

Substance flow analysis of parabens in Denmark complemented with a survey of presence and frequency in various commodities

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Abstract

Parabens are commonly used as preservatives due to anti-bactericidal and anti-fungicidal properties and they are ubiquitously present in personal care products, pharmaceuticals, food, industrial and domestic commodities. They are suspected of causing endocrine disrupting effects to aquatic organisms and adverse effects in humans and, thus, it is highly relevant to identify and quantify their sources and transportation pathways in the urban environment. Here a substance flow analysis (SFA) was performed in order to map and comprehend the substances' flow on a national basis. Many household commodities were found to contain parabens; cleaning detergents, slimy toys, and water-based paint. The presence and concentration of parabens are regulated in cosmetics and food. Use of parabens in pharmaceuticals as excipients is documented in Denmark. The import of parabens is increasing; although the number of industrial parabens containing commodities is decreasing and manufacturer reports phase-out of parabens. The vast majority of the paraben containing commodities has a durability of 18–30 months, thus the average lifetime of the paraben stock is perceived to be limited. The inflow was ca. 154 tonnes via pure chemicals and 7.2–73 tonnes via commodities in 2004. This corresponds to an average wastewater concentration of 640–900 $\mu\text{g/L}$, when excluding discharge to solid waste, soil, biodegradation and metabolism. This is in the same order of magnitudes as can be found in industrial wastewater but higher than that seen in domestic wastewater. The data needed for the SFA is sparse, dispersed, and difficult to access and associated with a great deal of uncertainty.

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1. Introduction

Cosmetics are used to beautify the human body as well as to protect the appearance or odour, for example sun-screen lotions and deodorants. Although the individual xenobiotic organic substances (XOCs) in the cosmetics often are regarded as safe for human application little is known about their effects on aquatic organisms. This is a substantial drawback since a vast majority of the cosmetics are used and removed in a way that they end up in wastewater, either directly by usage of so called “rinse-off products” or indirectly by the usage of “leave-on products” (76/768/EEC) [1]. The primary objective of wastewater treatment plans (WWTPs) is to remove bulk organic matter and nutrients. Accordingly are the processes generally effective but not optimal for removing XOCs such as industrial chemi-

cals, endocrine disrupting chemicals and pharmaceuticals (e.g., [2,3]). Hence, they are discharged into receiving water bodies in low but detectable and potentially harmful levels.

The parabens have been used as antimicrobial preservatives in cosmetics and pharmaceuticals since 1924 [4]. A review of the potential impacts on human health due to exposure to methyl and propyl paraben, proposed an acceptable daily intake (ADI) of 55 mg/kg/day for methyl paraben and propyl paraben [5,6]. However, parabens are also known to cause skin irritation and contact dermatitis in a small percentage of the general population suffering from paraben allergies [7] and it has afterward been reported that parabens were found in breast tumours [8]. Recently suspicions have been raised concerning their potential for causing endocrine disrupting effects in rainbow trout (ethyl, propyl and butyl parabens) [9–11] and medaka (propyl paraben) [12]. Though, in an *in vivo* study, the estrogenic pathway suggested difference in pharmacokinetic behaviour of butyl paraben was determined to be about 100,000 times weaker than estradiol [13].

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There are various tools for assessing the environmental impacts caused by and distribution exerted by this kind of XOCs, including environmental impact assessment (EIA), environmental risk assessment (ERA), life cycle assessment (LCA), material flow accounting, material flow analysis (MFA) and substance flow analysis (SFA). In Europe, ERA has received particular attention through the technical framework for human and environmental risk assessment first published in 1996 and revised in 2003 [14]. SFA and MFA are now receiving more attention as they have been successfully used in connection with urban water systems. For example, MFA was used in order to select relevant endocrine disruptors, which then were elucidated by pathways of environmental pollution [15]. SFA of nutrients and organic matter in the wastewater management system [16] were used to identify stressors on the receiving water. SFA was applied and aimed at identify the sources to six persistent polar pollutants, e.g., pharmaceuticals and pesticides, and determine the partial contribution of WWTP compared to the total emissions to surface water [17]. Afterwards, decision on improved treatment strategies and/or implementation of upstream source control can be determined to be most suitable to meet stipulated criteria. SFA is limited to a single substance or a small group of substances that simplify the in-depth analysis and enable the extension of the analysis to describe the exchange of substances between the lithosphere, biosphere and technosphere [18]. It aims at reducing or substituting harmful environmental fluxes [19] by tracing sources, discovering future problems and forming a basis for regulation. SFA can also be a support tool for data acquisition resulting in analyses of trends and their causes, and it can contribute to the identification and prediction of the effectiveness of source control measures or emission barriers in order to generate a ranking [20,21], hence, making it suitable to connect to LCA and ERA.

The overall objective of this study is to identify and quantify the sources and pathways for transporting parabens within the technosphere/urban environment. The outcome can be used to identify necessary actions to be taken in order to limit paraben occurrence and distribution. Subsequently, the following specific aims are as follows: (i) to perform a substance flow analysis of parabens for Denmark in year 2004 and (ii) to calculate plausible concentrations in wastewater.

2. Methodology

Substance flow analysis is an analytical tool to map and comprehend substance (i.e., elements, chemical compounds or groups of chemical compounds) flow through a given system in space and time such as a region or a company [18,20]. It is based on the thermodynamic law of mass conservation, and the basic principle is that mass is conserved in physical or chemical processes but transferred into a new compound or compounds [22]. Hence, the core method is the mass balance of the flow through a given system. van der Voet et al. [18] cover a general method for SFA, among others. In addition, in Denmark a specific paradigm for SFA has been developed for the Danish EPA [23]. The methodology can be used for identifying and quantifying key sources and sinks to pollution and facilitate linking

between concentrations in the environment with these sources. It can also be used for identification of accumulation or cessation of substances as it allows observations of the changes in the stocks. Raw materials are converted into manufactured chemicals, products, and goods (here called “commodities”) as well as wastes.

Items included in SFA ([18,23] among others):

- System definition
- Quantification of stock and flows
 - Source identification
 - Mapping of use patterns
 - Import and export
 - Waste (solid waste and wastewater) handling
- Evaluation of the results

The geographical boundary for this study is Denmark. Parabens can enter and exit Denmark via import and export in form of the pure chemicals or commodities containing parabens, but domestic production is also possible though known not to occur. Point sources are industry producing parabens or commodities containing parabens, whereas the uses of domestic commodities are considered as diffuse paraben sources. Diffuse emission is for example the use and subsequent discharge of “down-the-drain-chemicals”, i.e., cosmetics, personal care products and household cleaners. Parabens found in commodities accumulating in the society, which will be referred to as the stock, whereas the inflow refers to the difference between the export and the joint value of the import and national production. Depending on the commodity, some may stay for a long time and some may be used and then discarded shortly after production, depending on the lifetime of the commodity. As paraben containing commodities primarily are water-based emulsions the durability of these is perceived to be short in comparison to, e.g., building materials. Thus, stock is thought to marginal in comparison with the inflow, a hypothesis that will be tested.

The parabens may enter the environment via point, diffuse or accidental sources. The approach applied here will focus on the part discharge via wastewater, thus entering municipal WWTPs or on-site wastewater treatment plants, for example grey wastewater treatment, i.e., domestic wastewater without faecal contamination (showers, bathtubs, hand-basins, laundry and kitchen effluents). Inflow and stock data is focused on amounts (in ton or kg per year) and are based on official statistics on different levels.

The data is organised into several groups; bulk chemicals, “down-the-drain-chemicals”, pharmaceuticals, food products and other domestic and industrial commodities. The groups were chosen based on how the information was retrieved, the lifetime in the society and finally in the terms of domestic accessibility.

This study focuses on the stocks and flows of the short chain esters of *para*-hydroxy benzoic acid and the acid itself for 8 years (1998–2006), when available. If only limited information is available it will be focused on the year 2004, see Table 1.

Scientific publications, governmental and non-governmental reports and online databases were used. Trade organisations, producers and different experts in the field have been important.

Table 1
Compounds included in the present study

Compound	Chemical name	Abbreviation	CAS no.	E no.	ESIS [24]
Methylparaben	Methyl <i>p</i> -hydroxybenzoate	MP	99-76-3	E 218	LVPC
	Sodium methyl <i>p</i> -hydroxybenzoate	Na MP	5026-62-0	E 219	LVPC
Ethylparaben	Ethyl <i>p</i> -hydroxybenzoate	EP	120-47-8	E 214	LVPC
	Sodium ethyl <i>p</i> -hydroxybenzoate	Na EP	35285-68-8	E 215	
Propylparaben	Propyl <i>p</i> -hydroxybenzoate	PP	94-13-3	E 216	LVPC
	Sodium propyl <i>p</i> -hydroxybenzoate	Na PP	36457-20-2	E 217	
Iso-propylparaben	Isopropyl <i>p</i> -hydroxybenzoate	IPP	4191-73-5		
Butylparaben	Butyl <i>p</i> -hydroxybenzoate	BP	94-26-8		
Iso-butylparaben	Isobutyl <i>p</i> -hydroxybenzoate	IBP	4247-02-3		
Benzylparaben	Benzyl <i>p</i> -hydroxybenzoate	BzP	94-18-8		
4-Hydroxy benzoic acid		4HBA	99-96-7		LVPC

LVPC = low volume production chemical: between 10 and 1000 tonnes/year [24].

