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# Predicting environmental biodegradability using initial rates: mineralization of cellulose, guar and their semisynthetic derivatives in wastewater and soil

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Microplastic pollution is a growing concern, and natural materials are being increasingly sought as plastic alternatives. Semisynthetic biopolymers occupy a grey area between natural and synthetic materials and are often presented as green alternatives to conventional plastic. They can be water-soluble or insoluble, and are ubiquitous in commercial products as thickeners, films, filters, viscosity modifiers and coatings. This work compares the mineralization kinetics of cellulose, guar and several of their commercialized derivatives using a simple pseudo first-order kinetic model to extrapolate half-lives and lifetimes, while identifying the levers that influence the mineralization rates of these ubiquitous materials. Industrial composting rates were consistently faster than those of wastewater. While partially substituted biopolymers exhibited measurable degradation, kinetic analysis revealed this effect could be entirely accounted for by the fraction of unsubstituted biopolymer. Surprisingly, the initial rates of highly substituted biopolymers exhibited persistence on par with conventional plastics over the experimental durations studied.

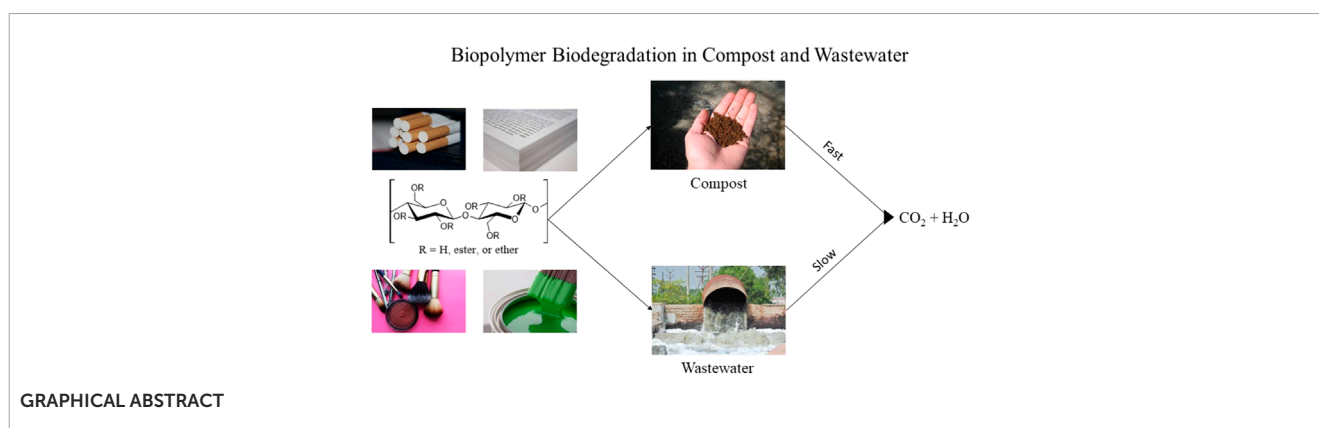
## KEYWORDS

modified celluloses, cellulose acetate, carboxymethyl cellulose, hydroxypropyl methylcellulose, hydroxyethyl cellulose, ethyl cellulose, green chemistry, environment

## Introduction

The use of persistent chemicals and materials has contributed to environmental pollution (Li et al., 2016; Andrady, 2017), biocide resistance (Stewart et al., 2001; Mathur and Singh, 2005; Green, 2007; Green and Owen, 2011) and hazards such as hormone disruption, (Hoffman et al., 2017), cancer (Vaughn et al., 2013), and birth defects (Ngo et al., 2006). A principal goal of the bioeconomy is the conversion of biological resources into products

**Abbreviations:** CMC, carboxymethyl cellulose; CA, cellulose acetate; HPMC, hydroxypropyl methylcellulose; HEC, hydroxyethyl cellulose; EC, ethyl cellulose; HPG, hydroxypropyl guar.



that are competitive with those derived from nonrenewable sources, thereby reducing our reliance on “fossilized” carbon feedstocks (Philp et al., 2013; Bugge et al., 2016; Kim et al., 2016; Matharu et al., 2016; Buckley et al., 2017). Synthetic modification of naturally-derived biochemicals, including biomacromolecules, can produce semisynthetic materials with value-added properties that are not present in the parent biomolecule (Debeaufort et al., 1998).

