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Abstract

Purpose: Cigarette filters degrade over time due to the action of weathering elements such as sun, wind, rain, and mechanical action in the environment. Biodegradation of common cellulose derivatives in different laboratory, natural and man-made environments can be solely a result of biodegradation or a combination of several processes, such as chemical, enzymatic hydrolysis, microbial population and their action including oxidation. This study aimed to assess the biodegradation of cigarette filters through a scientific investigation.

Methodology: This research investigated the biodegradation of cigarette filters in the natural soil environment through two methodologies: (i) the Cage Method and (ii) the Litterbag Method. to understand the fate of cigarette filter in the environment and the time required for complete biodegradation of cigarette filter mass.

Findings: Real-time biodegradation of cellulose acetate (CA) cigarette filter and combined material filter (CMF) was studied using cage method which demonstrated a 100% mass loss for CA cigarette filter and CMF in 33 months and 21 months respectively confirming that cigarette filters complete degrade in natural environment scenario. Biodegradation of cigarette filters in soil under controlled condition was studied using litter bag method which revealed a mass loss of 100% for CMF in 15 months. Similar studies on DE-Tow™ filters, CA filters and Saal wood revealed 100%, 99.9% and 45% mass loss respectively in 27 months which clearly demonstrates that cigarette filters biodegrade much faster compared to the known biodegradable material Saal wood. Furthermore, absence of residual secondary cellulose acetate in resulting biomass post biodegradation was confirmed using Gel Permeation Chromatography (GPC) and NMR analysis.

Unique Contribution to Theory, Practice and Policy: This research underscores the significance of the biodegradation environment and the chemical composition of cigarette filters, along with their degree of substitution, as crucial elements influencing the biodegradation process of these filters. The cigarette filter is constructed from secondary cellulose acetate, which differs from primary cellulose acetate in both physical and chemical characteristics. Contrary to popular belief, this study provides definitive evidence that cigarette filters made from secondary cellulose acetate are indeed biodegradable. The findings of this study will serve as a valuable resource for the scientific community, regulatory bodies, and manufacturers alike.

***Keywords*:** *Biodegradation, Natural Environment, Simulated Soil Environment, Cellulose Acetate (CA), Cigarette Filter*

JEL Code of Classification: *Q 01, Q20, Q26, Q28, Q53, Q55*

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INTRODUCTION

Cellulose acetate (CA) is a widely used natural polymer that is a derivatised polymer. Its applications vary from textile industry to plastic films, packaging, and cigarette filter tows. It is an eco-friendly material and majorly sourced from cellulose present in wood or cotton linters through reaction with acetic acid (Biodegradation of cellulose acetate, Chemical book, 2023,). Cellulose is a natural biopolymer and inherently degradable in suitable natural environment. In cigarette industry, secondary cellulose acetate (SCA) or commonly called as cellulose diacetate is used for manufacturing cigarette filter tow.

Cellulose acetate is defined in the Indian Standard IS 10335 entitled "Tobacco and Tobacco Products – Glossary of Terms" as "A secondary cellulose acetate commonly known as cellulose acetate (CA) in tobacco industry. It is a white, odourless, tasteless, non-toxic, hydrophilic, and biodegradable partially acetylated cellulose manufactured from natural, renewable resources such as wood pulp. It is used in making cigarette filter, textile, diapers and surgical products." Secondary cellulose acetate is significantly different from primary cellulose acetate (PCA) also known as cellulose triacetate, where all the hydroxyl groups are completely acetylated (Figure1). Since cigarette filter is manufactured by dry spinning of filter tow, using fully substituted PCA for filter tow is not suitable. Thus, PCA is partially hydrolysed wherein the degree of substitution (DS) is adjusted around 2.4 to produce SCA (Figure 2) which is then used in cigarette filter tow (Johannes and Hans-Peter, 2014).