The calculations depend on the source data available, type of commodities and available information of uses. When available two different methods were used and different values could be calculated and compared.

The quantifications of stock and flows were carried out via several different approaches:

- Literature review, including a review of Danish and European legislation regarding parabens and their related products.
- Utilization of statistics (import and export).
- Search in product registers.
- Review of consumer and non-governmental organisational studies of parabens.
- Contact with industry and trade organizations.

The literature review was performed via the Technical Knowledge Center of Denmark [25], Cambridge scientific abstracts, and Web of Science as well as via a general Internet search database (Google; AltaVista). Statistics were based on Danish, Scandinavian and European statistical databanks,

Table 2
Identified paraben sources

	Group	Source	References
Down-the-drain-chemicals	Personal care products	Cosmetics, dandruff shampoo, tooth paste, sun screen agents, self-tanning products	[1,30–37]
	Pet care products	Washing and grooming products	[38,39]
Pharmaceuticals	Pharmaceuticals	Oral, vaginal and injection solutions, medical personal care products	[40–44]
	Veterinary medicinal products	Oral, injection solution and for intra-mammary use	[40]
Food products	European origin	Meat, candy, snacks and food supplements	[45,46]
	Non-European origin	Soy sauce, seasoning, baked goods, beverages, cream pastes, artificially sweetened jam and jelly, syrups	[4,44,47–50]
Domestic and industrial commodities	Domestic products incl. cleaning agents	Cleaning materials, food packaging polyethylene (LDPE) film, household paper, pleasure gels, products for treatment of sports injuries and pains, slimy toys and textile dyes	[51,58]
	Pesticides	Embalming and preservation of animals	[54]
	Paint, lacquers and varnishes	Water-based paint, spray paint, glass and porcelain colours	[28,59,60]

whereas Danish and European regulations were accessed via the regulative organisations' homepages (the legal information system of the Danish State; the Official EU Law portal and the Nordic product register [26–28] and Nordic non-governmental organisation reports were accessed via their homepages, respectively. Individual industry and trade organisations were contacted directly via telephone, mail and email after identification of their relevance by accessing their homepages.

3. Results

3.1. Use and source identification

3.1.1. Uses

Parabens and their salts are widely used as preservatives for their bactericidal and fungicidal properties [4,29].

3.1.2. Sources

Parabens can be found in personal care products such as shampoos, shower gels, skin lotions, and toothpaste as well as

Table 3
Legislation governing paraben concentrations in food within Denmark [45] and the European Union [62]

Products	Maximum authorized concentration (mg/kg)	Items where the Danish legislation differ from the European one
Confectionery (excluding chocolate)	300 (E214–219)	Also excluding chewing gum
Snacks based on cereals, potatoes or starch, and coated nuts	300 (E214–219)	Includes snacks based on starch and processed nuts
Liquid dietary food supplements	2000 (E200, 202–203, E210–213, and/or E214–219)	
Surface treatment of dried meat products	Surface treatment of dried products q.s. (E214–219)	Excluding traditional Danish liver paste/pâté
Jelly coatings of meat products and pâté (non-heated, heat prepared)	1000 (E 200, 202–203 and/or E 214–219)	

q.s. (quantum satis): no maximum level has been established but the compounds are allowed to be used according to Good Manufacturing Practice in a level sufficient to achieve the sought effect.

topical pharmaceuticals. They are also applied as food additives and are present in a wide range of domestic and industrial commodities (Table 2).

3.1.3. Legislation governing use of parabens

The legislation and regulation governing the use of parabens in personal care products including cosmetics in Denmark are summarised in the Danish statutory order on cosmetic products [31]. Parabens permitted in food is described in the list of permitted food additives [45]. Both have been conformed around the corresponding Directives of the European Union (76/768/EEC; 95/2/EC [1,46]), see Table 3.

The maximum authorized concentrations of parabens in personal care products are 0.4% (acid) for one ester and 0.8% (acid) for a mixture of esters [1,31]. The Danish statutory order permits 4-hydroxy benzoic acid, its salts and esters but excludes the benzyl ester whereas the European directive includes the phenyl ester. Although contact has been made to both the Danish EPA and the European Environmental Agency, it has not been possible to neither confirm nor dispute that the two regulations refer to the same compound.

All compounds present in concentrations greater than 1% should be included in the list of contents in decreasing order of abundance [31]. Compounds present in concentrations less than 1% should also be included in the list of contents in optional order. However, impurities and technical excipients, such as solvents and carriers, are not included. Additionally, the European directive states that “Substances may be used in cosmetic products in concentrations other than stated in 76/768/EEC if intended for other specific purposes apparent from the presentation of the products, e.g., as deodorants in soaps or as anti-dandruff agents in shampoos” [1].

For the pharmaceuticals; the use of excipients such as sweeteners, preservatives and dyes are regulated by the Danish Statutory order no. 1210 [61] where it states that *para*-hydroxy benzoates and their esters (E214-219) should always be reported on the package leaflet due to the risk of allergic reactions.

Imported food into Denmark must also fulfil the requirements for food additives [45].

3.2. Mapping of production and use patterns

3.2.1. Production within and import to EU

In terms of production, four parabens are listed as low volume production chemicals (LVPC) (Table 1), i.e., produced or imported into the EU with a tonnage >10 tonnes/year but less than 1000 tonnes/year per producer or importer). Since the other parabens are reported as neither LVPC nor high volume production chemical (HVPC), they are produced or imported with a tonnage less than 10 tonnes/year. None of companies that produce or import parabens to Europe are located in Denmark [24].

3.2.2. Consumer related use of products containing parabens

In Denmark, the Danish Information Centre of Environmental and Health has a database which compiled tests of cosmetics and household chemicals from 2003 to 2006 [62]. This study utilizes information available from tests on the following personal care products: baby bathing products, baby lotions, children’s cosmetics, combined shampoo and body wash (2-in-1 products), conditioners, deodorants, hand soaps, make-up, suntan lotions and wet wipes. The level of detail in tests on these products varies and their aims were different. Therefore, some studies contain analyses or evaluations of all parabens while some only focus on BP and IBP, and hence the high weight of these to compounds in Table 4. However, since the table contains information on 751 different personal care products, it is regarded as a good reflection of the presence of parabens in Danish commodities. In total, 272 out of 751 products contained parabens, i.e., 36%, and BP was present in 13% of the products. Five parabens were found to be present on the declaration of contents on the investigated personal care products; MP, EP, PP, BP and IBP as well as the unspecified “paraben” parameter, i.e., the presence of parabens has been reported but not specified individually. In general MP and PP are the most frequently used parabens.

In a similar study of baby products in Sweden, it was found that six out of 69 products contained butyl paraben, i.e., 9% [65]. No other parabens were included in the study. In a Norwegian study of baby lotion, baby shampoo and soap, children’s sun lotion and baby wet wipes, 37 of 117 products, i.e., 32%, were

Table 4
Percent of investigated Danish personal care products containing parabens (in %)

	MP	EP	PP	BP	IBP	Parabens	Number of products tested
Baby bath [62]				9 ^a			11
Baby lotion [62]				27 ^a	20 ^a		15
Baby wash [62]				17 ^a			6
Baby lotion [63]						33	12
Baby wet wipes [62]			31				13
Children's cosmetics [62]				21 ^a	16 ^a		43
Children's cosmetics [62]	100	20	100	20	20		5
Conditioners [62]				9 ^a	0 ^a		23
Dandruff shampoo [32]	22	11	22	11			9
Deodorants [62]			3				31
Hand soaps [32]	12	12	12	4	4	4	25
Hand soaps [62]			2				45
Hair styling products [36]	23	8	16	9	4	2	328
Make up [62]				36 ^a	25 ^a		36
Nipple lotions [62]				20 ^a			5
Shampoo and products for men [62]				17 ^a			6
Shrovetide and theatrical make-up [64]	61	18	51	16			51
Skin creams and lotions [30]	85	43	70	36	9		67
Sunscreen lotions [62]				3 ^a			32

^a Studies with focus on only BP and IBP.

found to contain parabens. The most frequently present parabens were methyl and propyl parabens, present in 32% and 26% of the products, whereas BP was present in 11% of the products in a range from 7% in baby lotions to 19% in baby soaps [66].

In an empirical study of the consumption of household chemicals at a tenants owners' society, BO90, in Copenhagen (Denmark) [67,68], 92 household chemicals and personal care products were registered to have been used by 30 tenants [67]. Sixteen of these contained parabens, i.e., 17% (Table 5). However, no cosmetics were included and hence an important source of parabens is missing, indicating that the number originating from BO90 is an underestimation of the real number of paraben containing products. Cosmetics on the other hand frequently contain parabens as found by Masten [69] whom recorded parabens in 58% of the cosmetics.