However, while adding commercial viability, these synthetic modifications may also reduce the biodegradability of the resulting semisynthetic material by orders of magnitude (Wirick, 1974; Rivard et al., 1992; Buchanan et al., 1993; Puls et al., 2011). A primary example of this reduced degradability is cigarette filters which are made of cellulose acetate, a mostly natural biomacromolecule, that is a leading visible source of plastic pollution, especially in large bodies of water (Driedger et al., 2015).

Other common plastics such as polyvinyl alcohol, used to make laundry or dish pods, polyethylene, and polyethylene terephthalate (Morét et al., 2010), are also reportedly biodegradable under unique conditions, such as exposure to specific fungi, rare bacteria (Yoshida et al., 2016), or wax moth larvae (Bombelli et al., 2017). However, these examples illustrate a critical distinction between a material’s possible capacity for biodegradation versus the extent to which mineralization actually occurs in natural environments (Kint and Muñoz-Guerra, 1999; Kubowicz and Booth, 2017). Experimental determinations of biopolymer degradability are highly variable depending on the media, inoculum, and conditions employed (Puls et al., 2011), and natural environments generally do not mimic the conditions found in the lab. The incomplete breakdown of plastics generates microplastics that accumulate in the human body, foods, and the natural environment (Andrady, 2011). While previous studies have examined the thermochemical degradation of cellulose (Osman et al., 2022), and biodegradability of natural and semisynthetic celluloses (Kubowicz and Booth, 2017; Torgbo and Sukyai, 2020; Frank et al., 2021; Polman et al., 2021; Yadav and Hakkarainen, 2021), investigations comprehensively evaluating the effect of functional group, medium (e.g., wastewater, compost) and degree of substitution (DS) using standard degradation protocols are needed.

In this work, we compared the mineralization rates of semisynthetic polysaccharides to unmodified cellulose and guar in aerobic wastewater and soil compost environments (hydroxypropyl methylcellulose (HPMC), carboxymethyl cellulose (CMC), hydroxyethyl cellulose (HEC), ethyl cellulose (EC), cellulose acetate

(CA), hydroxypropyl guar (HPG)); Figure 1). These substances were selected due to their widespread use and commercial viability. A respirometry method was utilized to measure the mineralization of the cellulosic materials in the wastewater and compost matrices. Pseudo first-order kinetic analyses enabled the quantitative determination of relative biodegradability rates, half-lives and predicted lifetimes. Similar approaches (Hart-Cooper et al., 2015a; Hart-Cooper et al., 2015b; Flynn et al., 2020), which have been used to quantify chemical reaction rates, enables rapid determination of biodegradation half-lives, and their extrapolation to material lifetimes, typically defined as five half-lives, or ~97% conversion. The effect of functional group type (e.g., ether, ester, unmodified) on degradation was determined. A kinetic model was used to deconvolute how the degree of polymer substitution influenced degradation rate. Comprehensively, these metrics provide a facile, robust, and practical way to assess and characterize the biodegradability of semisynthetics. A kinetic framework for understanding the effects of functional group derivatization, degree of substitution and media is presented herein.

