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What difference can drop-in substitution actually make? A life cycle assessment of alternative water repellent chemicals

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ABSTRACT

Per- and polyfluoroalkyl substances (PFASs) are used in durable water repellents (DWRs) on outdoor garments and manufacturers are currently phasing out hazardous PFASs. A critical question is: which alternatives should be chosen? The answer should depend on a holistic assessment, but the published inventory data and methodological guidance for assessing PFAS in products is slim and typically limited to hazard assessment. We aim to provide a holistic assessment of the potential environmental consequences of this phase out of DWRs, going beyond the more traditional hazard-focused substitution assessment to also include a broad life-cycle-based assessment of PFASs and their drop-in alternatives.

In this study, potential environmental consequences of the phase out were evaluated by applying a life cycle assessment (LCA) to shell jackets with side-chain fluorinated polymer based (i.e., PFASs) or non-fluorinated alternative DWRs with the aim to support a substitution assessment. We demonstrated an innovative approach to impact assessment by inclusion of PFAS related fate and toxicity and invested effort towards contributing new primary inventory data by using a combination of industry dialogue and performance measurements from our larger project context.

From a methodological point of view, this paper demonstrates the state-of-the-art in product LCA of persistent textile chemicals and identifies the current limits of this assessment approach. It also delivers new LCI data of use to other analysts. The LCA results in this paper suggest that jackets without PFASs are environmentally preferable. Potential problem shifting due to increased washing and reimpregnation of the jackets did not outweigh PFAS-related potential toxicity impacts as indicated by LCA results. Based on the results presented here, specific DWRs within the non-fluorinated DWR group could not be identified as preferable to others. This LCA does however provide a relevant starting point for more detailed studies on specific DWR systems and it supports moves to phase-out PFASs from non-essential DWR uses.

1. Introduction

PFAS chemicals are used in clothing but this is now considered a problem. For many years, breathable and water repellent outdoor garments have been made of textiles impregnated with durable water repellents (DWRs). DWRs are chemical mixtures commonly added to the textile during industrial finishing. DWRs are also used by consumers at home to restore water repellency after wearing and washing garments. Different actors in the outdoor garment industry are currently phasing out either all DWRs based on side-chain fluorinated polymers (i.e. PFAS) or those that give rise to emissions of so called long-chain perfluoroalkyl acids (PFAAs), due to current and expected legal restrictions on such substances (European Commission, 2017), and public pressure (Cousins et al. 2019). This phase-out is possible, because fluorinated and non-fluorinated drop-in substitutes are available on the market, but it is also complicated by performance requirements and the presence of unwanted impurities in DWR products. Long-chain PFAAs have been defined as having an alkyl chain containing six or more carbons in molecules that are perfluorosulfonic acids (PFSAs: $C_nF_{2n+1}SO_3H$, $n \geq 6$) and seven or more carbons in molecules that are perfluorinated

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carboxylic acids (PFCAs: $C_nF_{2n+1}COOH$, $n \geq 7$) (Buck et al. 2011), and a number of these PFAA are recognized as persistent, bioaccumulative and toxic (PTB) (Scheringer et al. 2014). Glüge et al. (2020) recently reviewed the global use of PFAS substances and concluded that surface protection of textiles is a priority area for reduction or elimination of PFAS use. To support the substitution, Holmquist et al. (2016) mapped available DWR systems and their associated hazards. Four general types of DWR systems were identified, where the "active ingredient", the functional polymer providing the water (and oil) repellency, was a side-chain fluorinated polymer, a silicone-based polymer, a hydrocarbon-based polymer or "other". In the hazard assessment by Holmquist et al. (2016) the DWR of concern was a so-called C8, i.e. a DWR based on side-chain fluorinated polymers containing the C_8F_{17} moiety, as those can give rise to emissions of long-chain PFAAs. The assessment showed that alternatives to C8 DWRs had better profiles for human health and environmental hazards. The hazard assessment also showed that the alternatives are also associated with problematic chemical emissions. Alternative DWRs based on side-chain fluorinated polymers (so called C4, including the C_4F_9 moiety, and C6, including the C_6F_{13} moiety) caused emissions of less (eco)toxic but equally (extremely) persistent short-chain PFAAs. Toxic and persistent siloxanes octamethylcyclotetrasiloxane (D4) and decamethylcyclopentasiloxane (D5) and silicone degradation products were associated with silicone based DWRs. Holmquist et al. (2016) highlighted the need to go beyond hazard assessment, as emissions of DWR-related chemicals and the ultimate fate of the original compounds can be very different between the alternatives. Furthermore, the alternative DWRs are not identical in performance (Schellenberger et al. 2018), and substitution could cause "problem shifting" - increasing impacts in another part of the system (e.g. extra garment production compensating for shorter functional lifespans) while the (eco)toxicity impacts directly related to the DWR are reduced, or increasing impacts in other impact categories, such as e.g. climate change.

Life cycle assessment (LCA) provides a holistic environmental perspective including the full life cycle of the product under study and all relevant impact categories (Baumann and Tillman, 2004). Potential environmental and human health impacts are included in an "environmental LCA" and are hereafter described as environmental impacts. LCA quantifies environmental impacts of goods or services along their entire life cycles in a four phase approach: setting the goal and scope; life cycle inventory (LCI) analysis; life cycle impact assessment (LCIA); and interpretation. In the LCI phase, resources used and emissions released by the studied product are quantified as flows. In the LCIA phase, potential environmental impacts of these inventory flows are characterized using substance-specific impact characterization factors (CFs). This broad environmental perspective makes LCA suitable for identifying potential problem shifting in product design and selection of alternatives. In chemical alternatives assessment (CAA), the inclusion of a life cycle perspective, e.g. by use of LCA, is recommended to ascertain inclusion of life cycle impacts and potential for problem shifting (Fantke et al. 2015; Geiser et al. 2015; Tickner et al. 2015, 2019; Jacobs et al. 2016).

