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Review

Life cycle assessment (LCA) studies on flame retardants: A systematic review

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ABSTRACT

Considering the advantages and vast use of FRs in different sectors, there are serious concerns regarding negative impacts of FRs on the human health and the environment as we are exposed to FRs throughout our lives. Nonetheless, FRs are usually neglected in the studies on the environmental impacts of polymers with life cycle assessment (LCA). Firstly, this paper gives an overview over different FRs and the associated health and environmental concerns and policies. Afterwards, the LCA studies on FRs are systematically reviewed and discussed. This includes analyzing the LCA methodologies and data for different types of FRs and applications as well as the contribution of their different life cycle phases. The results of this review highlight the importance of preserving the interconnection between three domains of environmental impacts, human exposure and health concerns while considering the contribution of different life cycle phases. Furthermore, it brings insights into the pros and cons of current FR solutions, the existence of a "time lag" between production, use and end of life phases and the role LCA can play in technological development of FRs noting environmental and health concerns. The recommendations include considering fire occurrence as one of the end of life scenarios (fire-LCA), determining the optimum amount of FR with the minimum environmental and health impacts that can prevent fire occurrence and new approaches for the transition to a circular economy for FRs.

Contents

1. Introduction

The plastic production has increased twenty-fold in the last 50 years (Ellen MacArthur Foundation, 2016). Almost 335 Mt of

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Nomenclature

ATH	aluminum trihydroxide
BFR	brominated flame retardant
BPA-BDPP	bisphenol A bis(diphenylphosphate)
Br	bromine
CFR	chlorinated flame retardant
Cl	chlorine
CNT	carbon nanotube
CO	carbon monoxide
CO_2	carbon dioxide
CP	chlorinated paraffin
CPSC	consumer product safety commission
deca-BDE	decabromodiphenyl ether
ECHA	european chemicals agency
EFRA	european flame retardant association
EPA	environmental protection agency
EU	european union
FR	flame retardant
FU	functional unit
GHG	greenhouse gas
HBCD	hexabromocyclododecane
HFFR	halogen-free flame retardant
HFR	halogenated flame retardant
LCA	life cycle assessment
LCIA	life cycle impact assessment
LED	light emitting diode
MCCP	middle chain chlorinated paraffin
MDH	magnesium hydroxide
MPP	melamine polyphosphate
MSS	most sensitive species
NIEHS	national institute of environmental health sciences
octa-BDE	octabromodipheyl ether
PAF	potentially affected fraction
PAH	polycyclic aromatic hydrocarbon
PBB	polybrominated biphenyl
PBDE	polybrominated diphenyl ether
PCB	printed circuit board
penta-BDE	pentabromodiphenyl ether
PFR	phosphorus based flame retardant
POP	persistent organic pollutant
PU	polyurethane
PVC	polyvinyl chloride
RBDPP	resorcinol bis(diphenylphosphate)
RoHS	restriction of hazardous substances
SCCP	short chain chlorinated paraffin
TBBPA	tetrabromobisphenol A
TBDD	2,3,7,8-tetrabromodibenzo-p-dioxin
TCDD	2,3,7,8-tetrachlorodibenzo-p-dioxin
US	united states
WEEE	waste electrical and electronic equipment
WFD	water framework directive

plastics were produced in 2016 globally (Leal Filho et al., 2019) and this number is predicted to double in 20 years and almost quadruple by 2050 (Ellen MacArthur Foundation, 2016). However, the service life of 40% of plastic products is estimated to be less than a month. The question then arises, “What happens to all these plastic products afterwards?”. In Europe, for instance, in spite of all the technical developments and environmental concerns, half of the plastic waste ends up in landfill disposal (Hahladakis et al., 2018) where it may remain up to 1000 years (Aryan et al., 2019). Another remarkable fraction of plastic waste is openly burnt associated with carbon monoxide (CO) and carbon dioxide (CO_2) emissions (Hahladakis et al., 2018). Besides, at least 8 Mt of plastics end up in the ocean every year. This amount is equal to dumping the contents of one garbage truck into the ocean every minute. In a business as usual scenario, this is expected to increase to two per minute by 2030 and four per minute by 2050 (Ellen MacArthur Foundation, 2016).

Additives are the chemical substances which are added to the plastic products for improving their properties (Hahladakis et al., 2018) and increasing their durability (Van den Oever and Molenveld, 2017). Additives constitute around 10% of the weight of plastics and can account for 5%–40% of their cradle to grave greenhouse gas (GHG) emissions (Broeren et al., 2016). Flame retardants (FRs) are among the most commonly used additives (Hahladakis et al., 2018) and represent 18% of the global plastic additive sales by weight (Broeren et al., 2016). In 2015, the total consumption of FRs was 2.49 Mt globally (Grand View Research, 2017) and over 600,000 t in Europe (Delva et al., 2018). However, most studies on the environmental impacts of polymers with Life Cycle Assessment (LCA) do not consider additives such as FRs (Jonkers et al., 2016), while the environmental effects cannot be neglected.

