Short and medium chained chlorinated paraffins in buildings and constructions in the EU

PREFACE

I would like to thank my supervisors Daniel Müller and Gunnar Grini, for thoughtful advices and engagement in my work.

I would also like to thank Magne Gilberg, who has supported me throughout the entire process and helped me hold my spirit up.

ABSTRACT

A concern has arised regarding the potentially large amounts of short chained chlorinated paraffins (SCCPs) and medium chained chlorinated paraffins (MCCPs) accumulated in buildings and constructions and the future contaminated waste from the sector. The historic use of SCCPs and MCCPs in EU has been estimated and a dynamic material flow analysis has been applied to determine the stocks and waste flows. The waste flow of SCCPs is estimated to have been at it's largest in 2002 with approximately 1300 tons per year. Today (year 2012) it has decreased to almost 1000 tons per year. The waste flow of MCCPs was at it's maximum in year 2006 with around 15 000 tons and has decreased to around 14 000 tons today. The use of SCCPs has decreased evenly since the 1980s and the waste flow from buildings and construction is assumed to cease within the 2040s. The use of MCCPs has also decreased, but is still large. If the use of MCCPs is stopped today, it is estimated that it still will take more than 30 years before the building stock and waste flow is free from the substance. The data availability has been scarce and many assumptions are made. Improved knowledge on which products that contains SCCPs and MCCPs and where they are used is needed. This will improve the model and ensure that waste containing SCCPs and MCCPs is securely taken care of.

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1 INTRODUCTION

Short and medium chained chlorinated paraffins (SCCPs and MCCPs) are part of the larger group of organic pollutants called chlorinated paraffins (CPs). They are used as high-pressure additives in metal working fluids and plasticizers, flame-retardants and chemical- and water resistant agents in plastics, rubber, paints, adhesives, sealants, textiles, leather and carbonless copy paper.

SCCPs are classified as persistent, toxic and bioaccumulative (PTB) and are listed on the European Candidate list of substances of priority concern. In the past the major concern was their use in metal working fluids and fat liquoring of leather. In the EU there has been restrictions on the use of SCCPs for these applications since 2004 and the concern has shifted to comprise the possibly big amount of CPs in waste from buildings and constructions. There has been less attention on MCCPs, even though they have been found to have many similar properties as SCCPs. MCCPs are not yet restricted in the EU and some of the SCCP usage is believed to have been substituted with MCCPs.

The intention behind this thesis has been to answer the main question of concern – how much SCCPs and MCCPs can be found in builing and costruction waste today? And how big amounts can be expected in the future? These are fundamental questions that needs to be answered to be able to assess the scope of the problem and to ensure that necessary measures are in place.

A realistic estimate of future SCCPs and MCCPs waste amounts from buildings and constructions, must take into account the historic consumption of the substances. Buildings and constructions have long lifetimes and conciderable amounts of SCCPs and MCCPs may therfore be accumulated in the stock, before it exits use as waste. A study by Håvard Bergsdal on polychlorinated biphenyls (PCB) in the Norwegian building stock, showed that the accumulated amount of PCBs in the stock would continue to be a problem for demolition waste the entire century to come, althoug the use of PCB was faced out in the 1970s (Bergsdal, 2009). Dynamic material flow analysis (MFA) was used for the analysis of PCBs in the Norwegian building stock.

A literature reviewe on former studies applying MFA, revealed four studies concerning CPs. This included one study from 2011 on SCCPs in the EU (Umweltbundesamt et al. 2011). The study was based on SCCPs sales data from 1994 to 2009 and concerned all the main product groups containing SCCPs. The study did, however, mainly quantify flows and not accumulated amounts

(stocks) in society. The waste flows are therefore likely to be underestimated. The quantifications where in general very simplified and the system analyzed was not clearly defined. The other studies concerned Japan, Stockholm and Norway. None of them were dynamic MFAs and none of them concerned MCCPs (although the study for Stockholm concerned CPs in general). The previous MFA studies on CPs does therefore not manage to address what is believed to be the main future problem concerning CPs, namely waste from buildings and constructions with it's accumulated SCCPs and MCCPs.

Although CPs in general are attended with great concern, there is little knowledge on the amounts used and in which products/articles they can be found. The main research questions in this thesis have been to answer how big the accumulated amounts of SCCPs and MCCPs in buildings and constructions in the EU are and how big the amounts of SCCPs and MCCPs exiting use as building and construction waste are. However, several other questions needed to be answered beforehand:

- What has the historic consumption of CPs in the EU been?
- What has the distribution between SCCPs, MCCPs and the last subgroup of CPs – LCCPs – been?
- How big amounts have been used for the production of different products?
- How much of the products containing SCCPs or MCCPs are used in buildings and constructions?

The topic of the thesis was initiated by the Norwegian Building Authority, who initially wished to estimate CP stocks and waste flows from Norwegian buildings and constuctions. Data on use of CPs in Norway have been assessed, but concidered insufficient. Some data on historical amounts of SCCPs and MCCPs used in Norwegian production was available, but the data was very uncertain because of changing legislation on which substances that needed to be registered in the Norwegian product register. The data was also available for less than 20 years of past uses and amounts entering Norway through imported finished articles was not included. It is well known that most of the imports to Norway is from Europe. According to a report prepared for the Norwegian Climate and Pollution Agency, this is also the case for important product groups containing CPs (Cowi, 2010). Since more comprehensive data was available on the European level, it was decided to investigate the use of SCCPs and MCCPs in EU buildings and constructions. This will hopefully also provide information on a likely development of the use of the substances in Norway and give a picture of the scope at which SCCPs and MCCPs might have been imported into Norway. The situation in Norway regarding SCCPs and MCCPs will to some extent be commented on.

2 BACKGROUND

In this chapter background information is given on the substances in question. SCCPs, MCCPs, LCCPs and CPs in general are defined and their physical and hazardous properties are presented. Their use and function in relevant applications are described and relevant agreements and legislation are summarized.

2.1 DEFINITION OF SUBSTANCE

This thesis is mainly concerned with SCCPs and MCCPs, but some information on LCCPs and the group of CPs as a whole has been considered. LCCPs and CPs in general are therefore also defined in the following.

CPs consists of hydrogen and carbon atoms, which are bonded by single bonds (i.e., saturated compounds) without any cycles or loops, where some of the hydrogen atoms have been replaced with chlorine atoms (de Boer et al. 2010). The CPs are usually sub-divided by chain lengths. SCCPs are CPs with a carbon chain-length of 10-13 atoms. MCCPs are CPs with a carbon chain-length of 14-17 atoms, while LCCPs have a carbon chain length of 18-30 atoms.

Within each of the sub-groups, the degrees of chlorination and position of the chlorine atoms can vary. Usually commercially available SCCPs, MCCPs and LCCPs are mixtures of different carbon chain lengths, degrees of chlorination and with differing positions of the chlorine atoms (de Boer et al. 2010). It is also estimated that 1% of commercial MCCP mixtures are SCCPs (European Chemicals Bureau, 2005).

Commercial available SCCPs typically have a chlorine content of 49-70% by weight (European Chemicals Bureau, 2000). For commercially available MCCPs the chlorine content ranges from 40-63%, but is usually between 45-52% (European Chemicals Bureau, 2005). LCCPs can have chlorine contents ranging from 30-72% (Brooke, Crookes, & Merckel, 2009).

SCCPs and MCCPs are colorless or light yellow fluids, while LCCPs can be either a fluid or a solid wax (solid LCCPs have a chlorine content of 70-72%) (Brooke et al., 2009; European Chemicals Bureau, 2000, 2005).

The molecular formula of SCCPs is:
$C_xH_{(2x-y+2)}Cl_y$, where x=10-13 and y=1-13

The structure of $C_{10}H_{17}Cl_5$ is shown in Figure 1.

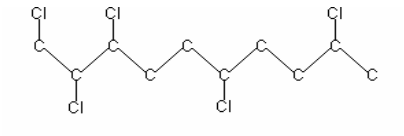

Figure 1: The structure of $C_{10}H_{17}Cl_5$. Source: (POPRC, 2010)

The molecular formula of MCCPs is:
$C_xH_{(2x-y+2)}Cl_y$, where x=14-17 and y=1-17

The structure of $C_{17}H_{29}Cl_7$ is shown in Figure 2

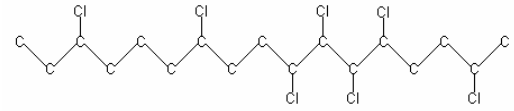

Figure 2: The structure of $C_{17}H_{29}Cl_7$. Source: (European Chemicals Agency, n.d.-a)

Synonyms for CPs are: alkanes, chlorinated; alkanes, chloro-(50-70%); chlorinated alkanes; chloroalkanes; chlorocarbons; polychlorinated alkanes; paraffins-chlorinated; chloroparaffin; chlorinated paraffin waxes and hydrocarbon waxes; paraffin oils and hydrocarbon oils, chloro.

2.2 PHYSICAL PROPERTIES

The physical properties of CPs differ between SCCPs, MCCPs and LCCPs, but also within each of these sub-groups. This is because of the range of possible chain-lengths and degrees of chlorination that can be represented within each class of CPs (as explained above). Despite this, CPs can generally be described as viscose, chemically stable, non-flammable and they have low water solubility's and vapor pressures (de Boer et al. 2010).

Their viscosity, density, thermal stability and resistance against hydrolysis increase with increased chlorine content, and by increased carbon chain length (de Boer et al. 2010). Their vapor pressure decreases with increased carbon chain length and increased degree of chlorination (at least this seems to be the case for both SCCP and MCCP) (European Chemicals Bureau, 2005).

In the following, the vapor pressure and water solubility of SCCP and MCCP is presented. These properties determine the emission ratios that are used in the analysis.

The vapor pressure of SCCPs with a chlorine content in the range of 45-52%, was found to be 0,0035-0,028 Pa at 25°C (K. G. Drouillard, Tomy, Muir, & Friesen, 1998). For SCCPs with a higher degree of chlorination, e.g. 55-61%, the vapor pressure was lower, $1,4*10^{-4}$ – $5,4*10^{-3}$ Pa at 25°C (used for textile applications). For SCCPs with more than 70% chlorine by weight, the vapor pressure has been estimated to be around $1,34*10^{-5}$ Pa at 25°C (used for rubber applications) (K. G. Drouillard et al., 1998; European Chemicals Bureau, 2008a).

There has been done less research on the vapor pressure of MCCPs, but it seems like the vapor pressure decreases with increasing carbon chain-lengths and chlorine content, as for SCCPs (European Chemicals Bureau, 2005). In the European Union Risk Assessment Report on MCCPs, the vapor pressure used in the assessment is $2,7*10^{-4}$ Pa at 20°C, as measured by Campbell and McConnel (1980) of a MCCP with 52% chlorine content.

Experiments have been conducted for determining the water solubility of SCCPs. There was no clear trend in the water solubility depending on chain-lengths and chlorine content (K.G Drouillard et al., 1998; Friesen et al., 1995). A water solubility of 0,15-0,47 mg/l is used in the European Union Risk Assessment Report on SCCPs (2008).

In the European Union Risk Assessment Report on MCCPs a water solubility of 0,027 mg/l is assumed to represent MCCPs reasonably well ((European Chemicals Bureau, 2005) according to (Madeley, Gillings, & Reynolds, 1983)).

Both SCCPs and MCCPs starts to decompose at temperatures above 200°C, and releases hydrogen chlorine in the process (European Chemicals Bureau, 2005), (European Chemicals Bureau, 2000).

2.3 HAZARDOUS PROPERTIES

The degree of toxicity, persistence, bioaccumulation etc. of the CPs differs depending on chain-length and chlorination. The hazardous properties mainly belong to SCCPs and MCCPs.

SCCPs are classified as persistent, toxic and bioaccumulative (PTB) substances (ECHA, 2008) in the EU. A substance is considered very persistent if it has a half-life > 60 days in marine, fresh- or estuarine water or >180 days in marine sediment, freshwater or estuarine sediment or soil. The half-live of a C10-13 with 65% chlorine, was determined to be around 1,630 days in freshwater and 450 days in marine sediment. A substance is considered to be very bioaccumulative if it has a bioconcentration factor of >5,000 l/kg. The highest measured bioconcentration factor value is around 7,816 l/kg (for freshwater fish). SCCPs are also considered toxic based on the fact that the lowest no effect concentration (NOEC) was found to be 0.005 mg/l (for Daphnia magna), when the limit for classifying a substance as toxic is that the NOEC is less than 0.01 mg/l.

SCCPs are also classified as suspected for causing cancer (carcinogen category 3), with the possible risk of irreversible effects (risk phrase R40) (European Chemicals Agency, n.d.-b). They are classified as dangerous for the environment (N), with the risk phrases "very toxic to aquatic organisms" (R50) and "may cause long-term adverse effects in the aquatic environment" (R53).

SCCPs have been found to have similar characteristics as known persistent organic pollutants (POPs) that can be transported over long distances (atmospheric half life's of 0,81-10,5 days and vapor pressure in the range of known POPs that undergo long range atmospheric transport) (POPRC 2010). SCCPs have also been found in biota and sediments in remote arctic areas and it is believed that SCCPs are transported via air or ocean currents.

SCCPs have also been found in breast milk of Inuit women and women in the United Kingdom (POPRC 2010).

Less research has been conducted on MCCPs, but it is considered that MCCPs meet the criteria's for being persistent and toxic (*ANNEX XV RESTRICTION REPORT; MCCPs*, 2008). More research is however needed for determining if MCCPs meet the bioaccumulation criteria. Anyhow, MCCPs are also classified as dangerous for the environment (N), very toxic to aquatic organisms and they may cause long-term effects in the aquatic environment (R50/53). They are also classified with the risk phrases "may cause harm to breastfeed children" (R64)

and “repeated exposure may cause skin dryness or cracking” (R66) (European Chemicals Agency n.d.).

According to Brooke et al. (2009), LCCPs are persistent, but not toxic and because of insufficient data they where not able to conclude whether LCCPs are bioaccumulative (Brooke et al., 2009). Even though LCCPs as a group is not considered as a PTB substances, some of the congeners and uses can be associated with risks.

2.4 APPLICATIONS

The first use of CPs were as a solvent in an antiseptic nasal and throat spray under world war one ((Howard, Santodonato, & Saxena, 1975) according to (Scheer, 1944)). In 1932 CPs where introduced as extreme pressure additives in lubricants/metal working fluids. This was the start of a large-scale commercial production. Nevertheless, the largest expansion came under world war two, due to the increased use of CPs for weather-and flame proofing of tents an camouflage nettings (Roberts, n.d.). Later on, the introduction of MCCPs as plasticizers in flexible PVC has been an important reason for the increased used of CPs.

Altogether, CPs are used or have been used in eight main product groups; metal working fluids, flexible PVC, rubber and other polymers (except PVC), sealants and adhesives, paint and coatings, back-coating of textiles, leather fat liquor and carbonless copy paper. The usage and function of the CPs in each of the product groups relevant for buildings and constructions is described in more detail in the following.

2.4.1 FLEXIBLE PVC

The major application of CPs in Europe at present is as a secondary plasticizer in flexible PVC, often in conjunction with the primary plasticizer phthalate DEHP (Euro Chlor 2011a). Only the MCCPs are used in PVC (at least in a big scale) and not SCCPs. In addition to being a plasticizer, the MCCPs enhance the flame retardancy of the PVC.

The majority of the MCCPs used in PVC have a chlorine content of 45% or 50-52% by weight ((European Chemicals Bureau, 2005) according to Euro Chlor, 1999). Less than 1% of the sold amounts have a higher chlorine content (e.g 56-58%) or a lower chlorine content (around 40%). The compatibility with PVC and the primary plasticizer increases with increased chlorination, but decreases with increased chain-length ((European Chemicals Bureau, 2005) according to (Beratergremium für umweltrelevante Altstoffe (BUA), 1992)).

The majority of flexible PVC is believed to be used in flooring, wall covering, upholstery and insulation of wire and cables (European Chemicals Bureau, 2005).

The MCCPs usually account for 10-15% of the resulting plastic article (European Chemicals Bureau, 2005).

2.4.2 RUBBER AND OTHER POLYMERES

Rubber products can be produced from natural rubber or several different types of synthetic rubber, which are all polymers.

SCCPs with high chlorine content (e.g. 70% Cl by weight) are used as flame-retardants in synthetic and natural rubber (Zitko & Arsenault, 1974). SCCPs with lower chlorine content are used both for their flame retardancy and plasticizing effect. MCCPs are usually functioning as both plasticizers and flame retardants (European Chemicals Bureau, 2005).

The SCCPs are generally added to the rubber in fractions ranging from 1-4%, but can account for up to 15% by weight of the application ((Beratergremium für umweltrelevante Altstoffe (BUA), 1992), (Zitko & Arsenault, 1974)). Similar fractions of MCCPs are applied (European Chemicals Bureau, 2005).

The major use of rubber containing SCCPs in the EU is in conveyor belts for under ground coal mining. But they are also used in technical articles such as hoses and gaskets for buildings and automotive applications (European Chemicals Bureau, 2008a). Less information on the use of MCCPs in rubber is available, but MCCPs have also been found to be used in conveyor belts and technical articles (Entec UK Limited, 2008).

The use of CPs in other plastics (not PVC) is mainly as a flame retardant (European Chemicals Bureau, 2005). One reported use is in polyurethane, for rigid foams and one-component foams (used for insulation, among others) (Entec UK Limited, 2008).

2.4.3 COATINGS

CPs are used in paint, varnishes and other coatings (European Chemicals Bureau, 2005). In the following, the term coating will be used to describe any material that is applied as a thin continuous layer to a surface.

CPs main function in coatings is to be a plasticizer, but they also improve the water resistance, chemical resistance and inflammability of the coating (European Chemicals Bureau, 2000). The coatings in which CPs are used are mostly solvent-born and chlorinated rubber and vinyl copolymer are the most common resin types (European Chemicals Bureau, 2005). Some examples of coating types and associated CP concentrations are shown in the table below.

Coating type	Chlorinated paraffin content (% by weight)
Organic solvent borne chlorinated rubber primers and topcoats	1-5
Organic solvent borne chlorinated rubber systems for swimming pools/fishponds	5-20
Organic solvent borne zinc rich (epoxy) primers	2-5
Organic solvent borne acrylic container coatings	2-10
Organic solvent borne chemical and water resistant coatings	5-20
Organic solvent borne vacuum metallising lacquers	1-5
Organic solvent borne flame retardant coating for wood	1-5
Organic solvent borne intumescent coating for structural steel	20-30
Organic solvent borne floor paints	5-10
Organic solvent borne water-proofing coatings for walls	5

Table 1: Coating types and associated CP concentrations. Source: (European Chemicals Bureau, 2005) according to British Coatings Federation 1999.

For MCCPs used in coatings, a chlorine content of 50-60% by weight has been reported (European Chemicals Bureau, 2005). The chlorine content of SCCPs used in coatings appear to be a bit higher, ranging from 60-70% by weight ((European Chemicals Bureau, 2000), (European Chemicals Bureau, 2008a) according to British Coatings Federation 1999).

The typical concentration of CPs in coatings is reported by Euro Chlor to be 4-15% in wet paint. This corresponds to 5-20% when the coating is dry (solvents have evaporated) (European Chemicals Bureau, 2005, 2008a).

LCCPs are according to Zarogiannis and Nwaogu (2010) the most commonly used CP in coatings.

2.4.4 SEALANTS AND ADHESIVES

Sealants and adhesives are sometimes difficult to distinguish from each other, as some sealants are used as adhesives and vice versa (Palmer & Klosowski, 1997). A sealant is described as a material that is used to keep wind, water, dirt or other contaminants from passing through gaps or joints. While adhesives are used to transfer loads and therefore have higher tensile and shear strength. However, it seems like only small amounts of CPs are used in adhesives and the term sealant will be used in the following to describe both sealants and adhesives.

The main function of CPs in sealants and adhesives is as a plasticizer and flame retardant ((European Chemicals Bureau, 2005) according to Euro Chlor and (Beratergremium für umweltrelevante Altstoffe (BUA), 1992). Examples of sealant where CPs are used are polyurethane, polysulfide, acrylic and butyl sealants. but it is not used in modern silicone rubber.

The MCCPs used in sealants typically have a chlorine content of 50-58% by weight (European Chemicals Bureau, 2005). SCCPs used in sealants are reported to have a chlorine content of 56-65% by weight (European Chemicals Bureau, 2000).

In the EU risk assessment for MCCPs (2005), the typically concentration of CPs in sealants is reported to 10-14% of the final sealant. In the risk assessment for SCCPs (2008) the range is given as 5-14% of the final sealant. In both cases the concentration could amount to 20% of the sealant, in exceptional cases.

2.5 LEGISLATION AND CLASSIFICATION

The EU posed restrictions on the use of SCCP in an amendment to the Marketing and Use Directive of June 2002 (76/769/EEC), (European Parliament 2002). The amendment states that SCCPs may not be placed on the market for use as a substances it self or as a constituent of other substances or preparations in concentrations higher than 1 % in metalworking and for fat liquoring of leather. EU members where required to adopt regulations to comply with the EU directive by January 2004.

The use of MCCPs is not restricted, but a proposal for regulating MCCPs in consumer articles is sent to the EFTA Surveillance Authority (ESA) for notification in the EU (Klima- og forurensningsdirektoratet 2011a).

In Norway SCCPs are more stringently restricted than in the EU. Production, import, export, sales and use of SCCPs and preparations and products containing more than 0,1 % by weight of SCCPs, has been prohibited since 2002 (Miljøverndepartementet, 2004a). MCCPs are not restricted in Norway. In stead, the Norwegian authorities has worked on banning MCCPs on the wider European level, since most MCCPs enter Norway through imported articles.

SCCPs where classified by the European Chemicals Agency's as a substances of very high concern in 2008 and included on the EU Candidate list (European Chemicals Agency 2011b). In Norway, both SCCPs and MCCPs are listed on the corresponding Norwegian list for substances of concern and it is resolved that emissions and use of substances on the list should be stopped within 2020 ((The Norwegian Climate and Pollution Agency n.d.), (The Norwegian Climate and Pollution Agency 2011)).

2.6 International agreements

2.6.1.1 UNECE LRTAP Convention: POPs Protocol

The United Nations Geneva convention on Long Range Transboundary Air Pollution (LRTAP) was established in 1979. The convention addresses environmental problems in the United Nations Economic Commission for Europe (UNECE). In 2009 SCCPs (C 10-13 and Cl 48% by weight) where added to the 1998 Protocol on Persistent Organic Pollutants (POPs) (UNECE 2009). The protocol prohibits all production and use of SCCPs except in rubber used for conveyor belts in the mining industry and in dam sealants.

