listed as emerging substances*

The ultimate goal is to identify substitutes that are increasing in use in Sweden and can already be found in the European environment. The hypothesis is that substances already found in surface waters in Europe are likely to be found in Swedish surface waters too given that they are increasingly used and possibly emitted to the environment. The final emission modelling and risk estimation needs to be done by Swedish officials that have access to emission data.

5) *Identification of substitutes as bases for future monitoring recommendations*

In order to develop monitoring recommendations for Sweden, the industrially used substitutes with increasing trend need to be compared to the existing monitoring data for surface waters gathered in the NORMAN EMPODAT database. EMPODAT holds all European monitoring data in various environmental matrixes. Given, that there is no entry for the increasing chemical and physio-chemical characteristics that indicate slow degradation in the environment, monitoring could be recommended.

3. Methods

In order to answer all research questions, a multitude of tests and queries has been conducted using the SPIN database and various other data sources described in detail below.

3.1. Data collection:

The relevant databases (SPIN SQL database, REACH candidate List, SIN list, NORMAN list, WFD substance list) were downloaded during May 2017 from following websites: REACH Candidate List (ECHA, 2017a), Norman List (http://www.normandata.eu/?q=node/19), Norman suspect list (http://www.norman-network.com/datatable/), SIN list (http://sinlist.chemsec.org/) SPIN database (http://www.spin2000.eu/), WFD priority substances and certain other pollutants (http://ec.europa.eu/environment/water/water-framework/priority_substances.htm). The REACH candidate list was completed with chemicals names and CAS numbers as some substances in the candidate list are listed as groups of substances. Therefore, individual CAS numbers needed to be allocated for all of the representatives of the respective group. The original 173 SVHC that included substance groups were processed into 250 individual substances with unambiguous CAS number. The individual CAS numbers were found via ECHA where support documents supply an exhaustive list of the CAS numbers (https://echa.europa.eu/de/candidate-list-table). This was accessed and conducted during May 2017. Moreover, an account for the NORMAN EMPODAT database (http://www.normandata.eu/empodat/) was created to access the European monitoring data for emerging substances. The SIN (http://sinlist.chemsec.org/) and SINimilarity (http://sinimilarity.chemsec.org/) list were used for qualitative analysis of the identified emerging substances

3.2. Definition of groups with water relevance:

wrSVHC

Surface water relevant SVHC were defined as SVHC present in the NORMAN list or on included in the WFD. As a result 16 wrSVHC were found on SPIN with end-to-end time trends fit for analysis. A total of 16 wrSVHC were defined 9 from the WFD of which 3 were also present on the NORMAN list and 7 that are exclusively found on the NORMAN list. All wrSVHC with respective abbreviation code and CAS number are found in the appendix 8.1.18.1.

Flame retardants

The list of flame retardants was compiled by research for common substitutes for the flame retardants wrSVHC [Tris(2-chloroethyl)phosphate (CAS: 115-96-8), Short Chain Chlorinated Paraffins (CAS: 85535-84-8), DecaBDE (CAS: 1163-19-5)], HBCDD (CAS: 3194-55-6)] on SUBSPORT and the SIN (substitute it now) list. Further, the list was extended with flame retardants from the NORMAN network supplemented by flame retardants from literature reviews (Covaci et al., 2011; Iqbal et al., 2017; R.W.P.M. Laane, 2000; Richardson and Ternes, 2014; NORMAN network, 2005). In total, the list holds 83 flame retardants and is found in chapter 8.1.2 in the appendix.

Plasticizers

The list of plasticizers was compiled by research for common substitutes of wrSVHC plasticizers [DEHP (CAS: 117-81-7), BBP (CAS: 85-68-7), DBP (CAS: 84-74-2), BPA (CAS: 80-05-7)] on SUBSPORT and the SIN list. Further, the list was supplemented with other plasticizers retrieved from the NORMAN network (NORMAN network, 2005) the German Environmental Specimen Bank (German Environmental Specimen Bank, 2017) and some relevant publications (Zheng et al., 2007; Fromme et al., 2002). The plasticizer list aims to cover all high volume plasticizers, holding 80 plasticizers found in chapter 8.1.3 in the Appendix.

Surfactants

The list of surfactants was compiled by research for common substitutes of wrSVHC surfactants [4-Nonylphenol, branched and linear, ethoxylated (CAS: 25154-52-3), 4-Nonylphenol, branched and linear (CAS: 104-40-5), 4-(1,1,3,3-tetramethylbutyl)phenol (CAS: 140-66-9)] on SUBSPORT and the SIN list (substitute it now). The surfactant list holds 133 of surfactants and was complemented by a literature Survey of Surfactants in the Nordic Countries (Johansson et al., 2012), the surfactants from the NORMAN network (NORMAN network, 2005), the HERA report (Environmental HERA report, 2013) and a book with an extensive list on APEs and AEs (Talmage, op. 1994) found in the appendix in chapter 8.1.4.

Exceptions

For Benzo[a]pyrene (CAS 50-32-8) no substitutes were found on SUBSPORT or SIN. BaP belongs to the polycyclic aromatic hydrocarbons (PAHs). They are formed as a side product during incomplete combustion or pyrolysis of organic material. Benzo[a]pyrene is not used on purpose and therefore will not be substituted by another chemical.

Persistent and mobile organic contaminants (PMOC)

The PMOC list was obtained by a cooperation with Dr. Reemtsma and holds 59 substances considered to be water relevant by an expert team (Schulze et al., 2017). Of those, 43 substances were found in SPIN of which 12 had end-to-end time lines. These are found in chapter 8.1.5 in the appendix.

3.3. Software and databases:

The SPIN database was operated and managed in Microsoft Access (Version: 14.0.7181.5000 (32-Bit)) and the relevant lists mentioned above (PMOC, plasticizers, etc) were incorporated for analysis. SQL queries were conducted and further data modification and analysis was carried out in Microsoft Excel (Version: 14.0.7181.5000 (32-Bit)).

3.4. Data processing:

Development of regulated wrSVHC in Sweden

In order to compare the situation before REACH and under REACH (“before and under”) - REACH scenarios) a percentage change in use volumes was calculated for individual wrSVHC. Due to annual fluctuation of the tonnages an average of the tonnages in the years 2000, 2001 and 2002 were calculated and compared to the average of years 2012, 2013 and 2014.

Development of unregulated PMOC

SPIN annual tonnage data for all PMOC was retrieved. 12 PMOC of quantitative relevance (<5t in 2004) were selected and plotted (see Figure 8: Time trends of unregulated PMOC in Sweden).

Comparison of regulated wrSVHC and unregulated PMOC in Sweden

Data for all wrSVHC and PMOC in 2004 and 2014 were retrieved via an SQL query in the database. These years were picked, as in the early years some substances were subject of underreporting and a ten year time span seemed adequate to compare before REACH and with REACH. All substances that yielded end-to-end time trends without confidential data were selected and plotted in **Fehler! Verweisquelle konnte nicht gefunden werden.**.

Development of flame retardants

The flame retardant list was subset into HFRs and HFFRs as seen in Appendix Table 2: Compiled flame retardants list. Annual tonnage data was retrieved via an SQL query from SPIN. However, aluminum hydroxide (CAS: 21645-51-2) had to be excluded from the HFFR list, due to it being a high volume substance that is predominantly used in water purification as flocculation agent and only secondarily used as flame retardant. In order to identify flame retardants substitutes SPIN was searched by an SQL query for all flame retardants present in Sweden. The resulting list held 77 flame retardants and was then manually filtered for substances exhibiting upwards trends, resulting in 11 substances. EPs were identified by intersecting the flame retardant list with the NORMAN list of EPs as attached in Appendix Table 3: Flame retardant ESs. In this table, flame retardants with increasing use volumes ("concerning trends") are marked in red.

Development of plasticizers

The plasticizer list was subset into phthalates and others as seen in the Appendix Table 4: Compiled plasticizer list. Plasticizer substitutes were identified by a SQL query in SPIN for all plasticizers present in Sweden. The resulting list held 68 plasticizers. It was manually filtered for substances exhibiting upwards trends, resulting in 16 substances. EPs were identified by intersecting the plasticizer list with the NORMAN list of EPs as attached. EPs with concerning trend are marked in red as shown in

Appendix Table 5: Plasticizer ESs. Of the plasticizer list with 80 substances 21 substances were listed on NORMAN and found in the Swedish SPIN data. Of the 21 Substances 2 have at least in one year confidential tonnages. 19 substances have end-to-end time trends. Of those 3 shows concerning trends.

Development of surfactants

The surfactant list was subset into alkylphenol ethoxylates, alcohol ethoxylates and other as seen in Appendix Table 6: Compiled surfactant list. Surfactant substitutes were identified by a SQL query in SPIN for all surfactants present in Sweden. EPs were identified by intersecting the plasticizer list with the NORMAN list of EPs as attached EPs with concerning trend are marked in red as shown in Appendix Table 7: Surfactant ESs

Appendix Table 7: Surfactant ESs. Of the 133 substances 11 substances were listed on NORMAN and found in the Swedish SPIN data. Of the 11 substances 5 have at least in one year confidential tonnages. 6 substances have end-to-end time trends. Of those 3 shows increasing trends.

4. Results

4.1. Regulative impact of REACH on SVHC use in Sweden

The following results show that industrial use of substance groups under strict regulation (REACH regulation, in combination with the WFD) strongly decreased over time. It was shown that each individual wrSVHC developed independently but nevertheless with similar trends.

4.1.1. Development of regulated wrSVHC in Sweden

The time trends for 15 wrSVHC show a clear decrease, however 1-2, dichlorethane is sharply increasing. This figure also serves as first impression of SPIN data, with strongly fluctuation curves. For unambiguous identification of the SVHCs see Figure 7: Cumulative time trends of regulated wrSVHC in Sweden

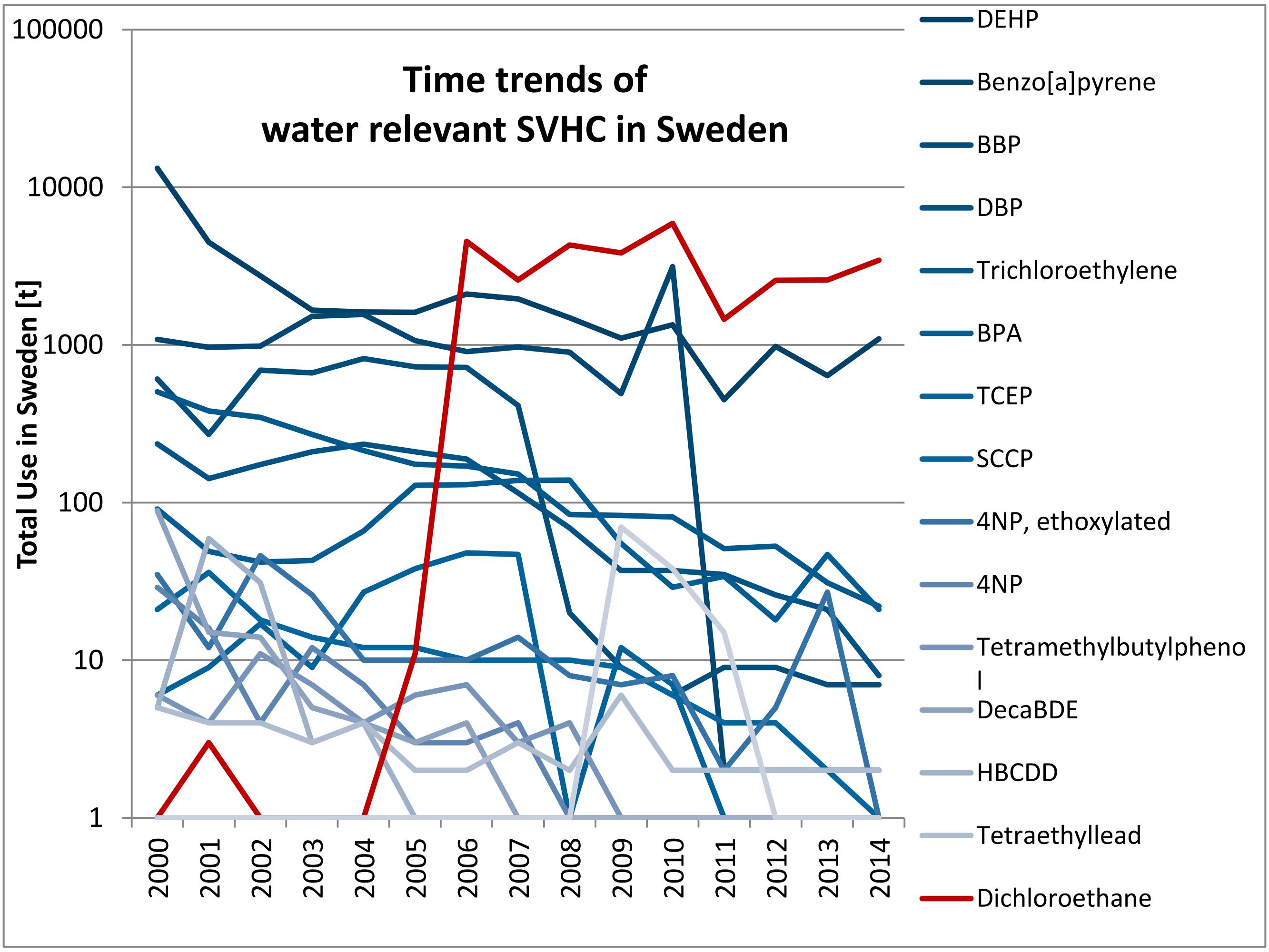

Figure 6: Time trends of surface water relevant SVHC use in Sweden

The sharp increase of 1-2, dichlorethane coul originate through a lack of reporting in the early years as shown in red in Figure 6. An other possible explaination is that 1-2, dichlorethane is used as an intermediate under strictly controlled conditions reacting to vinyl chloride monomers which is the precursor of polyvinyl chloride (PVC). Under REACH these intermediates are strictly controlled and therefore exempted from authorization, which means that they can be used unrestricted as intermediates. As 80% of the 1-2, dichlorethane is used as intermediated in PVC production, the use data is also not expected to decline. Therefore, 1,2- Dichlorethane is exempted from further consideration. All other wrSVHC are used less over time in Sweden.

The percentage change of every wrSVHC was calculated indicating a notable decrease in all substances across all chemical groups as shown in Table 2.

Table 2: Use volumes of individual wrSVHC, before REACH and under REACH

CAS	Name	Tons before-REACH	Tons under REACH	% change	Chemical Group
117-81-7	DEHP	6816	904	-87%	Surfactant
50-32-8	Benzo[a]pyrene	1011	2	-100%	Surfactant
85-68-7	BBP	524	8	-99%	plasticizer
84-74-2	DBP	184	18	-90%	Others
79-01-6	Trichloroethylene	411	35	-91%	plasticizer
80-05-7	BPA	61	29	-53%	Others
115-96-8	TCEP	11	1	-91%	plasticizer
85535-84-8	SCCP	25	2	-91%	Flame retardant
25154-52-3	4NP, ethoxylated	31	11	-65%	plasticizer
104-40-5	4NP	16	1	-94%	Others
140-66-9	Tetramethylbutylphenol	7	1	-86%	Flame retardant
1163-19-5	DecaBDE	39	1	-97%	Flame retardant
3194-55-6	HBCDD	32	1	-97%	Flame retardant
78-00-2	Tetraethyllead	4	2	-54%	Others
7440-43-9	Cadmium	1	1	0%	Others

Together, all wrSVHC (exempting 1-2, dichlorethane for previously named reasons) show a clear time trend pattern, confirming the restrictive impact of REACH and other regulations (see Figure 7).

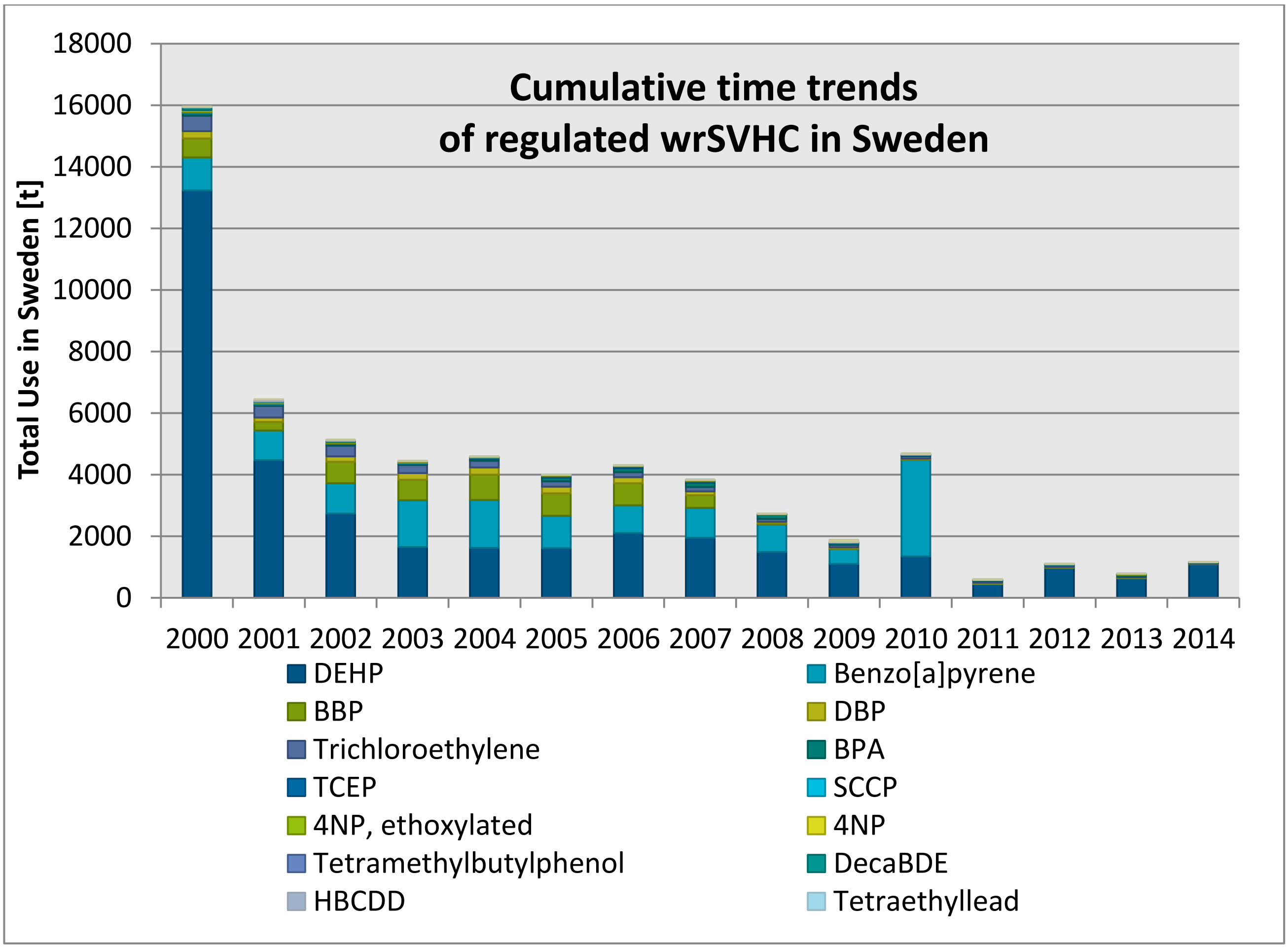

Figure 7: Cumulative time trends of regulated wrSVHC in Sweden

4.1.2. Development of unregulated PMOC

As compared to regulated SVHC there is no overall decrease pattern in unregulated PMOC as shown in Figure 8. PMOC are not in political focus and non- regulated, therefore depicting a control group of substances to the regulated wrSVHC.

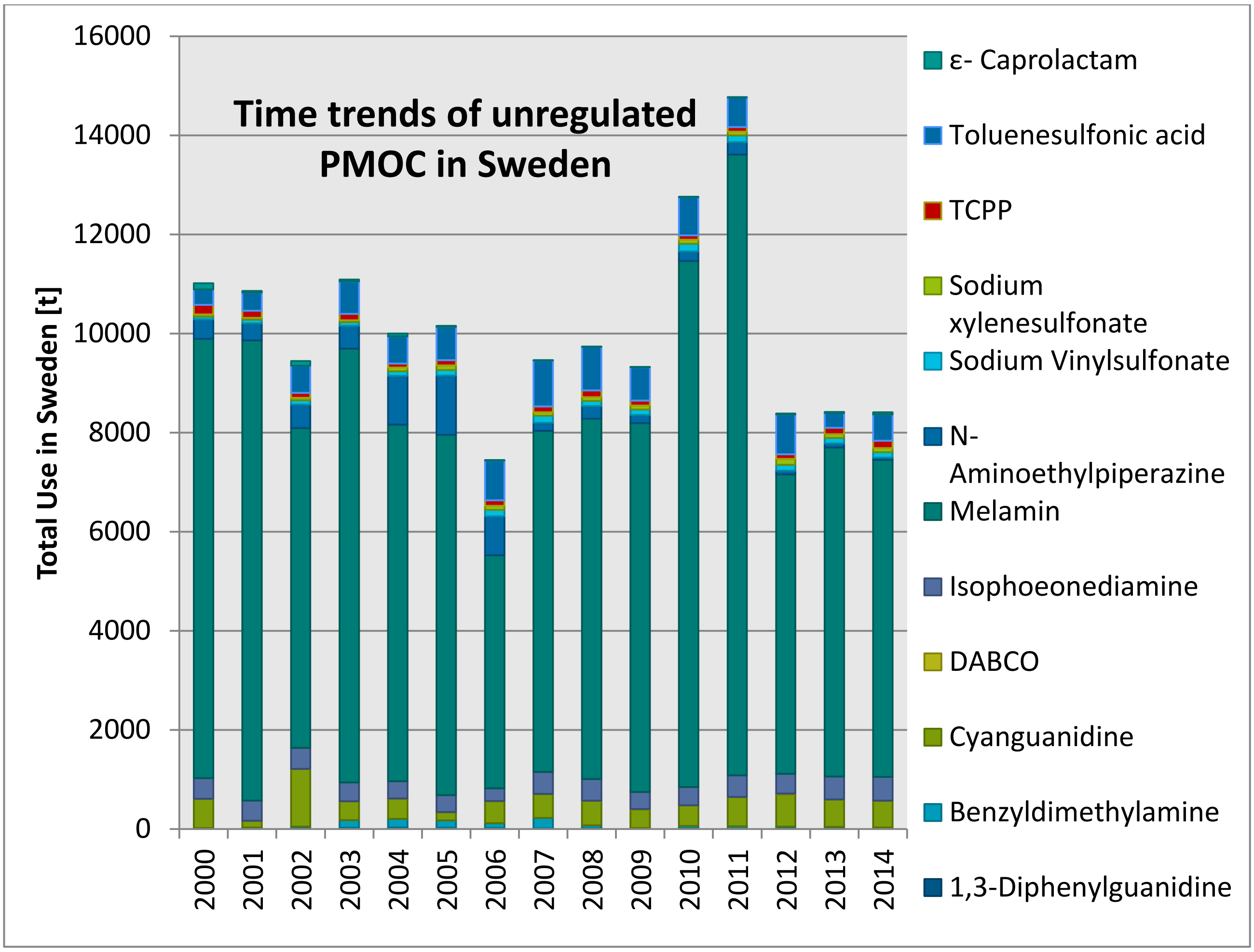

Figure 8: Time trends of unregulated PMOC in Sweden

In unregulated PMOC there is use trend found over time. Several fluctuations mainly in the highly used melamine constitute variety, however no clear trends are found. This confirms that the use of unregulated substances does not decrease over time.

4.1.3. Comparison of regulated wrSVHC and unregulated PMOC in Sweden

Given the clear decreasing time trend seen in the regulated group of wrSVHC and no trend pattern in unregulated PMOC, these groups are compared in a scatterplot with industrial use data from 2004 as pre- REACH scenario and industrial use data from 2014 as post- REACH scenario. As shown in **Fehler! Verweisquelle konnte nicht gefunden werden.** all regulated wrSVHC decreased from 2004 to 2014 whereas numerous of the PMOC stayed on similar levels.

