Hazardous compounds released from textiles and the associated load they place on Swedish sewage treatment plants

Umeå Universitet
Peter Haglund
Lisa Lundin
Per Liljelind
Maria Hjelt

IVL Svenska Miljöinsitutet
Mikael Renberg
Lennart Kaj

Stockholms Universitet (ACES)
Tomas Alsberg

NATIONELL MILJÖÖVERVAKNING PÅ UPPDRAG AV NATURVÅRDSVERKET

Sammanfattning

Bakgrund

Hushållens bidrag av tvättvatten från textiltvätt till de kommunala avloppsreningsverken uppskattas till ca 2% av det totala volymflödet. Registerdata över kemikalier som används vid tillverkning av textilier/kläder samt analyser av tvättvatten visar att textilfibrer, mikroplastfibrer och många miljöstörande ämnen når våra reningsverk via textiltvätt. Dessa fibrer och kemiska ämnen kan bidra till förorening av avloppsreningsslam som används för gödsling av åkermark eller av vattenmiljö nedströms reningsverken.

Ett av Sveriges miljömål är En Giftfri Miljö och dess delmål innefattar bl.a. information om farliga ämnen i varor (Ds 2012:23). Textilier är en av de varugrupper som Miljömålberedningen föreslagit (SOU 2012:38) bli föremål för ett regeringsuppdrag avseende innehåll av farliga ämnen och riskbegränsande åtgärder samt frivillig miljömärkning. I det svenska miljömålsystemet ingår också generationsmål om att materialkretsloppen skall vara så fria från farliga ämnen som möjligt och att våra konsumtionsmönster av varor ska ge så små hälso- och miljöproblem som möjligt även i varornas tillverkningsländer utanför Sverige. Generationsmålet innebär att svensk politik behöver ta hänsyn till den miljö- och hälsopåverkan som svensk konsumtion orsakar i andra länder. EU:s ramdirektiv för avfall (2008/96/EG) har vidare slagit fast en avfallshierarki som sätter återanvändning av uttjänta varor, t. ex. kläder, före materialåtervinning av avfall.

Syfte

Syftet med denna studie var att undersöka i vilken utsträckning som vattentvätt av fem klädtyper (t-tröjor av bomull med plasttryck, bomullsjeans, arbetsbyxor, fleecetröjor samt allvädersjackor) bidrar till förekomsten av miljögifter i slam samt i utgående vatten från representativt utvalda svenska reningsverk.

Utförande

Kläder av fem olika klädtyper, enligt specifikation från naturvårdsverket (baserat på en tidigare studie av Swerea) köptes in från affärer i Umeå under januari 2014. Det var 8 st t-tröjor av bomull med plasttryck, 3 st bomullsjeans, 2 st arbetsbyxor, 8 st fleecetröjor samt 3 st allvädersjackor.

Kläderna tvättades i en vanlig tvättmaskin 2 gånger efter varandra utan att torka mellan och allt tvättvatten samlades upp. Delprov av tvättvattnet togs ut och analyserades för 126 utvalda ämnena på tre olika laboratorier (Miljökemiska Laboratoriet, Umeå Universitet, Svenska Miljöinstitutet (IVL) och Stockholms Universitet (ACES)). Ämnena var processkemikalier såsom pentaklorfenol, och triklosan, funktionskemikalier såsom ftalater och organofosfater samt oönskade kemikalier såsom dioxiner, klorfenoler och klorbensener.

Resultat

Studien visar på att det mängdmässigt främst är funktionskemikalier som släpper från kläderna vid tvätt. Det här var väntat då dessa kemikalier är avsiktligt och oftast inte kemiskt bundet till tyget. Processkemikalier avges i mindre mängd och oönskade kemikalier såsom till exempel klorerade fenoler och bensener hittades i väldigt små mängder i tvättvattnet oavsett vilken typ av klädesplagg som tvättats. Vare sig processkemikalierna eller de oönskade kemikalierna borde finnas i plaggen och därför var det väntat att dessa kemikalier inte skulle hittas i samma utsträckning som funktionskemikalierna.

Om man ser till detektionsfrekvensen, d.v.s. hur ofta de ämnen som ingår i en ämnesklass påträffas, blir bilden delvis en annan. Mer än 75% av de funktionskemikalier (38 av 50), ca 50% av funktionskemikalierna (26 av 49) och ca. 30% av processkemikalierna (8 av 27) detekterades i tvättvattenproverna.

T-tröjor och skaljackorna var de klädtyper som avgav störst mängd kemikalier per kg 47 mg/kg (0.005% w/w) för t-tröjor följt av 23 mg/kg (0.002% w/w) för skaljackor. Jeans, arbetsbyxor och fleecetröjor släppte mycket mindre mängd kemikalier 0.001, 0.001 and 0.0005% vid tvätt till tvättvattnet.

De fem klädtyperna släppte alla bisfenol AF, organofosfater, ftalater, formaldehyd, bromerade och klorerade fenoler samt klorerade bensener till tvättvattnet vid de två första tvättarna. Några föreningar som inte kunde detekteras i tvättvattnet var 4 stycken siloxaner, 9 stycken olika aniliner och majoriteten av de 17 dioxinerna och furanerna som ingick i studien.

T-tröjor släppte mer textilfibrer (0,85 mg/kg) jämfört med de andra klädtyperna. De andra klädtyperna släppte betydlig mindre fibrer vid tvätt: jeans 0,46 mg/kg, skaljackor 0,02 mg/kg, arbetsbyxor 0,07 mg/kg och fleecetröjor 0,1 mg/kg.

Diskussion

Om man tar hänsyn till den årliga användningsvolymen av de olika klädestyperna avger t-tröjorna den största mängden kemikalier (469 kg funktionella kemikalier, 0,5 kg processkemikalier och 0,07 kg oönskade kemikalier) vid de två första tvättarna av plaggen. Arbetsbyxor var den klädestyp som släppte minst kemikalier (30 kg funktionella kemikalier, 7 kg processkemikalier och 0,9 g oönskade kemikalier).

Ftalater och organofosfater frigjordes i stora mängder från kläderna (302 kg och 7,6 kg) och bidrar med 50% respektive 5% vardera till vad som återfinns i utgående vatten och slam från avloppsreningsverken. Klorfenoler och perfluorerade ämnen frigjordes i betydligt mindre mängder (430 g och 300 g) men bidrar i teorin med mer (167% respektive 223%) än vad som återfinns i utgåendevatten och slam från avloppsreningsverken för respektive grupp, vilket är orealistiskt. Brister i dataunderlaget eller degradering av föroreningar i reningsprocessen kan vara möjliga orsaker till överskattningen. Det är dock klart att tvätt av kläder ger ett betydande bidrag till vad som återfinns i reningsverksvatten och slam.

Slutsatser

Kemikalier som är förbjudna enligt t ex Reach ska naturligtvis inte förekomma i kläder. Trots det så hittas de ändå ibland vid inspektion. Det är ett stort problem eftersom kemikalierna fortfarande kan vara lagliga att använda i visa länder. Exempelvis är det förbjudet att använda arylaminer inom EU, ändå återfinner vi en av dessa 4,4'-diaminodiphenylmethane i tvättvatten från alla typer av kläder i denna studie. Idag sker produktion av kläder över hela världen och det är svårt att få information om vilka kemikalier som har använts för ett visst plagg. Denna spårbarhet skulle behöva förbättras.

I den här studien har vi hittat 72 av 126 föreningar, alla icke-naturliga föreningar, i tvättvattnet. De föreningar som frigjordes i störst mängder till tvättvattnet i den här studien var BPS, ftalater (DBP, BBP, DEHP, DINP, DIDP), DINCH, organofosfater (TPP, TCEP, TCPP, TEHP, TBEP) och formaldehyd. Med hänsyn taget till nettotillförseln av nya kläder kommer den mängden kemikalier på årlig basis som avges från nya kläder som tvättas de två första gångerna att vara betydande.

Även om en del av de föreningar som avges från kläderna kommer att brytas ner under behandlingen av avloppsvattnet i avloppsreningsverken så kommer många av dem att hamna i det utgående vattnet eller i slammet. Dessa kommer hamna i recipienten eller där slam används för att tillföra näringsämnen.

Fortsatt arbete

För att få en ännu bättre bild av hur mycket kemikalier som frigörs från kläder vid tvätt skulle det vara intressant att studera fler klädtyper. Det skulle också vara av intresse att analysera kläderna i sig för att kunna avgöra hur stor andel av det som finns i kläderna som avges vid tvätt, men också vad som finns kvar i kläderna när de så småningom blir textilavfall.

Slutligen skulle det vara intressant att genom så kallad ”non-target analysis” av både kläder och tvättvatten få veta vilka andra föreningar som förekommer i både kläder och tvättvatten. Rätt använt skulle ”non-target anaysis” kunna fånga upp ett brett spektrum av kemikalier och ge en ”totalbild” av substansflödet från textilier, via tvättvatten och reningsverk, till olika recipienter.

Summary

Background

Water from household laundry has been estimated to make up about 2% of the total volume flowing into municipal wastewater treatment plants (WWTPs). Records of the chemicals used in the manufacture of textiles/clothing and analyses of both washed clothes and laundry wastewater indicate that a large number of environmentally harmful substances can potentially reach treatment plants. These substances, including fibers and micro plastics from laundry, may contribute to the pollution of sewage sludge used for fertilization of arable land, or pollute the receiving waters downstream of wastewater treatment plants.

Textiles are one of the groups of consumer goods that the Environmental Objectives Committee proposed (SOU 2012: 38) be subject to a government mandate regarding the use of hazardous chemicals, environmental risk reduction measures and voluntary eco-labeling. The government has also decided (Ds 2012: 23) on interim measures aimed at removing toxic material from the environment, including providing information on hazardous substances in clothing. The Swedish environmental objectives system also includes the so-called “Generation target”, that states that material life cycles should be as free as possible from hazardous substances and that consumption of goods should produce as few health and environmental problems as possible, including in all the countries where they were manufactured. The Generation target means that the Swedish government needs to take into account environmental and health impacts that Swedish consumption may cause in other countries. The EU Waste Framework Directive (2008/96 / EC) defines a waste hierarchy that puts the recycling of old products, such as clothing, before the recycling of waste. This study may inform those working on developing such directives.

Aim

The purpose of this study was to examine the extent to which laundering of five types of clothing (cotton t-shirts, cotton jeans, work trousers, fleece sweaters and weatherproof jackets) contributes to the presence of toxic pollutants in sludge and effluent water from a representative sample of treatment plants.

Experimental

The choice of clothing was based on the study “Kartläggning av kemikalieanvändning i kläder” (Swerea IVF, Report 09/52) and were purchased in Umeå during January 2014.

The different categories of clothing were washed twice in a washing machine, without drying them in between and all wastewater was collected from the washer. Immediately after washing, samples of this water were transferred into 2 L glass containers and were analyzed for 126 compounds by three different laboratories (Miljökemiska Laboratoriet, Umeå Universitet, Svenska Miljöinstitutet (IVL) och Stockholms Universitet (ACES)).

Results

The results show that the main types of chemicals that were released when the clothing was washed, regardless of the type of clothing, were process and functional chemicals. This was expected since functional chemicals are added to the garment and are usually not chemically bonded to the fabric, whilst the process chemicals should not be present in the final product at all. Chemicals belonging to the group unwanted chemicals were released in very small amounts to the wastewater whatever type of clothing washed.

The functional chemicals represented 30 % of the analyzed target compounds but accounted for up to 99% (for t-shirts) of the release when the clothing was washed. The lowest contribution of functional chemicals to the total release of chemicals was from weatherproof jackets. Process chemicals dominated those released from weatherproof jackets (90%) and fleece sweaters (72%); for working pants, the contribution was 41%. The unwanted chemicals were present in much lower amounts in the laundry wastewater than the functional and process chemicals: they represented 1% or less of the chemicals detected.

T-shirts is estimated to release the largest amount of chemicals (469 kg functional chemicals, 0.5 kg process chemicals and 0.07 kg unwanted chemicals) based on the yearly net supply and the first two washing cycles. Fleece sweaters released the least amount of chemicals; 1.8 kg functional chemicals, 2.9 kg process chemicals and 3 g unwanted chemicals.

Phthalates, DINCH (a phthalate substitute), bisphenols, formaldehyde, and organophosphates were the groups of chemicals estimated to be released in largest amounts from the five types of clothing included in the study, contributing 47%, 25%, 12%, 12%, and 3%, respectively, to the total amount.

Based on the yearly net supply of clothing included in this study, the estimated release of textile fibers varies between 100 kg for fleece sweaters up to 8,500 kg for t-shirts. T-shirts released 0.85 mg fibers per kg, jeans released 0.46 mg/kg, weatherproof jackets 0.02 mg/kg, working pants 0.07 mg/kg and fleece sweaters 0.1 mg/kg.

Discussion

Phthalates and organophosphates were estimated to be released in large amounts (302 kg and 7.6 kg) contribute with 50% and 5% respectively to the amounts found in effluents from wastewater treatment plants. Chlorophenols and perfluorinated compounds were estimated to be released in very low amounts (430 g and 300 g respectively). This is however still more, 167% and 223% respectively, than what is found annually in the effluents and sewage sludge of all Swedish WWTPs.

The estimated contribution to sewage sludge for the different compound classes was far higher than the calculated contribution to effluent. The estimation produced a contribution figure of over 100% for some compound groups. Short chain chloroparaffins and chlorophenols were estimated to contribute to the amount found in sewage sludge to such a large degree that it exceeded what is actually found in the sewage sludge. Chlorophenols are distributed between both effluent and sewage sludge, but reference data was only found for sludge, so this could be the reason for the overestimation of the amount that ends up in the sewage sludge. It can also not be excluded that the selection of clothing was not representative of what is on the market.

Conclusions

Chemicals that are banned according to legislation such as Reach should, in principle, not be present in clothing. Even so, they are sometimes found during inspections of manufacturing facilities and analyses of clothing. This is a large problem since the use of a chemical can be banned in some countries but not in others. Arylamines are, for example, forbidden within the EU, but one of those 4,4'-diaminodiphenylmethane could still be detected in all types of clothing. Now, the clothing that we wear comes from all over the world, and it is difficult to find information on which chemicals have been used in its production since that can take place in many different countries. This tractability needs to be improved.

In this study, we detected 72 out of 126 compounds that are non-naturally occurring compounds, in the laundry wastewater. Among the compound groups that could not be detected were anilines, triclosan, triclocarban, and siloxanes.

The compounds released in large amounts into the laundry wastewater in this study were the process chemical bisphenol S (BPS), and the functional chemicals phthalates (DBP, BBP, DINP, DIDP), DINCH, organophosphates (TPP, TCEP, TCPP, TEHP, TBEP) and formaldehyde. Considering the net supply of new clothing to Sweden, the estimated annual contribution of the release of such compounds from new clothing being washed for the first time will be substantial.

Even though some of these chemicals will be degraded during the treatment process in the WWTP, many of them will end up in effluent or sewage sludge and, to different degrees, contribute to the compounds that risk ending up in WWTPs or where nutrients are recycled from sewage sludge.

Future work

To obtain a better picture of the volume of chemicals flowing to the WWTPs and, potentially, the environment, originating from the laundering of clothing, it would be of interest to study the release of chemicals from a broader range of clothing types. It would also be interesting to include analysis of the fabric to see what proportion of chemicals are released during laundry, and what proportion remain and are then potentially released during later washing or enter the textile waste stream.

It would also be of great interest to carry out non-target analysis on both the textiles and the wastewater to form an even broader picture of which chemicals are present in the textiles and the wastewater.

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1. Aim

The purpose of this study was to examine the extent to which laundering of five types of clothing (cotton t-shirts, cotton jeans, work trousers, fleece sweaters and weatherproof jackets) contributes to the presence of toxic pollutants in sludge and effluent water from a representative sample of treatment plants.

In this study, brand new clothes made of cotton and synthetic fibers were washed to see if 126 selected substances could be found in the laundry wastewater.

The study will also contribute to knowledge about the release of micro plastic and other fibers from the laundering of textiles.

2. Project administration

This study has been coordinated by Umeå University (UmU).
The following people have been involved in the work:

UmU	Peter Haglund, Professor
UmU	Lisa Lundin, Fil. Dr.
UmU	Per Liljelind, Fil. Dr.
UmU	Maria Hjelt, Fo. Ing.
IVL	Mikael Remberger
ACES	Tomas Alsberg

Contacts at the Swedish EPA:
Linda Linderholm
Britta Hedlund

3. Background

Water from household laundry has been estimated to make up about 2% of the total volume flowing into municipal wastewater treatment plants (WWTPs). Records of the chemicals used in the manufacture of textiles/clothing and analyses of both washed clothes and laundry wastewater indicate that a large number of environmentally harmful substances can potentially reach treatment plants. These substances, including micro plastics in laundry, may contribute to the pollution of sewage sludge used for fertilization of arable land, or pollute the receiving waters downstream of wastewater treatment plants.

Textiles are one of the groups that the Environmental Objectives Committee proposed (SOU 2012: 38) be subject to a government mandate regarding the use of hazardous chemicals, environmental risk reduction measures and voluntary eco-labeling. The government has also decided (Ds 2012: 23) on interim measures aimed at removing toxic material from the environment, including providing information on hazardous substances in clothing. The Swedish environmental objectives system also includes the so-called "Generation target", that states that material life cycles should be as free as possible from hazardous substances and that consumption of goods should produce as few health and environmental problems as possible, including in all the countries where they were manufactured. The Generation target means that the Swedish government needs to take into account environmental and health impacts that Swedish consumption may cause in other countries. The EU Waste Framework Directive (2008/96 / EC) defines a waste hierarchy that puts the recycling of old products, such as clothing, before the recycling of waste. This study may inform those working on developing such directives.

3.1 Chemicals in clothing

The Swedish Chemicals Agency recently published a report (KemI, Report 3/15) that concluded that our consumption of textiles is increasing and thus the use of chemicals. In Sweden, consumers buy about 14 kgs of textiles per person per year; for the EU as a whole, consumption is 19 kg.