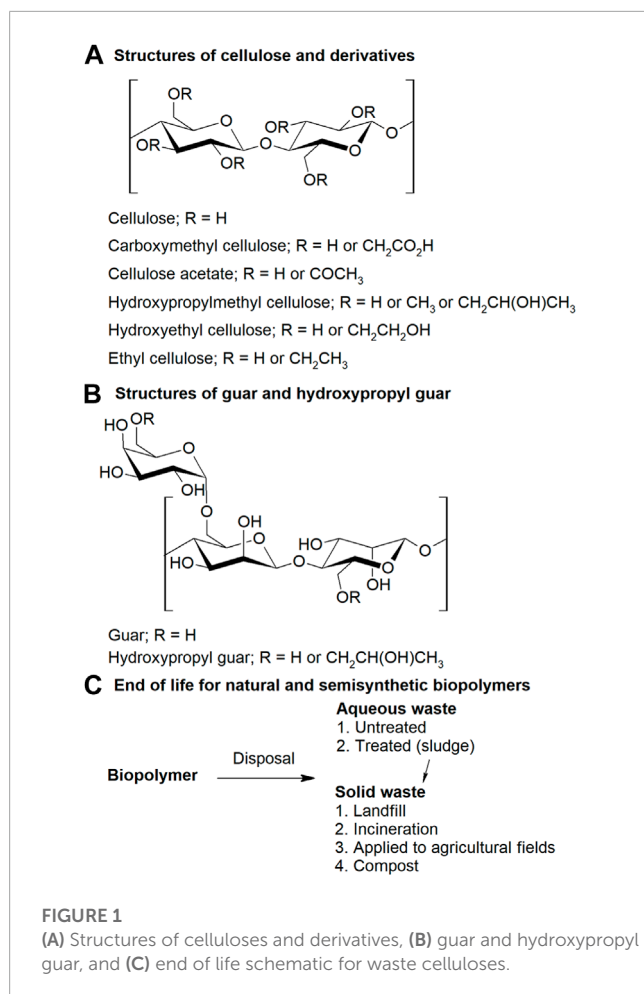
## Materials and methods

### Materials

Chemicals were acquired from commercial sources (Ashland: HEC, CMC; DuPont: EC, HPMC; Sigma Aldrich: CA; Rita Corp: HPG; Avicell: cellulose; Rhodia: Guar; Supplementary Table S1), with grades based on commercial availability. Molecular weight (MW) and degree of substitution (DS) are indicated in Table 1. Compost (Miracle-Gro Garden Soil All Purpose) was purchased from a local hardware store. The compost contained 0.09% total nitrogen, 0.05% available phosphate ( $P_2O_5$ ), and 0.07% soluble potash ( $K_2O$ ). Moisture content of the compost was determined using a moisture analyzer (Ohaus Model MB25, Switzerland).

### Respirometry

Respiration rates obtained via monitoring  $CO_2$  concentrations using a respirometer (Micro-Oxymax, Columbus Instruments, Columbus, OH). Since there is a baseline amount of  $CO_2$  produced by the compost or wastewater, the  $CO_2$  production attributable to



each treatment sample was determined by subtracting the baseline production (control) from the total CO<sub>2</sub> production detected in the respirometry chambers containing the sample material being tested. A first order rate constant was found for each data set using an exponential fit and used to calculate projected half-lives and lifetimes.

## Soil compost

The compost was prepared by sieving (14 mesh), adjusting the moisture content to 60% (w/w), and equilibrating overnight in a plastic bag. Polymer samples were prepared by milling to 40 mesh per ASTM D 5338 (ASTM D5338 et al., 2003). Glass respiration chambers (500 mL) were prepared by loading 40.0 g of compost and 0.500 g of milled sample, measured using an analytical balance. The contents of each respirometer chamber were thoroughly mixed before attaching to the respirometer via tubing. The respiration chambers were kept in an enclosed incubator (60°C ± 0.5°C) to maintain a stable temperature based on industrial composting standards. The experimental design included three replicates for each treatment. Four controls containing only compost were also included in each experiment for baseline determination. The net increase in CO<sub>2</sub> production for each treatment was calculated. The standard deviations for individual time points were 5%–15%.

## Wastewater

Respirometry determination in wastewater closely followed ISO 14851 (International Organization for Standardization, 2019). A standard test medium was prepared, composed from 10 mL solution A (8.5 g KH<sub>2</sub>PO<sub>4</sub>, 21.75 g K<sub>2</sub>HPO<sub>4</sub>, 33.4 g Na<sub>2</sub>HPO<sub>4</sub>×2H<sub>2</sub>O, 0.5 g NH<sub>4</sub>Cl; diluted to 1,000 mL), 1 mL each of solutions B (22.5 g MgSO<sub>4</sub>×7H<sub>2</sub>O; diluted to 1,000 mL), C (36.4 g CaCl<sub>2</sub>×2H<sub>2</sub>O; diluted to 1,000 mL), and D (0.25 g FeCl<sub>3</sub>×6H<sub>2</sub>O; diluted to 1,000 mL), and the resulting solution further diluted to a total volume of 1,000 mL with DI water. The activated sludge, obtained from East Bay Municipal Utilities District (Oakland, CA), was filtered through a 16-mesh sieve. Activated sludge (0.5 mL) with a solids content of 1.66% was added into each chamber to achieve a solids content of 830 mg dry weight. The milled sample (0.500 g) was added to the respirometer chambers and the contents were agitated before attaching them to the respirometer via tubing. Respirometry was carried out in an incubator maintained at 30°C (±0.5°C). CO<sub>2</sub> accumulation in the headspace of each chamber was continuously recorded. Each sample was run in triplicate with four controls to measure the baseline CO<sub>2</sub> production of the activated sludge. The net CO<sub>2</sub> production for each sample was used to determine the mineralization rates.