In the USA, the Household products database contains over 6000 consumer brands (non-food and non-pharmaceutical consumer products) [38] and the SkinDeep personal care product safety guide contains information on over 14,835 products [70]. The Household product database records parabens to be present in personal care products and pet care products and a total of 346 products were found to contain parabens, and most frequently hair dyes, skin moisturizers as well as eye and lip make-up. Most products contained two parabens, but some products were found to contain five and six different parabens and/or sodium salts [38]. In SkinDeep, parabens were

found to be present in Anti-aging, Body Wash/Cleanser, Brow Liner, Conditioner, Eye Shadow, Facial Cleanser, Facial Moisturizer/Treatment, Foundation, Lip Gloss, Liquid Hand Soap, Makeup Remover, Mascara, Moisturizer, Powder, Shampoo, and Sunscreen/Tanning Oil. MP were found to be present in 40% of the personal care products, EP 9%, PP 33%, BP 10% and IBP 10%, respectively [70]. Additionally, besides the five parabens included in this study SkinDeep also includes information of personal care products containing iso-propyl paraben and three unspecified parabens (*para*-hydroxybenzoate ester and parabens). No corresponding Danish databases were found, but it is assumed that a similar distribution is valid for Denmark. This is coherent with the commercial preservatives and blends available in Europe, i.e., MP, EP, PP, BP and IBP as well as their sodium salts [29].

3.2.3. Pet care products and patterns

In one study of pet care products on the Danish market were 455 products found of which 315 cat/dog products and 140 were intended for horses [39]. The authors reviewed products sold in shops, in online stores, veterinary hospitals, pet grooming salons and in connection with shows and assessed that they had covered 75% of the products sold in Denmark. They also noted that the majority of the products lacked lists of content and thus were only 157 products evaluated for their content. MP was the most frequently found paraben and IBP were found

Table 5
Parabens in personal care and domestic products found in an empirical study

Compound	Number of products	Type of products
Methylparaben	15	Cleaning detergent, conditioner, hand soap, shampoo, shaving foam, shower gel, skin lotion, toothpaste
Ethylparaben	7	Conditioners, hand soap, shampoo, shower gel
Propylparaben	10	Cleaning detergent, conditioner, hand soap, shower gel, skin lotion
Butylparaben	5	Conditioner, hand soap, shaving foam
Isobutylparaben	4	Conditioner, hand soap, shaving foam

Table 6
Number of products and paraben containing products in the SPC database [40]

	Pharmaceuticals	Veterinary medicinal products	Herbal medicinal products	Vitamins and mineral preparations
No. of products	5560	520	142	113
No. of products containing parabens	178	41	7	0
Percentage containing parabens	3.2	7.9	4.9	0

Table 7
Parabens in Danish pharmaceuticals [40]

Use type	MP	Na MP	EP	Na EP	PP	Na PP	BP	IBP	BzP	Total no. of pharmaceuticals
Oral	72	5	1	1	58	4	2			78
Rectal	13	1			6	1				14
Eye	1									1
Nose	3				3					3
Dermal	27	1			20		2			31
Shampoo	3		1		3		1	1		3
Injection	28				12					29
Plaster	3		1		3		1		1	3
Vaginal	1	1			1	1				2
Internal	14				6					14
Total no. of parabens	165	8	3	1	112	6	6	1	1	178

to be present in half the products containing BP. In total 24 products did contain parabens (15%). In Denmark, 40% of the families have pets and there are 546,000 dogs and 646,000 cats in Denmark [71].

3.2.4. Parabens in pharmaceuticals and veterinary medicinal products

The Danish Medicine Agency has two tools for pharmaceutical evaluation, summaries of product characteristics (SPC [40]) and annual statistics (grouped according to active compounds and to packets [72]) for the years 2000–2004. Statistics of additives such as the parabens are not included [73] but merely information regarding their presence. In the SPC database there is also information regarding veterinary medicinal products, and herbal medicinal products as well as vitamins and mineral preparations, see Table 6.

As the use of excipients is regulated [61] and the presence of parabens should be reported on the package, the found number of paraben containing products should be the correct value (Table 7). However, the regulation does not include the veterinary medicinal products and hence, the actual number of products containing parabens could be larger than what can be found from the package leaflets (Table 8).

Methyl and propyl parabens are the most frequently used parabens in pharmaceuticals, present in 93% and 63% of the

paraben containing pharmaceuticals, respectively. From Table 7 it can also be seen that parabens are primarily used in pharmaceuticals intended for oral and dermal use as well as injection solutions.

Methyl and propyl parabens are the only parabens used in veterinary medicine products. They are used in oral suspensions or pastes, injection solutions or for intra-mammary use. Similarly, in a study by the Swedish Medical Products Agency [74] MP were found to be present in ca. 170 medical products including ca. 20 veterinary medicine products, thus in the same order of magnitudes as found in this study.

Among the natural remedies all the seven products were found to contain MP and four contained PP. Four were oral liquids, one nose spray and two in tablet form. None of the Vitamins and mineral preparations was reported to contain parabens.

3.2.5. Paraben containing food products and pattern

Only three studies of the use or presence of parabens in food stuff (candy, meat and fish products) could be found for Danish conditions. Thus, a questionnaire was sent to 21 relevant Danish trade associations, non-Danish companies with production facilities or branches in Denmark and major Danish food producers were contacted in order to confirm general attitude towards parabens and their use of parabens in food, as shown in Table 9. Nineteen replied (90%) and only one were

Table 8
Parabens in Danish veterinary medicine products [40]

Use type	MP	Na MP	EP	Na EP	PP	Na PP	BP	IBP	BzP	Total no. of veterinary medicine products
Oral	13				13					13
Injection	27				15					27
Intra-mammary	1				1					1
Total no. of parabens	41				29					41

Table 9
Results of the questionnaire among food related trade organisations and producers

Number of associations/producers contacted in this study	Meat ^a	Snacks ^b	Candy	Food supplements	Spices, food additives incl. preservatives
Contacted	8	5	3	3	2
Replied	8	5	3	2	1
No reply	0	0	0	1	1
Using parabens	0	0	0	1	0

^a Salami and pâté of poultry, pork and beef, dried or heated products.

^b Coated nuts and snacks made of wheat, corn, rice or other grains.

found to use parabens in their products, i.e., liquid oral food supplements.

Based on this rather limited study it is indicated that Danish producers do not primarily use parabens but sorbates (E200, 2002–2003) for food preservation.

For filled chocolates, it was found that 9 out of 101 samples (8.9%) contained PP but neither MP nor EP was detected in the same study [75]. All the PP containing filled chocolates were of Danish origin. Two hundred ninety meats and fish (meat products made of whole pieces or processed meat and sliced pieces of these products; boiled, roasted, smoked, dried or salted fish and fish products; fish products, canned and in glass; and fish products, canned and heat-treated) were investigated for MP, EP and PP where EP was found in one fish spread [76]. However,

parabens are not permitted in fish products according to the Positive list [45] and in another study of 22 fish products were none of the parabens detected [77], hence, 1 of 59 tested fish products contained parabens (1.7%).

Other studies, of non-European origin has shown that parabens are present in a wide range of food products such as soy sauce, fish sauce, pickle condiment liquids and jams as well as beverages [47,49,78,81]. Food imported into Denmark must also fulfil the Danish requirements for food additives [45]. The producer or importer has the responsibility to reassure that the products imported to, or sold, in Denmark are harmonized to the current legislation. The imported goods are therefore controlled by the companies' "self supervision", where the companies are supervised by the control authorities located at the

Table 10
SPIN database tonnage and use of parabens in Denmark within industry and commercial businesses

Compound	Year	Number of preparations	Tonnes	National use category
MP	2004	82	0.3	Cleaning/washing agents, Cosmetics, General cleaning/washing agents (floor wash, basic cleaning), Hair care products, Hair shampoo, Polishing agents, Skin care products, Skin cleaners (Soap, shower gel, hand cleansing cream), Skin protection materials, Writing ink
	2003	113	1.4	
	2002	124	1.5	
	2001	122	1.6	
	2000	125	1.5	
EP	2004	31	0.0 ^a	Cosmetics, Hair care products, Hair shampoo, Skin care products, Skin cleaners (Soap, shower gel, hand cleansing cream)
	2003	43	0.1 ^a	
	2002	50	0.8	
	2001	50	0.8	
	2000	38	0.1 ^a	
PP	2004	56	0.1 ^a	Cleaning/washing agents, Cooling agents for metal processing, Cosmetics, Cutting fluids (for metal treatment), Hair care products, Hair shampoo, Polishing agents, Skin care products, Skin cleaners (Soap, shower gel, hand cleansing cream), Skin protection materials
	2003	95	a	
	2002	112	0.6 ^a	
	2001	106	0.6 ^a	
	2000	107	0.6 ^a	
BP	2004	27	0.0	Cosmetics, Hair care products, Hair shampoo, Skin care products, Skin cleaners (Soap, shower gel, hand cleansing cream)
	2003	44	0.1	
	2002	46	0.1	
	2001	42	0.1	
	2000	38	0.1	
IBP	2003	5	0.0	No record
	2002	5	0.0	
	2001	a	a	
	2000	a	a	
Na MP	2004	a	a	Paint, lacquers and varnishes
	2003	a	a	
	2002	a	a	
	2001	a	a	
	2000	5	0.0	

^a Contains confidential data. If the value is "0.0" it means that the volume is below the limit of accuracy, i.e., 100 kg.