In the manufacture of clothing, from the production of fiber (natural or synthetic) and the preparation of yarn or fabrics, to the production of the actual garments (making up), many different chemicals (KemI, Report 3/15) are used. Chemicals used in the textile industry can be divided into processing, functional and unintentional chemicals. The largest releases of chemicals occur in connection with the production of the fiber and the garments. Most of these chemicals do not remain in the fabric/garment but there are some chemical residues from the manufacturing process (KemI Report 3/15) with some chemicals being added after the garment is complete. For instance, fungicides exist in some clothing to prevent fabrics/garments from becoming moldy during transport. Significantly, people with sensitive skin are advised not to wear new clothes before they are washed.

3.2 Chemicals included in the study

The Swedish EPA has reviewed the current literature in the field and compiled a list of 126 compounds for target analysis. The compounds can be divided into three groups: i. *Process chemicals* (27 compounds), used during the manufacturing of the garment; ii. *Functional chemical*s (49 compounds), that produce specific properties in the material of the garment, and iii. *Unwanted chemicals* (50 compounds), that are substances not used intentionally in the production of textiles or added to the garment. The 126 target compounds are listed in **Table 3-5** and described in the following sections.

3.1.1 Process chemicals

Pentachlorophenol (PCP) has been used by the textile industry since 1930 as a biocide. It has been banned in Sweden since 1978, and in the EU since 1992. Unfortunately, PCP is still used in many parts of the world.

Nonylphenol ethoxylates are used during the washing, dyeing and printing processes in the production of textiles. They are odorless, light yellow liquids or waxes. They are molecules that contain a lipophilic (fat-friendly) nonylphenol part and a hydrophilic (water-friendly) ethoxylate part. Since the molecule contains these two different parts that prefer different surroundings, the molecule will lie in the interface between water and fat. Nonylphenol ethoxylates have been detected in grey water (Almquist and Hanaeus, 2006) as well as incoming and outgoing water and sludge from WWTPs (IVL, Report B1934, UmU screening 2012 and 2013).

Dyes can be derived from chloranil, which can then be found in those dyes as an impurity.

3.1.2 Functional chemicals

Functional chemicals remain in the fabric/garment and can have negative effects on the user. These chemicals often have physico-chemical properties that make them either persistent, bioaccumulative or toxic, or a combination of the three. If the chemicals are present in the laundry wastewater, they can enter the environment if municipal WWTPs are unable to clean the sewage sufficiently thoroughly.

Antibacterial chemicals are typically used to give textiles improved resilience against microorganisms (e.g. preventing destruction of polymers, discoloration) and to protect textiles against colonization by odor-forming bacteria. Antibacterial chemicals can also be added to provide the consumer with a product requiring less frequent or less harsh washing. Antibacterial substances that might be found in clothing are triclosan and triclocarban which are used for their bactericidal properties to prevent bad odor from, among other things, sweat. Triclosan is a skin irritant and can cause long-term adverse effects to the aquatic environment. The use of antibacterial substances can lead to the removal of beneficial bacteria and the generation of harmful, resistant strains.

Flame retardants are used to delay the ignition of textiles. Most of the flame retardants used are persistent because they function over the lifetime of the garment. This means that the flame retardant can spread far from the source and accumulate in the environment. Some of the flame retardants which may occur in cotton and synthetic materials are short chain chloroparaffins (SCCPs) and organic phosphate esters (OPs) such as tri (2-chloroethyl) phosphate and tris (2,3-dibromopropyl) phosphate. The latter can also be used as a plasticizer and is not chemically bound to the polymer but can move freely in and out of the material.

Perfluorinated compounds (PFCs) are a class of emerging persistent organic pollutants that consist of a fully fluorinated hydrophobic alkyl chain attached to a hydrophilic end group. Due to the unique structure of the fluoride-carbon bond, they exhibit significantly stable thermal and chemical properties. In the textile industry, they are used to produce materials that are waterproof, breathable and grease and dirt repellent; they are used in both cotton and synthetic materials. Siloxanes are also used for this purpose.

In recent years, large prints, often made of PVC, on t-shirts have become more popular. PVC is normally a hard plastic, so to make it possible to make it into a soft print on a t-shirt, softeners such as phthalates are needed. Phthalates are not banned in clothing; indeed, they are used in toys and childcare products. They are released from the plastic and can be absorbed by the body. Three phthalates – diethylhexyl phthalate (DEHP), dibutyl phthalate (DBP) and benzyl butyl phthalate (BBP) – have been classified as harmful to humans by the EU.

Formaldehyde can be used to provide resistance to dirt, for color fixation and fiber reinforcement, as well as to counteract shrinkage and wrinkling of textiles.

UV stabilizers are used to protect polymers, such as plasticized PVC, from degradation caused by light exposure, and can be found in printed textiles.

3.1.3 Unwanted chemicals

Dioxins and furans are not intended to be added to textiles but can be contaminates in other chemicals used in the production of the textile or formed during production. For instance, when pentachlorophenol is used, dioxins and furans can be found as contaminates in the final product. Pentachlorophenol is used as a biocide in the textile industry.

Arylamines are not used in the production of textiles, but are a degradation product of certain azo dyes. Arylamines are carcinogenic and banned in the EU.

4. Experimental set-up

The choice of clothing was based on the study "Kartläggning av kemikalieanvändning i kläder" (Swerea IVF, Report 09/52). In that study, the choice was based on the most commonly bought clothing in Sweden (SCB). **Table 1** and **Figure 1** show the different categories of clothing included in this study.

Table 1. Description of the clothing included in this study.

Type of clothing	T-shirts	Jeans	Working pants	Weatherproof jackets	Fleece sweaters
Number of pieces	8	8	2	4	**2**
Colors	Orange, red, blue, green, black and yellow	Dark blue	Black and grey	Blue, red and pink/purple (One, two and three layers)	Purple, green, blue, black, pink, red and white
Brand	Puma Jack&Jones InterSport + unknown brands	Lee, Levis, Diesel	HellyHansen PROF	Haglöfs, North Face and Norröna	Haglöfs, Adidas, Etriel, Tierra
Total Weight (kg)	**1.3**	**1.9**	**1.7**	**1.5**	**2.4**

The number of pieces of clothing were chosen to produce a total weight of 3 kg in each category. The t-shirts were estimated to weigh around 250 g each but, in reality, the mean weight was only 150 g. The jeans weighed around 450 g, the fleece sweaters 300 g, the working pants 780 and 900 g each and the all-weather jackets 273 g, 400 g and 830 g. In no category did we end up with a total weight of 3 kg. The colors of the t-shirts were specified by the Swedish EPA.

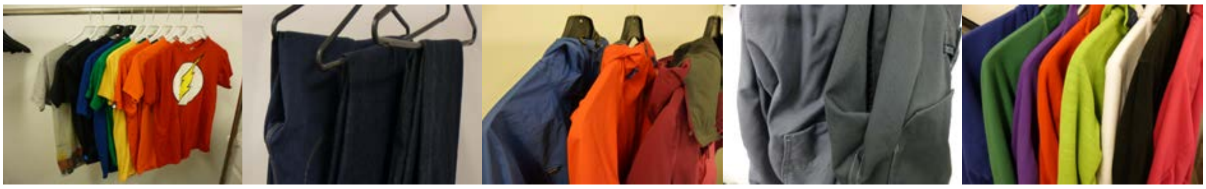

Figure 1. Picture of the clothing that was used in the study. From left to right: t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters.

4.1 Washing procedure

The different categories of clothing were washed twice in a washing machine, without drying them in between. The washing instructions for the clothing were followed. The machine used for the test was a Miele Softronic (front loading, Model W 3251). A popular household detergent was used (Via Color, Unilever). The wastewater was collected from the washer and directly split into two containers (**Figure 2**). Immediately after washing, samples of this water were transferred into 2 L glass containers.

Figure 2. Sampling set-up of wastewater from washing the clothing.

Table 2 shows the volume of detergent used, the laundry program used for each category of clothing and the resulting volume of water from the two washing cycles.

Table 2. Information on the washing of the clothing

Type of clothing	T-shirts	Jeans	Fleece sweaters	Weatherproof jackets	Working pants	Blank
Volume of detergent (mL)	20	20	20	20	20	20
Laundry program	Cotton, short program	Dark textiles/Jeans	Synthetic materials	Synthetic materials	Cotton	Cotton, short program
Temperature (ºC)	40	40	40	40	60	40
Total volume of water (L)	80	92	100	56	42	40

4.2 Compounds included in the study

The compounds included in the study have been categorized as process, functional and unwanted chemicals. **Tables 3 – 5** show all the 126 compounds included in this study, divided into compound category.

Table 3. Compounds included in the "Process chemicals" category.

NO.	CAS NUMBER	COMPOUND	ABBREVIATION
		Phenolic compounds	
19	87-86-5	Pentachlorophenol	PCP
23	128-37-0	Butylhydroxytoluene	BHT
24	80-05-7	Bisphenol A	BPA
25	80-09-1	4,4'-Sulfonyldiphenol	BPS
26	1478-61-1	Bisphenol AF	BF-AF
27	84852-15-3	4-nonylphenol, branched	4NP
28	140-66-9	4-tert octylphenol , branched	4tOP
		Alkylphenoletoxylates	
29	104-35-8	4-Nonylphenol (branched) -mono-ethoxylate	4NPEO1
30	20427-84-3	4-Nonylphenol (branched) -di-ethoxylate	4NPEO2
31	2315-67-5	4-tert-octylphenol-mono-ethoxylate	4tOPEO1
32	2315-61-9	4-tert-octylphenol-di-ethoxylate	4tOPEO2
		Chlorinated p-benzoquinones	
45	118-75-2	2,5-Cyclohexadiene-1,4-dione, 2,3,5,6-tetrachloro-	Chloroanil
		Arylmines (anilines/xylidines)	
89	95-69-2	4-chloro-2-methylaniline	4Cl2MeA
90	106-47-8	4-chloroaniline	4-ClA
91	87-59-2	2,3-dimethylaniline	2,3-DMA
92	95-68-1	2,4-dimethylaniline	2,4-DMA
93	95-78-3	2,5-dimethylaniline	2,5-DMA
94	87-62-7	2,6-dimethylaniline	2,6-DMA
95	95-64-7	3,4-dimethylaniline	3,4-DMA
96	108-69-0	3,5-dimethylaniline	3,5-DMA
97	90-04-0	2-methoxyaniline	2-MeOA
		Arylamines (others)	
98	92-67-1	4-aminodiphenyl	
99	122-39-4	Diphenylamine	DPA
100	101-72-4	N-Isopropyl-N'-phenyl-1,4-phenylenediamine	IPPA
101	91-59 -8	2-Naphtylamine	2-NA
102	(92-87-5)	Benzidine	
103	(101-77-9)	4,4'-Diaminodiphenylmethane	DADPM

Table 4. Compounds included in the “Functional chemicals” category.

NO.	CAS NUMBER	COMPOUND	ABBREVIATION
		Anti-bacterial / pesticides	
47	3380-34-5	Triclosan	
48	101-20-2	Triclocarban	
49	115-29-7	Endosulfan	
50	2385-85-5	Mirex	
		Quaternary ammonium compounds	
51	107-64-2	Dimethyldioctadecylammonium chloride	
52	8001-54-5	Benzalkonium chloride	
		Phthalates	
53	84-74-2	Di-n-butylphthalate	DBP
54	84-69-5	Di-isobutylphthalate	DIBP
55	85-68-7	Butylbensylphthalate	BBP
56	117-81-7	Di(2-etylhexyl)phthalate	DEHP
57	28553-12-0	Di(isononyl)phthalate	DINP
58	26761-40-0	Di(isodecyl)phthalate	DIDP
		Flame retardants/softeners	
59	85535-84-8	Short chained chloroparaffins C10-13	SCCP
60	115-86-6	Triphenylphosphate	TPP
61	115-96-8	Tris (2-chloroetyl) phosphate	TCEP
62	26248-87-3	Tris (3-chloro-n-propyl)phosphate	
63	13674-84-5	Tris (2-chloro-isopropyl) phosphate	TCPP
64	12645-31-7	Tris (2-etylhexyl) phosphate	TEHP
65	13674-87-8	Tris (1,3-dichloro-2-propyl) phosphate	TDCPP
66	78-51-3	Tris (2-butoxyethyl) phosphate	TBEP
67	31570-04-4	Tris(2,4-di-tert-butylphenyl)-phosphite	Irgafos 168
68	126-72-7	Tris(2,3-dibrompropyl)-phosphate	TRIS
69	38613-77-3	Tetrakis(2,4-di-t-butylphenyl)-4,4`-biphenylylene-diphosphonite	Irgafos P-EPQ
		Surface finishing agent / plasticizer and waterproofing	
70	541-05-9	Hexa methylcyclo-trisiloxane	D3
71	556-67-2	Octamethylcyclo-tetrasiloxane	D4
72	541-02-6	Decamethylcyclo-pentasiloxane	D5
73	540-97-6	Dodecamethylcyclo-hexasiloxane	D6
74	1763-23-1	Perfluorooctane sulfonic acid	PFOS
75		Perfluorobutyric acid	PFBS
76		Perfluorohexane soulfonic acid	PFHxS
77		Perfluorodecane sulfonic acid	PFDS
78		Perfluorohexanoic acid	PFHxA
79		Perfluoorheptanoic acid	PFHpA
80		Perfluoroctanoic acid	PFOA
81		Perfluorononanoic acid	PFNA
82		Perfluorodecanoic acid	PFDA
83		Perfluoroundecanoic acid	PFUnA
84		Perfluorododecanoic acid	PFDoA
85		Perfluorotridecanoic acid	PFTrA
86		Perfluorotetradecanoic acid	PFTA
87		Perfluoropentadecanoic acid	PFPeA
88		Perfluorooctanesulfonamide	PFOSA

Table 4 continued. Compounds included in the “Functional chemicals” category.

NO.	CAS NUMBER	COMPOUND	ABBREVIATION
		Stabilizers, incl. UV stabilizer	
104	6683-19-8	Tetrakis methylene(3,5-di-t-butyl-4-hydroxyhydrocinnamate)methane	Irganox 1010
105	85-60-9	4,4'-Butylidenbis(2-t-butyl-5-methyl)phenol	Lowinox 44B25
106	1843-05-6	2-Hydroxy-4-n-octoxybenzophenone	Chimasorb 81
107	2440-22-4	2-(2´-Hydroxy-5´-methylphenyl)-benzotriazole	UV P
108	25973-55-1	2-(2H-Bensotriazol-2-yl)-4,6-bis(1,1-dimethylpropyl)phenol	UV 328
46	126-86-3	TMDD (2,4,7,9-Tetramethyl-5-decyn-4,7-diol)	TMDD
126	50-00-0	Formaldehyde	

Table 5. Compounds included in the “Unwanted chemicals” category.

No.	CAS number	Compound	Abbreviation
		Polychlorinated phenols (PCPh)	
1		2-monochlorophenol	2-MoCPh
2		3-monochlorophenol	3-MoCPh
3		4-monochlorophenol	4-MoCPh
4		2,3-dichlorophenol	2,3-DiCPh
5	120-83-2	2,4-dichlorophenol	2,4-DiCPh
6		2,5-dichlorophenol	2,5-DiCPh
7		2,6-dichlorophenol	2,6-DiCPh
8		3,4-dichlorophenol	3,4-DiCPh
9		3,5-dichlorophenol	3,5-DiCPh
10		2,3,4-trichlorophenol	2,3,4-TriCPh
11		2,3,5-trichlorophenol	2,3,5-TriCPh
12		2,3,6-trichlorophenol	2,3,6-TriCPh
13		2,4,5-trichlorophenol	2,4,5-TriCPh
14		2,4,6-trichlorophenol	2,4,6-TriCPh
15		3,4,5-trichlorophenol	3,4,5-TriCPh
16	4901-51-3	2,3,4,5-tetrachlorophenol	2,3,4,5-TeCPh
17	58-90-2	2,3,4,6-tetrachlorophenol	2,3,4,6-TeCPh
18	935-95-5	2,3,5,6-tetrachlorophenol	2,3,5,6-TeCPh
		Polybrominated phenols (PBPh)	
20	118-79-6	2,4-dibromophenol	2,4-DiBPh
21		2,4,5-tribromophenol	2,4,5-TriBPh
22	608-71-9	Pentabromophenol	PeBPh
		Polychlorinated benzenes (PCBz), incl. nitrated benzenes	
33		1,2-dichlorobenzene	1,2-DiCBz
34		1,3- dichlorobenzene	1,3-DiCBz
35		1,4- dichlorobenzene	1,4-DiCBz
36	87-61-6	1,2,3-trichlorobenzene	1,2,3-TriCBz
37		1,2,4-trichlorobenzene	1,2,4-TriCBz
38		1,3,5-trichlorobenzene	1,3,5-TriCBz
39		1,2,3,4-tetrachlorobenzene	1,2,3,4-TeCBz
40		1,2,3,5-tetrachlorobenzene	1,2,3,5-TeCBz
41		1,2,4,5-tetrachlorobenzene	1,2,4,5-TeCBz
42		Pentachlorobenzene	PeCBz
43	82-68-8	Benzene, pentachlornitro-,	
44	81-19-6	Benzene, 1,3-dichloro-2-(dichloromethyl)-	

Table 5 continued. Compounds included in the "Unwanted chemicals" category.