2.6.1.2 Stockholm Convention on POPs

The Stockholm Convention on Persistent Organic Pollutions (POPs) is a global treaty under the United Nations Environment Program to protect the environment and human health against POPs (Secretariat of the Stockholm Convention n.d.). It was agreed upon in 2001 and entered into force in 2004. A list of twelve POPs where chosen, for which governments who have signed the treaty should take measures to eliminate or reduce. SCCPs are under review for listing on the global Stockholm Convention on POPs. The review comity (POPRC) has not been able to agree upon a risk profile of the SCCPs.

2.6.1.3 OSPAR Convention

OSPAR is a convention for protecting the North-East Atlantic. It is governed by 15 contracting parties/countries and the European Commission (OSPAR n.d.). In 1995 the OSPAR Commission adopted a decision on SCCPs – PARCOM Decision 95/1 (OSPAR n.d.). This decision required the contracting parties to phase out SCCPs as plasticizers in paints, coatings and sealants; as flame-retardants in rubber, plastics and textile; and their use in metalworking fluids. This should be achieved by 1999, except for the use of SCCPs as flame-retardants in conveyor belts in underground mining and plasticizers in dam sealants, which should be phased out by 2004.

2.7 Literature survey

SFAs of CPs has been conducted on three different geographical levels; regional (EU), country (Japan, Norway) and city (Stockholm). Three of the studies concentrate on SCCPs, while one of the studies includes all CPs. Stocks and flows are quantified for only one year. General problems for all the studies is the lack of knowledge on the historical amounts used, the division of amounts between SCCPs, MCCPs and LCCPs used and divisions of the amounts used for different applications.

On the request of the European Union, a study on waste related issues of newly listed POPs and candidate POPs was conducted by the German Federal Environmental Agency (Umweltbundesamt) and two consultant firms (BiPRO and Enviroplan) in 2011 (Umweltbundesamt et al. 2011). In this study simple SFA systems on all the main application areas of SCCPs where analyzed. The study is based on sales data in EU from 1994 until 2009 from the European chlorine organization Euro Chlor. The SFA study summarizes important data for conducting a SFA, but it mainly quantifies flows and not stocks, it does not clearly defined the system analyzed and it does not discuss imports and exports of CPs to the EU.

Two SFA studies are conducted on country level, Japan and Norway. The study in Japan was conducted for the year 2001 (Tsunemi & Tokai 2007). The estimations of stocks and flows were based on domestic production data of SCCPs in Japan from 1979 until 2001. The fraction of SCCPs of total CPs in 2001, is assumed to be the same for all years. It can be discussed if this is a good assumption. In Europe the use of SCCPs has decreased substantially in later years. In lack of data for where the SCCPs where used, a half by half division

between SCCPs used in metal working fluids and SCCPs used in products was also assumed and one emission factor was used for all SCCPs in products (differentiating between outdoor and indoor use). Imports and exports of SCCPs are not discussed.

The SFA of CPs in Norway was conducted for the year 1998 (Bjørnstad 1999). The study does not include imported SCCPs through finished articles; neither does it include emissions and wastes from historical consumption stored in stocks. The SFA is therefore just estimating the added amounts of SCCPs in flows and stocks from use in 1998. The earliest available information on use of SCCPs was from 1991. The system is not clearly defined and it includes only three processes.

A SFA of CPs in general was conducted for Stockholm for the year 2004 (Fridén & McLachlan 2007). This study is the most transparent study of the SFA studies discussed here. It estimates both flows and stocks of four applications paint, sealants, plastics- and rubber products and others. It manages to overcome the difficulties of estimating the stocks in lack of data by multiplying the usage for one application one year by the lifetime of the application. The latest available information of CPs use in Sweden was from 1995.

In addition to the SFAs mentioned, several mappings of SCCPs and MCCPs has been conducted in Norway. The Norwegian Climate and Pollution Agency (KLIF) publishes every year a review of environmental pollutants in products where SCCPs and MCCPs are included. They have also requested several surveys from consultant firms (Amlo, S. & Bakke, K. 2010),(Weholt 2005),(Cowi 2010a). A mapping of MCCPs in products from 2010, compared registered amounts of MCCPs in Norway with the consumption if an average EU per capita consumption was assumed. The results show that there might be an underestimation by one order of magnitude for the amounts registered in Norway (for 2009 12 tons were registered in Norway, while 395 tons was the result if an average EU consumption per capita was assumed). All the reports on CPs in Norway are estimations for of amounts used in different applications for a limited number of years from the 90s until today.

3 METHODOLOGY

This chapter describes the methodology and model used for quantifying the stocks and waste flows from building and construction in the EU. Thereafter, data and assumptions used in the calculations are presented. Lastly, uncertainties are discussed and the conditions for a sensitivity analysis are given.

3.1 MATERIAL FLOW ANALYSIS

Abel Wolman introduced the term "metabolism of cities" in an article for Scientific American in 1965 (Wolman, 1965). He viewed the city as a living organism with inputs, outputs and stocks of material and energy. Many disciplines studies flows and stocks of material or energy, but their individual methodologies are designed for analyzing specialized areas and is insufficient for a holistic study of an entire anthropogenic system. In the "Practical Handbook of Material Flow Analysis", Brunner and Rechberger present a methodology for analyzing such systems and their interaction with nature – identifying problematic flows or stocks and studying cause and effects. Brunner and Rechberger defines material flow analysis (MFA) as "the systematic assessment of the flows and stocks of materials within a system defined in space and time" (Brunner & Rechberger, 2004).

The stocks and flows of the defined system is quantified by data collected on the system, e.g. technologies used, emission ratioes, resource consumption, etc. In addition the principal of mass balance is utilized. Inflows, outflows and net changes in the stock of a process needs to be in balance, as well as the total inflows, outflow and net stock of the entire system.

In the following, relevant MFA terminology (as defined by Brunner and Rechberger) is described.

A **substance** is defined as a chemical element or compound composed of uniform units. Examples are: cadmium (Cd), carbon (C), carbon dioxide (CO_2), ammonia (NH_3).

A **material** is composed of several substances. Examples are: wood, steel, concrete.

A **system** can generally be described as a group of elements and the interactions between them. In MFA these elements are processes and the interactions between them are flows.

A **process** is in MFA something that represents transformation, transport, storage etc. of goods. Examples of processes are: the production of a mobile phone, mining of iron, an oil well (representing storage), waste collection, a market (with imports and exports), etc.

A **good** is defined as an economic entity of matter, with a positive or negative economic value. A good can be made up of one or several substances. Examples of goods are: drinking water, TV sets, automobiles, garbage, sewage sludge.

A **flow** is the amount of a good or a substance that is exchanged between two processes in the system per time, or in or out of the system.

A **stock** is the stored amount of a substance or material in a process. If the input flows and output flows of a process is not in mass balance, there will be a net change in the stock.

A dynamic MFA model is a time dependent model. This means that the flows and stocks of a system may change with time.

3.2 SYSTEM DEFINITION

The system shown in
Figure 3 represents production processes, application and use of the four main product groups relevant for building and construction in the EU; sealants, coatings, PVC and rubber/other polymers. Metal working fluids, textiles, leather and carbonless copy paper are not relevant, according to the definition of use in building and construction applied here (see section 3.2.11).

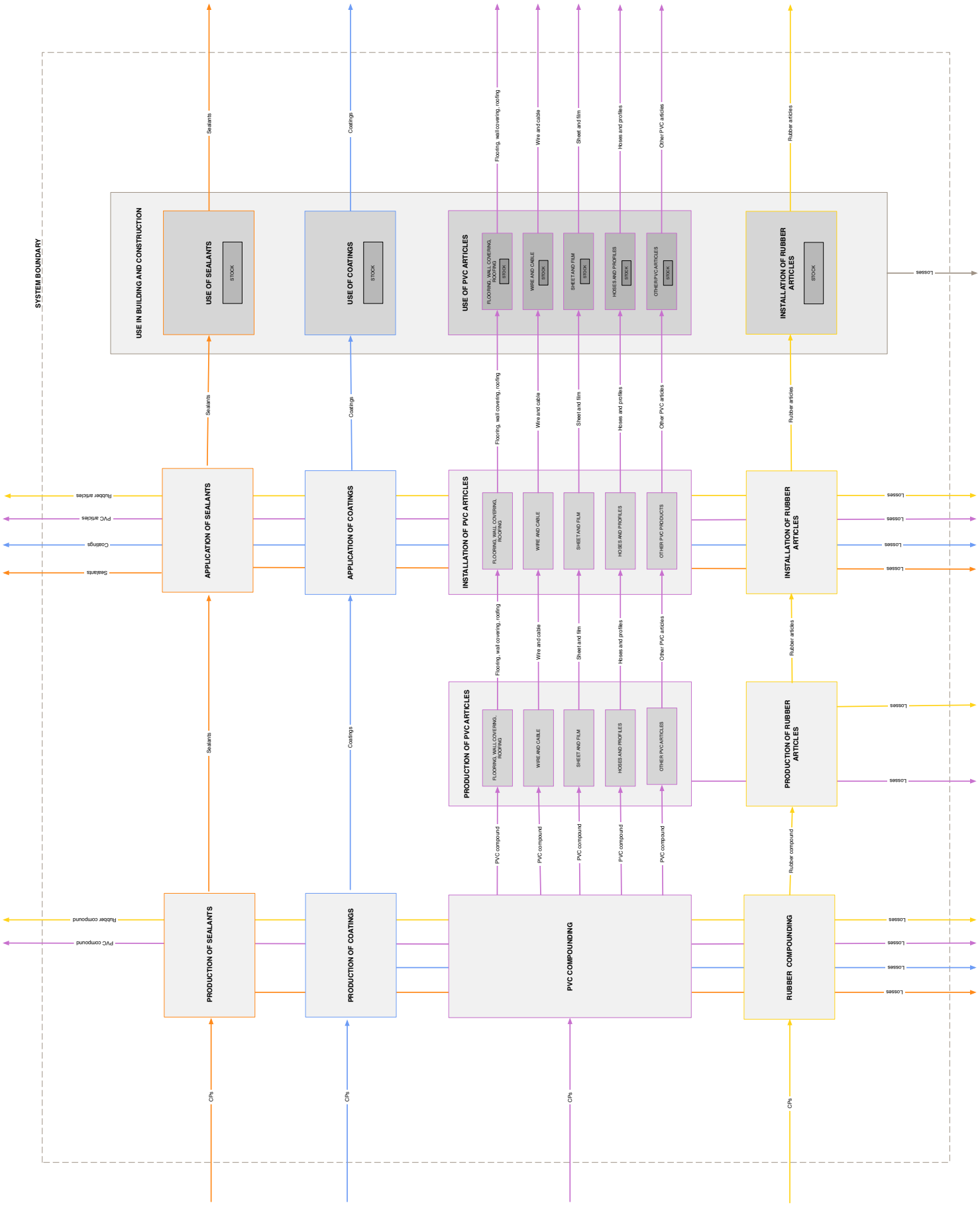

Figure 3: System definition

The flows and stock of both SCCPs and MCCPs are quantified according to the same system. However, SCCPs are not used in PVC. The processes involving PVC are therefore not relevant for SCCPs.

It has not been a goal to quantify the emissions of CPs to the environment. The flows labeled "losses" are therefore not necessarily losses to the environment, but could include losses through waste or emissions that are collected in wastewater and air treatment systems.

The production of CPs has not been included in the system. This is because data on production amounts and imports and exports of CPs to EU, were not available. Only consumption figures of CP use in the production of the main product groups where available (sealants, coatings, PVC, rubber/other polymers).

Imports and exports of CPs could occur for each of the goods exiting processes in the system. However, data was not available for quantifying this and imports and exports were assumed to be negligible. Imports and exports are therefore not modeled in the system.

It is possible that the CPs are incorporated in chemical mixtures before they are added to the production of sealants, coatings, PVC and rubber. However, no information describing this has been found, and the production of chemical formulations for use in sealants, coatings, PVC and rubber has therefore not been included in the system.

In the following, each of the processes visualized in the system is described.

3.2.1 PRODUCTION OF SEALANTS

This process represents the production of both sealants and adhesives. The distinction between the two is not always clear as some sealants are used as adhesives and vice versa (European Chemicals Bureau, 2005). It seems like only small amounts are used in adhesives, sealants will therefore be used in the following as a general term describe both goods.

Sealants are produced by mixing a viscous liquid polymer with required additives (European Chemicals Bureau, 2005). Most sealants are moister sensitive and the mixing is usually carried out under vacuum, to avoid moisture entering the process. Gentle heat can be added (up to around 40° C). Sealants are usually produced in batches.

When the mixing of the sealant is finished the sealant is filled directly from the mixing vessel to cartridges or tins.

Cleaning between batches is sought minimized, but is generally done by removing solid material by hand. The equipment can also be cleaned by using solvents.

MCCPs are used in both 1-part and 2-part sealants and the production methods are similar for both types.

3.2.2 APPLICATION OF SEALANTS

In this process the sealant is applied in a product (window, car etc.), a building or other infrastructure. Even though the sealants are applied in different products and at different sites, the process of applying them is assumed to be similar. The sealants are usually supplied in cartridges and squeezed onto the desired point of use (European Chemicals Bureau, 2005). The sealant may also be supplied in tins and filled into cartridges on site.

3.2.3 FORMULATION OF COATINGS

CPs can be used in paints, varnishes and other types of coatings (European Chemicals Bureau, 2005), (European Chemicals Bureau, 2000). Here the term coating is used to describe all of these goods (paint, varnishes etc.) and is defined as any material that is applied as a thin continuous layer to a surface.

The formulation of coatings includes four basic steps: dispersion, milling, finishing and filling (OECD, 2009a). Dispersion is the pre-mixing of raw materials and the addition of all powder pigments and extenders to form a pigment concentrate. Milling or grinding, is the reduction of the particle size of pigments and extenders. Finishing is the mixing of the pigment concentrate with all remaining liquid components. This also includes quality sampling and tinting of the mix to get the right color, viscosity, etc. Filling includes filtration and filling into final packages.

Formulation of coatings is a batch process (OECD, 2009a).

The process also includes: delivery, unloading and storage of material; storage and dispatch of finished goods; cleaning of manufacturing vessels; disposal of raw material packaging, cleaning and other wastes; containment and abatement of volatile organic compounds (VOC) emissions.

3.2.4 Application of Coatings

This process represents the application of a coating to a surface. Several methods can be used for the application of coatings. For example a roller or paintbrush could be used, or the coating could be sprayed on to the surface. There has not been found any information on which methods that are most likely to be used for the application of coatings containing CPs.

3.2.5 PVC compounding

Compounding is the mixing of plastic resins (pellets, flakes, powders) with other additives, such as pigments, stabilizers, reinforcements and plasticizers (Britannica, 2012). For the production of PVC, two different compounding processes are relevant; dry blending and plastisol blending (OECD, 2009b). A third process, Banbury mixing, seems to have been used to some extent, but this will not be investigated further, since it has similar emission ratios as dry blending.

Dry blending is the mixing of dry ingredients. In the case of PVC this could also include a liquid plasticizer, which could contain CPs (Britannica, 2012). The liquid plasticizer is then incorporated into the porous PVC powder and the final mixture still appears to be dry. The mixing is performed by a high-speed rotation agitator and may reach a temperature of 100-120°C by friction (OECD, 2009b). The mixing (and cooling of the resulting compound) is done in closed containers.

Plastisol blending is the mixing of plastic raw materials to a resulting liquid called plastisol (OECD, 2009b). Very fine PVC particles are dispersed in plasticizers (50% or more plasticizers) and the suspension is stirred until the polymer particles are dissolved in the plasticizers ((Britannica, 2012), OECD, 2009)). The mixing takes place in stirred vessels under ambient temperature. The vessels may be cooled to reduce the heat from friction.

Dry blending and plastisol blending can involve several processing steps and the use of various machines (OECD, 2009b).

3.2.6 Production of PVC articles

This process includes five internal processes; production of flooring, wall covering and roofing; production of wire and cable; production of sheet and film; production of hoses and profiles; production of other PVC articles. This has been done to be able to quantify the amounts of CP used for the different PVC articles.

Processing or conversion of the PVC compound into products can be done in a number of different ways. Even though the equipment and processes may differ, the principal is the same; the plastic compound is melted or softened (i.e. plastisol), formed to a shape and subsequently cooled. The most important conversion processes for PVC containing CPs are describes below. These include calendaring, extrusion, injection moulding and plastisol spread coating (European Chemicals Bureau, 2005).

The conversion methods can be categorized as open, partially open or closed systems. An open system is defined as a system in which materials are converted in almost completely enclosed environments. Partially closed systems are systems where the materials are exposed to ambient environment for some period at operating or near operating conditions. While closed systems are systems in which the material is exposed to ambient environment in an unrestricted way for some period, but possibly involving the use of some environmental control.

Extrusion can be seen both as a conversion process and an extension of the compounding process (OECD, 2009b). Polymers and additives are mixed, fed into an extruder and heated. The hot melt is then poured into dies producing pipes, profiles, sheets and wire coating, amongst others. Volatile emissions are vented at various points in the extruder. The process may be closed or partially closed depending on if the hot material is quenched immediately after exiting the die. In addition to being an independent conversion process, the extruder is often used only for heating the compound before it enters some of the other conversion processes described below.

Injection moulding is similar to extrusion apart from that the melted compound is projected into a mold instead of a die. The compound is often heated by an extruder, which delivers the melted compound to the mould (OECD, 2009b). Injection molding is a closed process, as both the moulding and cooling takes place in closed systems (European Chemicals Bureau, 2005; OECD, 2009b). Injection moulding can be used for the production of a number of small and medium sized articles (Store norske leksikon, n.d.).

Calendering is when a preheated compound is fed onto a calendar (OECD, 2009b). A calendar comprises of a number of rolls that compresses the compound to a thin layer. Calendering is used to produce films, sheets or to coat textiles and is an open process.

Plastisol spread coating produces flooring, wallcovering, tarpaulins etc. (European Chemicals Bureau, 2005). The plastisol is gelled in hot tunnel ovens (around 180° C). Plastisol spread coating is an open system process.

The conversion of plastisols may differ from the conversion of dry blends, because they are already liquid and do not have to be melted before they are formed. Plastisols can be spread onto fabrics/materials or given a shape just by dipping a form into the plastisol so that it sticks to it and takes it's shape (Store norske leksikon, n.d.). They can also be coated to a material, rotational moulded or cast into molds. After the plastisol has taken the wanted form, it is heated and the plastic and plasticizers melt together and solidifies.

3.2.7 INSTALLATION OF PVC ARTICLES

Flooring, hoses, cables etc. needs to be installed in buildings and constructions before they can be used. This means that flooring, hoses, sheets and cables needs to be fitted to their end use site. Installation of the products can involve different kind of processes depending on the product installed and the purpose of use. Installations of all kinds are included in this process, with the unifying characteristic of production of waste in form of cut-offs.

3.2.8 COMPOUNDING OF RUBBER

In this process polymers and additives are mixed to form a compound ((Gent, n.d.), (OECD, 2004)). The process and equipment is similar to compounding of PVC, although it is carried out under lower temperature (see section 3.2.5 for further details) (MCCP according to Kirk-Othmer 1997). When the polymer and additives are ready mixed, the compound is sheeted out, cooled by immersion in water and stored in piles.

3.2.9 PRODUCTION OF RUBBER ARTICLES

Shaping of the rubber is done in closed systems, by extrusion, injection or compression moulding (European Chemicals Bureau, 2005). These processes are described in section 3.2.6.

3.2.10 INSTALLATION OF RUBBER ARTICLES

As for PVC products, rubber products used in building and construction needs to be installed at the site before use. Installation of the products can involve

different kind of processes depending on the product installed and the purpose of use. Installations of all kinds are included in this process, with the unifying characteristic of production of waste in form of cut-offs.

3.2.11 USE IN BUILDINGS AND CONSTRUCTIONS

This process represents the use of the products produced and installed/applied in buildings and constructions. Buildings and constructions comprise residential, commercial and industrial buildings, bridges, roads, tunnels and other underground constructions, etc. It does however not include oil and gas constructions (e.g. platforms and pipelines) or the electricity grid.

Products used in building and construction comprises everything that is mounted/fixed to the building or construction. Wallpaper, flooring, windows, paint, cables fixed to the construction etc. is regarded as part of the building or construction. Furniture, carpets, curtains, blinds, and electrical devices (lamps, washing machines, computers etc.) are not regarded as part of a building/construction and it's use is not included in this process.

Use in building and construction is the only process in the system that is modeled with a stock.

3.3 MODEL

All stocks and flows in the system defined above, is to be quantified. Information on use patterns and wastes and emissions from the processes are needed for doing this. In addition the principle of mass balance is utilized. In the following, the model used for quantifying the system, is outlined.

F_{G-H}	–	flow from process G to process H (or from/to the system (O) as loss (Ol) or for other uses (Ou))
F_{Gi}	-	total input flow to process G
S_G	-	Stock of process G

α_{yt} – estimated parameter, changes with time t. y can be s for sealant, c for coating, pvc for PVC or r for rubber

L_G - parameter determining losses (emissions plus waste) from process G

D_{G-H} - parameter determining distribution from process G to H

The input flows to the system equals estimated parameters α_{yt}, where y labels if the input flow is for the production of sealants (s), coatings (c), PVC (pvc) or rubber (r).

$F_{O-G} = \alpha_{yt}$

α_{yt} is the only parameter that varies with time, but because all flows are dependent on the input to the system, all flows varies with time as well. The method used for determining α_{yt} is described in chapter 3.4.

From all processes in the system there is a flow of losses (waste and emissions), exiting the system (F_{G-Ol}). The loss from a process is determined by one or several parameters (here presented as one parameter) and the inflow to the process:

$F_{G-Ol} = F_{Gi} * L_G$

For processes where F_{G-Ol} is the only loss, the continuing flow from the process (G) to the next process (H) is determined by mass balance:

$F_{G-H} = F_{Gi} - F_{G-Ol}$

A second flow may exit the process and system, F_{G-Ou}. This flow is not composed of waste or emissions, but other goods for uses that are not relevant for building and construction. In this case a distribution parameter determining the ratio of the good, which is to enter the next process in the system, is needed.

$F_{G-H} = (F_{Gi} - F_{G-Ol}) * D_{G-H}$

The loss from a process to other uses can then be determined by mass balance:

$F_{G-Ou} = F_{Gi} - F_{G-Ol} - F_{G-H}$

Flows from the process "use in building and construction" (U) are calculated in a slightly different way. Emissions and waste are separated into two different flows; F_{U-Oe} and F_{U-Ow} where e labels emissions and w waste.

The emissions of CPs to air and wastewater from use in building and construction should ideally be distributed over the lifetime of the product in use. However, since the emissions are small and the data found gave accumulated emission estimates for the entire lifetime of the product, these accumulated

lifetime emissions are subtracted from the process the same year as the product enters use.

$F_{U\text{-}0e} = F_{Ui} * L_U$

For estimating the waste flow from building and construction ($F_{U\text{-}0w}$), a lifetime approach has been taken. This means that the CPs exits as waste dependent on the lifetime of the product it is contained in.