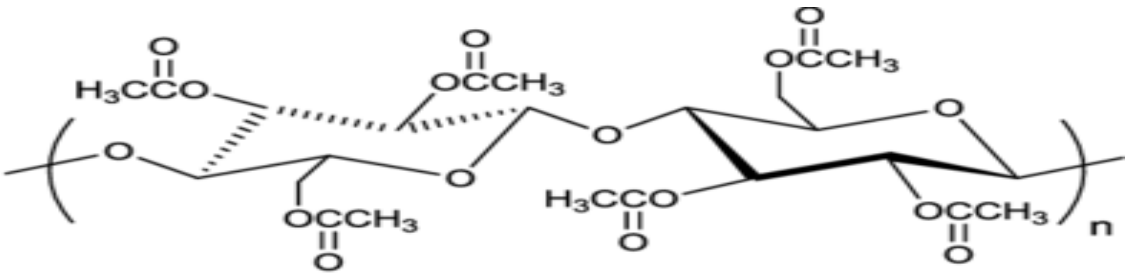

Figure 1: Structure of primary Cellulose Acetate

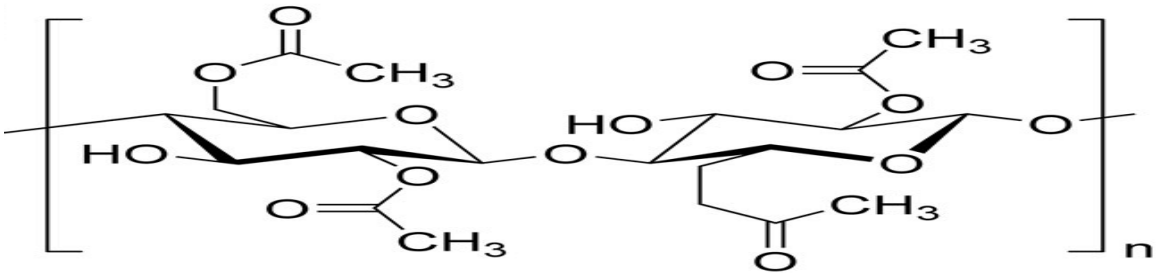

Figure 2: Structure of secondary Cellulose Acetate

Developing significance in product sustainability and a persistent interest in the environmental impact of secondary cellulose acetate-based cigarette filters has been driving rigorous research activities to understand the decomposition rate of cigarette filters.

Cellulose acetate is one of the simple cellulose esters. CA is industrially produced from the esterification of the hydroxyl groups at 2-, 3-, and 6-positions of the anhydro glucose units (AGUs) of cellulose with acetic anhydride in the presence of acetic acid as a solvent and concentrated sulfuric acid as an esterification catalyst (Malm et al., 1946). The degree of substitution (DS), i.e., the average number of acetyl groups per AGU, ranges from 1.8 to 2.5 in the acetone-soluble products. SCA is acetone soluble and has DS The properties of CA, such as solubility and viscosity, are closely related to the individual DS at the 2-, 3-, and 6-positions along the cellulose chains, as well as the overall DS (Fox et al., 2011 & Daiqiang Xu et.al,2012). Hence the solubility of SCA and PCA are completely different from each other. Cellulose acetate with different degrees of substitution was also characterized using NMR spectroscopy by Hiroyuki et.al.

According to Zhou and collaborators, cellulose acetylation dramatically alters the surface characteristics of product by altering strongly hydrophilic nature of cellulose by decreasing its polarity. As DS increases, there is also a consistent increase in hydrophobic character of cellulose (Zhou et.al., 2016). Hence PCA, which is hydrophobic in nature with high DS, whereas SCA is hydrophilic in nature. Therefore, SCA is different from PCA in its physical properties, chemical composition and also in biodegradation pattern.

Table 1: Physio-chemical Properties of Secondary Cellulose Acetate and Primary Cellulose Acetate

Parameter	**(Secondary CA)**	**(Primary CA)**
No. of cellulose hydroxyl groups that are acetylated	75-92%	>92%
Degree of substitution	<2.5	>2.5
Solubility	Soluble in acetone	Insoluble in acetone
Ability to absorb water	Hydrophilic	Hydrophobic
Tensile strength- Longitudinal (kg/mm2)	8.5 - 10	12-14
Transverse (kg/mm2)	16- 18	14-25
Melting point(^{o}C)	225-250	310-315

Note: The difference in physio-chemical properties of SCA and PCA are stated in Table-1.

