Do flushed biodegradable wet wipes really degrade?

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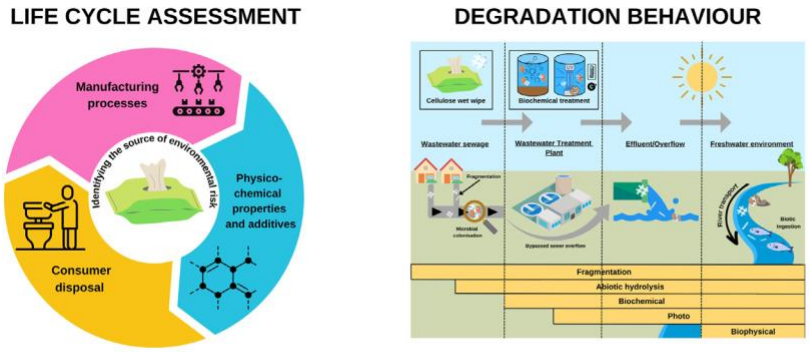
HIGHLIGHTS

- Cellulose wet wipes are an underestimated potential source of envi-ronmental pollution.
- Wipes labelled "biodegradable" shed many microfibres that do not degrade.
- Life-cycle assessments highlight where and how this degradation is limited.
- Urgent investigation into degradation pathways is required to guide decision-making.

GRAPHICAL ABSTRACT



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ABSTRACT

Consumer wet wipes sold as biodegradable and flushable have tripled in market size in the last decade (>$3 billion in 2022), spurred by concerns over their potential harmful impact. Whilst predominantly composed of cellulosic fibres such as cotton, rayon, or wood pulp, these have been found to persist in sewers and in the environment in near equal abundance to their 'synthetic' counterparts. This questions whether flushed biodegradable wet wipes really de-grade.

Working from first principles, we therefore explore the physicochemical composition, environmental interactions, and degradation processes throughout the entire life cycle of cellulosic wet wipe fibres, from production to environmental fate, to understand their degradation behaviour in wastewater and freshwater systems.

The results highlight that >50 % of biodegradable and flushable wipes are commonly manufactured with both biological biodegradable cellulose-based fibres and low-degradable synthetic fibres, and that they contain various property-enhancing chemical additives that can limit degradation. Whilst cellulose fibres in wet wipes are highly prone to physical fragmentation, their molecular degradation is difficult within the environment. This is due to the physicochemical manufacturing properties of wet wipes and the usually inadequate ambient conditions for its break-down, creating persistent and possibly biologically harmful microfibres.

We conclude that currently, most flushed biodegradable wet wipes do not really degrade, and that more empirical in-vestigations are needed on their in-situ degradation behaviour and the environmental and manufacturing processes that may influence this breakdown. In doing so, full life cycle approaches to wet wipes should be adopted, considering their manufacturing properties, consumer disposal behaviour, and environmental implications.

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Contents

1. Introduction

Multi-purpose, single-use wet wipes have surged in consumer demand worldwide, namely since the recent pandemic. Their inappropriate domes-tic disposal presents a significant route of contamination to aquatic ecosystems worldwide (Shruti et al., 2021; Zhang et al., 2021b) and can be easily spotted along rivers (McCoy et al., 2020). As an example, in the UK, their subsequent occurrence on beaches has increased by 94 % in 2016, with a 400 % increase over the last decade (Pantoja-Munoz et al., 2018), whilst >23,000 wet wipes were recovered from a single area of the Thames fore-shore (UK), with an average density of 201 wet wipes/m2 (Thames21, 2019).

Domestic wet wipes are mainly composed of non-degradable synthetic polymers such as polyethylene terephthalate (PET) and polypropylene (PP) (Pantoja-Munoz et al., 2018; Ó Briain et al., 2020). Over time, these wipes will fragment into smaller but still molecularly intact microfibres in the environment. At this point, they can be ingested by aquatic biota (Hu et al., 2022), with potential physical and ecotoxicological risks to their health (Stone et al., 2020). Ingested microplastic particles have also been found to travel through aquatic food webs (Windsor et al., 2019b; D'Souza et al., 2020), which may, ultimately, transmit these health risks to humans (Zhang et al., 2020; Jiang et al., 2021; Liao and Chen, 2021).

Incorrect wet wipe disposal also has significant social and economic im-pacts. Wet wipe pollution has increasingly gained media attention, often visualised by their presence in “fatbergs” – congealed masses of wet wipes and other wastewater substances that persist within the sewer network (Alda-Vidal et al., 2020). Roughly 50 % of blockages within UK sewage systems are caused by wipes and similar hygiene products (Pantoja-Munoz et al., 2018; Mitchell, 2019). These blockages can promote the surface growth of various microbial communities, thus creating the need for additional biological treatment processes within sewers (Lee et al., 2021). Furthermore, some of these microrganisms can produce toxic or corrosive by-product chemicals that cause damage to the surrounding wastewater infrastructure (Durukan and Karadagli, 2019). The operational and financial costs for wastewater companies can be high (Mitchell, 2019; Ó Briain et al., 2020), reaching annual damages up to $15 million in Australia, and £23 million in London (UK) alone (Fam et al., 2017). Combined sewage overflows (CSOs), release points for sewage and rainwater run-off from roads and urban areas during heavy rainfall (Scurlock, 2022), can act as additional pathways for wipes into freshwater ecosystems so that downstream, wipes are becoming a frequent source of aesthetic pollution (Horton et al., 2017; Besley and Cassidy, 2022).

Alternative wet wipes, marketed as biodegradable and flushable have been developed to address these issues (Zhang, 2010). For a wipe to be considered as biodegradable, it must be produced from natural polymers, such as cellulose, in order to decompose into harmless biomass (IWSFG, 2020). Alongside biodegradability, cellulose offers the advantage of favourable manufacturing properties (i.e. lightweight, high chemical adhesion, bio-compatibility, and natural abundance) and low cost (Ibrahim et al., 2018; Yun et al., 2020a; Jiang et al., 2021; Polman et al., 2021). Cellulose has, therefore, become a key polymer for commercially available biodegradable wet wipes.

However, there is limited knowledge on the abundance and degradation behaviour of these alternative wet wipes and their fibres, particularly once they enter the environment (Liao and Chen, 2021). There is growing evidence that many products labelled as biodegradable, such as wet wipes, do not, or do not fully, degrade under natural environmental conditions (Napper and Thompson, 2016; Manfra et al., 2021). This may be be-cause many wipes branded as biodegradable still tend to include plastic (Ó Briain et al., 2020). However, there is evidence that cellulose-based

fibres remain undegraded within wastewater effluents and freshwaters (Dris et al., 2018; Lares et al., 2018; Ó Briain et al., 2020; Zambrano et al., 2020b), marine environments (Adams et al., 2021) and municipal sewage sludges (Habib et al., 1998). This environmental persistence may also derive from the presence of additives - chemical substances commonly added to polymers during the manufacturing process to improve product functionality, degradation resistance, and performance (Lambert and Wagner, 2017; Hahladakis et al., 2018), but this has not been thoroughly analysed to date. Whilst outside the scope of this paper, understanding the extent of the environmental risk posed by cellulosic wet wipes and their fibres also requires both their in-situ identification and quantification
– analyses that are only just developing for plastics.

This paper explores the current knowledge on wet wipes, and questions whether biodegradable wet wipes, and particularly cellulose-based forms, are environmentally better alternatives to traditionally synthetic wipes. Working from first principles, we introduce wet wipes within the context of a life cycle assessment and explore their different manufacturing pro-cesses, physicochemical properties, consumer disposal, and fate to evaluate their potential environmental risks (Section 2). We then highlight the extent of likely biodegradable wet wipe breakdown within the aquatic environment, focusing on wastewater and freshwaters as their disposal routes. In doing so, we explore the environmental degradation mechanisms available (Section 3), the likely influencing environmental variables (Section 4), and the interplay between these that may influence break-down. Finally, we discuss whether flushed biodegradable wet wipes really degrade and make recommendations for future research (Section 5).

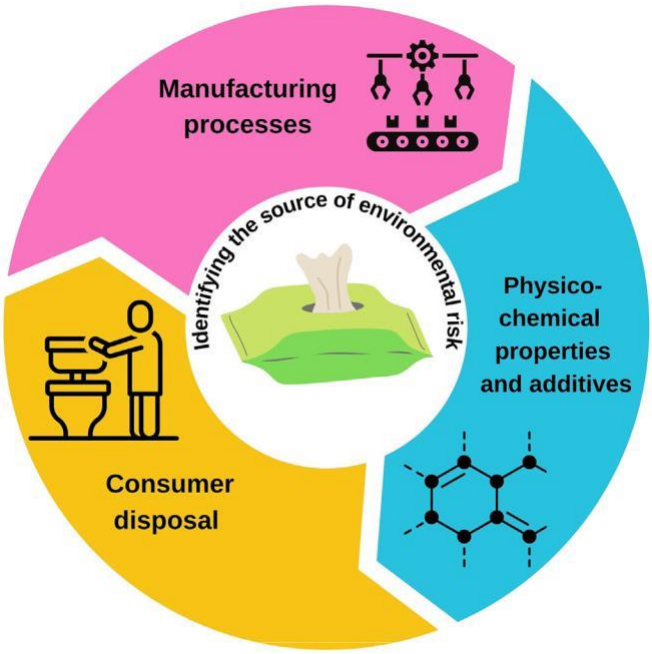

Fig. 1. Stages of the cellulosic wet wipe life cycle to investigate as sources of environmental risk.