For determining the lifetime of the products, a lifetime distribution P(t', t) is used. The lifetime distribution gives the probability for when a product exits use and becomes waste after a given time. This is dependent on the average lifetime of the product. A normal distribution function is used for determining the lifetime distribution for sealants, PVC and rubber. For coatings the Weibull distribution was used. This was done because the standard deviation was assumed to be relatively high for coatings, which gave an unrealistic lifetime distribution using the normal distribution. Further explanations of the choice of lifetime distribution is given in section 3.4.5.

The waste flow for a certain year is hence dependent on previous amounts entering use and the probability for the product to exit use after the number of years since it entered use.

$F_{U\text{-}0w} = \int_{t0}^{t} P(t',t) \times (F_i(t') - F_{0e}(t'))dt'$ where t: actual time
t': time of input
t_0: year when use began
P(t', t): Lifetime distribution

The stock of CPs in buildings and constructions is then determined by

$S(t) = \int_{t0}^{t} [F_i(t) - F_{U\text{-}0e}(t) - F_{U\text{-}0w}(t)]dt'$ where t: actual time
t': time of input

3.4 DATA AND ASSUMPTIONS

A top–down approach was chosen for the data collection and estimations. This means that overall production figures of CPs and information on the processing into final articles have been collected rather than detailed data on the use of different products in building and construction (bottom-up approach).

In the following, all parameters used for quantifying the system, are presented. Data on the input flows to the system, α_{yt}, is collected for both SCCPs and MCCPs. All other parameters used for quantifying the system, are equal for both SCCPs and MCCPs (except emission estimates for sealants in the use phase). This is a simplification, and is due to the lack of available data.

It has been desired to estimate the stock and flows of CPs from the starting point of the large-scale CP production in 1932. A large amount of data needed to be collected for achieving this. Data on amounts of SCCPs and MCCPs used in the production of each of the main product groups containing CPs, were available from the year 1994 to present. Prior to this there was no quantified information on the distribution of CPs for different applications. Information on the total European and global consumption was therefore collected, an estimate of the distribution between SCCPs, MCCPs and LCCPs was made, and the distribution of SCCPs/MCCPs for the production of the relevant product groups was assumed to equal the distribution in 1994. The methods used for estimating the yearly input flows, α_{yt}, are described in more detail in conjunction with the presentation of available data from literature.

Most of the data collected represents the EU, but EU has expanded considerably within the period analyzed. No adjustments have been made to correct for this. In the MFA by Bipro et al. (2011) on SCCPs in the EU, an adjustment of the consumption figures for SCCPs was made based on the relative increase of population within the region. The consumption of CPs in production processes within EU is however not directly coupled with population, but rather with the type of industry within the region. The expansion of the EU is also not the only complicating factor of the data collected. For some estimates, the numbers given where said to represent Europe or western-Europe. No further explanation of what was defined as western-Europe was given. To keep the estimates simple and transparent, it was decided not to adjust the numbers given in literature. All values found in literature representing Europe or western Europe are assumed to be representative for EU.

Information on imports and exports of CPs, or goods containing CPs, where scarce. The data presented is therefore production figures, or consumption figures for the production of different articles. Final articles or goods for further processing (for example PVC compound), may be imported or exported from the EU, but this has not been possible to account for.

3.4.1 TOTAL EUROPEAN AND GLOBAL CONSUMPTION OF CPS

Data on the long-term production and consumption of CPs in Europe is scarce. Since the start of the large-scale commercial production in 1932, only three reliable estimates of the total use of CPs in Europe (including all chain-lenghts) has been found. Not all estimates found are estimates of the consumption of CPs in Europe, but rather of the production of CPs. As mentioned, little information was found on imports and exports of CPs. Production levels are therefore assumed to be close to consumption levels.

The earliest estimate available for Europe is for the year 1985 and is an estimate of the production in Western Europe. 95 000 tons are estimated to have been produced in 1985 ((World Health Organization, 1990) according to (SRI International, 1986)). The following estimate gives the European consumption in 1993 as 100 000-120 000 tons per year (B Willis, Crookes, Diment, & Dobson, 1994). Lastly, a current production estimate for Europe (for the year 2010) was provided by Euro Chlor (organization representing the chlor-alkali industry in Europe) and given as 45 000 tons (van Wijk, 2012).

Global estimates of the production/consumption of CPs where retrieved from literature to help fill in the gaps of information on consumption in Europe (especially between1932 to1985). In 1961 Hardie reported the world production of CPs to be roughly 35-50 000 short tons (32-45 000 tons). For the year 1977 Campbell and McConnell (1980) gave a world consumption estimate, excluding Eastern Europe, at 230 000 tons. They also estimated the growth rate of consumption for the preceding five years to be 5%. World production in 1985 was estimated to 300 000 tons ((OSPAR, 1996) according to (Beratergremium für umweltrelevante Altstoffe (BUA), 1992)). In 1988 Mukherjee estimated the world production, excluding Eastern Europe, to be approx. 340 000 tons. This is coherent to Omori (1987) that reports the world production to be more than 300 000 tons (Omori, Kimura, & Kodama, 1987). However, Mukherjee (1990) also reports that Houghton (personal communication, ICI Chemicals and Polymeres, 1989) believes that the world production (including eastern Europe) is 400 000 tons (including eastern Europe). The most recent estimate found is for the year 2001, and estimates the world production to be 400 000 tons and have an annual growth of 1-2% (Lahaniatis, 2001). The estimation is according to a website that cannot be connected to any more and additional information about the estimate could therefore not be retrieved. Since Houghton estimated the production to be 400 000 tons already in 1988, this might be an outdated estimate.

After 2001, no reliable estimates of the world production have been found in literature. However, it is well known that the production in China has increased drastically (de Boer et al., 2010).

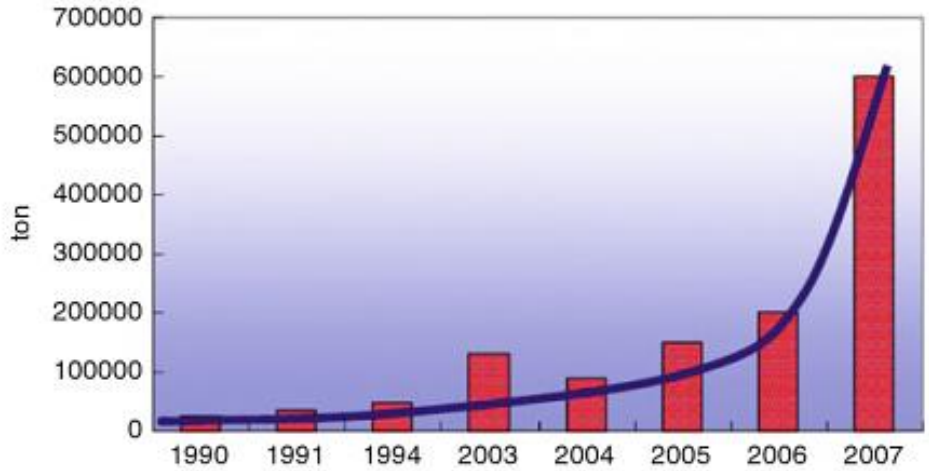

Figure 4: Production of CPs in China. Source: (de Boer et al., 2010).

The estimates of global production of CPs retrieved from literature are visualized in the graph below.

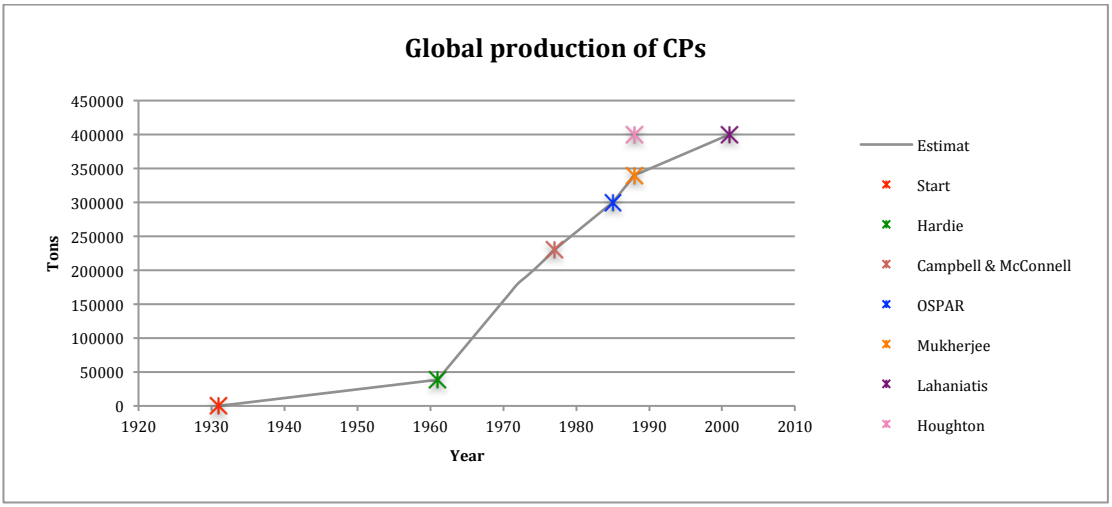

Figure 5: Global production of CPs. Source: (Howard et al., 1975) according to (Scheer, 1944), (Hardie, 1964), (Campbell & McConnell, 1980), (OSPAR, 1996) according to (Beratergremium für umweltrelevante Altstoffe (BUA), 1992), (Mukherjee, 1990) and (Lahaniatis, 2001).

Linear interpolation has been used for estimating the production between the given data points from literature (visualized in the graph as the grey line). The yearly estimates may be underestimated, as the numbers which where excluding Eastern Europe has been used. These where, however, the data points that gave the most likely and smooth growth in production.

For the year 1985, there is both an estimate of the European consumption and global production of CPs. The European consumption can therefore be

calculated to account for 31,7% of the global production this year. The relative fraction between European consumption and global production may have changed throughout the history of CPs, but for estimating the European consumption prior to 1985 it will be assumed that Europe consumed 31,7% of the global amount of CPs produced.

The European consumption of CPs after 1985 is estimated by linear interpolation between the given European consumption amounts retrieved from literature.

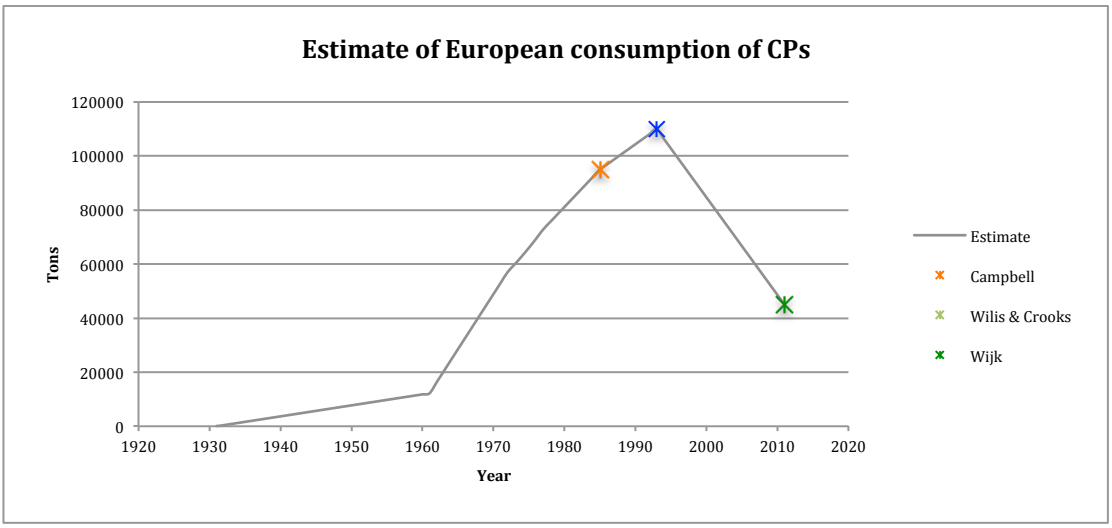

Figure 6: Estimate of European consumption of CPs. Source: (Campbell & McConnell, 1980), (B Willis et al., 1994), (van Wijk, 2012)

3.4.2 SCCPs, MCCPs and LCCPs in Europe

A few estimates of the amounts of SCCPs and MCCPs in Europe are available. Euro Chlor provided the National Institute for public health and environment in the Netherlands with sales data on SCCPs in EU from 1994 to 2008 (Zarogiannis & Nwaogu, 2010).

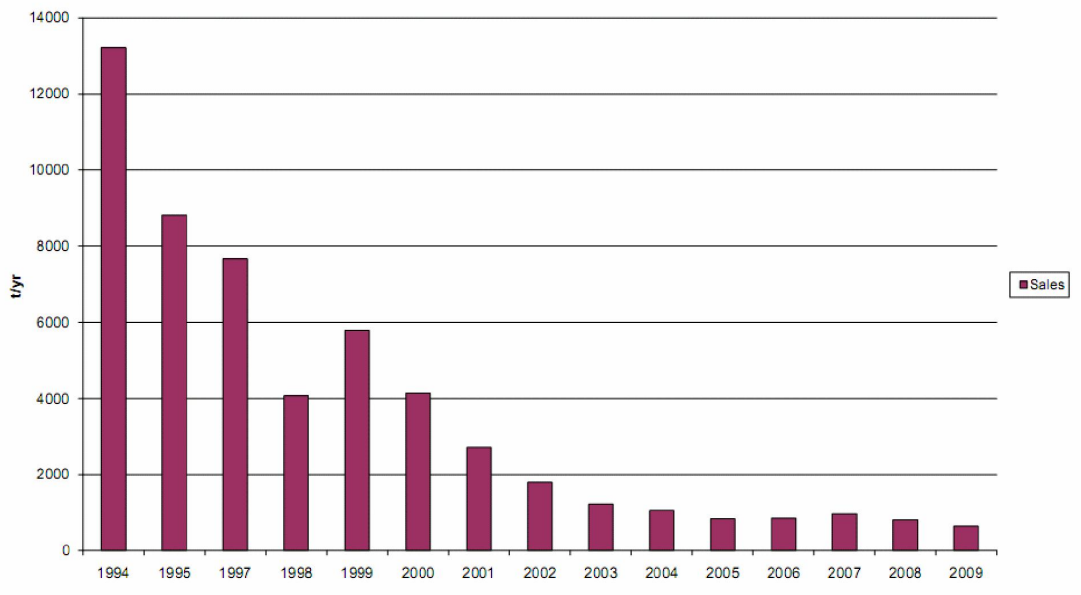

Figure 7: Sales data of SCCPs in EU15 for 1994-2003, EU25 for 2004-2006 and EU27 for 2007-2009. Source: (Zarogiannis & Nwaogu, 2010) according to EuroChlor.

The sales data covers the EU15, EU25 and EU27 as the union expands - the early sales data is not adjusted for the increasing number of countries included in EU. In a report for the European commission by Bipro et al., a 25% increase in the amounts where added to the reported figure for EU15 to account for the 12 new countries that has been included in EU. This was based on a per capita approach. As explained above, such adjustments have not been done in this thesis. It is believed that the trend in use is represented within a reasonable amount of uncertainty.

In an OSPAR document (1996) referring to a workshop on CPs in Brussels in 1994, annual sales data for SCCPs from 1989 to 1993 for western Europe, excluding Italy was given;16 400 tons in 1989, 14 200 tons in 1990, 12 750 tons in 1991, 11 650 tons in 1992 and 10 200 tons in 1993 (OSPAR, 1996). According to the European Chemicals Agency, the production in Italy was 1000 – 2000 tons in 1994/1995. Thousand tons were therefore added to the numbers from 1989 to 1993, to account for sales within Italy.

A few other sources for the amount of SCCPs consumed, where found. Euro Chlor gave the amount used in western Europe in 1994 to be 13208 tons (European

Chemicals Bureau, 2000). Zarogiannis & Nwaogu (2010) estimated the amount of SCCPs consumed in EU to be 530 tons in 2009. And in a report prepared for OSPAR, Bo Nyström (2009) gives the amount used in Europe in 1998 as 4075 tons.

Euro Chlor has also provided estimates of the amounts of MCCPs used in the EU from 1994 to 1997 and in 2003 (56 573, 58 671, 59 306 and 65 256 tons respectively) (European Chemicals Bureau, 2005) (Entec UK Limited, 2008). They also stated that the amount used in 1998 had fallen to a similar level as in 1994.

For the year 1993 the distribution of SCCPs, MCCPs and LCCPs consumed in Europe was estimated by Willis et al. to be 15% SCCPs, 70% MCCP and 15% LCCPs (B Willis et al., 1994). When using this fraction for calculating the total MCCP amount in 1993 (by using the estimated total CP consumption in Europe), it gives a value that is approx. 20 000 tons higher then the given amount for 1994 (57kt to 77kt). This seems unrealistic and the value is therefore not used. An explanation to the mismatch could be that the consumption data for MCCPs, SCCPs and LCCPs in 1994 are all EU figures, while the estimate for total CPs in 1993 is for the whole European region (the ratio 70% as well). The consumption of MCCPs in 1994 could therefore be underestimated in regard to the whole European region. Another pitfall could be that the 70% is more an estimate for the European production of MCCPs, rather than consumption (even though the source states that it is a rough estimate of the consumption). In a report for Defra it is found that almost 40% of the MCCPs produced in EU might be exported out of EU (Entec UK Limited, 2008). The value for SCCP calculated using the ratios given by Willis et al. is in relatively good agreement with the known value for 1994 (differ by 3300 tons). No estimates of the absolute amount LCCPs have been found, the calculated LCCPs could therefore not be compared to this.

In1977 Campbell & McConnel (1980) estimated the global use of CPs to comprise 60 kt of SCCPs (approx. 26%), 110kt MCCPs (approx. 48%) and 60 kt of LCCPs (approx. 26%). The same distribution is assumed to apply for Europe in 1977.

In addition to the values presented above, qualitative information is available for the early use of CPs. Initially, the availability of paraffin wax led to the development of LCCPs (Zarogiannis & Nwaogu, 2010) . Later on MCCPs were developed and replaced the LCCP waxes because their compatibility with PVC was better than the longer chain-lengths. The increased demand for SCCPs came even later, when the metal working fluid market expanded. SCCPs could hold a high chlorine content and had low viscosity and was therefore more suitable than MCCPs or LCCPs. Based on this it is assumed that the production of SCCPs and MCCPs each accounted for 10% of the CPs in 1932. In the 1950s the production of PVC increased dramatically. The amount of MCCPs produced is

therefore assumed to gradually increase to 15% by 1950, 20% by 1955 and 30% by 1960. The use of MCCPs is assumed to continue to increase and is set to account for 40% of the CPs in 1970 until it reaches 48% in 1977. The use of SCCPs is said to have increased when the market for metal working fluid expanded. It is not known when this took place, but a gradual increase to 15% in 1965 is assumed in the estimation, followed by an increase to 20% by 1970.

Figure 8 and Figure 9 gives the estimated total amounts and distribution of SCCPs, MCCPs and LCCPs in Europe.

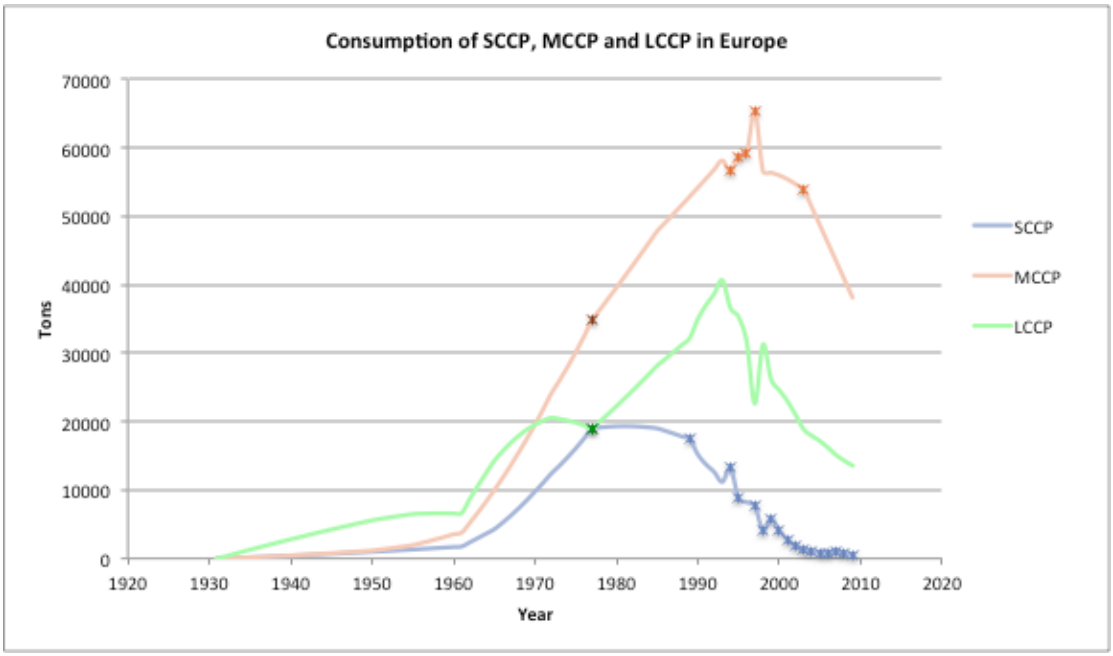

Figure 8: Estimated consumption of SCCPs, MCCPs and LCCPs in Europe. The values found in literature (described in the text above) are visualized in the graph as a star.