Considering the advantages and vast use of FRs in different sectors, we are exposed to FRs throughout our lives (Nazaré, 2009). By development of FRs, their presence and concentration in the environment has increased in recent years. Accordingly, the exposure in humans is reported through ingestion of dust, food and air, dermal absorption as well as disposal sites (Yasin et al., 2018). According to the director of the National Institute of Environmental Health Sciences (NIEHS) in the United States (US), 97% of the US residents have measurable quantities of toxic organohalogen FRs in their blood (“Consumer Product Safety Commission (CPSC),” 2017). Various authors such as Aschberger et al. (2017), Escamilla and Paul (2014), Hardy et al. (2003), Kim et al. (2006) and Yasin et al. (2018) highlight the negative health and environmental impacts of FRs. These concerns have gone so far that some authors such as Simonson et al. (2002) believed that “it is better to let things burn more often than to use flame retardants”. On the other hand, Nazaré (2009) believes that in spite of such concerns, there is limited knowledge about the long-term effects of FRs and former studies prove no certain risk of using them to human health or the environment. However, she acknowledges the undesirable health impacts of the release of highly toxic substances in case of fire and burning of FRs. She declares that the inhalation of toxic gases such as CO is reported to be the main cause of death in fire accidents beyond the burning. The discrepancy in the outcomes of different studies can be explained by various types of FRs and lack of information on their environmental and health impacts in different phases of their life cycle and the consequent human exposure. Noting the discussed concern on the use of different FRs and limited knowledge on their LCA, this paper aims at reviewing LCA studies on FRs. The research question is articulated as “what is the state of the art of the scientific literature on LCA studies of FRs?” First, we provide an overview over different FRs, their main categories and recent developments in FRs. Then, the LCA studies on FRs are systematically reviewed. Finally, the outcomes and conclusions of this review will be discussed.

2. Flame retardant (FR)

A flame retardant (FR) is an additive that aims at inhibiting, suppressing or delaying an ignition in plastics. The growing use of thermoplastics and thermosetting polymers in different applications as well as more fire safety requirements have led to an increase in use of FRs in different sectors over the last decades (Aschberger et al., 2017). Various authors have highlighted the demand for FRs in sectors such as textiles (Dahllöf, 2004), (Nazaré, 2009) and (Yasin et al., 2018), transportation (Nazaré, 2009) and (Escamilla and Paul, 2014), construction (Escamilla and Paul, 2014), military (Nazaré, 2009), electronics (Deng et al., 2016) and (Pourhossein and Mousavi, 2018) and upholstered furniture (Chivas et al., 2009). In 2014, the building and construction sector accounted for the largest use of FRs in Europe (37%) while wires and cables accounted for 20% and electronics and appliances for 12% (PINFA, 2017). According to the European flame retardant association (EFRA), 12 deaths and over 120 fire-related injuries are reported in Europe every day (Delva et al., 2018).

Various scholars such as Aschberger et al. (2017), Escamilla and Paul (2014), Nazaré (2009), Pereira and Martins (2014), Segev et al. (2009) and Yasin et al. (2016) have categorized FRs in different ways mainly based on their active chemical component. One of the most common ways to classify FRs is to divide them into halogenated FRs (HFRs) and halogen-free FRs (HFFRs). HFRs are based on halogens and have been used broadly in different applications. The most common types of HFRs are based on Bromine (Br) and Chlorine (Cl) and called Brominated FR (BFR) and Chlorinated FR (CFR), respectively (Aschberger et al., 2017). Following the extensive use of HFRs in different sectors, there is an increase in the concentration of HFRs in the environment as well as human tissues which arises questions in terms of health and environmental impacts. As HFRs are resistant, they can move long distances and bioaccumulate (Aschberger et al., 2017). Traces of HFRs are even reported in human breastmilk aside from indoor air dust, in seafood, commercial water containers and human serum samples (Hendriks et al., 2012a). HFRs can cause toxicity due to the halogen element synthesis, are an endocrine disrupter and can lead to dioxin formation upon incineration (Deng et al., 2016). Policymakers have been trying to limit and restrict their use. The fight against the use of HFRs in Europe was initiated in the early 90s by different environmentalists (Simonson et al., 2002). Nowadays, the European Union (EU) has restricted the use HFRs. Consequently, there is a growing interest in developing HFFRs based on organic (mainly phosphorus and nitrogen) and inorganic (metals, boron, etc.) elements (Aschberger et al., 2017). BFRs, CFRs, phosphorus based FRs (PFRs), nitrogen containing FRs and inorganic FRs are discussed in the following sections.

Table 1 summarizes the classification of the main types of FRs. While BFRs, CFRs and PFRs constitute the majority of organic FRs, it should be noted that some FRs contain neither halogen nor phosphorus. Besides, some novel FRs may contain both halogen and phosphorus. In addition to the above classification, FRs can be divided into gas-phase-active and condensed-phase-actives while for each of these two types, they can act physically or chemically. Moreover, FRs can be categorized into additive and reactive ones. The reactive ones are chemically reacted into the material and released at high temperatures while the additive ones that are normally added to the product without bonding or reacting with the polymer leach out more easily (Hirschler, 2013).