2. Wet wipes in their life cycle

Wet wipes are versatile nonwoven textiles that serve a variety of purposes, from disinfection to personal hygiene. They are composed of differ-ent polymer fibres and chemical additives (Russell, 2007; Durukan and Karadagli, 2019), that require distinct manufacturing processes and present unique disposal challenges. The breakdown of wet wipes in the environ-ment is largely determined by the choice of manufacturing process. To eval-uate the environmental impact of cellulose-based wet wipes, we adopt here a life-cycle approach (Section 2.1) that examines the effects of raw mate-rials and typical manufacturing processes (Section 2.2), as well as the phys-icochemical properties and additives (Section 2.3) that influence their degradability and fate. Additionally, we discuss how different disposal methods can affect the breakdown behaviours of wet wipes (Section 2.4).

2.1. Life cycle assessments of wet wipes

Life cycle assessments (LCA) can be valuable tools for evaluating the environmental impact of wet wipes throughout their entire life cycle, includ-ing manufacturing, usage, disposal, and fate (Fig. 1). A recent LCA of synthetic and bio-based wet wipes in China found that whilst the overall environmental impact of wet wipe manufacturing is relatively low (0.02 %– 7.17 % of the total manufacturing emissions produced across all stages) (Zhang et al., 2021b), the production of wood pulp fibres used in their production can lead to significant eutrophication due to chemical fertilizer and pesticide application. Besides this however, there are no LCAs on biodegradable wet wipes.

2.2. Raw materials and manufacturing processes

Wet wipes can be made from a variety of polymer fibres including: synthetic petrochemicals (e.g. PET, PP and polyethylene [PE]), natural sources such as cellulose (e.g. cotton and wood pulp) or chemically regenerated cellulose (e.g. viscose and lyocell) (Dris et al., 2018; Pantoja-Munoz et al., 2018; Orr and Karadagli, 2020).

Synthetic plastic fibres are petroleum-based polymers with added chemical additives that resist degradation but can leach harmful by-products and pollutants into aquatic environments (Webb et al., 2013; Miller et al., 2017; Stone et al., 2020). Most wet wipes currently available on the market contain at least one type of synthetic fibre (Ó Briain et al., 2020; Lee et al., 2021), as their ability to maintain shape and increase ten-sile strength when wet makes them more durable during consumer usage (Pantoja-Munoz et al., 2018; Lee et al., 2021).

Natural cellulosic plant fibres are bio-polymers derived from renewable plant feedstock sources (Shaghaleh et al., 2018; Liu et al., 2021). Among these, cotton is the most commonly used for manufacturing wet wipes, accounting for almost 30 % of all 113 million tonnes of polymer fibres produced in 2021 (Carr, 2017; Barrows et al., 2018; Textile Exchange, 2022). Cellulosic fibres make up over 50 % of all raw materials used in wet wipes and are added to increase biodegradability, softness, and water ab-sorbency (Zhang et al., 2018; Durukan and Karadagli, 2019; Orr and Karadagli, 2020; Harter et al., 2021). Regenerated fibres, including viscose, rayon, lyocell, are also derived from cellulose biopolymers, but have been physically and chemically modified during manufacturing (Gago et al., 2018). However, the inconsistent labelling of regenerated fibres as either natural or synthetic in environmental pollution studies makes it challenging to accurately estimate their environmental persistence and impact.

The manufacturing processes involved in producing wet wipes from selected raw materials include web formation, bonding, and finishing pro-cesses. Web formation involves arranging fibres of different sources into layers (EDANA, 2022), which can be achieved through dry-laid (i.e. web forming in a dry state), wet-laid (i.e. web forming in a liquid substance) or spunmelt methods (i.e. wet forming by heat) (Mitchell, 2019; Atasagun and Bhat, 2020). The fibrous web networks are then mechanically or chem-ically bonded to increase their strength (Russell, 2007; Pantoja-Munoz et al., 2018; Durukan and Karadagli, 2019; EDANA, 2022). Finally, different chemical additives are added during the finishing process to improve a specific product's functionality (EDANA, 2022). These additives can include antibacterial and antistatic properties, dyeing, flame retardancy, anti-shrink agents, and surface softening (Varadarajan and Venkatachalam, 2016; Dris et al., 2018; Stone et al., 2020; Harter et al., 2021; Jahandideh et al., 2021).

2.3. The specific properties of cellulose wet wipes

We focus in this section on the specific chemical (Section 2.3.1) and physical (Section 2.3.2) properties of cellulose fibres in wet wipes, as well

as applied chemical additives (Section 2.3.3) that may affect their degradation and fate.

2.3.1. Key chemical properties of cellulose

Cellulose is a bio-renewable polysaccharide that makes up most plant tissue (Yuan and Cheng, 2015; Adams et al., 2021). It consists of β-D-glucose repeating units covalently linked by acetal functions. Each mono-meric unit of cellulose has three highly reactive hydroxyl groups, giving the polymer its hydrophilic and biodegradable properties (Klemm et al., 2005; Shaghaleh et al., 2018).

Cellulose fibres are structured hierarchically as single chains, microfibrils, and macrofibrils (Fig. 2) (Belgacem and Gandini, 2011). Strong intra and intermolecular bonds formed by hydroxyl groups and oxygen atoms create highly-ordered crystalline regions in the microfibrils (Yuan and Cheng, 2015; Ghasemi et al., 2017). The degree of crystallinity deter-mines the overall strength, durability and biocompatibility of the polymer (Shaghaleh et al., 2018). For example, cotton has a higher degree of crystal-linity (~70 %) than other cellulosic fibres (Wood, 1988), making the poly-mer more resistant to degradation. In contrast, amorphous regions are loosely structured, susceptible to degradation processes, and tend to in-clude additional biopolymers such as hemicelluloses and lignin (Ghasemi et al., 2017). Therefore, the de-crystallisation process plays an essential role in the extent of degradation in cellulose (Ghasemi et al., 2018).

Cellulose has a strong affinity for itself and hydroxyl-containing materials such as water (Khazraji and Robert, 2013). Cellulose exhibits amphiphilic properties, due to its extensive intra and inter molecular hydrogen bonds and simultaneous hydrophobic molecular interactions, which can significantly impact its solubility in water and most organic solvents (Väisänen et al., 2021b).

The degree of polymerisation (DP), representing the average number of monomer repeat units in a polymer, varies depending on the origin and processing of cellulose (Klemm et al., 2005; Yuan and Cheng, 2015). For instance, filter paper consists of purified cellulose with a DP of 2900 (Malešič et al., 2005), whilst birch and dissolving pulps have DPs of 1800 and 3600, respectively (Sirviö and Lakovaara, 2021). However, the typically high DP of cellulose is a major obstacle to its chemical breakdown (Väisänen et al., 2021b).

Cellulosic fibres from different sources also vary in molecular weight (MW) which can affect the rate and extent of polymer degradation. You et al. (2021) reported the weight-average MWs (measured in kDa) of vari-ous cellulosic fibres, with raw cotton having the highest MW (444 kDa), followed by dissolving wood pulp (208 kDa), Tencel (lyocell) (127 kDa), and viscose (127 kDa).

2.3.2. Key physical properties of cellulose

Wet wipe manufacturers need raw materials with modifiable physico-chemical properties to meet a variety of consumer and industrial needs. These properties may include high strength, high absorbency for chemical additives, or biodegradability for fast disintegration in the natural environ-ment (Mitchell, 2019). Unmodified cellulose alone has poor adsorption ca-pacity and mechanical properties, but it has a large number of hydroxyl polar groups that can be modified to suit various applications (Habibi, 2014; Jiang et al., 2021).

The morphology of cellulosic fibres is important for their application and biological degradation (Klemm et al., 2005). Cellulosic fibres in wet wipes are typically short in length (0.3-10 mm) (Zhang et al., 2018; Mitchell, 2019). Regenerated fibres like viscose are longer than natural wood pulp fibres (Zhang et al., 2018). The length of fibres affects the breakdown of wipes. For instance, shorter fibres improve physical dispersibility and chemical degradation processes, whilst longer fibres increase wet tensile strength and resistance to breakdown (Tipper, 2016).