For garment manufacturers looking for guidance on the use of LCA in this context there are some critical gaps in the literature. One is the unavailability of suitable LCI data, a problem compounded by confidentiality issues in the chemical industry. A review by Roos et al. (2015a) found that in 88% of published textile LCA studies, LCI data on toxic emissions was absent. Worse, suitable methods for LCIA are in their infancy and in the same study, only 7% of the studies were able to characterize the impacts of toxic emissions. For unusually persistent and surface-active compounds like PFAS, the situation is worse: correct characterization of emission data is complicated by the many substances in the group and their different fate and effect patterns (Shi et al. 2015; Schulz et al. 2020; D'Ambro et al. 2021). Consequently, reports on practical use of LCA in substitution case studies are few.

To address these gaps, this paper describes an LCA on shell jackets with alternative DWRs. In particular, our research aims were firstly to test the practical feasibility and applicability of an approach to holistic assessment of the potential environmental consequences of the phase out of DWRs which generate emissions of long-chain PFAAs, going beyond the more traditional substitution assessment process which focuses on hazards of the function providing chemical and its alternatives. A secondary aim was to assess the influence of different design and use parameters on the impacts of garments impregnated with DWRs. The overall objective is to inform the substitution of hazardous highly fluorinated chemicals in DWRs on outdoor garments and to test the applicability of LCA in this context. The LCA was part of a publicly-funded project supporting sustainability transitions in the garment industry that arose from dialogue between various researchers and industrial companies, rather than the needs of any single organization. The intended audience for this work includes garment and DWR manufacturers, government policy makers and LCA practitioners.

2. Material and methods

The method for this study was essentially an LCA based on the four steps outlined in ISO14044. So it began with goal and scope definition to clearly define the ambit of the study, followed by collection of an inventory of resources and emissions associated with the function of the objects under study. Life cycle impact assessment and interpretation of results in the light of sensitivity analysis completed the study. The contributions via the inventory and impact assessment steps are in particular focus here.

2.1. Life cycle assessment goal and scope

The overall goal of this study was to contribute to a sustainable substitution of hazardous side-chain fluorinated polymer based DWRs on outdoor garments. The study had two main objectives; i) to demonstrate the feasibility of a new approach to assessing DWRs in a life cycle framework, and ii) to identify environmentally preferable DWRs and key parameters in the environmental performance of DWR systems. To achieve this goal a cradle-to-grave, attributional LCA of water repellent

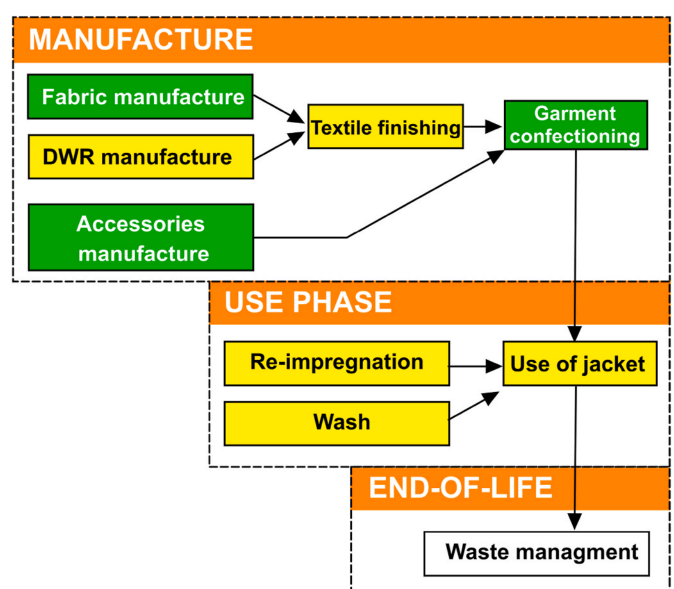

Fig. 1. Simplified system diagram for a DWR impregnated garment. Foreground processes are yellow, middle ground processes are green and background processes are white. Note that this is a simplification and each box contains more processes than are shown here. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

jackets with alternative DWR was performed. Five alternative DWR systems were selected for assessment (Fig. 2): two types of side-chain fluorinated polymers (C4 and C6); one silicone-based DWR; one hydrocarbon-based wax and a non-fluorinated DWR based on hyperbranched polymers (highly branched three-dimensional macromolecules). These five DWR systems give a good coverage of the types of DWR systems currently on the market (Holmquist et al. 2016). Additionally, a C8 system was selected as benchmark. The six DWR systems were applied to a medium performance, "standard" jacket to construct six hypothetical but realistic cases (i.e. different DWRs on an otherwise identical jacket). Industrial stakeholders; DWR producers, finishers and garment brand owners were involved in the goal and scope setting to achieve a realistic model with relevant functional criteria for the garments. The LCA was constructed to provide decision support to policy makers and garment manufacturers in the substitution process of C8 DWRs.

In breathable water repellent outdoor garments the outer fabric is impregnated with DWR but a membrane or an interior coating of the fabric makes the garment waterproof. The low surface energy of DWRs hinders water droplets from penetrating the outer fabric, which can otherwise cause a wet-out and make the wearer feel cold. The water repellency of the outer fabric can also affect the breathability of the laminate, e.g. if oily substances or water clog the pores, vapor transfer is retarded, and the wearer may feel sweaty and eventually cold. Furthermore, the additional function of oil- and stain-repellency provided by some DWRs can affect the need for washing and potentially also the garment life length. The functional unit (FU) was for the standard jacket set to "keeping the wearer warm and dry during one use (30 min) of the jacket". This use could occur during rain or dry conditions, as these kinds of garments are not solely used as rain protection. Technical performance of the DWR is key for the garment to deliver the desired function and specific criteria were identified in dialogue with industry stakeholders (S2.2). Data on the DWR performance described by standard tests of water repellency and oil repellency, before and after washing, abrasion and weathering, were obtained from recent research (see S2.1 and Schellenberger, 2019), and used to guide the selection of DWR systems for further study with LCA. The garment was considered to have reached its life length when the FU could no longer be achieved, despite reimpregnation. Garment life length could not be determined from the functional testing but was defined in dialogue with industry stakeholders.