2.1. Brominated FR (BFR)

BFRs decelerate or prevent the fire from starting by stopping the chemical chain reaction leading to flame formation. BFRs are the main use of bromine globally (Nazaré, 2009) and the most common BFRs are tetrabromobisphenol A (TBBPA), hexabromocyclododecane (HBCD) and three combinations of polybrominated diphenyl ethers (PBDEs), i.e. decabromodiphenyl ether (deca-BDE), octabromodipheyl ether (octa-BDE) and pentabromodiphenyl ether (penta-BDE). BFRs are frequently used for different applications due to their low impact on the characteristics of the polymer (Aschberger et al., 2017), high effectivity in comparison with other FRs and cheap price (Waaijers et al., 2013a). For instance, TBBPA is broadly used in light emitting diodes (LEDs) (Pourhossein and Mousavi, 2018). BFRs are persistent organic pollutants (POPs). POPs are the pollutants existing for a long time in the environment and resist against degradation and removal from environmental settings (air, water, soil, sediment) (Hendriks and Westerink, 2015). They have potential significant impacts on human health and the environment and can transport to the places where never been used (Alharbi et al., 2018), such as the poles of the earth. Nazaré (2009) states that BFRs are persistent and bioaccumulative and generate dixons and furans while burning. Simonson et al. (2002) also highlight the environmental concerns about the deca-BDE, being used as a FR in TV sets, due to POP molecules. The nervous and cholinergic systems have been reported (Hendriks et al., 2015) to be the most vulnerable targets of BFRs. Pourhossein and Mousavi (2018) highlight that high concentrations of TBBPA can impact hormone-sensitive parameters. For instance, the BFRs exists in most of the E-waste and can cause hazardous emissions such as dioxins and furans and negative health and environmental impacts.

The EU and North America started to voluntarily phase out the PBDE in 2003 (Hendriks et al., 2010). The EU Waste Electrical and Electronic Equipment (WEEE) and the Restriction of Hazardous Substances (RoHS) directives prohibited the use of polybrominated biphenyls (PBB) and PBDE in 2003 (European Commission, 2019). Deca-BDE and HBCD were also listed as candidates with very high concern by the European Chemicals Agency (ECHA) (European Commission, 2006). The EU prohibited the use of penta-BDE and octa-BDE in 2004 and listed them as hazardous substances under the Water Framework Directive (WFD). Deca-BDE and HBCD were also itemized as substances to be monitored. Eventually, the use of deca-BDE in electric and electronic products got prohibited in 2009 (Aschberger et al., 2017). The US Environmental Protection Agency (EPA) asked to phase out the deca-BDE by the end of 2013 (Brandsma et al., 2013a). The use of BFRs as well as chlorinated FRs is restricted and banned in some states of the US (Nazaré, 2009).

Hendriks et al. (2010) state that although the commercial production of PBDE is notably declined, the amount of PBDE in the

Table 1
Different types of FRs.

Category	Main chemical component	Example
Brominated FR (BFR)	Bromine	TBBPA, HBCD, PBDEs (deca-BDE, octa-BDE, penta-BDE)
Chlorinated FR (CFR)	Chlorine	Chlorinated paraffins (CPs)
Phosphorus based FR (PFR)	Phosphorus	Organophosphorus FRs
Nitrogen containing FR	Nitrogen	melamine derivatives
Inorganic FR	Metals (Al, Mg)	ATH, MDH

environment may still be growing. Accordingly, (Nazaré, 2009) believes that the demand for BFRs is increasing due to their preferable performance and low price. Abbasi et al. (2019) also highlight that in spite of the zero production of PBDE mixtures, the emission of PBDEs will last until 2050 due to a “time lag” between production, use and end of life phases. Hahladakis et al. (2018) explain that the separation of restricted BFRs is not possible in practice and this hinders the recycling of BFR containing plastics. However, Ragaert et al. (2017) discuss that detection of BFR containing plastics as well as extraction of BFR from the melt is feasible through techniques such as supercritical-fluid extraction or ultrasonic extraction. The Stockholm convention tolerates the recycling of PBDE containing plastics only under controlled circumstances (Hahladakis et al., 2018).

2.2. Chlorinated FR (CFR)

The most common type of CFRs are chlorinated paraffins (CPs) in the form of short chain chlorinated paraffins (SCCPs) and middle chain chlorinated paraffins (MCCPs). CPs are commonly used as FR and plasticizers in polyvinyl chloride (PVC) products globally. The release of halogen atoms into the gaseous phase prevents the material to reach the ignition temperature and contributes to retarding of polymer burning. SCCPs and MCCPs are carcinogenic, persistent, bioaccumulative and toxic substances and categorized as POPs (Kobetičová; Černý, 2018). The EU, Japan, US and Canada have already banned the production and import of SCCPs. The Council Regulation 793/93/EEC has labeled MCCPs as dangerous substances and the WFD has listed all CPs as “priority substances” for risk assessment. The Convention for the Protection of the Marine Environment of the North-East Atlantic-OSPAR also monitors MCCPs and SSCPs. On the other hand, these substances are used as FRs and plasticizers in the other parts of the world without any noteworthy control or prohibition. At the moment, China, Russia and India are the main producers and consumers of SCCPs as well as MCCPs in the near future (Kobetičová; Černý, 2018). However, China has also initiated “management methods for the restriction of the use of hazardous substances in electrical and electronic products” in 2016 (PINFA, 2017).