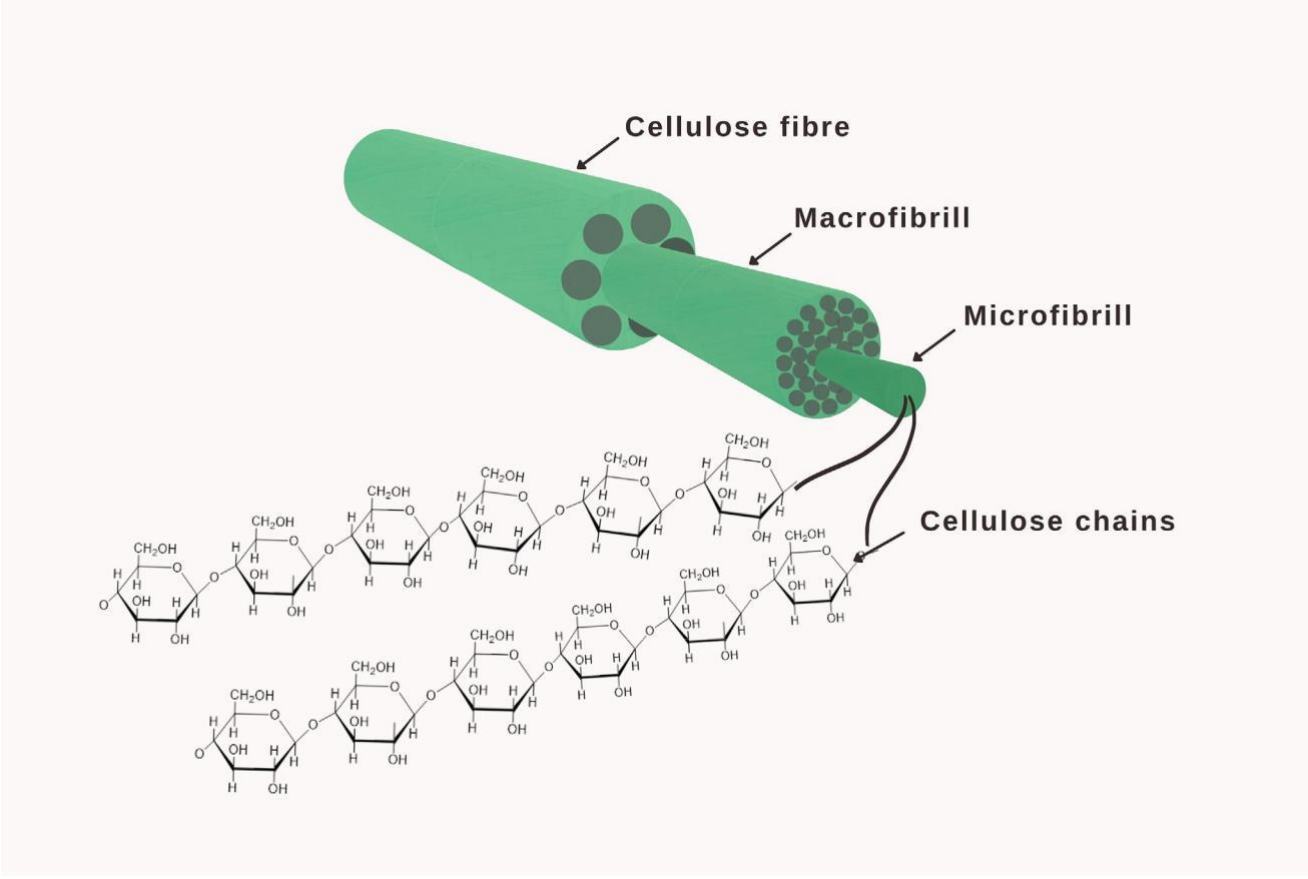

Fig. 2. Cross section of the different hierarchical levels of cellulose fibres. Adapted from Belgacem and Gandini, 2011.

The internal morphology of cellulosic fibres is also important for understanding the degradation of cellulose-based wet wipes (Abu-Rous et al., 2006). Amorphous regions in natural cellulose are more porous than crystalline regions, making them more susceptible to water and enzymatic hydrolysis (Holtzapple, 2003). Therefore, wipes with greater porous and cracked fibre structures may promote fibre fragmentation in aquatic envi-ronments (Enfrin et al., 2020; Duan et al., 2021). However, more porous wipes may also accumulate more small sewer solids during wastewater transport, increasing their density and reducing their porosity, and thus, hindering their transport and degradation (Durukan and Karadagli, 2019).

Few studies have explored the physical properties of wet wipes, especially biodegradable forms, and those that have been done are usually experimental in design and lab-based. Durukan and Karadagli (2019) found that the physical properties of dry and moist-state flushable and non-flushable wipes were very similar in terms of sheet mass, thickness, volume per unit mass, and moisture content, likely due to similar manufacturing processes.

High wet tensile strength and durability are desirable qualities in synthetic wet wipes (Orr and Karadagli, 2020). However, producing bio-based biodegradable wipes that balance wet strength, dispersibility and softness is challenging (Sawhney et al., 2012; Zhang et al., 2018; Yun et al., 2020a). A study comparing the wet and dry tensile strength of "flush-able" nonwoven wipes to toilet paper found that these wipes had almost 23 times greater wet breaking force and retained their strength when wet, un-like toilet paper (Durukan and Karadagli, 2019). The wet strength loss of flushable wipes was only 29 % on average, compared to 91 % for toilet paper.

Petrochemical-derived additives are commonly used to improve the wet strength of cellulose wet wipes (see Section 2.3.3), but they can limit degradation processes (Yun et al., 2020b). However, blends of cellulosic fibres can provide a balance of wet strength and degradability without the use of chemical additives. An experimental study on biodegradable wet wipes found that increasing the viscose content increased wet strength but de-creased degradation, whilst increasing the wood pulp content added soft-ness and degradability (Zhang et al., 2021b).

The orientation of individual fibrils within cellulose fibres affects their strength, elasticity, elongation at breakage, and therefore, their degrada-tion behaviour. Fibres with higher microfibril orientation are stronger but have lower elongation at breakage compared to fibres with low microfibril orientation (Klemm et al., 2005). Natural fibres and lyocell seem to have higher orientation than viscose (Abu-Rous et al., 2006). However, viscose and cotton have been found to share poor elasticity properties (Remy et al., 2015), suggesting that they may also have similar degradation behav-iour.

High liquid absorbency is critical in the production and application of wet wipes, and bio-based cellulosic fibres have a strong advantage in fulfill-ing this requirement. Natural and regenerated cellulose fibres have low wet strength and high absorption properties, which makes them more readily available to biodegrade (Hauser, 2015; Mitchell, 2019; Harter et al., 2021). Furthermore, fibres with greater absorption properties tend to have a deep-grooved geometry due to the higher specific surface area this creates (Soukupova et al., 2007; Duan et al., 2021). However, pre-treated cotton fibres tend to be more hydrophobic (Sawhney et al., 2012).

2.3.3. Chemical additives

Chemical additives are commonly added to materials during manufacturing to improve their properties and durability (Campanale et al., 2020). These additives are present in plastic products but also cellu-losic wet wipes, where they can act as strengthening agents or preservatives to prevent microbial growth (Hahladakis et al., 2018; Alvim et al., 2020; Adams et al., 2021). Examples of such additives include polyacrylic ester and urea-formaldehyde resin, phenoxyethanol, parabens, and benzoates (Obokata and a. I., A., 2007; Yun et al., 2020a; Pack et al., 2021). However, these additives can leach out over time since they are weakly bound to the molecular structure of polymers, leading to potential ecotoxicological is-sues (Horton et al., 2017; Windsor et al., 2019a; Luo et al., 2022). Moreover, additive contaminants can also adsorb onto cellulose fibres in the environment, resulting in similar issues (Jâms et al., 2020; Adams et al., 2021). It is currently unclear however, how different additives affect microfibre degradation in the environment (Henry et al., 2019; Ebrahimbabaie et al., 2022). Additionally, biodegradability tests for poly-mers do not consider the impact of any additives, although biodegradable additives are necessary for a polymer to be considered biodegradable (Lambert and Wagner, 2017; Harrison et al., 2018).

2.4. Disposal pathways and fate

Wet wipes are commonly disposed of through either domestic toilets (flushable) or domestic waste (non-flushable) pathways (Mitchell, 2019). This categorisation is based on a wipe's ability to be flushed down the toilet, known as "flushability" (INDA and EDANA, 2018; Orr and Karadagli, 2020). Ideally, the disposal pathway of wet wipes should also consider the biodegradability and dispersal of their polymers, and logically, syn-thetic petrochemical fibres should be disposed of through bin waste path-ways and bio-based fibres through toilet pathways. However, the actual process is more complicated, and fibres from wet wipes of different origins and designated disposal pathways are found undegraded in wastewater and river environments (Drinkwater and Moy, 2017; Mitchell, 2019; Ó Briain et al., 2020). This emphasises the need for a better understanding of the dif-ferences between flushable and non-flushable wipes (Section 2.4.1), and between biodegradable and non-biodegradable polymers (Section 2.4.2).