The user scenario for the standard jacket was based on the DWR providing comfort during activities such as walking to work for 30 min every day in the summer season (April to October, assuming the use of a different, insulated garment in winter) and in all weathers. The jacket life length was assumed to be 5 years, meaning 2129 uses (i.e. used twice a day over the seven-month period). In the basic scenario the jacket was assumed to be washed and dried twice a year (i.e. nine times during its life length) with reimpregnation every other wash (i.e. four times during its life length). Schellenberger et al. (2018) tested water repellency of polyester (PES) and polyamide (PA) fabrics and found that water repellency after washing could differ as much between textile samples employing one DWR system, as between samples with different DWR systems. Due to the large variability in performance, no DWR performance parameters specific for the different DWR scenarios could be defined. Instead one basic scenario as defined above was the starting

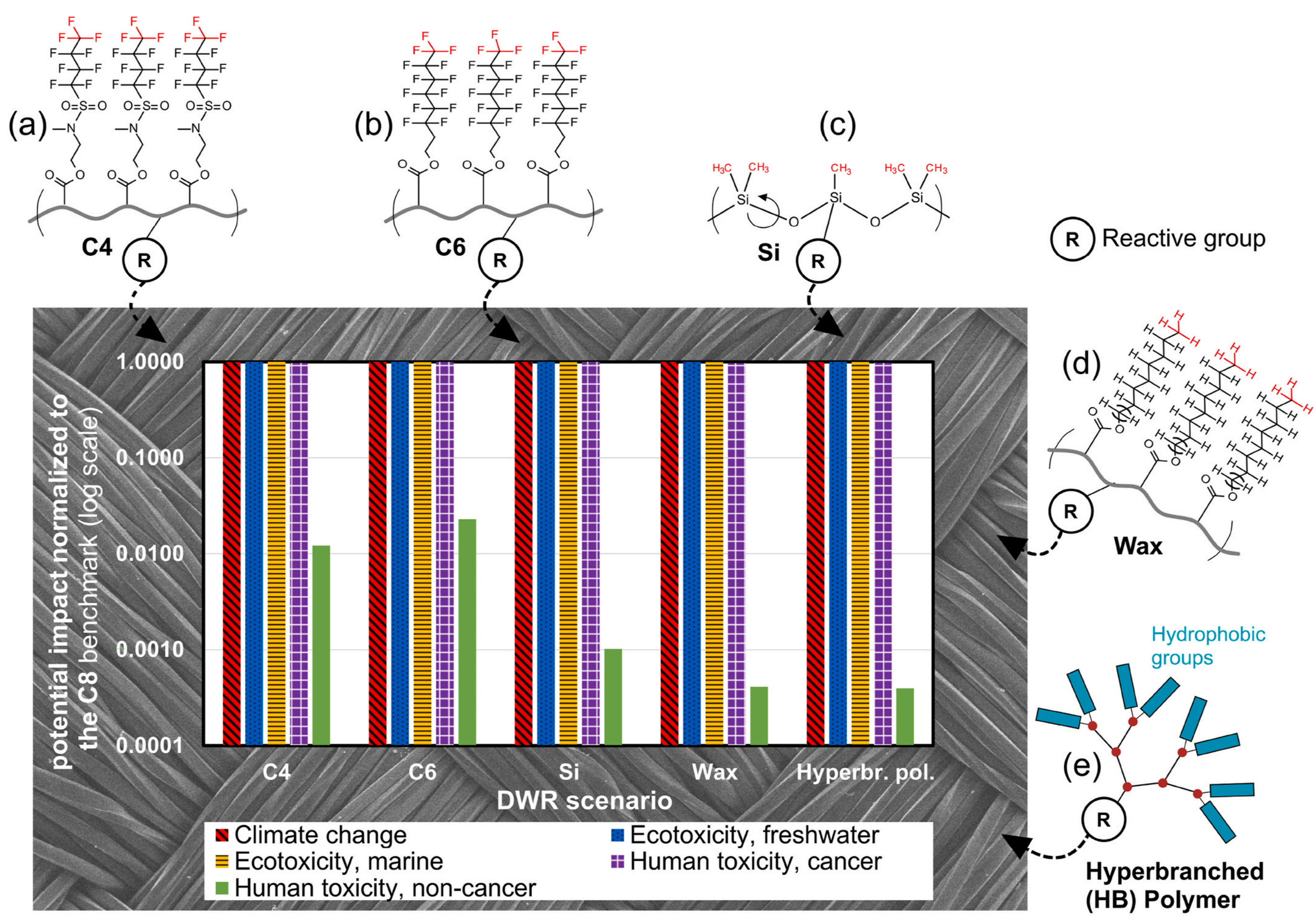

Fig. 2. Results for the standard jacket normalized against the benchmark C8 DWR over the full garment life cycle. Note that results for the PFAS-based DWRs were based on characterization factors based on roughly extrapolated epidemiological data and a 50% polymer degradation scenario. Note the log scale.

point of the assessment. Implications of deviations from this basic scenario due to better or worse technical performance were investigated using scenario analysis covering wash frequency, reimpregnation frequency and life length.