2.3. Phosphorus based FR (PFR)

PFRs can be classified into organic (organophosphorus) FRs and inorganic ones. Inorganic PFRs are based on compounds such as red phosphorus and ammonium phosphate (Weil, 1978) and organophosphorus PFRs include phosphate esters and phosphonates (Aschberger et al., 2017). Heating this type of FRs results in the release of phosphoric acid and, consequently, char formation and incomplete combustion (Aschberger et al., 2017). PFRs are considered as proper replacements for HFRs (Yasin et al., 2016) and can be used in numerous applications such as textiles, polyurethane (PU) foams, coatings and rubber (Aschberger et al., 2017). Organophosphorus FRs accounted for 14% of the global production of FRs while this number was 21% for BFRs (Brandsma et al., 2013a). Brandsma et al. (2013b) have investigated two types of organophosphorus FRs, i.e. Resorcinol bis(diphenylphosphate) (RBDPP) and bisphenol A bis(diphenylphosphate) (BPA-BDPP) as alternative HFFRs to be used in TV/flat screen housings.

2.4. Nitrogen containing FR

Pure melamine, melamine derivatives and homologues are commonly used as the base element of nitrogen containing FRs. This type of FRs is capable of generating cross-linked structures at high temperature which supports char formation. Consequently, the released inert nitrogen dilutes combustible gases and inhibits the chain reaction (Aschberger et al., 2017).

2.5. Inorganic FR

Hydroxides of aluminum and magnesium with aluminum trihydroxide (ATH) are the most common types of inorganic FRs. ATHs are frequently used in polymer and polymer composites due to their low cost, good properties and nontoxic smoke (Pereira and Martins, 2014). The water release and cooling down the flame zone is the base of this type of FRs and they are normally resistant up to 200 °C. However, Magnesium hydroxide (MDH) can resist up to 300 °C and, therefore, can be used when a higher temperature is required for the process (Aschberger et al., 2017).

3. LCA of FRs

As discussed in the previous sections, there are serious health and environmental concerns about the use of FRs. LCA is an environmental management technique for evaluation of the environmental (including toxicity) impacts of a product over its entire life cycle, “from cradle to the grave” (International Organization for Standardization, 2006). In this section, a systematic review on the LCA studies on FRs will be expounded. A systematic review usually aims at summarizing, evaluating and criticizing the available information and existing data and giving an expert opinion in a transparent and unbiased way. There is a growing interest in conducting systematic reviews in different disciplines, including LCA (Zumsteg et al., 2012).

Noting the complications in performing an LCA such as required time and resources, some authors have tried simplified methods for evaluating the environmental aspects associated with FRs. In 1997, Hardy et al. (2003) used a semi-quantitative environmental index to assess, i.e. rank, eleven generic FR cotton and polyester textiles. Broeren et al. (2016) have also utilized a so-called screening LCA for the purpose of material selection. This is conducted by means of a few selected impact categories, relying on the available data and skipping some phases which can be equal for different alternatives in comparative studies. Broeren et al. (2016) have utilized screening LCA for material selection and only considered non-renewable energy use, GHG emissions and agricultural land use impact categories. They believe that using screening-LCA is a middle ground between ignoring additives completely, (which happens often due to the difficulties in obtaining data), and performing a complete LCA, (which is resource demanding and time consuming). They suggest that in spite of all uncertainties, additives have a significant role in the LCA of plastics and by using techniques such as screening LCA, the evaluation of their impacts can be enhanced. In addition to full and simplified LCA models, ecotoxicity models such as Potentially Affected Fraction (PAF) and the Most Sensitive Species (MSS) are used to evaluate the health and environmental concerns of FRs (Kobetičová; Černý, 2018).

The occurrence of fire is normally omitted in LCA studies (Jonkers et al., 2016). The SP Swedish National Testing and Research Institute and IVL Swedish Environmental Research Institute developed a so-called fire-LCA model in which the environmental impacts of fire were integrated in the LCA study (Andersson et al., 2004). As Fig. 1 illustrates, the fire-LCA model is principally equivalent to a typical LCA approach with the inclusion of the emissions due to the fire and the dispersion of the emitted pollutants in the atmosphere. As a cradle to grave LCA, it takes into account the emissions associated with all life cycle phases. The FR production and the fire performance should be included by using modules to describe the fire behavior for different types of fires and quantification of the amount of used materials based on fire