2.4.1. Flushable vs non-flushable wipes

Wet wipes and other sanitary products are increasingly marketed world-wide as "flushable" (Atasagun and Bhat, 2020). Packaging instructions claim that these wipes can be disposed of through household toilets and into the sewage system (Orr and Karadagli, 2020; Harter et al., 2021). These wipes are mainly made of biodegradable bio-based fibres that have low tensile strength in their wet state and are typically used as moist toilet tissues and personal hygiene wipes (Zhang et al., 2018). In contrast, non-flushable products are made of synthetic fibres with much higher wet strength (Mitchell, 2019).

Ideally, a wipe should only be considered flushable if it is made entirely of biodegradable bio-based (i.e. cellulosic) fibres without any non-degradable components that could affect sewage infrastructure and fresh-water ecosystems (IWSFG, 2020). However, Joksimovic et al. (2020) found that all tested fabrics labelled as flushable, including wet wipes, only partly degraded when assessed against the International Water Ser-vices Flushability Group (IWSFG) standards.

The confusion about flushable wipes stems from inconsistent criteria and regulations for assessing flushability (Pantoja-Munoz et al., 2018). Manufacturers and consumers often assume that flushable products move easily through wastewater to treatment plants and degrade during or after transport (Durukan and Karadagli, 2019). However, this depends on their physicochemical properties, as well as infrastructural and mechanical factors in the wastewater system, including wastewater flow velocity and pipe dynamics (Orr and Karadagli, 2020). Both flushable and non-flushable wipes have caused sewage blockages (Drinkwater and Moy, 2017; Mitchell, 2019). To address these misperceptions, several industries have produced "flushability criteria" guidelines (INDA and EDANA, 2018; WaterUK, 2019; Wildlife and Link, C., 2021). Despite these methodological efforts, there are still no universally standardised and legally-binding regu-lations placed on manufacturers regarding what can be deemed flushable (Drinkwater and Moy, 2017).

2.4.2. Biodegradable vs non-biodegradable polymers

Biodegradable polymers are materials that can be broken down by nat-ural microorganisms, whereas non-biodegradable polymers, such as tradi-tional plastics, resist degradation due to strong apolar bonds (Fig. 3) (Sun et al., 2022b). However, the definition of biodegradable polymers can vary, and it is often mistakenly believed that bio-based and biodegradable mean the same thing (Filiciotto and Rothenberg, 2021). It is important to

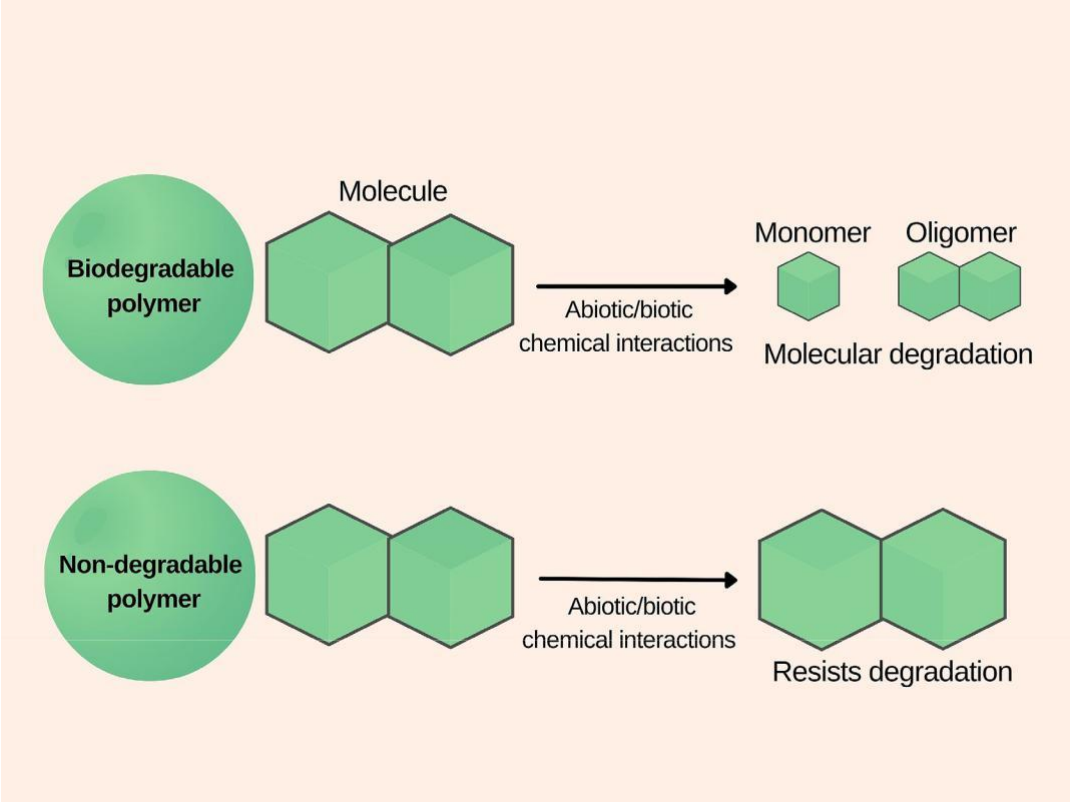

Fig. 3. Molecular degradation pathways for biodegradable and non-degradable polymers in response to abiotic/biotic chemical interactions.

distinguish between bio-based and biodegradable polymers to understand their environmental fate and impact. As pointed out by Lambert and Wagner (2017), bio-based materials come from renewable organic sources, but not all of them biodegrade, including some bio-based plastics. Instead, degradability depends on a polymer's chemical structure rather than its bi-ological source. Sun et al. (2022b) also emphasise that polymer degradation that lowers its molecular mass facilitates the most efficient biomass gener-ation and microbiological assimilation. The breakdown of a biodegradable polymers occurs through chemical reactions in the environment that weaken chemical linkages and polar bonds, breaking molecular chains into smaller compounds that microorganisms can assimilate (Wang et al., 2011; Law and Narayan, 2021).

Cellulose-based fibres in wet wipes are widely understood to be bio-based and biodegradable materials (Leja and Lewandowicz, 2010; Wei et al., 2021), but the biodegradation process is influenced by their manufacturing processes and chemical properties (Park et al., 2004). Stan-dard biodegradation test protocols require high temperature and moisture settings (Straub et al., 2017; Kjeldsen et al., 2018; GOV.UK, 2021), which are mostly limited to industrial and lab-based facilities, and the conditions required for biodegradation in natural environments vary significantly (Wei et al., 2021). Therefore, biodegradable polymers like cellulose may not efficiently degrade outside laboratory conditions (Lambert and Wagner, 2017) and have been found to accumulate as microparticles or fi-bres in aquatic environments (Brinsko et al., 2016; Dris et al., 2018; Wei et al., 2021). However, there are no studies to our knowledge that have properly assessed the fate impact of cellulose-based fibres across their en-tire life cycle.

3. Cellulosic wet wipe degradation mechanisms in different environments

Degradation refers to any process (chemical, physical or biological) that breaks down the large molecular chains of a polymer into smaller units with low molecular weight such as carbon dioxide, water, and methane (Guo and Wang, 2019; Ebrahimbabaie et al., 2022). The underlying principle of a biodegradable polymer is that it will completely degrade (Shen et al., 2020; Liao and Chen, 2021). The primary degradation processes in aquatic systems are mechanical fragmentation, photo-degradation, and biodegra-dation. These often occur together, making it challenging to predict the total life cycle and environmental fate of materials (Chen et al., 2019; Duan et al., 2021; Luo et al., 2022). To better understand the degradation complexity of biodegradable wet wipes that enter the aquatic environment, we investigate, based on first principles, how wet wipes might degrade within the different environments encountered throughout their disposal pathways from flushing (Section 3.1) to wastewater treatment plants (Section 3.2) to receiving freshwaters (Section 3.3) (Fig. 4). We also high-light their intermediate degradation states and show how the mechanisms found might interact (Section 3.4).

3.1. Degradation in the wastewater sewage environment

3.1.1. Fragmentation

When wet wipes are flushed down the toilet, they enter the sewage sys-tem and mix with wastewaters (Fig. 4). In this sewage transport stage, two degradation mechanisms are expected. The first process is fragmentation, whereby exposure to abrasive and mechanical forces from physical interac-tions with other (semi)solid particles break a material down into smaller pieces (Duan et al., 2021; Drummond et al., 2022). For wet wipes, physical interactions in sewers will likely result in material surface crack defections that initiate further fragmentation processes throughout the sewer, creating an abundance of increasingly smaller fibres (Enfrin et al., 2020; Drummond et al., 2022).

