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## Initial estimates of the lifetime of unsmoked cellulose diacetate and paper cigarette filters in the coastal ocean†

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Cigarette filters (CFs) are the most littered items on the planet, but their fate in the coastal ocean is unknown. The results of this study demonstrate that unsmoked cellulose diacetate and proposed replacement paper CFs have similar lifetimes, suggesting that policy aimed at transitioning to paper filters is unlikely to reduce CF pollution in coastal areas. Three alternative paths to curb CF pollution are discussed, focusing on research, infrastructure, and education.

### Introduction

Single-use plastics are major sources of plastic pollution with negative impacts on ecosystem and human health. Given the concerns with this emerging pollutant across diverse stakeholder groups, governmental agencies and legislators are seeking solutions to minimize the use and leakage of single-use plastics. For example, the United States National Oceanic Atmospheric Administration's (NOAA) Marine Debris Program, the 2019 no. 904 European Union Directive, and the negotiations UN Treaty on Plastic Pollution all aim to reduce the impacts of frequently leaked single-use plastic products, including beverage bottles, service ware, textiles, fishing gear, and cigarette filters (CFs).<sup>1–3</sup>

Today, practically every cigarette sold is single-use and has a filter.<sup>4</sup> CFs have been made from a wide range of materials, including cellulose diacetate (CDA), paper, polylactic acid (PLA), polypropylene (PP), and viscose.<sup>5–8</sup> However, about 98% of CFs are made from CDA.<sup>9</sup> Since the 1950s, this material has been preferred by smokers and the tobacco industry due to its performance properties, including, for example, pressure drop, tip firmness, and selective filtration for managing taste.<sup>10,11</sup>

An estimated five to six trillion cigarettes are smoked each year. Approximately 50% of smoked cigarettes are improperly

### Environmental significance

Cigarette filters (CFs) are ubiquitous in coastal regions and thus are the focus of policies aimed at curbing CF pollution. However, the lifetimes of current-use cellulose diacetate (CDA) and proposed alternative paper CFs have not been explored in depth. Herein, we report that unsmoked CDA and paper CFs are biodegradable and their lifetime in the coastal ocean is similar, raising questions about the effectiveness of proposed policies. Moreover, a comparison of economic and environmental metrics reveals that switching from CDA to paper CFs could yield substantial costs related to the freshwater footprint of manufacturing, while garnering marginal savings related to greenhouse gas emissions. These findings highlight the need for diverse stakeholder groups to better understand the fate and impacts of CFs on coastal ecosystems.

disposed of in the environment, making CFs the most widely littered item on the planet.<sup>12</sup> Routinely, global litter clean-up surveys conducted over several decades in coastal and urban areas report that CFs were the most abundant item recovered, comprising nearly one in five pieces of litter.<sup>13</sup> According to directive 2019 no. 904,<sup>1</sup> CFs are the second most found single-use plastic item on surveys of EU beaches. Given that about 20% of the global population smokes regularly, and the population is increasing faster than the decline in smoking rates,<sup>14</sup> it is expected that CFs will remain the most littered item for the foreseeable future. Therefore, identifying strategies to understand the fate of CFs in the environment and minimize such litter is critical.

The fate of CFs in the environment is poorly constrained, particularly in coastal ecosystems. To date, nearly all studies focused on the fate of CFs have been conducted in terrestrial, composting, or landfill conditions,<sup>15–23</sup> with all reporting signs of degradation on varying timelines. For example, one study showed progressive degradation in terrestrial ecosystems of up to 80% mass loss over five years.<sup>15</sup> While substantial degradation was measured, the degradation rates varied across terrestrial ecosystems (e.g., grassland vs. desert) and nitrogen availability.<sup>15</sup> Despite calls for an improved understanding of CF degradation in marine systems,<sup>24</sup> no study has estimated the lifetime of CFs in the coastal ocean.