An increased awareness of routes of biodegradation of cellulose acetate could prove useful to understand its persistence in the environment. SCA degrades by involving multiple degradation mechanisms (Puls, Wilson and Holter 2011). This involves the elimination of acetyl groups from the cellulose acetate followed by biodegradation of the cellulose chain. The key mechanism for biodegradation is an initial deacetylation step by chemical hydrolysis, thereby allowing the biodegradation of cellulose backbone with enzymes (cellulose & acetyl esterase) (Lindemannb, W. Ho, Martin 1983 and Komarek, Gardner, Buchanan and Gedon 1993). Hence life cycle of cellulose acetate does not harm the ecosystem as depicted in Figure 3.

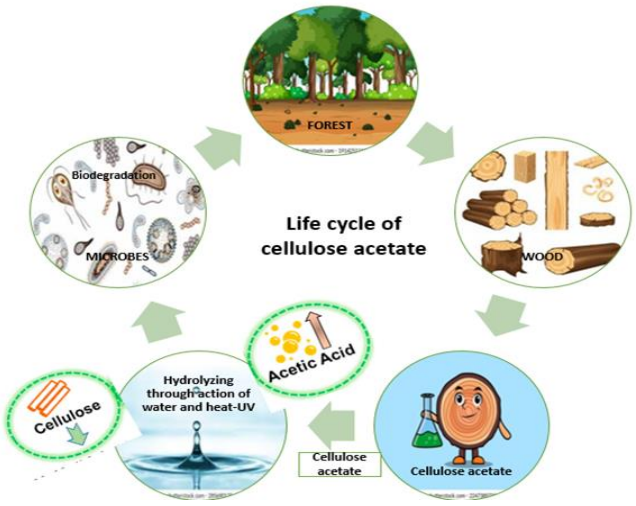

Figure 3: Mechanism of Degradation of Cellulose Acetate

Note: The complete lifecycle of cellulose acetate and degradation mechanism are depicted in Figure 3

Buchuan et. al. has studied the biodegradation of cellulose acetate films using in-vitro enrichment cultivation technique and activated sludge wastewater treatment system. The enriched culture was able to degrade cellulose acetate films within 2–3 weeks, as indicated by 67% weight loss. The industrial wastewater treatment system provided the same biodegradation pattern (Buchanan, Gardner and Komarek 1993).

Gu J.D et. al. has studied the biodegradability of cellulose acetate films with DS between 1.7 and 2.5 under controlled composting conditions where the materials were exposed to biologically active laboratory aerobic test vessels at 53^{o}C (9). It was found that the films were completely biodegraded after incubation for 7 and 18 days, respectively.

SCA appears to be labile to microbial attack in soil and wastewater environments (Buchanan et.al., 1993 and Gu, J.D, D. T. Eberiel 1993). Various studies have been conducted on the biodegradability of cellulose acetate, but very few studies have clearly explored the all-environmental aspect, including biological and chemical degradation mechanisms and assessed the persistence of SCA-based materials in environments along with its impact on plant germination and growth.

Bonanomi et. al. has conducted trials with cellulose acetate cigarette filters in soil under ambient conditions and observed 37.8% biodegradation in two years (Bonanomi et.al.,2015). The same group continued the study with same filters and reported 75% biodegradation in five years under same conditions (Bonanomi et al., 2020).

The latest review by Yadav et. al. on cellulose acetate biodegradation has mentioned that weight loss or simple growth of microorganism on the surface of the material or biodegradation of lower molar mass fraction in the material should not be considered as the proof of biodegradation (Yadav, N and M. Hakkarainen 2021).

A Task Force was chartered in 1993 by the CORESTA Technology Study Group to develop a method to predict the degradability of cigarette filters outdoors. It is known that filters will degrade over time due to the action of weathering elements such as sun, wind, rain, mechanical

action and biological actions. Similar conditions were simulated for the purposes of the study and the results obtained exhibited good corelation in terms of solar irradiance vs. weight loss records differences in climate were taken into account. The study was conducted for 12 months during which the average weight loss was approximately 26% for CORESTA Monitor2 and 24% for 1R3F Kentucky reference cigarette filters. Although, study was discontinued, the effort was both interesting and rewarding (Final Report, Task Force on Cigarette butt degradability-Aug 2000).