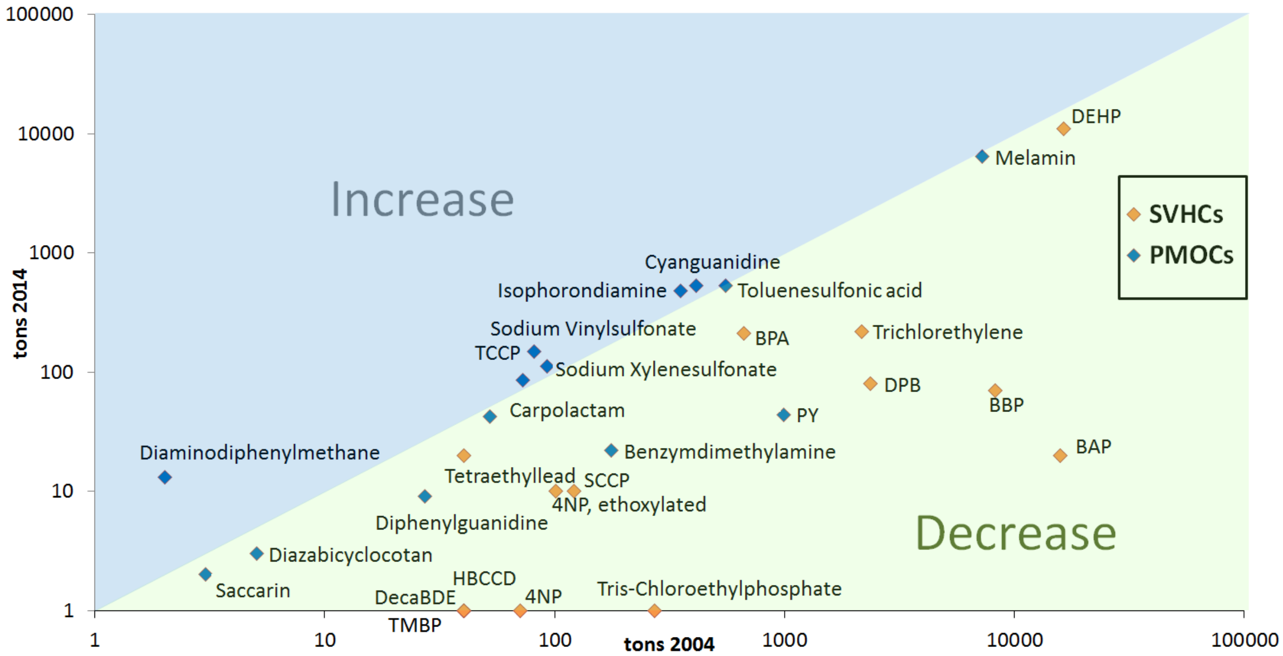

Figure 9: Comparison of regulated wrSVHC and unregulated PMOC

The decrease of wrSVHC is expected to be accompanied by an increase in substitutes according to the defined use of the chemicals. However, the decrease in regulated SVHC could also be partially be explained by other regulations. Therefore, a policy framework analysis for Swedish policies was conducted in order to identify potential confounding. A Swedish institution (IVL) conducted the policy framework analysis in collaboration. IVL compiled a review of EU regulations, EU directives and multilateral environmental agreements into a database (Lexén et al., 2015). WrPMOC and wrSVHC have been searched in database via a SQL-script to see if and in which legislation they are regulated in. As seen in Table 3, PMOC are seldom listed in regulatory frameworks, whereas all wrSVHC are by definition regulated under the REACH candidate list and additionally appear on numerous other regulatory frameworks.

Table 3: Regulatory framework analysis for PMOC and wrSVHC in Sweden

Class	Name	CAS	REACH Annex 14	REACH Annex 17	Drinking Water Directive	PRTR, E-PRTR	WFD	Other
PMOC	Melamin	108-78-1						
PMOC	Aminoethylpiperazine	140-31-8						
PMOC	Toluenesulfonic acid	104-15-4						
PMOC	Cyanguanidine	461-58-5						
PMOC	Isophoeonediamine	2855-13-2						
PMOC	Benzyldimethylamine	103-83-3						
PMOC	Sodium Vinylsulfonate	3039-83-6						
PMOC	TCPP	13674-84-5						
PMOC	Sodium xylenesulfonate	1300-72-7						
PMOC	ε- Caprolactam	105-60-2						Cosmetic Product Regulation
PMOC	Diphenylguanidine	102-06-7						
PMOC	DABCO	280-57-9						
wrSVHC	DEHP	117-81-7	x	x		x	x	Cosmetic Product Regulation
wrSVHC	Benzo[a]pyrene	50-32-8					x	Cosmetic Product Regulation
wrSVHC	BBP	85-68-7	x	x				Cosmetic Product Regulation
wrSVHC	DBP	84-74-2	x	x				Cosmetic Product Regulation
wrSVHC	Trichloroethylene	79-01-6	x		x	x	x	
wrSVHC	Bisphenol A	80-05-7						
wrSVHC	TCEP	115-96-8	x					Cosmetic Product Regulation
wrSVHC	SCCPs	85535-84-8				x	x	Cosmetic Product Regulation
wrSVHC	4NP, ethoxylated	25154-52-3		x			x	Cosmetic Product Regulation
wrSVHC	4NP	104-40-5					x	

wrSVHC	Tetramethylbutylphenol	140-66-9					x	
wrSVHC	DecaBDE	1163-19-5						
wrSVHC	HBCDD	3194-55-6					x	
wrSVHC	Tetraethyllead	78-00-2						Cosmetic Product Regulation, Rotterdam Convention
wrSVHC	1,2-dichloroethane	107-06-2	x		x	x		Cosmetic Product Regulation, Rotterdam Convention

Supplementary information:

Reach Annex 14: Authorization

Reach Annex 17 Restrictions

Drinking Water Directive (98/83/EC)

Ground Water Directive (2006/118/EC)

Protocol on Pollutant Release and Transfer (PRTR, E-PRTR)

Water Framework Directive (2000/60/EC) (WFD priority substances Annex I, WFD priority hazardous substances Annex I)

4.2. Flame retardants

4.2.1. Identification of substitute flame retardants

8 flame retardants are used in Sweden in clearly increasing amounts. It is reasonable to assume that these flame retardants are used as substitutes for strictly regulated flame retardants (which show decreases in the amount used), 7 of these substitutes are halogen-free flame retardants. The halogenated flame retardant, ammonium bromide is marked in red, indicating a regrettable substitution of a restricted halogenated substance by another, structurally closely related substance. However, as Figure 10 shows most increasing trends were found in halogen- free flame retardants.

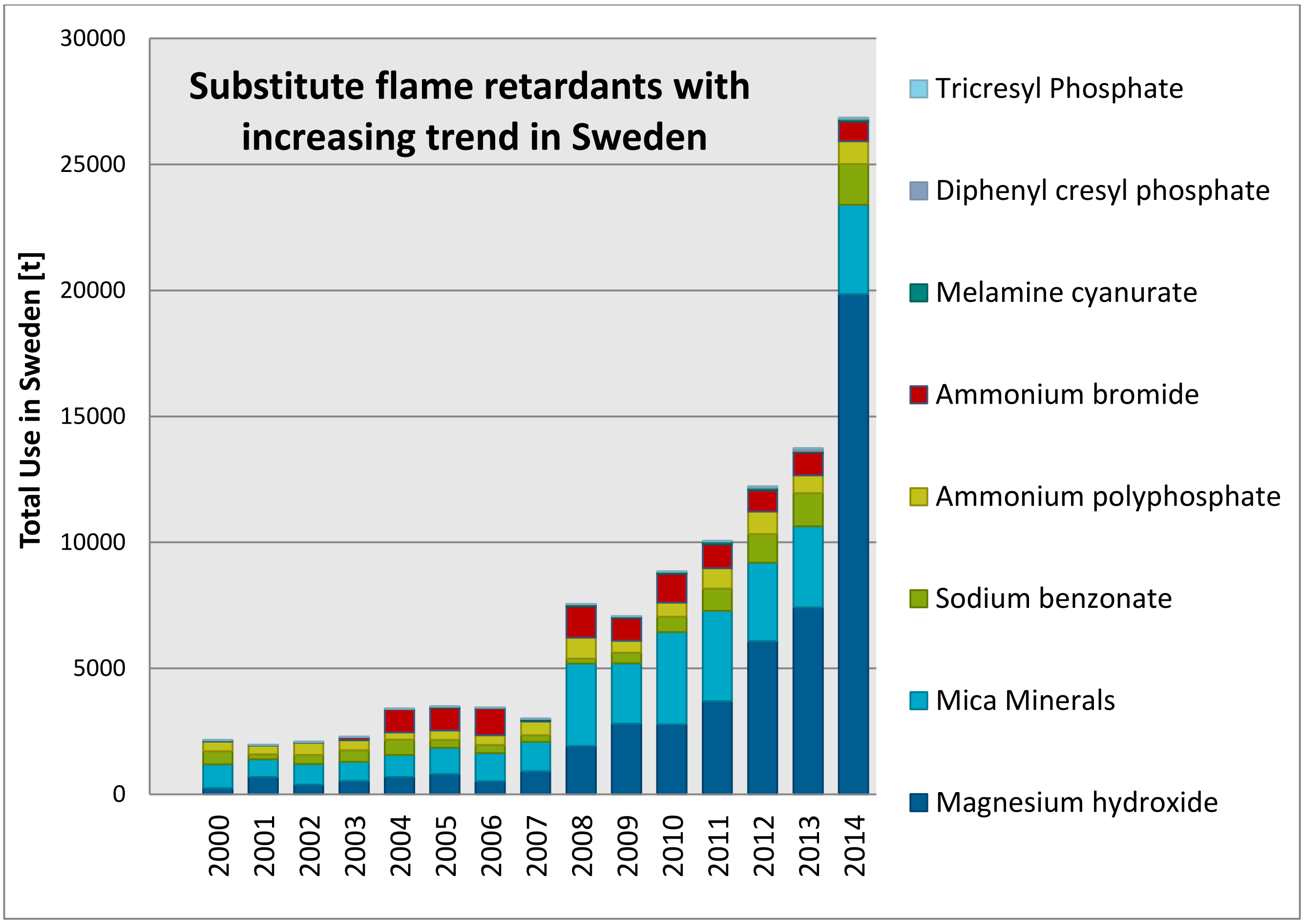

Figure 10: Flame retardants with increasing trend in Sweden

The CAS numbers of the respective flame retardants with clearly increasing trends are given in Table 4.

Table 4: Identified flame retardant substitutes

CAS	Name
1309-42-8	Magnesium hydroxide
12001-26-2	Mica Minerals
532-32-1	Sodium benzonate

68333-79-9	Ammonium polyphosphate
12124-97-9	Ammonium bromide
37640-57-6	Melamine cyanurate
26444-49-5	Diphenyl cresyl phosphate
1330-78-5	Tricresyl Phosphate

Some other flame retardants also showed also temporary increases, but were fluctuating. Thus, they are not listed.

4.2.2. Patterns of substitution for flame retardants

Figure 11 shows the increase of halogenated flame retardants (HFR) and a steady use of halogen free flame retardants (HFFRs). The quantitative importance of the regulated wrSVHC is shown as green line and negligible compared to the quantities of halogen free flame retardants.

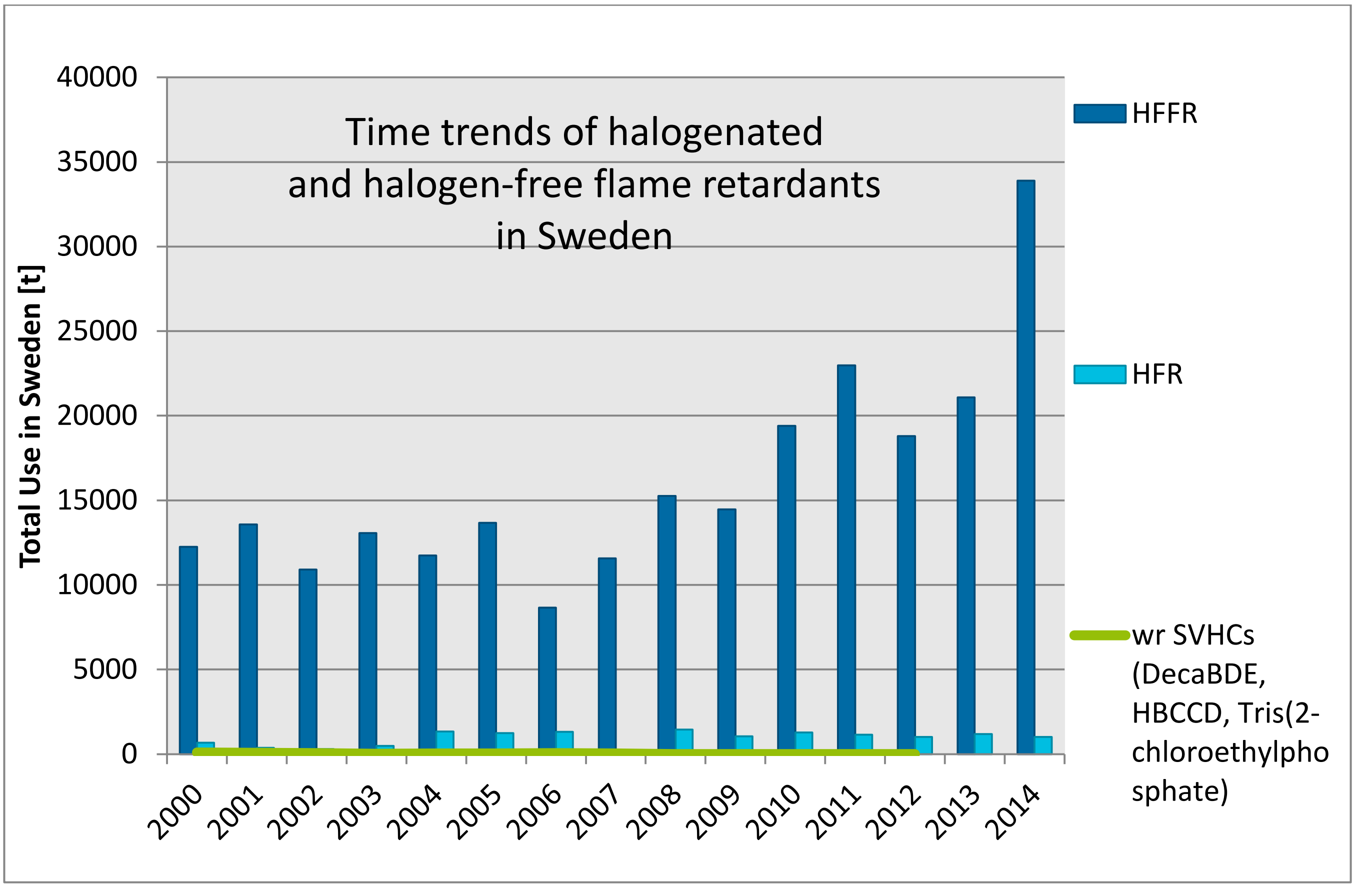

Figure 11: Time trends of halogenated and halogen-free flame retardants in Sweden

Unexpectedly, HFRs do not show a clear pattern of decrease. Yet, HFFRs are increasing from 2010 onwards. The increase is mainly attributed to the previously shown substitutes. It might be that the HFRs stay on a similar level because some of them are authorized under REACH for special applications (REACH- Annex XIV).

4.2.3. Identification of emerging flame retardant substitutes

Flame retardants, which are used in increasing amounts, and which at the same time are already listed on the NORMAN list of ES, are considered as potential emerging pollutants, depending on their environmental behavior. From the flame retardant list, some are already found in the environment. To identify the emerging flame retardants in the environment, the flame retardant list was compared to NORMAN list of ES and 4 substances show concerning trends. Three of which are HFFRs marked in blue and one HFR in red. HFRs are thought of as regrettable substitutes, as they are found to be detrimental to human health and the environment. However, also HFFRs are found in the environment as these findings show. This could indicate that regrettable substitutions could not only take place in HFRs but also in HFFRs.

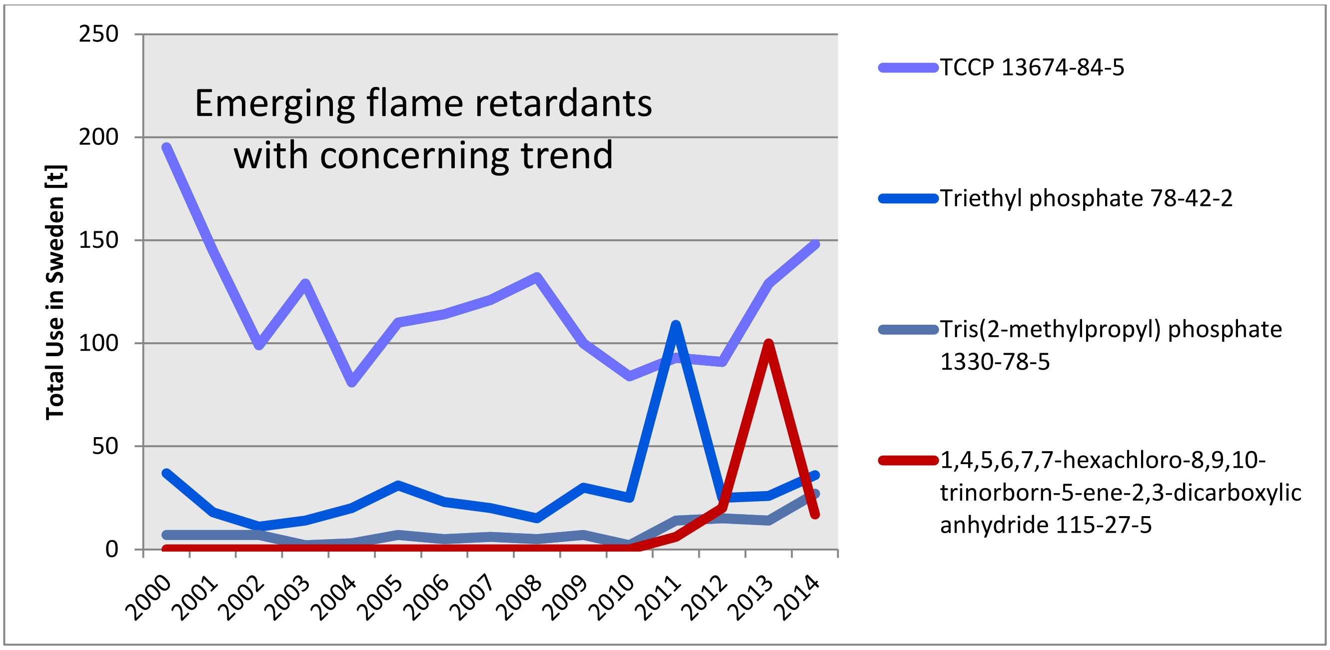

Figure 12: Emerging flame retardants with concerning trends in Sweden

4.3. Plasticizers

4.3.1. Identification of substitute plasticizers

14 plasticizers are used in Sweden in clearly increasing amounts. It is reasonable to assume that these plasticizers are used as substitutes for strictly regulated plasticizers (which show decreases in the amount used), 1 of these substitutes is phthalate marked in red, which shows a sharp increase from 2001 to 2010 and then decreases again. This suggests that it was used as a phthalate substitute for a major plasticizer such as DEHP in the beginning and then was substituted itself.

Some plasticizers showed increasing trends, but were not included due to small quantities. Of those, 3 were identified as phthalates however, their quantity is small compared to the other plasticizers

(<50 t in 2014) and therefore not included in Figure 13. Four non-phthalates are also excluded due to small quantities (<50 t in 2014).

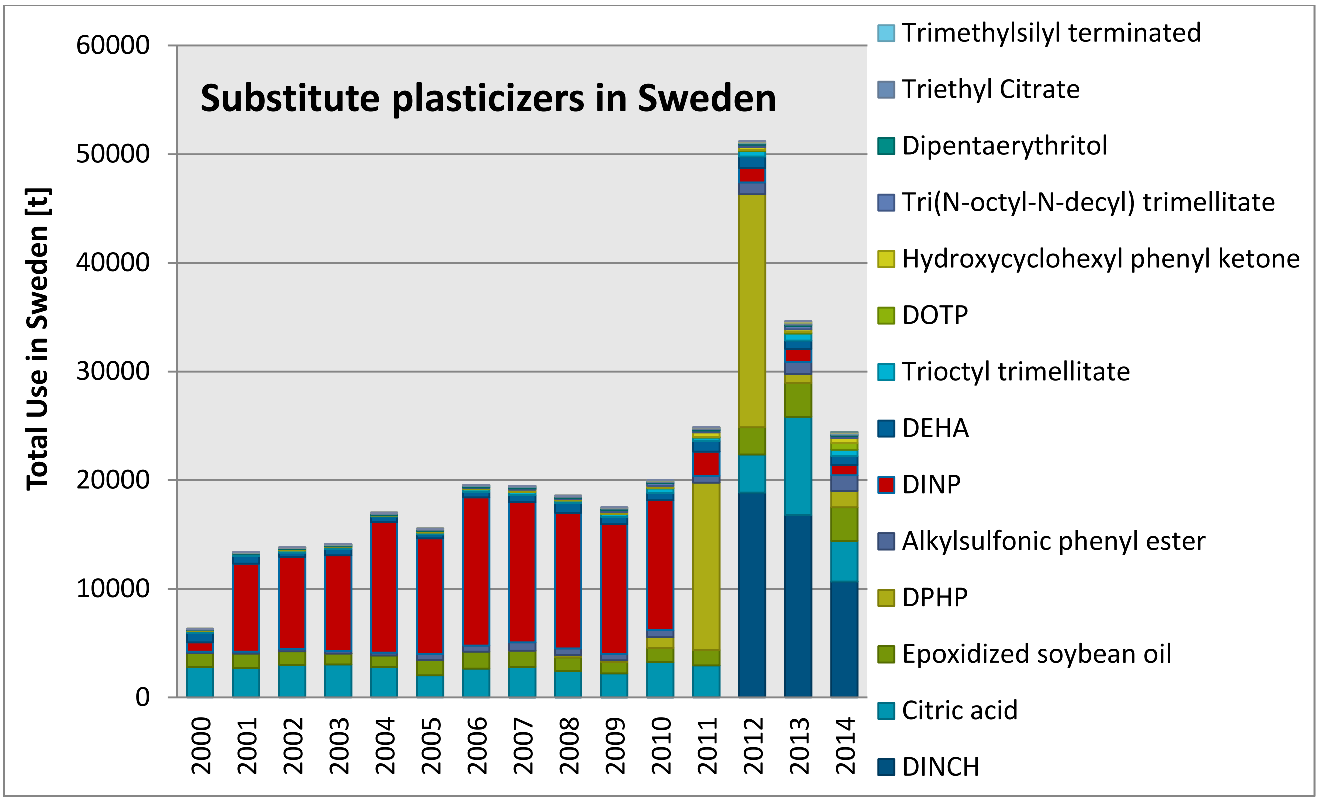

Figure 13: Substitute plasticizers in Sweden

Figure 13 shows the cumulative increase of all plasticizers with increasing trends, most of which are found in the group of "non-phthalates". This is corroborated by analyzing the time trends of plasticizers according to group. All quantitatively relevant plasticizer substitutes are listed with CAS in Table 5.

Table 5: Identified plasticizer substitutes

Name	CAS
DINCH	166412-78-8
Citric acid	77-92-9
Epoxidized soybean oil	8013-07-8
DPHP	53306-54-0
Alkylsulfonic phenyl ester	91082-17-6
DINP	28553-12-0
DEHA	103-23-1
Trioctyl trimellitate	3319-31-1
DOTP	6422-86-2
Hydroxycyclohexyl phenyl ketone	947-19-3
Tri(N-octyl-N-decyl) trimellitate	67989-23-5

Dipentaerythritol	126-58-9
Triethyl Citrate	77-93-0
Trimethylsilyl terminated	68037-59-2
Diundecyl phthalate	85507-79-5
Non- phthalate15	26444-49-5
Non- phthalate16	1330-78-5
Non- phthalate17	3622-84-2

4.3.2. Patterns of substitution for plasticizers

Figure 14 shows the development of plasticizers according to chemical class. As discussed in the introduction, phthalates are the class that has been widely discussed and efforts have been directed towards replacement of them, as confirmed here.