The overall physical damage caused to wet wipes significantly depends on their tensile strength and elastic modulus (Ghorbani et al., 2013), al-though their overall mechanical stability will be reduced as a result of these abrasive and mechanical forces (Duan et al., 2021). In the case of cellulose-based wet wipes, physical degradation is expected to be high as their fibres typically have weaker tensile strength and elasticity than their

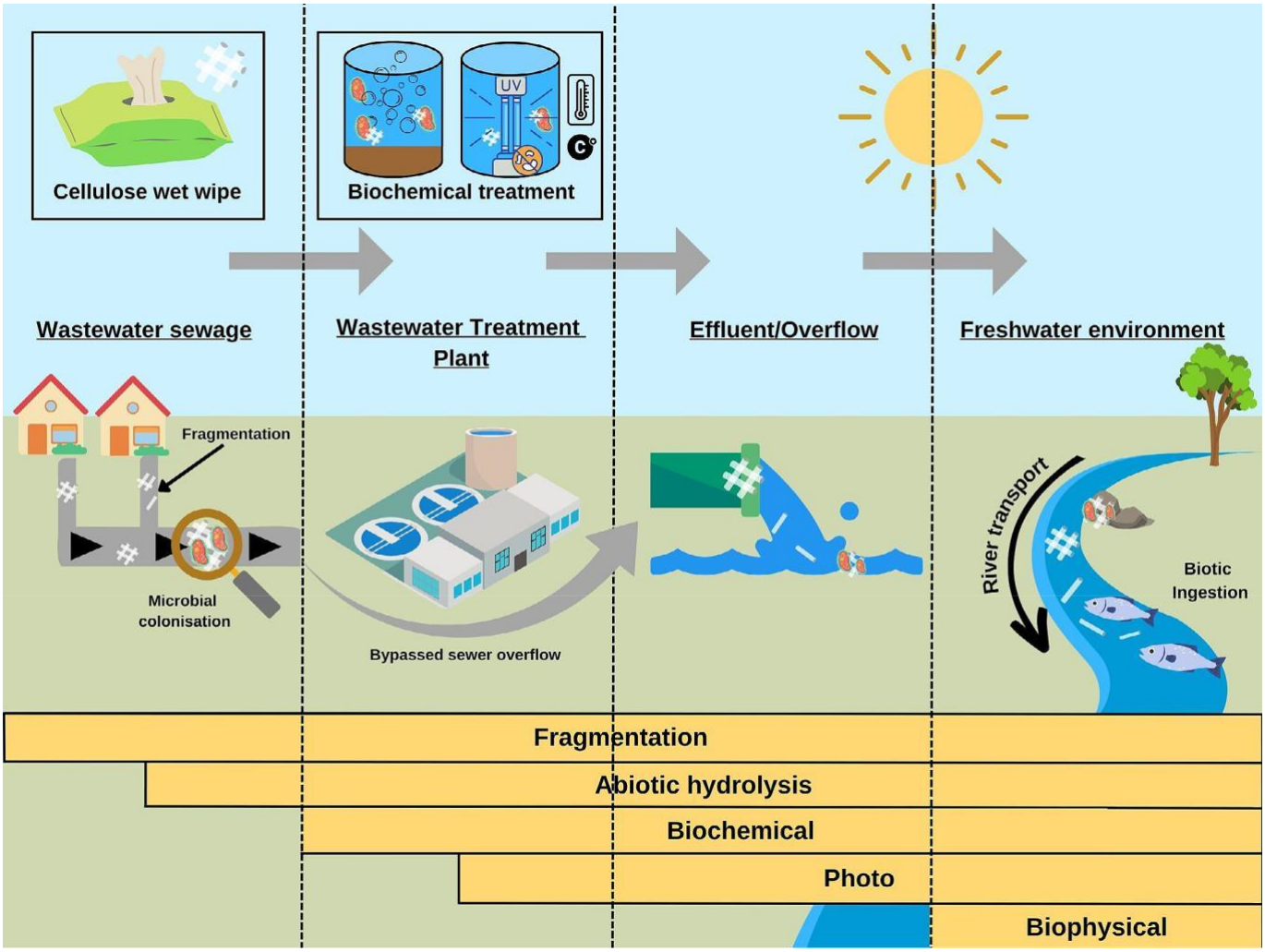

Fig. 4. Flushed cellulose wet wipe disposal pathway and expected degradation processes at each stage.

synthetic counterparts (Klemm et al., 2005; Remy et al., 2015). Recent experiments suggest that cellulose-based wipes may shed an average of 548,000 microfibres per gram in simulated wastewater conditions (Kwon et al., 2022).

3.1.2. Abiotic hydrolysis

The second degradation mechanism expected for cellulosic wet wipes abiotic hydrolysis (Fig. 4), a process where water reacts with the material's surface to break down its molecular chains (Speight, 2017). Hydrolytic degradation is influenced by the presence of hydrophilic monomers and end groups, degree of crystallinity, overall material size, and oxygen-containing functional groups in the polymer backbone (Kale et al., 2007; Farah et al., 2016; Filiciotto and Rothenberg, 2021). In biodegradable plas-tics, hydrolytic degradation occurs in two stages: 1) random hydrolytic scis-sion of ester bonds in the amorphous regions leading to increased crystallinity, and 2) inward degradation of the amorphous regions towards the polymers' core (Elsawy et al., 2017).

For cellulose, abiotic hydrolytic degradation involves the molecular scission of its glycosidic bonds, which typically requires industrial acidic or alkaline conditions that are not commonly found in wastewaters (Sharma et al., 2013). Hydrolytic degradation in cellulose is also made dif-ficult due to its high degree of insolubility, inflexible molecular chains, and self-aggregation of fibres with higher hemicellulose content into gelatinous substances in water (Ghasemi et al., 2017; Väisänen et al., 2021a; Väisänen et al., 2021b). Cellulose-based wet wipes exacerbate these issues by aggre-gating in wastewaters and causing blockages (Drinkwater and Moy, 2017).

In principle, for cellulose fibres to degrade hydrolytically, water must dif-fuse into its structure and affect both the amorphous and crystalline re-gions, however, the efficiency of this molecular degradation relies on the reduction of its crystalline region and the disentanglement of its molecular chains simultaneously (Cheng et al., 2012; Ghasemi et al., 2017; Ghasemi et al., 2018), which is often not possible by abiotic hydrolysis alone. There-fore, hydrolytic degradation of cellulose-based wet wipes is likely only ef-fective when other degradation mechanisms such as fragmentation are also occurring (Fig. 4).

Hydrolysis causes cellulose fibres to swell, especially in fibres with smaller diameters and lower crystallinity (Ghasemi et al., 2018; Väisänen et al., 2021a). It occurs as a result of the high number of hydroxyl groups within the secondary wall that allows water molecules to easily penetrate under osmotic pressure, increasing the overall solvent concentration inside (Peng et al., 2017). Fibre swelling can reduce the material's overall ability to degrade within aquatic environments and may be visually identifiable or detected through initial polymer weight gain (Khazraji and Robert, 2013; Harter et al., 2021). This raises concerns about the pre-treatment pro-cesses involved in making biodegradable wet wipes and their solubility for effective hydrolytic degradation within the aquatic environment.

3.2. Wastewater treatment plant

Following their sewer transport, cellulosic wet wipes and their fibres enter WWTPs and are subject to a range of filtration processes. These in-clude physical filtration processes that remove the larger fractions of wet

wipe; however, their smaller fragmented microfibres can pass through into the biochemical filtration stages of treatment. Physical fragmentation and abiotic hydrolysis processes are expected to continue during this wastewa-ter stage due to their sustained physical interactions with wastewater and solid particles. Furthermore, abiotic hydrolysis of cellulose-based wet wipes and their fibres may be enhanced by the introduction of two expected chemical degradation mechanisms during this stage – biodegradation and photodegradation (Fig. 4).

3.2.1. Biodegradation

Polymers in the environment can be biodegraded through two main pathways: biochemical microbial degradation and biophysical biotic inges-tion (Duan et al., 2021; Ebrahimbabaie et al., 2022). As cellulosic wet wipe fibres are fundamentally organic and biodegradable, biochemical degrada-tion is expected to play a key role in their breakdown in both wastewater systems and freshwater environments downstream.

Biochemical degradation is a complex process that occurs in multiple stages, some of which overlap with biophysical processes (Luo et al., 2022). In aquatic systems (Fig. 5), microorganisms first colonise the poly-mer surface, attracted by physical properties such as porosity, density, sur-face tension, and size (Windsor et al., 2019a; Zambrano et al., 2020b; Luo et al., 2022), as well as environmental factors including pH nutrient avail-ability (Roohi et al., 2017). As they colonise, the microorganisms secrete ex-tracellular polymer substances, which form a sticky biofilm on the polymer surface to aid further microbial growth (Roohi et al., 2017; Duan et al., 2021; Ngyuen et al., 2021). This is followed by a simultaneous biophysical and biochemical process in which hydrolase enzymes catalyse the hydroly-sis of the molecular chains, leading to progressive dissimilation of the poly-mer and surface fragmentation (Bano et al., 2017; Liao and Chen, 2021; Ebrahimbabaie et al., 2022). This process generates low molecular weight oligomers and monomers, which are released into the environment due to chemical degradation and can be used as biomass energy sources for mi-crobes through further bio-assimilation, mineralisation, and metabolite production (Bano et al., 2017; Duan et al., 2021; Liao and Chen, 2021; Ebrahimbabaie et al., 2022; Sun et al., 2022b).