Table 11
Number of the investigated consumer commodities containing parabens (in %)

	MP	EP	PP	BP	I-BP	Parabens	No. of commodities
Artificial blood [85]	100						1
Dyes (textile and pens etc) [52]						8	12
Finger paint [86]	42	5	32	11			19
Food packaging polyethylene [55]							–
Hand soap for industrial use [58]	8		2				25
Household cleaners [54,67]	50		50				2
Household cleaners (this study)						12	8
Household cleaners and chemicals [87]						18	39
Kitchen rolls (virgin fibre) [51]	9		18				11
Pleasure gels (incl. erotic massage oils and creams) [56]	36	18	24	18	15		33
Slimy toys [53]	17		17				18
Spray paint [59]						1	142
Treatment of sports injuries and pains [57]	8	3					39
Water-based paint [85]						X	

regional veterinary and food administration centres [82]. Thus, the contribution of parabens present in food such as soy sauce and jam ought to be small, but personal import during tourist trips and immigrants visits to their native countries cannot be excluded.

3.2.6. Other domestic products and industrial commodities

3.2.6.1. Industrial and commercial commodities. The Nordic SPIN database consists of data from all the Nordic countries (Norway, Sweden, Denmark and Finland). The Danish data consists of information regarding dangerous products used in industrial or other commercial businesses. Therefore, products used only by private persons, including cosmetics and pharmaceuticals as well as consumer commodities are not included in the database [83]. The SPIN database uses net data, equal to data on exported chemical products subtracted from a total of produced and imported chemical products [28]. The net paraben tonnage and corresponding uses for 2000–2004 are shown in Table 10.

One biocide manufacturer (not named due to a confidentially agreement) that previously had a paraben containing product on the market was contacted during this study and it became clear that he no longer produced any paraben containing products. In a Danish survey of biocides it was noted that embalming fluids contained parabens [54].

3.2.6.2. Consumer commodities. A wide range of domestic Danish products such as household cleaning agents, e.g., window washing liquids, universal cleaning liquids, and household paper and food packaging materials have been found to contain parabens (Table 11). MP and PP are the most commonly used parabens. Five shoe care products were tested for parabens (cleaning, care and freshener products) but none of them were found to contain MP, EP, PP or BP [84].

The parabens are the main biocides used for preservation of cleaning materials in Denmark [54] and the typical concentration of a mixture of parabens is 0.7%. No in-depth studies of parabens different types of household cleaners could however be found. To get an idea an examination of the presence of universal cleaners and washing-up liquids was carried out at three departments at

the Technical University of Denmark. The study was carried out once in nine different products and it was found that one of them, thus approximately 11%, contained parabens. A Swedish handbook on washing-up liquids, cleaning agents and washing detergents contains 39 product categories within the field and 7, i.e., 18% were recorded to contain parabens (universal cleaners, floor polish, “heavy duty cleaner”, washing-up liquids, metal polish, scrubbing polish, and liquid soap [87]).

In a survey of materials used in live role-playing and sold in specialised role-playing shops, it was found that the artificial blood is preserved with MP [85].

The products register SPIN shows that the category “Paint, lacquers and varnishes” contains parabens but only a limited number of reports, publications, etc. have been found to confirm this (glass and porcelain colours, spray paint and textile dye as well as dye pens). Hence, contact with two manufacturers of paints, lacquers and varnishes resulted in negative replies as they do not use parabens in paints or coatings. The Confederation of Danish Industries [85] reports that their members generally do not use parabens in water-based paint, lacquers or glue. Only one of their members replied in a questionnaire that they used raw materials containing parabens in “low mg/L levels” and thus the concentration in the final product would be substantially lower, however a more detailed reply could not be obtained. Similarly, a manufacturer of indoor paint, hobby paints and finger paint replied that their paint in average contained 0.27% biocides but no parabens.

Although the Danish EPA and the Danish Information Centre for Environment & Health have had focus on hobby paint for colouring of glass and porcelain etc. as well as modelling clay, the focus has not been on parabens and thus, no use pattern could be retrieved. There are more than 39 biocides registered as preservatives in paints in the Danish national database which provide the background information to SPIN; PROBAS [89]. The list of preservatives, however, was not available due to confidentiality but for most water-based paints, there is no obligation to declare the individual components to PROBAS, and thus the register cannot be used for quantification of the total content of biocides in paint [54]. However, one out of 142 tested spray paints contained parabens [59].

Table 12
Import and export of 4-hydroxybenzoic acid esters and their salts (code 29182930) retrieved from [90]

Annual (tonnes)	1998	1999	2000	2001	2002	2003	2004	2005	2006
Import	8.6	31	32	38	95	119	159	165	216
Export	0.33	0.0	0.0	2.0	3.3	5.3	4.4	6.5	0.6
National ^a	0	0.16	0	0	0	1.0	0	0	^b
Inflow	8.3	31	32	36	92	115	154	158	216

^a Sales, production, treatment or collection.

^b Data from the 4th quarter not available (Feb 2007).

3.3. Quantification of flows

3.3.1. Raw materials

The import and export statistics of external trade, i.e., the import and export to Denmark as well as the Danish national production of certain goods is available through the StatBank [90], the parabens are however grouped into one group consisting of “4-hydroxybenzoic acid and its salts and esters” (code 29182930) according to the Danish customs service [91]. Data for January 1999 to March 2006 were extracted. The numbers presented are the net weight i.e., the packing is not included [92] furthermore, the national production and sales comprises of produce and solid within Denmark as well as produced and sold outside Denmark [93]. The commercial business providing Denmark with the majority of the parabens via import confirms that only the parabens (MP, EP, PP, BP and IBP) and their sodium salts are sold to Denmark, hence, the acid (4-hydroxybenzoic acid) is not included.

Both the import and export of parabens has increased during the selected period (1998–2006) though the export is only 3–5% of the annual import (Table 12). The national sales, production, treatment or collection is minor in comparison, hence the inflow substantially exceed the outflow. Parabens in commodities, e.g., pharmaceuticals and personal care products do not contribute to the statistics in Table 12, as they are a part of the composite commodity, and hence, the actual import and export of parabens is greater than what can be expected from the above mentioned numbers. An overview of the flow of parabens in 2004 can be seen in Fig. 2. Between 2000 and 2006 the sales of cosmetics and personal care products (in Euros, €) increased by 31% [94], which partly can explain the increased import of the pure chemicals as the production volumes has increased correspondingly.

3.3.2. Personal care products

3.3.2.1. *Presence.* The regulation limits the use of single parabens to 0.4% and the sum of all parabens into 0.8% [1].

Table 13
Paraben content in personal care products (percentage of total weight)

Commodity type	MP	EP	PP	BP	IBP	Parabens total
Hand cream [95]	0.10	0.08	0.07			
Lip gloss for children [53]			2.3			
Skin creams and lotions [30]	0.007–0.409	0.005–0.100	0.005–0.206	0.012–0.100	0.002–0.048	0.024–0.511
Toothpaste for children [33]						0.017–0.052
Toothpaste of general use [33]						0.007–0.137
Toothpaste of medicinal use [33]						0.035–0.045

Typical usage concentrations suggested by a paraben manufacturer are for the individual preservatives 0.1–0.3% and for preservative blends 0.03–0.8% [29]. One Danish study of 98 skin lotions revealed that the actual paraben content ranged from 0.024% to 0.511% in the various items (Table 13). The highest individual concentrations were noted for MP and PP where MP actually exceeded the regulated limit. Studies of the content of paraben in toothpastes indicate that these conform to the stipulated limits for personal care products (Table 9).

3.3.2.2. *Import and export.* Import and export as well as Danish domestic production [90] of a range of potential paraben containing personal care products, selected from the eVITA database [91] and based on the sources identified in Table 2 is shown in Table 14. For the majority of the personal care products, the import exceeds both the export and the national production. It can be noted that only hand soap and toothpaste and related products are reported to be produced in Denmark. This is not entirely true as Danish industry also, e.g., produce topical skin lotions, etc. but these are sold and distributed within the framework of the pharmacy organisation and therefore included among the pharmaceuticals.

The national production refers to “national production, treatment or collection” and as all the parabens present in nationally produced commodities originate from imported pure chemicals there is an inherent risk of double bookkeeping of the amounts of parabens.