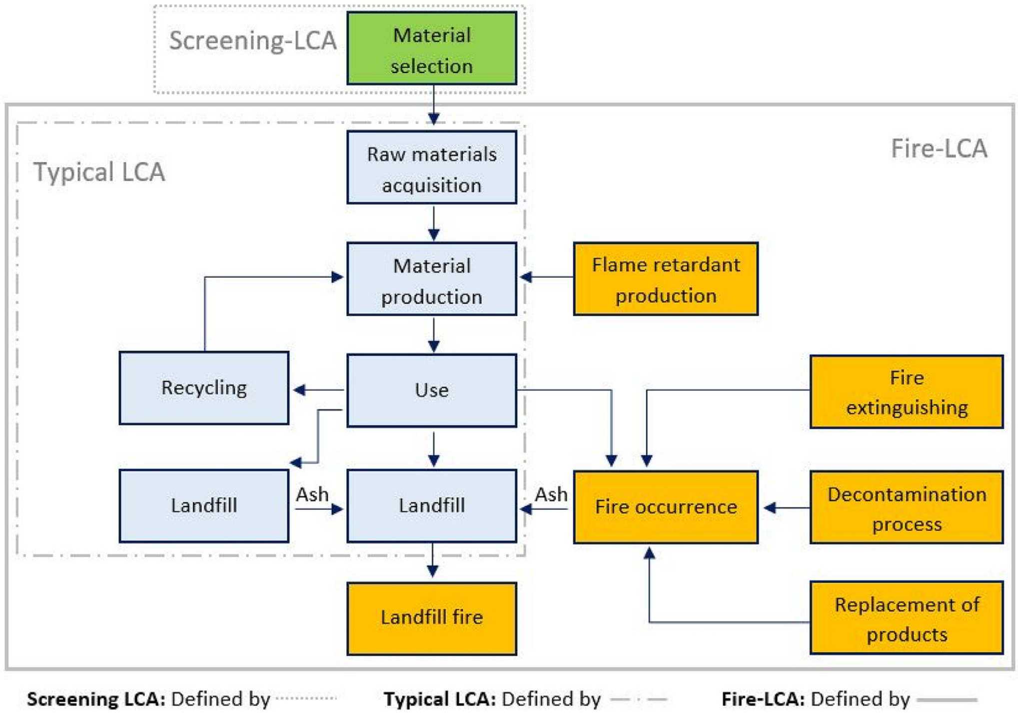

Fig. 1. Comparison of Screening LCA, Fire-LCA and Typical LCA, adapted from (Andersson et al., 2004).

statistics. Furthermore, the impact of the consequences of fire, such as replacing new materials, fire extinguishing and decontamination processes should also be included in such models (Andersson et al., 2004)(Chettouh et al., 2014). The fire-LCA has been used by other authors such as (Simonson et al., 2002), Chivas et al. (2009) and Chettouh et al. (2014) for evaluating the environmental impacts of FRs. Andersson et al. (2004) provide a comprehensive guideline for performing fire-LCA studies discussing modules to describe different fire behaviors based on fire statistics. They also highlight that performing fire-LCA allows us to evaluate/compare the environmental impacts of FRs more effectively as the fire occurrence is considered as one possible end of life scenario. Some scholars such as Jonkers et al. (2016) also considered the occurrence of fire, but did not mention the term fire-LCA.

In 2000, the National research council of the US published a comprehensive work (National Research Council, 2000) on toxicological risks (potential cancer or non-cancer effects) of 16 selected FRs used in residential furniture. This work was performed by analyzing human and animal data on neurotoxicity, immunotoxicity, reproductive and developmental toxicity, organ toxicity, dermal and pulmonary toxicity, carcinogenicity, and other local and systemic effects (Hirschler, 2013). ENFIRO (ENFIRO, 2016) was a European Commission-funded project which was aimed at substituting options for specific BFRs. This project was named “Life Cycle Assessment of Environment-Compatible Flame Retardants (Prototypical case study)” and resulted in various publications such as (Brandsma et al., 2013a), (Brandsma et al., 2013b), (Hendriks et al., 2010), (Hendriks et al., 2012a), (Hendriks et al., 2012b), (Hendriks et al., 2014), (Hendriks et al., 2015), (Hendriks and Westerink, 2015), (Jonkers et al., 2016), (Waaijers et al., 2013a), (Waaijers et al., 2013b) and (Waaijers et al., 2013c). However, only the study by Jonkers et al. (2016) expounds the LCA of FRs and the others mostly explore the chemical hazards, potential exposure and alternative substitutes for HFRs and HFFRs.

Considering the wide use of FRs in electronics, a number of studies have begun to examine the LCA of FRs in this industry. For instance, Simonson et al. (2002) compared the environmental impacts of two types of TV sets with and without FR by performing a fire-LCA model. This comparative study considered the whole life cycle, but mainly focused on different end of life scenarios. The results of their study show not much difference in terms of CO_2 emissions and energy use for both scenarios, but PAH emissions are remarkably higher for the product containing FR. For the 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) and 2,3,7,8-tetrabromodibenzo-p-dioxin (TBDD) equivalents, the product with FR has lower emissions for the current scenario (97% landfill), but notably higher emissions for the future scenario (10% landfill). The present and the future scenario considered incineration for 1% of end of life scenarios and do not include extinguishment. Deng et al. (2016) also compared the environmental impacts of a HFFR, i.e. melamine polyphosphate (MPP), with a HFR, i.e. TBBPA, for a printed circuit board (PCB) substrate. Their results show that use of MPP contributes significantly to reduction in terrestrial ecotoxicity and marine ecotoxicity categories. By pointing out the mandatory use of FRs for PCBs while currently 80% of them are based on TBBPA, they highlight the potential benefits of using HFFRs in electronic industry. Similarly, Jonkers et al. (2016) did a comparative LCA study on the use of BFRs and HFFRs for laptop computers. Two different scenarios of BFR and HFFR were considered and each scenario consists of different FRs from that FR family for different parts of laptop computers, i.e. cables, casing, etc. Their results highlighted notable environmental benefits in replacing BFRs by HFFRs. They also discuss that on LCA studies of electronics, the environmental impact of the used metals by far overshadows the plastic part. Furthermore, the used plastic is mostly considered to be a polymer ignoring the additive such as FRs. Broeren et al. (2016) have also utilized a screening LCA method to evaluate the use of FR for a side panel of a large multi-functional printer.