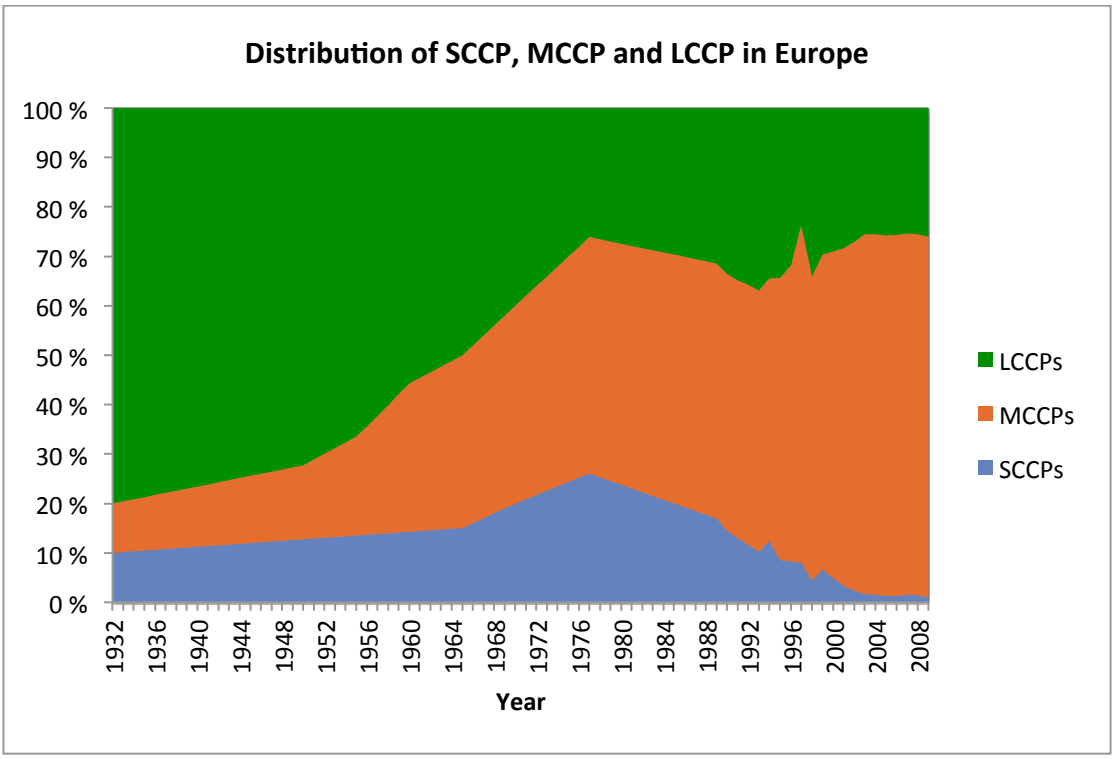

Figure 9: Distribution of SCCPs, MCCPs and LCCPs in Europe.

For LCCPs no estimations of the amount used in Europe has been retrieved (except the ratio of 15% given by Willis et al.). The fraction of LCCPs in the above graphs is therefor calculated using mass balance: LCCPs = CPs – SCCPs – MCCPs.

3.4.3 USE BY MAIN PRODUCT GROUPS

The final graphs given in this section, visualizing the consumption of SCCPs and MCCPs in the production of sealants, coating, rubber and PVC, are the input flows (α_{yt}) to the defined system in chapter 3.2. The estimates of SCCPs and MCCPs consumed in the production of sealants, coating, rubber and PVC are based on the estimate of the total European consumption of CPs and the distribution between chain-lenghts.

3.4.3.1 SCCPS BY PRODUCT GROUPS

The European Chemical Bureau collected data for the year 1994 from Euro Chlor for their risk assessment report on SCCPs in 2000. SCCP use was registered for metal working fluids, rubber, leather, paint and coatings, sealants and adhesives, and textiles. A small amount was also listed as “others”. This amount was said not to represent other uses then those mentioned above, but rather SCCPs that could not be tracked to their final application. The amount in the category

"others" has therefore been distributed to the listed applications by weighted fractions.

In a UNEP report estimates of the use of SCCPs in 1998 were given (Nyström, 2009). The product groups where however categorizes differently. Paint, adhesives and sealants were lumped together as well as rubber, flame retardants, textiles and polymers other than PVC. For MCCPs, the distribution between paint and sealants (and adhesives) has been reported by Euro Chlor to be 1/3 for paint and 2/3 for sealants ((European Chemicals Bureau, 2005) according to Euro Chlor). This is assumed to hold true for SCCPs as well. The amount listed as "rubber, flame retardants, textiles and polymers other than PVC" was distributed to the two categories rubber and textiles. To do this, the proportion between amounts listed as "rubber, flame retardants, textiles and polymers other than PVC" and "rubber" in two different sources for the year 1994 was used ((de Boer et al., 2010), (European Chemicals Bureau, 2000)). A small amount of SCCPs were listed as "others". As for the data given in 1994, the amount was distributed to each of the other product groups by weighted fractions.

For 2009 Zarogiannis & Nwaogu (2010) estimated the amount of SCCPs used rubber, sealants, coating and textiles.

In addition to the estimates found in literature, the use of metal working fluids and leather fat liquor is assumed to have ceased after the ban in 2004.

Between the given data points given in literature, linear interpolation has been used to calculate the yearly consumption within each product group. This is done to make the best estimate of the trend of use within each product group. To make sure that the sum of use for all applications a specific year, equals the estimate of total SCCPs used that same year, ratios for each application (based on the given numbers and the sum of them) are calculated and multiplied with the total estimated amount SCCP for that year.

The earliest estimations of amounts used for each of the product group, are for the year 1994. Therefore the distribution of use within product groups in 1994, is used for all previous years (1932-1994). The distribution is multiplied with the estimated total amount of SCCPs consumed in EU to obtain the absolute amounts.

The figure below shows the estimated total amount of SCCPs used in different product groups in EU.

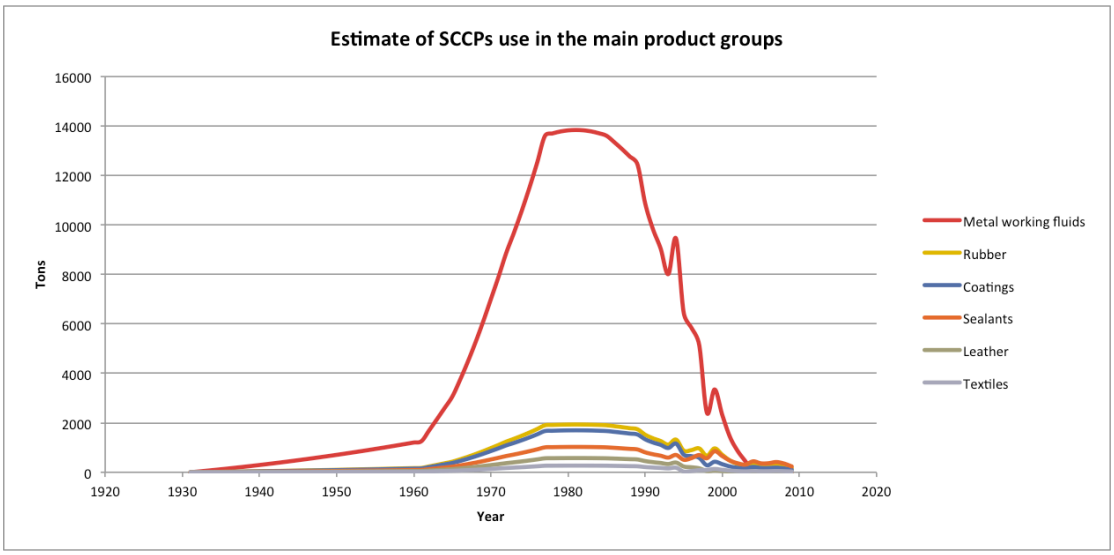

Figure 10: Estimates of SCCP use in main product groups.

The use in metal working fluids is clearly dominant. Figure 11 shows the amounts for rubber, coatings and sealants more clearly.

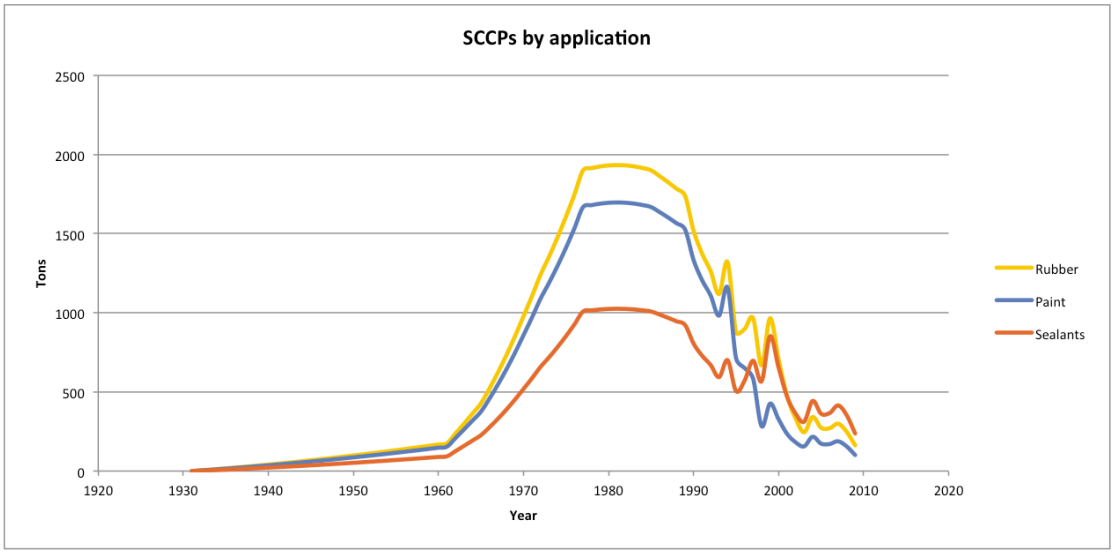

Figure 11: Estimated use of SCCPs in rubber, coatings and sealants in EU.

13 tons of SCCPs where reported to be used in PVC in 1998 (Nyström, 2009). No other estimates for use of SCCP in PVC have been found, and as it seems to be a minor use, it has not been modeled. However the use of SCCP in PVC might have been greater in the past (European Chemicals Bureau, 2008a).

3.4.3.2 MCCPs BY PRODUCT GROUPS

In the European Chemicals risk assessment for MCCPs (2005) amounts used per application is given from 1994 to 1997 (according to Euro Chlor). Estimates for

2003 and 2006, has also been found ((Entec UK Limited, 2008) and (Cowi, 2010) according to Entec). The product groups where MCCPs are used are; metal working fluids, PVC, rubber/other polymeres, paint and coatings, sealants and adhesives, leather and carbonless copy paper.

In all estimates found the product groups "paint and coatings" and "sealants and adhesives" are lumped together. 1/3 of the amount given is assumed to be used in paint and coatings, and 2/3 in sealants and adhesives, as indicated by Euro Chlor (European Chemicals Bureau, 2005).

Linear interpolation is used to achieve a realistic trend of MCCP use within each product group. To make sure that the sum of all interpolated values for a specific year equals the estimated total amount of MCCPs used in EU, ratios are calculated based on the sum of amounts used in all product groups and multiplied with the estimated total amount of MCCPs.

The earliest estimations of amounts used for each of the product group, are for the year 1994. Therefore the distribution of use within product groups in 1994, is used for all previous years (1932-1994). The distribution is multiplied with the estimated total amount of MCCPs consumed in EU to obtain the absolute amounts.

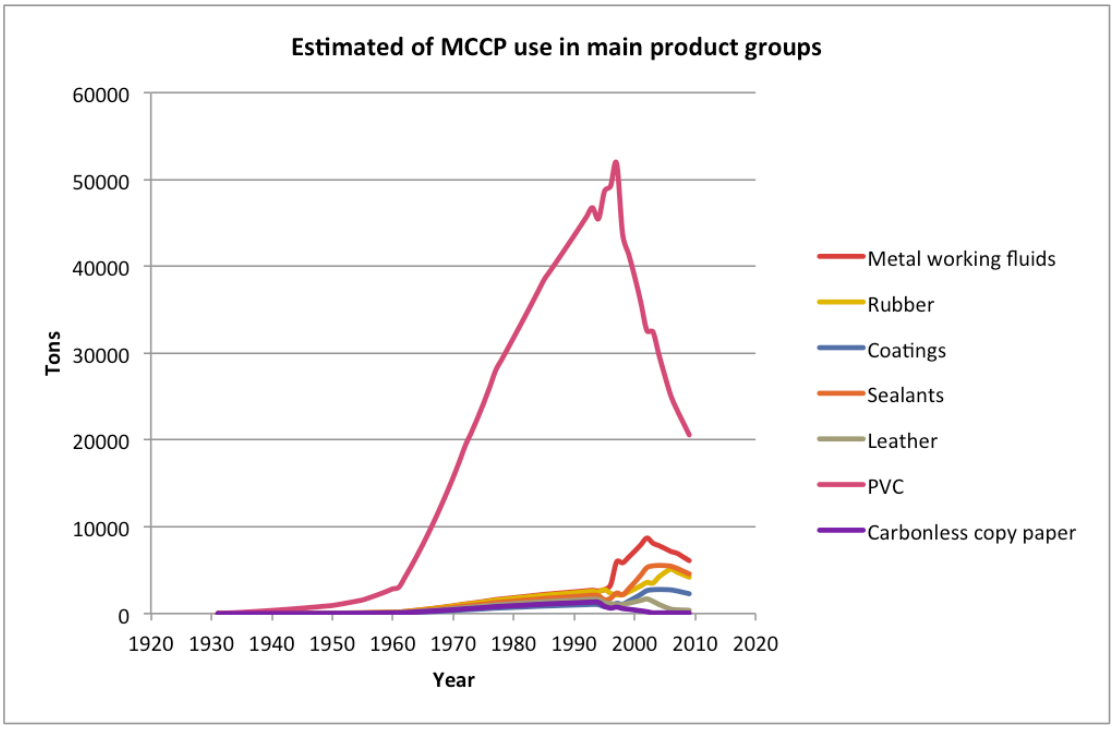

Figure 12: Estimate of MCCP use in main product groups.

For MCCPs flexible PVC is by far the dominant product group. The use of sealants, rubber and coatings are visualized in better detail in Figure 13.

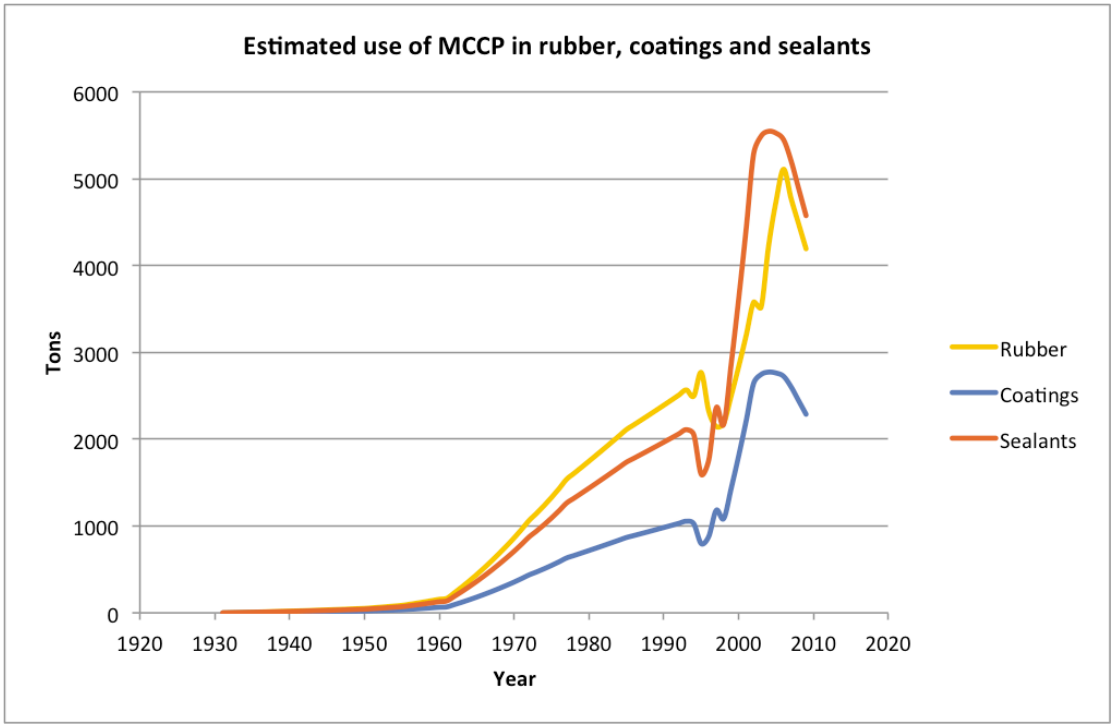

Figure 13: Estimated use of MCCPs in rubber, coatings and sealants.

Data for 2006 on the use of MCCPs, exceeds the amount estimated for total CP use in Europe for the same year. This means that the estimate of the total consumption of CPs in EU are underestimated (Cowi (2010) gives 63691 tons of MCCPs in use, while the estimate amount of total CP use was 63056 tons). The value used for estimating the total consumption of CPs in Europe between 1993 and 2010 (which determines the total MCCP consumption) was an estimate of the production in Europe. This may underestimate the consumption if additional CPs are imported to Europe.

When the given consumption figures for MCCPs in each product group is not truncated by the estimated total consumption of MCCPs, the consumption of MCCPs increase for metalworking fluids, sealants, rubber and coating from 2003 to 2006.

3.4.4 Distribution, emission and waste parameters

For calculating the flows between processes in the defined system, the parameters defined in the mathematical model in chapter 3.3, needs to be quantified. As mentioned earlier, these parameters do not change with time, and the same parameters are used for the calculations of both SCCPs and MCCP.

From all processes there will be losses, either as waste or as emissions to the environment. Several parameters may be used for quantifying the total flow of losses from the system.

Some processes have additional flows that leaves the system in the form of semi finished products or final articles. The semi finished products or articles are used in processes, which are not relevant for determining the stock and waste flows of SCCPs and MCCPs from building and construction. The flows of CPs from these processes are determined by distribution parameters.

Degradation losses of CPs are not included in the estimations.

In the following, distribution parameters and parameters determining losses, are presented for each of the product groups.

3.4.4.1 Sealants

3.4.4.1.1 Losses from production of sealants

All release figures are collected from the EU risk assessment on MCCPs and is based on a survey of CP use (both SCCPs and MCCPs) in the production of sealants in the UK in 1998 (European Chemicals Bureau, 2005). Twenty-two companies were contacted and response was received from ten of them. For the companies where CPs were used, a consistent picture of the industry was obtained.

Losses to water from the production of sealants are likely to be very low/close to zero, as water is not used in the process. Losses to air are also unlikely, as the process is usually carried out under vacuum. A **5% loss** of solid sealant waste could occur due to cleaning of machines. This figure will be used in the calculation as the total loss from the process, as it is the only reported loss.

3.4.4.1.2 Losses from application of sealants

1-part sealants are usually applied to its final site from cartridges (European Chemicals Bureau, 2005). A fraction of the sealant will remain in the cartridge and become waste. A typical cartridge contains 500 grams of sealants and 2-3 cm^3 would typically remain in the cartridge after use. 2-part sealants are usually supplied in tins and filled into cartridges on site.

In a report prepared for the European Commission by BiPro et al. (2011), the waste from application of sealants is assumed to be 5% (BiPRO, Umweltbundesamt, & Enviroplan, 2011). A total **loss of 5%** is therefore used in the calculations.

3.4.4.1.3 Use in building and construction

It has not been found any information on the amount of sealants containing CPs that could be used in building and construction versus other uses.

Reported uses for sealants containing CPs are (BiPRO et al., 2011):

- filling expansion and movement joints
- filling gaps around doors, windows and arches
- filling in double and triple glazed windows for buildings
- filling around automotive windows
- waterproof constructions such as roofs, bridges, culverts, basements, subways and hydropower dams. According to RPA SCCP 2010, dam sealants accounted for 5-20% of the total use of SCCPs in sealants.
- protect from oil and fuel spillages in petrol stations and sewage treatment works.

Reported uses for adhesives containing CPs are (Zarogiannis & Nwaogu, 2010):

- road marking tape
- military uses
- artificial grass

Based on this information it is evident that most sealants and adhesives are used in buildings and constructions. Other likely uses are applications in oil and gas constructions and transport carriers other than automotives (ships, trains, airplanes). These uses are however not confirmed.

There has not been found any additional information to help quantify the ratio of sealants entering these three different end use categories (building and constructions, oil and gas construction, transport carriers). In the estimations it is therefore assumed that **80%** is used in buildings and constructions, while the rest is used in the other end use categories. This assumption has been discussed with Norwegian Building Authority and is believed to be a likely, but crude assumption.

3.4.4.1.4 Emissions from use in buildings and constructions

Volatile, leaching and particulate losses of CPs from use of sealants and adhesives have been estimated for both SCCPs and MCCPs.

Losses of SCCPs over the entire service lifetime of sealants and adhesives are (BRE, IOM Consulting, & Entec, 2008):

0,25% volatilization
0,75% leaching
3,5% particulate loss (range given 2-5%)

Losses of MCCPs over the entire service lifetime of sealants and adhesives are (European Chemicals Bureau, 2005):

0,05% volatilization

0,15% per year leaching
2% particulate loss

3.4.4.2 Coatings

3.4.4.2.1 Losses from Formulation of coatings

The waste and emission estimates presented in the updated EU risk assessment of SCCPs are used for the calculation of losses from this process (European Chemicals Bureau, 2008a). The estimates are collected from an emission scenario document for formulation of solvent-borne coatings developed by the UK Environment Agency in 2003 and apply for both SCCPs and MCCPs (Environment Agency, 2003). The estimated losses are not developed for CPs especially, but for solvent-borne coatings in general. The actual document could not be retrieved, but OECD has made it's own emission scenario document for the coating industry based on the Environment Agency document (OECD, 2009a). This has been consulted whenever needed.

Emissions to air and wastewater are assumed to be negligible for low volatile liquids (European Chemicals Bureau, 2008a). Waste generation is therefore the only relevant source of loss.

The formulation of coatings is done in batches (OECD, 2009a). After each batch of coating, all equipment used in the process is usually cleaned with organic solvents. Losses of coating from cleaning are estimated to be 1% of the total amount raw material used. However, some of the solvents used for cleaning may be reused in the next batch of coating. This is assumed to reduce the amount of waste generated from cleaning of equipment to a total of 0,5%.

For the calculations a **0,5% loss** of coating is used, which implies a 0,5% loss of the input flow of CPs to the process.

Remainders of raw material in packaging and of coatings in filters used in the process, also contributes to the amount of waste generated (OECD, 2009a).This is assumed to be **0,5%** of the raw material input to the process.

3.4.4.2.2 Losses from application of coatings

Losses from the application of coating vary depending on the technic used and if it is industry or the general public that uses the coating (European Chemicals Bureau, 2003). The losses are generally lower when industry is responsible for the application of the coating, as opposed to an average consumer. Losses from spraying are also higher than losses from for example rolling of the coating on to a material.

Losses from the application of coating include, dripping, discarded tools with coating residues and discarded cans with coating residues (European Chemicals

Bureau, 2003). All of these losses are to soil or as waste. Emission estimates varies from source to source. In the EU Technical Guidance Document on risk assessment, a loss of 12% is assumed for uses by private consumers, while the losses from the use by professional painters is given as 4-6%. In the updated EU risk assessment for SCCPs, however, a loss from the application of coatings containing SCCPs ranging from 2,5% to 60,8% is considered (according to (Environment Agency, 2003)). This is also used in a report from the consulting firm BiPro in a report for the European Commission (BiPRO et al., 2011). An average loss of **30%** of the inflow of coatings to the process is therefore used in the calculations.

Losses of SCCPs and MCCPs to air and water are assumed to be minimal, due to the low vapor pressure and water solubility of CPs (European Chemicals Bureau, 2005, 2008a).

3.4.4.2.3 Use in building and construction

There has not been found any information on the fraction of coatings containing CPs, that can be considered used in building and constructions. Zarogiannis and Nwaogu (2010) have summarized reported uses of coatings containing SCCPs.