Using the data above (import and export statistics) and the pattern of parabens in personal care products, in which it was found that 36–40% of the personal care products contained parabens, as well as the maximum and minimum concentrations permitted and found in skin lotion (0.8 and 0.024%) from Table 13, the amount of parabens included in the complex matrix of blended personal care products can be estimated (Table 15). For more details see Fig. 2.

Table 14
 Import and export and national production of personal care products (tonnes per annum) [90]

		1999	2000	2001	2002	2003	2004	2005
Hand soap	I	3597	4052	3667	4846	4592	4567	4066
	E	902	1385	1463	2173	2687	1863	1438
	N	773	640	689	1478	2053	1763	699
	IF	3468	3307	2893	4151	3958	4467	3327
Skin lotion	I	2779	3512	3123	3170	2845	2128	2532
	E	2133	1936	2077	2126	1996	1808	1409
	N	0	0	0	0	0	0	0
	IF	646	1576	1046	1044	849	320	1123
Hair wash (330510)	I	6344	6819	7388	7805	8006	6288	9329
	E	3501	4244	7454	7147	7334	6999	6760
	N	0	0	0	0	0	0	0
	IF	2843	2575	−66	658	672	−711	2569
Beauty treatment incl. sun lotion	I	5789	6276	5667	6235	8278	7443	9631
	E	1219	1552	2609	2375	2673	2328	2424
	N	0	0	0	0	0	0	0
	IF	4570	4724	3058	3860	5605	5115	7207
Eye make-up (330410)	I	299	358	370	390	519	486	527
	E	36	52	67	90	143	142	170
	N	0	0	0	0	0	0	0
	IF	263	306	303	300	376	344	357
Lip make-up (330420)	I	385	474	253	365	383	391	363
	E	27	155	850	1023	1181	968	774
	N	0	0	0	0	0	0	0
	IF	358	319	−597	−658	−798	−577	−411
Face make-up	I	285	224	126	97	127	133	204
	E	33	31	23	41	108	450	420
	N	0	0	0	0	0	0	0
	IF	252	193	103	56	19	−317	−216
Oral hygiene products (3306)	I	172	173	325	272	318	352	471
	E	36	103	275	197	291	173	183
	IF	116	123	53	−19	−8	−496	−504
Toothpaste (3306)	I	5751	5706	7369	7770	12387	9598	9151
	E	3771	3894	4905	6518	8484	6051	5403
Toothpaste and related products	N	162	152	123	98	77	57	10
	IF	2142	1964	2587	1350	3980	3604	3758
Hair removal and others, e.g., deodorants and shaving related prod. (3307)	I	805	630	688	1124	1761	1875	2647
	E	614	744	1274	1153	1418	1347	1415
	N	0	0	0	0	0	0	0
	IF	191	−114	−586	−29	343	528	1232
Hair care products (33059000)	I	4087	6578	7043	7469	9835	11792	9470
	E	893	1044	633	477	637	1332	1707
	N	0	0	0	0	0	0	0
	IF	3194	5534	6410	6992	9198	10460	7763
Hair tonic (33059010)	I	589	430	213	322	193	405	478
	E	8	6	1	1	7	12	20
	N	0	0	0	0	0	0	0
	IF	581	424	212	321	186	393	458
Permanent treatment (330520)	I	264	213	134	145	188	121	98
	E	5	4	11	3	3	33	50
	N	0	0	0	0	0	0	0
	IF	259	209	123	142	185	88	48
Manicure etc. (330430)	I	491	564	525	415	266	272	259
	E	57	116	82	136	64	105	67
	N	0	0	0	0	0	0	0
	IF	434	448	443	279	202	167	192

I = import; E = export; N = national production, treatment or collection; IF = inflow, i.e., import + national production − export.

Table 15

Minimum and maximum paraben (tonnes) import, export and national production as a part of the commodity group personal care products

	%	2004		1998–2005 cumulative	
		Max	Min	Max	Min
The annual import of parabens via personal care products	36 40	132 147	4.0 4.4	91–143	2.7–4.3
The annual export of parabens via personal care products	36 40	68 76	2.0 2.3	38–78	1.1–2.3
The annual national consumption of parabens via personal care products production	36 40	5.2 5.8	0.16 0.18	2.0–6.1	<0.1–0.18

Table 16

Presence of parabens in pet care products in Denmark

Number present	MP	EP	PP	BP	IBP	Total no.
On the list of content	22	12	19	11	6	157
Stated concentration (%) ^a	0.1–0.2	0.1–0.2	0.1–0.2	0.1–0.2	0.1–0.2	
Measured concentration (%) ^b	0.0062–0.099	0.0025–0.018	<0.002–0.009	<0.001–0.02	^c	12

^a Provided by the producer(s).^b Chemical analyses (GC–MS).^c Not analysed.

Table 17

Concentrations of parabens in human pharmaceuticals (in mg/g)

	MP	EP	PP	BP	Total
Cough syrup (mg/mL) [42]	1.7				1.7
Cream a [97]	0.98–1.00			0.24–0.26	1.22–1.26
Cream b [97]	1.90–1.98			0.19–0.21	2.09–2.19
Cream c [97]	1.92–2.05		0.39–0.41	0.21–0.22	2.97–3.16
Lotion a [97]	1.42–1.44			0.18–0.20	1.60–1.64
Solution a [97]	1.65–1.68				1.65–1.68
Ointment a [97]	0.16–0.18			1.41–1.48	1.56–1.66
Bioadhesive gel (%) [98]				0.3	0.3

3.3.3. Pet care products

3.3.3.1. Presence. One Danish study focusing on pet care products (shampoo, conditioners, skin and fur oil, paw and hoof creams, etc.) was found to compare the declared concentrations of parabens with the actual concentrations measured within the study. Twelve pet care products were analysed and it was found that the actual concentrations of parabens were lower than declared [39] (Table 16).

3.3.3.2. Import and export. The pet care products, i.e., shampoo, conditioners and skin creams, etc. intended for domestic animals but excluding products with medicinal purpose, do not have a unique commodity code and subsequently no specific statistics can be extracted from the Danish StatBank [90]. They are included in the general codes for personal care and grooming, custom code 330790 (“others”), i.e., pre-shave, shaving or aftershave preparations, personal deodorants, bath preparations, depilatories and other perfumery, cosmetic or toilet preparations, not elsewhere specified or included; prepared room deodorizers, whether or not perfumed or having disinfectant properties not governed by the other codes under 3307 [96]. Thus, import and export of pet care commodities are included in Table 14.

3.3.4. Pharmaceuticals and veterinary medicinal products

3.3.4.1. Presence. The SPC does not contain any information on the concentration of parabens in individual human pharmaceuticals as this not required by the statutory order [61] but approximately 2/3 of all the veterinary medicinal product sheets contain this information. Hence a literature survey of concentrations in a range of different human pharmaceuticals was conducted as show in Table 17. Reports on actual concentrations (in mg/g or mg/mL) were sparse in the literature found.

The Danish veterinary medicines were found not only to contain more methyl paraben than propyl paraben based on the presence in products (Table 8) but also based on the concentrations declared for the individual products (Table 18), where

Table 18

Concentration of parabens in veterinary medicinal products [40]

	MP	PP
In solid form – max (mg/g)	2.6	0.6
In solid form – min (mg/g)	0.8	0.1
In liquid form – max (mg/mL)	1.8	0.5
In liquid form – min (mg/mL)	0.5	0.1

Table 19
Potential max and min contribution of parabens based on pharmaceuticals (tonnes)

	2000	2001	2002	2003	2004
In solid form – max	0.085	0.098	0.098	0.093	0.098
In solid form – min	0.035	0.041	0.040	0.038	0.040
In liquid form – max	0.858	0.905	0.867	0.838	0.814
In liquid form – min	0.234	0.247	0.236	0.228	0.222
Total max	0.94	1.0	0.96	0.93	0.91
Total min	0.27	0.29	0.28	0.27	0.26

methyl paraben was found in higher concentrations for both solid and liquid products.

3.3.4.2. Import and export. The import and export of pharmaceuticals and veterinary medicines are covered by the custom code 3000 (Pharmaceutical products) which covers everything from groups of vaccines, and band aids to protective closes used in surgery [91]. The official public available information is however not detailed enough to find information on individual or specific groups of chemicals. Import and export statistics of sub-groups (code 3003) “pharmaceutical consisting of two or more products for therapeutic or prophylactic use” and (code 3004) “mixed or un-mixed for therapeutic or prophylactic use” for 2004 showed that the total import were 21,100 tonnes and the export 16,400 tonnes, i.e., a surplus of 4700 tonnes. As there is no further information how many pharmaceutical preparations this corresponds to, it is not possible to calculate the paraben import and export within pharmaceuticals and veterinary medicine.

However, it is possible to use the information in the SPC database and the declared paraben presence in veterinary medicines in order to calculate the parabens spread within Denmark due to the use of pharmaceuticals and veterinary medicines (Table 19). It is assumed that the human pharmaceuticals contain the same amount of parabens as the veterinary medicines as indicated in Tables 17 and 18. The sales statistics retrieved showed data for only 133 out of the 178 paraben containing pharmaceuticals as the remaining products had not been sold during the investigated time period [99]. For 123 an evaluation based on volume per package needed to be performed.