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Grey literature estimates are highly uncertain, with some reports suggesting that CFs could persist for up to a decade<sup>25,26</sup> and others suggesting that CFs are non-biodegradable and thus persist forever.<sup>27,28</sup> In contrast, recent peer-reviewed research demonstrated that the CDA-based fibers comprising CFs biodegrade on timelines of months in the coastal ocean,<sup>29</sup> implying that CFs may not persist in the coastal ocean. However, during the manufacture of cigarettes, many processing steps modify and assemble CDA-based fibers into CFs, which has unknown consequences for degradation rates. The lifetime of CFs in the coastal ocean thus remains a knowledge gap, and until it is resolved, policies will continue to be decided with incomplete data.

In addition to transitioning away from single-use plastics that are frequently mismanaged, the NOAA Marine Debris Program, EU Directive, and UN Treaty propose the adoption of alternative materials that are biodegradable, cost-effective, and more sustainable.<sup>1-3</sup> This aim is expected to lead to an increase in the use and littering of paper CFs globally. However, no study has compared economic and environmental metrics to evaluate the best material to manufacture CFs and guide the design of functional and sustainable CFs. For example, it is unknown how switching from CDA to paper filters impacts the costs to consumers, the amount of greenhouse gas (GHG) emissions and freshwater required to manufacture filters, and the amount of litter in coastal ecosystems.

This initial study addressed two critical knowledge gaps related to the persistence and sustainability of unsmoked CDA and paper CFs. First, unsmoked CDA and paper CFs were incubated in a flow-through seawater mesocosm over six months to determine the first estimates of their lifetime in the coastal ocean. Presumably unsmoked CFs represent the shortest lifetimes for CFs, thus evaluating them constrains the lower limit for their persistence in the coastal ocean. Second, the first comparison of multiple economic and environmental metrics for CDA and paper CFs was conducted. Collectively, these findings contribute to the discussion on the potential effectiveness of policy aimed at curbing CF pollution and offer alternative strategies that may prove more tractable.

## Materials and methods

### Materials

The CFs used in this study included a CDA and paper CF provided by Eastman (labeled CDA 1 and Paper 1, respectively) and three other CFs (one CDA and two paper) cut from three different cigarette brands labeled (CDA 2, Paper 2, and Paper 3, respectively). CDA 1 and Paper 1 included plug wrap, while CDA 2, Paper 2, and Paper 3 included a complete wrapper of plug wrap, tipping paper, and adhesive (Fig. S1†). The CDA filters utilized 3.6 denier per filament (dpf), 31 000 total denier, and Y cross-section acetate tow. The filters cut from cigarettes had comparable lengths, circumferences, and pressure drops (Table S1†).

### Continuous flow natural seawater mesocosm

Experiments were conducted at the Environmental Systems Laboratory (ESL) at the Woods Hole Oceanographic Institution.

The mesocosm system included a 380 L aquaria tank supplied with a continuous flow of seawater by a head tank.<sup>30</sup> Martha's Vineyard Sound (41° 31' 52.0" N, 70° 38' 36.6" W) seawater was pumped to the ESL, tempered to 20 °C, collected in a head tank, and flowed to the aquaria tank with an average flow rate of 190 L h<sup>-1</sup>, yielding a residence time of ~120 minutes. Details of the seawater pumping, filtering, and temperature tempering system have been described previously.<sup>30</sup>

### Sample geometry

All CFs were cut to a length of ~25.4 mm pieces. Before placement in the mesocosm tank, all CFs were massed using a Mettler Toledo AG245 analytical balance (readability of 0.1 mg; repeatability of 0.1 mg). The same analytical balance was used for the entirety of the time series. The geometric dimensions (length and diameter) of each CF were measured with digital calipers (Mitutoyo CD-6" ASX; uncertainty of 0.02 mm, resolution of 0.01 mm). The pertinent properties of the CFs (mass, diameter, length, and density) are included in Table S1.†