Significant morphological changes occur during biochemical degrada-tion, including changes in overall MW, tensile strength, surface area, adhe-sion, and hydrophilicity (Luo et al., 2022). Enzymes play a vital role in these changes, leading to loss of mechanical stability and embrittlement (Yuan et al., 2020), as well as the breakdown of polymer additives, which can be released into the environment or promote further biological assimilation (Duan et al., 2021; Luo et al., 2022). However, microbial hydrolytic pro-cesses within the natural environment are influenced by various polymer properties, including stereochemistry (Shen et al., 2020; Liao and Chen, 2021), crystallinity (Emadian et al., 2017; Wei et al., 2021), MW (Wei et al., 2021), size and surface area (Duan et al., 2021), as well as environ-mental conditions and other degradation mechanisms such as light availability, water quality, nutrients, microorganism communities and me-chanical fragmentation forces (Zambrano et al., 2020b).

Limited research exists on the biochemical degradation of cellulosic wet wipes and their fibres in WWTPs. However, studies on the biodegradability of various cellulose-based materials in different environments provide valu-able insights into their fate. For instance, rayon fibres had the highest rate of biodegradation due to their low crystallinity, degree of orientation, and high moisture regain (Park et al., 2004). Moreover, microbial biodegrada-tion (>70 %) of cotton and rayon fibres was observed in WWTP activated sludge and freshwater, whilst poorer biodegradation occurred in seawater conditions and for cotton/polyester blends in general (Zambrano et al., 2020b). However, the extent of biodegradation depends on the types of en-zymes and microorganism communities present (Nagamine et al., 2022). Experiments in rivers and lakes with cellulosic cotton strips also show how their biodegradation varies with temperature, nutrient concentrations, sediment load, and acid-base status (Tiegs et al., 2013; Colas et al., 2019; Carballeira et al., 2020).

We expect biodegradation processes in cellulosic wet wipes to be most effective during their passage through WWTPs, where a wide range of aer-obic and anaerobic microbial communities are present and where environ-mental conditions are controlled to facilitate molecular breakdown. Additionally, the degradation may occur earlier in the wastewater system when cellulosic wet wipes contribute to sewer blockages and are colonised

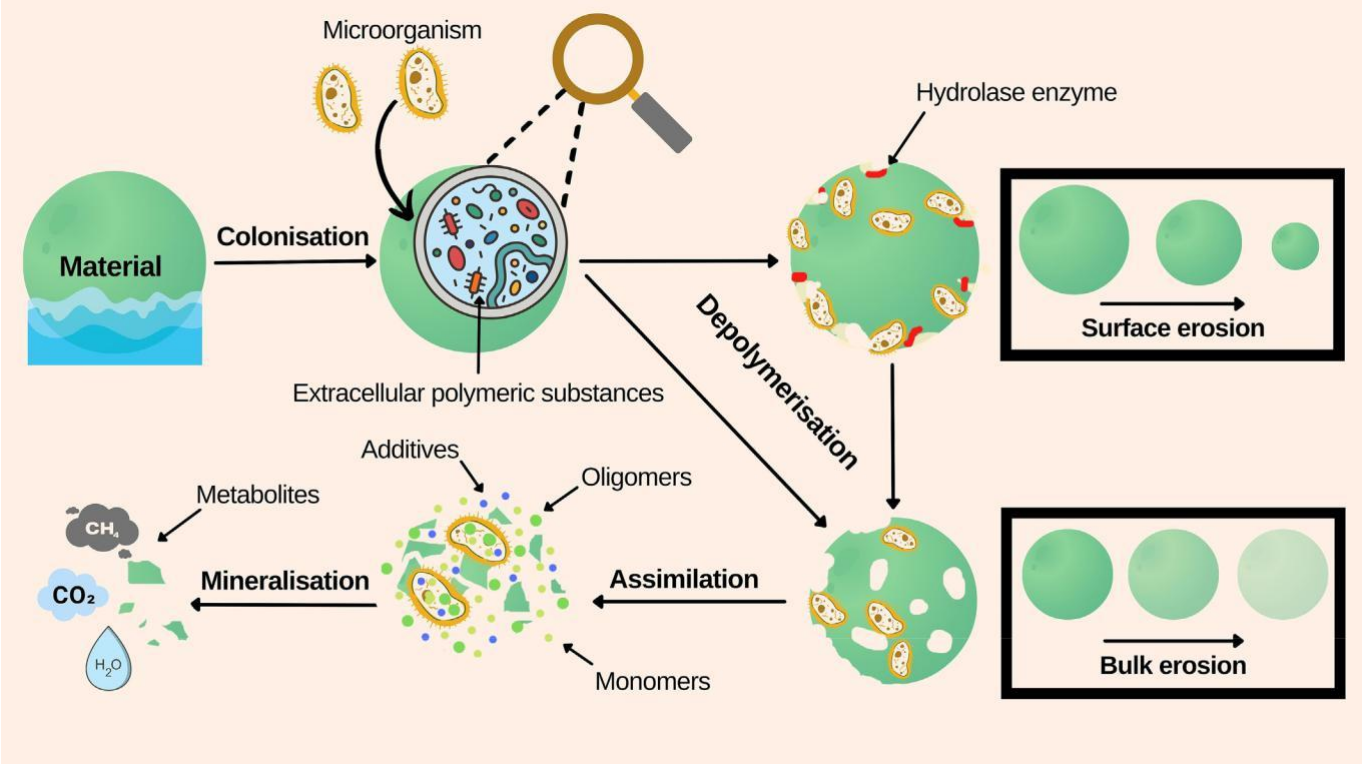

Fig. 5. Microbial degradation pathway for biodegradable materials in aquatic environments. Adapted from Duan et al. (2021).

by microbial communities, leading to subsequent biological breakdown (Drummond et al., 2022).

3.2.2. Photodegradation

Photodegradation is a significant cause of plastic breakdown (Zhang et al., 2021a), as well as cellulose in lab-based (Malešič et al., 2005), and quite possibly in aquatic environmental conditions (Fig. 4). This process is predominantly caused by ultraviolet (UV) energy from sunlight, which reacts with the photo-sensitive components of polymers (Luo et al., 2021). Photodegradation can cause molecular chain scission, surface oxida-tion processes, and the loss of mechanical properties, ultimately leading to MW loss into smaller harmless compounds (Rummel et al., 2017; Shao et al., 2018; Liao and Chen, 2021; Luo et al., 2022). UV filtration technology utilising photo-catalytic reactions is often used to treat and remove pollut-ants from wastewater (Ebrahimbabaie et al., 2022).

UV radiation with a wavelength>340 nm provides enough energy to de-grade cellulose with the formation of hydroxyl radicals and oxidation (Malešič et al., 2005). Previous studies on cellulose photodegradation have also found hydroxyl radical breakdown products to form in cotton cellulose at wavelengths between 330 and 360 nm (Phillips et al., 1966; Hon, 1976).

Oxidation plays a critical role in cellulose molecular degradation (Shao et al., 2018). Free radicals and reactive oxygen species form when UV energy photons are absorbed by photo-sensitive chromophores in the macromolecules, causing molecules to enter excited energy states (Ebrahimbabaie et al., 2022). These excited states are then transferred to the nearest C–C and C–H covalent bonds through intramolecular energy transfer processes, overcoming their bond enthalpies and initiating molec-ular breakages (Duan et al., 2021; Filiciotto and Rothenberg, 2021). Reactive oxygen species have also been found to continue molecular degra-dation by migrating into the deeper layers of polymers (Ebrahimbabaie et al., 2022).

Manufactured cellulosic fibres have been found to contain a small num-ber of chromophoric compounds, which are believed to derive from ther-mal, acidic, or basic condensation products of saccharides created during cellulose degradation (Rosenau et al., 2004). These same chromophores often show discolouration and surface damage in response to photo-degradation in the aquatic environment (Cai et al., 2018; Ebrahimbabaie et al., 2022). Photodegradation processes in microparticles and fibres are highly influenced by their crystallinity, with higher crystallinity causing light to scatter and reflect (Duan et al., 2021), reducing light penetration distance. As a result, most photodegradation processes occur around the surface layers of crystalline structures instead of the bulk (Luo et al., 2022). UV irradiation and oxidation changes the surface hydrophobicity and adsorption capacity of microparticles (Lin et al., 2020), and these mor-phological changes are likely increased by additional polymer degrading processes and environmental conditions (Duan et al., 2021). Although there is no study on the subject, it is expected that cellulosic wet wipes could also be impacted by these processes.