To investigate the relevance of garment design and use patterns, three additional garments are considered in less detail in the discussion in this article: an ambulance jacket, an "extreme" jacket and an insulated children's overall. An LCA model was set up for the ambulance jacket, while implications of material choices and user scenarios were qualitatively discussed for the extreme jacket and the children's overall. The standard jacket was modelled as a lightweight weave PA jacket with a waterproof, breathable polyurethane (PU) interior coating. The ambulance jacket was modelled as a heavier jacket with 2-layer PES and 3-layer PA weave and a polytetrafluoroethylene (PTFE) membrane. While the standard jacket can be expected to be washed a few times per year, the ambulance jacket was assumed to be washed weekly according to the routines at a Swedish ambulance station.

The LCA modelling was conducted in GaBi 8.7.0.18 with databases ecoinvent 2.2 and GaBi Professional 2018 (service pack 36) used in addition to LCI data inventoried by Roos et al. (2019) and data collected specifically for the DWR finish; from finishers, DWR producers and the literature. Selected parts of the technical description and life cycle inventory (LCI) are included below and detailed descriptions are available in the supporting information (SI).

2.2. Life cycle inventory

Detailed life cycle inventory (LCI) data is hard to find for clothing products (Roos et al., 2015a,b). The full life cycle of our focus garment was included in the LCA (Fig. 1) and the detailed LCI data are available for other analysts in the SI to this paper. The use phase and end-of-life occurred in Sweden, but the supply chain was global. Data selection choices were made to capture a realistic, full scale production system in the recent past. In the manufacturing phase, energy requirements were modelled with an Asian electricity mix according to Roos et al. (2015b, see S4.8.1).

The LCI was mainly conducted via dialogue with industry, databases and literature research. The modelled system was divided into background, "middle ground" and foreground processes. The foreground was considered to incorporate processes for the manufacture and application of the DWR, as well as use phase processes dependent on DWR performance. The middle ground included processes where the garment manufacturer can exert influence, other than those related to the DWR, and the rest are background processes. We focused on the foreground system, where communication with industry representatives, site visits, chemical analysis and technical performance experiments complemented literature data. The middle ground was based on collaboration with a number of garment manufacturers and garment material composition was based on real garments. Furthermore, the middle ground LCI was based on recent work by Roos et al. (2015b), Roos (2016), Roos et al. (2019) on inclusion of (eco)toxicity and chemical inventory in textile LCAs. The bill of material (BOM) for the standard jacket and the ambulance jacket, as well as detailed process descriptions for the full LCI (with anonymized DWR recipes) can be found in section S4.

2.2.1. The DWR systems

Based on an inventory of DWR products typical recipes were constructed, aiming for a realistic but brand-independent model, using proxies available in LCA databases. For each DWR system included in the study, ingredient lists were constructed based on data in material safety data sheets (MSDS), complemented with literature, data from communication with the industry or expert judgement when needed. The inventory made by Roos et al. (2019) was the starting point. The full DWR products, i.e. not only the water (and oil) repellent functional polymer, but also cosolvents and dispersing agents were included, and each ingredient was classified according to Table S1. See Holmquist et al. (2016 Table S1) for a description of a typical DWR product. The impurities identified for DWR products by Holmquist et al. (2016) were also included. To achieve a "time-integrated" emission inventory, i.e. including the primary pollutants and relevant degradation products, persistent degradation products were identified and included based on Roos et al. (2019) for general textile chemicals and Holmquist et al. (2020) for side-chain fluorinated polymer based DWRs. Crosslinkers used in the finishing step were included in the models for all products except the Si DWR. For the Si DWR the DWR recipe itself contained all chemicals needed for the DWR finishing so no additional products were included in the model.

The data inventory for the foreground system was complicated by confidentiality around DWR products and their manufacture. Therefore, a stepwise data collection strategy was implemented: first when "LCA-data" for the relevant process was available, that data was used and complemented with direct chemical emissions (by application of emission scenarios, see section 2.2.2) if relevant chemicals identified by Holmquist et al. (2016) were not already included in the emissions inventory. LCA-data process inventories attempted to cover the use of resources, chemicals, energy and water and the emission of pollutants. For example, industry data (complemented by secondary data) for an aggregated process for the manufacture of C-6 fluorinated acrylic copolymer by esterification of 2-(perfluorohexyl)ethanol by acrylic acid was available from Sphera. If such LCA-data were not available, unit process inventories were constructed based on information in the literature, see e.g. the LCI for manufacture of the N-methyl perfluorobutanesulfonamido vinyl polymer (i.e. the polymer assumed to be used in the C4 DWR) (S4.2.1), using LCA-data for similar components as proxies in order to achieve a complete inventory. Transports were not in focus and only added as a summary parameter in the garment model, excluding transports in the foreground system.

2.2.2. Chemical emissions

For consistent inclusion of chemical emissions in the inventory of the foreground processes, scenarios were created for the emissions to air, water and soil from industrial facilities (S3). The emission scenarios depend on system losses, i.e. emissions created at the industrial site, and further treatment, i.e. by wastewater treatment (WWTP). The influence of system losses on the results was evaluated in scenario assessment while the variability of downstream WWTPs was considered to be out of scope of this study.

Emissions also arise during textile use, e.g. when the clothes are washed (Haglund et al. 2016). These use-phase emissions were considered by employment of emission factors for diffuse emissions to rainwater, air and wash water of non-polymeric impurities and fiber bound polymers (Schellenberger et al., 2019b, Steffen Schellenberger pers. comm., Ike van der Veen, pers. comm. and S3.2).