### Sample collection for mass loss measurements

The CFs were threaded onto a fishing line with glass beads between each filter. The glass beads separated the CFs and ensured the lines were negatively buoyant. The lines were anchored to the mesocosm tank, suspending the CFs ~5 cm from the bottom of the tank (Fig. S2†). The mass loss experiment was conducted from June 29, 2022, to January 4, 2023. Throughout this period, lines were collected at designated time points. The CFs were removed, photographed, and placed into pre-weighed 2.5 mL polypropylene microcentrifuge tubes filled with Milli-Q water and incubated for ~30 minutes. On occasion, during collection from the tank, a few paper CFs detached from the fishing line and fell to the bottom of the tank. These paper CFs could not be retrieved without unraveling, so these samples were not included in the study. After the incubation, samples and tubes were lightly rinsed with Milli-Q water to remove detritus. Then, samples, in their respective uncapped tubes, were placed open to dry at 60 °C for 48 h in an IsoTemp 637G oven (Fisher Scientific). Samples in their tubes were then removed from the oven, closed, allowed to return to room temperature, and massed.

### Mass loss measurements

Each sample at each time point was evaluated for mass loss in triplicate unless otherwise noted (Table S2†). Mass loss was calculated as the relative mass loss (%) being the difference between the initial mass of the sample ( $m_0$ ) and the mass of the sample ( $m_t$ ) at the time point ( $t$ ) normalized to the initial mass of the sample (eqn (1)). The relative mass loss of each replicate at each time point for each article type is included in Table S3.† Measurements in the continuous flow natural seawater mesocosm have been shown to be reproducible and repeatable over multiple years and seasons.<sup>31</sup>

$$\text{Mass loss(\%)} = \frac{m_0 - m_t}{m_0} (100\%) \quad (1)$$



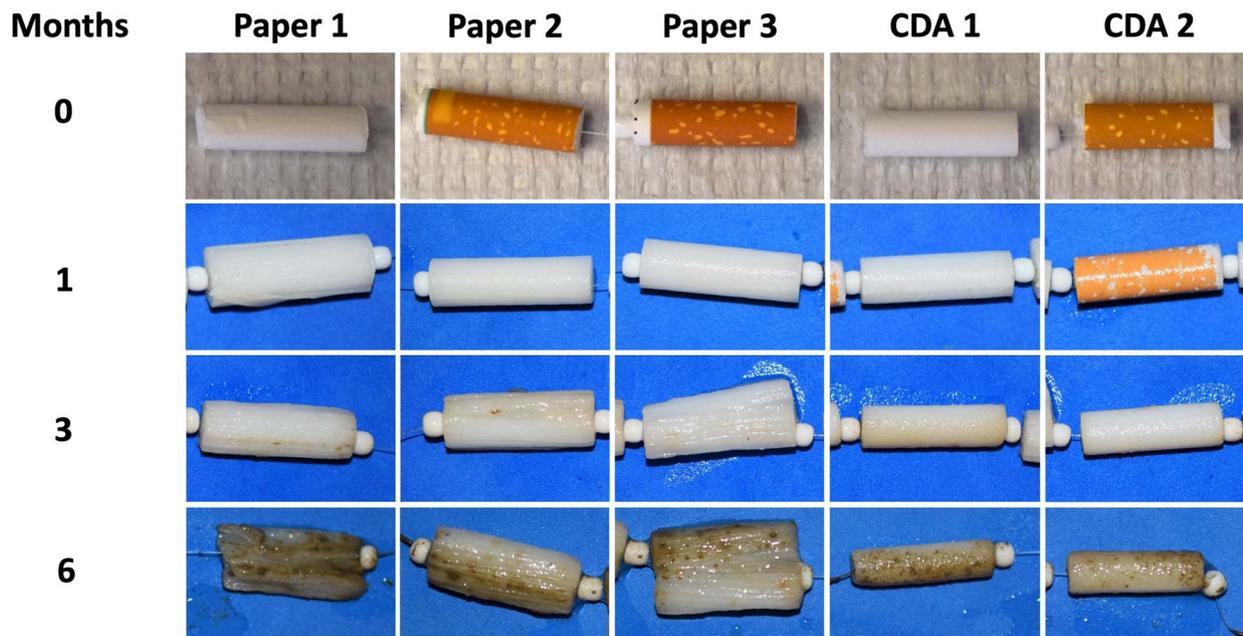


Fig. 1 Representative photographs of each CF initially and after 1, 3, and 6 months in a flow-through seawater mesocosm.