Recently, Sudesna et. al., (2022) found that SCA used in the cigarette industry can undergo aerobic biodegradation with 92.1% in 151 days, meeting biodegradation standards criteria. This study has proved that different cigarette filters viz., conventional and non-conventional cellulose acetate (CA) filters, infused paper filters, combined material filters (CMF), condensed tobacco end filters (CTEC) and bidi filters are biodegradable when tested accordingly to standard ISO 14855-1 (Sudesna et.al. (2022) and, ISO 14855-1, ASTM D 5338, ASTM D-6400, EN 13432, DIN EN ISO 14855-1, ISO 17088, EN ISO 14855-1, JIS K6953-1, IS 5402 Part 1).

Our hypothesis is grounded in the biodegradation mechanism of cigarette filters that incorporate secondary cellulose acetate. This substance is characterized by its hydrophilic properties and a lower degree of substitution, which together facilitate the attraction of microbial communities and improve biodegradation efficiency. As a result, this process yields carbon dioxide, water, and new biomass, with no residual secondary cellulose acetate left behind, ensuring that there are no environmental repercussions. Lack of any common definition and standard test protocol for evaluation of degradation of cellulose acetate is evident in the widely varied degradation rate of cellulose acetate reported by different research groups in all the aforementioned studies. In this context, cigarette filter degradability study in natural environment scenario will be instrumental in understanding the biodegradation mechanisms, potential degradation rates and post-disposal environmental persistence of cigarette filters.

Therefore, in this first-of-its-kind study, we report the evaluation of biodegradation of cellulose acetate-based cigarette filters in terms of complete mass loss both in natural environment as well as controlled simulated disposal environment. The findings on the environmental persistence of cigarette filters, its impact on soil quality and effect of resulting soil on plant growth will undoubtedly benefit the scientific community in multifaceted ways.

EXPERIMENTAL

This study employed several orthogonal yet complementary analytical techniques to track the behaviour and impact of SCA-based cigarette filters as well as positive control materials on soil characteristics within the dedicated environmental boundaries. The following methods were used for testing the environmental persistence of cigarette filters.

a. Cage method (Natural environment)
b. Litter Bag method (Controlled Simulated)

Instrumentation

The following glassware and equipment were used in the study: digital dual range balance (Sartorius-BSA224S-CW, Sartorius-BSA6202S-CW, Germany) to weigh test materials, orbitek shaker (Scigenic Biotech, LX-D, India), micropipettes (Brands, Germany), hot air oven (MMM Venticell Drying Oven set to 105 ± 10^{o}C), humidity chamber (MMM Climacell),

burette, measuring cylinder, beaker, and conical flask from Borosil India, Linear Smoking Machine (Cerulean SM 450, UK).

Experimental Setup

Soil Properties

A clean, vegetation free site of 3m × 3m within the ITC LS&TC campus, Peenya, Bangalore, India, which was free from shadow of trees was identified for experimentation purpose. Soil samples were collected from the several points (0-5 cm depth) inside the experimental sites, well mixed, and analysed for pH (1:5 soils: water suspension), total organic carbon content (CHNS-analyzer with pre-acid digestion method), and total nitrogen content (titrimetric method). Soil texture of the experimental site was determined using conventional mineral particle sizes method and was found to be sandy clay (ISSS classification scheme). All studies were performed in duplicates. The growth of natural vegetation was allowed and monitored throughout the course of the study (Figure 4).

The stainless cage with the specification length: 100 cm, width: 50 cm, length and height: 10 cm was assigned to each sample. Each cage had 10 compartments with 20 cm × 25 cm each with mesh diameter of 1 mm. The five cages were placed with at least one meter distance from each other.

Figure 4: Experimental Cages at Experimental Site and the Biodegradation Pattern of Cigarette Filter

Note: The experimental cages located at the testing site were assessed at the beginning of the study, and after a duration of 30 months, the condition of the cigarette filter within the cage was evaluated in relation to its biodegradation pattern.

Climatic Data

Data on the mean daily air temperature and the amount of rainfall at the site during the experimental period were obtained from the nearest meteorological stations (The Indian Meteorological Agency 2016 to 2019) depicted in Figure 5.