2.3. Life cycle impact assessment

Significant methodological advances have been made recently in the impact assessment of toxic chemicals in LCA frameworks. Relevantly for this paper, a new approach to assessing the ecotoxicity and human toxicity of PFAS chemicals has been developed that better considers their persistence and amphiphilic behavior. This paper is the first time this new approach has been employed to assess garments. *(Eco)toxicity* indicator results were based on USEtox CFs (Rosenbaum et al. 2008), as in the ILCD PEF recommendations (European Commission, 2013), complemented CFs calculated by Roos et al. (2018) and Holmquist et al. (2020). Holmquist et al. (2020) presented two sets of *human toxicity* CFs for PFAAs: CFs with effect factors based on laboratory derived rodent data, and higher CFs roughly extrapolated from human epidemiological data for perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS). Depending on the effect factor the CFs differed by orders of magnitude. In this LCA the higher CFs were used despite their more unconventional data basis. Implications of this data selection are discussed. The extreme persistence of the PFAAs was carefully considered in the CFs for adequate representation of chemical accumulation in the environment (Holmquist et al., 2020). The toxicity assessment was complemented by more traditional indicators covering acidification, climate change, eutrophication, ozone depletion, primary energy, resource depletion and water use. Indicator results for these indicators were calculated with the ILCD PEF v. 1.09 recommendations except for the primary energy (primary energy demand from renewable and non-renewable resources (gross cal. value), which is not included in the PEF/ILCD recommendations).

2.4. Interpretation via scenario assessments

The robustness of the results was evaluated and key parameters defining environmental performance of DWRs were identified in scenario assessments:

- Six drop-in substitution scenarios including five alternative DWR systems and a C8 benchmark.
- Four kinds of garment use phase scenarios: two sets of scenarios in which life length was either halved or reduced by a factor of 10, and two other sets of scenarios in which the intervals between washes and between reimpregnations were either doubled or increased by a factor of 10.
- Emissions from the foreground system were assessed in low and high emission scenarios. In the low emission scenario, losses in chemical industry and textile finishing were both set to 1% (10% and 5% in the basic scenario, respectively) and diffuse emissions in the use phase were reduced by setting the degradation rate of side-chain fluorinated polymers to zero (50% in the basic scenario). In the high emission scenario system losses were set to 30% in the manufacturing and finishing processes and polymer degradation was set to 100%.
- The importance of electricity sourcing was explored by corner-stone scenarios, exchanging the mainly fossil-based Asian energy mix in the basic scenario (i.e. the textile industry average electricity mix defined by Roos et al. (2015b)) with a scenario where only renewable energy (electricity, from a wind power plant in Europe) was used and a scenario where all energy was from coal (electricity, from a hard coal power plant in China).

3. Results

3.1. Life cycle impact assessment of the standard jacket

3.1.1. DWR scenarios

It was feasible to apply the new approach to the assessment of alternatives to the C8 DWR. Fig. 2 shows the LCA results normalized against the C8 system (which is not shown per se, but set to 100%). In the figure, the garment life length and user behaviors are held constant, to isolate potential impact from change of DWR. It shows large differences in the human toxicity non-cancer indicator scores – three orders of magnitude in some cases depending on the choice of DWR. The C4 and C6 DWRs have less than 5% of the non-cancer toxicity compared to the C8 DWR, and the non-fluorinated DWRs have less than 0.1% of its toxicity in the same category (if the CFs based on roughly extrapolated epidemiological data were used, but not if the analysis was restricted to rodent data effect factor based CFs). Excluding the garment (that is identical across systems) and looking only at the DWR finishing (Fig. S3, including cradle-to-gate impacts from the DWR manufacture, auxiliary chemicals and the finishing process but excluding garment and textile manufacture), the differences between the DWR systems become more pronounced, but still mainly in the human toxicity non-cancer indicator, where, again, C4 and C6 DWRs have less than 5% and non-fluorinated DWRs have less than 0.1% of C8 toxicity. On the other hand, there are only small variations in climate change, ecotoxicity and human toxicity cancer indicators. In those categories since differences were relatively small (indicator scores were within the same order of magnitude), so it is difficult to determine if they are due to actual differences between the DWR systems, or are a reflection of data uncertainty. Non-normalized results for all impact categories are reported in S5.

Regarding the key parameters contributing to the results, a contribution analysis for the climate change indicator showed that for the full life cycle of the garments the manufacture of the PA weave was the main contributor, mainly due to its electricity use in the textile manufacturing processes and the nylon 6.6 production. For the DWR finishing the combustion of natural gas and electricity required for the finishing was the main contributor in all systems. The energy required for finishing was equal for all DWR systems as the finishing process, the so-called pad-dry-cure (Schindler and Hauser, 2004), was the same in all systems. The DWR and auxiliary chemical manufacture contributed <50% across systems for finishing.

A contribution analysis for the (eco)toxicological indicators was made for the full garment life cycle. For the freshwater and marine ecotoxicity indicators, aluminum emissions to freshwater were the main contributor to the indicator scores (>90% across systems). The main contributors were indirect metal emissions generated from electricity production for PA fabric manufacturing. The PFAAs made a negligible contribution to the marine ecotoxicity indicator, despite their relatively high CFs in this category (cf. Holmquist et al., 2020 Fig. 2). For the human health cancer indicator, chromium (+VI) freshwater emissions were the main contributor (>70%). Also here the main contributor was the electricity production for the PA fabric manufacturing process. While the ecotoxicity and human health cancer indicator scores were dominated by energy related processes the picture is different for the human health non-cancer indicator for the DWRs based on side-chain fluorinated polymers, where PFAA emissions to water and soils, generated during the DWR manufacture and use phases of the LCA, constitute the main contributors (>30% and >50%, respectively). This PFAA contribution was dependent on the data selection for the CFs; results presented here use extrapolated epidemiological data for effect factors (see further Holmquist et al., 2020). For the non-fluorinated DWRs the energy related processes contributed most of the human health non-cancer indicator scores, mainly due to heavy metal emissions.