The carbon content of each sample was calculated and used to determine the theoretical yield of CO<sub>2</sub> if 100% of the sample carbon was mineralized. The percent mineralization of each sample was calculated by dividing the cumulative CO<sub>2</sub> produced by the theoretical CO<sub>2</sub> and multiplying by 100.

## Results and discussion

### Mineralization of cellulose and derivatives

We compared the biodegradability of natural cellulose to commercial semisynthetic derivatives. Semisynthetic derivatives were selected based on commercial utility and exhibited varying degrees of substitution (DS). Biodegradation was monitored by measuring accumulated CO<sub>2</sub> relative to a baseline that does not contain test material (Figure 2).

The accumulation of CO<sub>2</sub> was a direct measurement of the thermodynamic final product of biopolymer consumption and is a useful metric for assessing full biodegradation, as it measures only the final oxidation product of carbon. Like many methods to assess biodegradation, this approach is not a perfect model for environmental biodegradation, in that it does not include continual inputs of organic matter, and has a limited ability to determine which functionalities are being respired. Monitoring oxygen and starting material consumption alone does not account for the possibility that starting material could be consumed, yet not entirely mineralized to CO<sub>2</sub>. Plots of the percent mineralization versus time fit well to pseudo first-order kinetic models using Eq. 1 and shown in Figure 3):

$$[A] = [A]_0 e^{-k_{\text{obs}} t} \quad (1)$$

Where [A] represents the concentration of a substance at a given time [A]<sub>0</sub> is the starting concentration of the substance, *k*<sub>obs</sub> is the pseudo first-order rate constant and *t* represents time.

**TABLE 1 Kinetic parameters, projected half-lives, and projected lifetimes of biopolymer mineralization.**

| Substance (DS = degree of substitution molar ratio, provided by supplier) | Degree of substitution (DS, molar ratio) | Medium           | $k_{obs}$ (days <sup>-1</sup> ) | $k_{rel}$ | Projected half-life (days) | Projected lifetime (~97% mineralization) |
|---------------------------------------------------------------------------|------------------------------------------|------------------|---------------------------------|-----------|----------------------------|------------------------------------------|
| Cellulose                                                                 |                                          | Soil, 60°C       | 0.040                           | 100       | 17                         | 3 months                                 |
|                                                                           | n/a                                      | Wastewater, 30°C | 0.0031                          | 7.7       | 230                        | 3 years                                  |
| Carboxymethyl cellulose (CMC)                                             | 0.7                                      | Soil             | 0.0082                          | 20        | 85                         | 1 year                                   |
|                                                                           |                                          | Wastewater       | 0.0012                          | 3.1       | 560                        | 8 years                                  |
| Cellulose acetate (CA)                                                    | 3.0                                      | Soil             | 0.0012                          | 3.0       | 580                        | 8 years                                  |
|                                                                           |                                          | Wastewater       | 0.00045                         | 1.1       | 1,600                      | 22 years                                 |
| Hydroxypropyl methylcellulose (HPMC)                                      |                                          | Soil             | ≤0.001                          | ≤3        | ≥690                       | ≥10 years                                |
|                                                                           | 0.9 (HP),0.3 (Me)                        | Wastewater       | ≤0.0004                         | ≤1        | ≥1730                      | ≥24 years                                |
| Hydroxyethyl cellulose (HEC)*                                             | 1.5                                      | Soil             | ≤0.001                          | ≤3        | ≥690                       | ≥10 years                                |
| Ethyl cellulose (EC)*                                                     | 3.0                                      | Soil             | ≤0.001                          | ≤3        | ≥690                       | ≥10 years                                |
| Guar*                                                                     | n/a                                      | Soil             | 0.085                           | 210       | 8                          | 1 month                                  |
| Hydroxypropyl guar (HPG)*                                                 | 1.3                                      | Soil             | 0.0038                          | 9         | 184                        | 3 years                                  |

\*Material was evaluated only in soil, not wastewater.

Determination of first-order rate constants ( $k_{obs}$ ) enabled the calculation of reaction half-lives and projected material lifetimes, which represent five half-lives (Figure 3; Table 1).