No.	CAS number	Compound	Abbreviation
		Polychlorinated dioxins and furans (PCDD/Fs)	
109		2,3,7,8-Tetrachlorinated dibenzo-*p*-dioxin	2,3,7,8-TCDD
110		1,2,3,7,8-Pentachlorinated dibenzo-*p*-dioxin	1,2,3,7,8-PeCDD
111		1,2,3,4,7,8-Hexachlorinated dibenzo-*p*-dioxin	1,2,3,4,7,8-HxCDD
112		1,2,3,6,7,8-Hexachlorinated dibenzo-*p*-dioxin	1,2,3,6,7,8-HxCDD
113		1,2,3,7,8,9-Hexachlorinated dibenzo-*p*-dioxin	1,2,3,7,8,9.HxCDD
114		1,2,3,4,6,7,8-Heptachlorinated dibenzo-*p*-dioxin	1,2,3,4,6,7,8-HpCDD
115		Octachlorinated dibenzo-*p*-dioxin	OCDD
116		2,3,7,8-Tetrachlorinated dibenzofuran	2,3,7,8-TCDF
117		1,2,3,7,8-Penatchlorinated dibenzofuran	1,2,3,7,8-PeCDF
118		2,3,4,7,8-Pentachlorinated dibenzofuran	2,3,4,7,8-PeCDF
119		1,2,3,4,7,8- Hexachlorinated dibenzofuran	1,2,3,4,7,8-HxCDF
120		1,2,3,6,7,8- Hexachlorinated dibenzofuran	1,2,3,6,7,8-HxCDF
121		2,3,4,6,7,8- Hexachlorinated dibenzofuran	2,3,4,6,7,8-HxCDF
122		1,2,3,7,8,9-Hexachlorinated dibenzofuran	1,2,3,7,8,9-HxCDF
123		1,2,3,4,6,7,8-Heptachlorinated dibenzofuran	1,2,3,4,6,7,8-HpCDF
124		1,2,3,4,7,8,9-Heptachlorinated dibenzofuran	1,2,3,4,7,8,9-HpCDF
125		Octachlorinated dibenzofuran	OCDF

5. Chemical analysis

5.1 Chemical analysis at Miljökemiska Laboratoriet, Umeå University

5.1.1 Extraction

Prior to extraction, internal standards were added. The samples (ca. 1.5 L) were extracted by liquid extraction using dichloromethane (DCM) as the organic solvent. The laundry wastewater was extracted with 4 portions of 75 ml DCM for analysis of PCDD/F, organophosphates and chloroparaffins. The extract was then divided into three equal portions (1:1:1). In addition, two 0.5 L portions of laundry wastewater were extracted with 4 portions of 50 mL DCM for analysis of chlorobenzenes (PCBz) and phenolic compounds, respectively.

5.1.2 Clean-up of dioxins and furans (PCDD and PCDF)

The first clean-up stage used a multilayer silica column consisting of basic silica gel, neutral silica gel, acidic silica gel and sodium sulfate with n-hexane as the eluent. The sample was further purified using a column comprising a mixture of AX-21 carbon and Celite (7.9:92.1). The carbon columns were eluted with n-hexane, dichloromethane 1/1 (v/v) and toluene. The n-hexane and dichloromethane fraction was discharged, and the toluene fraction used for the determination of dioxins and furans. The toluene fraction was concentrated with 40 µl tetradecane after addition of a ^{13}C-labeled recovery standard prior to analysis by gas chromatography-high resolution mass spectrometry (GC-HRMS).

5.1.3 Clean-up of organophosphates (OPs)

A Florisil column was used to separate the organophosphates from other compounds. The column was washed with 100 mL of hexane-diethyl ether (9:1 v:v) prior to elution with 100 mL of methanol-diethyl ether (17:83 v:v). The methanol-diethyl ether fraction was then concentrated with 1 ml toluene. A ^{13}C-labeled recovery standard was added prior to analysis using GC-HRMS.

5.1.4 Clean-up of short chain chlorinated paraffins (SCCP)

A Florisil column was used to separate the organophosphates from other compounds. The column was washed with 100 mL of hexane-diethyl ether (9:1 v:v) prior to elution with 100 mL of methanol-diethyl ether (17:83 v:v). The methanol-diethyl ether fraction was then concentrated with 1 ml toluene. A ^{13}C-labeled recovery standard was added prior to analysis using GC-HRMS.

5.1.5 Clean up of polychlorinated benzenes (PCBz) and other related compounds

The extract was added to a column containing deactivated silica with 10% water and some Na_2SO_4 on top. The column was eluted with 80 mL of hexane. The extract was concentrated with 0.5 mL of toluene. A ^{13}C-labeled recovery standard was added prior to analysis using GC-HRMS.

5.1.6 Clean up of polychlorinated phenols (PCPh) and other phenolic compounds

The extract was concentrated and dried in a sodium sulfate column prior to derivatization with acetic anhydride. The samples were shaken for 1 hour to ensure complete derivatization. A ^{13}C-labeled recovery standard was added prior to analysis using GC-MS.

5.1.7 Analysis

PCDD/F, OPs, PCBz and related benzenes were analyzed using GC-HRMS (Waters AutoSpec Ultima) with a DB-5MS column (60m x 0.25 mm), electron ionization (EI) and selected ion monitoring (SIM).

SCCP and phenols were analyzed using GC-MS (Agilent 5975 MSD) with a ZD-5MS column (30m x 0.25mm). Phenols were analyzed using EI; SCCPs were analyzed using electron capture negative ion chemical ionization (ECNI).

5.1.8 Quality assurance/ Quality control

To ensure the correct concentrations were measured, the following rules for the analyses were followed:

- An internal standard should have similar chemical and physical properties to those of the compounds being analyzed. The project used C^{13} or D isotopes.
- Irrespective of the method of analysis, a calibration curve was determined for each substance and the linear range identified. All samples were adjusted so that the concentration of the target substance fell within this range.
- For the identification of the chosen substances, the compounds have to have identical retention times as the reference standards for PCDD/F, PCBz, PCPh. For the other compounds, native compounds in a separate standard are to be used to identify the correct retention time.
- The ratio of the two ions in the molecular ion-cluster must be within +/- 15% (according to EN / EN 1948: 3) for PCDD/F.
- For each batch of samples processed and analyzed, a number of so-called blanks i.e. a sample consisting of solvent treated with the real samples, should also be analyzed to check for background contamination from solvents and laboratory equipment.
- All glassware should be washed and then heated in the oven at 400 ºC. Solvents used should be of the highest quality and purity.
- Recovery concentrations of the internal standards should be calculated to verify that the method worked (acceptable limits are 50 – 120% of SS / EN 1948: 3).
- The laboratory participates annually in inter-calibration activities for dioxins and dioxin-like PCBs in different matrixes.

5.2 Chemical analysis at Svenska Miljöinstitutet (IVL)

5.2.1 UV stabilizers except Irgafos 168 and Irganox 1010

Ascorbic acid (100 mg) and EDTA (50 mg) were added to 35 g of sample prior to acidification with HCl to a pH of 2 – 3. The internal standard (UV9; CAS 2170-39-0) and IPA (3,5 ml) were added. The samples were extracted using hexane:MTBE (3:1) for 10 min. The emulsion produced during the extraction was broken up by vigorously shaking the extract with NaCl (2 g). The extract was dried over Na_2SO_4 and evaporated to dryness. The evaporation residue was dissolved in acetone and transferred to a new test tube, where it was evaporated again. The clean-up procedure to remove some undissolved matrix compounds in acetone was as follows. Reagent MSTFA was added and heated to 85°C for 15 min. Then, 1 ml of hexane and volumetric (injection) standard (biphenyl) were added. The compounds were analyzed using GC-MS/MS.

5.2.2 Anilines, DCHA and IPPD

The internal standard (d5-anilin, 4-t-butylanilin, 4-*iso*-propylanilin) was added to the sample (35 ml). KOH was added to basic pH together with NaCl (1 g). The samples were extracted with MTBE twice. The extracts were combined and dried over Na_2SO_4. HCl in methanol was added as a ”keeper”. The extract was then evaporated to dryness. The evaporation residue was dissolved in a carbonate buffer and reacted with *iso*-butyl chloroformate (IBCF). The derivative was extracted into hexane after the addition of a carbonate buffer. Volumetric standard (biphenyl) was added and the compounds were analyzed using GC-MS/MS.

5.2.3 Cancinogenic amines

The extraction and clean-up was carried out as for the anilines. Benzidine and 4,4'-Diaminodifenylmetan did not react with IBCF. The derivatization of these compounds was achieved using pentafluoropropic acid (PFPAA). The evaporation residue of the extract was dissolved in hexane:MTBE. Pyridine and PFPAA were added and the sample was incubated at 80 ° C for one hour. The reagent was washed away with phosphate buffer (pH 6). Volumetric standard (biphenyl) was added and the compounds were analyzed using GC-MS / MS.

5.2.4 Chloranil, TEHP and DPA

The internal standard (Benzylcinnamat) was added to the sample (15 mL) and extracted with hexane:MTBE (9:1). The extract was dried over sodium sulfate and concentrated (2 ml). TEHP and DPA can be analyzed with GC-MS without derivatization whereas p-chloranil needs to be reduced and derivatized. The extract was thus derivatized by reductive acetylation with acetic anhydride, sodium acetate and zinc powder at 85 °C for 45 min. The derivative was fractionated with a silica gel column. Volumetric standard (biphenyl) was added and the compounds were analyzed using GC-MS / MS.

5.2.5 BF-AF and BPS

The extraction and clean-up procedure was the same as that for UV stabilizers but hexane:MTBE (9:1) was used as the extraction solvent. This extract was derivatized with MSTFA and used for determination of BF-AF. The sample was then extracted again with aspects of the BPS with hexane:MTBE (1:1) after the addition of the internal standard (d14 bisphenol A) and NaCl (2 g). The concentrated and dried sample extract was derivatized with MSTFA (see UV filter). Volumetric standard (biphenyl) was added and the compounds were analyzed using GC-MS / MS.

5.2.6 Formaldehyde

To 20 mL of the sample, potassium hydrogen phthalate (KHP) was added as a pH buffer (pH 4) with 2,4,5-F acetophenone recovery standard. Freshly prepared reagent O-(2,3,4,5,6-penta fluoro benzyl) hydroxylamine (PFBHA) was added to the sample and mixed well. The sample was placed in an oven at 35 °C for 2 hours. The sample was shaken 2 – 3 times during the course of the reaction. It was then acidified with H_2SO_4 and extracted using hexane (4 mL). The extract was diluted ten times. A volumetric standard (pentachlorobenzene) was added prior to GC-ECD.

5.2.7 Siloxanes

Wastewater samples (8 ml) were extracted with 4 ml of n-hexane after the addition of an internal standard (hexaethyldisiloxane). The phases were separated using centrifugation. The extraction was repeated once. The combined extract was dried, evaporated to 0.5 ml and analyzed using GC-MS / MS.

5.2.8 Phthalates

The internal standard (3F-DEP, d4-DBP, d4-DEHP) was added to the waste water samples (10 ml) and diluted with 10 ml Milli-Q water. The sample was then concentrated on a C18, 500 mg column. The column was rinsed with 2 ml Milli-Q water and dried. The phthalates were eluted with 2 ml MeOH, 3 ml MTBE and finally 2 ml n-hexane. 6 ml Milli-Q water was added and the extract was shaken for 2 minutes and centrifuged. The organic phase was dried over Na_2SO_4 and concentrated with N_2. It was then cleaned on a PSA, 500 mg which was eluted with 5 ml n-hexane:MTBE (3:1). Prior GC-MS/MS analysis a recovery standard (biphenyl) was added to the sample.

5.2.9 Quality assurance/ Quality control

Blanks were analyzed, and to measure extraction efficiency internal standards and recovery standards were used. The retention time was expected to match those of the standard compounds to within ± 0.1 min. The ratios (measured using GC-MS) of the selected ions (target and qualifier) were within ± 15% of expected ratios. Spiking tests for evaluating the extraction efficiency were carried out. The signal-noise ratios were measured at less than 3:1.

5.3 Chemical analysis at Stockholm University (ACES)

5.3.1 Extraction and clean-up

Some 50 ml of the sample were ultra sonicated for 10 minutes. The 50 ml were decanted for extraction. Then, 0.25 ml formic acid and 8 mg NH_4Oac were added as along with 25 µl ISTD solution. The sample was then extracted using an Oasis HLB Plus SPE cartridge (0.25 g) eluted with 8 ml methanol, and concentrated to approx. 0.2 ml. Finally, 25 µl RSTD solution were added to the sample. The detergent caused problems when analyzing the samples with the LC / MS so the original samples were diluted and reanalyzed.

5.3.2 Analysis

The samples were analyzed using ultra-high performance liquid chromatography tandem mass spectrometry (UHPLC-MS/MS) (Waters Acquity UPLC/Xevo TQ-S equipped with a PFC isolator column and a Waters Acquity BEH C18 separation column (1.7 um, 2.1 x 50 mm with a 2.1 x 5 mm pre-column)) using a gradient elution, a five-point calibration curve, electrospray ionization in negative ion mode, and Multiple Reaction Monitoring (MRM) for data acquisition.

5.3.3 Quality assurance/Quality control

Blank: Laboratory Blank (method blank) consisting of 50 mL of HPLC water processed the same way as the sample wastewater.
Workup Matches: Sample processing losses and matrix effects were compensated for by using internal standards (ISTDs). Losses of ISTDs were controlled using recovery standards (RSTDs).
Precision and accuracy: The precision was controlled through duplicate analyses and participation in inter-calibration studies.

5.4 Gravimetric determination of released textile fibers (UmU)

To determine the weight of released textile fibers during the washing of the five different clothing types, between 1.5 and 3.5 litres of laundry wastewater were filtered through a 300µm mesh filter. The filters were weighed before filtration and after the filters (containing the textiles fibers) were dried in the fume hood.

5.5 Compounds omitted from the study

Due to difficulties during clean-up and analysis, the compounds listed in Table 6 have been omitted from the study.

Table 6. Compounds that, for various reasons**,** were omitted from the study.

No.	CAS number	Compound	Abbreviation
24	80-05-7	Bisphenol A	BPA
27	84852-15-3	4-nonylphenol, branched	4NP
28	140-66-9	4-tert-octylphenol, branched	4tOP
29	104-35-8	4-Nonylphenol (branched) -mono-ethoxylate	4NPEO1
30	20427-84-3	4-Nonylphenol (branched) -di-ethoxylate	4NPEO2
31	2315-67-5	4-tert-octylphenol -mono-eteoxylate	4tOPEO1
32	2315-61-9	4-tert-octylphenol -di-etoxylat	4tOPEO2
46	126-86-3	2,4,7,9-Tetramethyl-5-decyn-4,7-diol	TMDD
49	115-29-7	Endosulfan	
51	107-64-2	Dimethyldioctadecylammonium chloride	
52	8001-54-5	Benzalkonium chloride	
62	26248-87-3	Tris (3-chloro-n-propyl)phosphate	
67	31570-04-4	Tris(2,4-di-tert-butylphenyl)-phosphite	Irgafos 168
69	38613-77-3	Tetrakis (2,4-di-t-butylphenyl)-4,4`-biphenylylene-diphosphonite	Irgafos P-EPQ
98	92-67-1	4-aminodiphenyl	

5.6 Compounds added to the study

Two phthalates that are considered emerging compounds were added to the study:

- Di (2-propyl heptyl) phthalate (DPHP). It is a C10-phthalate replacing DINP.
- 1,2-cyclohexanedicarboxylic acid di-*iso*-nonyl ester (DINCH): the same molecule as DINP but not aromatic, it is saturated.

We also included hexachloro benzene (HCB) in the results.

6. Results

The results show that t-shirts and weatherproof jackets released most chemicals per kg, 47 mg/kg (0.005% w/w) for t-shirts followed by 23 mg/kg (0.002% w/w) for weatherproof jackets (**Figure 3**). Jeans, working pants and fleece sweaters released far less 0.001% w/w, 0.001% w/w and 0.0005% w/w respectively. The main types of chemicals that were released when the clothing was washed, regardless of the type of clothing, were process and functional chemicals (**Figure 3**). This was expected since functional chemicals are added to the garment and usually not chemically bonded to the fabric, whilst the process chemicals should not be present in the final product at all.

The functional chemicals represented 30 % of the analyzed target compounds but accounted for up to 99% (for t-shirts) of the release when the clothing was washed (**Figure 4**). The lowest contribution for functional chemicals to the total release of chemicals was from weatherproof jackets (8%). Process chemicals dominated those released from weatherproof jackets (92%,) and fleece sweaters (62%); for working pants, the contribution was 18%. The unwanted chemicals were present in much lower amounts in the laundry wastewater than the functional and process chemicals: they represented 1% or less of the chemicals detected.

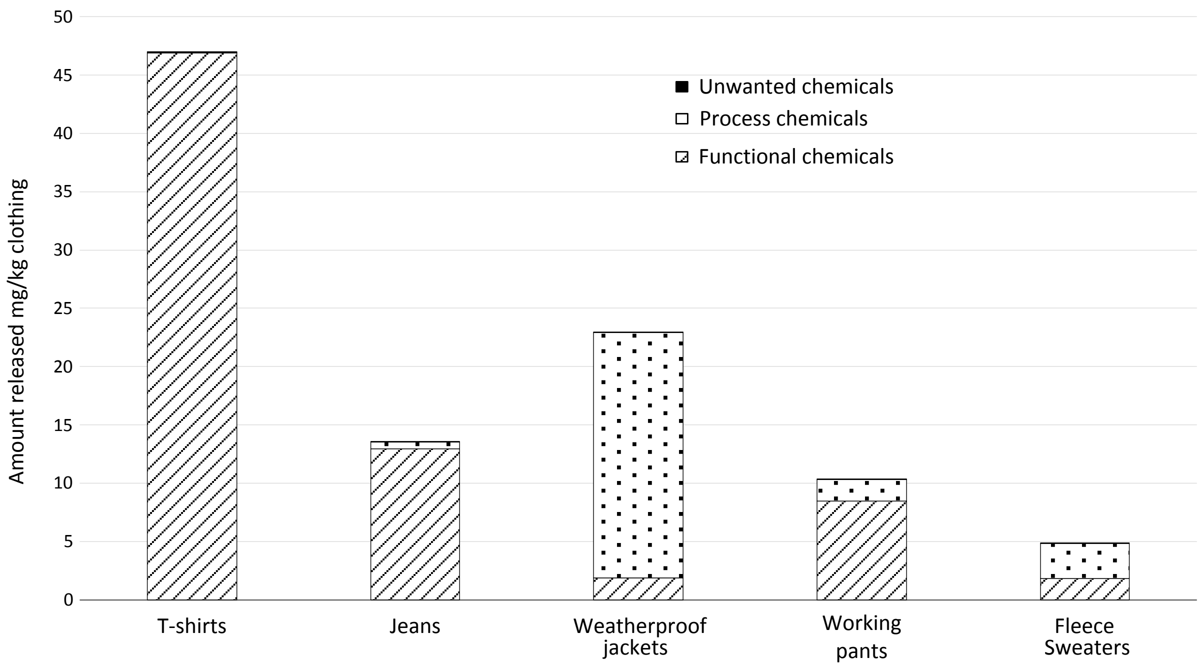

Figure 3. Release of chemicals from the five types of clothing divided into process, functional and unwanted chemicals, all in mg/kg of clothing**.**

From all five types of clothing, we detected the release of bisphenol AF, organophosphates, phthalates, formaldehyde, brominated and chlorinated phenols as well as chlorinated benzenes.