The results of a contribution analysis for the DWR finishing part of the system in isolation (Fig. S3) were similar to those of the full system. For the side-chain fluorinated polymer DWRs, emissions of the PFAAs, mainly from DWR manufacture, were main contributors to the human health non-cancer impact category (again, dependent on the choice of CF). In the human health non-cancer indicator mercury air emissions and arsenic freshwater emissions were important contributors in all non-fluorinated systems, these emissions were mainly generated from energy producing processes and are dependent on the energy source, e.g., the use of German electricity in a dataset for silicones included in the PA wet treatment model. The toluene diisocyanate in finishing processes cross-linkers contributed <1% of the human toxicity cancer indicator scores across systems. PFOA contributed <5% the human toxicity cancer indicator score for the C8 system.

The life cycle stage that contributes most to impacts depends on the impact category and DWR scenario. Looking at the life cycle of the whole garment, the majority of the impacts originated from the energy consumption in the manufacturing life stage, both for climate change and three of four (eco)toxicity categories (ecotoxicity and human health cancer) for all DWR scenarios. In the human toxicity non-cancer indicator however, the manufacturing stage dominated the wax and hyperbranched polymer scenarios, while for the side-chain fluorinated and silicone DWR scenarios, the use phase instead dominated the overall results. For the side-chain fluorinated polymer scenarios, PFAA emissions in the use phase were the main contributors to the indicator score, while for the Si-system it was mercury emissions, and for this origin in energy systems is a plausible explanation.

Predicted emissions of DWR related substances (Holmquist et al. 2016) were higher in the use phase compared to manufacturing (Table 1). More than 80% of PFAA emissions were associated with the use phase. Fiber loss (with a 50% polymer degradation scenario, see Holmquist et al. (2020)) and diffuse release of impurities contributed 30–50% of PFAA mass flows in the use-phase. These differences between the DWR scenarios have implications for the identification of key parameters for the environmental performance.

3.1.2. Garment care and life length scenarios

Regarding key scenario variables, the scenario assessment (Table 2 for wash scenarios, Table S28 for full scenario results, Fig. S17) showed that wash, reimpregnation and life length changes have different effects on the indicator scores per FU depending on what DWR-system is used on the jacket. For all DWR scenarios, i.e., independent of DWR system, garment life length was important. Halving the life length means that over a five-year period the user would need to buy two jackets and thus all potential impacts originating from the manufacturing would double. However, for jackets with DWRs based on C4, C6 or C8, life length was less important for the human health non-cancer toxicity indicator, as use-phase emissions were more important due to the high potential toxicity of the PFAAs emitted from the side-chain fluorinated polymer DWRs. Compared with the modelled life length changes, the changed wash frequencies (Table 2) had relatively moderate effects on garment environmental performance, and reimpregnation frequencies had little effect, again with the exception on human health non-cancer indicator scores for the side-chain fluorinated polymer based DWRs. Comparing the scenario with 90 washes (i.e., washing more than twice a month during the use period) for jackets with the non-fluorinated DWRs with the basic scenario (9 washes) for jackets with C4 and C6 DWRs shows that the predicted potential impact in the non-cancer toxicity was still higher for the fluorinated systems (Fig. S18).

3.1.3. Uncertainty in (eco)toxicity assessment

The CFs calculated in USEtox (or any LCIA method) are associated with uncertainty and the results must differ by several orders of magnitude to be robust (Rosenbaum et al. 2008), which is why results are here interpreted with caution. Herein all available CFs have been used: both recommended and indicative USEtox CFs; CFs that did not meet minimum data quality criteria in Roos et al. (2018) and also CFs based on roughly extrapolated human epidemiological data by Holmquist et al. (2020). We acknowledge that CF uncertainty is high for some substances but argue that leaving flows uncharacterized would generate equal or higher uncertainty.

In the foreground system sensitivity of the (eco)toxicological indicator scores to the emissions scenarios and polymer degradation rates was tested in additional scenario assessment as described in section 2.3. The effects of changes were limited, except for human toxicity non-cancer effects, in particular for the side-chain fluorinated polymer DWR systems (Table S29). This is in line with the observation that background processes make the largest contributions to climate change and (eco) toxicity indicator results in these models.

The chemical constituents of the DWR products have been approximated based on MSDSs and other information sources. MSDSs do not always describe exact chemical content and assumptions have been necessary to derive the LCI. For example, exact polymer structures were not known, "glycols" have been approximated with ethylene glycol and dispersing agents were assumed (S4). Dialogue with industrial stakeholders revealed that those approximations are not representative of specific popular products, but this was not the intent of this LCA, and in any case, foreground DWR-related emissions make relatively small contributions to the aggregated indictor scores so this uncertainty is of relatively small importance.

The energy mix used in textile manufacturing in the basic model scenario (i.e. the electricity mix by Roos et al. (2015b)) can be considered a global textile industry average for both climate change and (eco) toxicity. The wind power scenario resulted in reductions in indicator scores across impact categories and DWR systems, while the coal scenario resulted in increases (Table S30). These changes were smaller than many of the changes modelled in the use phase scenarios.

Table 1
Predicted emissions (μg) of the DWR related substances per functional unit and life cycle stage in the basic scenario. On account of the disposal mechanism, garment disposal was predicted to cause no emissions.