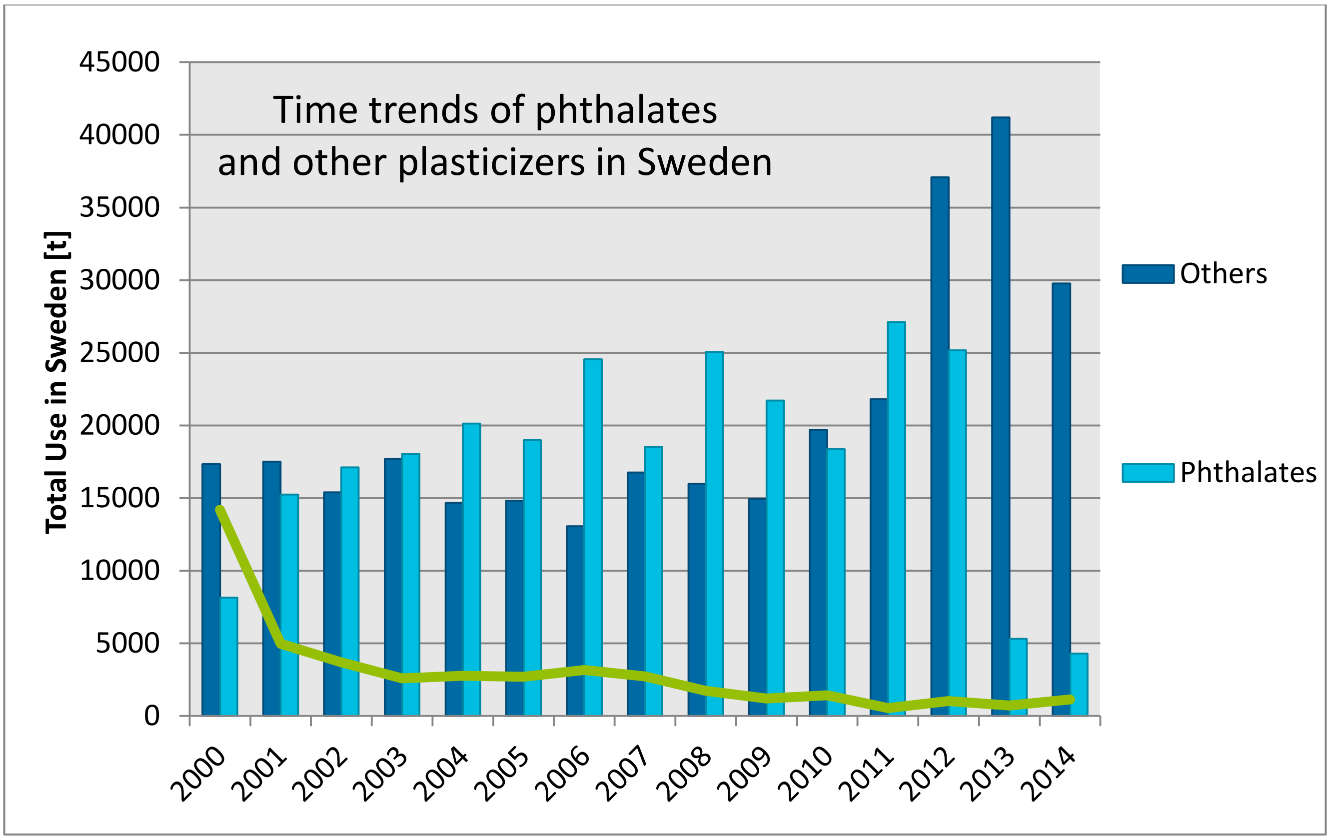

Figure 14: Time trends of phthalates and other plasticizers in Sweden

Phthalates decrease notably in 2013, whereas an increase in non-phthalate plasticizers can already be observed gradually from 2009 onwards. The quantitative importance of the regulated wrSVHC is important and it can be derived, that even though phthalates such as DEHP were regulated and decreased they were substituted first by other phthalates up until 2012, where a regime shift from phthalates to non-phthalates occurred.

4.3.3. Identification of emerging plasticizer substitutes

Four substances were listed on NORMAN and found in the Swedish SPIN data. Of those two plasticizers one was a phthalate marked in red, which has confidential data after the year 2007 but shows a sharply increasing trend before. It is marked in red, as phthalates are considered as generally as regrettable substitutes by the scientific community. Whereas non-phthalates could be functional substitutes, given enough (eco) toxicological data is available.

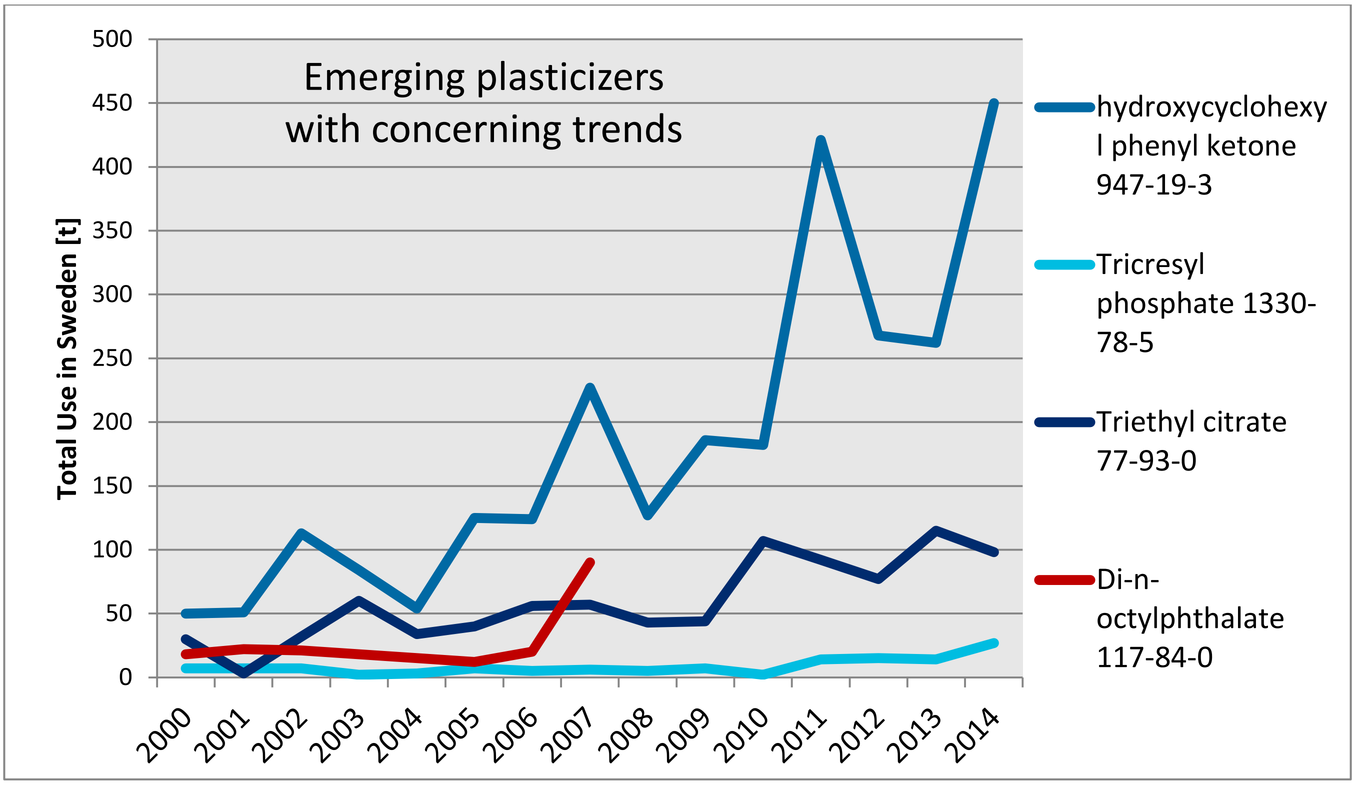

Figure 15: Emerging plasticizers with concerning trends in Sweden

4.4. Surfactants

4.4.1. Identification of substitute surfactants

20 surfactants are used in Sweden in clearly increasing amounts. It is reasonable to assume that these plasticizers are used as substitutes for strictly regulated surfactants. SPIN was searched by an SQL query for all surfactants in Sweden. The resulting list held 109 surfactants and was then manually filtered for substances exhibiting upwards trends, resulting in 20 substances. Of these 20 surfactants the 7 surfactants with smallest quantity were excluded (< 400 t in 2014). Figure 16 shows 13 quantitatively relevant surfactants with increasing trend in Sweden.

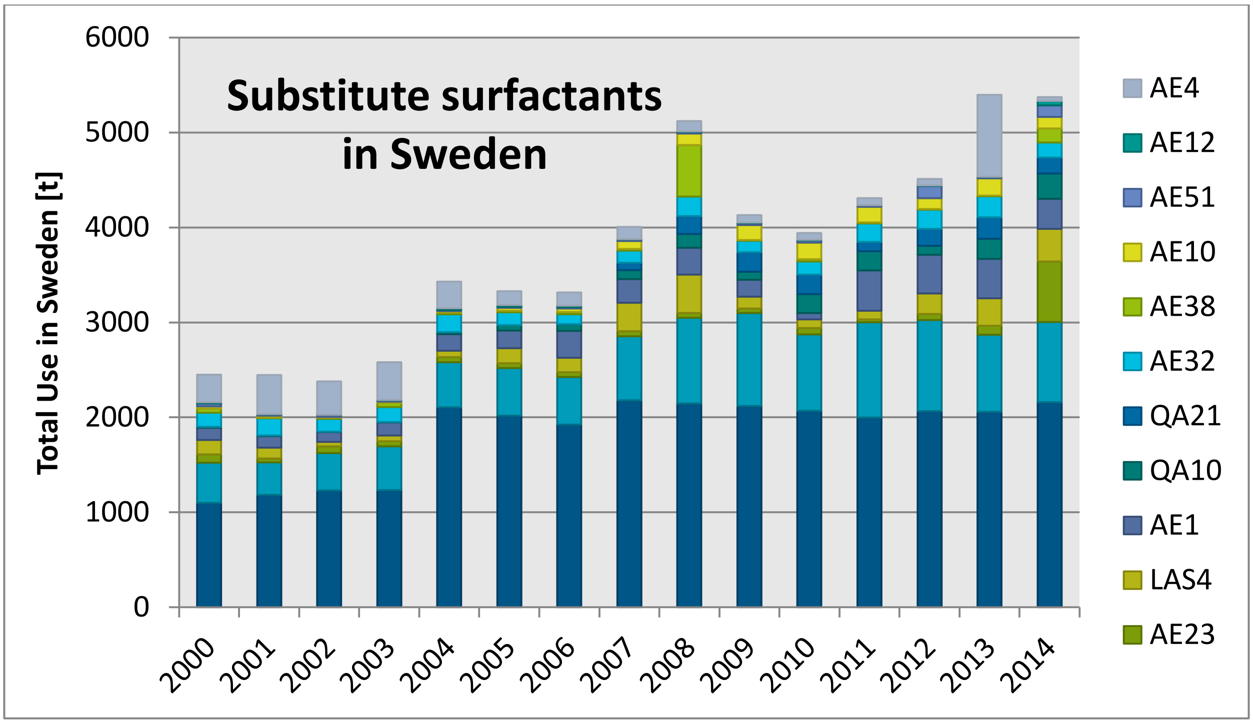

Figure 16: Substitute surfactants in Sweden

Table 6 shows the names, abbreviations and CAS numbers of the substitute surfactants in Sweden.

Table 6: Identified surfactant substitutes

Full Name	Abbreviation	
Alcohols, C12-15, ethoxylated	AE23	68131-39-5
Alcohols, C9-11, ethoxylated	AE31	68439-46-3
Alcohols, C12-18, ethoxylated	AE23	68213-23-0
Sodium alkylbenzene sulfonate	LAS4	68411-30-3
Polyalkylenglycolether	AE1	9002-92-0
Benzalkoniumchloride	QA10	68424-85-1
C12-14 alkylmethylaminethoxylatmethylchlorid	QA21	863679-20-3
Polyalkylenglycolether	AE32	68439-49-6
Alcohols, C12-16, ethoxylated	AE38	68551-12-2
Decanol, ethoxylated	AE10	26183-52-8
Alcohols, C10-16, ethoxylated	AE51	69227-22-1
n-Octyl-oligo-oxyethylene	AE12	27252-75-1
Polyethylene Glycol Monooleyl Ethe	AE4	9004-98-2

The chemicals groups of surfactants with increasing trends are diverse. However, the most discussed group alkylphenol ethoxylates is not represented here.

4.4.2. Patterns of substitution for surfactants

Figure 17 depicts the pattern of substitution for the distinct substance classes. As discussed in the introduction, alkylphenol ethoxylates are the class that has been widely discussed in the scientific community and efforts has been directed towards replacement of them, as also shown here.

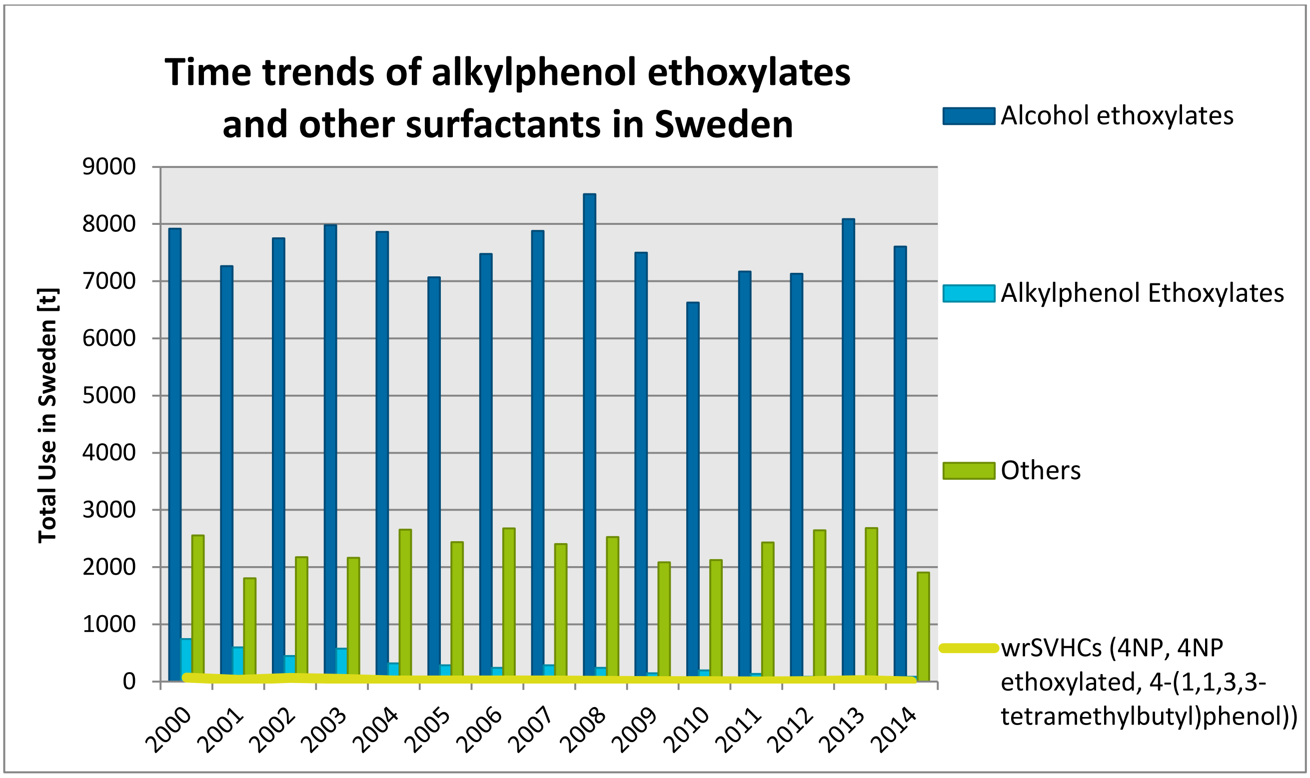

Figure 17: Time trends of alkylphenol ethoxlates and other surfactants in Sweden

Alkylphenol ethoxylates decreased gradually from 200 onwards. The quantitative importance of the regulated wrSVHC is negligible. However, later research with other CAS numbers of the nonylphenol entry on the Candidate list (CAS: 84852-15-3), showed a tremendous increase (1683 t) in one specific nonylphenol that was not used before at all in Sweden. It can be considered regrettable substitution and distorts the results presented. This increase is not shown in the figure. This later discovery of an unknown CAS number is a good example for the non- exhaustiveness of the analysis. The lack of all some CAS numbers of Candidate List entries is discussed further in chapter 5.1.2.

4.4.3. Identification of emerging surfactant substitutes

In order to identify EPs with increasing trends the surfactant list was compared to NORMAN two substances were listed on NORMAN and found in the Swedish SPIN data with increasing trends. Of those one is a QAs marked in red, and one is an LAS as shown in Figure 18.

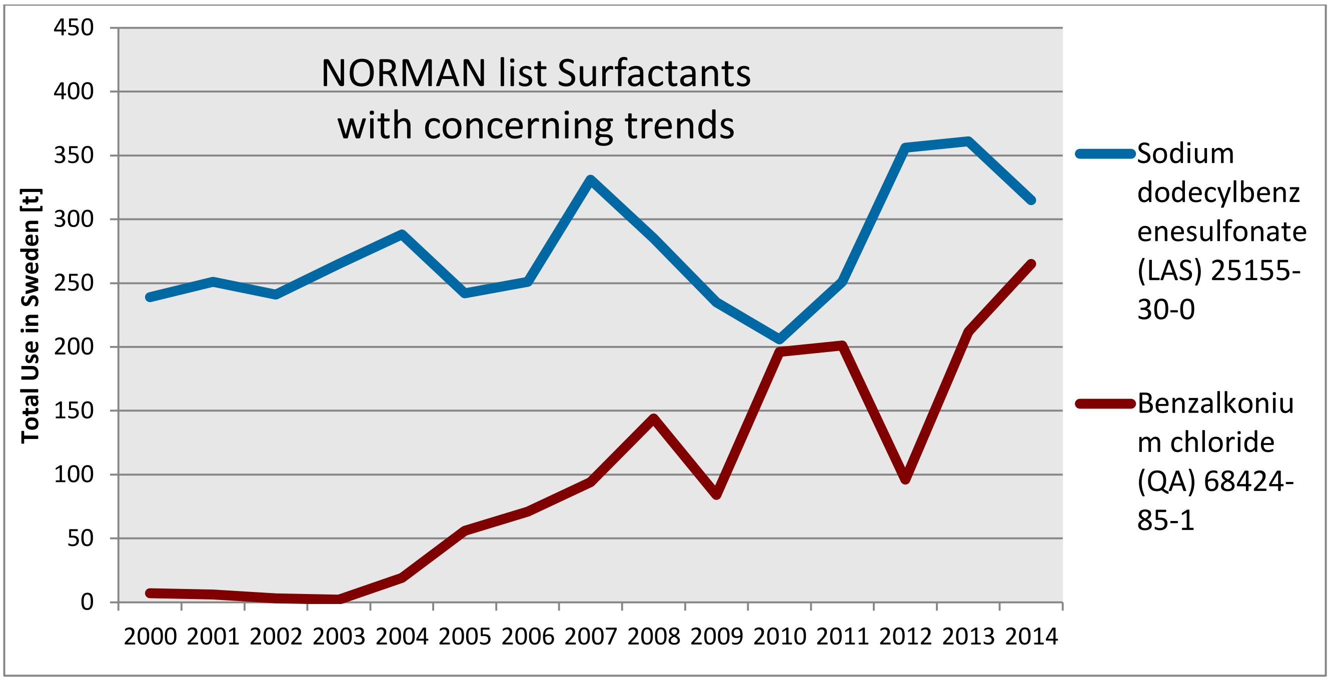

Figure 18: NORMAN list surfactants with concerning trends in Sweden

These findings show that there might be regrettable substitution in groups of chemicals, which are not inherently perceived as threatening.

5. Discussion

The results presented in the previous chapter demonstrate that the Total Use data from SPIN is a workable and data-rich source to develop time trends for problematic substances and their substitutes. This enabled the elaboration on the following four research questions as stated in chapter 2: Goal & research questions: II) Does strict regulatory action (such as placing on the REACH Candidate list) cause a reduction in the amount of water relevant chemicals used? III) Does strict regulatory action cause increased use of substitutes and can patterns of substitution be detected? IV) Can substitutes be identified which have the potential to be future emerging substances and possible pollutants? V) Does the identification of substitutes allow for future monitoring recommendations?

All of the stated research questions are discussed in detail as follows:

5.1.1. Regulative impact of REACH on SVHC use in Sweden

Time trends were used to evaluate the impact of REACH candidate list regulation on water relevant problematic substances. In this thesis it was shown that the placement of water relevant problematic substances on the REACH candidate list had a clear impact on the time trends of the amount of the wrSVHC were used in Sweden. It was shown that regulated wrSVHC were decreasing over time as compared to an unregulated set of chemicals the PMOC, which did not show clear time trend patterns. Until today there were no studies that attest the expected decrease of SVHC use in any EU member state. Therefore, this thesis provides the first data on the impact of the REACH candidate list regulation on the time trends of SVHC used. However, the decrease of wrSVHC cannot exclusively be accounted for by REACH, as wrSVHC are also frequently regulated under other regulatory frameworks in Sweden, especially the WFD. To decrease the impact of confounding originating from the WFD, an analysis of the time trends of all 173 SVHC was conducted and is found in the Appendix Figure 1 showing a clear pattern of decrease. Hence, the regulatory impact of the REACH candidate list can be regarded as successful in Sweden. Similarly the time trends of total SVHC use were found to decreasing in Finland and Denmark as shown in Appendix Figure 2 and Appendix Figure 3 respectively. However, Norway as a non-EU country does not show a clear pattern of decrease as illustrated in Appendix Figure 4.This might be due to the fact that Norway is not an EU member state. Nonetheless, REACH theoretically applies to all the EEA (European Economic Area) including Norway (EUROPA/GROW, 2015). This suggests that there is no proper implementation of REACH in Norway. Alternatively it could be assumed that the reporting of the Norwegian officials is erroneous or SVHC do can be used further. Conclusive, SPIN data confirmed that regulation lead to a decrease of wSVHC use in Sweden. Further research should be conducted on the unexpected pattern of SVHC use in Norway including a detailed analysis which substances are dominating the time trends in the distinct Nordic countries. SPIN could be further used as an indicator for policy assessment in chemical regulations, as it provides clear and workable use trends.

5.1.2. Identification of substitute chemicals and patterns of substitution

The hypothesis that hazardous chemicals are substituted was confirmed when looking at patterns of substitution. For each of the three substance groups (flame retardants, plasticizer and surfactants) numerous substances with increasing trend were identified and assumed to be substitutes for regulated chemicals. It was assumed that the increasing trends are likely to be due to the need for substitutes for regulated substances. However, it is possible that some substances with increasing time trend are merely due to increasing production volumes of a particular product. For instance steadily increasing production of PVC containing a plasticizer can also drive the increasing use of this substance and does not necessarily be a substitute for another substance. Nonetheless, it is expected that most of the increasing trends originate due to the need for substitution chemicals to replace strictly regulated chemicals. Previous studies conducted for the European Commission suggest that by regulating a substance on the REACH candidate list the substance is likely to be substituted. This is represented by a request for substitution of SVHC from suppliers (51%). Moreover, 27% of the companies that use SVHC launch initiatives to develop new substances to substitute the used SVHC (CESS, 2012). The results presented in chapter 4 shows these developments for the first time in a quantitative manner.

To address the patterns of substitution all technical use classes were split into two chemical classes respectively: One class that is considered hazardous and is also mostly regulated and the second class containing all other chemicals that are not widely discussed as hazardous. For “flame retardants”, “plasticizers” and “surfactants” the use tonnages of chemical classes that are considered less hazardous increased. Across two of the three technical use categories addressed (“plasticizers” and “surfactants”) a decrease in chemical classes considered hazardous (phthalates and alkylphenol ethoyxlates respectively) was observed while the use in chemical classes that are not under discussion increased. Of the classes considered hazardous various chemicals are regulated under the REACH Candidate List, such as DEHP in phthalates and nonylphenols in alkylphenol ethoyxlates. The general observation of patterns of substitution confirms the hypothesis of substitution. The patterns of substitution are further discussed for each technical use category separately.

Flame retardants

For flame retardants the widely discussed halogenated flame retardants of which some are also regulated (DecaDBE, HBCCD) did not decrease. This was unexpected and could be due to two causes: 1) Regulated halogenated flame retardants are to authorize for specific uses according to the authorization list. However,this was not confirmed. DecaDBE is not any more used after 2008 and HBCCD also decreased to zero 2005 in Sweden. However, a steady increase ammonium bromide (CAS: 12124-97-9) was observed, indicating regrettable substitution of regulated halogenated flame retardants for a non-regulated HFR. Regardless of this regrettable substitution in the realm of halogenated flame retardants an increase in halogen free flame retardants was observed. This however does not guarantee that the substitutes could not also be regrettable eventually, due to a lack of knowledge with regard to the substitute's characteristics. To identify if a substitute is functional according to state of the art knowledge, the SIN (substitute it now!) list and SINimilarity can be searched. SIN and SINimilarity show chemical similarities to hazardous substances. Similar substances are likely to behave in a similar manner, and therefore the chances for regrettable substitution increase.