The amounts of parabens used in pharmaceuticals were calculated according to:

$$\begin{aligned} & \text{Number of packages sold} \times \text{amount or volume of} \\ & \text{pharmaceutical per package} \\ & \times \text{concentration of parabens} \times \frac{133}{123} \text{ products} \\ & = \text{total amount of parabens} \end{aligned} \quad (1)$$

For 2004, the pharmaceuticals would contribute with 0.26–0.91 tonnes of parabens, primarily in liquid form (Fig. 2).

Danish Institute for Food and Veterinary Research collects statistics on consumption and sale of veterinary medicine products in Denmark (VetStat). An excerpt from the database for 2005 showed that only 37 of the 41 paraben containing prod-

Table 20
Potential contribution of parabens based on veterinary medicine products (2005) in tonnes

Paraben type	Paraben volume (tonnes)
Number of products	37
MP	0.0994
PP	0.0092
Total	0.109

ucts had been marketed/used in 2005 [100]. The SPC database included declared concentrations of parabens for 30 out of the 37 veterinary medicine products in the range shown in Table 18. The remaining 7 products were estimated to contain the highest range of parabens found, i.e., 2.6 mg/g of MP and 0.6 mg/g of PP. No data could be obtained for 2004 but the data for 2005 is estimated to be close to what had been used during the prior year (Table 20). The amounts of parabens in veterinary medicinal products were calculated according to:

$$\begin{aligned} & \text{The annual total amount or volume of veterinary medicine} \\ & \text{products} \times \text{declared concentration of parabens} \\ & = \text{total amount of parabens} \end{aligned} \quad (2)$$

Thus, the total contribution of parabens in veterinary medicines is estimated to be 0.1 tonne (Fig. 2).

3.3.5. Import and export of food products and additives

3.3.5.1. Presence. Based on the questionnaire to 21 Danish food producers it is not anticipated that parabens are used frequently. Thus it is estimated that only 2% of the food permitted to contain parabens actually do so and that the levels used are the maximal permitted concentrations, see Table 2.

3.3.5.2. Import and export.

3.3.5.2.1. Snacks, nuts and candy. The review of the import and export of snacks, nuts and candy shows that the import generally exceeds the export, i.e., there is a net inflow of snacks. However, Denmark exports potato based snacks; wine gum and jelly; as well as hard coated, sometimes filled candy hard candy [90].

Using the amounts of snacks (rice, potato, corn and wheat based) and candy (excluding chocolate and chewing gum) available in StatBank in combination with the maximal limit of 300 mg/kg as well as the estimation that only 2% contain parabens, an import of 0.2 tonnes and an export of 0.3 tonnes for 2004 is yielded (Table 21 and Fig. 2).

Using statistics for filled chocolate (with and without alcoholic content) [90] in combination with the frequency of paraben

Table 21
Estimated amounts of parabens in snacks, nuts and candy (in tonnes)

	1999	2000	2001	2002	2003	2004
Import (2%)	0.2	0.2	0.2	0.2	0.2	0.2
Import (10%)	1.2	1.0	1.1	1.0	1.1	1.2
Export (2%)	0.2	0.2	0.3	0.3	0.3	0.3

presence of 8.9% (see Section 3.2.5) and the detected concentration range 7.9–180 mg/kg [75] the contribution of parabens in filled chocolates can be calculated. Based on these figures <0.1–0.19 tonnes is imported, <0.1–0.13 is exported and a <0.1–0.18 tonnes is used by domestic industry.

3.3.5.2.2. Meat and fish. Based on statistics of sausages, dried sausages, processed meat products and products made from liver [90], the import to Denmark ranged during 2000 to 2005 from 4750 to 17500 tonnes and export ranged from 37100 to 47700 tonnes, thus the export vastly exceeded the import, especially pork-based food. If assuming that the products contained their maximum allowed amount of parabens (1000 mg/kg) and that 2% of the products contained parabens, it corresponds to an import of 0.095–0.74 and an export of 0.35–0.96 kg parabens on an annual basis. However, the Danish food producers report that they do not use parabens as found in Table 9; hence the exported amount may be an overestimation.

One fish spread (buckling pâté) has been confirmed to contain 95 mg/kg [76]. The Danes consume 11–17 g of fish and fish products per day, which corresponds to 28,400 tonnes per annum [101]. Herring and mackerel are the most frequently eaten fishes but even if we assume that 20% of the whole fish consumption consists of buckling pate, the paraben contribution would be less than 10 kg per annum.

3.3.5.2.3. Seasoning. Although seasoning should be paraben free as described in the legislation, an estimation of how much parabens that theoretically could be imported via soy sauce was made as parallel and illicit import cannot be excluded. In 2004 originated 26% of the soy sauce (code no. 2103100000) imported to Denmark from countries that have reported paraben presence in soy sauce [49,78–80], i.e., China, Korea and Taiwan. If the imported soy sauce contained the amounts of paraben reported by Lin and Choong [80] which ranged from 33.4 to 250 mg/L soy sauce, the import of parabens via soy sauce could have been in 2004 between 0.010 and 0.077 tonnes. No official institution in Denmark collects detailed official statistics of other seasonings such as fish sauce and thus no similar comparison could be made.

3.3.6. Food supplements

According to the StatBank, 6170–7180 tonnes of Vitamins and mineral preparations (non-pharmaceuticals) in aqueous solutions were imported to Denmark in 2004–2005, 2980–3270 tonnes were exported and 4850–4960 tonnes produced nationally [90]. None of the pharmaceutical Vitamins and mineral preparations was found to contain parabens (Table 6) but one Danish food supplements producer reported to use parabens in his products (Table 9), thus the market share of paraben containing food supplements is estimated to be 2%. The maximum level of parabens in food supplement is set to 2000 mg/kg (Table 3) indicating that the food supplement could contribute with import of 0.3 tonnes, export of 0.1 tonnes and national consumption of 0.2 tonnes of parabens during production (Fig. 2).

3.3.7. Domestic and industrial commodities

The number of industrial and commercial commodities reported in the SPIN database [28] to contain parabens, as

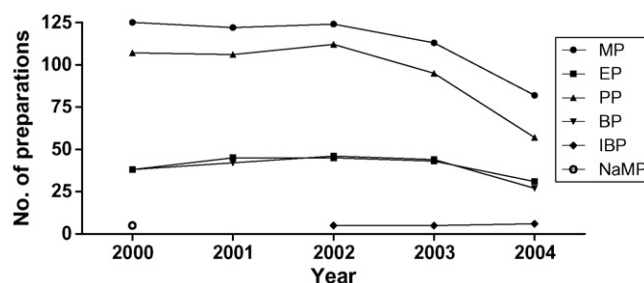


Fig. 1. Trends of use of parabens in commodities in Danish industry and commercial business.

seen in Table 10 and Fig. 1 is decreasing with respect to the parabens included in this study. The cumulative amounts, assuming that <0.1 category comprises of 0.09 tonnes ranges from 0.8 to 3.3 tonnes per annum (Fig. 2).

Personal communication with a Danish personal care products manufacturer and a Danish pharmaceutical company revealed that they when carrying out reformulation of their products exchange parabens for other preservatives. Similarly, one personal care products manufacturer will develop paraben-free baby products due to demand in the Danish society, but reformulation of existing products for adults have not been considered.

3.3.8. Other domestic products and industrial products

Water-based paints for indoor and outdoor purposes, and paint for dyeing for leather and textiles as well as glass and porcelain colours, were estimated to contain 0.27% parabens corresponding to the biocide level reported by a major paint manufacturer, see above. Some dyes, for example textile dye and dye pens were found to contain higher concentrations of parabens (0.6%) [52] but since it was not possible to distinguish between the different commodities in the custom service information all are treated under the same umbrella. Paraben containing paint is estimated to have a 2% market share and in combination with retail statistics of water-based paint (Table 22) from the StatBank in 2007 [90] it yields an import of 1 tonne, export of 1.3 tonnes and internal consumption of 3.1 tonnes of paraben for 2004.

150–170 tonnes of spray paint are used in Denmark on an annual basis. In a study to the Danish EPA only one out of 142 tested spray paints contained parabens and the concentration in this paint was <0.1% [59], hence the annual contribution of parabens on the Danish market is <1.2 kg.