As the screening LCA was used as a part of product development process and material selection in this study, the compared alternatives (FR panel and a non-FR panel) had different compositions (share of additives, biobased carbon content, weight, etc.).

Textile products have been one of the main applications of FRs for several years (Yasin et al., 2016) and FRs account for 3 to 30 weight percent (wt%) of the textile products (Nazaré, 2009). Dahllöf (2004) has performed a comprehensive comparative study for three types of fiber, i.e. conventional cotton, wool with 15% polyamide and a FR polyester. Her results show that cultivation of the cotton and subsequent processes such as dyeing are associated with high environmental impacts so that the FR polyester has less overall environmental impacts in comparison. In the case of the wool/polyamide alternative, the sheep farming, ring spinning, nylon fiber production and wool scouring are found to have high environmental impacts. Yasin et al. (2018) have also performed a LCA study on the FR cotton curtain by focusing on different end of life scenarios. They also explain that the cotton cultivation requires a notable amount of fertilizers containing phosphorus and nitrogen chemicals. Table 2 summarizes and compares the studied literature on the LCA of FRs.

The Functional unit (FU) is especially important in evaluating alternative FRs. It can be seen that in the reviewed studies, the FU is normally the product itself, e.g. curtain (Yasin et al., 2018), laptop computer (Jonkers et al., 2016), TV set (Simonson et al., 2002), sofa fabric (Dahllöf, 2004) or a PCB substrate (Deng et al., 2016). Defining a correct FU is critical in performing a LCA study. It should be noted that comparing the results of different literature is only valid while having the same FU and toward that, the functionality of the product must be taken into consideration. Broeren et al. (2016) point out that for innovative materials, such as biobased plastics, the LCA is mostly on a kg basis.

Lack of data is the main challenge in any LCA study and especially is the case for FRs. All of the studied LCA literature used different sources of data, either primary or secondary, such as literature, industrial partners, experiments as well as existing databases for completing the puzzle of environmental impacts of FRs. Admitting the existence of uncertainties on the obtained data, sensitivity analyses have been mostly performed by using techniques such as high and low end values (Broeren et al., 2016) or considering possible future scenarios (Simonson et al., 2002). All of the studies have used data from different countries. This can be explained by the reasons such as having different countries for different phases (e.g. production in one country and use in another) (Yasin et al., 2018) or having data available only for average European or OECD countries for specific product/processes (Jonkers et al., 2016).

Different impact assessment methods such as EPS 2000 (Dahllöf, 2004), ReCiPe (Deng et al., 2016) and (Jonkers et al., 2016) and USES-LCA2 (Jonkers et al., 2016) have been used in the LCA studies to evaluate the environmental impacts of FRs. The variation in the life cycle impact assessment (LCIA) methods may result in discrepancy in the results. Some scholars such as Broeren et al. (2016) developed their own methodology for this purpose. Some studies have referred to the use of software tools such as SimaPro (Deng et al., 2016) and LCAiT (Simonson et al., 2002), but most of them have not mentioned any references.

For better understanding of the environmental aspects of FRs, all of the studied literature have performed comparative studies. Sometimes the use of FRs are compared with conventional alternatives that do not use FRs. For instance, Simonson et al. (2002) have compared two scenarios for a TV with and without FR, Broeren et al. (2016) have performed a comparative study for a side panel of a large multi-functional printer with and without FR and Dahllöf (2004) have compared a FR polyester with two fiber types without FR. Some other studies have compared different types of FRs. For instance, Deng et al. (2016) have compared the use of HFFR with HFR and (Jonkers et al. (2016) have compared the use of HFFR with BFR. Some scholars such as Yasin et al. (2018) have focused on different end of life scenarios in their comparative study.

One of the most important issues in LCA studies on FRs is the considered phases of the life cycle. This concern is not limited to the question whether all the phases from the 'cradle to grave' are taken into consideration, but also to the contribution of different phases. Broeren et al. (2016) highlight the importance of eco-design and suggest that environmental aspects must be taken into consideration at the product design stage. Nonetheless, Yasin et al. (2018) discusses the example of textile products for which the use phase is the phase with the most environmental impacts due to washing and tumble drying. Therefore, the environmental concerns should not be limited to the design and chemistry of the product and a significant reduction can be achieved by changing the user behavior such as less frequent washing, using lower temperature for washing and avoiding tumble drying. Yasin et al. (2018) state

Table 2
LCA studies on FRs.