Mass loss is a reasonable measure for the degradation of CDA materials because it is well-established that these materials biodegrade to  $\text{CO}_2$  in the coastal ocean.<sup>29</sup> Additionally, because surface area-to-volume ratio (SA/V) controls the rate of biodegradation, if any mass loss were attributed to physical disintegration (fragmentation),<sup>31</sup> this would only increase the fragments' mass loss rate (eqn (2)). Samples in our system experienced negligible mechanical deformation and abrasion, and were without irradiation; the low flow rates were unable to deflect hanging samples (average flow velocity:  $\sim 0.3 \text{ mm s}^{-1}$ ), indicating a very low shear rate; and the use of seawater filtered to particulate less than  $200 \mu\text{m}$  in combination with low flow rates presumably minimizes any abrasive removal of material. Previous experiments have determined that no mass loss occurred in sterilized controls.<sup>32</sup>

### Surface erosion model

The relative mass loss data (Table S3<sup>†</sup>) was fit to a phenomenological surface erosion model (eqn (2)) in which  $\frac{\partial m}{\partial t}$  is the change in mass with time,  $m$  is the instantaneous mass,  $k_d$  is the specific surface degradation rate,  $A_s$  is the surface area, and  $V$  is the volume.<sup>30,33</sup>

$$\frac{\partial m}{\partial t} = -mk_d \frac{A_s}{V} \quad (2)$$

Eqn (2) was solved for a cylinder of initial length  $l_0$  and initial radius  $r_0$ , yielding eqn (3).

$$\text{Mass loss}(\%) = 100\% \left( 1 - \frac{(l_0 - 2k_d t)(r_0 - k_d t)^2}{l_0 r_0^2} \right) \quad (3)$$

For the mass loss measurements in our continuous flow-through seawater mesocosm,  $k_d$  was considered the apparent

surface erosion rate of the CF due to biodegradation processes. The data sets were fit to eqn (3) using nonlinear least-squares regression after an outlier (ROUT) removal step with a coefficient  $Q$  of 1% to clean the data of any outlying points. Identified outliers are indicated in bold in Tables S2 and S3.<sup>†</sup> All fits had  $R^2 > 0.90$ . All regressions were performed in GraphPad Prism 10.1.0 (264).

Projected environmental lifetimes ( $t_L$ ) were calculated using eqn (4), which was determined from the roots of 1 minus eqn (3), representing the remaining mass of the CF.

$$t_L = \frac{r_0}{k_d} \quad (4)$$

### Sustainability metrics

Material property data was used to calculate sustainability metrics for material efficiency (mass), material cost efficiency, GHG emissions, water usage, and environmental lifetime. Metrics for material cost efficiency, GHG emissions, and water usage were calculated by multiplying the mass of the CF by the specific price, specific GHG emissions, and specific water usage of the CF material. Data was collated from literature sources<sup>11,34</sup> for material properties not measured in this study (Table S4<sup>†</sup>).