Reported uses are:

- road marking paints (one of the key applications)
- anti-corrosive coatings for metal surfaces
- swimming pool coatings
- decorative paints for internal and external surfaces
- masonry paints (pliolite is often used as masonry paint for external façades)
- primer for polysulphide expansion joint sealants
- intumescent coatings
- textile printing inks

The EU risk assessment on MCCPs gives a similar picture of the use. The main areas of use is said to be in corrosion and water-resistant coating for steel constructions, ships, containers, industrial flooring, swimming pools, facades and road marking ((European Chemicals Bureau, 2005) according to (Beratergremium für umweltrelevante Altstoffe (BUA), 1992)).

MCCPs are most commonly used in chlorinated rubber and vinyl copolymer-based paints (European Chemicals Bureau, 2005). The chlorinated rubber paints are known to be used in aggressive marine and industrial environments, while the vinyl copolymer-based paints are used most commonly for protection of exterior masonry.

According to Zarogiannis and Nwaogu (2010) road marking paint accounts for around half of the entire SCCP related coating production. Chemical resistant top

coats, coatings for concrete pools, and quick-drying single layer paints are also important products in regard of amounts.

It has also been suggested that CPs could be used for car undercoating. This has however not been confirmed (European Chemicals Bureau, 2005).

Similar to sealants, the main end use of coatings containing CPs is assumed to be in buildings and constructions, oil and gas constructions and transport carriers. However, it seems to be a lesser ratio going to building and construction compared to sealants. A big fraction might go to anti-corrosive coating of oil and gas platforms and ships. It is therefore assumed that **70%** of the paint and coatings containing CPs are used for building and construction. This has been discussed with the Norwegian Building Authority, and is considered a crude assumption.

3.4.4.2.4 Emissions from use in building and construction

Emission factors for calculating the loss of SCCPs and MCCPs from coatings during the service lifetime are given in literature ((BRE et al., 2008), (European Chemicals Bureau, 2005)). When comparing the emission factors for SCCPs and MCCPs they were found to be equal, even though they were presented in different ways.

Emission factors for SCCPs and MCCPs during the service lifetime of coating are:

0,4% volatilization
1% leaching
4,25% particulate loss (range given as 2-6,5%)

3.4.4.3 PVC

3.4.4.3.1 Losses from PVC compounding

There are two main processes relevant for the compounding of PVC containing CPs; dry blending and plastisol blending. The **share of dry blending versus plastisol blending** is given as approximately **70%** and **30%** respectively (OECD, 2009b). Only MCCPs are modeled for use in PVC in this thesis, even though some SCCPs may be used in PVC as well. Emission ratios are therefore collected only for MCCPs.

The emission ratios given in the risk assessment for MCCP is rather detailed and differentiates between MCCPs with 45% chlorine by weight and 52% chlorine by weight, which are the two main MCCPs used as secondary plasticizers in PVC ((European Chemicals Bureau, 2005) according to Euro Chlor). The use of the two MCCPs gives slightly different emissions because of slightly different properties.

Emissions of **MCCPs to air** from **dry blending** is **0,03%** for MCCPs with **45% chlorine by weight** and **0,014%** for MCCPs with **52% chlorine by weight** (European Chemicals Bureau, 2005). These emission factors are based on measurements of the phthalate DEHP being vaporized in the dry blending process and comparisons of the capacity of MCCPs and DEHPs being vaporized. 50% of the losses to air is assumed to condensate and enter waste water (OECD, 2009b).

Emissions from plastisol blending are assumed to be negligible, as the temperatures involved are lower (European Chemicals Bureau, 2005).

In addition to releases from the compounding process, small losses could occur in the raw material handling, by **spillage** (OECD, 2009b). This is proposed to be **0,01%** of the MCCPs transported to the processing site.

Losses of MCCPs from cleaning of equipment have not been mentioned in the reports regarded.

3.4.4.3.2 Distribution between products

There has not been found any satisfactory information on the amounts of MCCPs that are used in different products. A survey prepared by an EU supplier of MCCPs has indicated that 20% of the MCCPs are used in flooring, 11% in wall covering, 6,6% in coated products, films and sheets and 17% in cables (European Chemicals Bureau, 2005). These figures are however incomplete as they only cover 55,6% of the MCCPs used. It is also uncertain if they reflect the average EU use.

It is indicated by several sources that MCCPs often are used as secondary plasticizer together with the phthalate DEHP. Until more accurate data is available for the use of CPs in flexible PVC products, it is assumed that the MCCPs follow the use pattern of the phthalate DEHP. This has been done to some extent in the EU risk assessment of MCCPs (2005) and is also assumed in the Norwegian survey on MCCP use in products ((Cowi, 2010), (European Chemicals Bureau, 2005).

The Norwegian survey on MCCP use in products summarized the tonnage of DEHP used in PVC products marketed in EU and calculated the percentage of use in each end-product, according to information from COWI et al. (COWI A/S, IOM, & Entec, 2009).

Product group	Consumption Tonnes	Percentage of total
Flooring, wall covering, roofing	47,600	18
Wires and cables	64,100	24
Film/sheet and coated products made by calendering	44,000	16
Coated fabric and other products from plastisol	43,800	16
Hoses and profiles	34,700	13
Moulded products	3,000	1
Shoe soles	19,400	7
Other polymer applications	12,300	5
Total	268,900	100

Table 2: Use of DEHP in PVC. Source: ((Cowi, 2010) *according to (COWI A/S et al., 2009))*

The products assumed to be relevant for end-use in building and construction is "flooring, wall covering, roofing", "wires and cables", "film/sheet and coated products made by calendaring" and "hoses and profiles". In addition "moulded products" and "other polymere applications" could be used in building and construction. The percentages given in the table above are used as distribution parameters in the calculations. The percentage of use in moulded products" and "other polymere applications" are added to represent flows and stocks of "other PVC products" in the defined system.

3.4.4.3.3 Losses from production of PVC products

Emissions to air from the production of PVC products are calculated based on if it is an open, partially open or closed production system that is used. For the production of flexible PVC, the **breakdown between open, partially open or closed systems** is **26%, 49%** and **25%** respectively ((European Chemicals Bureau, 2005) according to UCD 1998).

Emissions to air from the use of MCCPs with **45% chlorine by weight in open, partially open or closed systems** are **1,5%, 0,3%** and **0,3%** respectively (European Chemicals Bureau, 2005). For MCCPs with **52% chlorine by weight** the emission ratios are **0,7% for open systems** and **0,14% for partially or closed systems.** These figures assume that no air treatment equipment is used. Air treatment equipment is now used in most production processes, but the loss of MCCPs from the system will be the same even though the emissions to air are reduced, as MCCPs are captured in filters or incinerators and thus destroyed or collected as waste.

The **distribution of MCCPs with 45% chlorine by weight versus 52% chlorine by weight** is given as approximately **1/3** and **2/3** respectively (European Chemicals Bureau, 2005).

3.4.4.3.4 Losses from installation of PVC products

It is assumed to be a **5% loss** from the installation of the PVC products at their final use site.

3.4.4.3.5 Use in building and construction

It has not been possible to find out how much flooring, cables, sheets or hoses that are used in building and construction contra in other uses, e.g. cars, ships, electric infrastructure, consumer goods and oil and gas installations. The fractions of the products ending up in building and construction are therefore assumed, based on knowledge of where they are used.

Flooring, wallpaper, roofing

Buildings and constructions and transport carriages are believed to be the two categories where flooring, wallpaper and roofing is used. Since CPs improves the fire resistance of PVC products, it is likely that such products are used in public (and private) transport applications, where fire requirements are high.

The fraction of flooring, wallpaper and roofing used in building and construction is assumed to be **90%**.

Sheets and films

Sheets and films are one of the main categories of use for DEHP, and are also assumed to be so for CPs. It is not very clear what sheets and films are and what they are used for, but they seem to be used mostly in consumer products; upholstery, packaging, luggage, clothing, stationary products (European Chemicals Bureau, 2008b).

It is however believed that some sheets and films are used in buildings and constructions. Thin plastic sheets are used in buildings as vapor barriers, wind barriers, radon barriers, bathroom membranes, roofing membranes, etc. (Edvardsen & Ramstad, 2007). It is not known to what degree these contain CPs. Foils for vapor barrier has been suggested as a possible use, but the sole material for this use is said to be polyethylene and not PVC. PVC is however mentioned for use in roofing membranes.

Even though it has not been reported that sheets and films containing CPs are used in building and construction, the building and construction sector is a large industrial sector and flexible PVC is used for many applications. It is therefore assumed that **50%** of the films and sheets containing CPs are used in building and construction.

Wire and cable

Wires and cables are today used in innumerable products and applications and information on where cables containing CPs could be used is scarce. According to a report written for the Norwegian Agency for Building Quality, all different

kinds of plastics are used for insulating wire and cables ((Hjellnes Consult, 2011) according to COWI Denmark). However, PVC is the most common, and DEHP and MCCPs has traditionally been added. Cables containing CPs could be cables for applications in buildings or used in electrical and electronic equipment. Underground electric transmission cables are also to some extent insulated with PVC (according to a manufacturer of cabels) and could potentially contain CPs (Nexans Norway AS, 2009). In a mapping conducted by COWI (2009) around 15% of the cable and wires containing DEHPs are assumed to be used in cable and wire covered by soil (COWI A/S et al., 2009). 81% is assumed to be used in indoor applications, however it is not clear what kind of indoor applications this could represent.

For the calculations it is assumed that **20%** of the wires and cables used for indoor applications are used in buildings and constructions.

Hoses and profiles

Hoses and profiles are assumed to be used in buildings and constructions, oil and gas installations, transport carriers and consumer goods. It is believed that flexible hoses are not used in building and constructions. Profiles can be used in building and construction, but also in transport carriers. In total it is therefore assumed that **30%** of the hoses and profiles are used in buildings and constructions.

Other products

It is assumed that **50%** of the other products (unknown which kind), eg. "moulded products" and "other polymere applications" in table…, are used in building and construction.

3.4.4.3.6 Emissions from use in building and constructions

Because of the diversity of PVC products used in building and construction, the calculations of losses are more complex than for sealants and coatings. Parameters determining losses have collected for each of the PVC product categories and emission factors differ for indoor- and outdoor uses.

The emission estimation follows an alternative approach to the main emission calculation presented in the EU risk assessment of MCCPs, given in appendix F in the report. The alternative approach has been used to be able to cover all losses of MCCPs and not only losses to water and air (which are minimal).

The alternative approach is based on the fact that emissions are dependent on the surface area of the products. Based on assumptions of the density of the PVC products and the thickness of them, the surface area could be calculated from the estimated tonnage in use.

The **density of PVC** (flooring, wall covering, films and sheets, hoses and profiles) was given as **1300 kg/m^3** (European Chemicals Bureau, 2005). The thicknesses of the different products were assumed in the risk assessment to be:

Thickness of flooring	2 mm
Thickness of wall covering	0,5 mm
Thickness of roofing	1,5 mm
Thickness of films and sheets	1 mm
Thickness of cable	1,5 mm
Thickness of hoses and profiles	3 mm

For the category **"other products"** a **thickness of 2mm** was assumed.

To figure out the individual tonnages of flooring, wall covering and roofing, which where all lumped together in one category, information on the amounts of DEHP used for each of these products was used (COWI A/S et al., 2009). **69,3%** is assumed to be **flooring**, **23,1%** to be **wall covering** and **7,6%** to be **roofing**.

The emissions factor for MCCPs from PVC to **indoor air** was given as; **15,8 mg/m^2 per year** ((European Chemicals Bureau, 2005), (Campbell & McConnell, 1980)). The lifetimes of the products were also needed for calculating the losses over the entire lifetime of the products. These are presented in section 3.4.5.

Flooring, wall covering and sheets and films where assumed to be used only indoor. Roofing is used only outdoors. For each of the other products the amount used indoor and outdoor needed to be determined. Information of this was also found for products containing DEHP (COWI A/S et al., 2009). Based on this it was assumed that **81%** of the **wire and cables**, **89%** of **hoses and profiles** and **85%** of **other products** were **used indoor.**

Leaching losses indoor where only assumed to be relevant for flooring. **Leaching from washing PVC flooring** (diffusion loss - not abrasion losses) was given as **0,0005 g/m^2 per cleaning event** (European Chemicals Bureau, 2005). This assumes that MCCP have a similar potential for leaching as DEHP. The flooring was assumed to be cleaned every tenth day.

It has been reports claiming that some PVC flooring is coated with a polyurethane layer to increase the wear and stain resistance. This decreases the volatilization and leaching of MCCPs. **50%** of the flooring was assumed to be **coated with polyurethane** and **reduce** the **emissions by a factor of ten** (European Chemicals Bureau, 2005).

An abrasion loss was also calculated for flooring where walking is frequent. This was assumed to be the case for **50% of the flooring** and an **abrasion loss of 6,25%** was assumed (European Chemicals Bureau, 2005).

Emission factors for outdoor applications included both volatilization, leaching and degradation losses. The emission factors given were:

0,28 g/m^2 per year for uncovered roofing
0,65 g/m^2 per year for gravel covered roofing
0,28 g/m^2 per year for hoses and profiles

The emission factor for **"other products"** was assumed to be **0,28 g/m^2 per year.** The ratio for **uncovered roofing** was not given, but assumed to be **20%** as well.

In addition a particulate loss from **outdoors applications** were calculated for roofing, and hoses and profiles. This loss was given as 2-10% depending on the wear. In the calculations it is assumed that **roofing, and hoses and profiles** have a low wear rate and a **particulate loss of 2%** is used.

3.4.4.4 Rubber and other polymeres

3.4.4.4.1 Compounding

As for PVC compounding, the CPs used are handled in bulk using enclosed storage systems (European Chemicals Bureau, 2005). This minimizes losses from **spillage**, but a **0,01%** loss is considered in the calculations.

CPs used in rubber have a higher chlorine content than those used in PVC, which results in a lower volatility of the substance during compounding (European Chemicals Bureau, 2005). The volatility is similar or slightly lower than the volatility of the phthalate DEHP. **Releases to air** are therefore given as **0,01%** (same as for DEHP). 50% of these emissions will eventually enter wastewater.

3.4.4.4.2 Distribution between products

There has not been found any satisfying information on the amount of CPs used for different rubber products. It is however clear that most of the rubber containing SCCPs has been used in conveyor belts for the underground coal mining industry (or in other environments vulnerable for fire). This use is assumed to account for 75-90% of the consumption of SCCPs in rubber, by Zarogiannis and Nwaogu (2010).

Other reported uses of SCCPs in rubber is in gaskets and hoses and possibly in car carpets and other vehicle accessories (except tyres).(Zarogiannis & Nwaogu, 2010). CPs in general has also been used in rubber shoe soles and industrial sheeting.

In a survey developed by Entec, disseminated by the European Association of the Rubber Industry (answered by companies in Germany representing 40% of the EU market share), MCCPs were found to be used in conveyor belts and tubes for

compressed air in the mining industry; bellows for buses, metros and trains and; profiles for fireproofed doors (Entec UK Limited, 2008).

In addition to being used in rubber, MCCPs are reported to be used in polyurethane, especially in rigid foams and one-component foams (used for insulation, among others) (Entec UK Limited, 2008).

Based on the information given above, the use of MCCPs and SCCPs seems fairly similar, but there are no quantifiable estimations on the use of MCCPs that can confirm it. It is also a lack of information on the distribution of SCCPs among other products than conveyor belts.

Bipro et al. (2011) assumes that 80% of the SCCPs are used in conveyor belts (according to Zarogiannis and Nwaogu, 2010) and that the remaining 20% is distributed equally between gaskets and hoses. However, it seems likely that SCCPs are used in other products as well. For the calculation, 80% is assumed to be used in conveyor belts and the remaining **20%** in other products. This distribution is used for both SCCPs and MCCPs.

3.4.4.4.3 Losses from production of products

Nearly all rubber compounds containing flame-retardants are said to be converted in closed systems ((European Chemicals Bureau, 2005) according to UCD 1998). The losses are assumed to be similar as for losses of DEHP in closed conversion systems and are given as **0,1%**. 50% of these losses will eventually enter wastewater.

3.4.4.4.4 Losses from installation of products

A **5%** loss as cut-offs during installation is assumed.

3.4.4.4.5 Use in buildings and constructions

Conveyor belts are not relevant for use in building and construction. The remaining 20% of CPs in other rubber products is believed to be used in buildings and constructions, transport carriers and machines and in the oil and gas industry.

Because of the lack of information, it is assumed that the remaining 20% are distributed equally between the three end use categories, e.g. almost **7%** is used in building and construction.

3.4.4.4.6 Losses from use in buildings and constructions

Volatile losses from rubber and other polymers (except PVC) during the service life, is given as **0,05%** for MCCPs (European Chemicals Bureau, 2005). Losses to water is assumed negligible. A **particulate loss of 2%** was given for **outdoor applications**. It is not known how big amounts of rubber products that are **used outdoors,** a similar fraction as for PVC is therefore used; **15%.** The same losses are assumed for SCCPs.

3.4.5 LIFETIME AND LIFETIME DISTRIBUTIONS

The lifetime distribution L(t',t) of a product determines the probability of a product entering the use phase at time t' to exits use at time t. The lifetime distributions of products used in building and construction is not known. Different functions could be used for estimating the lifetime distribution. For simplicity the normal distribution function is used. As the lifetime of a product cannot be zero, the probability given by the lifetime distribution should approach zero for the first year of use. If the standard deviation is large compared to the mean lifetime of the product, this may not be the case. For such circumstances the Weibull distribution function can be used in stead, as it always starts at zero. For coatings the standard deviation was assumed to be high compared to the average lifetime, the Weibull distribution function has therefore been used for estimating the lifetime distribution of coatings.

The lifetime of sealants is assumed to be 20 years, as in the EU risk assessment reports for SCCPs and MCCPs (European Chemicals Bureau, 2000, 2005). The standard deviation was assumed to be 7 years.

For PVC products, several lifetime estimates was found in the EU risk assessment report for MCCPs:

PVC flooring	20 years
PVC wall covering	7 years
PVC films and sheets	7 years
PVC cabel and wire	30 years
PVC hoses and profiles	10 years
PVC roofing material	20 years

For the categories "flooring, wall covering, roofing" and "other products" the average lifetime was assumed to be 15 years (based on the lifetimes given above). For the other categories, the above given lifetimes were used. The assumed standard deviations were:

Flooring, wall covering roofing	5 years
Cable and wire	10 years
Sheets and films	2 years
Hoses and profiles	3 years
Other products	5 years

For rubber products the lifetime and standard deviation was assumed to be similar as for PVC products, 15 years and 5 years respectively.

For coatings the Weibull parameters α and β was assumed to be 2,5 and 23 respectively. This gives the highest probability for the coating to exit use after approximately 20 years.

3.5 UNCERTAINTIES

A number of assumptions and estimations have been made to be able to quantify the system.

The framework for the analysis, is a simplification of reality and limits the accuracy. It was assumed that all parameters, except the inflow parameter α_{ty}, were constant. However, it is very likely that there have been variations in use patterns, emissions and waste production from processes throughout the approx. 80 years CPs have been used. Such data is, however, very difficult to obtain and is rarely used in MFA. The system in it's self, it's processes and interactions, may also have changed, but this is overlooked for the same reason. Additional processes, which have not been described in literature, could also be overlooked.

The same system and parameters (except the inflow) was used for quantifying stocks and flows of both SCCPs and MCCPs. This is also a simplification, but again data did not allow for a more detailed analysis. The processes involved are similar for both SCCPs and MCCPs, but the distribution parameters and emissions are not necessarily the same.

The major uncertainty is however the input flows to the system. Estimates of the European CP consumption and distributions between chain-lengths are incorporated in the estimates of the yearly input flows to the production of each of the product groups. This means that a mistake in the estimate of the European consumption will be carried on to the other estimates. As mentioned earlier, it is likely that the last data point used for the estimate of the European consumption is an underestimation. The data point is a production figure and not a consumption figure, and the data from literature giving the consumption of MCCPs exceeds the total estimated European CP consumption. There was however no other estimate of the European consumption between 1993 and today and the consumption figure was needed for estimating the distribution of chain-lengths (which again was needed for the estimate of historic use of SCCPs and MCCPs within product groups). The data on use of SCCPs and MCCPs for the production of the different product groups is however relatively detailed from 1994 to present. This data could therefore have been used independently of the overall European consumption figure. It was however decided to stick to the principal of a top-down study and adjust the values to match the overall European consumption. In addition, there was not data of use within product groups for all years between 1994 to present and interpolation between data points was applied. When adding up interpolated values, the total may be out of proportion. To conclude, it should be emphasized that the input flows to the

system is most likely underestimated for the last 20 years (which are the most important for determining the future waste flow).

It should also be stated that there most likely has been changes in the distribution of consumption between product groups prior to 1994, which has not been taken into account.

A general challenge when collecting data, was the unclear definitions given in literature of. It was not always clear what the data found represented. Use, consumption, sales and production figures where sometimes confused. And the region that the data represented could be blurred (e.g. western Europe, Europe excluding eastern Europe, etc.).

Data on imports and exports of CPs or goods containing CPs, was scarce and import and export flows to the processes in the system have not been included. This is a major lack of the model. Because of scarce data, production and consumption figures have also been used interchangeably. This can explain why the MCCP consumption exceeds the estimated total European CP consumption. Without including imports and exports in the system, it is uncertain how well the calculated stocks and flows represent the actual consumption in EU.

The distribution parameters determining the fraction of the products used in building and construction are believed to be the second most uncertain parameters. The values are merely based on incomprehensive information of where products containing CPs are used and qualified suggestions of how much of these uses are in building and construction. Although the assumptions are discussed with the Norwegian Building Authority, they had no specialized qualification for informing the assumptions besides their general knowledge on buildings and constructions.

For PVC and rubber, addition assumptions where made for determining how much CPs where contained in the different products. For PVC products it was assumed that CPs follow the use pattern of DEHP. This is a very crude assumption to make, as DEHP and CPs will not always be used together. Especially in later years, the use pattern might have changed, as DEHP has been replaced by other phthalates that are less compatible with CPs.

However, this was believed to be the best assumption, based on the available data. Very little data was available for rubber. The only possible differentiation between rubber products was between conveyor belts and other rubber products. However, the assumed 80% going to conveyor belts seems to be relatively certain.

For determining the waste flows and stocks of the individual products in building and construction, the lifetime distribution is decisive. To decide on one

average lifetime for use in both buildings and constructions and all the possible applications therein, is not a simple task. This was especially difficult for sealants and coatings. Some sealants may be removed when a building and construction is refurbished (e.g. around windows or doors), while others will remain in the building or construction until it is demolished (e.g sealants used between concrete slabs). Even when a choice has been made between determining the lifetime according to refurbishment or demolition, the question of what a joint average lifetime (or refurbishing rate) of a building and construction is, will arise. Use in building and construction is defined very widely in this thesis and contains everything from a residential building to a hydropower dam.