Quantitatively important substitute flame retardants hat are not suspected from SIN or SINimilarity and therefore considered functional substitutes are magnesium dihydroxide (CAS: 1309-42-8), Mica-group minerals (CAS: 12001-26-2), Polyphosphoric acids, ammonium salts (CAS: 68333-79-9) and Graphite (CAS: 7782-42-5). Magnesium dihydroxide is, similar to aluminum dihydroxide expected to be used in waste water treatment as flocculation agent and therefore is of high use. Similarly for graphite, the main uses are steel-making, foundry moldings, refractories, auto parts, and as lubricants. Flame retardant uses are expected to be small. More concerning are high use volume chemicals that are suspected from SIN/SINimilarity, such as sodium benzoate (CAS: 532-32-1). None of the chemicals listed above has monitoring data, neither in Europe nor Sweden according to the NORMAN EMPODAT database. As mentioned before, the emission to the environment is essential to derive sensible recommendations for monitoring. SPIN offers the exposure tool, which estimates the exposure of substances to various matrixes in the environment on a scale from 1 to 5, with 5 being high exposure. SPIN estimates the exposure to of the mentioned substances to surface waters as medium exposure with following values: magnesium dihydroxide =4, Mica-group minerals = 4, Graphite= 4. For the inorganics and polyphosphoric acids= 4, no threat to surface waters is expected as they are not listed on SIN or SINimilarity and are (except for polyphosphoric acids) inorganics. However, sodium benzoate which is also widely used food preservative has an exposure of 4. According to ECHA the biodegration rate is 50-97 % over 60 days (ECHA, 2017c). Therefore, no monitoring is required, unless it is found to be emitted from point sources in very high quantities to surface waters.

Plasticizers

The technical use category "plasticizers" was subset into phthalates and non- phthalates. Phthalates are widely discussed in the scientific community with regard to their hazardous impact on the environment and the endocrine disrupting character (Gao and Wen, 2016). In the use data analysis, phthalates showed a clear decreasing trend while non-phthalate plasticizers depicted an increasing trend. The wrSVHC are quantitatively of importance in the early 2000, due to the very high use of DEHP that was regulated under REACH as SVHC since 2008. The strong decrease of the wrSVHC, especially DEHP occurred before 2008, which could be explained by public discussions thereby publicly increased awareness of consumers in the Nordic countries that triggered industrial change. While DEHP was decreasing drastically from 2000 onwards, the general use of other phthalates increased further until 20012, from where a clear decrease in phthalates can be observed. Meanwhile, the non-phthalate plasticizers show a sharp increase from 2011 onwards. It is likely that during the last years numerous of the phthalates were substituted by non-phthalates as shown in Appendix Figure 6. It can be clearly seen, that regulated phthalates like DEHP decreased and were replaced by other phthalates. Especially bis(2-propylheptyl) phthalate (CAS: 53306-54-0) was a very prominent phthalate in2011 and 2012, with over 20000 t used in Sweden in 2012. However, from 2013 the use shrinks to a minute tonnage, while non-phthalates sky rocket after 2012, replacing the declining phthalates. Conclusive it can be said that regulated phthalates declined first and were replaced by other non-regulated phthalates that were ultimately replaced by non-phthalates. However, the fact that the substitutes are non-phthalates is no reason to cut back on risk assessment. Contrarily, it should be strongly encouraged to find and test the high use volume chemicals, which are supposedly replacing phthalates, such as DINCH (CAS: 166412-78-8), melamine (CAS: 108-78-1), citric acid (CAS: 77-92-9) and Epoxidized soybean oil (ESBO) (CAS: 8013-07-8). For neither melamine, epoxidized soybean oil, DINCH nor bis(2-propylheptyl) phthalate there are monitoring data available on NORMAN EMPODAT. However, citric acid, is expected to degrade rapidly. SPIN estimates the exposure to of the mentioned substances to surface waters as medium exposure with following values: DINCH=3, melamine= 3, citric acid= 4, Epoxidized soybean oil (ESBO)= 4, bis(2-propylheptyl) phthalate= 3. Combined with the high use volume, this could constitute results that are detectable in the environment. Given their quantitative relevance and high emission ranking, biodegradation data needs to be collected and in case of slow degradation, monitoring should be initiated in Sweden. Monitoring matrixes chosen should be based on the physio-chemical properties of individual substances.

Surfactants

The technical use category "surfactants" was subset into alcohol ethoxylates, alkylphenol ethoxylates (APEs) and others. The alkylphenol ethoxylates showed a clear decreasing trend, however compared to the tonnages of alcohol ethoxylates with the used CAS numbers. The decrease in APEs is expected as it encompasses nonylphenol ethoxylates which are restricted under REACH and the WFD. However, in later analytics it showed that other CAS numbers from a alkylphenol ethoxylates entry the "nonylphenols" were not exhaustive and the decreasing alkylphenol ethoxylates detected and shown in results were substituted by another nonylphenol (CAS: 84852-15-3), that is registered with a highly concerning volume of 1682 tons in 2015. This can be considered a remarkable example of regrettable substitution. Moreover, it shows the incompleteness of the data used. This is discussed in more detail in the limitation section. Generally, all nonylphenols need to be surveilled, not only in use but also in surface waters and other environmental matrixes. For the regrettable substitute found (***84852-15-3***) no monitoring data is available on NORMAN EMPODAT, however on Spin the Exposure to surface water is 4. This is very concerning. Officials need to check the use categories and to corroborate the emission to the environment. Rising of ***monitoring data is highly recommended.***

With regard to functional surfactant substitution in the surfactant category, literature suggests that APEs are commonly substituted by alcohol ethoxylates AEs (Burlington Chemical Co., L. L.C and Leuk, 2011). However, if also functional substitution occurred is difficult to evaluate from the results obtained from SPIN. This is due to annual fluctuation of AEs that do not allow for clear trends. Nonetheless, a clear decrease in APEs is evident for the regulated APEs, while APEs that are only suspects from the SIN/ SINimilarity list do not vary greatly shown in Appendix Figure 7. The substances that are not suspected on SIN/SINimilarity and therefore considered functional and increased rapidly to high volumes are AE31 (CAS: 68439-46-3) and AE23 (CAS: 68131-39-5). Another quantitatively important substance is a suspect from SINimilarity, Benzenesulphonic acid, C10-13 alkyl derivatives (CAS: 68411-30-3). Exposure of the substances to surface waters is estimated by SPIN for both AEs and Benzenesulphonic acid as high (index= 4). The AEs are expected to be readily biodegradable, however Benzenesulphonic acid needs to be checked for potential slow biodegradability and, given slow degradability needs to be monitored.

5.1.3. Identification of substitutes listed as emerging substances

The ultimate goal of the thesis was to identify substitutes that are increasing in use in Sweden and can already be found in the European environment. The hypothesis was that substances already found in surface waters in Europe (NORMAN list of emerging substances) are likely to be found in Swedish surface waters too given that they are increasingly used and possibly emitted to the environment. The emission modelling could be done with use categories, which indicate in what applications the substances are used. For example a substance used as fertilizer is more likely to be emitted to surface water than a substance that is used in a closed system approach in the industry. Yet, as described in the introduction, the use categories in the SPIN are insufficient and therefore the emission cannot be modelled due to lack of data. The final emission modelling and evaluation of biodegradability and (eco) toxicity needs to be conducted by experts. Nonetheless, the identified substances were tentatively researched to understand the state of the art knowledge about these substances and their hazard potential in the environment, especially surface waters.

Flame retardants

Four flame retardants which are used in increasing amounts were listed on the NORMAN list of emerging substances. 1,4,5,6,7,7-hexachloro-8,9,10-trinorborn-5-ene-2,3-dicarboxylic anhydride is an HFR with a peak trend cumulating in 100t in 2013. Yet, the use strongly declines afterwards to 25 t in 2014. This peak behavior is probably due to the attempted substitution of another HFR, with the late understanding that the replacement was a regrettable substitution. ECHA has no public registered data on the routes by which this substance is most likely to be released to the environment, and further, no environmental fate data is available. However, studies describe that in aqueous solution, the substance is rapidly hydrolyzed to chlorendic acid, with a half-life of approximately one hour (International Programme on chemical safety, 1996). Therefore, risk from bioaccumulation is not expected. Given the substance is emitted in small quantities, the threat to surface waters can be considered minimal, especially given the decline of use after the “try- out” peak.

However, the other three flame retardant EPs with increasing trend (Triethyl phosphate (CAS:78-42-2), Tris(2-methylpropyl) phosphate (CAS:1330-78-5), Tris(2,3-dichloropropyl) phosphate (TCCP) (CAS:13674-84-5)) are more interesting from an environmental hazard perspective. All are classified as organophosphates (OPs). The increase of OPs as a result of restricted use of HRFs has been reported in the US and the burden shift from HFRs to HFFRs or OPs (Stapleton et al., 2009). OPs are used as both, flame retardants and plasticizers in flexible PVC and polyurethane foam and other applications such as computer housings or hydraulic fluids (Bollmann et al., 2012). As they are not covalently integrated into the polymers, they are likely to leach from plastic materials. OP leaching from plastic films used for greenhouses was found an important source of import to surface waters (Cho et al., 1996). Emissions of OPs into the environment are still increasing, thus, OPs can be classified as “re-emerging pollutants” (Reemtsma et al., 2016). Concerning is that the Exposure indexes suggest values of 3 and 4 for the exposure of OP to the environment.

Quantitatively, the most concerning OP is TCCP, with an annual use of 150 t in 2014. TCCP is a chlorinated OP that is mostly contaminating surface waters by emissions from WWTPs. For chlorinated OPs the removal rate in WWTP is minimal resulting in high effluent concentrations (Reemtsma et al., 2008). According to a survey of WWTPs in several European countries TCPP is routinely detected in WWTP effluents, at concentrations of a few hundred ng/L. Furthermore, chlorinated OPs are likely to bio- accumulate as they do not photodegrade like the non-chlorinated OPs (Bollmann et al., 2012). Based on WWTP-effluent concentrations and their persistence, TCPP was found to be almost as problematic a contaminant as the commonly known pharmaceutical carbamazepine in partially closed water cycles (Reemtsma et al., 2006). An analysis on the EMPODAT database created by the NORMAN network exhibited that there are no environmental measurement data for TCCP in Sweden, where the increasing use trend was identified. However, in Germany and other countries there is plenty of data showing concerning levels of up to 0.47 µg/l in river water measured in Mylau in 2012 (NORMAN EMPODAT, 2017). Given the increasing use of TCCP in Sweden and the high exposure index of 4, more attention should be placed on surveillance of TCCP in Swedish surface waters. Furthermore, political effort should be placed on preventing the regrettable substitution of HFRs and/or plasticizers by TCCP.

Plasticizers

Four plasticizers that were listed on the NORMAN list of emerging substances were identified.

Triethyl citrate (CAS: 77-93-0) is a non-hazardous substitute for common plasticizers with total use volumes around 100tpa in 2014, trend increasing. It is also used as food additive (E number E1505). This suggests that Triethyl citrate is a functional substitute, as it is even allowed as food additive. However, also sucralose is used in the food industry and is lately in scientific focus due to its slow biodegradability and accumulation in surface waters. Currently ECHA has no data on the biodegradability of triethyl citrate or the environmental pathways. Triethyl citrate is marketed as safe and environmentally friendly plasticizer alternative. The data safety sheet states that there is low risk of bioaccumulation and that triethyl citrate is mostly found as vapor in the atmosphere. Vaporphase triethyl citrate will be degraded in the atmosphere by reaction with photochemically-produced hydroxyl radicals; the half-life for this reaction in air is estimated to be 4 days, according to Pucbem (Pubchem, 2017). However, biodegradation data in water were not available (Pubchem, 2017). Given on data for acetyl tributyl citrate, a structural analog, triethyl citrate may be expected to biodegrade rapidly. Yet, in NORMAN EMPODAT for the Netherlands concentrations up to 0.23 µg/l in surface waters were found in 2014. For Sweden there is no data available and the exposure index is 3. ECHA states that there is no hazard for aquatic organisms identified. Nonetheless, it is suggested to introduce spot sample monitoring for Swedish surface waters for triethyl citrate. Even though the substance might appear harmless on its own it can contribute to unexpected mixture effects with other chemicals. To prevent mixture effects it is important what substances are emitted and in what concentrations they can be found in the environment.

Di-n-octylphthalate (CAS: 117-84-0) is a phthalate plasticizer that with a use volume of 90 tpa in 2007, with an increasing trend pattern, which is confidential from 2008 onwards. The exposure index is estimated at 2 on SPIN. However, Di-n-octylphthalate can leach from materials such as plastic, rubber and textiles and release to the environment likely to occur from leaching from products. However, ECHA has no detailed information on DOP about these issues. According to the data safety sheet of Di-n-octylphthalate the substance can be damaging fertility or the unborn child. Yet, there is nothing as known about persistence and biodegradability. Known are the aquatic toxicity values EC50/48 h > 100 mg/l for daphnia magna and EC50/72 h > 100 mg/l for algae (Kolar, 2011). Due to the bad solubility in water, Di-n-octylphthalate is concentrated in sewage sludge. According to the NORMAN EMPODAT database Di-n-octylphthalate was found in Swedish sewage sludge in 2007 in concentrations up to 650 µg/kg dry weight in Ryaverket STP. This is the highest measured concentration in the whole database. Interestingly, a sharp increase in Di-n-octylphthalate is shown from 2006 to 2007, probably as a response to the decrease in DEHP. However, after 2007 the data is confidential. Therefore, it is highly recommended to KEMI, who have access to the confidential data, to check these time trends and to encourage more monitoring measurements to fill data gaps. Furthermore, assessments regarding the PBT criteria should be encouraged.

Tricresyl phosphate (CAS: 1330-78-5) is used as PVC plasticizer in plastics such as polystyrene with a use tonnage of 20 tpa in 2014, trend increasing. The exposure index is defined for Sweden at 4, thus very high. Due to the physicochemical properties of TCP, there is a high potential for bioaccumulation. According to ECHA tricresyl phosphate is very toxic to aquatic life with long lasting effects and suspected of damaging fertility or the unborn child. Already in the 1990 it was reported that rainbow trout is adversely affected by TCP concentrations below 1 mg/l, with sign of chronic poisoning and has synergistic effect on organophosphorus insecticide activity (Lassen et al., 1999). According to NORMAN EMPODAT, the highest levels measured were in the Danube in Hungary with 0.054 µg/l in 2013. For Sweden there is no data available. Therefore, it is recommended to raise monitoring data to evaluate if there is a threat from tricresyl phosphate in Swedish surface waters.

N-butyl-benzenesulfonamide (CAS: 3622-84-2) is a plasticizer, which was not used in Sweden until it appeared in the use data with 19 t. The exposure to surface waters is with 2 relatively low. ECHA found that N-n-butylbenzenesulphonamide does not biodegrade in a simulation test with natural surface water, according to test guideline OECD 309. LC50 for freshwater fish was determined at 37 mg/L. According to the NORMAN EMPODAT database, monitoring data exists, however always below LoQ. Yet, no monitoring data is available for Sweden. Given the increase in use for N-butyl-benzene sulfonamide monitoring of this substance in Swedish surface waters will probably result in similar results like obtained in other countries where the concentration was below the limit of detection. Therefore, monitoring would probably be in vain. However, the time trends of the use of N-butyl-benzenesulfonamide need to be followed closely.

Surfactants

Two surfactants that were listed on the NORMAN list of emerging substances were identified. Sodium dodecylbenzenesulfonate (CAS: 25155-30-0), is a LAS with a use tonnage around 250tpa in Sweden. It shows trend fluctuation but is increasing. The exposure index is with 4 very high, as expected for detergents. According to ECHA Sodium dodecylbenzenesulfonate is often used as pH regulator and water treatment products, coating products, metal surface treatment products, laboratory chemicals, polymers and washing & cleaning products. The release to the environment is likely to occur from liquids/detergents, care products, paints or fragrances. Sodium dodecylbenzene sulfonate is classified as readily degradable. There is no entry in the EMPODAT database for Sodium dodecylbenzenesulfonate, therefor there is no monitoring data available. As the substance appears on the NORMAN list of emerging substances and no monitoring data is available it is recommended to raise those data based on the use trend of the substance in Sweden.

Benzalkonium chloride (CAS: 68424-85-1) is a QA used as a biocide and cationic surfactant with strongly a use volume of around 250tpa in Sweden in 2014, trend increasing with an exposure index of 4. There is no entry in the EMPODAT database for Benzalkonium chloride. However, ECHA states that Benzalkonium chloride is very toxic to aquatic life with long lasting effects and indicates release to the environment is likely to occur from the manufacture of the substance. The effective toxic and genotoxic concentrations of benzalkonium chloride were found to be far lower than measured occurrence in surface waters (μg/L) a concerning environmental risk cannot be excluded (Lavorgna et al., 2016).Therefore, it is strongly recommended to update the NORMAN EMPODAT database with the monitoring data, this research is based on and to implement a monitoring in Sweden. These were tentative qualitative analyzes for the identified emerging substances. However, the final emission modelling and risk estimation needs to be done by Swedish officials that have access to quantitative emission data.

5.1.4. Identification of emerging substances as bases for future monitoring recommendations

All identified substances that were present in high quantities or had increasing trends were compared to the NORMAN EMPODAT database in order to estimate if the substances are already occurring in the environment. For all substances, unavailable on EMPODAT, data could be could potentially be available on IPCHEM, a supra-database, redirecting to other databases, such as NORMAN EMPODAT. Possibly there is internal Swedish monitoring data that is not publicly available. Swedish officials should check internal databases for such data and make it available on the NORMAN EMPODAT database for emerging pollutants.

The recommendations that derive for monitoring, mainly address the use time trends. As mentioned before a complete qualitative analysis holding emission data, toxicological profiles and physio-chemical properties need to be discussed further to evaluate if a substance needs to be monitored. This was done tentatively, however a sound validation of the risks of the identified chemicals needs to be addressed by experts and is beyond the scope of this thesis.

5.1.5. Limitations

There are limitations with regard to data quality and assumptions made in this thesis that need to be addressed. An important limitation of the results was shown in chapter 4.4.2 where the lack of one CAS number in the Candidate list entry "nonylphenols" was taken into account. This is due to the lack of an official and exhaustive list of CAS numbers that belong to one Candidate List entry. There are lists provided by ECHA that list various CAS identifiers, but ECHA states that those are "non-exhaustive" lists. During the last analysis it turned out by chance that this regulated nonylphenol increased tremendously in 2014. Therefore, it is important to emphasize that some substances can slip through the analysis not only with regard to this thesis but also official screenings. Possibly the industry is aware of substances that fall under an entry on the candidate list but not clearly identified with a CAS as in the described example. This could be used for regrettable substitution as it happened in Sweden with nonylphenol, where a nonylphenol with well stated CAS number (104-40-5) was replaced by one that was less accessible (*84852-15-3)*. The first step is to have official exhaustive lists by ECHA of isomers that belong to one Candidate List entry.

The same non- exhaustiveness applies to the self- compiled lists of surfactants, plasticizers and flame retardants in this thesis. There is always the possibility and high likelihood that relevant substances were not taken into account. There are thousands of industrially used flame retardants, surfactants and plasticizers, which could not all be researched and included in this thesis. Nonetheless, the used lists were compiled with the aspiration of covering most of the quantitative relevant substances within the respective technical use categories.

As a general limitation for the substitution patterns it needs to be stated, that the listed flame retardants, plasticizers and surfactants are likely to have uses additional to their application as flame retardants, plasticizers or surfactants. This means that a substance can be used mainly as flame retardant but also in other uses such as flocculation agent, such as seen in the examples of aluminum hydroxide and magnesium hydroxide. For this reason it was originally planned to work with the Use Category tonnages from SPIN, where tonnages of certain substance for particular uses were ment to be listed. However,as described in detail in chapter 8.3 Utility of the SPIN use categories (Use category UC62)", these showed tremendous data gaps and were therefore not workable. Yet, total use data gives clear hints and patterns for chemicals independent of their additional uses.

6. Conclusion

The goals of this thesis were an understanding of the impact of regulation on the use of SVHC, the patterns of substitution of chemicals (which chemicals are substituted and by what) and the identification of substances that could constitute future EPs in Swedish surface waters. Time tends for various problematic substances and substance groups based on the Swedish SPIN data were retrieved. SPIN contains data on annual use of industrial chemicals. Those time trends were used to 1) analyze the regulative impact of REACH candidate list on wrSVHC use in Sweden 2) identify substitute chemicals and patterns of substitution 3) single out potentially hazardous substitutes by the NORMAN list of emerging substances. Further, the identification of substitutes was used to make tentative monitoring recommendations that need to be revalidated by experts.

It was found that REACH regulation lead to a clear decrease of wrSVHC use in Sweden. These findings were compared to a control group of unregulated wrPMOC, which showed no decrease in industrial use. It can be concluded that regulations such as REACH or the WFD decreases the industrial use of the substances regulated. This result was expected but never proven before on the base of industrial use data. The results shown can be considered as validation of the SPIN total use data.

Further, multiple substitute chemicals with increasing industrial use trends were identified and are presented according to technical use category in results. For each technical use category the patterns of substitution were identified. In Sweden, plasticizers showed a clear substitution pattern from phthalates to non-phthalate plasticizers with a regime shift around 2012. A sharp decline of phthalates was observed in 2013, accompanied in a gradual increase in non-phthalate plasticizers starting from 2009. Flame retardants experienced no decrease in halogenated flame retardants, however an increase in halogen- free flame was observed. Surfactants showed a decreased use for alkylphenols with clearly stated CAS in ECHA starting from 2000 with complete disappearance in 2012. Yet, one alkylphenol with a little accessible CAS number (nonylphenol, CAS: (84852-15-3) skyrockets in 2015 from 0 tons to over 1600t, demonstrating regrettable substitution. Additionally, substitutes that are already found in the environment were singled out by comparison of the identified substitutes with the NORMAN list of emerging substances. Generally, the found substances are not highly utilized substances with all chemicals being used < 300tpa. The NORMAN EMPODAT database did not hold sufficient data to any of the identified chemicals, probably requiring monitoring in the future.

The greatest challenge with regard to emerging pollutants currently is the determination of mixture effects of chemicals in the aquatic environment. Nonetheless, the determination of mixture effects does not solve the problem of chemical burden and emerging pollutants in surface waters and other environmental matrixes. In order to combat emerging substances in the environment sustainably, use and emission of hazardous substances need to be minimized. It is essential to have a good database for analysis of reduction and patterns of substitution such as SPIN. On a European scale, REACH offers a database with registration data. However, use categories for emission modelling are absolutely necessary and not available yet. Therefore, more effort should be placed at the creation of solution oriented data collection with industrial use data holding use categories to derive reliable exposure estimations. Moreover, surveillance the chemical composition of imported articles needs to be implemented. Further, the SPIN database for Nordic countries could be extended by pesticides and chemicals in articles. Industrial use time trends have shown to be a useful tool in detecting substitution patterns for EP screening. Not only in Scandinavia but also on European scale a database with time trends for industrial use is a long term goal for preventative measures with respect to regrettable substitution.

Most importantly, future efforts need to be directed at functional substitution of hazardous chemicals. Regrettable substitution of hazardous chemicals needs to be prevented, by analyzing chemical use databases such as SPIN for substitution patterns. Once regrettable substitution is identified, information based approaches directed to companies using these substances need to be applied to inform the responsible company about tools and possibilities to avoid regrettable substitution (Blum et al., 2017). Examples for such tools are GreenScreen, SIN, SINimilarity, Subsport among other sustainable chemistry initiatives such as the newly founded ISC[3].