On the other hand, some of the compound groups could not be detected in any of the wastewater: these were siloxanes (4 compounds), anilines (9 compounds) and PCDD/F, except for the hepta-chlorinated and octa-chlorinated compounds.

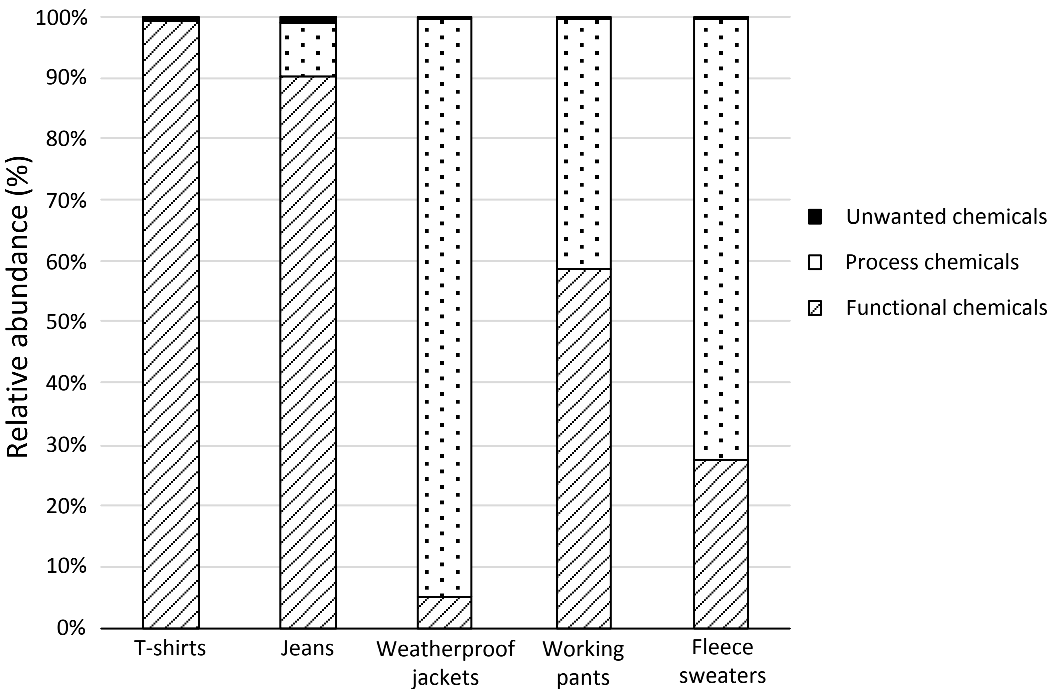

Figure 4. Relative distribution of chemicals released from the clothing when washed.

6.1 Release of Functional chemicals

Most of the chemicals released into the laundry wastewater in this study were functional chemicals (**Table 4**), used for a specific function in the clothing. From weatherproof jackets and fleece sweaters process chemicals contributed more (**Figure 4**). Forty-nine compounds out of 126 in this study were classified as functional chemicals. Of these 49 compounds, 26 were detected in the laundry wastewater. **Figure 5** show the distribution of the compound groups that were detected in the laundry wastewater from the five different clothing types. The concentrations of the individual compounds in the laundry wastewater are shown in **Table 7**.

Table 7. Concentrations of functional chemicals (µg/kg cloths) detected in wastewater from washing five types of clothing. Chemicals that were below limit-of-quantification are listed in Appendix 1.

Compound	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
Phthalates					
DBP	160	690	120	86	95
Di-isobutylphthalate	190	320	100	60	170
BBP	<120	570	130	<50	<83
Di(2-etylhexyl) phthalate	1600	450	410	1100	660
DINP	22000	<1500	<1100	1100	<1300
DIDP	4700	<1500	<1100	<740	<1300
DPHP*	n.d.	n.d.	n.d.	3000	n.d.
DINCH*	16000	<970	<750	2700	<830
Flame retardants/softeners					
SCCP (C10-13)	6.1	2.6	1.0	180	3.3
TPP	180	4.4	12	n.d.	0.63
TCEP	38	12	49	n.d.	12
TCPP	100	32	97	n.d.	26
TEHP	6.2	1500	2.1	n.d.	5.8
TBEP	12	380	21	n.d.	100
TRIS	0.62	<0.05	<0.04	<0.02	<0.04
Surface finishing and waterproofing agents					
PFDS	n.a.	n.a.	2.8	10	n.a.
PFHxA	n.a.	n.a.	50	<1.5	n.a.
PFHpA	n.a.	n.a.	11	3.2	n.a.
PFOA	n.a.	n.a.	3.8	4.2	n.a.
PFNA	n.a.	n.a.	<1.1	2.2	n.a.
PFDA	n.a.	n.a.	<1.1	1.1	n.a.
PFUnA	n.a.	n.a.	1.4	<0.74	n.a.
Stabilizers					
Lowinox 44B25	20	9.2	23	6.9	12
Chimasorb 81	58	<34	37	35	37
2-(2´-Hydroxy-5´-methylphenyl)-benzotriazole	330	<4.8	8.2	11	<4.2
2-(2H-Bensotriazol-2-yl)-4,6-bis(1,1-dimetylpropyl)fenol	33	12	82	6.1	13
Formaldehyde	1900	9200	630	250	630

n.a. not analyzed
n.d. below detection limit
* emerging phthalates

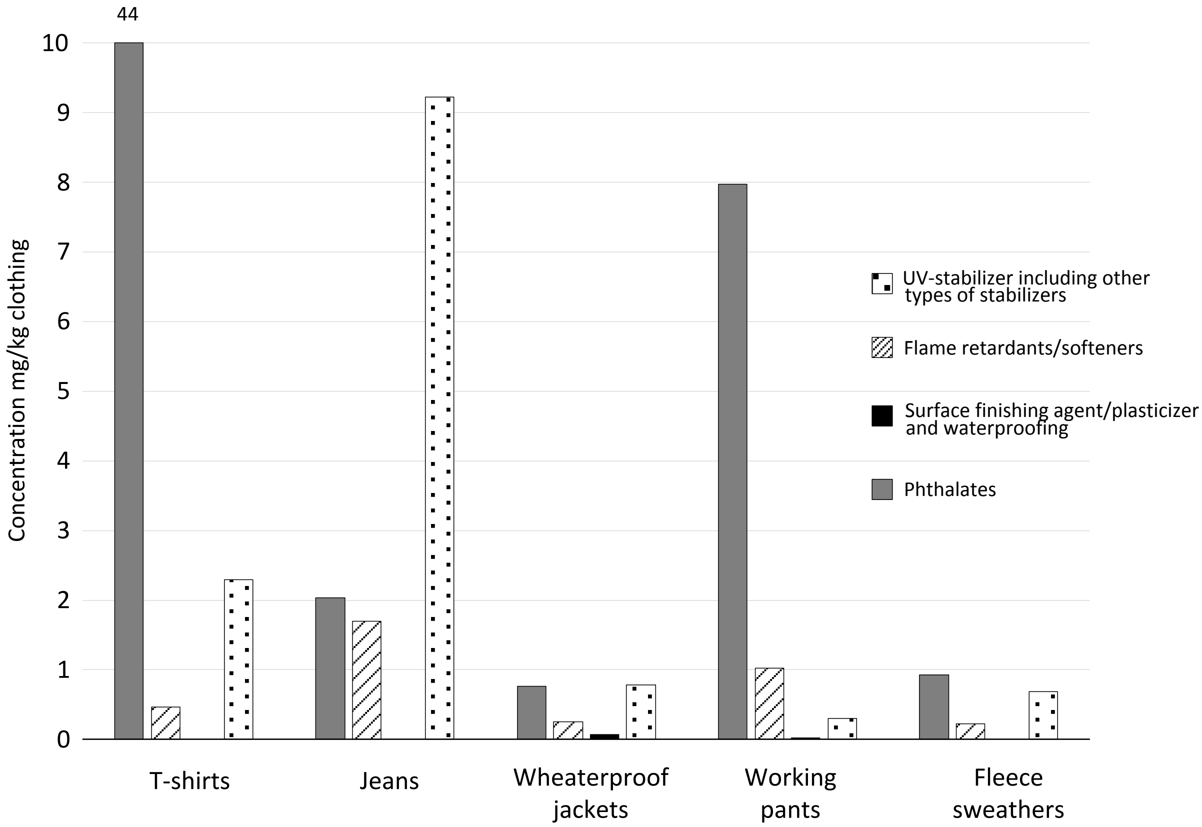

Figure 5. Distribution of functional chemicals between compound groups.

The perfluorinated compounds (included in the category "surface finishing agent/plasticizer and waterproofing") were only analyzed in the laundry wastewater from weatherproof jackets and working pants and the results show that the weatherproof jackets released higher amounts of these chemicals than the working pants did. Siloxanes were included in the same category, and they were not detected in the wastewater from any of the clothing types.

6.2 Release of Process Chemicals

Chemical substances used in the production of textiles are categorized as process chemicals; 27 out of the 126 compounds in this study were classified as such. Of these 27 compounds, 8 were detected in the laundry wastewater.

The laundry wastewater from t-shirts and working pants contained the lowest percentage of process chemicals, less than 1%. From the other categories of clothing, the percentage of process chemicals was 9%, 95%, 41% and 72% for jeans, weatherproof jackets, working pants and fleece sweaters respectively. The high percentages are due to the high amount of BPS released during washing.

The process chemicals that could be detected in the samples were phenolic compounds and arylamines. For the phenolic compounds, BPS was the main contributor (99%), the others being pentachlorophenol and bisphenol AF. BHT was detected in all samples. However, BHT was also detected at relatively high concentrations in the laundry wastewater blank, so it was excluded from the results.

The distribution of the compound groups that were detected in the laundry wastewater from the five different clothing types is shown in **Figure 6**. The individual compounds that were detected in the laundry wastewater are shown in **Table 8**.

Table 8. Concentrations of process chemicals (μg/kg clothing) detected in laundry wastewater from five types of clothing. Chemicals that were below limit-of-quantification are listed in Appendix 1.

Compound	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
Phenolic compounds					
PCP	2.9	27	220	n.a.	1000
BPS	<31	150	21000	1800	2900
Bisphenol AF	2.5	2.4	1.5	0.99	1.7
Arylamines					
IPPD	9.8	<4.8	<3.7	<2.5	<4.2
DADPM	33	420	140	17	92

n.a. not analyzed due to analytical problems

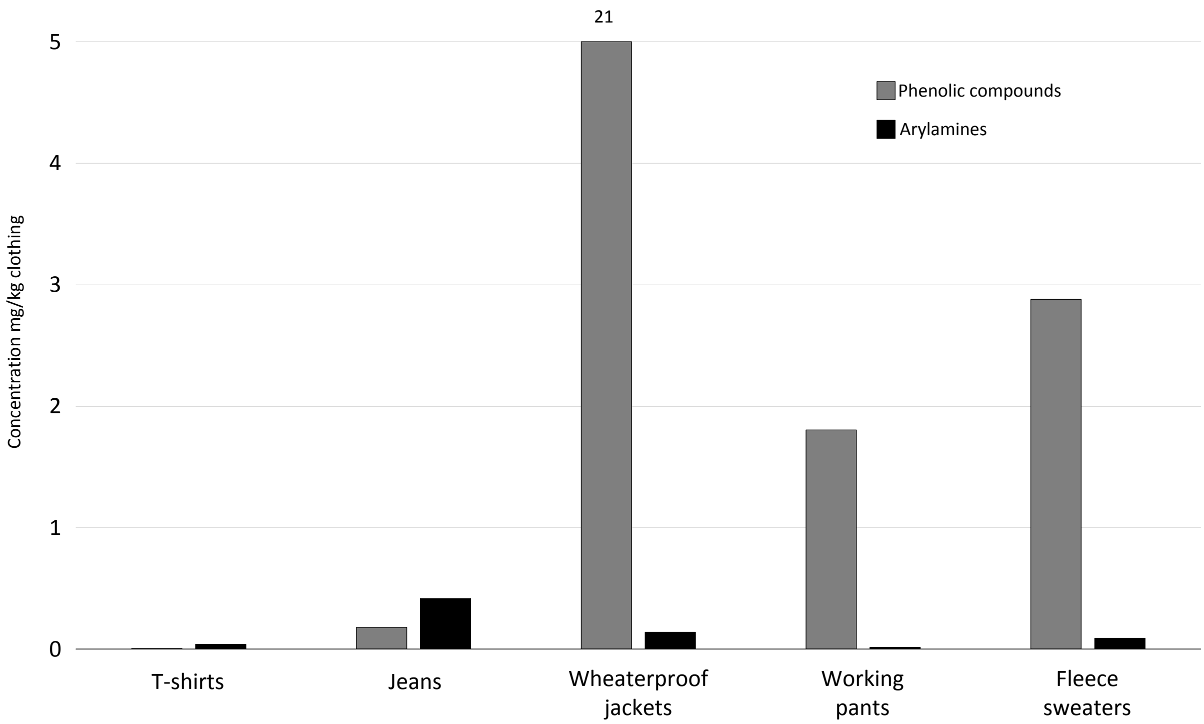

Figure 6. Distribution of compound groups for process chemicals.

6.3 Release of Unwanted Chemicals

Fifty compounds out of the 126 in this study were classified as unwanted chemicals, which means that these chemicals were not intentionally used in any step of the production of the clothing. Of these 50 compounds, 38 were detected in the laundry wastewater. This study found that the percentage of unwanted chemicals released to the laundry wastewater was below 1% of the total released weight of chemicals. Polychlorinated phenols were the largest contributor to the release of unwanted chemicals (**Figure 7**), followed by polybrominated phenols and polychlorinated benzenes. Dioxins and furans could be detected, but at very low concentrations (**Table 9**).

Table 9. Concentrations of unwanted chemicals (µg/kg clothing) detected in laundry wastewater from five types of clothing. Chemicals that were below limit-of-quantification are listed in Appendix 1.

Compound	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
Polychlorinated phenols					
2-MoCPh	0.08	0.07	0.06	n.a.	0.10
3- MoCPh	0.14	0.10	0.05	n.a.	0.15
4- MoCPh	0.52	0.54	0.27	n.a.	0.14
2,3-DiCPh	0.17	0.25	0.16	n.a.	0.87
2,4- and 2,5-DiCPh	0.28	0.14	0.20	n.a.	0.63
2,6-DiCPh	0.10	0.05	0.03	n.a.	0.008
3,4- DiCPh	0.13	0.18	0.12	n.a.	0.57
3,5- DiCPh	0.15	0.06	0.05	n.a.	0.05
2,3,6-TriCPh	0.17	<0.15	<0.11	n.a.	<0.3
2,4,5- TriCPh	0.18	<0.15	<0.11	n.a.	<0.3
2,4,6- TriCPh	0.44	0.70	<0.11	n.a.	<0.3
3,4,5- TriCPh	0.37	0.32	0.08	n.a.	0.46
2,3,4,5-TeCPh	0.65	2.0	0.57	n.a.	<0.3
2,3,4,6-TeCPh	0.62	31	<0.11	n.a.	<0.3
2,3,5,6-TeCPh	0.41	<0.11	<0.11	n.a.	<0.3
Polybrominated phenols					
2,4-DiBPh	0.07	0.01	0.01	n.a.	0.03
2,4,5-TriBPh	1.3	0.96	0.06	n.a.	0.21

n.a. not analyzed due to analytical problems

Table 9 continued. Concentrations of unwanted chemicals (μg/kg clothing) detected in laundry wastewater from five types of clothing. Chemicals that were below limit-of-quantification are listed in Appendix 1.

Compound	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
Polychlorinated benzenes					
1,2-DiCBz	0.20	0.062	0.05	0.05	0.05
1,3-DiCBz	0.35	0.18	0.25	0.03	0.03
1,4-DiCBz	0.06	0.08	0.06	0.02	0.06
1,2,3-TriCBz	0.04	0.02	0.03	0.005	0.02
1,2,4-TriCBz	0.13	0.06	0.05	0.02	0.08
1,3,5-TriCBz	0.024	0.009	0.02	0.01	0.004
1,2,3,4-TeCBz	0.013	0.009	0.007	0.005	0.003
1,2,3,5-TeCBz	0.02	0.009	0.01	0.01	0.005
1,2,4,5-TeCBz	0.02	0.01	0.008	0.006	0.008
PeCBz	0.02	0.01	0.005	0.01	0.006
HCB	0.06	0.04	0.01	0.06	0.02
Benzene, pentachlornitro-,	0.04	n.d	n.d.	n.d.	n.d.
Benzene, 1,3-dichloro-2-(dichloromethyl)-	0.05	0.01	0.06	0.02	0.008
Dioxins and furans					
1,2,3,4,6,7,8-HpCDD	0.0004	<0.0002	0.0001	0.0003	<0.0001
OCDD	0.001	0.0004	0.0004	0.0004	0.0003
2,3,4,7,8-PeCDF	<0.004	<0.0002	<0.0002	0.0002	<0.0001
2,3,4,6,7,8-HxCDF	<0.0003	<0.0001	<0.0001	0.0002	<0.0001
1,2,3,4,6,7,8-HpCDF	0.0009	<0.0001	<0.0001	<0.00008	<0.0001
OCDF	0.002	<0.00008	0.004	0.00006	0.0003

n.d. not detected.