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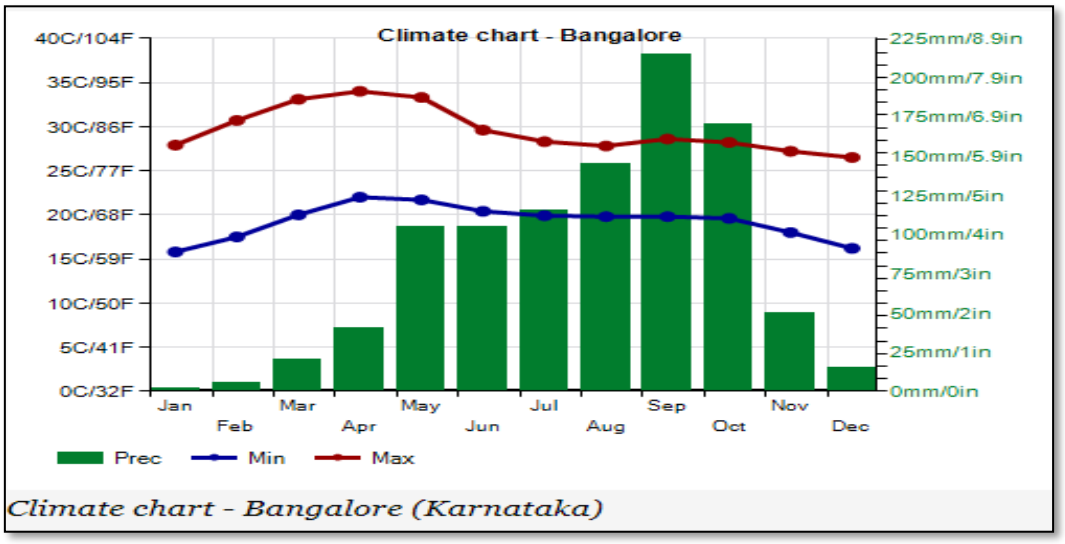

Figure 5: Annual Climate Charts of the Experimental Region from the Indian Meteorological Agency

Test Sample Preparation

Two cigarette brands with secondary cellulose acetate filter and combined material filter were selected for the study. The cigarette samples were smoked using linear smoking machine as per ISO regime (puff volume: 35 cc, puff duration: 2 sec, puff frequency: 60 sec) following the standard ISO 3308 titled "Routine analytical cigarette-smoking machine- Definitions and standard conditions". After machine smoking of cigarettes, butts were collected. The remaining portion of tobacco and cigarette paper were removed. The cigarette filter sample thus prepared was considered as test sample. Before the start of study, smoked as well as non-smoked cigarette filters were analysed to determine total weight, dry weight, volume of cigarette filter, weight of CA & non- CA content. Cigarette filters without smoking were considered as control for each brand.

Composite degradation value (CDV) value was formed using 20 smoked cigarette filters and 20 unsmoked cigarette filters. 200 smoked and 100 unsmoked cigarette filters from each brand were distributed equally among the 10 compartments of each cage. Samples in cages were arranged in a way to ensure the sample comes in contact with the soil.

Parameters Assessed during Biodegradation

Two cigarette filter samples from each of the 10 cages constituting 20 cigarette filter samples in total were collected every three months after cleaning the vegetation in the experimental site and samples are cleaned using brush for dust and other foreign materials. These samples were then subjected to analysis.

Total weight as-is basis, total weight dry weight basis, visual observation (especially with respect to overwrap and overall shape of sample), volume, CA content and non-CA content were measured. Additionally, a snap shot of samples after sampling were taken each time and compared with the mass loss trend of cigarette filters. The change in external appearance of the cigarette filters were assessed, the degradation was estimated visually, and rating was assigned as per the criteria depicted in the Table 2.