7. References

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8. Appendix

8.1. Substance groups and time trends

8.1.1. Water relevant Substances of Very High Concern (wrSVHC)

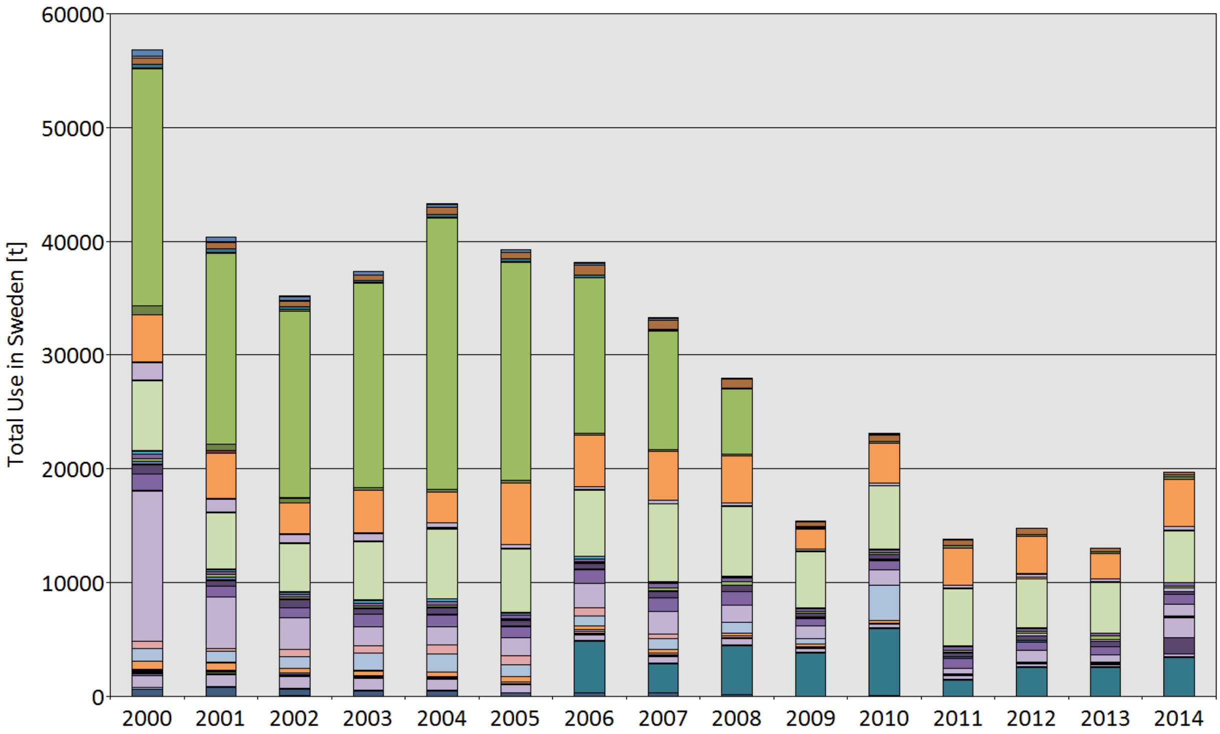
In this thesis, water relevant SVHC are defined as SVHC from the REACH Candidate list, which are also included in the NORMAN list of emerging pollutants or in the list of priority substances of the WFD. All SVHC were compared to the WFD list. 10 SVHC have been found on the WFD list. One of these substances has insufficient data in SPIN. For nine substances time trends could be derived from SPIN. These nine substances have been further assessed in this thesis. In addition, 20 SVHC have been found on the NORMAN list of ES. Only seven of these substances had end-to-end time trends. They were further assessed in the thesis. In total, 16 SVHC have been identified as water relevant SVHC. They are listed in the following Appendix Table 1

Appendix Table 1: List of 16 water relevant SVHC

	CAS	Name	Abbreviations	Source
1	104-40-5	4-Nonylphenol, branched and linear	4NP	WFD
2	107-06-2	1,2-dichloroethane	Dichlorethane	WFD
3	117-81-7	Bis (2-ethylhexyl)phthalate (DEHP)	DEHP	WFD
4	140-66-9	4-(1,1,3,3-tetramethylbutyl)phenol	Tetramethylbutylphenol	WFD
5	25154-52-3	4-Nonylphenol, branched and linear, ethoxylated	4NP, ethoxylated	WFD
6	50-32-8	Benzo[def]chrysene (Benzo[a]pyrene)	Benzo[a]pyrene	WFD
7	7440-43-9	Cadmium	Cadmium	WFD
8	79-01-6	Trichloroethylene	Trichloroethylene	WFD
9	85535-84-8	Alkanes, C10-13, chloro (Short Chain Chlorinated Paraffins)	SCCP	WFD
10	115-96-8	Tris(2-chloroethyl)phosphate	TCEP	NORMAN
11	85-68-7	Benzyl butyl phthalate (BBP)	BBP	NORMAN
12	84-74-2	Dibutyl phthalate (DBP)	DBP	NORMAN
13	80-05-7	4,4'-isopropylidenediphenol Bisphenol A; BPA	BPA	NORMAN
14	1163-19-5	Bis(pentabromophenyl) ether (decabromodiphenyl ether) (DecaBDE)	DecaBDE	NORMAN
15	3194-55-6	Hexabromocyclododecane (HBCDD) and all major diastereoisomers identified: 1,2,5,6,9,10-hexabromocyclodecane	HBCDD	NORMAN
16	78-00-2	Tetraethyllead	Tetraethyllead	NORMAN

8.1.1.1. Cumulative SVHC time trends Sweden and other countries

In order to assess the regulative impact of the REACH Candidate list on the use of SVHC (not only the water relevant SVHC) queries were constructed to compare the development of SVHC use in all Nordic countries. Appendix Figure 1: Time trends of cumulative SVHC tonnage in Sweden shows the time trends for Sweden, where a clear decrease over time is observed.



Appendix Figure 1: Time trends of cumulative SVHC tonnage in Sweden

The graph is dominated by some highly used substances; therefore the biggest shares are indicated.

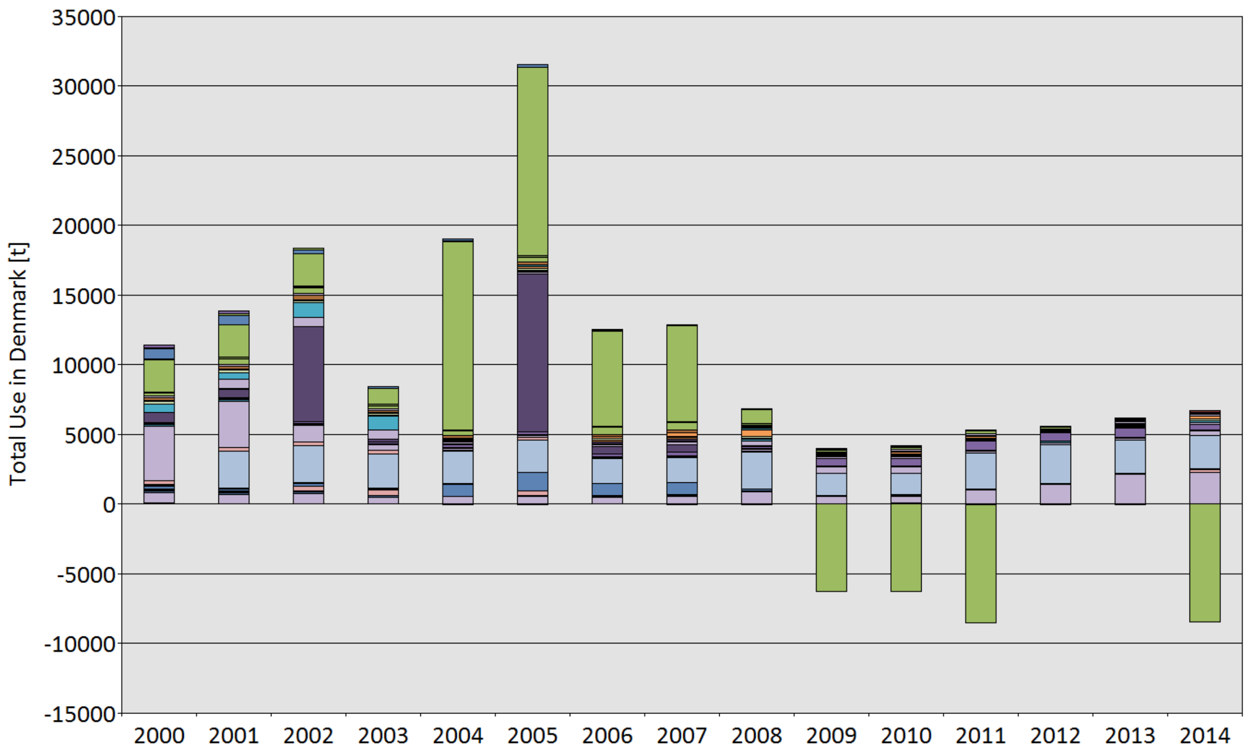
Bright light green bars = pitch coal tar, orange = methyloxirane

Light green = disodium tetraborate, greyblue = 1,2- dichlorethane.

Lavender = DEHP.

The slight increase in 2014 is due to the regrettable substitution of 4NP (CAS: 84852-15-3) that peaked in 2014 and some other increases in already existing SVHCs.

In Denmark the decrease of SVHCs becomes apparent after 2005 as shown in Appendix Figure 2: Time trends of cumulative SVHC tonnage in Denmark. Again, the trends are dominated by few substances as shown in the legend below.

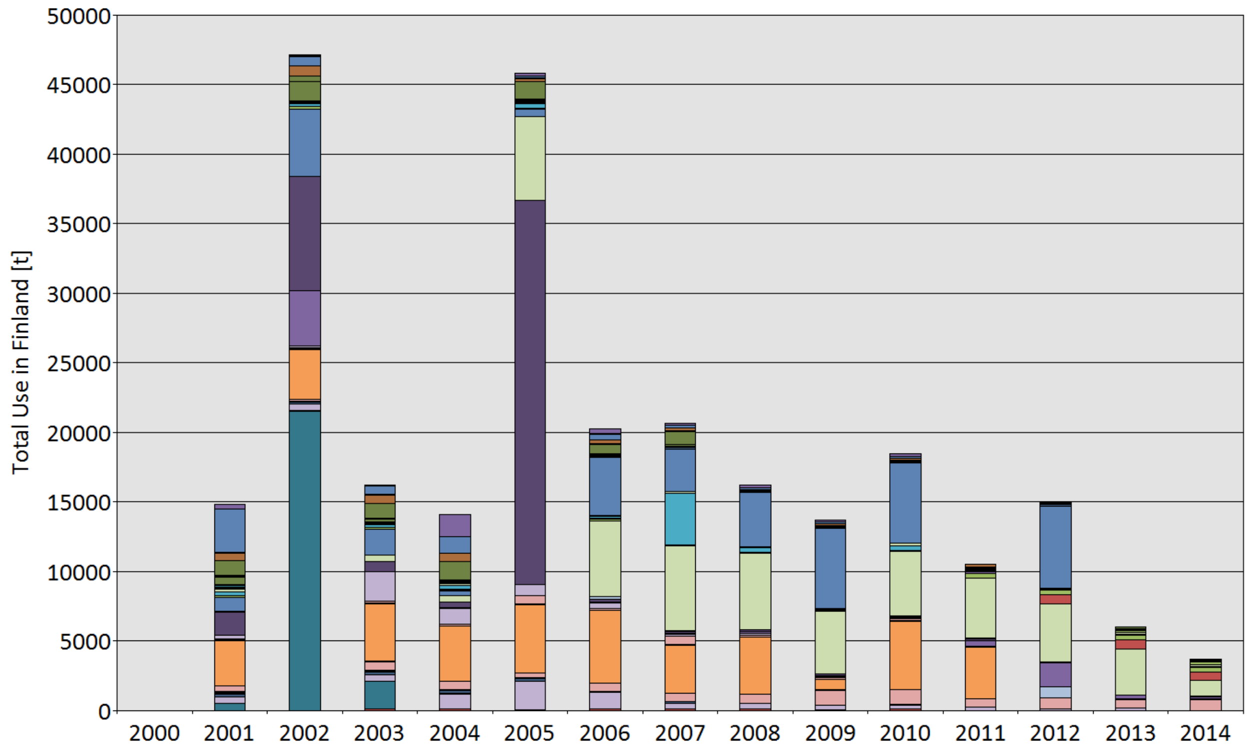


Appendix Figure 2: Time trends of cumulative SVHC tonnage in Denmark

- Pitch, coal tar, high temp.
- Benzo[def]chrysene (Benzo[a]pyrene)
- 1-Methyl-2-pyrrolidone (NMP)

Interestingly, Denmark exports pitch. Generally a slight increase in SVHC can be detected over the last years, mainly caused my NMP and a multitude of SVHC in small amounts.

In Finland the decrease of SVHC becomes also apparent after 2005 as shown inAppendix Figure 2: Time trends of cumulative SVHC tonnage in Denmark. Again, the trends are dominated by few substances as shown in the legend below.



Appendix Figure 3: Time trends of cumulative SVHC tonnage in Finland

Orange bars = Acrylamide

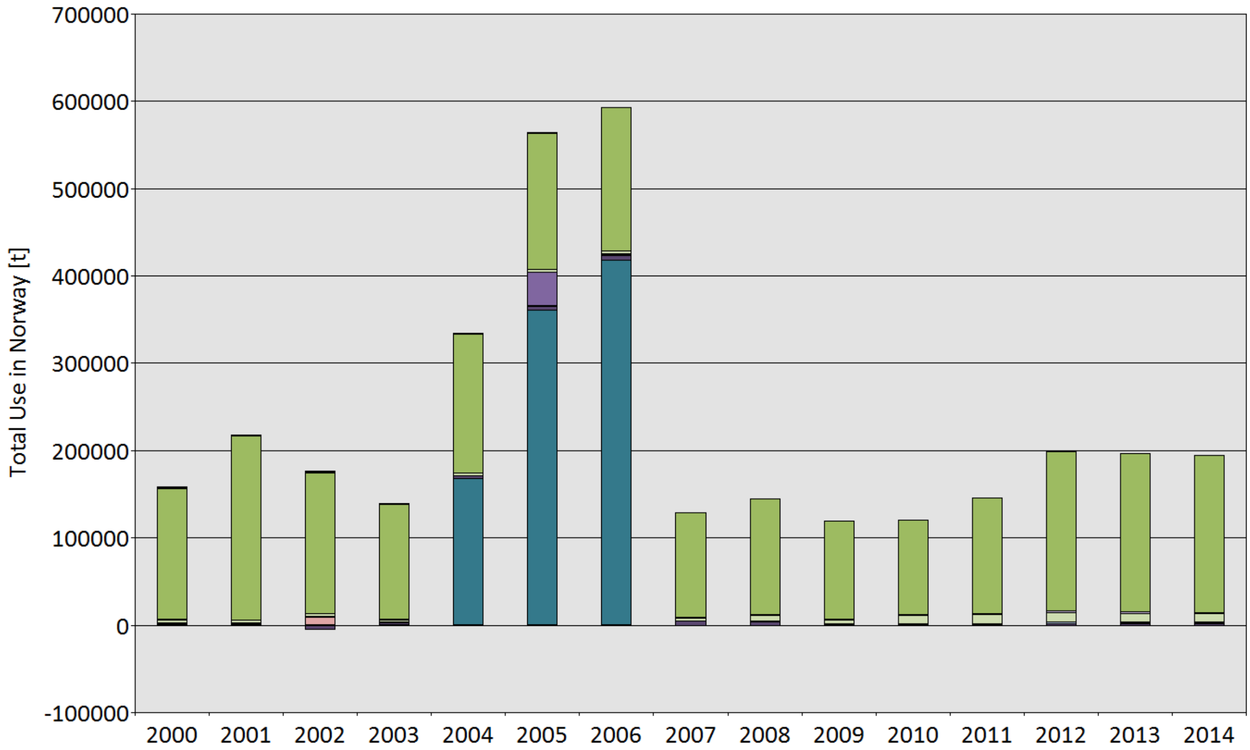
Light green = Cobalt (II) sulfate

Blue bars = Formamide

Red bars = Diarsenic trioxide

Interestingly, Finland started 2012 with the use of diarsenic trioxide.

In Norway the decrease of SVHC becomes apparent after 2006 as shown inAppendix Figure 2: Time trends of cumulative SVHC tonnage in Denmark. Again, the trends are dominated by few substances as shown in the legend below. Norway is dominated by pitch coal tar use, which is increasing. The decrease after 2006 is attributed to the decline of 1,2- dichlorethane.



Appendix Figure 4: Time trends of cumulative SVHC tonnage in Norway

light green bars = pitch coal tar

dark green bars = 1,2- dichlorethane

8.1.2. Substance groups: flame retardants

The flame retardant list consists of two contributions: Substances from the NORMAN network list of emerging pollutants, which are characterized as "flame retardants and flame retardants reported in literature. In total, the list contains 83 substances. They are divided in two classes: halogenated flame retardants and halogen-free flame retardants.

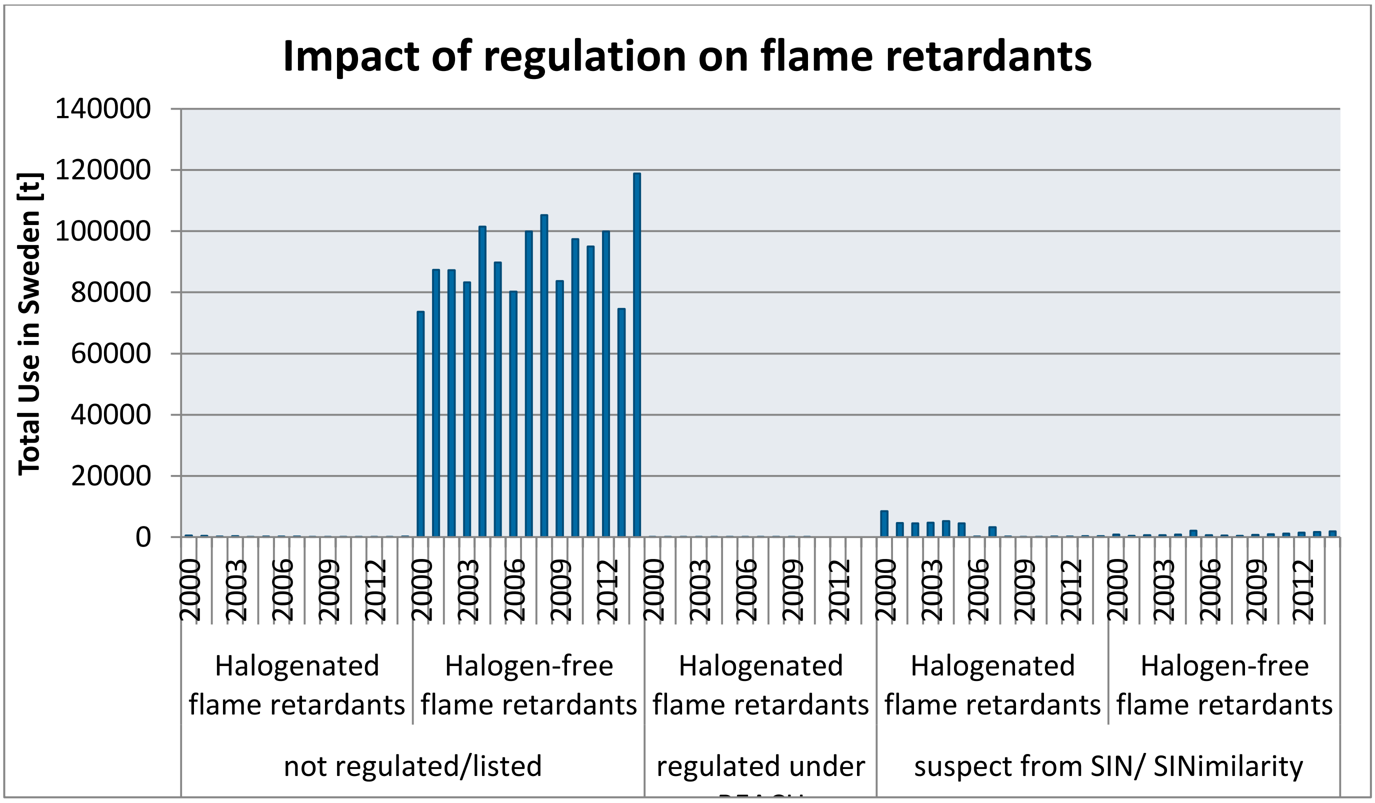
Appendix Table 2: Compiled flame retardants list

	Name	CAS	Class
1	Hypophosphite, calcium salt	7789-79-9	HFFR
2	triphenyl phosphate	115-86-6	HFFR
3	tetraphenyl m-phenylene bis(phosphate)	57583-54-7	HFFR
4	Tris(2-methylpropyl) phosphate	1330-78-5	HFFR
5	diphenyl tolyl phosphate	26444-49-5	HFFR
6	sodium benzoate	532-32-1	HFFR
7	Dimethyl methyl phosphonate (DMMP)	756-79-9	HFFR
8	Magnesium dihydroxide	1309-42-8	HFFR
9	Aluminum Hydroxide/Alumina Trihydrate (ATH) (recommended with further addition of phosphorus or brominated FRs)	21645-51-2	HFFR
10	tributyl phosphate	126-73-8	HFFR
11	triethyl phosphate	78-40-0	HFFR
12	Tris(2-ethylhexyl)phosphat (TEHP)	78-42-2	HFFR
13	Tris(2-butoxyethyl) phosphate	78-51-3	HFFR
14	Triisobutyl phosphate	126-71-6	HFFR
15	6H-dibenz[c,e][1,2]oxaphosphorin 6-oxide	35948-25-5	HFFR
16	Bisphenol-A bis(diphenyl phosphate)	5945-33-5	HFFR
17	2,2-Bis(bromomethyl)-1,3-propanediol	3296-90-0	HFR
18	Boehmite (Al(OH)O)	1318-23-6	HFFR
19	ammonium bromide	12124-97-9	HFR
20	Polyphosphoric acids, ammonium salts	68333-79-9	HFFR
21	Phosphoric trichloride, reaction products with bisphenol A and phenol	181028-79-5	HFFR
22	Graphite	7782-42-5	HFFR
23	Melamine	108-78-1	HFFR
24	Melamine cyanurate	37640-57-6	HFFR
25	Melamine-phosphate	41583-09-9	HFFR
26	Melapur M 200	218768-84-4	HFFR
27	Mica-group minerals	12001-26-2	HFFR
28	Polyetherimide	61128-46-9	HFFR
29	Phenol, isobutylenated, phosphate	68937-40-6	HFFR
30	Pentaerythrityl phosphate	5301-78-0	HFFR
31	Decabromdiphenylethan	84852-53-9	HFR
32	tris(2,3-dibromopropyl) phosphate	126-72-7	HFR
33	2,4,6-Tribromophenyl allyl ether	3278-89-5	HFR

34	bis(allyl ether) of tetrabromobisphenol A	25327-89-3	HFR
35	1,1,2,3,4,4-Hexabromo-2-butene	36678-45-2	HFR
36	Tetrabromocyclooctane	31454-48-5	HFR
37	Dibromoethyldibromocyclohexane	3322-93-8	HFR
38	Chloropentabromocyclohexane	87-84-3	HFR
39	2,4,6-tribromophenol	118-79-6	HFR
40	Tetrabromobisphenol A	79-94-7	HFR
41	tris[2-chloro-1-(chloromethyl)ethyl] phosphate	13674-87-8	HFR
42	bis(2-ethylhexyl) tetrabromophthalate	26040-51-7	HFR
43	1,6,7,8,9,14,15,16,17,17,18,18-dodecachloropentacyclo[12.2.1.16,9.02,13.05,10]octadeca-7,15-diene	13560-89-9	HFR
44	BDE No 153 solution	68631-49-2	HFR
45	BDE No 47 solution	5436-43-1	HFR
46	BDE No 28 solution	41318-75-6	HFR
47	1,1'-(isopropylidene)bis[3,5-dibromo-4-(2,3-dibromopropoxy)benzene]	21850-44-2	HFR
48	tris(2,3-dichloropropyl) phosphate	78-43-3	HFR
49	Tris(2,3-dichloropropyl) phosphate	13674-84-5	HFR
50	Alkanes, C18-28, chloro	85535-86-0	HFR
51	Tetrabromophthalate diol(TBPD)	20566-35-2	HFR
52	1,2-Bis(tetrabromophthalimido) ethane	32588-76-4	HFR
53	Chlorendic acid	115-28-6	HFR
54	1,4,5,6,7,7-hexachloro-8,9,10-trinorborn-5-ene-2,3-dicarboxylic anhydride	115-27-5	HFR
55	2,2-bis(chloromethyl)trimethylene bis(bis(2-chloroethyl)phosphate)	38051-10-4	HFR
56	hexachlorocyclopentadiene	77-47-4	HFR
57	tetrachlorophthalic anhydride	117-08-8	HFR
58	1,2,4,5-tetrabromo-3,6-bis(pentabromophenoxy)benzene	58965-66-5	HFR
59	Tris(tribromoneopenthyl)phosphate	19186-97-1	HFR
60	1,3,5-Triazine, 2,4,6-tris(2,4,6-tribromophenoxy)-	25713-60-4	HFR
61	1,1'-(isopropylidene)bis[3,5-dibromo-4-(2,3-dibromo-2-methylpropoxy)benzene]	97416-84-7	HFR
62	N-(2,3-dibromopropyl)-4,5- dibromotetrahydrophthalimide	93202-89-2	HFR
63	1,1,2,3,4,4-Hexabromo-2-butene	72108-73-7	HFR
64	Hexabromohexene	125512-87-0	HFR
65	Tetrabromocyclooctane	3194-57-8	HFR
66	BDE No 206 solution	63387-28-0	HFR
67	BDE No 183 solution	207122-16-5	HFR
68	BDE No 154 solution	207122-15-4	HFR
69	2,2',4,5'-Tetrabromobiphenyl	60044-24-8	HFR
70	2,4-Dibromophenol	615-58-7	HFR
71	Dibromoneopentyl glycol	68928-70-1	HFR
72	Benzene, ethenyl-, homopolymer, brominated	88497-56-7	HFR
73	Hexachlorocyclopentadiene	1195978-93-8	HFR
74	Carbonic dichloride, polymer with 4,4'-(1-methylethylidene)bis[2,6-dibromophenol], bis(2,4,6-tribromophenyl) ester	71342-77-3	HFR
75	Carbonic dichloride, polymer with 4,4'-(1-methylethylidene)bis[2,6-dibromophenol] and phenol	94334-64-2	HFR

76	Poly(pentabromobenzyl acrylate)	59447-57-3	HFR
77	Benzene, ethenyl-, ar-bromo derivs	148993-99-1	HFR
78	1,2,5,6,9,10-hexabromocyclodecane	3194-55-6	HFR
79	DecaBDE	1163-19-5	HFR
80	diphenyl ether, octabromo derivative	32536-52-0	HFR
81	tris(2-chloroethyl) phosphate	115-96-8	HFR
82	1,1'-[ethane-1,2-diylbisoxy]bis[2,4,6-tribromobenzene]	37853-59-1	HFR
83	Hexabromobenzene (HBB)	87-82-1	HFR

Appendix Figure 5: Impact of regulation on flame retardants shows the time trends of various flame retardant groups according to their regulation status. Generally, not regulated, halogen free flame retardants increase steadily. Moreover, it can be seen that there are no halogen free flame retardants that are regulated under REACH. Moreover, halogenated flame retardants that are suspect from SIN/SINimilarity decline over time, whereas halogen free flame retardants that are suspects from SIN/SINimilarty slightly increase. However, there are some halogen free flame retardants that are suspect to be detrimental to humans or the environment based on their structural similarity to other hazardous chemicals assessed by SIN/SINimilarity.