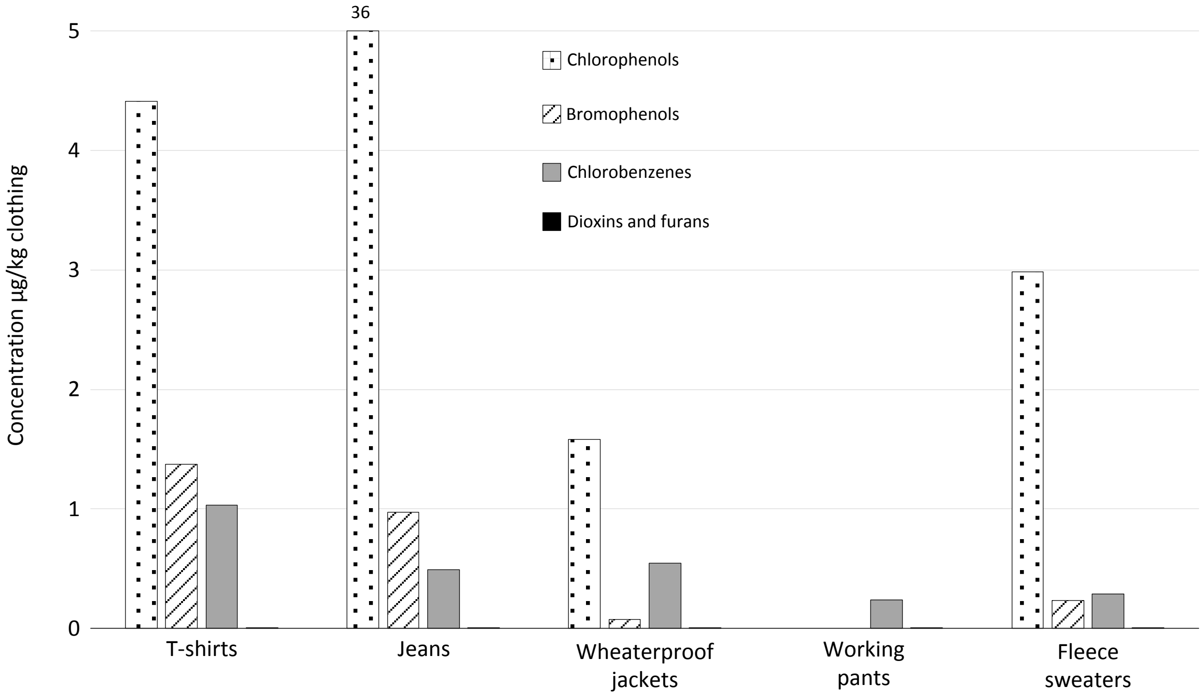

Figure 7. Distribution of compound groups for unwanted chemicals.

6.4 Release of textile fibers from clothing

T-shirts released more textile fibers than the other clothing types, indeed, double the weight of that released by jeans (**Table 10**). Weatherproof jackets, working pants and fleece sweaters released far less textile fibers than t-shirts and jeans.

Table 10. Release of textile fibers from the five different clothing types.

Concentration	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
mg/L wastewater	0.014	0.0096	0.0006	0.0017	0.0039
mg/kg clothing	0.85	0.46	0.02	0.07	0.10

7. Discussion

7.1 Occurrence

The net supply (in ton) of the different clothing types included in this study was provided by SCB (Statistics Sweden) and is shown in **Table 11**.

Table 11. Net supply in ton of the clothing types included in this study, provided by SCB.

	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
Net supply (ton)	10,000	6,000	3,200	3,600	1,000

Using these material flow data and the release data from the laundry tests (Tables 7-9) it is possible to calculate the relative contributions from the various groups of chemicals to the total amount of chemicals (670 kg) released from the five categories of clothing and two wash cycles (**Figure 8**).

Phthalates and the phthalate substitute DINCH dominates the substance flow of chemicals that follows laundry water to WWTPs, together accounting for almost 75% of the total amount. Formaldehyde and bisphenols contributes and an additional 23%, and the remaining 5% is split between organophosphate esters (OPs) and the remaining compound groups.

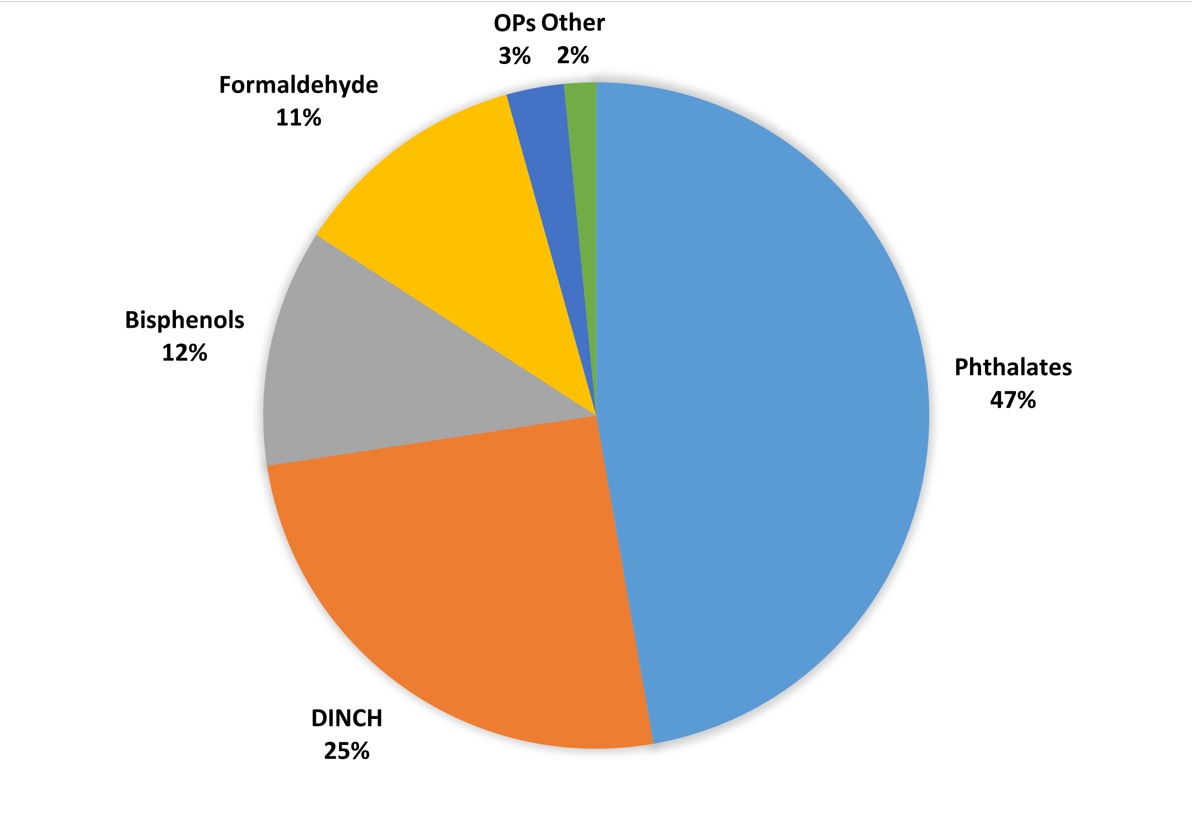

Figure 8. Contributions of various compounds groups to the total amount of chemicals released annually in Sweden upon laundry of the five types of clothes under study.

7.2 Transport and fate of urban water contaminants

The discharge of contaminants into WWTPs is a suitable release pathway into the aquatic environment; the presence of water contaminants is a global environmental problem. Households are the main source for many of the chemicals that are found in effluent and sludge from WWTPs. There are many factors that influence the fate of the chemicals travelling through urban sewer systems from households, via WWTPs, to the environment.

Of particular importance for this project are compounds that are released from clothing during washing and that are not degraded on the way to, or during the treatment in, the WWTP. These end up in either the effluent (outgoing water) or the sludge.

Non-polar compounds tend to bind to particulates, ending up in the sludge due to their high hydrophobicity (Ratola et al., 2012). However, it been shown that some non-polar contaminants can remain in the treated effluent (Rossi et al., 2004). Based on the partitioning coefficient (K_{ow}) of octanol-water, the sorption to particles has been estimated (Rogers, 1996) as:

- Log $K_{ow} < 2.5$ — low sorption potential
- Log $K_{ow} > 2.5$ and < 4.0 — medium sorption potential
- Log $K_{ow} > 4.0$ — high sorption potential

It is also important to account for a compound's susceptibility to chemical degradation (abiotic processes e.g. hydrolysis), biodegradation and volatilization.

Considering these distributions and loss processes, the compounds included in this study could be distributed in water and sludge as shown in **Table 12**. The majority of the compounds with a known log K_{ow} have a value above 4, which indicates that they should be found in the sludge.

Table 12. Estimated distribution of chemicals between water and sewage sludge based on particle sorption potential (log K_{ow}).*

Water (low sorption)	Uncertain (medium sorption)	Sludge (high sorption)
Chlorophenols (mono)	Chlorophenols (di and tri)	Chlorophenols (tetra and penta)
Organophosphates (TCEP)	Organophosphates (TCPP and TBEP)	Organophosphates (TPP)
	Chlorobenzenes (di and tri)	Chlorobenzenes (tetra-hexa)
		Dioxins/Furans
		PFC
		Phthalates

* The octanol-water partitioning coefficient has not been determined for all the compounds in this study and these are therefore not included in the table.

Analyses of influent, effluent and sludge from WWTPs in Sweden show that many of the targeted compounds in this study (50 out of 126 compounds) have been detected in at least one of the three (**Table 13**). The presence of this study's targeted chemicals in such material was described in both "Miljöövervakning av utgående vatten och slam från svenska avloppsreningsverk (Umeå Universitet, 2014) and "Regional Screening 2008" (IVL, 2010), which are two screening studies of contaminants in influent, effluent and sewage sludge from WWTP in Sweden. The concentrations of PCA were taken from Filipovic et al. (2015).

Table 13 confirms that compounds can be found in effluent even though their log K_{ow} indicates that they prefer to be particle bound and should therefore associate with the sludge. It can also be seen from **Table 13** that the opposite can occur, that is, compounds with a low log K_{ow} can be present in the sewage sludge. The properties and fate of the target compound classes are discussed below.

7.2.1 Chlorophenols

The physicochemical properties for chlorophenols vary somewhat between the different degrees of chlorination. The log K_{ow} ranges between 2.16 for monochlorophenols and 4.74 for pentachlorophenol. The water solubility ranges between 45 mg/L for pentachlorophenol and 12,700 mg/L for the monochlorinated phenols. The distribution of the chlorinated phenols is dependent on the number of chlorine atoms in the molecule.

Chlorophenols were detected in both effluent and sludge (**Table 13**) and, even though the most chlorinated congeners were expected to be present in the sewage sludge, they were only detected in the effluent. In addition to this, only monochlorinated and dichlorinated congeners were detected in the sewage sludge when we would expect to find these congeners only in the effluent.

Thus, there is a shortage of data for mass balance calculations. When calculating how the amount of chlorophenols released from laundry wastewater contribute to what is found in WWTPs, the total amount released from the clothing during laundry was therefore compared to the total amounts reported for effluent and sewage sludge.

7.2.2 Chlorobenzenes

As with the chlorophenols, the physicochemical properties of chlorobenzenes vary somewhat between the different degrees of chlorination. The log K_{ow} ranges between 3.28 for dichlorobenzene and 5.86 for hexachlorobenzene. The water solubility ranges between 0.3 mg/L for hexachlorobenzene and 104 mg/L for dichlorobenzenes. Again, the distribution of the chlorinated benzenes is dependent on the number of chlorine atoms in the molecule.

Only data on chlorobenzenes have been found for sewage sludge (**Table 13**), with all degrees of chlorination from di up to hexa being detected.

When calculating the amount of chlorobenzenes released from laundry wastewater into WWTPs, the total amount released from the clothing during washing was used, and compared to the amounts reported for sewage sludge. Because the dichloro- through hexachlorobenzenes has medium to high sorption potential, this is a reasonable assumption.

Table 13. Compounds detected in incoming (influent) and outgoing (effluent) water (µg/L) and in sewage sludge (µg/kg dry weight) in screening studies of Swedish WWTPs by IVL, Umeå University (UMU) and Filipovic et al. (2015).

Compound	Influent	Effluent	Sludge	Ref
DBP	0.39	0.053	17	IVL
DIBP	0.11	0.014	13	IVL
DEHP	4.2	0.57	9800	IVL
DINP	3.6	-	9800	IVL
DIDP	1.3	-	9800	IVL
SCCP	-	-	1.1	UMU
TPP	-	0.021	160	UMU
TCEP	-	0.18	17	UMU
TCPP	-	1.7	3300	UMU
TEHP	-	<	n.a.	UMU
TBEP	-	5.4	1500	UMU
BHT	-	<5000	2000	UMU
1,2-DiCBz	-	-	15	UMU
1,3-DiCBz	-	-	0.6	UMU
1,4-DiCBz	-	-	8	UMU
1,2,3-TriCBz	-	-	0.5	UMU
1,2,4-TriCBz	-	-	11	UMU
1,3,5-TriCBz	-	-	0.3	UMU
1235/1245-TeCBz	-	-	0.2	UMU
1,2,3,4-TeCBz	-	-	0.2	UMU
PeCBz	-	-	2	UMU
HCB	-	-	6	UMU
4-MoCPh	-	0.011	-	UMU
2-MoCPh	-	-	0.022	UMU
2,6-DiCPh	-	0.013	-	UMU
2,3-DiCPh	-	-	0.022	UMU
24/25-DiCPh	-	0.016	0.042	UMU
2,4,6-TriCPh	-	0.024	-	UMU
2,3,4,6-TeCPh	-	0.011	-	UMU
PCP	-	0.036	-	UMU
1,2,3,6,7,8-HxCDD	-	-	0.004	UMU
HpCDD	-	-	0.14	UMU
OCDD	-	-	0.78	UMU
2,3,7,8-TeCDF	-	-	0.002	UMU
1,2,3,4,6,7,8-HpCDF	-	-	0.024	UMU
OCDF	-	-	0.064	UMU
PFDS	0.00029	<	0.27	Filipovic
PFHxA	0.0038	0.0079	1.1	Filipovic
PFHpA	0.0018	0.0018	0.34	Filipovic
PFOA	0.0032	0.0047	3.2	Filipovic
PFNA	0.00075	0.00066	0.33	Filipovic
PFDA	0.00067	0.00046	0.89	Filipovic
PFUnA	0.00039	0.00013	0.33	Filipovic

\- No data available
< Below LOD, but no data given in the reference
n.a. not analyzed

7.2.3 Phthalates

The physicochemical properties for phthalates vary widely, depending on the compound. The log K_{ow} for the compounds included in this study ranges between 4.61 and 10.36 with water solubility ranging between $1.0x10^{-5}$ and 6.4 mg/L. Based on the physicochemical properties, the majority of the phthalates should end up in the sewage sludge, which was also concluded by Clara et al. (2012). It has also been reported that sludge from WWTP treating only household wastewater contains less phthalates than sludge from WWTP treating both household and industrial wastewater, with DEHP and DnBP being the most common phthalates in sewage sludge (Meng et al., 2014). The major sources of DEHP are floor and wall coverings as well as textiles (Rule et al., 2006; ICON 2001).

Based on the volume of wastewater treated in Swedish WWTPs, the amount of sewage sludge produced yearly and literature data (**Table 13**), DBP and DIBP are mostly present in effluent while DEHP, DINP and DIDP are present in sewage sludge.

Therefore, when calculating the amount of phthalates released into effluent and sewage sludge, the total amount released from the clothing during laundry was used, but the amount of DBP and DIBP was only compared to the amount in the effluent and the amount of DEHP, DINP and DIDP was only compared to the amount in the sewage sludge.

7.2.4 Organophosphates

The physicochemical properties of organophosphates included in the study vary but not to the same extent as the phthalates and the chlorophenols. The lowest log K_{ow} is 1.63 for TCEP and the highest 4.70 for TPP. Their water solubility ranges between 4.7 mg/L for TPP and 5,597 mg/L for TCEP. IVL (2008, **Table 13**) found more TCEP in the effluent than in sludge, which is reasonable in respect of log K_{ow} and the water solubility. However, more TBEP was found in the effluent than in sludge which, according to log K_{ow} and the water solubility, would be more prone to associate with the sludge. It is known that aliphatic organophosphates (e.g. TBP) are more easily degradable than aromatic organophosphates (e.g. TPP), and that the chlorinated organophosphates (e.g. TCPP) are the most persistent (Saeger et al., 1979; WHO, 1991a; 1991b; 1998). TCEP and TPP are both persistent and water soluble, meaning that they can pass through WWTPs, something demonstrated by Marklund et al. (2005).

For calculating the contribution of organophosphates, TCEP, TCPP and TBEP were assumed to only end up in the effluent and TPP in the sewage sludge.

7.2.5 Perfluorinated compounds

The physicochemical properties of perfluorinated compounds (PFC) included in the study vary to the same extent as those of phthalates and the chlorophenols. Log K_{ow} ranges between 2.20 for PFHxS and 10.16 for PFDoDA (not detected in any of the samples in this study). Their water solubility ranges between $6.1x10^{-7}$ mg/L for PFDoDA and 0.85 mg/L for PFHxA. Flipovic (2015, **Table 13**) found PFC both in effluent and sludge, most likely due to the compounds being bonded to particles in the effluent because of the very low water solubility of the PFC.