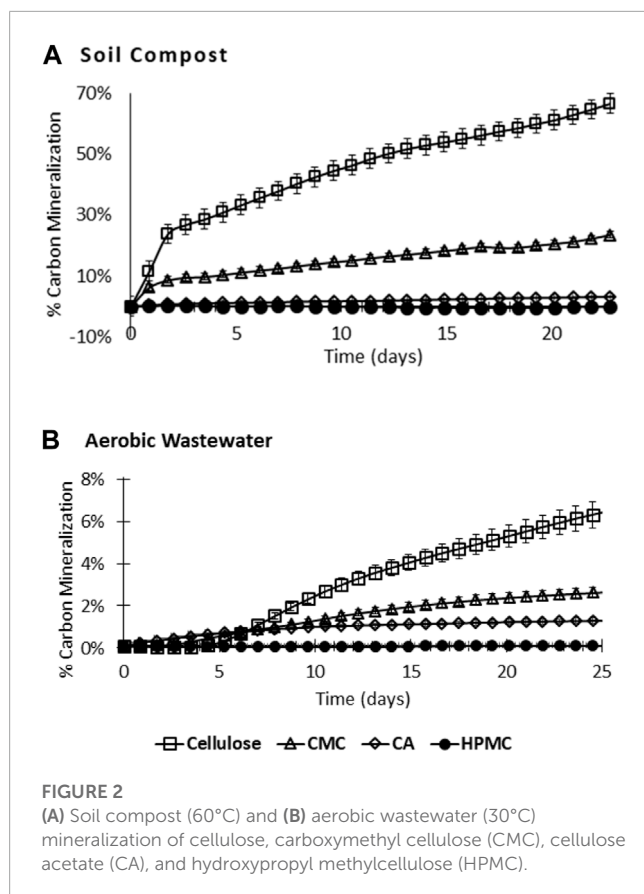
A perpetual challenge in biodegradation studies is the high variability observed between inoculum compositions. To address this issue in our measurements, we calculated relative mineralization rates,  $k_{rel}$ , which represents the ratio of a given biodegradation rate divided by the minimum rate observed in the experimental set, shown in Eq. 2. This value ( $k_{rel}$ ) provides a robust comparison of an inoculum's pseudo first-order biodegradation rate relative to a positive control (cellulose). In our hands, relative biodegradation rates spanned up to two orders of magnitude, indicating that HPMC, EC and HEC are at minimum 100 times more persistent than cellulose.

$$k_{rel} = \frac{k_{fast}}{k_{slow}} \quad (2)$$

In soil compost (60°C), cellulose mineralized rapidly at an average rate of 3% per day for the first 23 days, with an extrapolated lifetime of 3 months. Carboxymethyl cellulose (CMC; DS = 0.7) degraded slightly slower, on average 1% per day (first 23 days) with a lifetime of 1 year. This projected lifetime is remarkably close to those reported in previous studies of CMC, DS = 0.7 (Nie et al., 2004). Cellulose acetate (DS = 3) mineralized an order of magnitude more slowly than the CMC sample (0.1% per day; lifetime of 8 years, assuming no induction period). Given the high DS of CA, this observed mineralization rate is lower than what is typically observed with less substituted analogues (Buchanan et al., 1993; Puls et al.,

2011; Yadav and Hakkarainen, 2021). The remaining synthetically modified celluloses (HPMC; HEC; EC) did not measurably degrade. We attribute the higher degradability rate of CA compared to EC to CA's ester bonds, which are more rapidly cleaved than an ethyl ether bond. This result led us to establish a maximum estimate of their biodegradation rates based on detection limits, which were comparable to the minimum measurable biodegradation rate observed in compost ( $k_{obs} \leq 0.001$  days<sup>-1</sup>, lifetime of ≥10 years, assuming no induction period).