Substance	Manufacturing	Use phase	Sum
Benchmark C8			
Perfluorooctanoic acid (PFOA)	9.1	43	52
C4			
Perfluorobutanesulfonic acid (PFBS)	8.8	67	76
C6			
Perfluorohexanoic acid (PFHxA)	8.5	40	49
Si			
D4, Octamethylcyclotetrasiloxane	0.021	0.24	0.26
D5, Decamethylcyclopentasiloxane	0.021	0.24	0.26
Dimethylsilanediol (DMSD)	53	250	310
Hydroxytrimethylsilane (trimethylsilanol) (TMS)	0.010	0.097	0.11
Wax (emulsion)			
Paraffins	76	280	350
Hyperbranched polymer			
Unknown	–	–	–

4. Discussion: implications for researchers and the clothing industry

4.1. Consequences of emerging (eco)toxicological knowledge

The comparison of DWR systems on a garment basis was able to show that DWRs based on side-chain fluorinated polymers are associated with large potential human health non-cancer impacts. This LCA combines emissions, fate and effects, and further substantiates the seriousness of extreme persistence combined with possible adverse effects highlighted by Holmquist et al. (2016). The LCIA for the PFASs was based on a newly developed LCIA framework for (eco)toxicity (Holmquist et al., 2020). As part of the uncertainty assessment for that framework, PFAA effects were quantified using new recommendations from EFSA, where effect thresholds for PFOA and PFOS were reduced by orders of magnitude compared to previous threshold values (EFSA CONTAM Panel *et al.* 2018). These PFOA and PFOS effects were extrapolated to the short-chain PFAAs (perfluorobutanesulfonic acid, PFBS and perfluorohexanoic acid, PFHxA) by use of toxicokinetic data. Hence the LCIA framework went beyond the hazard assessment in the quantification of effects, mainly because between the studies EFSA published new recommendations, but also because the data collection procedures in LCIA are less prescriptive than the hazard assessment scheme applied by Holmquist et al. (2016). Only when CFs are based on roughly extrapolated epidemiological data could side-chain fluorinated DWRs be identified as worse than other DWR alternatives. Using instead the rodent based CFs no differentiation between DWR systems was possible. On the one hand those extrapolated CFs are highly uncertain, as the use of epidemiological data in CF calculation is not common practice, but also because it is not known if those low-dose effects as seen for PFOA are relevant for the short-chain PFAAs. On the other hand, the fact that EFSA has lowered PFOA thresholds by orders of magnitude based on epidemiological data, indicates that the more conventional CFs based on rodent data are also uncertain. Based on a precautionary approach, applying the higher CFs, it can be concluded that the LCA indicates that non-fluorinated DWRs are preferable to DWRs based on side-chain fluorinated polymers.

Table 2
Results of washing scenario assessment for the standard jacket showing increased impacts per functional unit (%) from increasing wash frequency by 2 or 10 times. Hy. br. = hyperbranched polymer DWR, C4, C6, C8 = side-chain fluorinated polymer based DWR, Si = silicone based DWR. Changes <10% are marked in green, >10% < 50% in yellow and ≤50% in red.

	×2	×10	×2	×10	×2	×10	×2	×10	×2	×10	×2	×10
	C8	C8	C4	C4	C6	C6	Wax	Wax	Si	Si	Hy.br.	Hy.br
Climate change	3.6%	33%	3.6%	33%	3.6%	33%	3.6%	33%	3.6%	32%	3.7%	33%
Ecotoxicity, freshwater	17%	150%	17%	150%	17%	150%	17%	150%	17%	150%	17%	150%
Ecotoxicity, marine	16%	150%	16%	150%	16%	150%	16%	150%	16%	150%	16%	150%
Human toxicity, cancer	20%	180%	21%	190%	21%	180%	21%	190%	20%	180%	21%	190%
Human toxicity, non-cancer	0.59%	5.3%	1.4%	13%	1.1%	10%	26%	240%	11%	95%	27%	250%

4.2. Relevance of a systems perspective

The inclusion of DWR treatments in a wider context (i.e. in an LCA) showed that, except for the PFAS related potential impacts, the importance of the chemicals directly emitted by the DWR system was small in comparison to energy related (indirect) emissions. Any environmentally motivated intervention in DWR selection, manufacture or finishing practices, must thus focus not only on direct chemical emissions but also on energy use, and do so with a life cycle perspective. However, the importance of these indirect emissions could be overestimated as: i) metal emissions related to energy production may be overestimated (Roos, 2016), ii) CFs are available for many metals, while organic substances more often lack CFs, and iii) direct chemical emissions may be underestimated, as database processes rarely contain such inventories and the recipe-based emission estimates include only known content. Other impurities, degradation products or chemicals used for e.g. cleaning of equipment might not be fully captured. Although a major effort was made here to include textile relevant chemicals in the LCIA there are still substantial data gaps and uncertainties. These results should serve as a reminder to consider energy related indirect (eco) toxicity but at the same time they are a reason for further data collection and model development for inclusion of direct (eco)toxicity in LCIA.

4.3. What about other types of garments?

The focus in this article has been on a "standard jacket". To extend the discussion of the results, an LCA was also conducted for an ambulance jacket (see S5.3), while an "extreme jacket" and a children's overall were considered qualitatively.

A comparison between the standard and ambulance jacket showed that predicted environmental impacts were higher for the latter as it was a heavier jacket (more material needs to be produced) and because its user scenario included extensive washing. However, the relation between the different DWR treatments remained. Throughout this work reimpregnation was modelled with the same DWR product as was used in the industrial step. Ambulance personnel told us that non-fluorinated DWRs can be used in reimpregnation of a jacket that originally was treated with side-chain fluorinated polymer DWR (the same can of course apply to any garment). In such a scenario, with weekly washing but no additional use of PFASs, washing impacts (i.e., not PFAS related) might potentially outweigh the PFAS impacts also in the non-cancer human toxicity indicator scores modelled with the roughly extrapolated CFs based on epidemiological data. One important difference between the standard jacket and the ambulance jacket is that for the former, oil-repellency (i.e. a function provided by the use of side-chain fluorinated polymers in DWR) is not a safety issue, while it can be so for the ambulance jacket, e.g. in interventions at traffic accidents where oil absorbance could mean a fire hazard or reduced visibility to drivers. Thus, for the ambulance jacket, the avoidance of PFAS-related environmental impacts might not yet be possible.