PFOA and PFOS has been identified in outdoor clothing in the range up to 30 ng/g (Brigden et al., 2013; Greenpeace 2013; Knepper et al., 2014): in this study, we found 7 of the 15 targeted perflourochemicals (**Table 7**) in the laundry wastewater from work trousers or weatherproof jackets.

The widespread use of products containing PFCs in households leads to it ending up in the environment (Herzke et al., 2012). This is because PFCs are not eliminated by WWTPs as would be preferred (Guo et al., 2010). It has been observed that even negative removal or in-plant production can occur (Heidler et al., 2008; Pasquini et al., 2014). The fate of PFCs in a typical WWTP was predicted by Arvaniti (2014): short chain PFCs, with less than 10 carbons, were expected to end up in the treated wastewater and be released into the environment. Ratolo et al. (2012) concluded that improving the understanding of how PFCs pass through WWTPs was vital, particularly as their physicochemical properties are not as well-known as for other contaminants.

Even though PFCs should end up in the sewage sludge according to their physicochemical properties, they have been detected in the effluent of WWTPs. Therefore, when calculating the contribution of PFC to effluent and sewage sludge, the whole amount released from clothing during laundry has been used as a worst-case scenario for both effluent and sewage sludge. This is because possible in-plant production of PFC is very hard to estimate.

7.2.6 Dioxins and furans

The physicochemical properties of the 17 toxic dioxins and furans included in the study vary depending on the number of chlorines in the molecules. Log K_{ow} ranges between 6.29 for 2,3,7,8-TeCDF up to 9.50 for OCDD. Their water solubility ranges between $9.3x10^{-6}$ mg/L for OCDD up to $3.1x10^{-2}$ mg/L for 2,3,7,8-TeCDF. In the screening studies presented in **Table 13,** no PCDD and PCDF were detected in influent or effluent, and only at low concentrations in sludge.

Dioxins and furans have been found in new cotton clothing (Horstmann and McLachlan, 1995a) with the congeners found in textiles being mainly heptachlorinated and octachlorinated. The origin of the dioxins was attributed to the use of pentachlorophenol as an herbicide during the production of cotton (Horstmann and McLachlan, 1995a). PCDD and PCDF have also been shown to originate from dyes derived from chloranil. PCDD and PCDF have previously been shown to be present in rather high concentrations in household wastewater (Horstmann and McLachlan, 1995b), between 1 and 15 pg TEQ/L, but a great deal has changed in the last 20 years with respect to legislation and restriction of use of these chemicals. It has also been shown that levels of OCDD have decreased in Swedish sewage sludge since 2004 (Olofsson et al., 2012).

For the calculation of the contribution of PCDD and PCDF to effluent and sewage sludge, no data were found for effluent, therefore, the whole amount released from clothing during washing was used for the calculations of percent contribution to sewage sludge.

7.3 Estimated contribution of the targeted compounds to the amounts found in effluents and sewage sludge in WWTPs

Household contributions of wastewater from the washing of fabrics is estimated to be 2% of the volume flow into the municipal sewage treatment plant.

The total volume of water that is treated in Swedish WWTPs annually ($V_{treated\ wastewater}$) is estimated to be 1.2 billion m^3 with the amount of sludge produced annually being estimated to be 240,000 tons of dry matter. The following formulae were used to calculate the amount of a compound ($M_{compound}$) that moved from laundry wastewater to effluent (R):

$$M_{compound} = C_{laundry\ wastewater} \left(\frac{g}{kg}\right) x\ M_{cloth}\ (kg)$$

$$M_{effluent} = C_{\ effluent} \left(\frac{g}{m3}\right) x\ V_{treated\ wastewater} \left(\frac{m3}{year}\right)$$

$$R = M_{compound}/M_{effluent}$$

where M_{cloth} is the net supply of clothing in Sweden (Table 11), $C_{laundry\ wastewater}$ and $C_{effluent}$ is the concentration of the compound in wastewater and effluent, respectively,

The compounds included in the calculations for the contribution to effluent and sewage sludge are described in 7.1.1 to 7.1.6. A similar calculation was also carried out for sewage sludge.

Table 14. Estimated weights (g) of the functional chemicals released from five types of clothing annually, based on the net supply shown in **Table 11**.

	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
Phthalates					
DBP	1600	4200	380	310	94
DIBP	1900	1900	320	220	170
BBP		3400	430		
DEHP	16000	2700	1300	4000	660
DINP	215000			3900	
DIDP	47000				
DPHP*				11000	
DINCH*	160000			9600	
Flame retardants/					
SCCP	61	16	3.2	650	3.3
TPP	1800	26	37		0.63
TCEP	380	70	160		12
TCPP	1050	190	310		26
TBEP	120	2300	67		100
Surface finishing agent /					
PFDS			9.0	37	
PFHxA			160		
PFHpA			35	12	
PFOA			12	15	
PFNA				8.1	
PFDA				3.9	
PFUnA			4.5		
Stabilizers					
Lowinox 44B25	200	55	74	25	12
Chimasorb 81	580		120	120	37
UV P	3300		26	40	
UV 328	330	73	260	22	13
Formaldehyde	18000	55000	2000	890	630
Total weight functional chemicals	**467000**	**69900**	**5710**	**30900**	**1760**

*Emerging phthalate

Table 15. Estimated weights (g) of the process chemicals released from five types of clothing annually, based on the net supply shown in **Table 11**.

	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
Phenolic compounds					
PCP	28	160	0.71		1.0
Arylamines					
IPPA	98				
DADPM	330	2500	440	60	92
Total weight of process chemicals	**456**	**2660**	**440**	**60**	**93**

Table 16. Estimated weights (g) of the unwanted chemicals released from five types of clothing annually, based on the net supply shown in **Table 11**.

	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
Polychlorinated phenols					
2-MoCPh	0.84	0.40	0.21		0.10
3-MoCPh	1.4	0.61	0.16		0.15
4-MoCPh	5.2	3.2	0.85		0.14
2,3-DiCPh	1.7	1.5	0.52		0.87
2,4- + 2,5-DiCPh	2.8	0.87	0.63		0.63
2,6-DiCPh	1.0		0.08		
3,4-DiCPh	1.3	1.1	0.37		0.57
3,5-DiCPh	1.5	0.39	0.17		0.05
2,3,6-TriCPh	1.7				
2,4,5-TriCPh	1.8				
2,4,6-TriCPh	4.4	4.2			
3,4,5-TriCPh	3.7	1.9	0.25		0.46
2,3,4,5-TeCPh	6.5	12	1.8		
2,3,4,6-TeCPh	6.2	190			
2,3,5,6-TeCPh	4.1				
Polybrominated phenols					
2,4-DiBPh	0.67	0.06	0.04		0.03
2,4,5-TriBPh	13	5.8	0.19		0.21
Polychlorinated benzenes					
1,2-DiCBz	2.0	0.37	0.15	0.18	0.05
1,3-DiCBz	3.5	1.1	0.79	0.12	0.03
1,4-DiCBz	0.64	0.47	0.19	0.08	0.06
1,2,3-TriCBz		0.12	0.08	0.02	0.02
1,2,4-TriCBz	1.3	0.34	0.16	0.06	0.08
1,3,5-TriCBz	0.24	0.06	0.06	0.04	0.004
1,2,3,4-TeCBz	0.13	0.05	0.02	0.02	0.003
1,2,3,5-TeCBz	0.20	0.06	0.04	0.04	0.005
1,2,4,5-TeCBz	0.15	0.06	0.03	0.02	0.008
PeCBz	0.18	0.07	0.02	0.05	0.006
HCB	0.59	0.24	0.04	0.20	0.02
Pentachloronitrobenzene	0.41				
1,3-dichloro-2-(dichloromethyl)-benzene	0.54	0.06	0.18	0.06	0.008
Dioxins and furans					
1,2,3,4,6,7,8-HpCDD	0.004		0.0003	0.001	
OCDD	0.01	0.003	0.001	0.002	0.0004
2,3,4,7,8-PeCDF				0.0007	
2,3,4,6,7,8-HxCDF				0.008	
1,2,3,4,6,7,8-HpCDF	0.009				
OCDF	0.02		0.01		0.0003
Total weight unwanted chemicals	**68**	**220**	**7.1**	**0.9**	**3.5**

Comparing the total weights of chemicals released from the five categories of clothing studied show that the functional chemicals are released in the largest amounts (**Table 14**) followed by process chemicals (**Table 15**) and unwanted chemicals (**Table 16**). In each category of chemicals, “T-shirts” was the clothing category that was estimated to release the highest weight (469 kg functional chemicals, 0.5 kg process chemicals and 0.07 kg unwanted chemicals) of chemicals annually from the first two washes. The smallest release was from “Fleece sweaters” for all three categories of chemicals with 1.8 kg functional chemicals, 3 kg process chemicals and 0.003 kg of unwanted chemicals.

Table 17 shows the contribution of 7 compound groups to that found in effluent and sewage sludge from Swedish WWTPs. Phthalates and organophosphates were estimated to be released in very high amounts (302 kg and 7.6 kg) and contributed 50% and 29% respectively. Chlorophenols and PFCs were estimated to be released in very low amounts (430 g and 300 g respectively) and contributed more than was found in the effluents and sewage sludge, 167% and 223% respectively.

Table 17. Comparison between estimated contributions of different compound groups in laundry wastewater and amounts detected in effluent and sewage sludge (combined) from Swedish STPs.

	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters	Total
Chlorophenols	75%	47%	20%		25%	167%
Chlorobenzenes	4%	1%	0.9%	0.3%	0.1%	6.3%
Phthalates	25%	18%	2.6%	2.1%	1.2%	50%
Organophosphates	21%	2.6%	4.3%		0.3%	29%
Perfluorinated compounds			134%	89%		223%
Dioxins and furans	0.3%	0.001%	0.08%	0.006%	0.002%	0.39%
SCCP	0.02%	0.006%	0.001%	0.3%	0.001%	0.23%

For the other compound groups not estimated to contribute significantly to the amounts released to the environment, it has to be taken into account that some compounds that were detected in the laundry wastewater are not present in effluent or sewage sludge data used to estimate the contribution of chemicals from the clothing.

The contribution of the compounds included in this study (for which comparison data are available, **Table 13**) to effluent and sewage sludge can be seen in **Figures 9** and **10**. According to this, t-shirt and jeans contribute the most to the amounts reaching WWTPs, followed by weatherproof jackets, working pants, and fleece sweaters. The contribution of the different compound groups differed depending on the clothing type.

The estimated contribution from laundry to amounts found in effluent from WWTPs was low overall for working pants and fleece sweaters (**Figure 9**). T-shirts, jeans and weatherproof jackets contributed substantially more. For t-shirts, the main compounds released were phthalates and organophosphates, for jeans phthalates, and for weatherproof jackets perfluorinated compounds, which were estimated to contribute most to the overall discharge of the individual compounds into the environment.

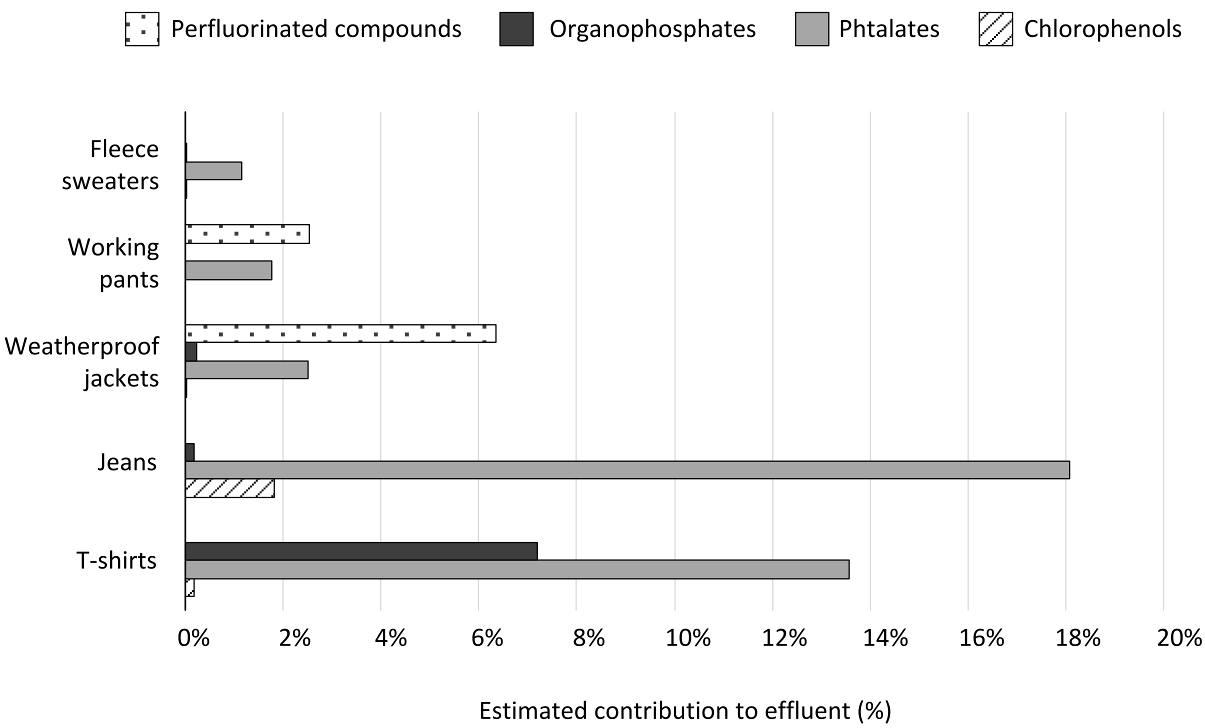

Figure 9. Estimated contribution of compound groups to effluent from WWTPs in Sweden.

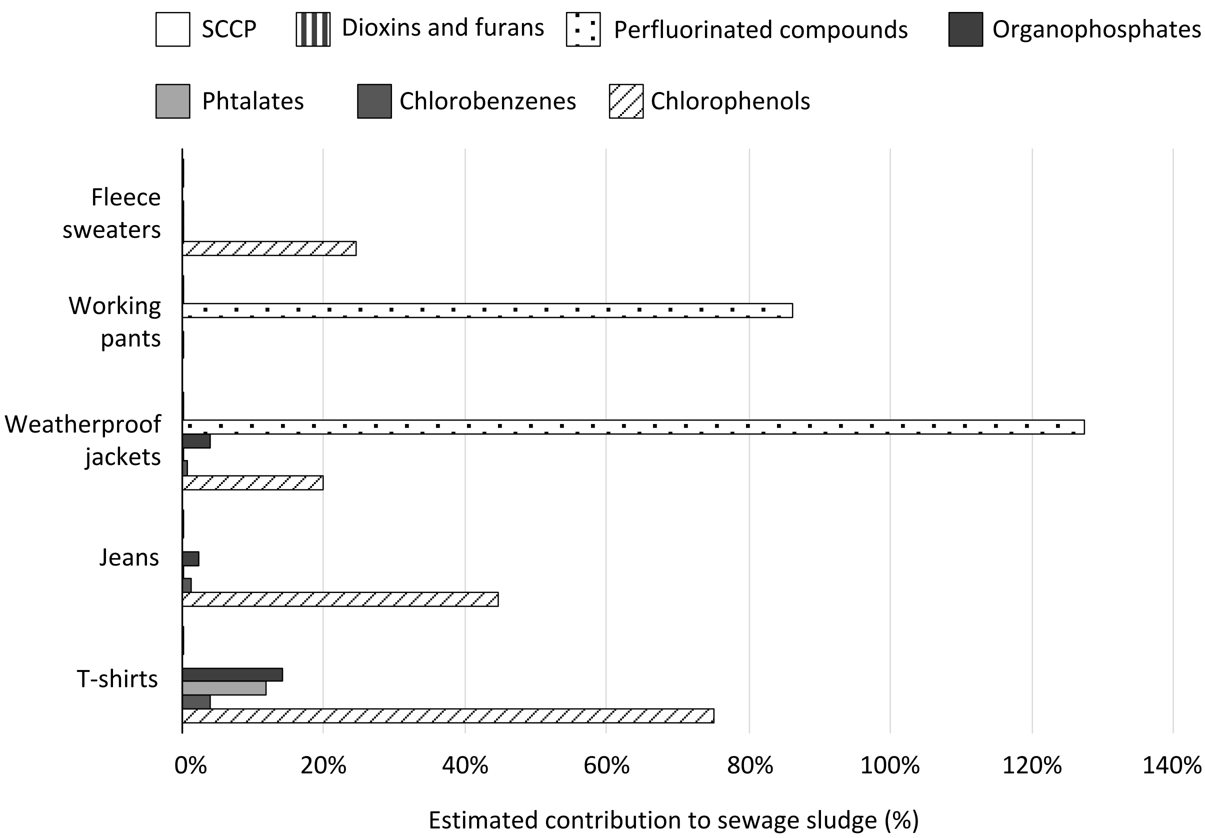

Figure 10. Estimated contribution of compound groups to sewage sludge from WWTPs in Sweden.

The estimated contribution to sewage sludge for the different compound classes was far higher than the calculated contribution to effluent (see **Figures 9** and **10** and **Tables 14 – 16**). The estimation produced a contribution figure of over 100% for some compound groups, which is not realistic. PFC and chlorophenols were the compound groups that were estimated to contribute to the amount found in effluent and sewage sludge to such a large degree that it exceeded what is actually found in the effluent and sewage sludge (**Table 17**). Chlorophenols are distributed between both effluent and sewage sludge so this could be the reason for the overestimation of the amount that ends up in the sewage sludge. It is also possible that chlorophenols and/or PFCs are degrading in the WWTP process, which could explain their (apparently) high contributions.

The estimated contribution of each compound group to effluent and sewage sludge from WWTPs is described below.