Coating was a generally difficult product group to treat. As for sealants, the lifetime was hard to determine. The coating can flake off after just a couple of years or a new layer of coating can be applied, so that the coating sticks to the construction until it is demolished. To adjust the lifetime distribution for this, a larger dispersion was assumed. In addition it was not clear how much of the coating could be regarded as building and construction waste, and how much that should be regarded as emissions. The parameters found for calculating the emissions from coatings in the use phase, did not include abrasion losses. All abrasion losses are therefore defined as building and construction waste, even though they may not be collected and treated as waste.

There is also some uncertainty related to the parameters determining the losses from the processes in the system. The parameters are collected from risk assessments, where the purpose is to determine the loss of the substance to environmental compartments and not to estimate how much of the initial amount consumed that reaches end use in a product. This means that estimates of waste production from a processes is not a priority. For some of the processes information on waste production was given, but this was generally not the case. It is therefore uncertain if the parameters used for estimating the losses comprises all losses from the process, or if additional waste losses should have been included. When no data was given on waste amounts, but it was obvious that waste would be produced during the process (as for installation of PVC and rubber products) – a ratio determining waste amounts was assumed.

Information on rubber processing and resulting losses from the processes was scarce. It was assumed to be similar to PVC processing, but according to a document developed by OECD, more water seems to be used in the processing of rubber compared to PVC (OECD, 2004). It is therefore likely that there will be some losses to water from the process, but this is not included in the calculations.

Many of the parameters determining losses are worst-case losses. The typical losses to air and water may therefore be smaller than calculated. On the other

hand, the total losses are believed to be greater than what is calculated, since waste production is not believed to be included for all processes.

Degradation losses are not included in the study. No information has been available for calculating this. Degradation losses could decrease the stock and waste flows of CPs from building and construction.

To conclude; although a lot of information was collected for determining the losses from the processes, the losses can generally not be viewed as comprehensive.

3.6 SCENARIOS

For predicting the future stock and waste flows from building and construction, one future scenario has been developed for SCCPs and three for MCCPs.

The use of SCCPs are already approaching zero. The only likely future scenario was therefore believed to be a continued decrease of it's use. The use was therefore assumed to reach zero in 2020.

For MCCPs the use in PVC decreases, but the use in sealants, coatings and rubber seems to be increasing. Scenario 0, assumes that the consumption of MCCPs will continue as in 2009 for all product groups. Scenario 1 assumes that the use of MCCPs in all product groups will reach zero in 2040. Lastly, scenario 2 assumes increased use of MCCPs in sealants, rubber and coating and continued use as in year 2009 for PVC.

The figures depicted below, shows the total input to use in building and construction according to the different scenarios. The figures are stacked area graphs, which means that the inputs of each product group has been put on top of each other to show the total input to buildings and constructions.

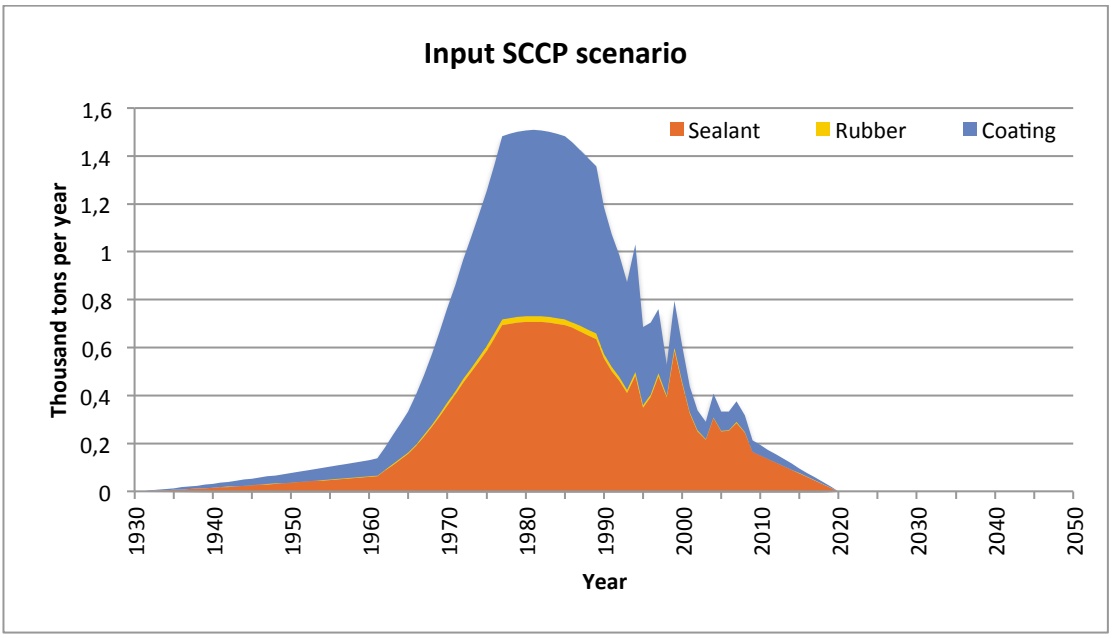

Figure 14: Scenario for input of SCCPs. Stacked area graph.

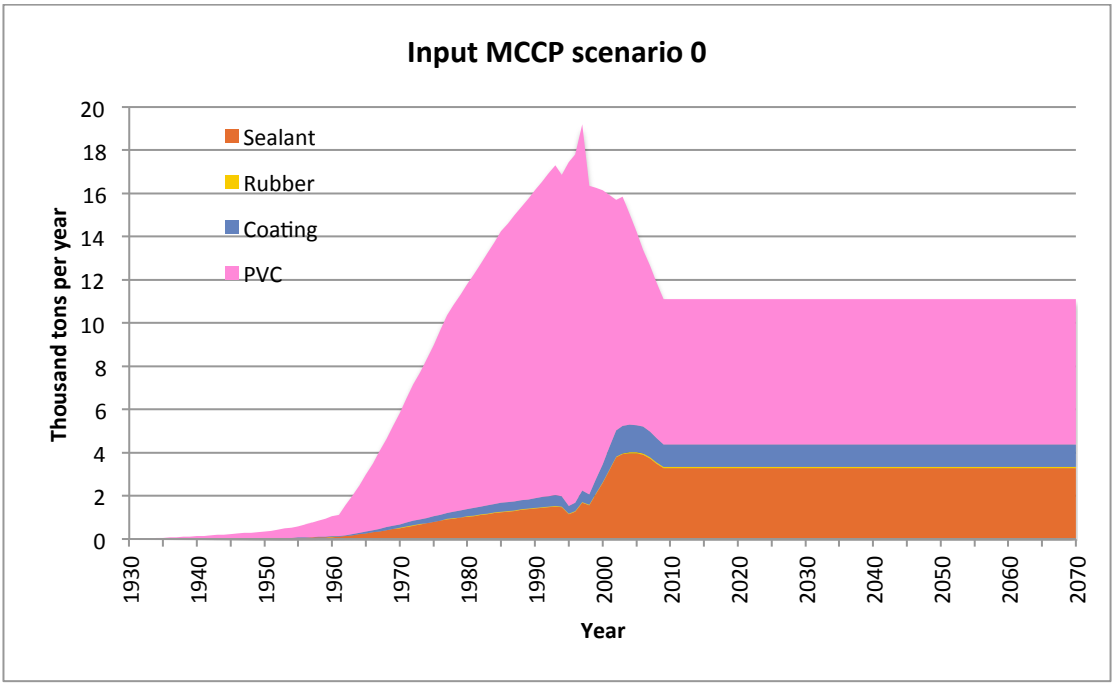

Figure 15: Scenario 0 for input of MCCPs. Stacked area graph.

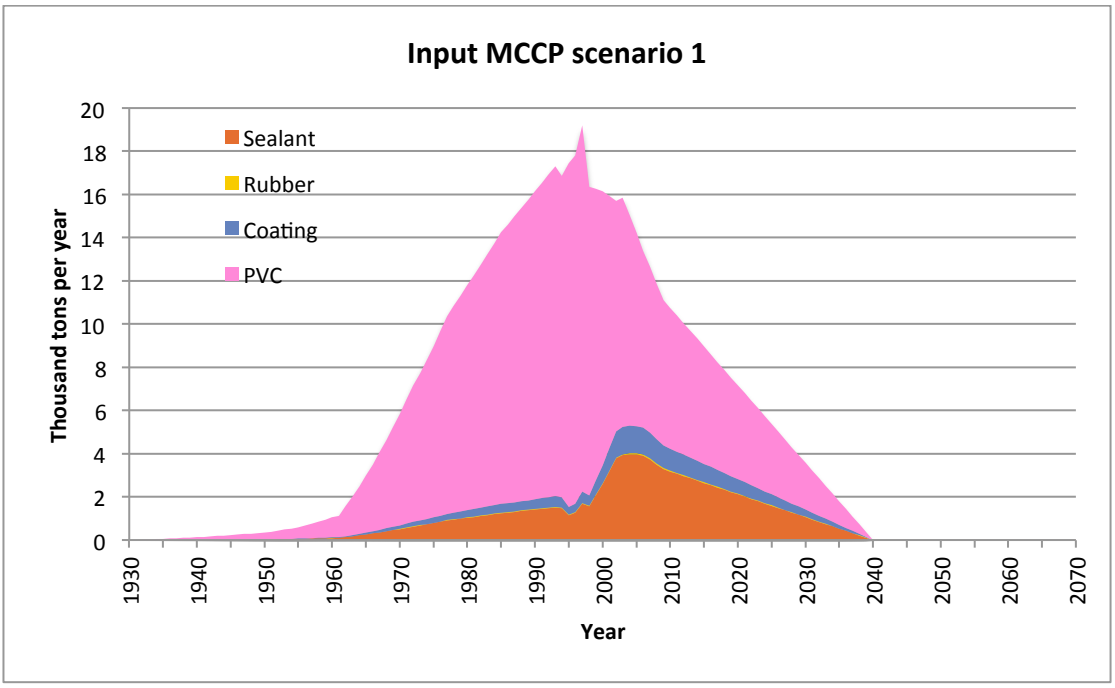

Figure 16: Scenario 1 for input of MCCPs. Stacked area graph.

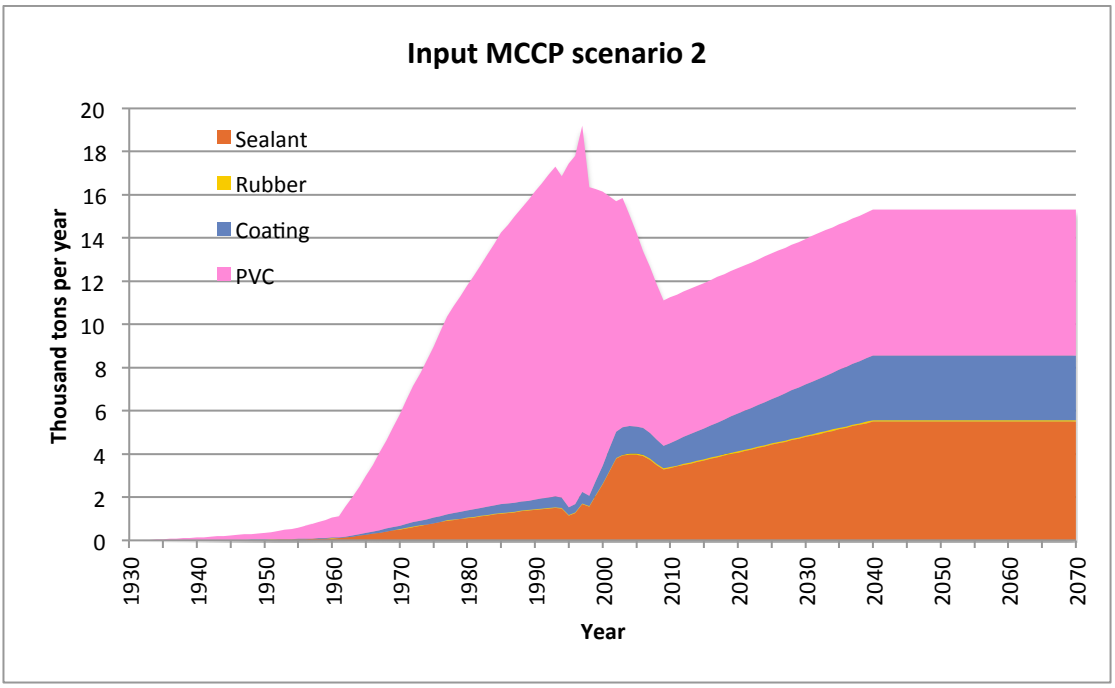

Figure 17: Scenario 2 for input of MCCPs. Stacked area graph.

4 RESULTS

4.1 SCCPs

Figure 20 (shown below) depicts the total, accumulated stock of SCCP in building and construction with an assumed continued decrease in consumption from 2009 to 2020 (stacked area graph). The stock of SCCPs in building and construction can be seen to have been on it's largest in 1992 with approximately 25 100 tons. Today (year 2012), the stock is around 12 000 tons.

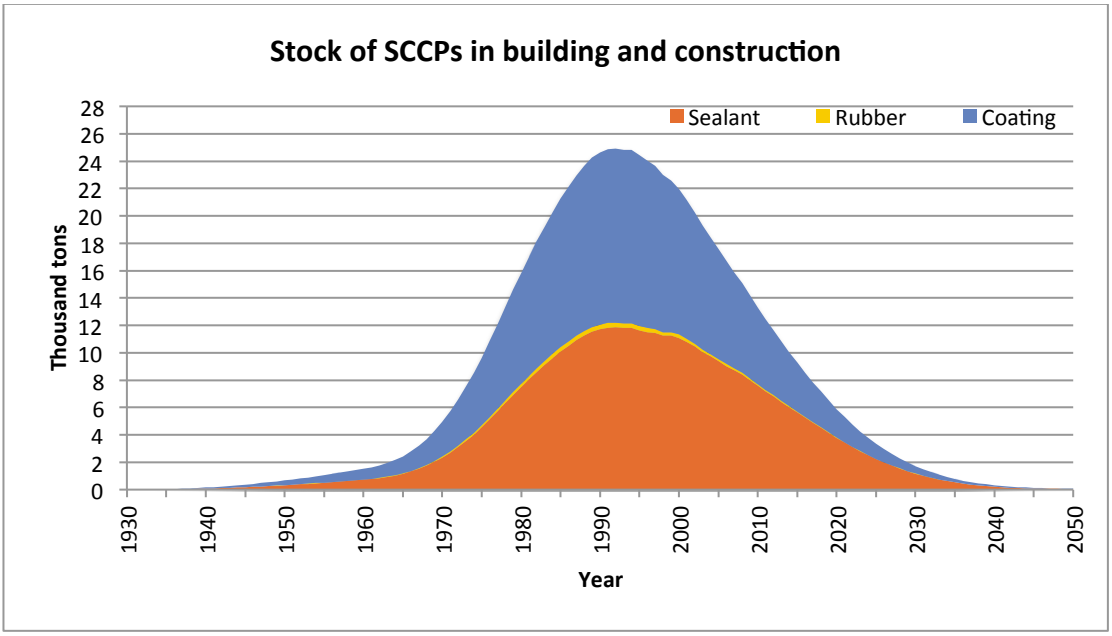

Figure 18: Stock of SCCPs in building and construction. Stacked area graph.

The amount of SCCP in sealants and coatings clearly constitute the largest part of the stock, while SCCPs in rubber is negligible. The stock of SCCP in sealants and coating follows a similar pattern, but the stock in coatings decreases slightly more rapid than the stock in sealants.

Figure 21(shown below) shows the waste of SCCPs per year from sealants, rubber and coatings used in buildings and constructions (stacked area graph, in colors). The total input of SCCPs from all three product groups and total stock of

SCCPs in all three product groups is also plotted for comparison (line plots). The waste flow can be seen to have been approximately 1300 tons per year at it's maximum (year 2002). The maximum waste flow occurs 21 years after the maximum input of SCCPs to the stock (year 1981). Today (year 2012) the waste flow is almost 1000 tons per year.

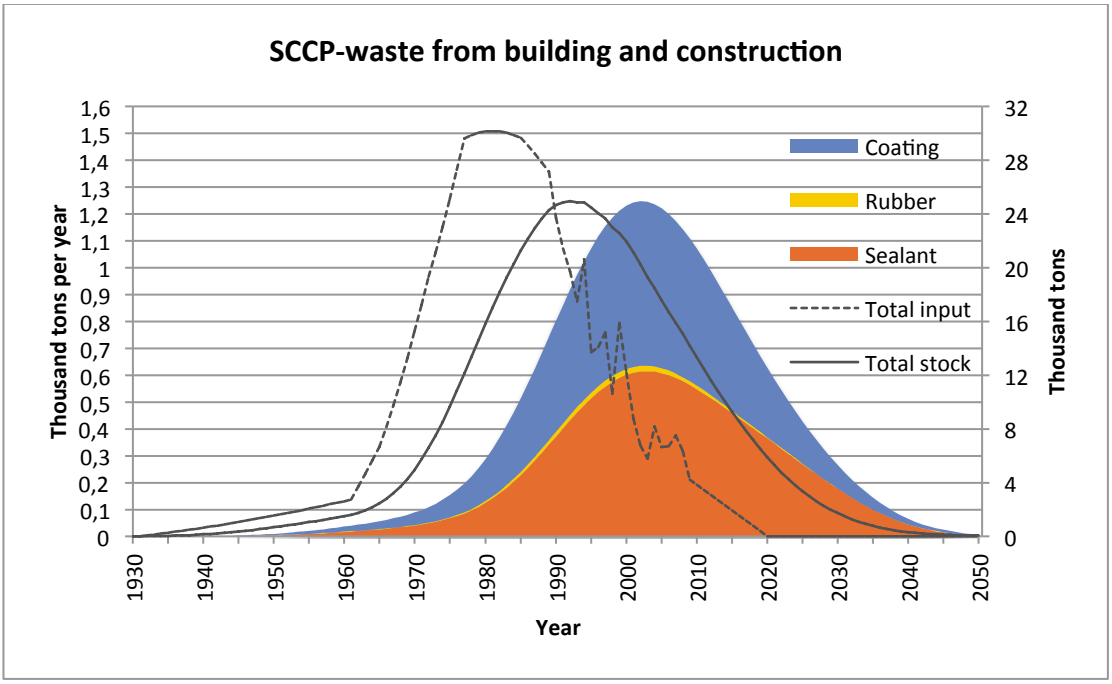

Figure 19: SCCP-waste from building and construction. Stacked area graph (lines are not stacked).

4.2 MCCPs

Figure 22 (shown below) shows the stock of MCCPs in sealants, coatings, PVC and rubber used in buildings and constructions according to scenario 0 – continued consumption as in 2009 (stacked area graph).

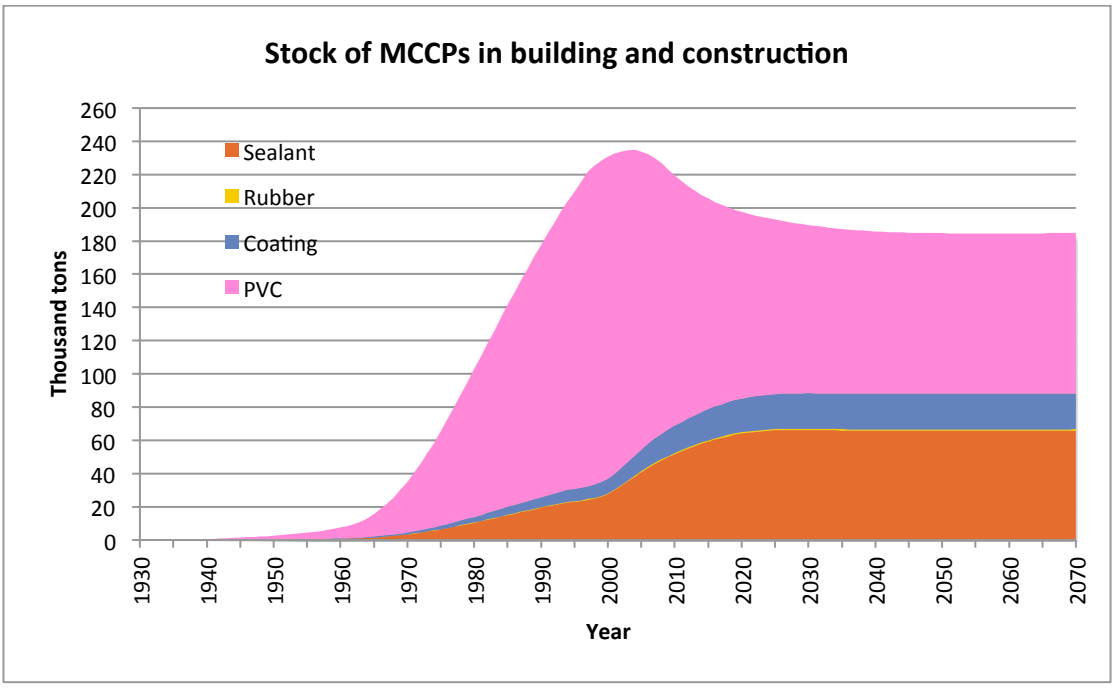

Figure 20: Scenario 0, Stock of MCCPs in building and construction. Stacked area graph.

The stock of MCCP in PVC products is clearly dominant. However, the stock of MCCP in PVC has decreased since year 2000, while the stocks in coatings, rubber and sealants have increased. The stock of MCCPs in rubber is negligible (660 tons in 2009).

The maximum stock of MCCPs in building and construction amounted to almost 235 000 tons in 2004 – almost ten times the stock of SCCPs. Today (year 2012) the stock is estimated to be around 213 000 tons. If the use of MCCPs continues as in 2009, the stock will stabilize at around 185 000 tons within 2040.

Figure 23 shows the stock of MCCPs in the different PVC products used in buildings and constructions. MCCPs in flooring, wall covering and roofing constitute the largest amount of MCCPs in PVC products, followed by wire & cable and films & sheets.