LCA results for the four garments would not be directly comparable as they provide different functions. However, their differences, in construction and use, serve to illustrate important parameters defining a garments environmental performance, and the relative importance of DWR potential impacts. Life length and material consumption will inevitably be key parameters for environmental performance for any garment. Our study shows that wash frequency and DWR impregnation can also be of high importance but that this is dependent on characteristics of the DWR product. Depending on the expected function of a product, a small change in design or material can lead to a larger change in how the product is used, example, a reduction in stain repellency (Schellenberger et al., 2019a) could lead to more frequent washing of a children's overall, or a slight reduction in water repellency that could lead to dangers for users of the "extreme jacket" as there could be long periods outdoor without access to shelter for these users.

4.4. What if technical performance changes?

Our results present an interesting counterpoint to other life cycle studies on DWRs. Typically, studies of clothing life cycle impacts are dominated by impacts prior to garment sale (Peters et al. 2021). This LCA is consistent with those findings, but also shows a situation in which the use phase dominates certain indicators. This is partially consistent with an LCA on DWR treatments for a waterproof, windproof and breathable jacket (W. L.Gore & Associates GmbH 2015) in which the authors conclude that durability of the jacket and the DWR is key. The non-fluorinated DWR in their study generated an increased wash and reimpregnation frequency, which in turn affected (eco)toxicity indicators substantially. In another study DWRs were compared in an LCA focusing on the finishing step (Fierro and Martínez, 2018). DWRs based on side-chain fluorinated polymers were compared to silicone, hyperbranched, perfluorosilicone and paraffin based DWRs. The side-chain fluorinated DWRs were shown to have higher potential impacts compared to the non-fluorinated DWRs with toxicity and ozone depletion being the two most important impact categories. The jacket LCA might not have included extensive PFAS considerations in the LCIA because CFs for relevant substances were unavailable. The DWR finishing study (Fierro and Martínez, 2018) made use of the CFs calculated by Roos et al. (2018) and could therefore capture PFAS related toxicity potential impacts. In the results reported herein the LCIA step was further extended by use of the PFAS (eco)toxicity LCIA model by Holmquist et al. (2020), and CFs based on epidemiological data, indicating an even higher importance of the DWR-related PFAS emissions.

The scenario assessment varying wash, reimpregnation and life length parameters can be used to design a strategy for alternative DWR selection based on garment environmental performance. Garment life length was a key parameter for all DWR-systems, as it is for textile products in general (Sandin et al. 2019). This is a natural consequence of the large share of the environmental impacts that the manufacturing life cycle stage causes. Thus, interventions prolonging garment life length will improve a garment's environmental performance. One such intervention could be to improve the users' garment care strategies to minimize washing and reimpregnation rates.

4.5. Can life cycle assessment support substitution processes?

The LCA of the standard jacket was constructed to capture potential environmental and human health impacts of a generic jacket impregnated with alternative DWRs. As such it can guide in a general selection of DWR. If a garment brand owner would like to compare particular alternative DWRs for selection to use in their further production, an LCA model specified to those exact DWR products and finishing systems can be set up and that model will be able to further differentiate between alternatives. Our results demonstrate that including the garment care and reimpregnation products in the concept is feasible and essential for the environmental performance. Furthermore, the inclusion of four different types of garments in this study further substantiated the need to construct a model for a specific garment if results are needed for that specific garment. The LCA results presented in this paper can then be used to guide modelling choices and support in the interpretation of results.

This LCA demonstrated the feasibility of evaluating alternatives in the light of a change in DWR function from keeping the wearer warm and dry, to stain repellency, as the evaluation is based on end user function requirements, and product changes in relation to that function can be included in the model. The functional requirements in a chemical substitution assessment cannot be static but need always be re-evaluated in relation to end user requirements. In some cases it may also be that environmental performance criteria outweigh functional criteria (cf. Hansson et al. 2011). Certain DWR applications may still require functions that can only be given by the side-chain fluorinated polymers, e.g. certain medical textiles and work wear (Schellenberger et al. 2019). In these cases, the essential use concept as proposed by Cousins et al. (2019) is a useful tool to evaluate whether PFAS can be phased-out or not for a specific product.

Regarding the overall objective of this LCA study. the results suggest four interventions for industrial and regulatory stakeholders to improve environmental performance of garments with DWRs:

- Substitute DWRs based on side-chain fluorinated polymers to non-fluorinated DWRs where possible.
- Select the DWR and other garment components to give the garment as long life length as possible and to need as few wash and care cycles as possible.
- Actively work with energy sourcing in the value chain to reduce both climate change and (eco)toxicity impacts.
- Keep levels of impurities low in DWR formulations.

This application of LCA to provide relevant information to a substitution assessment and shows that LCA can be a key method to assess trade-offs to avoid sub-optimization. Results are however dependent on careful data selection as was shown herein by the dependency on whether PFAA effect parameters were based on human or rodent data.

CRediT authorship contribution statement

Hanna Holmquist: Conceptualization, Methodology, Data curation, Formal analysis, Visualization, Writing – original draft, Writing – review & editing. **Sandra Roos:** Resources, Formal analysis, Writing – review & editing. **Steffen Schellenberger:** Resources, Formal analysis, Writing – review & editing, Visualization. **Christina Jönsson:** Conceptualization, Writing – review & editing, Funding acquisition, Project administration. **Gregory Peters:** Conceptualization, Methodology, Validation, Formal analysis, Writing – review & editing, Funding acquisition, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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