Appendix Figure 5: Impact of regulation on flame retardants

Appendix Table 3: Flame retardant ESs shows the flame retardants from the self- compiled list of FRs that is found in the environment according to the NORMAN network. Some of these substances are found on SPIN, some show confidential data and others have no entries at all.

Appendix Table 3: Flame retardant ESs

	Name (FR with concerning time trend marked in red)	CAS	Used amount 2014 [t]
1	Tris(2,3-dichloropropyl) phosphate	13674-84-5	148
2	triphenyl phosphate	115-86-6	97
3	Triethyl phosphate	78-42-2	36
4	Decabromodiphenyl ethane	79-94-7	34
5	Tris(2-methylpropyl) phosphate	1330-78-5	27
6	1,4,5,6,7,7-hexachloro-8,9,10-trinorborn-5-ene-2,3-dicarboxylic anhydride	115-27-5	17
7	Brominated anionic styrene polymer + bis(pentabromophenyl) ethane	84852-53-9	9
8	Tris(2-ethylhexyl) phosphate	78-51-3	9
9	Tris(2-butoxyethyl) phosphate	126-71-6	8
10	tetraphenyl m-phenylene bis(phosphate)	57583-54-7	7
11	tributyl phosphate	126-73-8	6
12	triethyl phosphate	78-40-0	7
13	bis(2-ethylhexyl) tetrabromophthalate	26040-51-7	1
14	carbon tetrachloride	56-23-5	0
15	hexachlorocyclopentadiene	77-47-4	confidential
16	N,N'-ethylenebis(3,4,5,6-tetrabromophthalimide)	32588-76-4	0
17	Tetrabromobisphenol A (TBBPA)	20566-35-2	0
18	1,1'-(isopropylidene)bis[3,5-dibromo-4-(2,3-dibromopropoxy)benzene]	21850-44-2	confidential
19	1,1'-[ethane-1,2-diylbisoxy]bis[2,4,6-tribromobenzene]	37853-59-1	0
20	1,2,4,5-tetrabromo-3,6-bis(pentabromophenoxy)benzene	58965-66-5	N/A
21	1,2,5,6,9,10-hexabromocyclodecane	3194-55-6	confidential
22	1,4,5,6,7,7-Hexachloro-8,9,10-Trinorborn-5-ene-2,3-Dicarboxylic acid	115-28-6	N/A
23	1,6,7,8,9,14,15,16,17,17,18,18-dodecachloropentacyclo[12.2.1.16,9.02,13.05,10]octadeca-7,15-diene	13560-89-9	confidential
24	1-Propanol, 3-bromo-2,2-bis(bromomethyl)-, 1,1',1''-phosphate	19186-97-1	confidential
25	2,2-bis(chloromethyl)trimethylene bis(bis(2-chloroethyl)phosphate)	38051-10-4	N/A
26	6H-dibenz[c,e][1,2]oxaphosphorin 6-oxide	35948-25-5	confidential
27	DecaBDE	1163-19-5	N/A
28	diphenyl ether, octabromo derivative	32536-52-0	N/A
29	Phosphoric acid, P,P'-[(1-methylethylidene)di-4,1-phenylene] P,P,P',P'-tetraphenyl ester	3296-90-0	confidential

30	TCEP	51805-45-9	confidential
31	tetrachlorophthalic anhydride	117-08-8	N/A
32	tris(2-chloroethyl) phosphate	115-96-8	confidential
33	Tris(2-chloroethyl) phosphate	5945-33-5	confidential

The list shows all the flame retardants that were found on NORMAN list of ES. This means that those substances were already identified in the environment. ES with concerning trends are marked in red.

8.1.3. Substance groups: plasticizers

The plasticizer list holds 80 plasticizers, 34 phthalates and 46 others plasticizers retrieved from the NORMAN network (NORMAN network, 2005) the SIN list (The International Chemical Secretariat, 2017), the German Environmental Specimen Bank (German Environmental Specimen Bank, 2017) and some relevant publications (Zheng et al., 2007; Fromme et al., 2002). All substances that were retrieved from the described sources are listed in Appendix Table 4.

Appendix Table 4: Compiled plasticizer list

No	Name	CAS Number	Class
1	Benzylbutylphthalate	85-68-7	Phthalates
2	Bis(2-ethylhexyl)phthalate;DEHP	84777-06-0	Phthalates
3	Bis(2-methoxyethyl)phthalate	117-82-8	Phthalates
4	DEHP (bis(2-ethylhexyl) phthalate)	117-81-7	Others
5	dihexylphthalate(DHP)	605-50-5	Phthalates
6	Diisobutyl phthalate	84-69-5	Phthalates
7	Diisodecylphthalate	71888-89-6	Phthalates
8	Diisodecylphthalate	776297-69-9	Phthalates
9	Diisononyl phthalate	28553-12-0	Phthalates
10	Diisononylphthalat	68515-48-0	Phthalates
11	Di-n-butylphthalate	84-74-2	Phthalates
12	Di-n-octylphthalate	117-84-0	Phthalates
13	Di-n-octylphthalate	131-18-0	Phthalates
14	1,2-benzenedicarboxylicacid,di-C8-10-branchedalkylesters,C9-rich	68515-50-4	Phthalates
15	Bisphenol A	80-05-7	Others
16	Di-n-hexyl phthalate	84-75-3	Phthalates
17	N-Methyl-2-pyrrolidone	872-50-4	Others
18	BEH-TEBP	26040-51-7	Phthalates
19	Diethyl phthalate	84-66-2	Phthalates
20	DiisononylPhthalatese	84-61-7	Phthalates
21	Diundecyl	3648-20-2	Phthalates
22	LCCPs, Cn≥18	63449-39-8	Others
23	Tris(2-methylpropyl) phosphate	68411-66-5	Others
24	Tris(2-ethylhexyl)trimellitat	3319-31-1	Others

25	Tricresyl phosphate	1330-78-5	Others
26	Tri-(heptyl, nonyl) trimellitate LTM	67989-23-5	Others
27	oxydiethylene dibenzoate	120-55-8	Others
28	Monomethyl adipate	627-91-8	Others
29	Isopropylphenyl diphenyl phosphate / IPPDPP	56803-37-3	Others
30	Diphenyl cresyl phosphate	26444-49-5	Others
31	Dioctyl terephthalate	6422-86-2	Others
32	Dioctyl adipate	123-79-5	Others
33	Dimetylphthalate	131-11-3	Phthalates
34	diisononyl adipate	33703-08-1	Others
35	Diisodecyl phthalate	26761-40-0	Phthalates
36	Dibutyl sebacate	109-43-3	Others
37	Cresyl diphenyl phosphate / CDP	28108-99-8	Others
38	bis(2-propylheptyl) phthalate	53306-54-0	Others
39	Bis(2-ethylhexyl)adipat	103-23-1	Others
40	2-Ethylhexanoic acid 2-ethylhexyl ester	7425-14-1	Others
41	1,2-Benzenedicarboxylic acid, di-C9-11-branched alkyl esters, C10-rich	68515-49-1	Phthalates
42	Triethylene glycol dihexanoate	94-28-0	Others
43	Triethyl citrate	77-93-0	Others
44	Tributylacetylcitrate	126-71-6	Others
45	Tributyl phosphate	126-73-8	Others
46	Tetraethylene glycol diheptanoate	70913-85-8	Others
47	tert-Bytulphenyl diphenyl phosphate / TBPDPP	68937-40-6	Others
48	Siloxanes and Silicones, di-Me, Me hydrogen	68037-59-2	Others
49	Siloxanes and Silicones, di-Me, hydrogen-terminated	70900-21-9	Others
50	N-ethyl toluene sulfonamide	8047-99-2	Others
51	N-butyl-benzenesulfonamide	3622-84-2	Others
52	N-(2-hydroxypropyl) benzene sulfonamide	35325-02-1	Others
53	Melamine	108-78-1	Others
54	MCCPs	85535-86-0	Others
55	hydroxycyclohexyl phenyl ketone	947-19-3	Others
56	Epoxidized soybean oil (ESBO)	08.07.8013	Others
57	Dipentaerythritol	126-58-9	Others
58	Dipentaerythriol	78-24-0	Others
59	Dimethyladipat	627-93-0	Others
60	Dimethyl adipate	474919-59-0	Others
61	Diisopropylnaphthalene	38640-62-9	Others
62	Diisononyl adipate DINCH	166412-78-8	Others
63	Dibutyl maleate	105-76-0	Others
64	Citric acid	77-92-9	Others
65	Chlorinated paraffins	85535-84-9	Others
66	Alkylsulfonic phenyl ester	91082-17-6	Others
67	Acetyltributylcitrate	77-90-7	Others
68	2,2,4-trimethylpentane-1,3-diol diisobutyrate	6846-50-0	Others
69	Glycerides, castor-oil mono-, hydrogenated, acetates/ COMGHA	736150-63-3	Others
70	Dimethyl phthalate	131-17-9	Phthalates

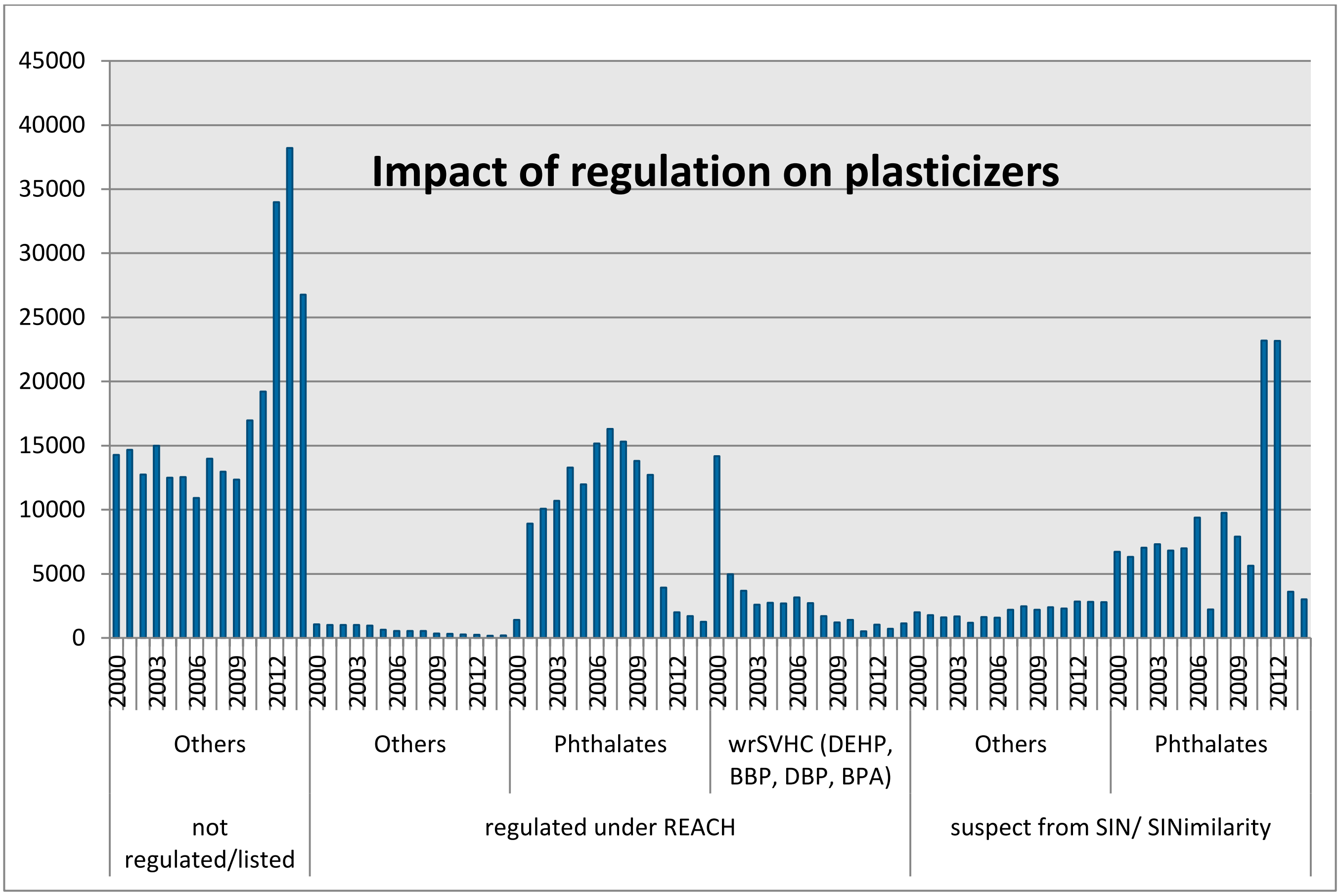
71	Diethyl phthalate	131-16-8	Phthalates
72	Diallyl phthalate	84-64-0	Phthalates
73	Di-n-propyl phthalate	146-50-9	Phthalates
74	Di-n-butyl phthalate	41451-28-9	Phthalates
75	Diisobutyl phthalate	89-19-0	Phthalates
76	Butyl cyclohexyl phthalate	27554-26-3	Phthalates
77	Di-n-pentyl phthalate	119-07-3	Phthalates
78	Dicyclohexyl phthalate	85507-79-5	Phthalates
79	Butyl benzyl phthalate	119-06-2	Phthalates
80	Di-n-hexyl phthalate	68515-47-9	Phthalates

Appendix Table 5: Plasticizer ESs

	Name (EPs with concerning trends marked in red)	CAS	Used amount 2014, Sweden [t]
1	2,2,4-trimethylpentane-1,3-diol diisobutyrate	6846-50-0	220
2	2-Ethylhexanoic acid 2-ethylhexyl ester	7425-14-1	2
3	Acetyltributylcitrate	77-90-7	5
4	BEH-TEBP	26040-51-7	1
5	Benzylbutylphthalate	85-68-7	7
6	Bisphenol A	80-05-7	21
7	Diethyl phthalate	84-66-2	9
8	Diisodecyl phthalate	26761-40-0	932
9	Diisononyl phthalate	28553-12-0	967
10	Diisopropylnaphthalene	38640-62-9	19
11	Dimetylphthalate	131-11-3	85
12	Di-n-butylphthalate	84-74-2	8
13	Di-n-octylphthalate	117-84-0	confidential
14	hydroxycyclohexyl phenyl ketone	947-19-3	450
15	LCCPs, Cn≥18	63449-39-8	191
16	N-butyl-benzenesulfonamide	3622-84-2	19
17	N-Methyl-2-pyrrolidone	872-50-4	196
18	Tributyl phosphate	126-73-8	6
19	Tributylacetylcitrate	126-71-6	8
20	Tricresyl phosphate	1330-78-5	27
21	Triethyl citrate	77-93-0	98

The list shows all the plasticizers that were found on NORMAN list of ES. This means that those substances were already identified in the environment. ES with concerning trends are marked in red.

Appendix Figure 6: Impact of regulation on plasticizers shows the time trends of various plasticizer groups according to their regulation status. Generally, not regulated, non-phthalate plasticizers increase steadily. Interestingly, phthalates that were not regulated under REACH increased until 2012. This indicates regrettable substitution of regulated phthalates in the early years of REACH. Since then, other plasticizers skyrocket. However, there are some other plasticizers that are increasing and are suspect to be detrimental to humans or the environment based on their structural similarity to other hazardous chemicals assessed by SIN/SINimilarity.



Appendix Figure 6: Impact of regulation on plasticizers

8.1.4. Substance groups: surfactants

The surfactant list holds 133 of surfactants and was compiled based on a literature Survey of Surfactants in the Nordic Countries (Johansson et al., 2012) complemented with the surfactants from the NORMAN network (NORMAN network, 2005), the SIN list (The International Chemical Secretariat, 2017) the HERA report (Environmental HERA report, 2013) and a book with an extensive list on APEs and AEs (Talmage, op. 1994) . All surfactants are listed as shown in Appendix Table 6.

Appendix Table 6: Compiled surfactant list

	Name	CAS	Class	Subclass
1	20-(4-nonylphenoxy)-3,6,9,12,15,18-hexaoxaicosan-1-ol	27942-27-4	APOs	Alkylphenols
2	4-Nonylphenol, branched, ethoxylated	127087-87-0	APOs	Alkylphenols
3	Nonaethylene glycol p-nonylphenyl ether	14409-72-4	APOs	Alkylphenols
4	4-TERT-NONYLPHENOLDIETHOXYLATE	156609-10-8	APOs	Alkylphenols
5	2-[2-[2-[2-(4-nonylphenoxy)ethoxy]ethoxy]ethoxy]ethanol	7311-27-5	APOs	Alkylphenols
6	2-[2-(4-nonylphenoxy)ethoxy]ethanol	20427-84-3	APOs	Alkylphenols
7	4-Nonylphenol, ethoxylated	26027-38-3	APOs	Alkylphenols
8	4-Nonyl Phenol Monoethoxylate	104-35-8	APOs	Alkylphenols
9	2-[2-[2-[2-[2-[2-(4-nonylphenoxy)ethoxy]ethoxy]ethoxy]ethoxy]ethoxy]ethanol	34166-38-6	APOs	Alkylphenols
10	2-[2-[4-(1,1,3,3-tetramethylbutyl)phenoxy]ethoxy]ethanol / 4-Octylphenol di-ethoxylate	2315-61-9	Others	Alkylphenols
11	2-[4-(1,1,3,3-tetramethylbutyl)phenoxy]ethanol / 4-Octylphenol mono-ethoxylate	2315-67-5	Others	Alkylphenols
12	Isononylphenol-ethoxylate	37205-87-1	APOs	Alkylphenols
13	Phenol, nonyl-, branched	90481-04-2	APOs	Alkylphenols
14	4-(1,1,3,3-tetramethylbutyl)phenol	140-66-9	Others	Alkylphenols
15	Nonylphenol, ethoxylated	9016-45-9	APOs	Alkylphenols
16	p-nonylphenol	104-40-5	APOs	Alkylphenols
17	Nonylphenol	25154-52-3	APOs	Alkylphenols
18	Nonylphenol, branched, ethoxylated	68412-54-4	APOs	Alkylphenols
19	Perfluorooctanoic acid (PFOA)	335-67-1	Others	Perfluorinated compounds
20	Nonadecafluorodecanoic acid (PFDA)	335-76-2	Others	Perfluorinated compounds
21	PFHxS	355-46-4	Others	Perfluorinated compounds
22	2-(2-(4-Nonylphenoxy)ethoxy)acetic acid	106807-78-7	APOs	Alkylphenols
23	4-Octylphenoxy acetic acid	15234-85-2	Others	Alkylphenols
24	C10-C14-LAS	69669-44-9	Others	LAS
25	C12-LAS	25155-30-0	Others	LAS
26	Surfinol-104	126-86-3	Others	LAS
27	(1,1,3,3-tetramethylbutyl)phenol	27193-28-8	Others	Alkylphenols
28	lithium heptadecafluorooctanesulfonate, lithium perfluorooctane sulfonate	29457-72-5	Others	Perfluorinated compounds
29	PFOS, heptadecafluorooctane-1-sulfonic acid, perfluorooctane sulfonic acid	1763-23-1	Others	Perfluorinated compounds
30	ammonium heptadecafluorooctanesulfonate, ammonium perfluorooctane sulfonate	29081-56-9	Others	Perfluorinated compounds
31	Sulfluramid (PFOSA)	4151-50-2	Others	Perfluorinated compounds
32	potassium heptadecafluorooctane-1-sulfonate, potassium perfluorooctanesulfonate	2795-39-3	Others	Perfluorinated compounds
33	diethanolamine perfluorooctane sulfonate	70225-14-8	Others	Perfluorinated compounds

34	8:2-diPAP	678-41-1	Others	Perfluorinated compounds
35	4-Nonylphenoxy acetic acid	3115-49-9	APOs	Alkylphenols
36	Naphthalene sulphonic acid	120-18-3	Others	LAS
37	Benzenesulphonic acid, C10-13 alkyl derivs., sodium salts	68411-30-3	Others	LAS
38	Sodium decylbenzenesulphonate	1322-98-1	Others	LAS
39	Benzenesulphonic acid, mono-C10-13 alkyl derivs., sodium salt	90194-45-9	Others	LAS
40	Benzenesulphonic acid, mono-C10-14 alkyl derivs., sodium salt	85117-50-6	Others	LAS
41	OPE1	9002-93-1	APOs	APOs
42	OPE2	9036-19-5	APOs	APOs
43	OPE3	68987-90-6	APOs	APOs
44	LAS1	68081-81-2	Others	LAS
45	LAS3	68584-22-5	Others	LAS
46	LAS4	68584-24-7	Others	LAS
47	LAS5	70024-69-0	Others	LAS
48	LAS6	70024-71-4	Others	LAS
49	LAS7	85117-49-3	Others	LAS
50	LAS9	85536-14-7	Others	LAS
51	LAS10	90194-26-6	Others	LAS
52	LAS11	90194-27-7	Others	LAS
53	LAS12	90194-45-9b	Others	LAS
54	LAS13	115733-10-3	Others	LAS
55	LAS14	134759-03-8	Others	LAS
56	LAS15	156105-31-6	Others	LAS
57	LAS16	722503-68-6	Others	LAS
58	LAS17	722503-69-7	Others	LAS
59	QA1	61789-18-2	Others	QAs
60	QA2	61789-71-7	Others	QAs
61	QA3	61789-77-3	Others	QAs
62	QA4	61791-10-4	Others	QAs
63	QA5	63449-41-2	Others	QAs
64	QA6	68002-60-8	Others	QAs
65	QA7	68139-30-0	Others	QAs
66	QA8	68153-30-0	Others	QAs
67	QA9	68391-01-5	Others	QAs
68	QA10	68424-85-1	Others	QAs
69	QA11	68911-87-5	Others	QAs
70	QA12	68953-58-2	Others	QAs
71	QA13	68989-03-7	Others	QAs
72	QA14	71011-24-0	Others	QAs
73	QA15	71011-25-1	Others	QAs
74	QA16	71011-26-2	Others	QAs
75	QA17	71011-27-3	Others	QAs

76	QA18	85409-22-9	Others	QAs
77	QA19	121888-66-2	Others	QAs
78	QA20	121888-68-4	Others	QAs
79	QA21	863679-20-3	Others	QAs
80	AE1	9002-92-0	AEs	AEs
81	AE2	9004-77-7	AEs	AEs
82	AE3	9004-95-9	AEs	AEs
83	AE4	9004-98-2	AEs	AEs
84	AE5	9005-00-9	AEs	AEs
85	AE7	9008-57-5	AEs	AEs
86	AE8	9038-95-3	AEs	AEs
87	AE9	24938-91-8	AEs	AEs
89	AE10	26183-52-8	AEs	AEs
90	AE12	27252-75-1	AEs	AEs
91	AE13	27306-79-2	AEs	AEs
92	AE14	31729-34-8	AEs	AEs
93	AE15	34398-05-5	AEs	AEs
94	AE16	60828-78-6	AEs	AEs
95	AE17	61702-78-1	AEs	AEs
96	AE18	61791-28-4	AEs	AEs
97	AE19	66455-14-9	AEs	AEs
98	AE20	66455-15-0	AEs	AEs
99	AE21	68002-96-0	AEs	AEs
100	AE22	68002-97-1	AEs	AEs
101	AE23	68131-39-5	AEs	AEs
102	AE24	68131-40-8	AEs	AEs
103	AE25	68154-%-1	AEs	AEs
104	AE26	68154-97-2	AEs	AEs
105	AE27	68154-98-3	AEs	AEs
106	AE28	68213-23-0	AEs	AEs
107	AE29	68213-24-1	AEs	AEs
108	AE30	68439-45-2	AEs	AEs
109	AE31	68439-46-3	AEs	AEs
110	AE32	68439-49-6	AEs	AEs
111	AE33	68439-50-9	AEs	AEs
112	AE34	68439-51-0	AEs	AEs
113	AE35	68439-54-3	AEs	AEs
114	AE36	68526-94-3	AEs	AEs
115	AE37	68526-95-4	AEs	AEs
116	AE38	68551-12-2	AEs	AEs
117	AE39	68551-13-3	AEs	AEs
118	AE40	68551-14-4	AEs	AEs
119	AE41	68603-20-3	AEs	AEs
120	AE42	68603-25-8	AEs	AEs
121	AE43	68937-66-6	AEs	AEs
122	AE44	68987-81-5	AEs	AEs
123	AE45	68991-48-0	AEs	AEs
124	AE46	69012-85-7	AEs	AEs
125	AE47	69013-18-9	AEs	AEs
126	AE48	69013-19-0	AEs	AEs