## Results and discussion

### Mesocosm incubations and lifetimes

Visual inspection of the CFs throughout the incubation in the flow-through mesocosm revealed similarities and differences between the paper and CDA filters. All CFs experienced biofouling within the first month, and the extent visually increased over time (Fig. 1). This timeline is consistent with other paper and plastic articles studied with the same



experimental system (e.g., films,<sup>29,31,32</sup> foams,<sup>29,31,32</sup> fabrics,<sup>29,32,35</sup> straws<sup>30</sup>). The tipping paper immediately unraveled from the paper CFs, but it took slightly over a month to do so for the CDA CFs. This difference is likely attributed to different adhesives for bonding and sealing the tipping paper and plug wrap to paper and CDA CFs.<sup>36</sup> The tipping paper, plug wrap, and adhesives accounted for ~26–28% of the mass of the CFs (Table S1†). The released tipping paper accumulated at the bottom of the mesocosm tank and was visually observed to degrade as well (Fig. S2†). All filters lost their plug wrap and tipping paper by two months (Fig. 2A), which was reflected by the mass loss between the one- and two-month time points approaching or exceeding the mass of those components (Table S1†). The extent of swelling was more prominent for the paper CFs, an observation likely attributed to differences in construction. Paper CFs are rolled bundles of crimped tissue paper, whereas CDA CFs are crimped bundles of CDA fibers fused by triacetin,<sup>37,38</sup> forming an open porous network (Fig. S3†).

Absolute mass loss (in mg) was calculated monthly throughout the six-month incubation and revealed that the paper CFs had a faster mass loss rate than CDA CFs (Fig. 2A). The paper CFs lost, on average, between 129.3 and 151.0 mg of

mass over six months. Meanwhile, on average, the CDA CFs lost between 69.4 and 89.2 mg of mass. The observation that paper has a faster mass loss rate than CDA is consistent with previous studies comparing paper and CDA drinking straws and films.<sup>30,31</sup>

While absolute mass loss was faster for paper than CDA CFs, relative mass loss (%) was similar between the two products, resulting in similar projected lifetimes in the coastal ocean. The paper CFs lost, on average, between 53.6 and 58.4% of their mass, while the CDA CFs lost, on average, between 49.2 and 49.9% of their mass (Fig. 2B). The mass loss data was fit to a surface erosion model (eqn (3)), with  $R^2$  values ranging between 0.90 and 0.95 for all the CFs (Fig. S4†).

The projected environmental lifetimes of the paper CFs ranged between 1.4 and 1.6 years, while those of the CDA CFs were 1.8 years (Fig. S4†). The ~2-year projected lifetime in the coastal ocean for CDA CFs is notably shorter than the decadal timescale commonly reported by diverse stakeholders in the grey literature.<sup>25,26</sup> Moreover, these findings strongly question the accuracy of statements made in the peer-reviewed literature and by non-governmental organizations that CFs are non-biodegradable.<sup>27,28</sup>

The similar projected lifetimes between the paper and CDA CFs, despite the faster degradation rates of the paper filters, emphasize the need to consider a product's material properties and functional performance when assessing environmental lifetimes and designing for degradation. Paper CFs used ~1.6-fold greater material mass than CDA CFs to achieve their intended performance (Table S1†), such as pressure drop (drag) and filtration efficiency for taste profiles.<sup>11</sup>

This finding is consistent with previous studies on the degradation timelines of consumer drinking straws in the coastal ocean.<sup>30</sup> For example, paper straws had a ~3-fold faster degradation rate than polyhydroxyalkanoate (PHA) and CDA straws. However, because of material and processing limitations compared to PHA and CDA, the paper straws were 2-fold thicker to achieve product performance.<sup>30</sup> Consequently, all the straws tested had similar degradation timelines.<sup>30</sup> It is thus critical to consider both material properties and product morphology during design, thereby optimizing for degradation without sacrificing product performance.

### Material selection using sustainability metrics

Sustainability metrics were calculated for economic and environmental design considerations, revealing notable tradeoffs when evaluating which material is optimal for CF construction (Fig. 3). Specifically, we considered the material mass, material cost, freshwater usage, and GHG emissions required to produce 1000 paper and CDA CFs, as well as the environmental lifetime of each CF in the coastal ocean. Two key findings resulted from this analysis.