Reference	Simonson et al. (2002)	Dahllöf (2004)	Deng et al. (2016)	Broeren et al. (2016)	Jonkers et al. (2016)	Yasin et al. (2018)
LCA method	Fire-LCA	LCA	LCA	Screening LCA	LCA	LCA
Product	TV set	Sofa fabric	Printed circuit board substrate	Side panel of a large multi-functional printer	Laptop computer	FR cotton curtain
Comparison	With FR vs without FR	Cotton vs Wool with 15% polyamide vs FR polyester	HFR vs HFFR	With FR vs without FR	BFRs vs HFFRs (12 alternatives)	Eco-path (removing FR) vs landfill vs incineration
Functional unit	1 TV set	Surface covering a 3-seat sofa with 10 years life time	1 m^2 of PCB substrate with a thickness of 1.6 mm	1 kg of panel	1 Laptop computer	1 kg of FR cotton curtain
System boundaries	Cradle to grave	Cradle to grave	Cradle to grave	Cradle to grave	Cradle to grave	Cradle to grave
Database	Industry, experiments, literature	Industry, literature	Ecoinvent, literature	Ecoinvent, literature, reports	Project partners, Ecoinvent	EIME software database, experiments, literature
Software	–	LCAiT	–	SimaPro	–	–
Location	Sweden, OECD countries	Various	Europe, US	The region of production, Europe	Europe	Turkey, Italy
Impact assessment method	–	EPS 2000	ReCiPe	Own methodology	USES-LCA2, ReCiPe	–
Sensitivity analysis	✓	✓	✓	✓	✓	–

that almost all of the LCA studies on textiles have focused on reduction of the environmental impacts at the use phase, as this phase accounts for 80% of the CO_2 emissions. Jonkers et al. (2016) also highlight that in the LCA community, the use phase receives the main attention when it comes to “any product with a plug”. In the comparison of HFRs and BFRs for a laptop computer, Jonkers et al. (2016) concluded that BFRs have higher impact at the use phase. This can be explained by their larger impact at human toxicity and freshwater ecotoxicity. However, the terrestrial and marine ecotoxicity impacts are higher for the HFFRs. Some authors such as Yasin et al. (2016), Aschberger et al. (2017) and Hahladakis et al. (2018) highlight that during the end of life phase, FRs can affect the human health in numerous ways. Landfill disposal can be associated with ecotoxicity and may lead to exposure to humans (Yasin et al., 2016). Moreover, there is a limitation in landfill space and landfill gas and leachate are hazardous (Yasin et al., 2018). Nazaré (2009) states that as textiles are not categorized as hazardous waste, most of the textiles are disposed as municipal waste neglecting the potential impacts of the impregnated FRs. The incineration can also cause toxic gas emissions (Yasin et al., 2016). Hahladakis et al. (2018) point out that incineration of BFRs may emit hazardous substances. Even recycling is not hazardous free and BFRs containing POPs, PFRs and phthalates are reported to be found in children toys manufactured from recycled plastics. In spite of all the global efforts to enhance the end of life of FRs, there is still space for significant improvements in both developing and industrial countries (Yasin et al., 2018). Jonkers et al. (2016) show that the BFRs have higher impact than HFFRs at the end of life cycle phase for a FRs containing laptop. However, they discuss that the total environmental impact of the BFR containing product over its life cycle is only slightly higher than the one with HFFR.

The importance of different phases of the life cycle of FRs in LCA studies is closely interconnected with their environmental exposure. The exposure to FRs occurs during the different stages of their life cycle. People can be exposed at the manufacturing, use and end of life (either landfill, incineration or recycling) phases of the life cycle of FRs (Aschberger et al., 2017). Nazaré (2009) lists various factors such as the volume of FR, its chemical and physical properties, biotic and abiotic degradation, the amount of FR released at different life cycle phases, emission control measures, persistence and bioaccumulation as the influential factors in the environmental exposure of FRs. Yasin et al. (2018) highlight that it is extremely challenging to measure the release of the FR substance in a textile product at each phase of its lifespan. In this case, the fiber production and subsequent processes such as dyeing, printing and finishing are associated with the use of chemicals and the possibility of release to the environment (Nazaré, 2009). Similarly, during the use phase some FRs may transmit from textile products to the environment and human beings (Yasin et al., 2016). The release may happen due to various reasons such as volatilization, laundry and leaching because of rain, solvents, oil, etc. The direct exposure of FRs to human beings may happen through dermal or oral exposure as well as inhalation (National Research Council, 2000). In the case of dermal exposure, a contact with sweat, blood, urine, skin cream or body lotion can cause leaching of the FR. Inhalation occurs upon the release of toxic emissions from the FRs in fire accidents. Even low concentrations of HFRs burning at low temperatures can lead to acute (immediate) and chronic (long-term) toxicity. Oral exposure can happen either directly (oral contact with FR textile product) or indirectly (FR presence in the water, food or air) (National Research Council, 2000). The health impact of FRs in this sector varies from skin irritation to neurotoxicity and carcinogenicity depending on the type and concentration of FR (Hendriks and Westerink, 2015). Yasin et al. (2018) notably highlights that a life cycle phase having a small share of the overall cradle to grave emissions could still be the most vital one in terms of exposure to human and other organisms. As an example, the use phase of polymeric FRs has low impacts in comparison with other phases, but is associated with human toxicity and freshwater toxicity emissions. Similarly, the end of life cycle has normally lower emissions in comparison with manufacturing and use phase, but might have serious negative impacts, such as ecotoxicity and human exposure (Yasin et al., 2018).