Sticky toys, modelling clay/clay dough, finger paint available via Danish statistics are not the public recorded in such detail that it is possible to extract information on these specific commodities [90]. But a Swiss study has shown that almost half of the finger paints contain parabens [86]. Modelling clay and finger paint are not recommended to children of 3 years or younger and it perceived that these activities cease at the age of 10, thus in the ages of 4–9 years, there were 0.42 million children in 2004. It is estimated that if 50% of the children use 400 g of finger paint, 600 g of modelling clay and 50 g of sticky toys per year, 50% with a content of 0.27% of parabens,

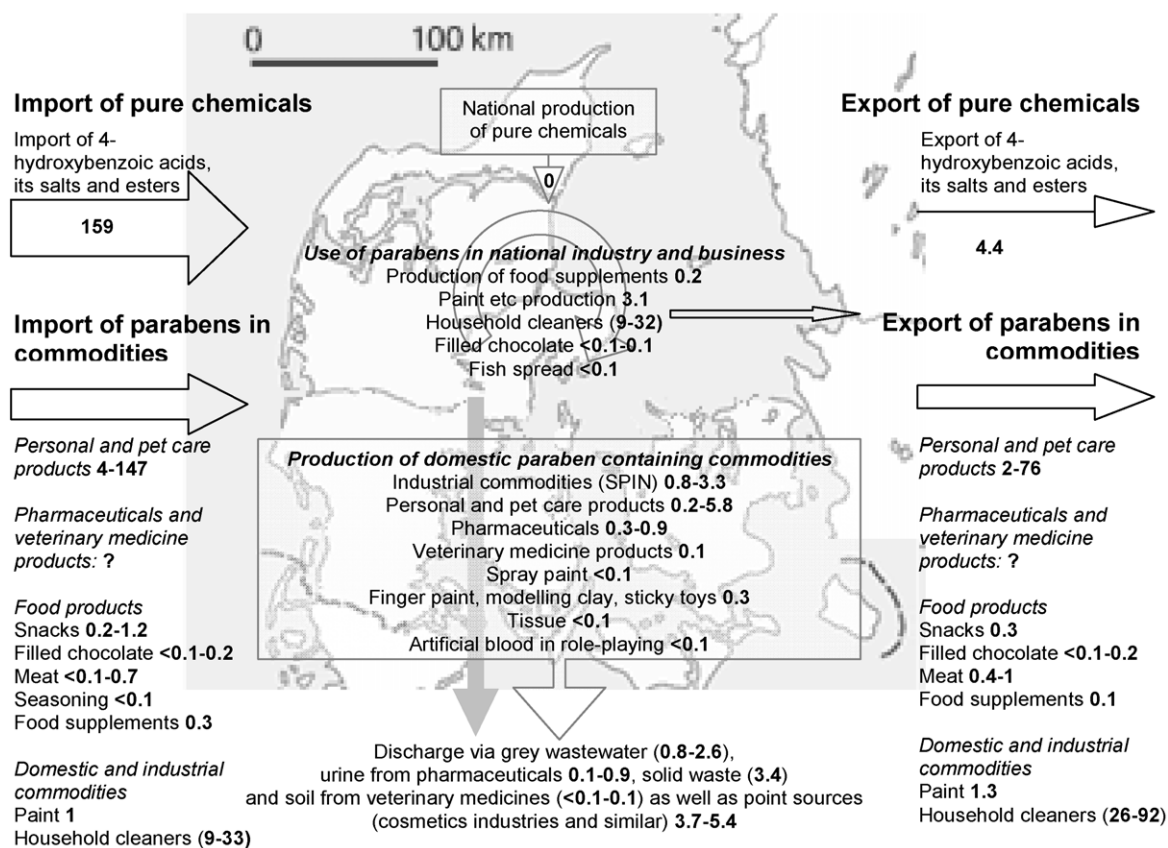


Fig. 2. Overview of the flow of parabens in the Danish society for the year 2004 (in tonnes).

these commodities would contribute with ca 0.3 tonnes in the Danish society (Fig. 2). The estimate is based on the input from two day-care centres and six parents with small (<10 year) children.

No studies or statistic of parabens in packing materials could be found. The use of industrial soap and its contribution to the parabens in the society are covered by SPIN, see Table 10.

National statistics of import and export of various household cleaners are gathered under one commodity code whereas the national production is available in a more detailed manner; hence, the data may not refer to identical commodities. If the household cleaners in average contained 0.7% a mixture of parabens [54] or 0.2% of a single paraben the input would in 2004 had been 9–33 tonnes import, 26–92 tonnes in export and 9–32 tonnes in national production. The frequency of paraben presence in cleaning commodities is estimated to be 11%, as shown in Table 11. This it however connected to high uncertainty as the calculations have been supported with much less data than for example the personal care products.

With an interview with role-playing retailers, it became clear that about 500 units of artificial blood were used in Denmark in 2005 and the product safety sheets listed the MP concentration to be 0.2% [88]. The size of the blood containers as indicated in the report [88] is small, <1 L, thus role-playing contributes with less than 1 kg of parabens.

Tissues used in Europe, e.g., kitchen rolls are predominantly used of virgin fibre [102] and each European is using 13 kg per year. If this also applied to Danes and the fact that 2 out of 11 virgin fibres contain parabens (Table 17) is used, it would indicate that every Dane used 2.36 kg of paraben containing tissue or 12,800 tonnes for the whole of Denmark. Vingaard et al. observed both MP and PP in virgin fibre kitchen rolls [51]. The highest levels confirmed were for PP 2.9 and 3.1 mg/kg, respectively. It corresponds to 37–40 kg parabens used in tissue on an annual basis, Fig. 2.

Neither for pleasure gels nor products for treatment of sports injuries and pains are there designated commodity codes available, thus, it is not possible to attain any statistic for their import,

Table 22
Import and export and national production of water-based paint (tonnes per annum) and calculated paraben contribution

		1999	2000	2001	2002	2003	2004	2005	2006
Water-based paint	I	14677 (0.8)	14183 (0.8)	14678 (0.8)	14791 (0.8)	15879 (0.9)	19332 (1.0)	27737 (1.5)	35638 (1.5)
	E	16625 (0.9)	19815 (1.1)	17827 (1.0)	18518 (1.0)	20858 (1.1)	24973 (1.3)	28304 (1.5)	36109 (1.9)
	N	52849 (2.9)	58941 (3.2)	59180 (3.2)	54856 (3.0)	53230 (2.9)	56555 (3.1)	52007 (2.8)	52333 (2.8)

Table 23

Comparison between actual and theoretical consumption of personal care product (in g/person/week) [67] and unpublished data from the same study

Product type	Actual consumption	Theoretical consumption (average 1999–2005)
Hair conditioner	3.0	12–39*
Hair styling products	0.6	
Deodorant	0.5	11–26**
Shaving foam	0.4	
Shower gel	2.1	
Hand soaps	8.2	10–16
Oral hygiene products	4.4	5–14
Shampoo	11.8	0***–10
Skin care products	2.5	1–6

*Hair tonic and hair care, **Beauty treatment incl. sun lotion, ***Negative inflow.

export, national production or general use [56,57]. Other uses such as in biocidal products and embalming fluids has been reported with the typical level of 0.7% [54] more information has not been possible to obtain due to the confidentiality of industrial products [89].

All the information collated within this study is presented in Fig. 2 which represents the sources and flow of parabens. The majority of the data has been available for the year 2004, thus this year has been selected to represent the substance flow analysis of parabens in Denmark.

3.4. Accumulation in society

Personal care products and cosmetics are required to be labelled with the year and month for minimum durability. However, if the durability is more than 30 months, it shall be indicated how long the product can be used after opening without any harm to the consumer shown with a symbol of an open container (76/768/EEC; [1]). In this study, it was found that the use of 12 months for minimum durability and an indication of 12 months keeping qualities as opened are the most frequently use for rinse-off and leave-on products available on the Danish market. Similarly, pharmaceuticals are provided with expiration dates labelled on the containers. These range from a few days to a few years depending on the pharmaceutical, but the paraben containing liquid solutions generally seem to have a shorter shelf-life than pharmaceuticals in tablet form. The Danish statutory order 1308 [103] regulates the labelling of minimum durability or ultimate “best before” for various food products. They are classified into three categories: <3, 3–18 and >18 months depending on the level of preservation. Thus, even if the food is preserved with parabens it has a relative short time period set a side for consumption. In general, the down-the-drain chemicals, pharmaceuticals and food are not commodities stored in households during long time periods or subjected to long-term accumulation in society and thus, the stock is not so relevant in comparison with the inflow.

Similarly, children’s modelling clay, sticky toys and finger paint have limited life-times whereas paint, for example indoor wall paint, may have a life time of 10–20 years. In all, the stock of parabens in the society perceived to be renewed every couple of years; hence, no significant accumulation is expected.

3.5. Comparisons and calibrations of the results

In order to validate the substance flow analysis, the outcome was compared with other studies, methods and approaches. Additionally, the flow to different waste fractions was calculated.

3.5.1. Personal care products

The annual balance of import, export and domestic production of personal care products (theoretical consumption; TC_{PCP}) can be compared with the actual consumption found in a tenants owners’ society in Copenhagen (BO90) [67,68].

The theoretical consumption was calculated as:

$$TC_{PCP} = \frac{(\text{import} + \text{national production} - \text{export})}{\text{No. of Danes} \times \text{weeks/year}} \quad (3)$$

Note that the Danish domestic production contains items both sold on the domestic market as well as items exported [93]. However, there is no data supporting the calculation of the separate amounts due to trade secrets, hence it is an estimate.