First, freshwater usage is the most substantial difference between paper and CDA CFs. Paper CFs may require ~11-fold more freshwater usage to manufacture compared to CDA CFs. Water usage is an important design consideration because the quantity and quality of freshwater resources are diminishing globally,<sup>39,40</sup> non-peer-reviewed studies estimate that the

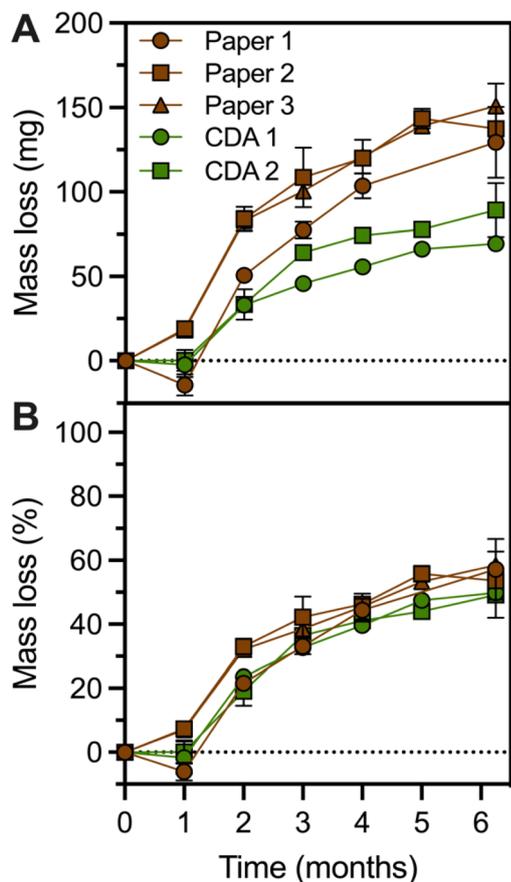


Fig. 2 (A) Absolute mass loss (in mg) and (B) relative mass loss (%) of the paper (brown) and CDA (green) CFs in the flow-through mesocosm throughout the six-month experiment. Data are presented as the mean  $\pm$  standard deviation. In some cases, error bars are smaller than the symbol. Values and replication are presented in Tables S2 and S3.†