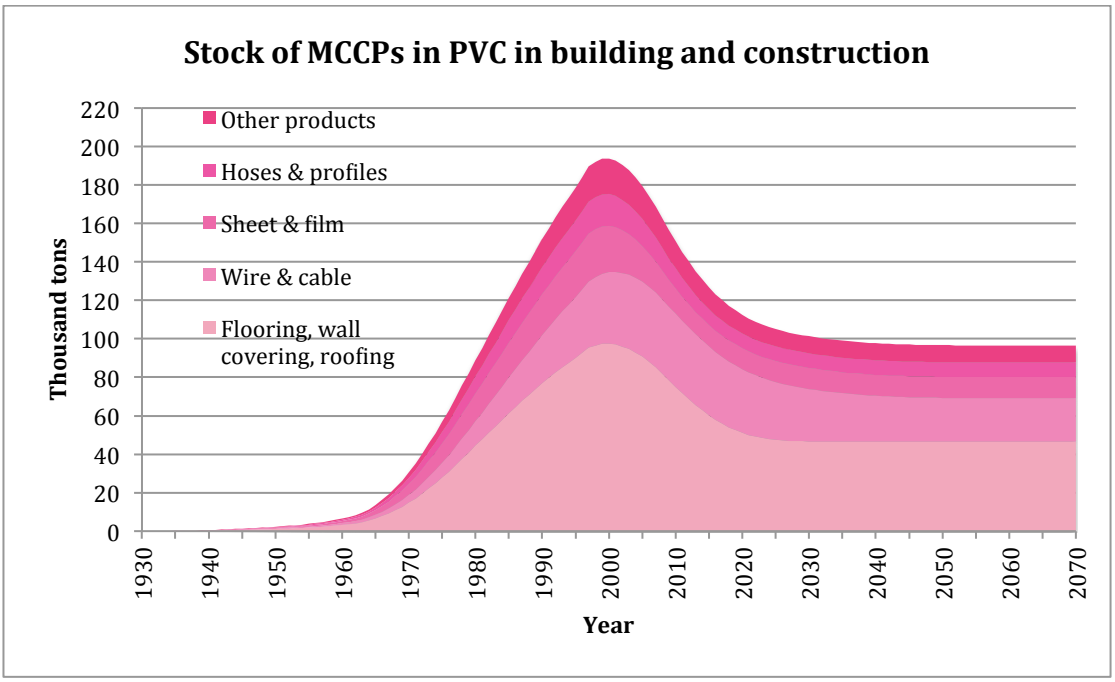

Figure 21: Scenario 0, Stock of MCCPs in PVC in building and construction. Stacked area graph.

Figure 24 depicts the MCCPs waste flow per year from sealants, coating, rubber and PVC (stacked area graph, in colors). The total input and stock is also visualized in the graph for comparison (line plots). The waste flow of MCCPs was at it's maximum in year 2006 with around 15 000 tons. Today it is around 14 000 tons. If the consumption of MCCPs continues as in year 2009, the amount of MCCP-waste will decrease until it reaches an equilibrium at approximately 11 000 tons from about year 2040.

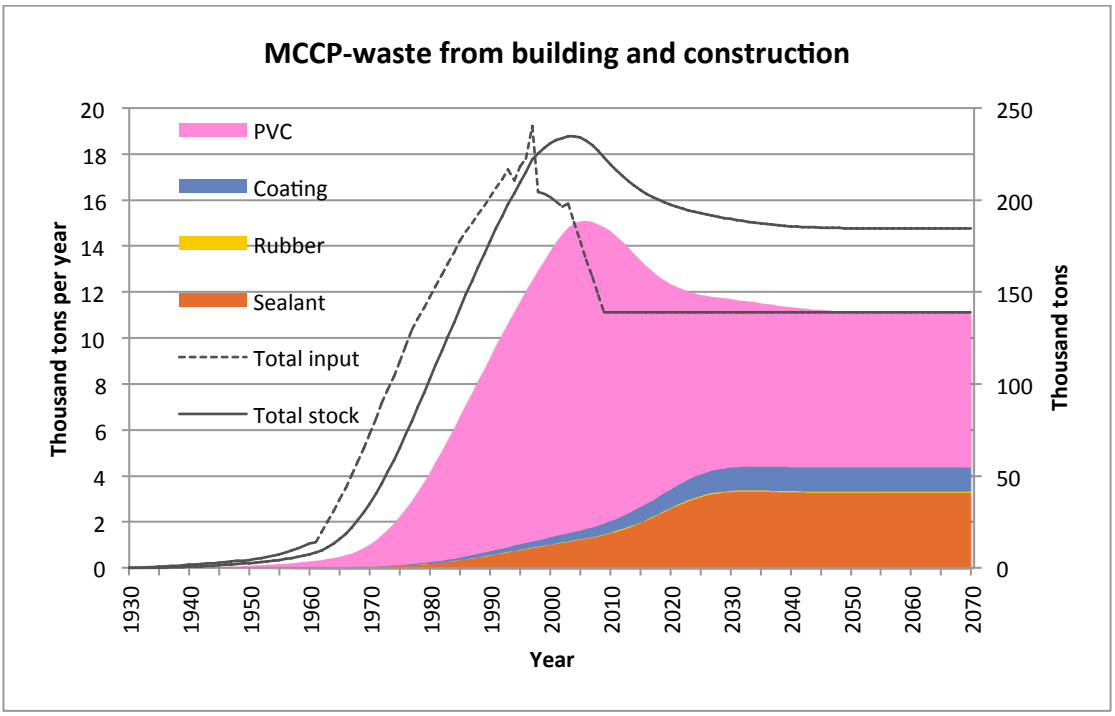

Figure 22: Scenario 0, MCCP-waste from building and construction. Stacked area graph. Lines are not stacked.

The maximum amount of MCCP-waste occurs 9 years after the maximum input to building and construction (1997). This seems a bit short. There was a peak in the input in 1997, however the big amount of input can be seen to have entered some years earlier.

Figure 25 shows the waste flow of MCCP in PVC products (stacked area graph, in colors). The total input flow of all PVC products and the total stock of all PVC products is also visualized (line plots). Although the stock of wire & cable is greater than sheet & film, the annual waste flow of sheet and film surpasses the waste flow of wire & cable. Flooring, wall covering and roofing still constitute the biggest fraction of the MCCP waste from PVC products.

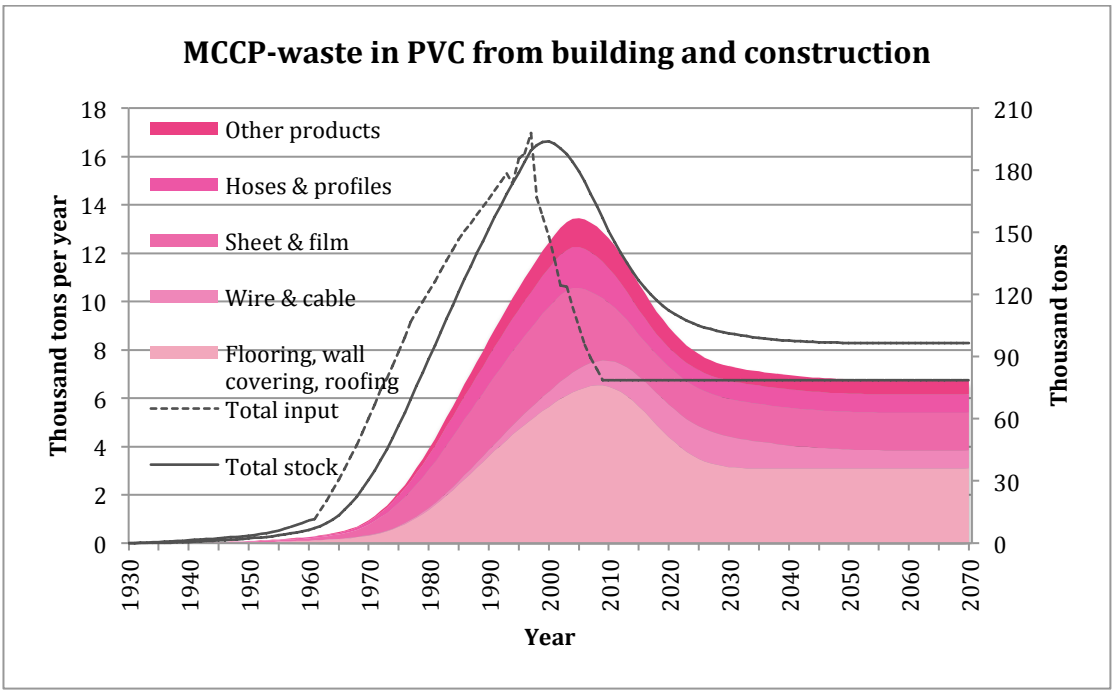

Figure 23: Scenario 0, MCCP-waste in PVC from building and construction. Stacked area graph. Lines are not stacked.

Scenario 1 and 2 are identical to scenario 0 from the year 1930 til the year 2009. The estimates of today's stock and waste flow of MCCPs according to scenario 1 and 2 are therefore similar to scenario 0 and will not be commented on. The long-term development is however different.

Figure 26 and 27 shows the stock and waste flow according to scenario 1. The stock and waste flow approaches zero in year 2070 – 30 years after the input is assumed to cease.

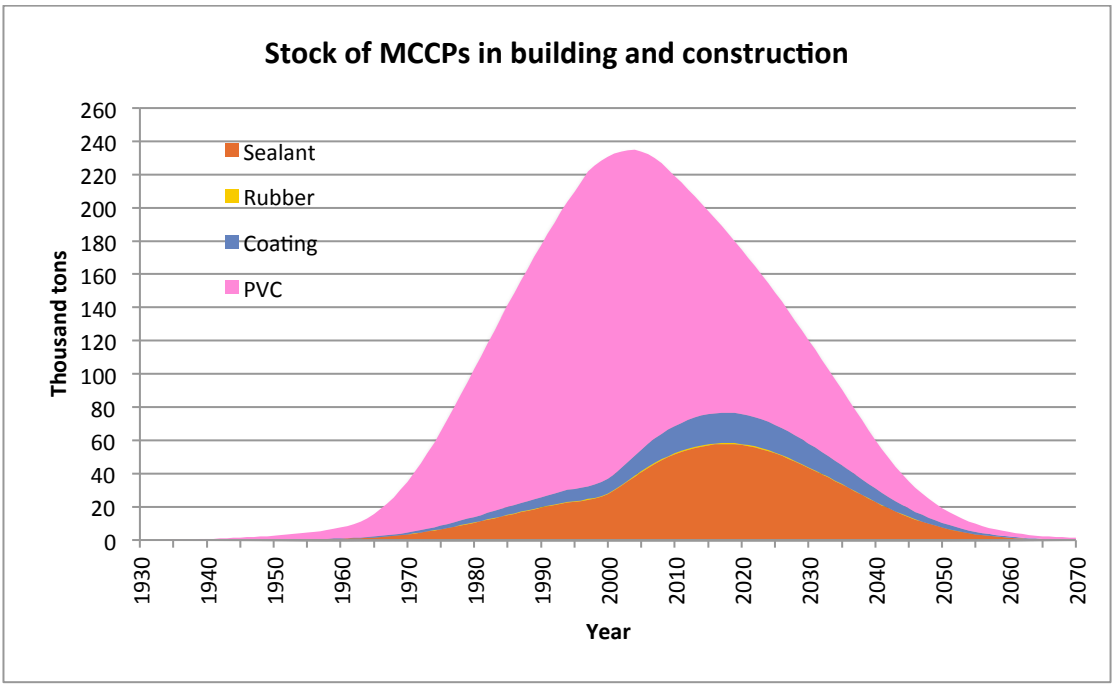

Figure 24: Scenario 1, Stock of MCCPs in building and construction. Stacked area graph.

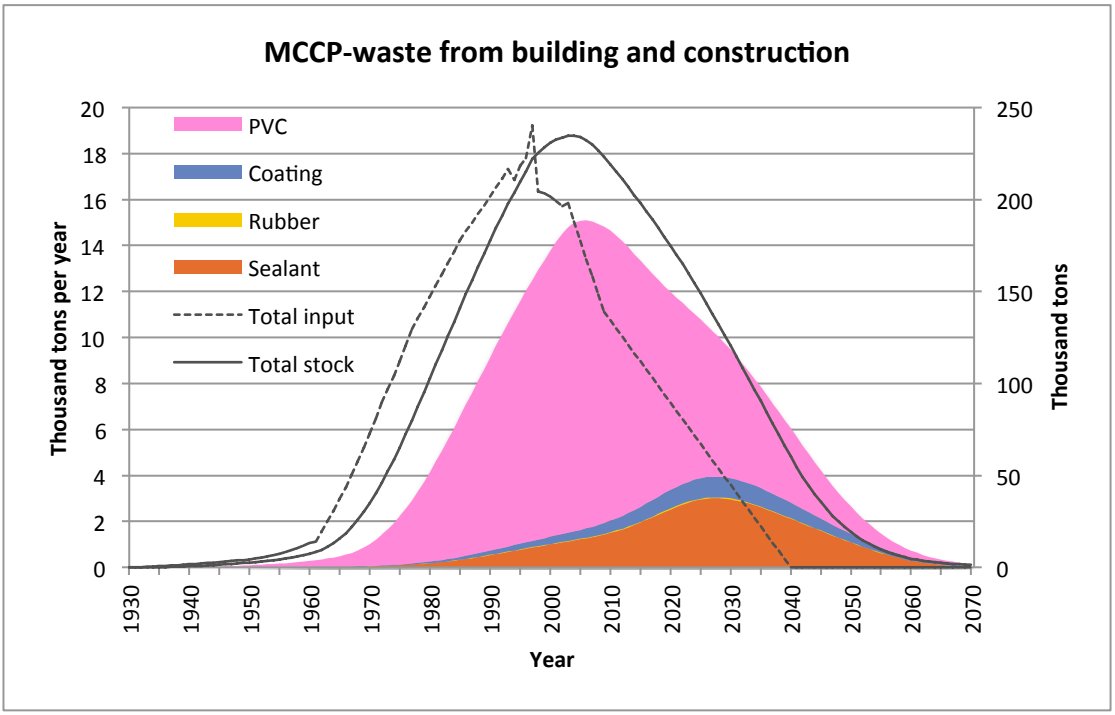

Figure 25: Scenario 1, MCCP-waste from building and construction. Stacked area graph. Lines are not stacked.

Figure 28 and 29 shows the stock and waste flows according to scenario 2. In this scenario the stock grows larger than in 2002 and reaches almost 270 000

tons before it stabilizes. The waste flow does also increase and reaches more than 15 000 tons by 2070.

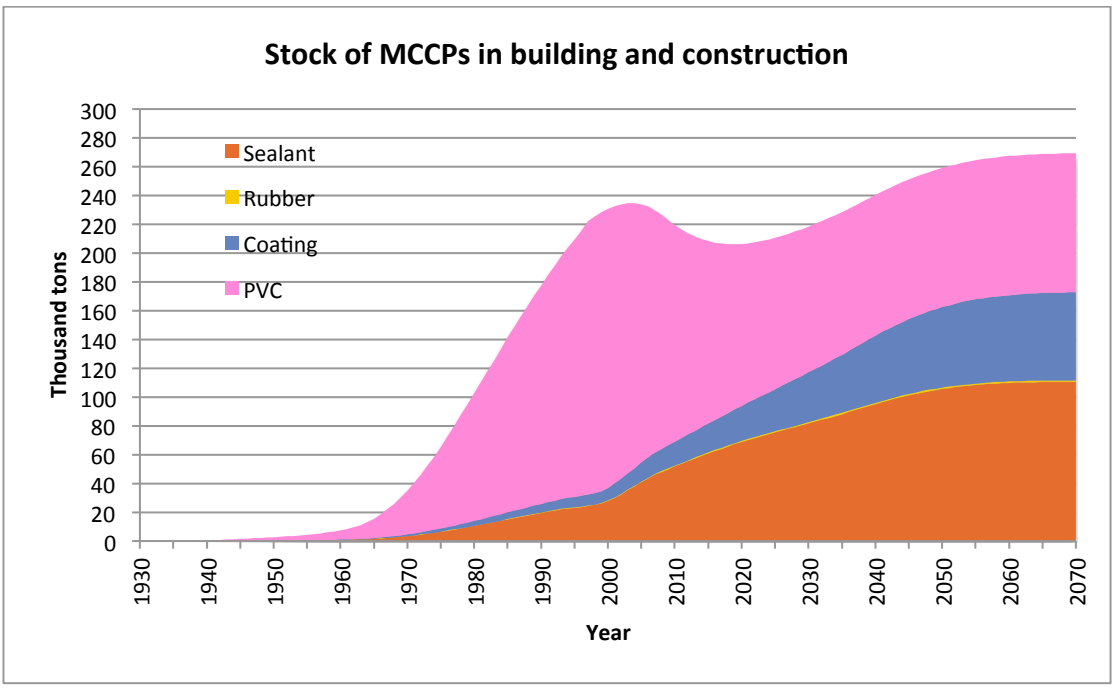

Figure 26: Scenario 2, Stock of MCCPs in building and construction. Stacked area graph.

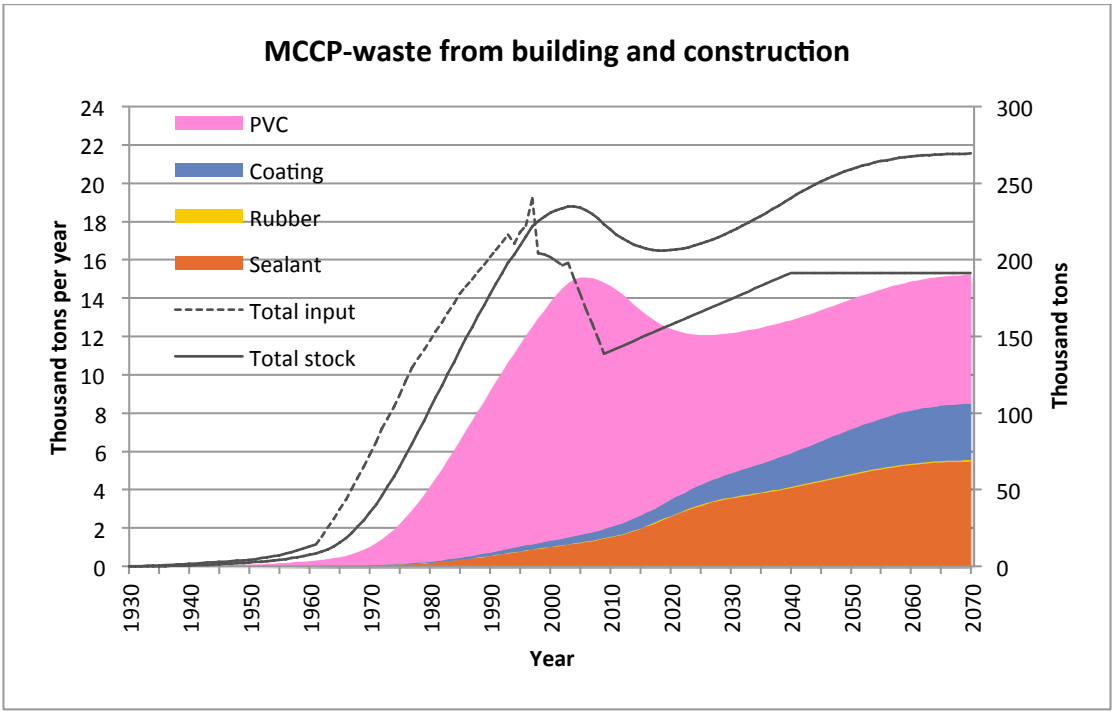

Figure 27: Scenario 2, MCCP-waste from building and construction. Stacked area graph. Lines are not stacked.

4.3 SENSITIVITY ANALYSIS OF PARAMETERS

Many parameters used in the calculations are uncertain and could have been chosen for conducting a sensitivity analysis. The input flows to the system (α_{yt}) and the distribution parameters determining the fraction of use in buildings and constructions were believed to be the most uncertain. Developing alternative input flows would be a time-consuming task. The distribution parameters have therefore been changed to see which effect this has on the resulting total amount of waste.

The distribution parameter determining the amount of sealants entering use was changed from 80% to 70% (a factor of 0,875). The other distribution parameters were also changed by the same factor – 0,875. The resulting total waste flows (including all product groups) are shown together with the original total waste flow in figure 30 and 31, for MCCP and SCCP respectively. For MCCP it can be seen that the resulting waste flow from a change in the distribution parameter for PVC, gives the biggest change in waste flow compared to the original waste flow. The resulting waste flow from a change in the distribution parameter for rubber is so small that it overlaps the original waste flow. This is why the original waste flow is difficult to see in the figure. For SCCP a change in the distribution parameter for sealants and coatings gives almost identical waste flows (the graph representing sealants is therefore difficult to see). These parameters are therefore equally sensitive – and equally important to have accurate estimates of.

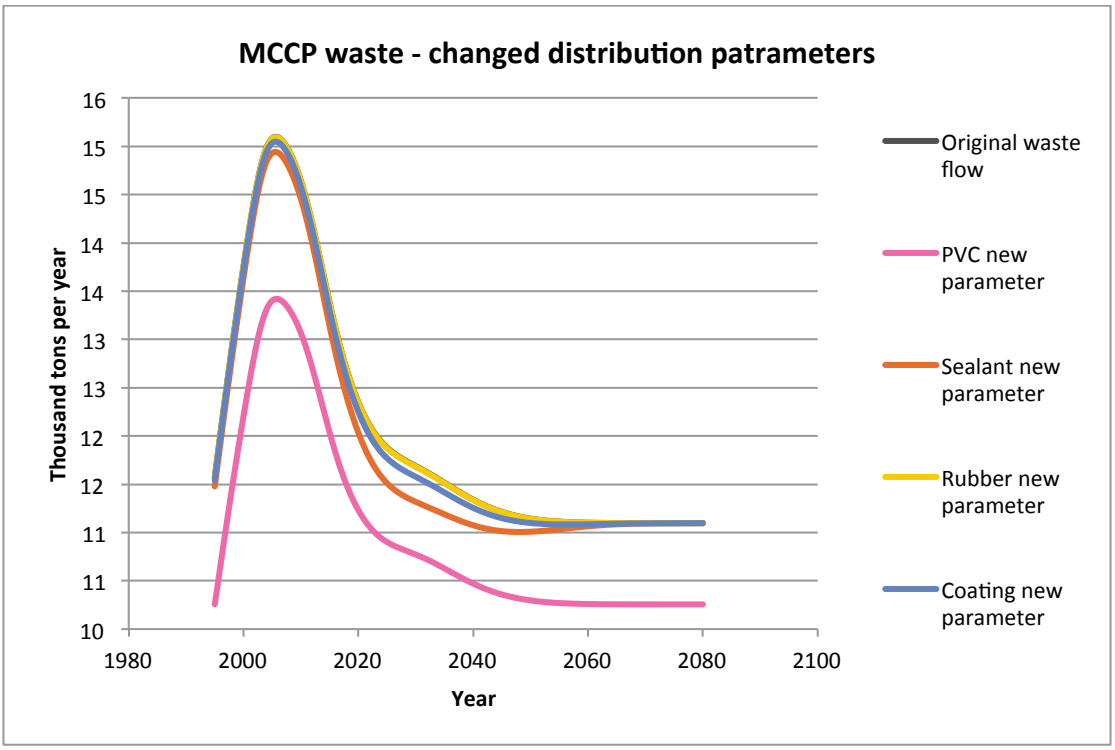

Figure 28: Sensitivity analysis, using scenario 0. The line graphs show the original total waste flow of MCCPs and the total waste flow of MCCPs when the distribution parameter for PVC, sealants, rubber and coatings are changed by the same factor.