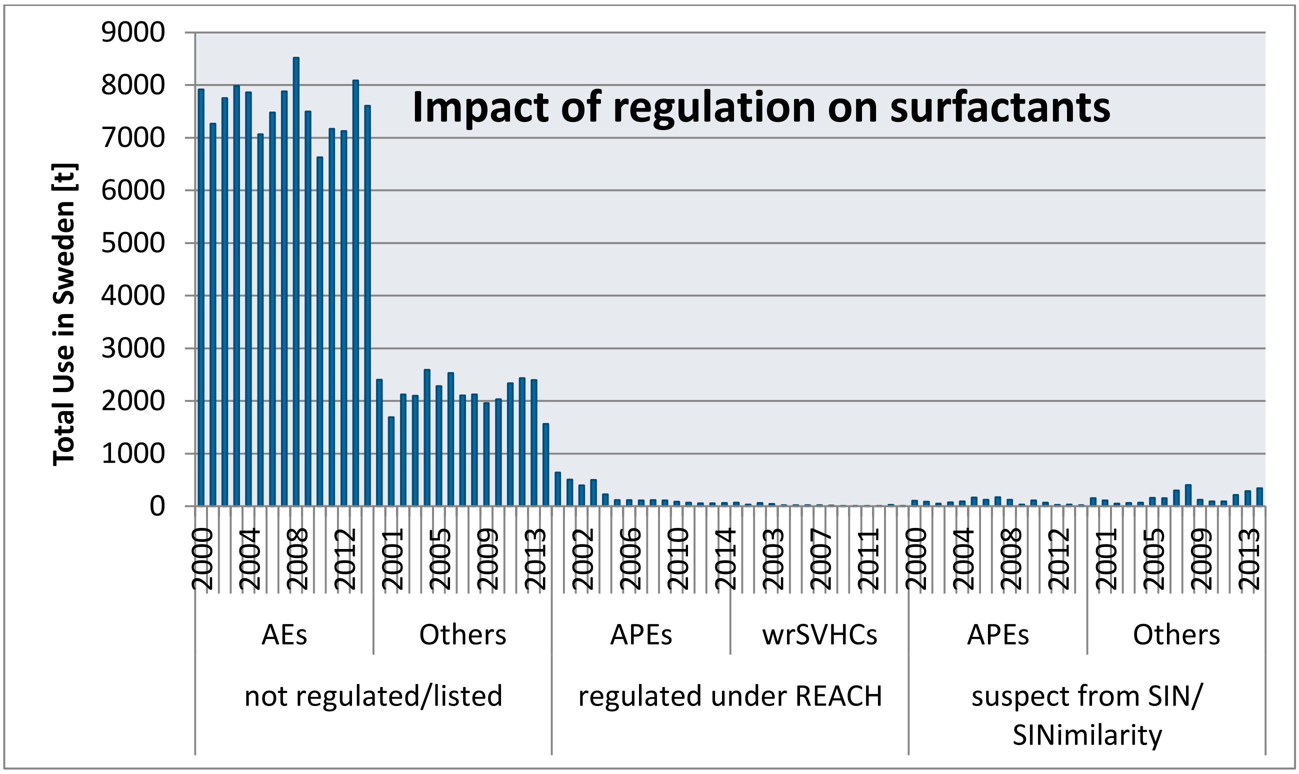
127	AE49	69227-20-9	AEs	AEs
128	AE50	69227-21-0	AEs	AEs
129	AE51	69227-22-1	AEs	AEs
130	AE52	70879-83-3	AEs	AEs
131	AE53	71011-10-4	AEs	AEs
132	AE54	71060-57-6	AEs	AEs
133	AE55	71243-46-4	AEs	AEs

Appendix Table 7: Surfactant ESs

	Name (EPs with concerning trend marked in red)	CAS	Used amounts, 2014 [t]
1	Sodium dodecylbenzenesulfonate (LAS)	25155-30-0	315
2	Benzalkonium chloride (QA)	68424-85-1	265
3	Surfinol-104	126-86-3	88
4	Benzalkonium chloride (QA)	85409-22-9	10
5	QA9	68391-01-5	5
6	QA1	61789-18-2	1
7	4-Nonylphenoxy acetic acid	3115-49-9	confidential
8	C10-C14-LAS	69669-44-9	confidential
9	Naphthalene sulphonic acid	120-18-3	confidential
10	Perfluorooctanoic acid (PFOA)	335-67-1	confidential
11	Sulfluramid (PFOSA)	4151-50-2	confidential

The list shows all the surfactants that were found on NORMAN list of ES. This means that those substances were already identified in the environment. ES with concerning trends are marked in red.

Appendix Figure 7: Impact of regulation on surfactants shows the time trends of various surfactant groups according to their regulation status. Generally, not regulated, AEs and others stay on the same level over the years. As seen in the results and discussion the surfactant list was not exhaustive neglecting some relevant substances. This is why no clear patterns can be detected.



Appendix Figure 7: Impact of regulation on surfactants

8.1.5. Persistent and mobile organic contaminants (PMOC)

The list of PMOC obtained from Dr. Reemtsma as referenced below (Schulze et al. 2017) has 59 PMOC with unambiguous CAS of which 42 were found on SPIN and 12 were identified as quantitatively relevant (> 5 t in 2000) in the Swedish SPIN data as shown in

Appendix Table 8.

Appendix Table 8: Quantitative relevant PMOC in Sweden

	PMOC Name	CAS	Used amounts, 2014 [t]
1	Melamin	108-78-1	6398
2	N-Aminoethylpiperazine	140-31-8	44
3	Toluenesulfonic acid	104-15-4	531

	PMOC Name	CAS	Used amounts, 2014 [t]
4	Cyanguanidine	461-58-5	534
5	Isophoeonediamine	2855-13-2	481
6	Benzyldimethylamine	103-83-3	22
7	Sodium Vinylsulfonate	3039-83-6	112
8	TCPP	13674-84-5	148
9	Sodium xylenesulfonate	1300-72-7	85
10	ε- Caprolactam	105-60-2	42
11	1,3-Diphenylguanidine	102-06-7	9
12	DABCO	280-57-9	3

8.2. SPIN : Limitations and opportunities

8.2.1 Potential relevance of substance not found on SPIN

For some of the substances that were relevant to this thesis no data has been found in the SPIN database. In order to estimate the magnitude of this limitation, the potential relevance of substances not covered has been assessed for the group of SVHC.

By July 2017, The REACH Candidate list has 173 entries (substances and substance groups), of which five had no CAS number. A total of 22 of the 173 entries no data were found in the Swedish SPIN data. In total, 27 substances or substance groups have been identified which belong to these entries and which are not found in SPIN. They are listed in the following Appendix Table 9. This list contains five substance groups, marked in yellow in the table (e.g. Aluminosilicate Refractory Ceramic Fibres). For these groups, no CAS numbers have been identified and could therefore not be searched for on SPIN. For the remaining 22 substances, CAS numbers have been found. The 22 substances that were not found on SPIN are amongst the products that do not need to be reported to SPIN according to the Swedish tariff codes. Most are pesticides or heavy metal compounds which by definition are expected from SPIN.

Appendix Table 9: SVHC not found in Swedish SPIN database

	Name	CAS No
1	Aluminosilicate Refractory Ceramic Fibres are fibres covered by index number 650-017-00-8 in Annex VI, part 3, table 3.1 of Regulation (EC) No 1272/2008 of the European Parliament and of the Council of 16 December 2008 on classification, labelling and pa	-

2	Zirconia Aluminosilicate Refractory Ceramic Fibres are fibres covered by index number 650-017-00-8 in Annex VI, part 3, table 3.1 of Regulation (EC) No 1272/2008 of the European Parliament and of the Council of 16 December 2008 on classification, labelli	-
3	4-(1,1,3,3-tetramethylbutyl)phenol, ethoxylated [covering well-defined substances and UVCB substances, polymers and homologues]	-
4	Reaction mass of 2-ethylhexyl 10-ethyl-4,4-dioctyl-7-oxo-8-oxa-3,5-dithia-4-stannatetradecanoate and 2-ethylhexyl 10-ethyl-4-[[2-[(2-ethylhexyl)oxy]-2-oxoethyl]thio]-4-octyl-7-oxo-8-oxa-3,5-dithia-4-stannatetradecanoate (reaction mass of DOTE and MOTE)	-
5	5-sec-butyl-2-(2,4-dimethylcyclohex-3-en-1-yl)-5-methyl-1,3-dioxane [1], 5-sec-butyl-2-(4,6-dimethylcyclohex-3-en-1-yl)-5-methyl-1,3-dioxane [2] covering any of the individual stereoisomers of [1] and [2] or any combination thereof	-
6	Lead titanium trioxide	12060-00-3
7	Triethyl arsenate	15606-95-8
8	Henicosafluoroundecanoic acid	2058-94-8
9	Tricosafluorododecanoic acid	307-55-1
10	Nonadecafluorodecanoic acid (PFDA) and its sodium and ammonium salts: Nonadecafluorodecanoic acid	335-76-2
11	Trilead diarsenate	3687-31-8
12	Perfluorononan-1-oic-acid and its sodium and ammonium salts: Perfluorononan-1-oic-acid	375-95-1
13	Heptacosafluorotetradecanoic acid	376-06-7
14	Acetic acid, lead salt, basic	51404-69-4
15	4,4'-bis(dimethylamino)-4''-(methylamino)trityl alcohol [with ≥ 0.1% of Michler's ketone (EC No. 202-027-5) or Michler's base (EC No. 202-959-2)]	561-41-1
16	Lead dipicrate	6477-64-1
17	Silicic acid (H2Si2O5), barium salt (1:1), lead-doped [with lead (Pb) content above the applicable generic concentration limit for ’toxicity for reproduction’ Repr. 1A (CLP) or category 1 (DSD); the substance is a member of the group entry of lead compoun	68784-75-8
18	Pentacosafluorotridecanoic acid	72629-94-8
19	N-pentyl-isopentylphthalate	776297-69-9
20	Calcium arsenate	7778-44-1
21	Lead hydrogen arsenate	7784-40-9
22	Cadmium fluoride	7790-79-6
23	N-methylacetamide	79-16-3
24	Dinoseb (6-sec-butyl-2,4-dinitrophenol)	88-85-7
25	Anthracene oil, anthracene paste	90640-81-6

26	Fatty acids, C16-18, lead salts	91031-62-8
27	Anthracene oil, anthracene paste, anthracene fraction	91995-15-2

To analyze if the substances that are not found on SPIN are of mayor quantitative relevance/ high volume substances, the list of non-listed SVHC was compared to the ECHA registration list. This list gives cumulative volumes (tonnage ranges of produced or imported chemicals) for the entire EU per year.

Form the 22 SVHC not found in SPIN, 10 are not yet registered under REACH. This means that they are used in tonnages below < 10 t/a in the EU (for these substances, registration had already been required). For the substances with tonnages below 10 tonnes registration deadline is by May 2018. Given a total volume of around 200.000 tonnes (in 2014) for all SVHC in Sweden listed in SPIN, these not registered substances (with a maximum tonnage of 100 tonnes) has no significant influence on the total tonnage. 12 substances, which have not been found in SPIN have been registered under REACH and are found in Appendix Table 10.

Appendix Table 10: Quantitative relevance of SVHC absent on SPIN

	Name	CAS	T/a
1	Acetic acid, lead salt, basic	51404-69-4	0 - 10 tonnes per annum
2	Lead titanium trioxide	12060-00-3	10 - 100 tonnes per annum
3	4,4'-bis(dimethylamino)-4''-(methylamino)trityl alcohol [with ≥ 0.1% of Michler's ketone (EC No. 202-027-5) or Michler's base (EC No. 202-959-2)]	561-41-1	10 - 100 tonnes per annum
4	Silicic acid (H2Si2O5), barium salt (1:1), lead-doped [with lead (Pb) content above the applicable generic concentration limit for 'toxicity for reproduction' Repr. 1A (CLP) or category 1 (DSD); the substance is a member of the group entry of lead compoun	68784-75-8	10 - 100 tonnes per annum
5	Dinoseb (6-sec-butyl-2,4-dinitrophenol)	88-85-7	100 - 1000 tonnes per annum
6	Fatty acids, C16-18, lead salts	91031-62-8	10000 - 100000 tonnes per annum
7	Trilead diarsenate	3687-31-8	Intermediate Use Only
8	Acetic acid, lead salt, basic	51404-69-4	Intermediate Use Only
9	Calcium arsenate	7778-44-1	Intermediate Use Only

10	Anthracene oil, anthracene paste	90640-81-6	Intermediate Use Only
11	Triethyl arsenate	15606-95-8	Tonnage Data Confidential
12	N-methylacetamide	79-16-3	Tonnage Data Confidential

For two substances, tonnage information are confidential. This is an indication that the volume of these substances is low. Four substances registered for intermediate use. This means the substance is reacting further to another product. Under REACH, registration as intermediate requires use under strictly controlled condition. Therefore, emissions surface waters can be expected to be very low. Therefore, these substances can be excluded from further analysis related to water relevant substances. From the remaining six substances, one has been registered for very small volumes (0 – 10 tons per annum) and three for small volumes (10 – 100 tons per annum).Only one of the registered substances, which are not found in SPIN is registered for a medium volume (100 – 1000 tons), and only one for a high volume (10.000 – 100.000 t/year). The high volume chemical is lead salts of Acetic acids. As mentioned above, compounds with heavy metals do not have to be notified in SPIN.

The total volume of the eight substances with tonnage information mounts up to a maximum values of 101.310 tons/year. Given a total tonnage of SVHC in Sweden of around 200.000 t in 2014, only the high volume chemical could significantly influence the total tonnage. The tonnage band in the registration dossier refers to all producers or importers in Europe. Country-specific data are not available. In 2004 Sweden had around 2% share of the chemical industry in the EU. This analysis shows that it is reasonable to assume that the SVHC not listed in SPIN are only of minor significant for the total volume of SVHC used in Sweden.

8.2.2 Influence of Confidential data

With regard to confidentiality, the SPIN data available for the candidate substances for all years (2000- 2015) and countries (Denmark, Finland, Sweden and Norway) is 6234 entries. Of these 3908 (63%) are non- confidential, while 2326 (***37%***) are confidential, meaning that the substance is used by less than 3 companies and/or in less than 4 products. The exact evaluation confidentiality for each substance is evaluated manually according to the 3 companies/ 4 product guidelines by KEMI staff. This raises the question in how many preparations an average substance is found, if 3 companies is the cut-off limit. 1-Methyl-2-pyrrolidone (NMP) for example was used in 882 products in 2011 and other substances are far above the 4 preparation cut- off limit.

Nonetheless, it is possible that one company holds the monopole on one product requiring large volumes of a specific SVHC and therefore relevant data is not accounted for. Yet KEMI stated that it is a rare scenario in Sweden (Eriksson 2017) but according to the Norwegian officials is a common scenario (Wigaard 2017). In order to check the potential influence of confidential data on the total volume of SVHC in Sweden, a more detailed analysis has been made for the SPIN data provided from 2004 to 2014. The following table shows the total use data for the 144 SVHC entries (groups or single substances) covered by SPIN, for the time period between 2004 and 2014. They are shown with decreasing volumes (in 2014). Data gaps due to confidentiality are indicated in red. The first substance which shows a significantly part of confidential data is substance Nr. 72. The 71 substances before have no significant data gaps due to confidentiality. These 71 substances together have a cumulative use volume of 19738 tons in 2014. Substance Ne. 71 has a total use volume of 0 tons in 2014.

Appendix Table 11: Total Use data for SVHC from 2000 to 2014 with confidential data

	Name	Cas	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014
1	Methyloxirane (Propylene oxide)	75-56-9	4088	3961	2710	3723	2654	5417	4506	4306	4125	1734	3538	3230	3270	2184	4124
2	Disodium tetraborate, anhydrous	12179-04-3	0	0	0	0	318	260	2	794	863	710	930	494	202	207	3920
3	1,2-dichloroethane	107-06-2	0	3	0	0	0	11	4548	2576	4307	3834	5912	1453	2568	2578	3439
4	linear	84852-15-3	1	0	1	3	4	9	133	12	6	6	5	1	3	1	1758
5	2-Methoxyethanol	109-86-4	6	51	0	0	0	1	0	0	0	0	0	0	0	0	1378
6	Bis (2-ethylhexyl)phthalate (DEHP)	117-81-7	13226	4477	2746	1658	1620	1611	2102	1957	1489	1105	1343	449	977	639	1096
7	Boric acid	10043-35-3	1447	949	876	1086	1007	968	1227	1189	1216	674	794	864	781	746	867
8	Disodium tetraborate, anhydrous	1303-96-4	6071	4886	4147	5032	5752	5321	5771	6019	5301	4233	4579	4567	4124	4181	591
9	Lead monoxide (lead oxide)	1317-36-8	1546	1124	731	663	413	322	238	267	215	172	231	253	255	235	311
10	anhydride	85-42-7	63	78	53	64	71	43	47	29	22	12	12	12	251	175	282
11	Diboron trioxide	1303-86-2	394	219	210	241	286	288	267	334	322	209	295	301	262	254	278
12	Strontium chromate	7789-06-2	563	596	521	497	655	580	853	818	771	405	570	456	549	303	232
13	Chromium trioxide	1333-82-0	743	524	715	514	524	478	475	559	525	350	373	308	344	463	213
14	azodi(formamide)) (ADCA)	123-77-3	193	231	177	168	183	104	99	283	334	140	143	200	175	261	199
15	N,N-dimethylformamide	68-12-2	781	554	431	239	290	230	176	138	180	120	125	291	143	173	197
16	1-Methyl-2-pyrrolidone (NMP)	872-50-4	1070	1019	1013	1013	973	642	546	555	539	353	314	287	248	183	196
17	Pitch, coal tar, high temp.	65996-93-2	20785	16706	16413	17940	23775	19146	13643	10409	5721	100	[redacted]	[redacted]	[redacted]	[redacted]	159
18	anhydride	19438-60-9	[redacted]	3	[redacted]	4	53	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	130	90	128
19	Acrylamide	79-06-1	723	636	421	451	389	446	284	254	213	174	185	54	46	51	84
20	triazinane-2,4,6-trione (TGIC)	2451-62-9	157	86	49	63	49	53	57	79	72	39	45	51	48	45	68
21	oxo-8-oxa-3,5-dithia-4-	15571-58-1	112	109	47	53	19	18	7	3	4	[redacted]	[redacted]	27	23	34	36
22	methylenedianiline	101-14-4	[redacted]	[redacted]	[redacted]	[redacted]	29	42	54	34	35	19	29	29	40	21	31
23	Sodium dichromate	10588-01-9	67	78	68	44	69	72	28	37	23	13	21	12	12	11	23
24	Trichloroethylene	79-01-6	504	381	347	270	214	175	170	152	84	83	81	51	53	31	22
25	Bisphenol A; BPA	80-05-7	91	49	42	43	66	129	130	138	139	55	29	34	18	47	21
26	Diisobutyl phthalate	84-69-5	94	79	114	85	66	49	62	55	42	41	30	24	23	10	14
27	(MDA)	101-77-9	2	3	3	2	2	1	4	4	3	1	0	2	0	6	13
28	Hydrazine	302-01-2	2	4	1	15	10	10	10	17	8	14	9	10	11	18	13
29	Dibutyl phthalate (DBP)	84-74-2	236	142	174	210	235	210	189	115	69	37	37	35	26	21	8
30	Benzyl butyl phthalate (BBP)	85-68-7	609	271	693	665	820	727	721	412	20	9	6	9	9	7	7
31	Disodium tetraborate, anhydrous	1330-43-4	40	41	53	58	48	26	26	24	19	12	9	8	3	5	6
32	anhydride	25550-51-0	5	5	11	10	12	6	5	4	2	1	3	3	8	10	4
33	linear, ethoxylated	37205-87-1	36	18	20	10	17	6	5	4	4	3	2	2	2	3	3
34	(Benzo[a]pyrene)	50-32-8	1083	965	985	1519	1564	1064	907	972	901	491	3143	2	2	2	2
35	Pigment Yellow 34)	1344-37-2	68	52	32	35	23	18	21	17	38	4	2	0	5	3	2
36	p-(1,1-dimethylpropyl)phenol	80-46-6	1	1	1	1	1	1	1	1	2	0	1	1	1	0	2

37	Tetraethyllead	78-00-2	5	4	4	3	4	2	2	3	2	6	2	2	2	2	2
38	dihexyl ester, branched and linear	68515-50-4			1		3	1	3	2	2	2	1	1	1	0	1
39	deterpentylphenol (UV-328)	25973-55-1	6	5	7	4	4	9	6	6	2	3	4	1	2	1	1
40	chlorobenzotriazol-2-yl)phenol (UV-	3864-99-1	2	2	3	3	3	6	6	6	4	2	1	10	1	0	1
41	linear, ethoxylated	25154-52-3	35	12	46	26	10	10	10	14	8	7	8	2	5	27	1
42	Chain Chlorinated Paraffins)	85535-84-8	21	36	18	14	12	12	10	10	10	9	6	4	4	2	1
43	Cobalt(II) dinitrate	10141-05-6	4	3	4	3	1	1	3	2	3	1	1	1	2	1	1
44	Lead dinitrate	10099-74-8	1	1	1	0	0	0	0	0	0	0	0	1	0	1	1
45	N,N-dimethylacetamide	127-19-5	12	257	5	5	0	0	4	2	1	0	0	1	4	5	1
46	Sodium peroxometaborate	7632-04-4	300	284	253	150	242	212	205	93	3	2	1	1	1	0	1
47	benzhydrylidene]cyclohexa-2,5-	548-62-9	5	6	6	2	1	2	0	0	0	0	0	4	1	0	0
48	11-branched and linear alkyl	68515-42-4	573	777	623	457	453	268	265	289	135	6	49	0	0	0	0
49	2-Ethoxyethanol	110-80-5	16	20	7	3	6	1	1	2	0	0	0	0	0	0	0
50	2-Ethoxyethyl acetate	111-15-9	21	16	10	2	1	4	1	1	0	0	0	0	0	0	0
51	4-(1,1,3,3-tetramethylbutyl)phenol	140-66-9	6	4	11	7	4	6	7	3	4	0	0	0	0	0	0
52	linear	104-40-5	29	16	4	12	7	3	3	4	0	0	0	0	0	0	0
53	linear, ethoxylated	0	0	33	31	0	0	0	0	0	0	0	0	0	0	0	0
54	linear, ethoxylated	26027-38-3	33	5	4	5	1	2	2	0	0	1	0	0	0	0	0
55	Bis(2-methoxyethyl) ether	111-96-6	0	1	0	0	0	0	0	0	0	0	0			0	0
56	Boric acid, crude natural	11113-50-1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
57	Cadmium	7440-43-9	1	1	0	0	0	0	0	0	0	70	38	15	0	0	0
58	Cobalt(II) sulphate	10124-43-3	0	0	0	0	0	1	1	0	0	2	1	3	0	0	0
59	Diarsenic trioxide	1327-53-3	10	7	1	0	0									0	0
60	Formamide	75-12-7					0	0	0	1	0	0	0	0	0	0	0
61	imidazoline-2-thiol)	96-45-7	3	1	1			0	0	0	0	0	0	10	0	0	0
62	sulphate red (C.I. Pigment Red 104)	12656-85-8	19	29	19	30	16	20	21	18	20	5	1	1	1	1	0
63	Methoxyacetic acid	625-45-6	1	1	1	1	1	1	0	1	1	1	0	0	0	0	0
64	o-aminoazotoluene	97-56-3													0	0	0
65	Orange lead (lead tetroxide)	1314-41-6	28	24	18	0	0			2	0	0	1	1	1	0	0
66	o-Toluidine	95-53-4	10	0	0	0	0	0	0	0	0	0	0	0	0	0	0
67	Pentazinc chromate octahydroxide	49663-84-5	2	2	1	0	0	0	0	0	0	0	0	0	0	0	0
68	Phenolphthalein	77-09-8	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
69	Potassium dichromate	7778-50-9	28	14	12	7	5	8	6	5	7	2	3	1	0	0	0
70	hydroxyoctaoxodizincatedichromat	11103-86-9	1	0	1	0			0	0	0	0	0	0	1	0	0
71	Sodium dichromate	7789-12-0	26	31	35	43	24	14	21	15	15	5	4	1	0	0	0
72	(dimethylamino)phenyl]methylene	2580-56-5	2	0	0	0				0							