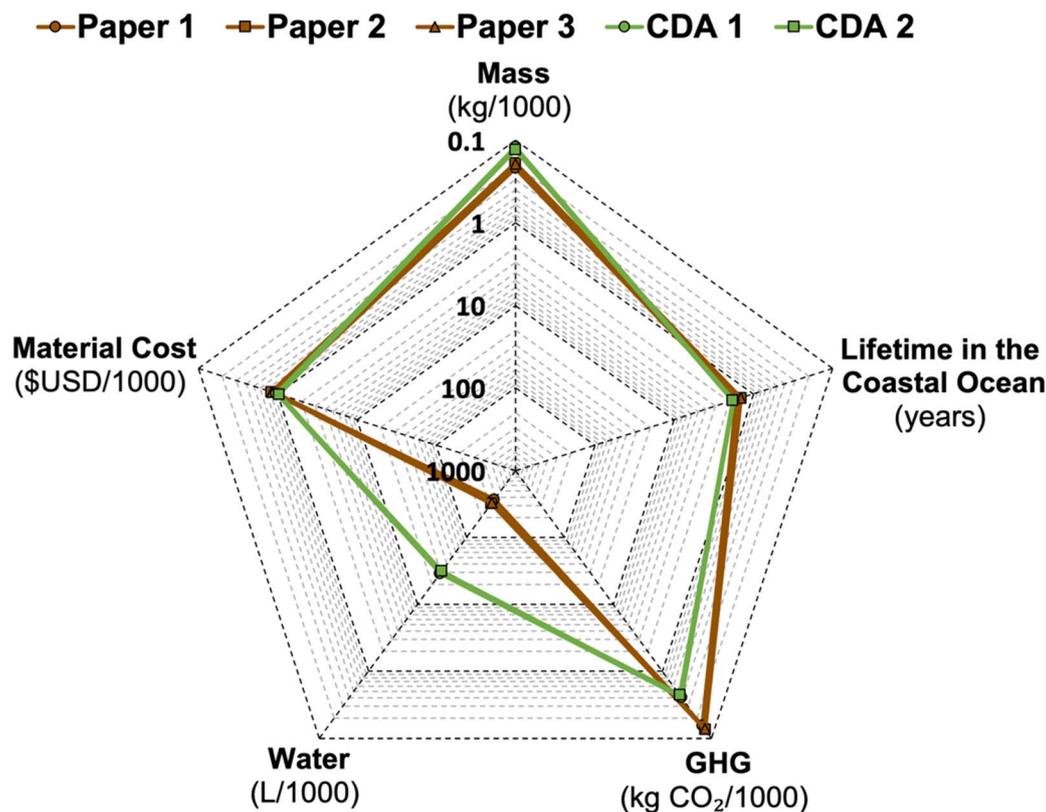


Fig. 3 Comparison of performance, economic, and environmental sustainability metrics for paper (brown) and CDA (green) CF plugs. Metrics for mass (kg), material cost (\$USD), water (L), and GHG emissions (kg CO<sub>2</sub>) are presented to produce 1000 CF plugs. Data to calculate these metrics are presented in Tables S1 and S4.†

societal value of freshwater ecosystems is \$58 trillion USD,<sup>41</sup> and paper and pulp mills pollute these ecosystems and diminish their value.<sup>42</sup>

Second, and relative to the differences in water usage, all other economic and environmental impact metrics generally tracked with one another. Paper is 1.8-fold cheaper (in \$ per kg) than CDA, has 4.8-fold less embodied GHG emissions (in CO<sub>2</sub> per kg), and degrades ~3-fold faster in the coastal ocean (in  $\mu\text{m}$  per year) compared to CDA (Table S4†). However, because of its material properties, 1.6-fold more paper (in kg) is required compared to CDA to achieve acceptable CF performance, thereby closing the gap between these economic and environmental impact metrics for paper and CDA CFs (Fig. 3). This result again emphasizes the need to consider material properties during design because numerous economic and environmental metrics are directly proportional to the amount of material required for a product to be functional. Currently, computer-based software is used to optimize properties for consumer preference, manufacturing, and the cost of CFs,<sup>37,43</sup> presenting an opportunity for including sustainability metrics in these platforms to design more sustainable CFs without compromising performance.

#### Potential Limitations

While this preliminary study provides the first estimates for the lifetimes of paper and CDA CFs in the coastal ocean, it is not

without limitations. The filters used in this study were unsmoked. It is well-documented that cigarette smoke contains a complex mixture of organic compounds, some of which sorb onto the CFs during smoking and may impact microbial functioning in closed system incubations.<sup>4,44</sup> The extent to which smoke residue on the CFs deters microbial degradation in open system incubations reflective of coastal ocean conditions is unknown. Presumably, there is a similar distribution of organics on smoked paper and CDA CFs, suggesting that any hindrance of microbial degradation would be comparable for paper and CDA CFs. Testing this hypothesis should be prioritized moving forward.

This study focuses on biodegradation in the coastal ocean at 20 °C and translation of the findings to colder waters or other ecosystems should be interpreted cautiously. Degradation rates in colder waters are expected to be slower;<sup>45</sup> however, the sensitivity of biodegradation of paper and CDA CFs to temperature is unknown. Moreover, lifetimes in ecosystems where nutrient and water availability is limited may be longer. For example, a study of the degradation of smoked CFs in terrestrial ecosystems with limited nutrients showed steady degradation of up to 80% in five years,<sup>15</sup> which is notably slower than the projected environmental lifetimes of ~1.8 years reported herein for the nutrient replete coastal ocean. Further work should expand this analytical framework to improve projections of lifetimes across diverse environmental conditions.