As can be seen in Table 23, the actual and the theoretical consumption of personal care product were found to be in the same order of magnitude. For all products were the commodity types not perfectly matched; for example, statistics for oral hygiene products and toothpaste were merged into one category and were found to be marginally larger than found in BO90. “Beauty treatment incl. sun lotion” also includes shaving products and deodorants thus a broad range of products (Table 14), more varied than found to have been used at BO90 and hence that theoretical consumption is larger than the actually seen consumption. Cosmetics were not included in the BO90 study so no such comparison could be made.

3.5.2. Pharmaceuticals and veterinary medicinal products

Here, it is assumed that all parabens in pharmaceuticals are transported to wastewater treatment plants and that all parabens in veterinary medicinal products are discharged directly to soil. It is an overestimation since, the Danish pharmacies are required to collect unused pharmaceuticals (2004/27/EC [104]), though detailed statistics are not available (based on type of or individual pharmaceuticals), which subsequently is sent for destruction via incineration. There may also be contribution from pharmaceuticals bought outside Denmark, e.g., during vacations which are not registered in the Danish system.

It has been found that in humans and dogs who digested parabens that they primarily excreted them via urine, either as the paraben itself (MP 83%, EP 50–95%, PP 50–95% and BP 40% [105] or as the acid of hydrolysis, 4-hydroxybenzoic acid. Thus, the pharmaceuticals excreted via urine contribute with 105–865 kg to wastewater and veterinary medicines with 44–104 kg primarily to soil and solid waste.

3.5.3. Dietary habits in the Danish population

The Danes eat 4 (adults) to 9 (children) grams of snacks per day on average [101] which yields a consumption of 8500 tonnes per annum. Correspondingly the inflow of snacks extracted from the StatBank [90] for 2004 is 10,200 tonnes, thus in total a discrepancy between the numbers of 1700 tonnes or 20%. Unfortunately, due to the lack on information of what kind of foods that was included in the reported food groups. No similar comparison could be made for any of the other potentially paraben containing foods.

3.5.4. Solid waste handling in Denmark

In Denmark garden waste, paper, glass, bulky wastes, electronic waste are collected and reused whereas hazardous waste are collected and secured. The new action plans for solid waste are related to the European framework waste legislation aiming at waste minimization and recovery. Many municipalities have during the last couple of years started of to collect plastic containers in recycling collection points along side, e.g., glass bottles. However, in general are personal care products packing and wrappings discarded into the remnant waste that is primarily incinerated. A minor questionnaire among the staff (35 staff members) at the Institute of Environment and Resources revealed that people usually throw away packing with residues of personal care products and household cleaners still inside the containers, i.e., they are not washed and dried before waste disposal. This was supported by the findings by Riber and Christensen [107] that non-rigid plastic, plastic bottles and clear glass contained 10–14% of water/moisture whereas tissue paper contained 47% of water/moisture.

According to the Danish StatBank [90] the annual domestic production of waste was in 2000, 1.7 million tonnes. We use the fact that 36–40% of the bottles, containers etc may have originally contained paraben preserved products (from Section 3.2.2) and assume that as much as 1% of the original product was remnant in the containers. Furthermore, it is assumed that all the commodities had contained the maximal level of one paraben of 0.4%. This yields a load of 3.4 tonnes of parabens to solid waste (Fig. 2).

3.5.5. Measured and plausible presence in wastewater

As most of the personal care products are rinse-off products, i.e., applied to the body, hair etc and then washed off, the paraben content will primarily be washed down the drain with the wastewater. Cosmetics applied onto the skin such as skin lotions may, however, result in dermal uptake and it has been found that 0.3% of BP was excreted in urine after application of a topical cream [106]. This is confirmed by an exposure index relying on volumes of chemicals and commodities, their closeness to

Table 24
Paraben balance overview

	Minimum (tonnes)	Maximum (tonnes)	
			<i>Import</i>
Pure chemicals	159	159	
Commodities	15	185	
National use	15	46	
			<i>Export</i>
Pure chemicals	4.4	4.4	
Commodities	30	171	
Estimated discharge ^a	8.1	13	
			<i>Inflow</i>
Pure chemicals	154	154	
Commodities	7.2	73	

^a To solid waste, soil, urine and greywater.

selected exposure targets and inherent properties, parabens are estimated to end up in the wastewater treatment plant in high numbers. MP and Na MP as “6”, EP and PP as “5”, BP, IBP and 4HBA as “4” on a scale from 1 to 7, where 7 corresponds to the highest exposure [113].

The average number of inhabitants in Denmark in 2004 was 5,401,320 people and the domestic use of water 250 million cubic meters [90], hence a daily consumption of 127 L/person. A survey of water use in Danish household yielded an estimation of that 74% of the water used in households’ generated grey wastewater and 37% originated from personal hygiene in bathrooms [108]. Parabens have been measured in two different bathroom derived grey wastewaters in Copenhagen. The collated concentrations of the parabens and 4-*para*-hydroxyl benzoic acid, if available, range from 4.2 to 22.2 µg/L [67,112]. Thus, it could be expected that 0.8–2.6 tonnes of parabens entered the wastewater stream on an annual basis via bathroom grey wastewater (e.g., wastewater without toilets; Fig. 2). In untreated wastewaters collated paraben concentrations between 0.4 and 4.4 µg/L have been observed [109] whereas in hospital sewer the concentrations ranged from below the limit of detection to 2.7 µg/L [110].

A grab sample taken from the effluent from a Swedish cosmetics factory was found to contain 190 µg/L of EP and 560 µg/L of PP [111], no similar investigation has been done in Denmark but it is assumed that similar facilities are present in Denmark. The mentioned cosmetics factory estimates that the maximum discharge of parabens are 2.4–3.4% of their total paraben use on an annual basis due to cleaning of production equipment and spills. Within a Danish perspective, this would correspond to 3.7–5.4 tonnes of point source emissions to wastewater if 2.4–3.4% of the imported pure chemicals are discharged in the described manor (Fig. 2).

The inflow of pure chemical was 154 tonnes whereas the inflow via commodities was 7.2–73 tonnes, Table 24. If this load of parabens is dispersed evenly by the total volume of water consumed in 2004 (250 million m³, see above), it yields wastewater concentrations of 640–900 µg/L. This is higher than actually noted in domestic wastewater but in the same order of magnitude as seen in industrial wastewater. It should be kept in mind

that it is an overestimation as not all the parabens used may enter the wastewater system and a significant part taken up by ingestion or dermal uptake will be metabolised and excreted as metabolites and degradation products. Hence, the estimations based on the substance flow analysis are feasible.

3.5.6. Uncertainty analysis

Here, it was found that single data, such as grab samples and statistics from one source, is common which limit the use of statistical evaluations or stochastic modelling. Moreover, reliability of data has come into question as faulty list of declaration, both with respect to the present substances as well as with respect to their content, has been observed for different skin creams and pet care products [30,39]. For skin creams, 22% were labelled incorrectly and too many, or too few, parabens were found by the chemical analyses compared with what was reported by the producer [30]. The presence of the acid of hydrolysis, 4BHA has only been noted in a few studies to be present in skin lotions [30], grey wastewater [67] and finger paint [85]. IPP has only been found in hair styling products [36] and several studies have been focusing on BP and IBP and neglecting information regarding the other parabens, hence biasing the material. This highlights the obstacles associated with collecting and evaluating information for SFA. In most cases, substances' presence in commodities, as percentage of the total volume, is unknown, or confidential, and thus can only be estimated from a few measurements or the limit values stipulated by the legislation. The observed consumer consumption patterns and the mapped use data may not be representative for the general public, so it cannot be disregarded that the outcome may have been quite different if more data had been available. Furthermore, statistical data not available for individual commodities or detailed groups of commodities, such as modelling clay and finger paint, due to the lack of custom commodity codes, which has severely hampered this study and has resulted in increased uncertainty as estimations based on larger pooled data had to be made. Assumptions made by the authors where no information could be retrieved also add to the overall level of uncertainty. Additionally, there may also be sources to parabens which have remained unknown to the authors.

4. Conclusions

Parabens are present in widely differing commodities such personal care products, pharmaceuticals, food as well as industrial and domestic commodities.

The import of parabens in Denmark is increasing, although the number of industrial commodities which contain parabens is decreasing and manufacturer reports phase-out of parabens in selected commodities. The commodity type contributing to the major inflow of parabens are the personal care products.

The average lifetime of the paraben stock is perceived to be limited to a few years since the vast majority of the paraben containing commodities are labelled with "best before" dates or maximum durability.

Data needed for the SFA is sparse, scattered and difficult to access, as statistics are not available in sufficient details or

should be handled confidentially. Thus, the study is associated with a great deal of uncertainty.

The calculated wastewater concentrations are in the same order of magnitudes as seen in industrial wastewaters but higher than that seen in domestic wastewater, indicating that the fraction of parabens entering the wastewater system may be overestimated. But also that metabolism and degradation are not sufficiently known.

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