7.3.1 Chlorophenols

The estimated contribution of chlorophenols from clothing to effluent is negligible. The estimated amount that is released from clothing is largely what is found in sewage sludge. It should also be stated that only 6 and 4 congeners out of the 17 chlorophenols were detected in the effluent and sewage sludge respectively. In the laundry wastewater, we detected 17 congeners so the fate of the other 11 congeners whilst on their way to the WWTP or during the wastewater treatment is unknown. The total contribution of chlorophenols from these four clothing types exceeds the amount of chlorophenols found in sewage sludge (160%), which indicates that most chlorophenols that leave the household via the wastewater are degraded, either on the way to the WWTP or during the treatment of the wastewater at the WWTP.

The estimated contribution of chlorophenols in the wastewater from washing t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters to effluent and sewage sludge is shown in **Figures 9** and **10** as well as **Table 16**. We have only included the congeners that have been detected in effluent and the sewage sludge even though more congeners were actually detected in the laundry wastewater. The estimated contribution of chlorophenols to effluent was 0.2%, 2%, 0.01% and 0.01% for t-shirts, jeans, weatherproof jackets and fleece sweaters respectively. For sewage sludge, the contributions were 75%, 45%, 20% and 25%, respectively. Chlorophenols could not be analyzed in wastewater from washing working pants due to analytical problems.

7.3.2 Chlorobenzenes

The estimated contribution of chlorobenzenes in the laundry wastewater from t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters to sewage sludge is shown in **Figure 10** and **Table 16**. The contribution of chlorobenzenes to sewage sludge was estimated as 4%, 1.3%, 0.9%, 0.3% and 0.1% for t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters respectively.

7.3.3 Phthalates

The estimated contribution to effluent was, in total, 40%, which is rather high for only two compounds (DBP and DIBP). The contribution of phthalates to sewage sludge from these five categories of clothing was lower than for effluent (13%) assuming that DEHP, DINP and DIDP associates with the sewage sludge. This indicates that phthalates are degraded during the treatment of the wastewater in the WWTP.

The estimated contribution of phthalates in the laundry wastewater of t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters to effluent and sewage sludge is shown in **Figures 9** and **10** and **Table 14**. For effluents, the contributions were estimated as 14%, 18%, 3%, 2% and 1% for t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters respectively; for sewage sludge, the estimates were 12%, 0.1%, 0.06%, 0.3% and 0.03%, respectively**.**

7.3.4 Organophosphates

This study showed that the organophosphates released from clothing during laundry contribute to the amounts in effluent and sewage sludge. T-shirts were the largest contributor. The total contribution to effluent and sewage sludge were 8% and 22% respectively. This indicates that the organophosphates are not affected by the treatment of the wastewater by the WWTP.

The estimated contribution of organophosphates in the laundry wastewater of t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters to effluent and sewage sludge is shown in **Figures 9** and **10** and **Table 14**. The estimated contribution of organophosphates to effluent was 7%, 0.2%, 0.2% and 0.01% for t-shirts, jeans, weatherproof jackets and fleece sweaters respectively; for sewage sludge, the estimates were 14%, 2%, 4% and 0.3%, respectively. None of the organophosphates that were found in effluent and sewage sludge were detected above the detection limit in the clothing type “working pants”.

7.3.5 Perfluorinted compounds

The calculated contribution of PFC to effluent from weatherproof jackets and working pants is substantial (9%), whereas the calculated contribution to sewage sludge (210%) indicates that PFC may be subjected to degradation during wastewater treatment.

The estimated contribution of PFCs from the laundry wastewater of weatherproof jackets and working pants to effluent and sewage sludge is shown in **Figures 9** and **10** and **Table 14**. For effluent, the figures were 6% and 3% for weatherproof jackets and working pants respectively; for sewage sludge, they were 127% and 86%. In both cases, 5 of the 7 detected compounds in effluent and sewage sludge were detected in the laundry wastewater (**Table 7** and **13**).

7.3.6 Dioxins and furans

Dioxins and furans were found in the laundry wastewater in this study in low concentrations (**Table 10**) but the estimated contribution to the amounts found in sewage sludge was negligible, so washing clothing in households cannot be considered as a relevant source of dioxins and furans in Swedish sewage sludge.

The estimated contribution of dioxins and furans in the laundry wastewater of t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters to sewage sludge is shown in **Figure 10** and **Table 16**. The estimated contribution of HpCDD/F and OCDD/F to sewage sludge are 0.3%, 0.001%, 0.08%, 0.006% and 0.002% for t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters respectively.

7.3.7 Short chain chloroparaffins

Approximately 4% of the polychlorinated alkenes (to which compound category SCCP belongs) used in Sweden ends up in sewage sludge, according to Olofsson et al. (2012). The clothing in this study contributed very small amounts, less than 1% of what is detected in sewage sludge.

The contribution of SCCP in the laundry wastewater of t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters to sewage sludge is shown in **Figure 10** and **Table 14**. The contribution of SCCP to sewage sludge is 0.02%, 0.006%, 0.001%, 0.25% and 0.001% for t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters respectively.

7.4 Estimation of the contribution of textile fibers to the effluent of WWTPs

Based on the net supply of clothing included in this study (**Table 11**), the estimated release of textile fibers varies between 100 kg for fleece sweaters up to 8,500 kg for t-shirts (**Table 18**).

Table 18. Estimated weight of textile fibers released from the five clothing types and the weight released to effluent if the removal efficiency is 70% and >99%.

	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
Amount (kg)	8,500	2,800	70	250	100
RE 70% (kg)	2,600	830	21	74	30
RE >99% (kg)	85	28	0.7	2	1

According to IVL (report B2208 and B2255), disc filters enhance the separation of fibers from the effluent. Comparing an WWTP where the effluent is passed through 15 – 20 µm disc filters to an WWTP without a corresponding disc filter, the number of particles >300 µm in the effluent were significantly reduced. The removal efficiency (RE) of particles >300 µm was 70 – 100% without the disc filter and >99% with the disc filter. The amount of textile fibers >300 µm released into laundry wastewater which can then be released to a WWTP can be reduced if the WWTP uses disc filters with a pore size of around 20 µm.

8. Conclusions

Chemicals that are banned according to legislation such as Reach should, of course, not be present in clothing. Even so, they are sometimes found during inspections of manufacturing facilities and analyses of clothing. This is a large problem since the use of a chemical can be banned in some countries but not in others. Now, the clothing that we wear comes from all over the world, and it is difficult to find information on which chemicals have been used in its production since that can take place in many different countries. Furthermore, the finding of carcinogenic arylamines (primarily 4,4'-diaminodiphenylmethane), currently banned in the EU, in laundry wastewater of all types of clothing under study also illustrates the limitations of the current legislation and the need for a harmonized global legislation.

A non-toxic environment is one of the environmental objectives that the Swedish government is working towards. This means that concentrations of non-naturally occurring compounds should be close to zero and the impact on human health and on ecosystems should be negligible.

In this study, we detected 72 out of 126 compounds that are non-naturally occurring compounds, in the laundry wastewater. Among the compound groups that could not be detected were anilines, triclosan, triclocarban, and siloxanes.

The compounds released in large amounts into the laundry wastewater in this study were the process chemical BPS, and the functional chemicals phthalates (DBP, BBP, DEHP, DINP, DIDP), DINCH, organophosphates (TPP, TCEP, TCPP, TEHP, TBEP) and formaldehyde. Considering the net supply of new clothing to Sweden, the estimated annual contribution of the release of such compounds from new clothing being washed for the first time will be substantial.

Even though some of these chemicals will be degraded during the treatment process in the WWTP, many of them will end up in effluent or sewage sludge and, to different degrees, contribute to the compounds that risk ending up in the recipient waters or where nutrients are recycled from sewage sludge.

Some of the compounds were released in very low concentrations and, with respect to the net supply of clothing to Sweden, but these compounds in laundry wastewater may still pose a threat, by themselves, to the environment if the biological potency is high. This is, for instance, often the case for endocrine disrupting chemicals, such as estrogens or dioxins and furans, among others. From a principal point of view we should always aim to have zero tolerance for unwanted chemicals in clothing.

9. Future studies

To obtain a better picture of the volume of chemicals flowing to the WWTPs and, potentially, the environment, originating from the laundering of clothing, it would be of interest to study the release of chemicals from a broader range of clothing types.

It would also be interesting to include analysis of the fabric to see what proportion of chemicals are released during laundry, and what proportion remain and are then potentially released during later washing or enter the textile waste stream.

It would, finally, be of great interest to carry out non-target analysis on both the textiles and the wastewater to form an even broader picture of which chemicals are present in the textiles and the wastewater.

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Appendix 1. Concentrations of the 126 compounds in wash water from t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters (wash water from a wash without cloths) per kilo clothing.

	Compound	Unit	T-shirts	Jeans	Weatherproof jackets	Working pants	Fleece sweaters
1	2-monochlorophenol	ng/kg	84	68	65	omitted	100
2	3-monochlorophenol	ng/kg	140	100	49	omitted	150
3	4-monochlorophenol	ng/kg	520	540	270	omitted	140
4	2,3-dichlorophenol	ng/kg	170	250	160	omitted	870
5	2,4-dichlorophenol	ng/kg	280	140	200	omitted	630
6	2,5-dichlorophenol	ng/kg	Co-eluting with 2,4	Co-eluting with 2,4	Co-eluting with 2,4	omitted	Co-eluting with 2,4
7	2,6-dichlorophenol	ng/kg	100	48	25	omitted	8.3
8	3,4-dichlorophenol	ng/kg	130	180	120	omitted	570
9	3,5-dichlorophenol	ng/kg	150	65	54	omitted	54
10	2,3,4-trichlorophenol	ng/kg	<120	<150	<110	omitted	<290
11	2,3,5-trichlorophenol	ng/kg	<120	<150	<110	omitted	<290
12	2,3,6-trichlorophenol	ng/kg	170	<150	<110	omitted	<290
13	2,4,5-trichlorophenol	ng/kg	180	<150	<110	omitted	<290
14	2,4,6-trichlorophenol	ng/kg	440	700	<110	omitted	<290
15	3,4,5-trichlorophenol	ng/kg	370	320	80	omitted	460
16	2,3,4,5-tetrachlorophenol	ng/kg	650	2000	570	omitted	<250
17	2,3,4,6-tetrachlorophenol	ng/kg	620	31000	<11	omitted	<250
18	2,3,5,6-tetrachlorophenol	ng/kg	410	<11	<11	omitted	<250
19	Pentachlorophenol	ng/kg	2900	37000	220	omitted	1000
20	2,4-dibromophenol	ng/kg	67	11	12	omitted	26
21	2,4,5-tribromophenol	ng/kg	1300	960	60	omitted	210

22	Pentabromophenol		omitted	omitted	omitted	omitted	omitted
23	Butylhydroxytoluene	ng/kg	498000	150000	52000	173000	7900
24	Bisphenol A		omitted	omitted	omitted	omitted	omitted
25	4,4'-Sulfonyldiphenol	µg/kg	<31	150	21000	1800	2900
26	Bisphenol AF	µg/kg	2.5	2.4	1.5	1.0	1.7
27	4-nonylphenol, branched		omitted	omitted	omitted	omitted	omitted
28	4-tert octylphenol , branched		omitted	omitted	omitted	omitted	omitted
29	4-Nonylphenol (branched) -mono-ethoxylate		omitted	omitted	omitted	omitted	omitted
30	4-Nonylphenol (branched) -di-ethoxylate		omitted	omitted	omitted	omitted	omitted
31	4-tert-octylphenol-mono-ethoxylate		omitted	omitted	omitted	omitted	omitted
32	4-tert-octylphenol-di-ethoxylate		omitted	omitted	omitted	omitted	omitted
33	1,2-dichlorobenzene	ng/kg	200	62	46	49	46
34	1,3- dichlorobenzene	ng/kg	350	180	250	33	31
35	1,4- dichlorobenzene	ng/kg	64	78	58	23	63
36	1,2,3-trichlorobenzene	ng/kg	44	20	26	4.6	17
37	1,2,4-trichlorobenzene	ng/kg	130	57	51	16	80.4
38	1,3,5-trichlorobenzene	ng/kg	24	9.2	18	11	4.2
39	1,2,3,4-tetrachlorobenzene	ng/kg	13	8.8	7.1	4.5	2.5
40	1,2,3,5-tetrachlorobenzene	ng/kg	20	9.2	12	9.6	5.4
41	1,2,4,5-tetrachlorobenzene	ng/kg	15	10	7.9	5.6	7.5
42	Pentachlorobenzene	ng/kg	18	12	4.7	13	6.3
	Hexachlorobenzene	ng/kg	59	40	12	55	15
43	Benzene, pentachlornitro-,	ng/kg	401	n.d.	n.d.	n.d.	n.d.

44	Benzene, 1,3-dichloro-2-(dichloromethyl)-	ng/kg	54	9.6	58	16	7.6
45	2,5-Cyclohexadiene-1,4-dione, 2,3,5,6-tetrachloro-	µg/kg	<120	<97	<75	<50	<83
46	2,4,7,9-Tetramethyl-5-decyn-4,7-diol		omitted	omitted	omitted	omitted	omitted
47	Triclosan	ng/kg	<12	<10	<5	<8	<8
48	Triclocarban	ng/kg	<12	<10	<5	<8	<8
49	Endosulfan		omitted	omitted	omitted	omitted	omitted
50	MIREX	ng/kg	<30	<25	<19	<13	<21
51	Dimethyldioctadecylammonium chloride		omitted	omitted	omitted	omitted	omitted
52	Benzalkonium chloride		omitted	omitted	omitted	omitted	omitted
53	Di-n-butylphthalate	µg/kg	160	690	120	86	95
54	Di-isobutylphthalate	µg/kg	190	320	100	60	170
55	Butylbensylphthalate	µg/kg	<120	570	130	<49	<83
56	Di(2-ethylhexyl)phthalate	µg/kg	1600	450	410	1100	660
57	Di(isononyl)phthalate	µg/kg	22000	<1500	<1100	1100	<1300
58	Di(isodecyl)phthalate	µg/kg	4700	<1500	<1100	<740	<1300
59	Short chain chloroparaffins C10-13	ng/kg	6100	2600	1000	180000	3300
60	Triphenylphosphate	ng/kg	178000	4400	12000	n.d.	630
61	Tris (2-chloroethyl) phosphate	ng/kg	38000	12000	49000	n.d.	12000
62	Tris (3-chloro-n-propyl) phosphate		omitted	omitted	omitted	omitted	omitted
63	Tris (2-klor-isopropyl) phosphate	ng/kg	105000	32000	97000	n.d.	26000
64	Tris (2-etylhexyl) phosphate	ng/kg	6200	155000	2100	n.d.	5800
65	Tris (1,3-dichloro-2-propyl) phosphate		omitted	omitted	omitted	omitted	omitted
66	Tris (2-butoxyethyl) phosphate	ng/kg	12000	383000	21000	n.d.	100000

67	Tris(2,4-di-tert-butylphenyl)phosphite		omitted	omitted	omitted	omitted	omitted
68	Tris(2,3-dibrompropyl)-fosfat	ng/kg	<62	<48	<37	<25	<42
69	Tetrakis(2,4-di-t-butylphenyl)-4,4`-biphenylylenediphosphonite, Irgafos P-EPQ		omitted	omitted	omitted	omitted	omitted
70	Hexa methylcyclo-trisiloxane	µg/kg	<620	<480	<370	<250	<420
71	Octamethylcyclo-tetrasiloxane	µg/kg	<250	<190	<150	<100	<170
72	Decamethylcyclo-pentasiloxane	µg/kg	<180	<150	<110	<75	<130
73	Dodecamethylcyclo-hexasiloxane	µg/kg	<120	<100	<75	<50	<80
74	Perfluorooctane sulfonic acid	µg/kg	n.a.	n.a.	<0.4	<0.2	n.a.
75	Perfluorobutyric acid	µg/kg	n.a.	n.a.	<0.2	<0.2	n.a.
76	Perfluorohexane soulfonic acid	µg/kg	n.a.	n.a.	<0.4	<0.2	n.a.
77	Perfluorodecane sulfonic acid	µg/kg	n.a.	n.a.	2.8	10	n.a.
78	Perfluorohexanoic acid	µg/kg	n.a.	n.a.	50	<1.5	n.a.
79	Perfluoorheptanoic acid	µg/kg	n.a.	n.a.	11	3.2	n.a.
80	Perfluoroctanoic acid	µg/kg	n.a.	n.a.	3.8	4.2	n.a.
81	Perfluorononanoic acid	µg/kg	n.a.	n.a.	<1.1	2.2	n.a.
82	Perfluorodecanoic acid	µg/kg	n.a.	n.a.	<1.1	1.1	n.a.
83	Perfluoroundecanoic acid	µg/kg	n.a.	n.a.	1.4	<0.7	n.a.
84	Perfluorododecanoic acid	µg/kg	n.a.	n.a.	<0.3	<1.0	n.a.
85	Perfluorotridecanoic acid	µg/kg	n.a.	n.a.	<0.2	<0.1	n.a.
86	Perfluorotetradecanoic acid	µg/kg	n.a.	n.a.	<0.4	<0.2	n.a.
87	Perfluoropentadecanoic acid	µg/kg	n.a.	n.a.	<0.4	<0.2	n.a.
88	Perfluorooctanesulfonamide	µg/kg	n.a.	n.a.	<1.9	<1.2	n.a.
89	4-chloro-2-methylaniline	µg/kg	<12	<10	<8	<5	<8