Table 2: Visual Assessment Rating for Plug Wrap and Filter Filters

Plug wrap		Shape of filter	
Visual analysis	**Rating**	**Visual analysis**	**Rating**
No change in plug wrap	100	No change in filter shape	100
Partial loss of plug wrap	66	Partially broken filter shape	66
Major loss in plug wrap	33	Considerably broken filter shape	33
All plug wrap degraded	0	Completely broken filter shape	0

Initial weight of the filter was recorded after cleaning using a soft brush. The filters were then dried in an oven for 1 hour at a constant temperature of 105°C followed by allowing the cigarette filters to attain room temperature in desiccator. The filters were then conditioned in a stability chamber at 22 ± 2°C temperature and 60 ± 5%for 30 mins RH for 24 hours and weighed. The mass loss was examined by percentage loss of weight of cigarette filter during the study with respect to the baseline weight.

$$\textbf{Mass Loss}(\%) = \frac{\textbf{Baseline Wt. of filter butt}(g) - \textbf{Final Wt. of filter butt}(g)}{\textbf{Baseline Wt. of filter butt}(g)} \times \mathbf{100}$$

The conditioned cigarette filter filters were individually extracted with 20 mL of acetone, the solution was filtered through filter paper and the filtrate was evaporated to dryness. Both the filter paper and residue obtained upon drying the filtrate were further dried at a constant temperature of 105°C and conditioned in a stability chamber at 22 ± 2°C temperature and 60 ± 5% RH for 24 hours. CA content was determined by weighing the residue obtained upon drying the filtrate and the weight of the residual particles in the filter paper indicated the non-CA content.

A 10 ml graduated cylinder was first filled with 5 mL of 250-micron glass beads while ensuring proper packing of the beads and this initial volume was noted. After this, in a 10 ml graduated cylinder, half of the glass beads is first layered ensuring no air-pockets followed by adding the pre-dried cigarette filters and finishing with a second layer of glass beads with proper packing. The final volume is then recorded. The difference between the final and initial value gives the post-degradation volume of the cigarette filters.

Loss in volume of the filter pre- and post-degradation is determined in percentage using the following formula. These results are further evaluated based on the visual assessment criteria described in the table 3.

$$\textbf{Loss in volume}(\%) = \frac{\textbf{Baseline volume}(mL) - \textbf{Post degradation Volume}(mL)}{\textbf{Baseline Volume}(mL)} \times \mathbf{100}$$

Table 3: Visual Assessment Rating for Volume of Plug Wrap and Filter Filters

Filter weight		Filter volume	
Percentage remaining	**Rating**	**Percentage remaining**	**Rating**
100-90	100	100-90	100
89-90	80	89-70	80
79-60	60	69-40	60
59-40	40	39-30	40
39-20	20	29-15	20
19-0	0	14-0	0

Final evaluation:

The biodegradation of the cigarette filters is finally evaluated by using composite degradation value (CDV). CDV is calculated based on the weighted average of the percentage of appearance, weight percentage remaining and volume percentage remaining of the original filter. It is defined by the formula as:

CDV = (% Appearance + %Weight remaining + % Volume remaining)/3

The percentage of appearance is derived by the following formula:

$$\%Appearance = (Plug\ wrap\ rating\ +\ Shape\ of\ filter\ butt\ rating + filter\ butt\ \%weight\ \%\quad remaining\ rating + filter\ butt\ \%volume\ \%\ remaining\ rating)/4$$

As long as any part of the test piece filter is remaining in the soil at the experiment site, the study was not terminated (Figure 4).

Litter Bag method (Controlled Simulated)

Material Collection

Regular filtered cigarettes with three common types of filters i.e., secondary Cellulose acetate filter (SCA), combined material filter (CMF) and secondary cellulose acetate filter with additives (DeTow^{TM}) were smoked in a linear smoking machine as per ISO 3308. A comparatively slow-decomposing standard organic substrate i.e., wood sticks cut from Saal tree branches (Southern India) was used. The cigarette filters and wood sticks showed different initial chemical features as follows: i) cigarette filters (N content = 0.31% of dry matter; C/N ratio = 154.8; ii) wood sticks (N content = 0.12%; C/N ratio = 440.7; lignin content = 38.54 % of dry matter).