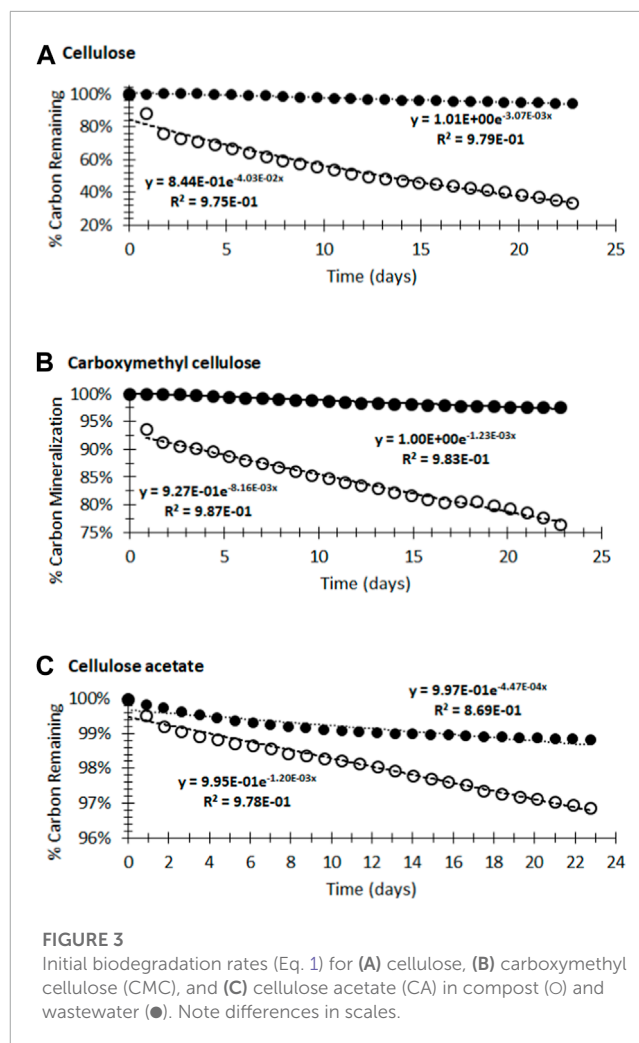
A polysaccharide's degree of substitution (DS) represents, on average, how many hydroxyl moieties in each monomer are functionalized. High degrees of substitution are associated with lower degradation rates compared to the unmodified polymer, an effect that can be attributed to, for example, reduced enzyme affinity for the modified substrate, and bulk polymer properties, such as reduced water solubility or bioavailability. To determine whether the inhibiting effects of synthetic modification on mineralization are general to other polysaccharides, guar and hydroxypropyl guar (HPG; DS 1.3) were evaluated (Supplementary Figure S1). Guar mineralized at approximately twice the rate of cellulose and 22 times faster than HPG. Unlike HPMC, HEC, and EC, the mineralization of HPG was fast enough to be measured. We attribute this effect to the fact that a monomer of guar contains three sugar subunits: one galactose and two mannoses. DS = 1.3 implies that on average, at least one subunit of sugar would not be derivatized per guar monomer. The observed small levels of mineralization can be attributed to microbial consumption of unmodified galactose and



mannose. Mineralization rates in soil compost decreased in the following order: guar > cellulose > CMC > HPG > CA > HPMC, HEC, EC.

A subset of cellulose derivatives representing a range of soil mineralization rates were additionally evaluated in wastewater (Table 1). Relatively low-temperature wastewater (30°C) mineralization rates were approximately an order of magnitude slower than in higher temperature soil compost (60°C) conditions, except for cellulose acetate, where the magnitude of the effect was smaller,  $k_{rel} \sim 3$ ; Figure 2B. Thermal effects on reaction rate are likely significant contributors for these differences. Within viable temperature ranges, most biological reaction rates increase by a factor of 2-3 with every 10°C increase in temperature (Reyes et al., 2008). The slower mineralization rate in wastewater may be attributed to the low microbial density and reduced diversity. Microbial strains present in compost will also be different than those in wastewater. All these factors impact the overall mineralization rate and can account for the observed differences between wastewater and compost conditions.

An apparent induction period of several days occurred for cellulose, suggesting microbial acclimation. Relative mineralization rates among different materials followed the same trend as observed in soil. As with compost, HPMC did not measurably mineralize to CO<sub>2</sub> and was assigned a maximum estimate for its biodegradability rate constant based on detection limits ( $k_{obs} \leq 0.0004 \text{ days}^{-1}$ , lifetime of  $\geq 24$  days, assuming no induction period). Unlike soil compost, wastewater treatments included no additional carbon source aside



from the test substance, allowing for an improved signal to noise ratio and a lower maximum rate estimate.