Thermal degradation treatment is sometimes used to improve the per-formance of anaerobic digestion of pollutants and sewer sludge dewatering processes (Hii et al., 2014). This process involves the absorption of energy (i.e. heat) to overcome the bond dissociation energy of C–C and C–H bonds, leading to chain scission, crosslinking, oxygenated free-radical for-mation, and a loss of molecular weight (Zhang et al., 2021a; Luo et al., 2022). Unlike photodegradation, thermal degradation affects the entire polymer rather than just its surface layers (Lambert, 2013). However ther-mal degradation is often expensive and not feasible in wastewater treat-ment plants due to its high operating costs, and it typically requires temperatures exceeding 300 °C for natural cellulosic fibres (Jandura et al., 2000; Yang et al., 2007). This makes it an unlikely mechanism for cel-lulose wet wipes both in industrial and natural environments.

3.3. Freshwater environment

Two disposal scenarios exist for wastewater into freshwater environments (Fig. 4). Raw wastewater either enters WWTPs, undergoes filtration and is then pumped out into the aquatic environment, or this pathway is bypassed, and wet wipes enter the aquatic environment through sewer spills – for example from combined sewer overflows. Spills are the predom-inant source of wet wipes directly into the freshwater environment (Besley and Cassidy, 2022). Given the evidence so far on freshwater pollution from cellulose fibres (Dris et al., 2018; Stanton et al., 2019; McCoy et al., 2020; Ó Briain et al., 2020), it is likely cellulosic wet wipe fibres might enter from both raw sewage spills and from treated sewage effluents.

Within freshwater environments, all previously highlighted degrada-tion mechanisms for cellulosic wet wipes (Fig. 4) are expected to continue, particularly, physical fragmentation processes, due to the abrasive and me-chanical forces caused by riverbed sediment, solid particles, and hyporheic processes (Duan et al., 2021; Drummond et al., 2022). Fragmentation rates for fibres can vary depending on their size, density, and transport. Larger and denser fibres are likely to sink, become buried within river sediments, and thus fragment more slowly (Nizzetto et al., 2016), although this may depend on the benthic and hyporheic exchange rates within freshwater sources (Eerkes-Medrano et al., 2015; Drummond et al., 2022). No studies were found on the impacts of physical degradation on cellulosic wet wipes within freshwater ecosystems. Nevertheless, there have been reports of wipes composed of cellulose and synthetic fibre blends found in down-stream sewage effluent and freshwater that have likely been fragmented through crack defects and shear stress forces of mechanical mixing from WWTPs (McCoy et al., 2020; Ó Briain et al., 2020; Lee et al., 2021). During this freshwater transport stage, cellulosic wet wipes and fibres are likely to also be degraded through biophysical interactions with aquatic life.

Biophysical degradation comes from the physical interaction and conse-quent ingestion and digestion of debris by biota within the natural environ-ment. This is a well-evidenced process in freshwater for microfibres of natural, regenerated and synthetic origin (Remy et al., 2015; McGoran et al., 2017; Miller et al., 2017; Dris et al., 2018; Mateos-Cárdenas et al., 2020; Stone et al., 2020; Mateos-Cárdenas et al., 2021). Overall, it is seen as an ecologically negative process, due to the possible toxic and physically harmful nature of manufactured particles and their chemical additives (Ó Briain et al., 2020; Luo et al., 2022). Microparticle interactions with organ-isms may lead to the reduction in foraging behaviour, malnutrition, and physical blockages of the gastro-intestinal tract (Graham and Thompson, 2009; Ebrahimbabaie et al., 2022; Sun et al., 2022a). These impacts might also be enhanced if the fibres have absorbed pharmaceutical, agricul-tural and industrial chemicals found in WWTP's and untreated freshwater (Jiang et al., 2021; Ebrahimbabaie et al., 2022).

Whilst much of the focus has been on aquatic biota interactions with synthetic microplastics (Dris et al., 2018; Sun et al., 2022a), some biotic in-teractions with cellulose-based fibres have been identified. Lusher et al. (2013) found that the majority of microparticle polymers ingested by fish within the English Channel were rayon-based. Remy et al. (2015) identified viscose fibres within the gut content of macroinvertebrates on the Mediter-ranean coast. Cotton fibres were identified within the stomachs of the Clupeiformes fish (Collard et al., 2015). Jamieson et al. (2019) reported the biotic ingestion of cellulose, rayon and lyocell fibres by deep-sea Lysianassoidea amphipods in the Pacific Ocean. Fibre ingestion also repre-sents a potential biological pollutant pathway to higher trophic levels and other ecosystems (Lusher et al., 2013; Remy et al., 2015). Evidence of inges-tion of cellulosic fibres by biota (other than microorganisms) is however still lacking in freshwaters (Dris et al., 2018).

Physical ingestion and digestion can also contribute to the continued fragmentation of materials that may either remain in an organism's diges-tive tract or be excreted back into an aquatic environment (Straub et al., 2017; Dawson et al., 2018). This has been identified in a lab-based study by Mateos-Cárdenas et al. (2020), of freshwater amphipods, Gammarus duebeni, which ingested PE microparticles, physically fragmenting them into smaller size orders in process. This fragmentation may be continued and catalysed by gastrointestinal enzymatic action (i.e. from amylase, cellu-lase, esterase, protease and lipase etc.) within the organism (Song et al., 2020; Duan et al., 2021). A follow-up study was conducted by the authors (Mateos-Cárdenas et al., 2021), and found a greater abundance of ingested

cellulose fibres than PE by Gammarus duebeni, although it presented no clear ecotoxicological impacts or fragmentation and excretion behaviour.

3.4. Interacting degradation mechanisms and end products entering natural environments

Understanding the behaviour of these different degradation mecha-nisms within the environment is made difficult by the often interacting and overlapping nature of these processes. Nevertheless, this is key to better understand and quantify the overall environmental fate and extent of risk from cellulose based wipes.

For example, in freshwater systems, photodegradation mechanisms that cause embrittlement on the polymer surface simultaneously promote the rate of mechanical fragmentation by physical abrasion (Song et al., 2017; Duan et al., 2021). Photo-oxidation processes within the surface of a poly-mer, can speed up the rate of microbial biodegradation and enzymatic hy-drolysis (Filiciotto and Rothenberg, 2021). Biological ingestion can alter the physicochemical properties of polymers, promoting fragments and fi-bres, and if excreted, can influence their river transport and subsequent me-chanical fragmentation behaviour (Windsor et al., 2019a). Physical fragmentation and the formation of crack defects can increase the surface area for microbial colonisation and for enzymatic hydrolysis reactions to take place (Zambrano et al., 2020b; Ngyuen et al., 2021). Abiotic degrada-tion mechanisms that cause chain scission near the polymer surface may re-sult in the formation of oxygen-containing groups, increasing its hydrophilicity and thus improving its biodegradation potential (Gewert et al., 2015; Ebrahimbabaie et al., 2022).

Hydroxyl radical-induced degradation is a common but important molecular breakdown subprocess. It occurs within polymers during exposure to a range of different chemical degradation mechanisms, such as photodegradation, oxidative-degradation, biodegradation, and hydrolytic degradation (Shao et al., 2018; Tian et al., 2019; Shao et al., 2020; Zhang et al., 2021a; Luo et al., 2022). In essence, highly reactive radicals attack the carbon backbone of the polymer, causing further chain scission and crosslinking processes to occur, and they play an important role in cellulose degradation. They can initiate hydrogen cleavage of the C–H bonds on cellu-lose's pyranose rings in the presence of oxygen, and cause subsequent chain scissoring of the β (1 → 4) glycosidic bonds (Haskins and Hogsed, 1950). They can also form peroxyl radicals that interact with cellulose photolytically leading to further radical formation and molecular breakdown (Duan et al., 2021). As hydroxyl-radical degradation is caused by various chemical break-down processes found in aquatic environments (e.g. abiotic hydrolysis and photodegradation), theoretically, it is expected to occur within cellulose-based wet wipes throughout their wastewater and freshwater transport.

Overall, the likely end-products of biodegradable wet wipe entering nat-ural aquatic environments are the result of five main degradation mecha-nisms discussed in Sections 3.1–3. These end-products may consist of the following components: 1) Fibres of varying sizes, which occur due to phys-ical fragmentation processes along the domestic disposal pathway, and bi-otic ingestion in the downstream environment; 2) Molecular breakdown products (e.g. oligomers, monomers, additives, and metabolites) generated through hydrolytic, ultraviolet, and microbial-induced degradation mecha-nisms at different stages of the wastewater disposal pathway (See Fig. 4); and 3) Swollen wet wipe fibres and their undegraded amalgamations, par-ticularly observed in sewer systems and when exposed to water.

4. Variables controlling degradation in man-made or natural water environments

4.1. Oxygen

Environmental oxygen levels can play a significant role in the extent of all previously mentioned degradation mechanisms within aquatic environ-ments (Duan et al., 2021; Issac and Kandasubramanian, 2021). To date, however, research on the anaerobic degradation of MPs (non-degradable and biodegradable) within the environment is scarce (Ebrahimbabaie et al., 2022). The presence of oxygen can alter the metabolic pathway of mi-crobial action to polymers (Duan et al., 2021), and may partially influence thermal degradation. In oxygen-rich environments, microorganisms use polymers as an energy source and produce carbon dioxide and water by-products in addition to heat, whilst in anaerobic conditions, microbes pro-duce methane gases and biomass with less thermal energy produced (Bátori et al., 2018).