73	[Phthalato(2-)]dioxotrilead	69011-06-9															
74	1,2,3-Trichloropropane	96-18-4															
75	10-alkyl esters	68515-51-5	29	16	25	15	9										
76	8-branched alkyl esters, C7-rich	71888-89-6	2	4	10	9	8	19	19	23	13	9					
77	dipentylester, branched and linear	84777-06-0															
78	(TEGDME, triglyme)	112-49-2				1	0	2	4	1	1	1	1	0			
79	1,2-Diethoxyethane	629-14-1															
80	epoxypropyl]-1,3,5-triazine-2,4,6-	59653-74-6															
81	bromide)	106-94-5															
82	butyl)-6-(sec-butyl)phenol (UV-350)	36437-37-3															
83	2,4-Dinitrotoluene	121-14-2	147	69	70	79	74	48	67	84	64	59	33	19	6		
84	butylphenol (UV-320)	3846-71-7															
85	bis(dimethylamino)benzophenone	90-94-8															
86	linear	1987-50-4													0		
87	linear	72624-02-3															
88	linear, ethoxylated	11066-49-2															
89	linear, ethoxylated	7311-27-5															
90	linear, ethoxylated	90481-04-2															
91	(Musk xylene)	81-15-2				0				0	0	0	0	0	0	0	
92	cresidine)	120-71-8															
93	trioxide and their oligomers:	13530-68-2															
94	trioxide and their oligomers:	7738-94-5									19	6	22	18	13	14	
95	Ammonium dichromate	7789-09-5	0	0													
96	pentadecafluorooctanoate (APFO)	3825-26-1															
97	Anthracene	120-12-7															
98	Anthracene oil	90640-80-5															

99	Anthracene oil, anthracene-low	90640-82-7															
100	Biphenyl-4-ylamine	92-67-1															
101	(decabromodiphenyl ether)	1163-19-5	89	15	14	5	4	3	4	1	1				0		
102	Bis(tributyltin)oxide (TBTO)	56-35-9	0	1	0	0	0										
103	Cadmium oxide	1306-19-0	0	0	0	0	89	75	67	53		36	97	195	153		
104	Cadmium sulphate	10124-36-4	0	0	0	0	0	0		0					0		
105	Cadmium sulphide	1306-23-6															
106	Cobalt dichloride	7646-79-9	0	0	0	0						0	0	0			
107	Cobalt(II) carbonate	513-79-1															
108	Cobalt(II) diacetate	71-48-7	0	0	0	0											
109	Diarsenic pentaoxide	1303-28-2	284	233													
110	Dibutyltin dichloride (DBTC)	683-18-1															
111	Dichromium tris(chromate)	24613-89-6															
112	Diethyl sulphate	64-67-5															
113	Dihexyl phthalate	84-75-3	2	2	1												
114	Diisopentylphthalate	605-50-5															
115	Dimethyl sulphate	77-78-1															
116	Dioxobis(stearato)trilead	12578-12-0	20														
117	Dipentyl phthalate (DPP)	131-18-0									11	15	11	5			
118	products with aniline	25214-70-4	0	0	1												
119	Furan	110-00-9															
120	and all major diastereoisomers	3194-55-6	5	59	31	3	4										
121	and all major diastereoisomers	25637-99-4															
122	anhydride	57110-29-9															
123	Hydrazine	7803-57-8	27	1	14	22	13	1									
124	Lead bis(tetrafluoroborate)	13814-96-5	0			0	0	0									
125	Lead chromate	7758-97-6	4	4	4	5	4	3	3	2	2						
126	Lead di(acetate)	301-04-2	0	0	0	0	0										
127	Lead diazide, Lead azide	13424-46-9															
128	Lead oxide sulfate	12036-76-9															
129	Lead styphnate	15245-44-0															
130	Lead(II) bis(methanesulfonate)	17570-76-2	0	0		0											
131	methylenedianiline (Michler's	101-61-1															

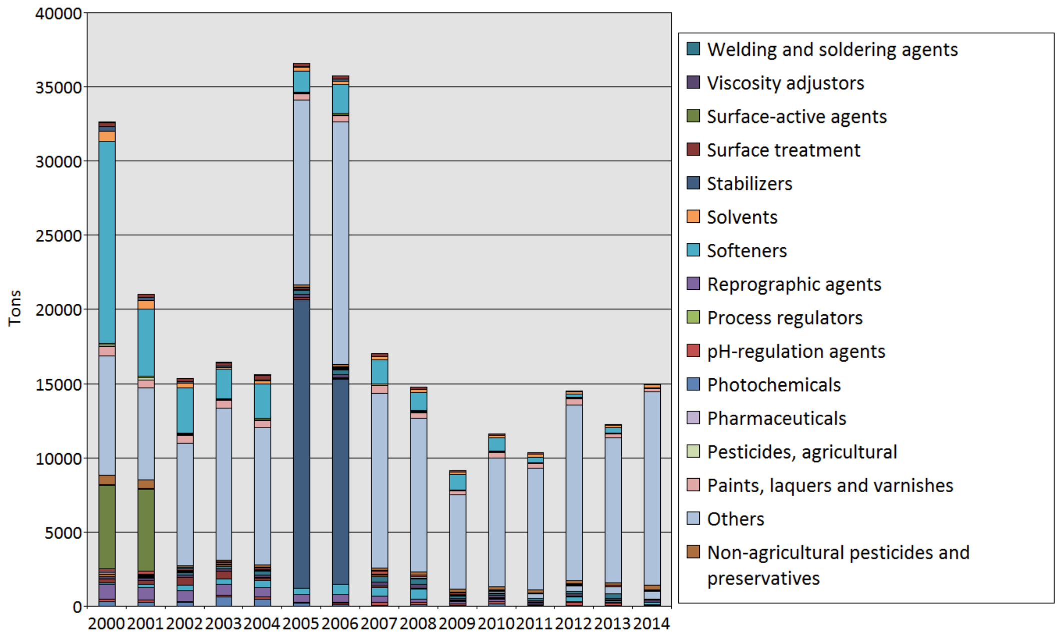
132	Nitrobenzene	98-95-3															
133	(PFOA)	335-67-1															
134	Pentalead tetraoxide sulphate	12065-90-6	4		6	4	1										
135	Potassium chromate	7789-00-6															
136	Silicic acid, lead salt	11120-22-2															
137	Sodium chromate	7775-11-3	6	5	7	7	8	6	7	6							
138	Sodium perborate	11138-47-9															
139	Solvent Blue 4	6786-83-0	5	3	0	0	0	0	0	1	0	1	1				
140	Sulfurous acid, lead salt, dibasic	62229-08-7															
141	hydrate	12267-73-1															
142	Tetralead trioxide sulphate	12202-17-4	154	40	14	12	8	8	6	1	0	0					
143	Trilead bis(carbonate) dihydroxide	1319-46-6	0	4	4	4	0	0	1								
144	Trilead dioxide phosphonate	12141-20-7	59	34	27	24	19	5	0	0							
145	Tris(2-chloroethyl)phosphate	115-96-8	6	9	17	9	27	38	48	47		12	7	0			
146	Trixylyl phosphate	25155-23-1	2	2	0	3	2	4	2	2	0	0	3	2	1	1	

The remaining (146-71=) 75 substances (from substance nr.72 – 146) have severe data gaps. However,looking at the time trends individually, it becomes clear that most of the substances are probably used by few companies in amounts so small, that the data becomes confidential. As seen with for example substance no. 146, the volume is known and decreases over the years. It is reasonable to assume that in 2013 one of the few companies using the substance stopped using it completely, and this is why the data became confidential. Nonetheless there is the possibility of disguising big tonnages under confidential data. As seen in substance no. 109, diarsenic pentoxide, just two data points are available in year 200 and 2001. It could be that after that one the data came into the confidentiality zone, but one big company is still using large amounts. There is always the risk that one company holds the monopole in substance use and therefore does not have to report it. However, looking at the data individually most of them are used so small amounts before and after the confidential years that the data there is quantitatively not of major relevance. There are only three substances that are in the range of 100t and more. Undeniably, the substances that have just confidential data from the start are impossible to estimate. For a first impression, the quantity index on SPIN in the ExposureToolbox can be used (see chapter 8.4). Yet the quantity index seems not to match the quantities reported in TotalUse for the non-confidential substances, therefore the reliability is unclear. In this case KEMI can be contacted to check the quantity and report it in tonnage bands.

8.3 Utility of the SPIN use categories (Use category UC62)

SPIN contains not only total use data, but also data on the use pattern of substances (see chapter1.4). For this purposes, 62 use categories have been introduced in the so-called "UC 62" system. Examples for such use categories are pesticides, welding and soldering agents, softeners etc. However, the UC62 is not exhaustive. In this thesis, the UC 62 information has been analyzed in order to estimate the part of the total use which is of relevance for surface water. The premise was that use categories as "fertilizers" are more likely to contribute to contamination of surface waters than other categories. For this purpose, the use information has been retrieved from SPIN for all SVHC reported in SPIN. The results are shown in Appendix Figure 8: Dominance of category "Others" in UC62 in Sweden.

The figure shows that for the largest part of the tonnage recorded in SPIN the use category "others" is selected. This makes the Use Category data very uncertain and unreliable. Therefore, no further analysis of emission of substances according to their use category was undertaken. This means, no specific data on the use are available. Therefore, the information on uses from the Use category database UC 62 is insufficient clarify the main uses of the SVHC recorded in SPIN. Moreover, it is seen that the amounts regisztered in the UC62 do not add up to the amounts registered in totalUse, but are slightly less. It is probable that some companies do not know what to fill in for UC62, when they registere and some tonnages are lost that way.



Appendix Figure 8: Dominance of category "Others" in UC62 in Sweden

8.4 Use of the ExposureToolbox for prediction of emerging pollutants

It has been assessed whether the ExposureTool in SPIN can be used to predict future emerging pollutants. A typical output of the Exposure toolbox for a specific substance is shown in figure x below.

EXPOSURE

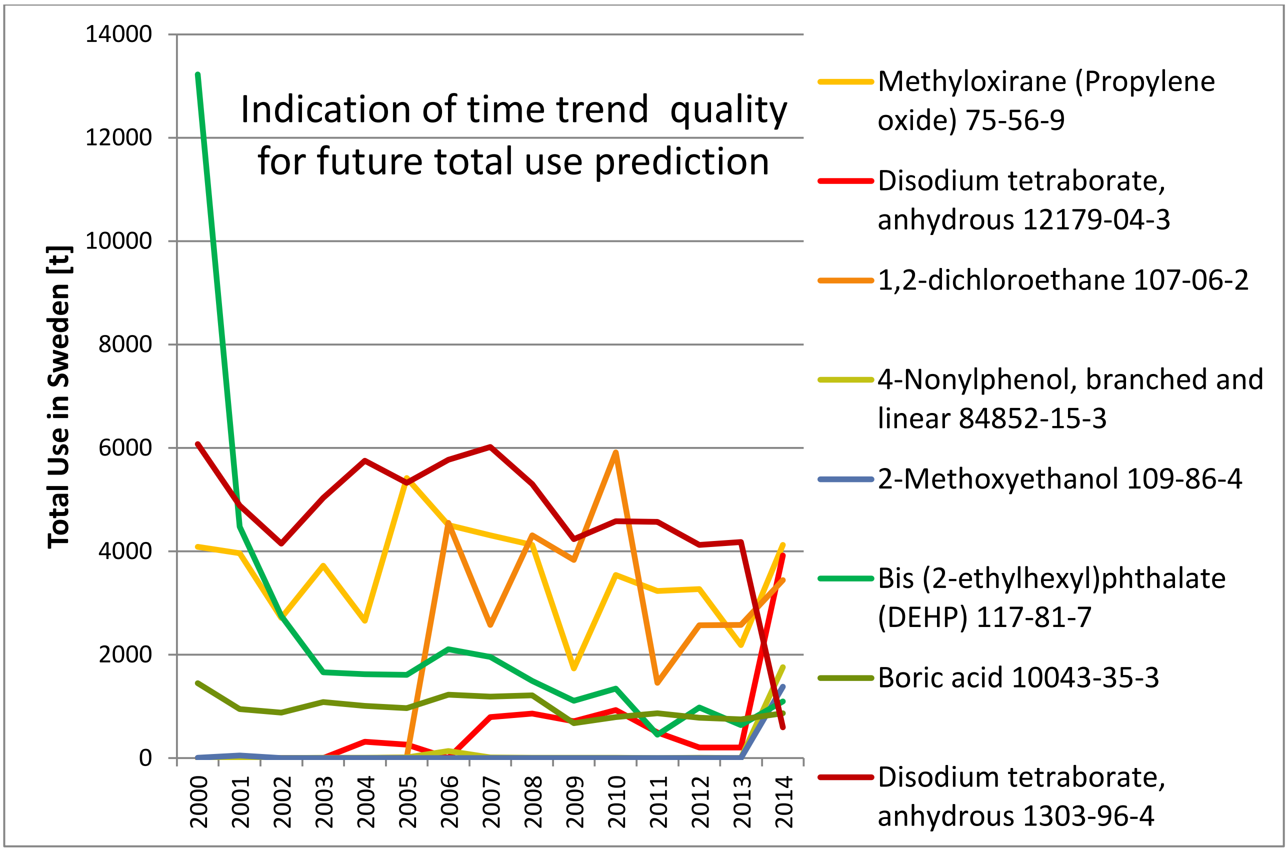
Country	Latest Year	Quantity	Use Index (UI)						Range of Use (RoU)	Article Index (AI)
			Surface water	Air	Soil	Waste water	Consumer	Occu-pational		
DK	2014	3	3	4	4	2	4	5	2	3
NO	2014		4	3	4	3	4	5	3	3
SE	2014	2	3	2	3	4	3	5	3	3

Appendix Figure 9: Typical ExposureToolbox output

The Exposure toolbox consists of a Use Index (UI) providing information on possible risks of certain substances in mixtures to the environment (surface water, air, soil, waste water) and to consumers and workers. The UI was developed to satisfy the increasing demand of exposure information needed under the REACH legislation. The numbers are derived from the Use category (in total 62, therefore UC62) such as “cleaning agents” or “absorbents and adsorbents” and the Industrial category such as “accommodation” or “services to buildings and landscape activities”. These categories give insights of how the products are handled and used and therefore form the bases for the UI algorithm. SPIN admits that there could be biases regarding the UI, as the members of the committee developing the UI did not have the same level of expertise on all industrial categories and use categories. However, SPIN advocates that the UI can be used as “a check tool for ECHA and national competent authorities when assessing a request for e.g. waiving of tests” and as “a screening tool for priorization” (SPIN 2017). As stated above the UI represents direct exposure to “soil”, “air”, “surface water”, “sewage treatment plant” (STP) “consumer” and “occupational”. The category “surface water” indicates the direct emission to surface water, such as chemical products used in or in the proximity of lakes/seas, or used in industrial plants that are likely to have separate discharge systems (i.e. not connected to municipal sewage treatment plants) (SPIN 2017). The exposure toolbox does not take physio-chemical properties such as “vapour- pressure” or “water solubility” or most importantly “biodegradability” into consideration. Therefore, the UI risk assessment is limited to the near surroundings, and cannot be used for an EU project but under reserve for local emerging point pollutants. However, also here precaution is advised as inconsistencies of the TotalUse data and the tonnage bands indicated in the Exposure tool were found, therefore raising doubt about the reliability of the ExposureTool itself. Therefore, it has not been further used in this thesis.

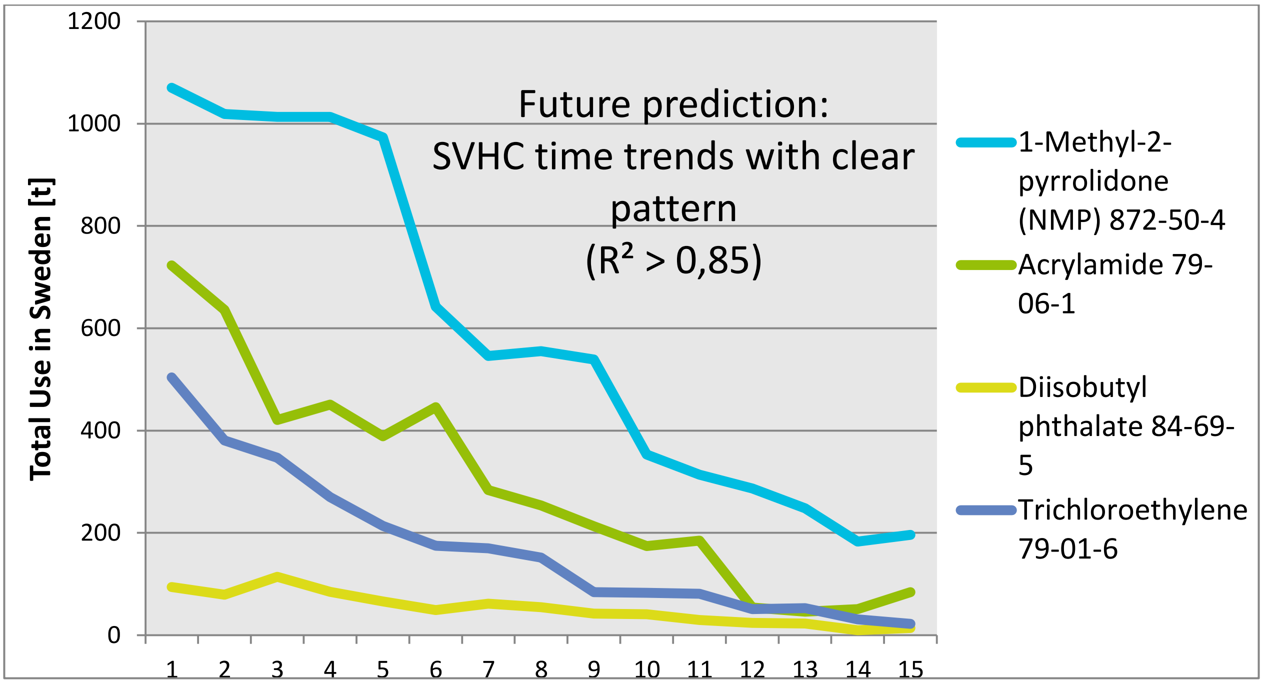
8.5 SPIN total use data for future time trend prediction

As seen through this thesis, SPIN offers data qualities from end-to end time trends to all confidential data. As described in chapter 8.2.2 Influence of Confidential data it is impossible to exclude the possibility that confidential data disguise big tonnages. However, in doubt KEMI can be contacted. It was seen that from the 144 SVHC entries covered in SPIN 71 had enough data to see clear time trends. Given complete datasets for individual substances theoretically future predictions could be derived. However, given the big variability and fluctuation in numerous substance time trends, it is impossible to make reliable predictions for future total use development for most of the substances. As shown in Appendix Figure 10, most of the time trends are fluctuating strongly. Trends that are not fit to predict future time trends are indicated in red. For some, a trend direction is recognizable; these are marked in orange or yellow. Generally for all of the end-to end time trends predictions could be made. However,it is questionable if these predictions can be accurate for most of the data. Some substances show clear trends, such as DEHP with an exponential decrease from 2000 onwards. Based on steady time patterns predictions are more likely to occur.



Appendix Figure 10: Indication of time trend quality for future total use prediction

In order to select the substances with time trends that could be fit for future prediction, the coefficient of determination (R^2) was calculated for some SVHC and as shown in Appendix Figure 11.



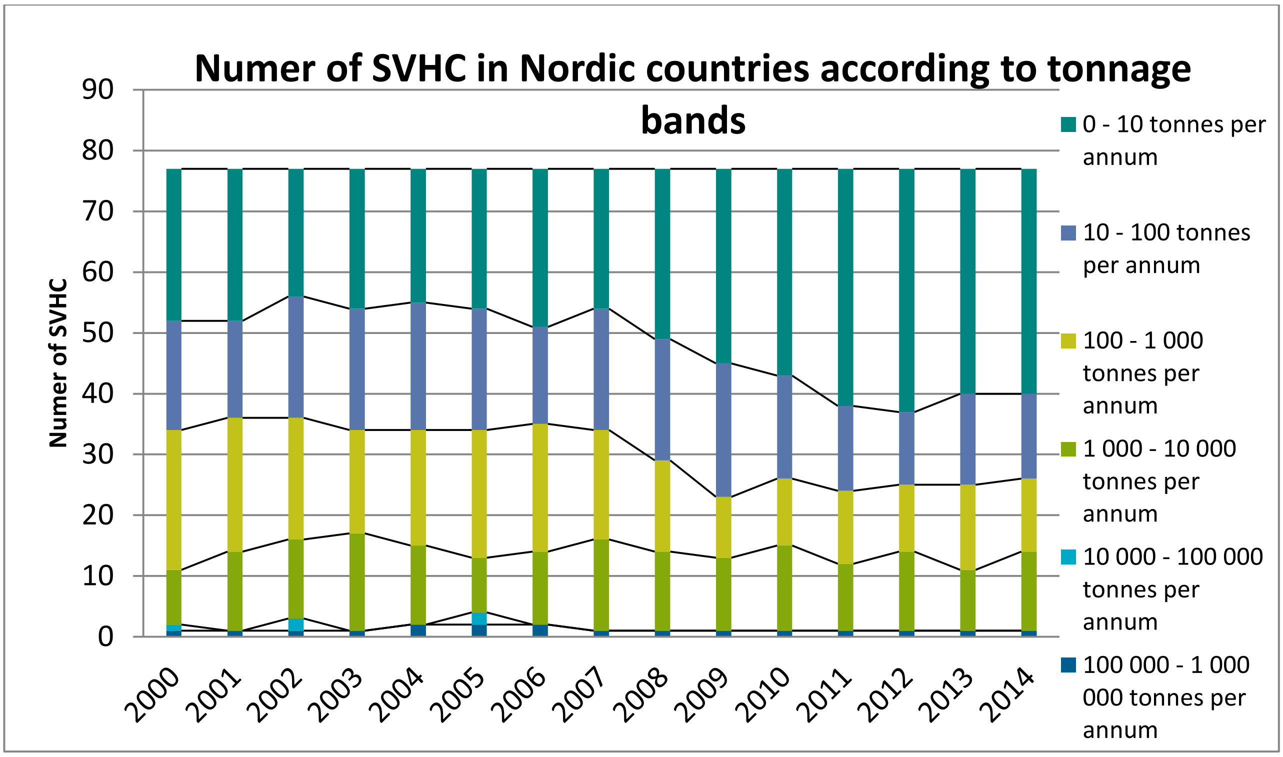
Appendix Figure 11: SVHC time trends with clear pattern

Nonetheless accurate prediction of future total use might be difficult, depending on regulative action or changes in the industry that can influence time developments strongly as shown in this thesis. Generally, predicting future use of substances is statistically possible but in the realm of chemical use data without indications of future regulation and market developments expected to be little reliable.

8.5 SPIN total use data as indicator for regulative impact

Even though future predictions are estimated to be difficult, SPIN is an excellent tool to measure the impact of regulation on the total use of chemicals. It was shown that regulative impact strongly decreases the amount of individual substances used. Therefore, it is recommended to use SPIN further as indicator for the success of chemical regulations such as REACH.

For all the end-to end time trends of all SVHC in the four Nordic countries it was found, that the amount of SVHC registered in high tonnage bands decreases over time. Simultaneously, the small tonnage bands (0-10t) increase in amount of SVHC over time. Concluding, the amount of SVHC that are high use volume chemicals decreases, while small use SVHC increases, indicating the success of the regulation through the REACH candidate list.



Appendix Figure 12: Number of SVHC in Nordic countries according to tonnage bands

Appendix Figure 12: Number of SVHC in Nordic countries according to tonnage band shows that substances that were used in big tonnages decrease and almost disappear. Contrastingly, SVHC used in small amounts increase over time. In 2000 there were around 20 substances used in small tonnages while in 2014 their share increased to almost 40 substances of very high concern that were used in small quantities.

These assessments show the suitability of SPIN as valuable data base for time trends for industrial chemicals. These time trends can facilitate the screening for EPs from industrial chemicals and can be used to assess chemical regulations.

Ehrenwörtliche Erklärung

Hiermit erkläre ich, dass die Arbeit selbständig und nur unter Verwendung der angegebenen Hilfsmittel angefertigt wurde.

Freiburg, den

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Kathrin Sackmann