A probable biodegradation mechanism for substituted cellulose derivatives is depicted in Figure 4. A substituted fragment (1) undergoes slow deprotection ( $k_1$ ) to afford an unsubstituted, cellulose fragment (2), which is quickly depolymerized ( $k_2$ ) and the resulting monomers (4) rapidly mineralized to CO<sub>2</sub> ( $k_5$ ). A conceivable alternate pathway ( $k_3k_4$ ) involving depolymerization of 1 followed by deprotection of the substituted monomer (3) is thought to be prohibitively slow, as many semisynthetic celluloses with high DS are relatively inert to cellulases ( $k_2 \gg k_3$ ) (Puls et al., 2011). The slow mineralization rates of highly substituted celluloses, combined with the simultaneous decreases in molecular weight and DS observed during semisynthetic biodegradation (Gu et al., 1993; Puls et al., 2011; Yadav and Hakkarainen, 2021) are consistent with the  $k_1k_2$  pathway, where  $k_1$  is rate-limiting.

$$k_{obs} = k_a f_a + k_b f_b \quad (3)$$

Intrinsic chemical reactivity rates predict that biodegradability would proceed more rapidly with esterified CA than the alpha-ether CMC. The observed inversion ( $k_{obs}(\text{CA}) < k_{obs}(\text{CMC})$ ) could result from enzymatic catalysis or differences in DS (3

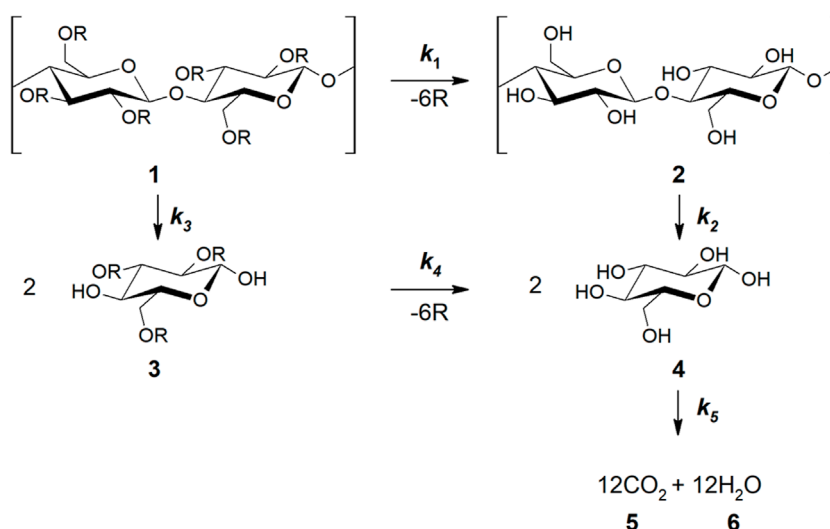


FIGURE 4

General mechanism for the mineralization of cellulose and semisynthetic derivatives. R represents a semisynthetic group as specified in Figure 1A. This framework is consistent with prior reports (see reference 16).

TABLE 2 Kinetic parameters of cellulose and carboxymethyl cellulose (CMC; DS 0.7) in soil and wastewater conditions.

| Substance (DS = degree of substitution)     | Medium (°C)    | $f_A$ | $f_B$ | $k_a$         | $k_b$  | $k_b/k_a$ |
|---------------------------------------------|----------------|-------|-------|---------------|--------|-----------|
| Cellulose                                   | Soil, 60       | 0     | 1     | n/a           | 0.040  | n/a       |
|                                             | Wastewater, 30 |       |       | n/a           | 0.0031 | n/a       |
| Carboxymethyl cellulose (CMC)<br>(DS = 0.7) | Soil, 60       | 0.64  | 0.36  | $\leq 0.0010$ | 0.0220 | $\geq 22$ |
|                                             | Wastewater, 30 |       |       | $\leq 0.0001$ | 0.0031 | $\geq 31$ |

and 0.7, respectively). Nonlinear curve fitting enabled us to determine whether differences in DS alone could account for this anomaly. Eq. 3 (Hart-Cooper et al., 2015a), which weights the mineralization rates of derivatized ( $k_a$ ) and nonderivatized ( $k_b$ ) cellulose with their respective carbon mol fractions ( $f_A$ ,  $f_B$ ), was used to deconvolute these steps separately when incorporated into Eq. 4 (Supplementary Material). When  $DS > 1$ ,  $f_A = 1$  and  $f_B = 0$ , reducing Eq. 3 to Eq. 1, where  $k_{obs} = k_a$ . In the case of cellulose,  $f_A = 0$ ,  $f_B = 1$  and  $k_{obs} = k_b$ . Fitting Eq. 3 to the mineralization curves of CMC revealed  $k_b$  values that were comparable (wastewater) or slightly less (compost) than that of cellulose (Table 2; Supplementary Figure S2).