WWTPs use both aerobic and anaerobic microbial conditions for bio-degradation as an effective secondary treatment for removing organic mat-ter within sewage systems (AEAL, 2009), with anaerobic microbial action commonly occurring within sewage sludge biosolids. Biodegradation is po-tentially more limited within unconditioned freshwater environments be-cause there is a less diverse range of microbial communities available (Zambrano et al., 2020b). Oxygen levels within freshwater can also be changed by chemically toxic contaminants, often stemming from wastewa-ter effluent and agriculture (Jamee and Siddique, 2019), which may have an influence on the rate of environmental degradation processes.

4.2. Water

Water itself can have varying impacts on environmental degradation processes. Photodegradation in freshwaters may be limited by light scatter-ing on the water surface (Duan et al., 2021), although the extent of this hin-drance would most likely depend on polymer buoyancy and the depth of the water source. Water plays an important role in influencing biodegrada-tion, by impacting the types of microorganism communities available (in-cluding algae, bacteria, fungi, protozoa etc.), their polymer colonisation and ability to form biofilm, and the rate of enzymatic hydrolysis (Yuan et al., 2020). Water content makes up a large proportion of wet wipes (>90 %), which may therefore promote their microbial colonisation and biofilm formation within aquatic environments (Salama et al., 2021).

Polysaccharides such as cellulose are generally hydrophilic in nature due to the high presence of hydroxyl groups, and as such, tend to react strongly to hydrolysis mechanisms from aquatic environments, especially within less organised amorphous regions (Karamanlioglu et al., 2017). However, as cellulose fibres are molecularly strong-bonded, water molecules are only capable of breaking already weakened bonds from other environmental degradation mechanisms (Khazraji and Robert, 2013). Nevertheless, cellulose-based wipes have been reported to easily tear and fragment when agitated in water (Ó Briain et al., 2020).

4.3. Temperature

Specific temperature conditions are important to many environmental degradation processes, but no more so than in biotic mechanisms. The bio-degradation of biopolymers, as found in standardised industrial composting environments, partly relies on high temperature conditions (55 °C) (Liao and Chen, 2021) which are seldom found in the natural environment. Fresh-water systems can vary in temperature by season and geography, but usually range anywhere between 2 and 24 °C. WWTPs typically operate at slightly lower temperatures for biological filtration processes, and at slightly higher temperatures around 31 °C for aerobic biological treatment and 37.5 °C for anaerobic sludge digestion (Alisawi, 2020). Higher temperatures not only promote thermal degradation of the molecular bonds (Luo et al., 2022), but also stimulate greater microbial and enzymatic activity (Duan et al., 2021), leading to a higher rate of hydrolytic breakdown in the polymer backbone (Kale et al., 2007). Zambrano et al. (2020a) found cellulosic cotton textile fabrics to shed more microfibres at higher temperatures during home laun-dering experiments. Research on cotton strip bioassays employed in freshwa-ter streams have also identified temperature as a significant factor influencing cellulosic biodegradation (Griffiths and Tiegs, 2016).

4.4. pH level

The pH level of the receiving waters can also regulate degradation. For example, physical fragmentation of biodegradable polymers is reported

faster in seawater compared to freshwaters, as a result of higher rates of hydrolysis linked to less acid conditions (Wei et al., 2021). However, hydrolytic degradation can occur in both pH extremes (Elsawy et al., 2017), and depolymerisation of cellulose is commonly achieved under both acidic and alkaline conditions (Väisänen et al., 2021a), with glycosidic bond cleavage possible under acidic conditions and general hydroxyl radical-induced degradation purporting to be enhanced by both (Shao et al., 2018; Shao et al., 2020). Microparticle absorption behaviour in aquatic en-vironments is known to vary with pH, with consequences for pollutant transport, and this could also apply to cellulose (Xu et al., 2018; Tirkey and Upadhyay, 2021; Sun et al., 2022b).

4.5. Organic matter, nutrients, and contaminants

Manufactured particles in aquatic systems can act as a vector for various toxic/nontoxic chemical elements, such as metal ions and natural organic matter, that could influence the surface morphology and degradation mechanisms (Li et al., 2018; Zhou et al., 2020). For example, microparticles in freshwaters are often covered in dissolved organic matter (i.e. humic acid) that carry negative ionic charge and interactive oxygen-containing functional groups, which may alter their surface properties and polymer degradation reactions (Chen et al., 2018; Sun et al., 2022b), particularly photodegradation since organic matter presents highly abundant chromo-phores that can act as photosensitisers (Duan et al., 2021).

The concentration of organic matter could also influence the abundance and variety of microbial communities that colonise onto wet wipes (Windsor et al., 2019a). For example, for microparticles, high concentra-tions of nutrients and organic matter within wastewater environments have been found to increase microbial activity, improving carbon and nitro-gen mineralisation and bio-assimilation, and enzymatic degradation (Tiegs et al., 2013; Colas et al., 2019; Zambrano et al., 2020b; Schell et al., 2022). Furthermore, it is likely cellulosic fibres that are degraded into their glucose units could also in turn provide nutrients that promote microbial colonisa-tion and biofilm formation (Salama et al., 2021).

4.6. Summary of expected degradation behaviours built on first principles

Based on the theorised mechanisms discussed in Section 3, and their likelihood within real-life environmental conditions highlighted in Section 4, we expect physical fragmentation to be the predominant degra-dation process throughout the disposal pathway of a flushed biodegradable wet wipe. This process may occur independently, through physical interac-tions during transport, or in conjunction with physico-chemical breakdown mechanisms. Of the expected molecular degradation mechanisms, it seems abiotic hydrolysis is likely to be the most widespread, due to the interac-tions that flushed wet wipes will have with water throughout their trans-port. However, initial fragmentation and other molecular degradation processes are required for this to become effective. Moreover, based on our analysis in Sections 3 and 4, it seems that biochemical degradation is likely to be an effective mechanism for the molecular breakdown of cellu-losic wet wipes in aquatic environments, especially within sewer systems and WWTPs where the most diverse range of microbial communities are likely to be found. Ultimately, the extent of these degradation processes in biodegradable wet wipes will be influenced by their specific fibre compo-sition and physicochemical properties.

5. Original contributions and perspectives

We asked if flushed wet wipes labelled as biodegradable really de-graded on their routes through wastewater systems and into freshwater en-vironments. Whilst mostly cellulose based, our findings indicate that these wipes often consist of a combination of bio-based and synthetic materials, including various chemical additives that are highly likely to limit their en-vironmental degradation.

Given the current lack of research on the degradation mechanisms in-volved, we identified five likely aquatic degradation mechanisms for cellulosic wet wipes based on first principles: photodegradation, abiotic hydrolysis, physical fragmentation, biophysical ingestion, and biochemical microbial degradation. Of these mechanisms, fragmentation and biochem-ical degradation were likely to be the most dominant in real-world condi-tions but the extent of the latter process is still limited.

We thus demonstrated that cellulosic wet wipes are likely to contribute significantly to microfibre pollution in aquatic environments due to limited molecular breakdown and significant physical fragmentation processes during transit from toilet disposal to aquatic environments.

Consequently, and despite being marketed as “biodegradable” and “flushable”, these products are likely to persist in the environment and pose significant threat to aquatic ecosystems. Lack of international regula-tion for biodegradable and flushable wet wipes will no doubt compound current consumer confusion around appropriate disposal methods.

To advance our understanding of the environmental impact of wet wipes and to support better decision-making for their management and reg-ulation, the following research directions are proposed:

1) Conduct more field research to better understand the sources, abun-dance, and degradation mechanisms involved in biodegradable wet wipes and their microfibres within wastewater, sewage sludge and freshwater environments.
2) Investigate the role of textile additives applied during wet wipe manufacturing, which may significantly influence the degree of environmental degradation and harm.
3) Apply a wider spatio-temporal context that captures the transport pathways of wet wipes and their fibres, and their possible degradation processes along this transport. This context should consider wider life cycle assessments of the product, from raw material source to disposal and final environmental fate.
4) Consider the biophysical interactions of wet wipes and their fibres in a wider ecological context, assessing the transference capability and risk of ingested bio-based microfibres to higher trophic levels within and be-yond freshwater ecosystems.
5) Investigate further the environmental variables that influence physical and chemical breakdown, particularly for cellulosic fibres and wet wipes.
6) Clarify methodologies towards field sampling, including common units of measurement, chemical identification, and sampling techniques, to promote easier and more effective data communication and comparable results across research.

CRediT authorship contribution statement

Thomas Allison: Conceptualization, Methodology, Writing – original draft, Writing – review & editing. Benjamin D. Ward: Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Funding acquisition, Supervision. Michael Harbottle: Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Funding acquisition, Supervision. Isabelle Durance: Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Funding acquisition, Supervision.

Data availability

No data was used for the research described in the article.

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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