A final potential limitation is that the study solely focuses on biodegradation, which may overestimate the projected lifetimes of paper and CDA CFs in the coastal ocean. It is well established that physical and photochemical processes can contribute to the degradation of paper and CDA, resulting in shorter lifetimes. For example, mechanical abrasion of paper and CDA filters can result in fragments with a higher SA/V than the parent article. Because SA/V is a key control of biodegradation,<sup>31</sup> these fragments likely degrade even faster than the parent filters. Our study design did not capture this process because there was insufficient shear in the mesocosm to cause gross fragmentation.<sup>31,32</sup> Moreover, sunlight exposure can directly mineralize CDA to CO<sub>2</sub> and can initiate chain scission reactions that accelerate biodegradation,<sup>35</sup> neither of which were considered in this study. Considering the broader array of degradation pathways in future studies may lead to shorter lifetime estimates.

### Implications for policy and paths forward

All stakeholder groups agree that tractable solutions to the plastic pollution crisis are needed. However, there is little consensus on the best path to achieve this goal. One path sought by government agencies and legislation (*e.g.*, the NOAA Debris Program,<sup>2</sup> the EU Directive 2019 N. 904,<sup>1</sup> and the UN Treaty on Plastic Pollution<sup>3</sup>) is to ban single-use plastics and replace them with alternatives that do not persist in the environment. Regarding CFs, the most frequently littered plastic item on the planet and one of the most common items collected in beach surveys,<sup>12,13</sup> adherence to such legislation implies switching from CDA to paper CFs. While additional research is needed (see Potential Limitations section above), the results from this initial study indicate that switching from CDA to paper filters will not reduce CF litter in coastal areas because these products share similar lifetimes. Moreover, the switch would come with substantial costs related to the freshwater footprint of manufacturing, while only marginal savings of GHG emissions are likely to be made.

In no way do the findings of this study support smoking cigarettes or littering CFs. Moreover, this study does not contribute to the ongoing debate about the value of cigarette filters from a public health perspective.<sup>46</sup> Nevertheless, one in five adults routinely smoke cigarettes, and the global population is increasing faster than the decline in smoking rates,<sup>14</sup> ensuring that CFs will continue to be the most littered item on the planet for the foreseeable future. Therefore, all stakeholders should prioritize tractable solutions to this ubiquitous pollutant.

We offer three alternative strategies to initiate a discussion about approaches that may prove more effective than adopting a policy that simply switches filter materials from CDA to paper. First, design for degradation. Invest in research that identifies alternative materials that simultaneously meet the consumer performance targets and degrade even faster in the environment, building on research initiated in the 1990s.<sup>7</sup> This includes the identification of economically viable and safe photocatalysts to directly mineralize the filter material to CO<sub>2</sub>

and stimulate microbial activity (*i.e.*, alternatives to titanium dioxide<sup>35</sup>), as well as embedded enzymes to rapidly degrade the filter material upon wetting.<sup>47</sup> Given the interdisciplinary nature of this problem and the need to scale solutions quickly, funding bodies should embrace industry-academic partnerships.<sup>48</sup>

Second, modernize waste management infrastructure and evaluate waste valorization strategies. Paper and CDA are compostable materials, yet industrial and home composting facilities are insufficient in most of the world, potentially contributing to poor end-of-life management for CFs. Evaluating and adopting such infrastructure could reap benefits that extend far beyond better management of CF waste, including minimizing methane emissions related to food waste disposal in landfills.<sup>49</sup> Forward looking, the valorization of CF waste into useful products should be explored.<sup>50–53</sup>

Finally, educate consumers. No matter the material CFs are made of, until consumer behavior changes and littering rates decrease, CF litter will continue to pose risks to human and ecosystem health. Educating consumers so that they understand the beginning- and end-of-life of the products they use and the waste they generate is a critical step needed to mitigate the risks of plastic pollution.

### Data availability

The data supporting this article have been included as part of the ESI.†

### Conflicts of interest

The authors declare the following competing financial interest(s): C. P. W and C. M. R acknowledge funding and scientific support from Eastman, a manufacturer of biodegradable plastics. The remaining authors declare no competing interests.

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