Of the partially substituted CMC copolymer, its 30% unmodified cellulose portion could readily account for the observed mineralization rate in both soil and wastewater. An upper limit for  $k_a$  was determined to be at least an order of magnitude smaller than the rate of cellulose mineralization ( $k_b$ ), implying environmental lifetimes on the order of years for the carboxymethyl-substitution portion of CMC (DS 0.7), which is comparable to the fully substituted semisynthetics that were tested. Upper estimates of CMC deprotection rates ( $k_a$ ) were comparable to  $k_{obs}$  of CA mineralization, which is consistent with the notion that ester hydrolysis is at least as fast as carboxymethyl cleavage.

A lower DS thus accounted for the faster mineralization of CMC relative to CA.

## Conclusion

Commercial grade semisynthetic celluloses, which are common ingredients in consumer products especially single-use items, packaging, and cigarette filters, mineralized up to 100 times more slowly than cellulose when degraded in soil compost. Semisynthetic derivatives, CA and CMC, afforded measurable degradation rates in this typical garden soil compost, as well as in aerobic wastewater, while no measurable degradation was observed with HPMC, EC, and HEC. A similar effect was observed with guar and HPG. Biodegradation rates decreased in the order guar > cellulose > CMC > HPG > CA > HPMC, HEC, EC. Mineralization in wastewater (30°C) was generally an order of magnitude slower than compost (60°C). These results are consistent with a biodegradation mechanism involving rate-limiting deprotection to yield the natural polymer, which is rapidly mineralized. Deprotection rates for semisynthetic biopolymers are at least one order of magnitude slower than mineralization of the natural biopolymer (cellulose or guar). This observation implies that highly synthetically substituted

biopolymers may, when water-soluble, persist invisibly for up to decades in the natural environment. Based on initial rates, this degree of persistence is comparable to some conventional plastics (Chamas et al., 2020).

Respirometry experiments have limitations. Experiments are conducted in isolated containers under controlled conditions. Biodegradation rates are dependent upon the selected conditions, temperature, microbial population levels and compositions. Mineralization rates in an uncontrolled environment would vary considerably with temperature fluctuations, the introduction of organic matter, moisture levels, oxygen availability, the activity of microbial compositions. For example, organic matter that could biodegrade relatively quickly in some environments may persist for centuries in a dry, cold, or toxic environment, such as a landfill (Bogner, 1990). Additionally, predicting material lifetimes by extrapolation carries the assumption that the entire degradation process will follow a pseudo first-order relationship. Mineralization rates may deviate from first-order kinetics and slow down, or even accelerate, later in the degradation process. Nevertheless, determining the relative mineralization rates in environments to which materials may be most likely exposed, and providing an estimate of their persistence, is important in anticipating the extent to which these materials will impact the environment in the long-term.

While longer duration studies are needed, it is conceivable that highly substituted biopolymers may exhibit comparable persistence to conventional plastics. Perhaps of most concern is that with the degradation of the polymer backbone, the substituted synthetic groups could be released into the environment as a form of microplastic. Microplastics are a concern due to the many health conditions associated with their accumulation in the human body and their discovery in most ecosystems (Hasan et al., 2021; Osman et al., 2023).

Despite their benefit as renewable materials, the slow mineralization kinetics of these semisynthetic biopolymers highlight the need and opportunity to develop more rapidly degradable compositions for non-durable products where biodegradation is desired. Environmental pollution resulting from highly substituted semisynthetic biopolymers may be greatly reduced by replacing them with materials with less or no substitution. Additional research on non-synthetic modification of material properties is likewise needed. For example, hydrothermal processing is a simple, low cost, and environmentally friendly process for modifying the properties of starches, without the use of chemical treatments and resulting hazardous waste (Dutta and Sit, 2022).

## Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

## Author contributions

WH-C: Conceptualization, Data curation, Formal Analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing–original draft, Writing–review and editing. NK: Conceptualization, Data curation, Formal Analysis, Methodology, Writing–review and editing. AK: Data curation, Investigation, Methodology, Writing–review and editing. LT: Conceptualization, Methodology, Validation, Writing–review and editing. GG: Investigation, Methodology, Project administration, Resources, Writing–review and editing. JC: Methodology, Software, Validation, Visualization, Writing–review and editing. KJ: Conceptualization, Investigation, Methodology, Writing–review and editing. WO: Project administration, Resources, Writing–review and editing.

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## Conflict of interest

Author KJ was employed by Method Products.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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## Supplementary material

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fmats.2023.1331308/full#supplementary-material>

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