4. Discussion and conclusions

This systematic review on LCA of FRs summarizes the state of the art of the scientific literature on LCA of FRs. In spite of all discussed environmental and health concerns regarding the use of FRs and numerous LCA studies on plastics, there is a huge gap in the literature on the LCA of additives and particularly FRs. Reviewing the related literature showed that those studies that aimed at evaluating the FRs are mainly focused on health concerns. This includes exploring the human exposure to FRs through different means during the different stages of their life cycle as well as consequent health impacts on vulnerable organs. On the other hand, the LCA studies on polymers usually ignore additives such as FRs. This lack of attention can be explained by undervaluing the magnitude of the environmental impacts of FRs as well as challenges in obtaining data. LCA studies of FRs can highlight new opportunities to improve the current solutions and develop new suitable alternatives toward a circular economy.

While comparative studies are the common practice and are shown to be suitable options to evaluate the environmental impacts of FRs, defining the alternatives is crucially important in the first place. In addition to different types of FRs, comparing a material (polymer) with and without using a specific FR is recommended. For the alternative of not using FR, fire occurrence as one of the end of life scenarios should be taken into consideration (fire-LCA). Although considering this scenario is challenging due to the difficulties in finding specific data based on statistics, it can bring valuable insights into LCA of FRs and answer whether the use of FR is actually preferable. Ultimately, different amounts of the relevant FR can be examined to find the optimum amount of FR with the minimum environmental and health impacts that can prevent fire occurrence. Furthermore, it should be noted that the definition of the FU is commonly and incorrectly based on the product rather than the function in the literature. When it comes to data collection, the confidentiality of the composition and production routes of FRs are important barriers in getting insights into their environmental impacts. Besides, assumptions are often inescapable in LCA studies of FRs due to uncertainty of specific data. This can especially be the case for the textile industry where the use phase is related with many different users with different habits of washing and tumble drying for instance. Moreover, specific products and processes may have different countries associated with different phases (e.g. production in one country and use in another) and the data for average European or OECD countries are often used. Therefore, performing a sensitivity analysis on these aspects is strongly suggested for LCA studies of FRs.

The inclusion of different life cycle phases of FRs is typically decided based on their contributions. However, the target for the contribution seems to be more human exposure rather than environmental impacts. This review pointed out that the contribution to the environmental impacts highly depends on the sector, the type and use of the product as well as the type of FR. Furthermore, the contribution to the exposure depends on numerous influential factors, such as the volume of FR, its chemical and physical properties in addition to the type of exposure (direct or indirect). The consequent impacts on the human health can also vary between minor and severe as well as immediate and long-term. While environmental and health concerns are interconnected, a phase with lower emissions, such as end of life, may be associated with negative impacts like ecotoxicity and human exposure. These outcomes highlight the importance of taking into account all of the life cycle phases while preserving the interconnection between three domains of environmental impacts, human exposure and health concerns. This also highlights the role of LCA as a tool for environmental impact assessment that covers both health and environmental aspects.

In evaluating the environmental and health concerns of FRs, the type of FR showed to be absolutely influential. Consequently, recognizing different types of FRs and their environmental and health impacts is inevitable in developing novel harmless ones. In spite of the implemented policies restricting the use of HFRs, they found to be still existing and may be growing in the environment. In the case of BFRs, this can be explained by their preferable performance and lower price as well as a “time lag” between production, use and end of life phases. In the case of CFRs, the main producers and consumers, i.e. China, Russia and India, do not have effective preventive policies yet. The current solution of stopping the production of hazardous FRs is not implemented in all countries and as a sole measure is not enough. Recycling of such materials has also proven to be unsafe as it may result in human exposure again. Moreover, the separation of specific FRs is challenging and their recycling is tolerated under controlled circumstances. These evidences imply that we should reconsider technical and biological cycles of FRs at the first place so that they will neither be recycled nor discarded as waste. Circular economy principally suggests to replace the traditional linear model of “take, make and dispose” by a circular model of restorative and regenerative design. This involves designing out waste and pollution, keeping product and materials in use by strategies such as reuse, repair and remanufacture for the technical cycle and regenerating natural systems. In the case of FRs, a transition to a circular economy can be achieved by finding a balance between developing novel harmless FRs for both environment and human health and emerging innovative waste management techniques. This requires a close collaboration of all stakeholders, i.e. governments, policymakers, industries and users.

Considering the serious environmental concerns about HFRs and the discussed policies prohibiting their use, further research could usefully explore the LCA of recently developed HFFRs. This should be performed not only by comparing these innovative FRs with formerly developed FRs, but also by evaluating their environmental impacts throughout the life cycle. Considering the importance of bio-based materials in the transition towards a circular economy, future studies could explore the LCA of bio-based FRs such as tannic acid, phytic acid and lignin. There is also a growing interest in using nanoparticles such as carbon nanotubes (CNTs) as well as nanoclays in the FRs which can improve the fire performance especially in the case of HFFRs. The current available databases for LCA do not contain such data and the results of these studies can be extremely helpful. In addition to the data, LCA tools could usefully provide the possibility of performing fire-LCAs. Noting the undervalued role of additives in LCA studies of plastics and presence of typically more than one additive in plastics, future studies could also assess the LCA of FRs in combination with other additives such as plasticizers, stabilizers, colorants and nucleating agents.

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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