90	4-chloroaniline	µg/kg	<12	<10	<8	<5	<8
91	2,3- dimethylaniline	µg/kg	<12	<10	<8	<5	<8
92	2,4- dimethylaniline	µg/kg	<12	<10	<8	<5	<8
93	2,5- dimethylaniline	µg/kg	<12	<10	<8	<5	<8
94	2,6- dimethylaniline	µg/kg	<12	<10	<8	<5	<8
95	3,4- dimethylaniline	µg/kg	<12	<10	<8	<5	<8
96	3,5- dimethylaniline	µg/kg	<12	<10	<8	<5	<8
97	2-methoxyaniline	µg/kg	<6	<5	<4	<3	<4
98	4-aminodiphenyl		omitted	omitted	omitted	omitted	omitted
99	Diphenylamine	µg/kg	<6	<5	<4	<3	<4
100	N-Isopropyl-N'-phenyl-1,4-phenylenediamine	µg/kg	9.8	<5	<4	<3	<4
101	2-Naphtylamine	µg/kg	<1.2	<1.0	<0.7	<0.5	<0.8
102	Benzidine	µg/kg	<4	<3	<3	<2	<3
103	4,4'-Diaminodiphenylmethane	µg/kg	33	420	140	17	92
104	Tetrakis methylene(3,5-di-t-butyl-4-hydroxyhydrocinnamate)methane	µg/kg	<120	<100	<75	<50	<83
105	4,4'-Butylidenbis(**2**-t-butyl-5-methyl)phenol	µg/kg	20	9.2	23	6.9	12
106	2-Hydroxy-4-n-octoxybenzophenone	µg/kg	59	<34	37	35	37
107	2-(2´-Hydroxy-5´-methylphenyl)-benzotriazole	µg/kg	330	<5	8.2	11	<4
108	2-(2H-Bensotriazol-2-yl)-4,6-bis(1,1-dimethylpropyl)phenol	µg/kg	33	12	82	6.2	13
109	2,3,7,8-Tetrachlorinated dibenzo-*p*-dioxin	pg/kg	<80	<50	<40	<20	<41

110	1,2,3,7,8-Pentachlorinated dibenzo-*p*-dioxin	pg/kg	<430	<230	<270	<140	<200
111	1,2,3,4,7,8-Hexachlorinated dibenzo-*p*-dioxin	pg/kg	<340	<200	<160	<90	<180
112	1,2,3,6,7,8-Hexachlorinated dibenzo-*p*-dioxin	pg/kg	<330	<180	<160	<90	<170
113	1,2,3,7,8,9-Hexachlorinated dibenzo-*p*-dioxin	pg/kg	<320	<200	<160	<90	<170
114	1,2,3,4,6,7,8-Heptachlorinated dibenzo-*p*-dioxin	pg/kg	360	<160	100	350	<120
115	Octachlorinated dibenzo-*p*-dioxin	pg/kg	980	440	450	440	350
116	2,3,7,8-Tetrachlorinated dibenzofuran	pg/kg	<80	<50	<40	<20	<40
117	1,2,3,7,8-Penatchlorinated dibenzofuran	pg/kg	<360	<180	<140	<190	<150
118	2,3,4,7,8-Pentachlorinated dibenzofuran	pg/kg	<380	<190	<140	190	<150
119	1,2,3,4,7,8- Hexachlorinated dibenzofuran	pg/kg	<310	<150	<120	<130	<130
120	1,2,3,6,7,8- Hexachlorinated dibenzofuran	pg/kg	<310	<130	<110	<120	<70
121	2,3,4,6,7,8- Hexachlorinated dibenzofuran	pg/kg	<330	<130	<110	230	<120
122	1,2,3,7,8,9-Hexachlorinated dibenzofuran	pg/kg	<370	<150	<120	<130	<140
123	1,2,3,4,6,7,8-Heptachlorinated dibenzofuran	pg/kg	860	<140	<130	<80	<120
124	1,2,3,4,7,8,9-Heptachlorinated dibenzofuran	pg/kg	<330	<100	<110	<100	<160
125	Octachlorinated dibenzofuran	pg/kg	1500	<80	3700	57	280
126	Formaldehyde	µg/kg	1800	9200	630	250	630

n.a. - Not analyzed in this clothing; n.d. - not detected

Appendix 2. Concentrations of the 126 compounds in laundry water from t-shirts, jeans, weatherproof jackets, working pants and fleece sweaters (wash water from a wash without cloths) per liter of laundry water.

	Compound	Unit	T-shirts	Jeans	Weaterproof jackets	Working pants	Fleece sweathers	Wash water Blank
1	2-monochlorophenol	ng/L	1.4	1.4	1.7	omitted	2.4	0.34
2	3-monochlorophenol	ng/L	2.3	2.1	1.3	omitted	3.5	<0.2
3	4-monochlorophenol	ng/L	8.5	11	7.1	omitted	3.5	1.0
4	2,3-dichlorophenol	ng/L	2.7	5.2	4.3	omitted	21	1.4
5	2,4-dichlorophenol	ng/L	4.5	3.0	5.2	omitted	15	1.3
6	2,5-dichlorophenol	ng/L	Co-eluting with 2,4	Co-eluting with 2,4	Co-eluting with 2,4	omitted	Co-eluting with 2,4	Co-eluting with 2,4
7	2,6-dichlorophenol	ng/L	1.7	<1.0	0.7	omitted	<0.2	<0.2
8	3,4-dichlorophenol	ng/L	2.1	3.8	3.1	omitted	14	1.3
9	3,5-dichlorophenol	ng/L	2.4	1.3	1.5	omitted	1.3	<0.2
10	2,3,4-trichlorophenol	ng/L	<2.0	<3.0	<3.0	omitted	<7.0	<0.2
11	2,3,5-trichlorophenol	ng/L	<2.0	<3.0	<3.0	omitted	<7.0	<0.2
12	2,3,6-trichlorophenol	ng/L	2.7	<3.0	<3.0	omitted	<7.0	<0.2
13	2,4,5-trichlorophenol	ng/L	3.0	<3.0	<3.0	omitted	<7.0	<0.2
14	2,4,6-trichlorophenol	ng/L	7.2	14	<3.0	omitted	<7.0	<0.2
15	3,4,5-trichlorophenol	ng/L	6.1	6.6	2.1	omitted	11	<0.2
16	2,3,4,5-tetrachlorophenol	ng/L	11	42	15	omitted	<6.0	<6.0
17	2,3,4,6-tetrachlorophenol	ng/L	10	646	0.0	omitted	<6.0	<6.0
18	2,3,5,6-tetrachlorophenol	ng/L	6.6	0.0	0.0	omitted	<6.0	<6.0
19	Pentachlorophenol	ng/L	47	551	6.0	omitted	24	6.9
20	2,4-dibromophenol	ng/L	1.1	0.22	0.33	omitted	0.61	0.22
21	2,4,5-tribromophenol	ng/L	21	20	1.6	omitted	5.0	0.64

22	Pentabromophenol		omitted	omitted	omitted	omitted	omitted	omitted
23	Butylhydroxytoluene	ng/L	8100	3100	1400	7000	190	4100
24	Bisphenol A		omitted	omitted	omitted	omitted	omitted	omitted
25	4,4'-Sulfonyldiphenol	µg/L	<0.5	3.1	560	73	69	<0.5
26	Bisphenol AF	µg/L	0.04	0.05	0.04	0.04	0.04	0.05
27	4-nonylphenol, branched		omitted	omitted	omitted	omitted	omitted	omitted
28	4-tert octylphenol , branched		omitted	omitted	omitted	omitted	omitted	omitted
29	4-Nonylphenol (branched) -mono-ethoxylate		omitted	omitted	omitted	omitted	omitted	omitted
30	4-Nonylphenol (branched) -di-ethoxylate		omitted	omitted	omitted	omitted	omitted	omitted
31	4-tert-octylphenol-mono-ethoxylate		omitted	omitted	omitted	omitted	omitted	omitted
32	4-tert-octylphenol-di-ethoxylate		omitted	omitted	omitted	omitted	omitted	omitted
33	1,2-dichlorobenzene	ng/L	3.26	1.28	1.22	1.97	1.10	<0.21
34	1,3- dichlorobenzene	ng/L	5.61	3.62	6.57	1.34	0.74	<0.09
35	1,4- dichlorobenzene	ng/L	1.04	1.62	1.56	0.92	1.52	<0.18
36	1,2,3-trichlorobenzene	ng/L	0.72	0.41	0.69	0.19	0.40	<0.06
37	1,2,4-trichlorobenzene	ng/L	2.16	1.18	1.37	0.66	1.93	<0.14
38	1,3,5-trichlorobenzene	ng/L	0.38	0.19	0.48	0.43	0.10	<0
39	1,2,3,4-tetrachlorobenzene	ng/L	0.21	0.18	0.19	0.18	0.06	0.03
40	1,2,3,5-tetrachlorobenzene	ng/L	0.33	0.19	0.31	0.39	0.13	0.03
41	1,2,4,5-tetrachlorobenzene	ng/L	0.25	0.21	0.21	0.23	0.18	0.13
42	Pentachlorobenzene	ng/L	0.29	0.25	0.13	0.53	0.15	0.08
	Hexachlorobenzene	ng/L	0.95	0.82	0.32	2.22	0.36	0.15
43	Benzene, pentachlornitro-,	ng/L	0.66	n.d.	n.d.	n.d.	n.d.	n.d.

44	Benzene, 1,3-dichloro-2-(dichloromethyl)-	ng/L	0.88	0.20	1.55	0.65	0.18	0.88
45	2,5-Cyclohexadiene-1,4-dione, 2,3,5,6-tetrachloro-	µg/L	<2	<2	<2	<2	<2	<2
46	2,4,7,9-Tetramethyl-5-decyn-4,7-diol		omitted	omitted	omitted	omitted	omitted	omitted
47	Triclosan	ng/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
48	Triclocarban	ng/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
49	Endosulfan		omitted	omitted	omitted	omitted	omitted	omitted
50	MIREX	ng/L	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
51	Dimethyldioctadecylammonium chloride		omitted	omitted	omitted	omitted	omitted	omitted
52	Benzalkonium chloride		omitted	omitted	omitted	omitted	omitted	omitted
53	Di-n-butylphthalate	µg/L	2.6	14	3.2	3.5	2.3	6.6
54	Di-isobutylphthalate	µg/L	3.0	6.7	2.7	2.4	4.1	6.8
55	Butylbensylphthalate	µg/L	<2	12	3.6	<2	<2	2.0
56	Di(2-ethylhexyl)phthalate	µg/L	26	9.3	11	45	16	9.5
57	Di(isononyl)phthalate	µg/L	350	<30	<30	44	<30	<30
58	Di(isodecyl)phthalate	µg/L	77	<30	<30	<30	<30	<3
59	Short chain chloroparaffins C10-13	ng/L	99	54	27	7300	79	<15
60	Triphenylphosphate	ng/L	2900	90	310	n.d.	15	11
61	Tris (2-chloroethyl) phosphate	ng/L	610	240	1300	n.d.	290	n.d.
62	Tris (3-chloro-n-propyl) phosphate	ng/L	omitted	omitted	omitted	omitted	omitted	omitted
63	Tris (2-klor-isopropyl) phosphate	ng/L	1700	670	2600	n.d.	620	1100
64	Tris (2-etylhexyl) phosphate	ng/L	100	32000	56	n.d.	140	15

65	Tris (1,3-dichloro-2-propyl) phosphate		omitted	omitted	omitted	omitted	omitted	omitted
66	Tris (2-butoxyethyl) phosphate	ng/L	200	7900	560	n.d.	2400	50
67	Tris(2,4-di-tert-butylphenyl)-phosphite		omitted	omitted	omitted	omitted	omitted	omitted
68	Tris(2,3-dibrompropyl)-phosphate	ng/L	<1	<1	<1	<1	<1	<1
69	Tetrakis(2,4-di-t-butylphenyl)-4,4`-biphenylylenediphosphonite, Irgafos P-EPQ		omitted	omitted	omitted	omitted	omitted	omitted
70	Hexa methylcyclo-trisiloxane	µg/L	<10	<10	<10	<10	<10	<10
71	Octamethylcyclo-tetrasiloxane	µg/L	<4	<4	<4	<4	<4	<4
72	Decamethylcyclo-pentasiloxane	µg/L	<3	<3	<3	<3	<3	<3
73	Dodecamethylcyclo-hexasiloxane	µg/L	<2	<2	<2	<2	<2	<2
74	Perfluorooctane sulfonic acid	µg/L	n.a.	n.a.	<0.01	<0.01	n.a.	<0.01
75	Perfluorobutyric acid	µg/L	n.a.	n.a.	<0.006	<0.008	n.a.	<0.002
76	Perfluorohexane soulfonic acid	µg/L	n.a.	n.a.	<0.01	<0.01	n.a.	<0.01
77	Perfluorodecane sulfonic acid	µg/L	n.a.	n.a.	0.075	0.419	n.a.	0.144
78	Perfluorohexanoic acid	µg/L	n.a.	n.a.	1.346	<0.06	n.a.	<0.06
79	Perfluoorheptanoic acid	µg/L	n.a.	n.a.	0.289	0.13	n.a.	0.09
80	Perfluoroctanoic acid	µg/L	n.a.	n.a.	0.101	0.169	n.a.	<0.02
81	Perfluorononanoic acid	µg/L	n.a.	n.a.	<0.03	0.091	n.a.	<0.01
82	Perfluorodecanoic acid	µg/L	n.a.	n.a.	<0.03	0.044	n.a.	<0.02
83	Perfluoroundecanoic acid	µg/L	n.a.	n.a.	0.038	<0.03	n.a.	0.02
84	Perfluorododecanoic acid	µg/L	n.a.	n.a.	<0.007	<0.04	n.a.	<0.019
85	Perfluorotridecanoic acid	µg/L	n.a.	n.a.	<0.005	<0.005	n.a.	<0.005
86	Perfluorotetradecanoic acid	µg/L	n.a.	n.a.	<0.01	<0.01	n.a.	<0.01

87	Perfluoropentadecanoic acid	µg/L	n.a.	n.a.	<0.01	<0.01	n.a.	<0.01
88	Perfluorooctanesulfonamide	µg/L	n.a.	n.a.	<0.05	<0.05	n.a.	<0.05
89	4-chloro-2-methylaniline	µg/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
90	4-chloroaniline	µg/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
91	2,3- dimethylaniline	µg/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
92	2,4- dimethylaniline	µg/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
93	2,5- dimethylaniline	µg/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
94	2,6- dimethylaniline	µg/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
95	3,4- dimethylaniline	µg/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
96	3,5- dimethylaniline	µg/L	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
97	2-methoxyaniline	µg/L	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
98	4-aminodiphenyl	µg/L	omitted	omitted	omitted	omitted	omitted	omitted
99	Diphenylamine	µg/L	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
100	N-Isopropyl-N'-phenyl-1,4-phenylenediamine	µg/L	0.16	<0.1	<0.1	<0.1	<0.1	0.32
101	2-Naphtylamine	µg/L	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
102	Benzidine	µg/L	<0.07	<0.07	<0.07	<0.07	<0.07	<0.07
103	4,4'-Diaminodiphenylmethane	µg/L	0.53	8.6	3.7	0.67	2.2	0.04
104	Tetrakis methylene(3,5-di-t-butyl-4-hydroxyhydrocinnamate)methane	µg/L	<2	<2	<2	<2	<2	<2
105	4,4'-Butylidenbis(**2**-t-butyl-5-methyl)phenol	µg/L	0.33	0.19	0.62	0.28	0.29	0.29
106	2-Hydroxy-4-n-octoxybenzophenone	µg/L	0.95	<0.7	0.98	1.4	0.89	1.1
107	2-(2´-Hydroxy-5´-methylphenyl)-benzotriazole	µg/L	5.4	<0.1	0.22	0.45	<0.1	0.14

108	2-(2H-Bensotriazol-2-yl)-4,6-bis(1,1-dimethylpropyl)phenol	µg/L	0.54	0.25	2.2	0.25	0.31	0.64
109	2,3,7,8-Tetrachlorinated dibenzo-*p*-dioxin	pg/L	<1.3	<1	<1	<1	<1	<1.2
110	1,2,3,7,8-Pentachlorinated dibenzo-*p*-dioxin	pg/L	<7	<4.8	<7.2	<5.6	<4.8	<7.6
111	1,2,3,4,7,8-Hexachlorinated dibenzo-*p*-dioxin	pg/L	<5.6	<4.1	<4.4	<3.8	<4.2	<4.7
112	1,2,3,6,7,8-Hexachlorinated dibenzo-*p*-dioxin	pg/L	<5.4	<3.8	<4.2	<3.5	<4	<4.4
113	1,2,3,7,8,9-Hexachlorinated dibenzo-*p*-dioxin	pg/L	<5.2	<4.2	<4.4	<3.7	<4	<4.6
114	1,2,3,4,6,7,8-Heptachlorinated dibenzo-*p*-dioxin	pg/L	5.8	<3.4	2.7	14	<2.9	<3.7
115	Octachlorinated dibenzo-*p*-dioxin	pg/L	16	9	12	18	8.3	5.3
116	2,3,7,8-Tetrachlorinated dibenzofuran	pg/L	<1.3	<1	<1	<1	<1	<1.2
117	1,2,3,7,8-Penatchlorinated dibenzofuran	pg/L	<5.9	<3.8	<3.7	<7.7	<3.6	<4.7
118	2,3,4,7,8-Pentachlorinated dibenzofuran	pg/L	<6.1	<3.9	<3.8	7.8	<3.5	<5.2
119	1,2,3,4,7,8- Hexachlorinated dibenzofuran	pg/L	<5.1	<3	<3.2	<5.1	<3	<4.3
120	1,2,3,6,7,8- Hexachlorinated dibenzofuran	pg/L	<5	<2.7	<3	<4.8	<1.7	<3.7
121	2,3,4,6,7,8- Hexachlorinated dibenzofuran	pg/L	<5.3	<2.7	<3	9.5	<2.9	<4.3
122	1,2,3,7,8,9-Hexachlorinated dibenzofuran	pg/L	<6	<3.1	<3.3	<5.3	<3.3	<5

123	1,2,3,4,6,7,8-Heptachlorinated dibenzofuran	pg/L	14	<2.8	<3.6	<3.2	<2.8	<5.2
124	1,2,3,4,7,8,9-Heptachlorinated dibenzofuran	pg/L	<5.4	<2.1	<3	<4.2	<3.7	<6.8
125	Octachlorinated dibenzofuran	pg/L	25	<1.6	100	2.3	6.6	<1.3
126	Formaldehyde	µg/L	30	190	17	10	15	1.1