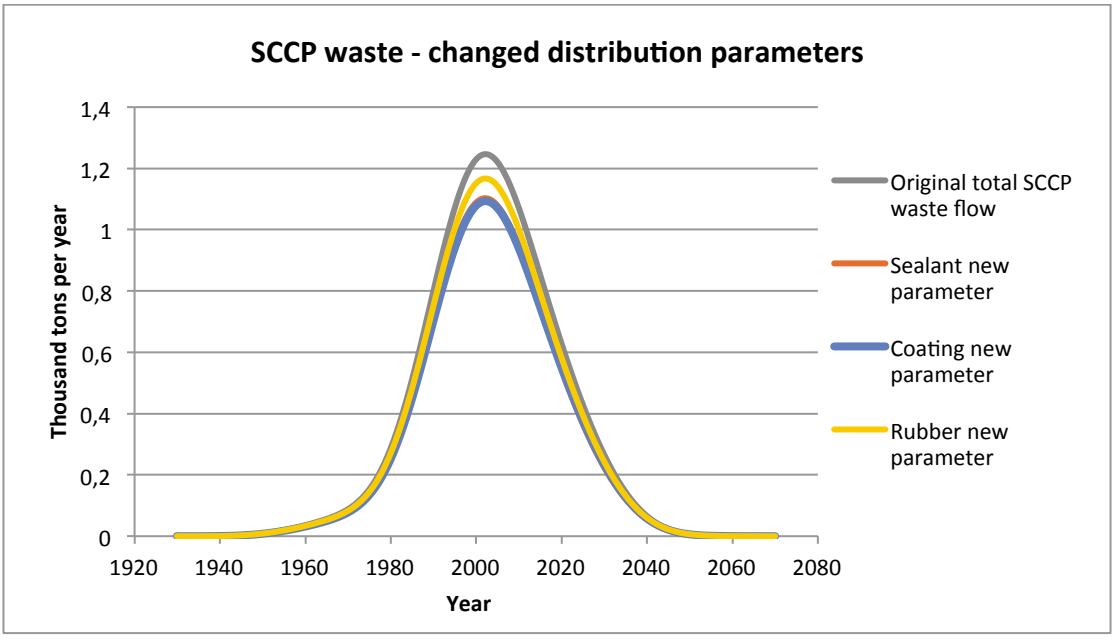

Figure 29: Sensitivity analysis. The line graphs show the original total waste flow of SCCPs and the total waste flow of SCCPs when the distribution parameter for PVC, sealants, rubber and coatings are changed by the same factor.

The result of changing the distribution parameters is that the corresponding waste flow is proportionally scaled. This gives the simple relation that the distribution parameter corresponding to the originally greatest waste flow is the most sensitive. In the case of MCCPs, PVC obviously makes up the greatest waste flow originally, and flooring, wall covering and roofing accounts for the greatest amount within the group. For SCCPs, sealants and coatings accounts for equally big waste amounts.

5 Discussion and conclusion

The lack of data and information on where CPs are used and by which amounts, has been a great challenge. The results presented in chapter 4 are therefore based on many assumptions and must be regarded as uncertain (see section 3.5). It is however believed that the general trend in stocks and waste flows are realistic and that some important findings can be drawn based on this.

One of the important findings is that the amount of MCCPs in the stock and waste flow from buildings and constructions is considerable and much greater than the amount of SCCPs. It is therefore important to determine whether MCCPs are persistent, toxic and bioaccumulative and if specific measures concerning MCCPs needs to be introduced in the EU. In this context it is also important to remember that 1% of all commercial MCCP mixtures are SCCPs. The actual SCCP stock and waste flow from buildings and constructions is therefore higher than what is shown in the results. Because of this, SCCPs will not be entirely phased out in buildings and constructions before MCCPs are phased out or the commercial MCCP mixtures are free from SCCPs.

Regardless of which scenario that is chosen it is clear that the waste flows of SCCPs and MCCPs will continue to be considerable for decades to come. Even if the consumption of SCCPs and MCCPs would cease today, they would still be present in the building and construction stock because of the long lifetime of the building products. Their presence in the built environment will therefore continue to be a potential threat that cannot easily be removed.

The data collected from literature shows that the use of SCCPs has decreased evenly since the 1980s. The use of SCCPs will therefore most probably soon be phased out and the remaining environmental issue concerning SCCPs is to take care of contaminated wastes. The use of MCCPs has decreased since the 1990s, mainly due to a reduction in PVC. The use is however still considerable and it continues to prolong a potentially big environmental problem.

The total stocks and waste flows of SCCPs and MCCPs are also believed to have reached their maximum recently and are now decreasing. However, the individual stock and waste flow of MCCPs in coatings and sealants seems to be increasing. If the use in these products continues to increase (as in scenario 2), the total stock of MCCPs in buildings and constructions will soon increase again. Focus should therefore lie on reducing the amount of MCCPs in coatings and sealants – in addition to PVC – if the positive decreasing trend is to be maintained.

It seems as SCCPs have been substituted with MCCPs (compare figure 10 and 12 in chapter 3). Soon after the use of SCCPs decreased, the use of MCCPs increased for those applications where SCCPs had been used. The use of MCCPs in PVC is however much more important than the uses where MCCPs have substituted SCCPs.

Estimates of the amount of SCCPs and MCCPs waste in Norway, has not been conducted. The estimates of SCCP and MCCP waste in EU can however be used as basis for estimating the amout of waste in Norway. The EU amounts can simply be scaled based on population, construction activity or other appropriate parameters. Such results can at least be a complement when data on historical amounts is lacking, or it can be a basis of comparison to other independent estimates. It can also be used as basis for estimating the amount of SCCPs and MCCPs entering Norway through products imported from EU.

The Norwegian Building Authority, waste treatment organizations and the building industry were especially interested in knowing the total tonnage of waste containing SCCPs and MCCPs that could be classified as hazardous. In Norway the limit for classifying waste containing SCCPs or MCCPs as hazardous is 2,5 gram per kg (Amlo, S. & Bakke, K., 2010; Miljøverndepartementet, 2004b). In literature, the typical concentration of CPs in sealants ranged from 4-20% (European Chemicals Bureau, 2005, 2008a). For coatings the concentration ranged from 5-20% and for PVC 10-15%. For rubber, the typical concentration is assumed to be similar as for PVC. If these concentrations are considered, all product groups containing SCCPs or MCCPs would most likely be classified as hazardous, in Norway. The total amount of SCCPs and MCCPs in each product group can then simply be divided by the concentration of the substances in the product group, to achieve an estimate of hazardous waste. Several samples of products used in Norway have however shown concentrations of SCCPs and MCCPs below the limit for hazardous waste (Amlo, S. & Bakke, K., 2010). This could support the contention that most products used in Norway are imported from countries in Europe using little CPs in their production (e.g. Germany, Sweden) (Cowi, 2010). The typical concentrations found in literature may therefore not be representative for products on the Norwegian market and must be reevaluated for Norway before an estimate can be given.

In the EU, waste containing significant amounts of SCCPs will most certainly be regarded as hazardous, according to Euro Chlor (Euro Chlor, 2011). It is however unclear what is meant by “significant” amounts. CPs with longer chain lengths can, according to Euro Chlor, not be classified as hazardous in the EU.

Due to the unclear definition of hazardous waste in EU, an estimation of the total tonnage of hazardous waste containing SCCPs and MCCPs is not possible. A rough estimate of total waste tonnages in EU can however be made by

multiplying the total amounts of SCCPs and MCCPs with ten (a CP concentration of 10% is assumed for all product categories). The tonnage of waste containing SCCPs and MCCPs in the EU will then be around 10 000 tons SCCPs and 142 000 tons MCCP (using scenario 0) in 2012. This is however based on the assumption that the waste fractions collected are composed of pure PVC or rubber (which is not the case for for example reinforced PVC or rubber), and that coating and sealant waste does not contain concrete or other materials it was fixed to. In reality, the actual amount is therefore believed to be higher.

It has not been possible to evaluate whether the present legislation and waste handling in EU is sufficient for safe handling of building and construction waste containing CPs, within the scope of this thesis. A satisfactory assessment of this would require much more exact knowledge on the products in which CPs are used and where they can be found. Since EU comprises several different countries, with differing legislation and waste handling solutions (although there are some overarching principles given by EU) it is not a simple question to answer.

Some comments on waste treatment can nevertheless be given. If waste is classified as hazardous, it must be assumed that it will be collected and treated in a responsible manner. Products containing hazardous substances may however not always be recognized as hazardous. If PVC and rubber is not recognized as hazardous, it is likely that it will be incinerated due to it's high calorific values. As CPs decompose at temperatures above 200°C, this is not believed to be problematic. It is however possible that PVC and rubber also is landfilled. The EU has introduced regulations to limit biodegradable municipal waste from being landfilled (The council of the European Union, 1999). This does however not seem to cover building and construction waste. And even if it would, plastic and rubber would most probably not be defined as biodegradable because most plastics biodegrades very slowly (c.f. (Miljøverndepartementet, 2004b)). Sealant and coating waste will most likely remain on the material it is fixed to. If the material it is fixed to is a metal, it is likely that it will be melted and recycled at temperatures above 200°C. This will destroy the CPs. If the sealant or coating is applied to concrete, it is more likely to be landfilled or crushed at the construction site and used for backfilling. In this case, the CPs will not be destroyed and they can pose a risk to health and environment.

For future research on the topic, the most important task is to improve the knowledge on which types of products that contain SCCPs and MCCPs and how large the amount in each product group is. This is important for improving the model and for ensuring proper waste collection and treatment. As the use of MCCPs in PVC accounts for the largest fraction of CPs in buildings and constructions, it is especially important with improved knowledge on this use.

Knowing the year CPs were introduced in the different products would also ease the waste collection and improve the model.

To improve the knowledge on historic consumption of CPs and the distribution between SCCPs, MCCPs and LCCPs, is also important. There are most probably many people and companies that have more information on production quantities and sales. It is however not easily available and needs to be collected and summarized. Euro Chlor provided almost all production/consumption estimates of CPs in EU that could be found in literature. Their estimates does however not go far enough back in time to answer when the use of SCCPs where on top historically and how big it was (van Wijk, 2012). They only have a few estimates of the use of MCCPs in EU and no estimates of LCCP use. One of the reasons for this is that the European CP market is small with few suppliers. Of confidential reasons, all information can therefore not be official. Global estimates would not encounter the same problems, but are not as useful for analyzing European conditions. There are however not many estimates of global production quantities of CPs available either. Euro Chlor is working with the establishment of a global organization for CP producers, which hopefully can give more information on this. Improved knowledge on imports and exports of SCCPs and MCCPs to Europe (especially from China) should also be a priority task.

It has not been a goal to estimate emissions to environmental compartments (air, water, soil). It is however much more information available on emissions to air, water and soil than on products containing SCCPs and MCCPs and where they are used. The model could therefore be expanded to include estimates of emissions to air, water and soil.

In this thesis it was decided to focus on buildings and constructions and their SCCP and MCCP waste. To narrow down the task to concern only buildings and constructions, is important since this sector probably is the greatest producer of CP waste. Hopefully this will increase the awareness within the sector and help it take it's responsibility. It was also thoght to be useful for developing effective measures and legislation for the sector. But to investigate in which sector products are used, is a difficult task. Within the scope of this thesis it was not possible to find out exactly which products and how much of them that acctually where used in buildings and constructions contra other sectors. The estimates are therfore only based on those assumptions outlined in section 3.4.4. Knowledge on which sectors products containing SCCPs and MCCPs are used in, should be imporved to increase the credibility and usefulness of further studies.

Although the correctness of the many assumptions and results in this thesis can be discussed, the thesis has revealed an unsatisfactory lack of data concerning the use of a hazardous substance that is of priority concern. One cannot be sure

that the SCCPs and MCCPs are handled in a safe way, before it is known where these substances are and how much there is of them. This thesis is an attempt to improve this knowledge and hopefully it will inspire to increased focus on the problem.

6 References

European Chemicals Agency (ECHA). *Annex XV restriction report; MCCPs*. (2008). Retrieved from http://echa.europa.eu/en/web/guest/search?p_p_id=echasearch_WAR_echaportlet&p_p_lifecycle=0&p_p_state=normal&p_p_mode=view&p_p_col_id=column-2&p_p_col_count=1

Amlo, S. & Bakke, K. (2010). *Kartlegging av nyere fraksjoner av farlig avfall i bygg*. The Norwegian Climate and Pollution Agency, Norconsult.

B Willis, Crookes, M. J., Diment, J., & Dobson, S. D. (1994). *Environmental hazard assessment: Chlorinated paraffins* (p. 47). Toxic Substance Division, Directorate for Air, Climate and Toxic Substances, Department of the Environment. London, UK.

Bre, Iom Consulting, & Entec. (2008). *Data on manufacture, import, export, uses and releases of alkanes, C10-13, chloro (SCCPs) as well as information on potential alternatives to its use. ECHA/2008/2*. European Chemicals Agency (ECHA).

Beratergremium für umweltrelevante Altstoffe (BUA). (1992). *Chlorparaffine. Paraffinwachse und Kohlenwasserstoffwachse, chloriert. BUA-Stoffbericht 93*. Gesellschaft Deutscher Chemiker.

Bergsdal, H. (2009). *Dynamic modeling and analysis of stocks and flows in building systems*. Phd thesis, Norwegian University of Science and Technology (NTNU)

BiPRO, Umweltbundesamt, & Enviroplan. (2011). *Service request under the framework contract No. ENV.G.4/FRA/2007/0066: Study on waste related issues of newly listed POPs and candidate POPs*. European Commission.

Britannica. (2012). *Plastic; Compounding*. Encyclopedia Britannica. Retrieved May 30, 2012, from http://www.britannica.com/EBchecked/topic/463684/plastic/82472/Compounding

Brooke, D. N., Crookes, M. J., & Merckel, D. (2009). *Environmental risk assessment: long- chain chlorinated paraffins*. Environment Agency; England and Wales.

Brunner, P. H., & Rechberger, H. (2004). *Practical Handbook of Material Flow analysis*. Lewis publishers.

COWI A/S, IOM consulting, & Entec. (2009). *Data on manufacture, import, export, uses and releases of Bis(2 -ethylhexyl)phthalate (DEHP) as well as information on potential alternatives to it's use*. European Chemicals Agency (ECHA).

Campbell, I., & McConnell, G. (1980). *Chlorinated Paraffins and Environment. 1. Environmental occurance*. Environmental Science and technology, 14(10).

Cowi. (2010). *Medium chained chlorinated paraffins (MCCPs): A survey of products in Norway. TA 2735*. The Norwegian Climate and Pollution Agency.

Drouillard, K. G., Tomy, G. T., Muir, D. C. G., & Friesen, K. J. (1998). *Volatility of chlorinated n-alkanes (C10-C12): Vapour pressures and Henry's Law constants*. Environmental Toxicol. Chem., 17, 1252–1260.

Drouillard, K.G, Hiebert, T., Tran, P., Tomy, G. T., Muir, D. C. G., & Friesen, K. J. (1998). *Estimating the aqueous solubilities of individual chlorinated n-alkanes (C10-C12) from measurements of chlorinated alkane mixtures*. Environmental Toxicol. Chem., 17, 1261–1267.

de Boer, J., Fiedler, H., Nikiforov, V. A., Tomy, G. T., Muir, D., El-Sayed Ali, T., Legler, J., et al. (2010). *The Handbook of Environmental Chemistry; Chlorinated Paraffins*. Springer-Verlag Berlin Heidelberg.

European Chemicals Agency (ECHA). (2008). *Support document for identification of alkanes, C10-13, chloro as a substance of very high concern*. Retrieved from http://echa.europa.eu/web/guest/candidate-list-table/-/substance/241/search/ /del/20/col/INCLUSIONDATECL/type/desc/pre/4/view

Edvardsen, K. I., & Ramstad, T. (2007). *Håndbok 53; Trehus*. Norges byggforskningsinstitutt. Oslo, Norway.

Entec UK Limited. (2008). *Environmental risk reduction strategy and analysis of advantages and drawbacks for medium chain chlorinated paraffins (MCCPs). Updated stage 4 report (draft)*. Department for Environment, Food and Rural Affairs, United Kingdom.

Environment Agency. (2003). *Emission scenario document: Chemicals in the coatings industry: Paints, Laquers and Varnishes*. Environment Agency. UK.

Euro Chlor. (2011). Classifications and regulations. Retrieved May 21, 2012, from http://www.eurochlor.org/chlorinated-paraffins-(cpsg)/waste-management/classification-and-regulations.aspx

European Chemicals Agency. (n.d.-a). Alkanes, C14-17, chloro. Retrieved December 14, 2011, from http://apps.echa.europa.eu/registered/data/dossiers/DISS-9ebcd9d5-5f92-56b4-e044-00144f67d031/AGGR-0e52c845-d756-4518-961e-70e867095eff_DISS-9ebcd9d5-5f92-56b4-e044-00144f67d031.html#section_3_5

European Chemicals Agency. (n.d.-b). Alkanes, C10-13, chloro. Retrieved December 14, 2011, from http://apps.echa.europa.eu/registered/data/dossiers/DISS-9d970c25-ddc8-22ff-e044-00144f67d249/AGGR-5a6482c2-1584-47ab-a2b2-78a751a08dfe_DISS-9d970c25-ddc8-22ff-e044-00144f67d249.html#section_1.1

European Chemicals Bureau. (2000). *European Union Risk Assessment Report of alkanes, C10-13, chloro (SCCP)*. Institute for Health and Consumer Protection, Joint Research Centre, European Commission.

European Chemicals Bureau. (2003). *Technical Guidance Document, Part 4*. Institute for Health and Consumer Protection, Joint Research Centre, European Commission.

European Chemicals Bureau. (2005). *European Union Risk Assessment Report of alkanes, C14-17, chloro (MCCP)*. Institute for Health and Consumer Protection, Joint Research Centre, European Commission

European Chemicals Bureau. (2008a). *European Union updated risk assesssment report of alkanes, C10-13, chloro (SCCP)*. Institute for Health and Consumer Protection, Joint Research Centre, European Commission.

European Chemicals Bureau. (2008b). *European Union Risk Assessment Report, bis(2-ethylhexyl)phthalate (DEHP)*. Institute for Health and Consumer Protection, Joint Research Centre, European Commission.

Friesen, K. J., Hiebert, T., Drouillard, K., Fisk, A. T., Bergman, Å., Tomy, G. ., Tittlemier, S. ., et al. (1995). *Synthesis, properties and bioaccumulation of chlorinated paraffins*. Organohalogen Compounds, 24, 409–413.

Gent, A. N. (n.d.). *Rubber*. Britannica. Retrieved June 2, 2012, from http://www.britannica.com/EBchecked/topic/511800/rubber

Hardie, D. (1964). *Chlorocarbons and chlorohydrocarbons: Chlorinated paraffins*. Kirk-Othmer Encyclopedia Chemical Technology.

Hjellnes Consult. (2011). *Miljøgifter og byggprodukter, vurdering av substitusjon og bransjeavtaler*. Direktorat for byggkvalitet.

Howard, P. H., Santodonato, J., & Saxena, J. (1975). *Investigation of selected potential environmental contaminants; chlorinated paraffins*. U.S. Environmental Protection Agency.

Lahaniatis, M. (2001). *Quantifizierung von kurz- und mittelkettigen Chlorparaffinrückständen in ausgesuchten fetthaltigen Lebensmitteln*. Technische Universität München.

Madeley, J. R., Gillings, E., & Reynolds, L. F. (1983). *Determination of the solubility of four chlorinated paraffins in water*. ICI Confidential Report BL/B/2301.

Miljøverndepartementet. (2004a). *Forskrift om begrensning i bruk av helse- og miljøfarlige kjemikalier og andre produkter (produktforskriften)*. Miljøverndepartementet. Retrieved from http://www.lovdata.no/for/sf/md/xd-20040601-0922.html#2-17

Miljøverndepartementet. (2004b). *FOR 2004-06-01 nr 930: Forskrift om gjenvinning og behandling av avfall (avfallsforskriften)*. Retrieved from http://www.lovdata.no/for/sf/md/xd-20040601-0930.html#map028

Mukherjee, A. B. (1990). *The use of chlorinated paraffins and their possible effects in the environment*. National Board of Waters and the Environment. Helsinki, Finland.

Nexans Norway AS. (2009). *Kabelboka*. Retrieved June 17, 2012, from http://www.nexans.no/eservice/Norway-no_NO/fileLibrary/Download_540200470/Norway/files/Kabelboka 090206.pdf

Nyström, B. (2009). *Background document on short chain chlorinated paraffins*. OSPAR Commission.

OECD. (2004). *Emission scenario document on additives in rubber industry*.

OECD. (2009a). *Emission scenario document on coating industry (paints, laquers, varnishes)*.

OECD. (2009b). *Emission scenario document on plastic additives, Env/jm/mono(2004)8/rev1*. Paris. doi:Env/jm/mono(2004)8/rev1 ?

OSPAR. (1996). *Point and Diffuse Sources. Short Chained Chlorinated Paraffins*.

Omori, T., Kimura, T., & Kodama, T. (1987). Bacterial cometabolic degradation of chlorinated paraffins. *Applied Microbiology and Biotechnology*, *25*(6). doi:10.1007/BF00252016

Persistent Organic Pollutants Review Committee (POPRC). (2010). *Revised draft risk profile: short-chained chlorinated paraffins*. United Nations Environmental Program, Stockholm convention on POPs.

Palmer, R. ., & Klosowski, J. M. (1997). *Sealants*. Kirk-Othmer Encyclopedia of Chemical Technology.

Roberts, H. . (n.d.). *Chlorinated paraffins*. Kirk-Othmer Encyclopedia Chemical Technology.

SRI International. (1986). *Chemical economics handbook*. Menlo Park, CA.

Scheer, W. E. (1944). *Properties and uses of chlorinated paraffins*. Journal of the Society of Chemical Industry.

Store norske leksikon. (n.d.). *Plast*. Retrieved May 31, 2012, from http://snl.no/plast

The council of the European Union. (1999). *Council directive 1999/31/EC (on the landfill of waste)*.

van Wijk, D. (2012). Personal communication. Ph.d. in Euro Chlor.

Wolman, A. (1965). *The metabolism of cities*. Scientific American, 213(3), 178–193.

World Health Organization (WHO). (1990). *IARC Monographs on the evaluation of carcinogenic risks to humans. Some flame retardants and textile chemicals, and exposure in the textile manufacturing industry*. Volume *48*.

Zarogiannis, P., & Nwaogu, T. (2010). *Evaluation of Possible Restrictions on Short Chain Chlorinated Paraffins (SCCPs)*. National Institute for Public Health and the Environment (RIVM) The Netherlands. Risk & Policy Analysts Limited UK.

Zitko, V., & Arsenault, E. (1974). *Chlorinated paraffins: Properties, uses and pollution potential*. Environment Canada.