Decomposition Study

The decomposition of matter in field conditions mainly depends on organic matter quality, water availability and temperature condition (Roberts, 2011). In this study, we decomposed organic substrates in simulated laboratory conditions. The laboratory experiment was included to reduce the importance of water availability and temperature on decay rate variation and to isolate the effect of cigarette filter biochemical quality. Decomposition experiments were carried out according to the litterbag method (Banonomi, 2015). The wood sticks were collected from the branch of Saal tree (Shorea Robusta) and used as control in the study. Wood sticks were cut with scissors to obtain pieces of each weigh approximately 200 mg & 200 mg

of cigarette filters from the selected brands were also weighed. Litterbags filled with 50 pieces (approx. 5.0 g) of each material along with grassland soil from the region of Mysore, Karnataka, southern India and placed inside a tray in the laboratory. Grassland soil was selected because it is one of the common natural environments where cigarette filters are discarded and it shows large differences in soil texture and fertility in comparison to other soil types. Soil texture was determined by sedimentation method and carbon and nitrogen content in soil was determined by elemental analyser. The main composition of grassland soil included 42.0% sand, 35.0% silt, 23.0% silt (loam soil). Other soil characteristics are listed in table 4.

Table 4: Soil Characteristics

Parameters	Result
pH	7.8
Soil texture	Sandy Clay
Nitrogen (%)	1.2
Carbon (%)	9.99

Litterbags were constructed from rayon fibre with a mesh size of 1.2 mm× 1.4 mm by heat sealing. Mesh size is generally chosen to optimize access of all soil microorganisms to the litter while minimizing excessive particle loss. Mesh size can also be manipulated to exclude functional groups of litter decomposers. Very small mesh size will not only exclude certain organisms, but also prevent particle loss to mineral soil as well. Square-shaped litterbags with 15-cm side length were filled with about 5.00 g of the air-dried cigarette filter and wood stick. Thirteen bags were taken per set and were set out next to one another with 3 to 5 cm gap between them and the litterbags were covered with 1-2 cm of soil. The experiment was done in duplicate and was planned for 36 months.

In the laboratory, litterbags were placed in a growth chamber under controlled temperature (22 ± 2°C night and 25 ± 2°C Day) and water (watered with distilled water every seven days to water holding capacity, previously determined equal to be 250%) conditions.

After retrieval, litterbags were opened and contents washed to separate litter residues from contaminants, e.g., soil and visible fauna. Residues were then oven-dried at 40°C (until constant weight was reached) and weighed to the nearest 0.001 g. Filters and wood sticks from each replicate were retrieved for each of twelve planned dates from the start of the experiment (i.e., 3, 6, 9, 12, 15, 18, 21, 24, 27, 30, 33, 36 months), totalling 1200 filters or wood sticks. The substrates retrieved after 3, 6, 9, 12, 15, 18, 21,24, 27 months of decomposition were analyzed.

Chemical Characteristics

Smoked cigarette filters, both undecomposed and decomposed for 27 months, were characterized for total C and N content by flash combustion of micro samples (5 mg each) in an Elemental Analyser. Additionally, undecomposed CBs (control) as well as CBs decomposed for 27 months were obtained in solid state under the same conditions and characterised by 13C-CPMAS NMR, thus allowing comparative analysis of the resulting spectra. 13C-CPMAS NMR analysis was done to highlight the differences between control cigarette filters made of cellulose acetate with the resulting biomass of cigarette filter. The spectrometer used was a Bruker AV-300 equipped with a 4 mm wide-bore MAS probe. Spectral regions and corresponding signals were monitored: 0–45 ppm = alkyl C; 46–60 ppm = methoxyl and N-

alkyl C; 61–90 ppm = O-alkyl C; 91–110 ppm = di-O-alkyl C; 111–140 ppm = H- and C-substituted aromatic C; 141–160 ppm = O-substituted aromatic C (phenolic and O-aryl C); 161–190 ppm = carboxyl C. Notably the carboxylic C signal in 161–190 ppm indicates the presence of acetate functional group. The degree of acetylation of cellulose was assessed by dividing the integral of the methyl C signal by the C-1 signal.

Gel Permeation Chromatography (GPC) technique is used to measure the absolute molecular weight of derivatised as well as natural polymer (Hintersteiner et.al., 2015). Herein, GPC techniques was used to measure molecular weight of pre-biodegradation smoked filters as well as the resulting biomass of the biodegraded cigarette filter samples of cellulose acetate, CMF and DE-Tow^{TM} to understand the extent of biodegradation and determine the persistence of any trace of cellulose acetate material.

The surface topography and cross-sectional morphology of the pre-biodegradation CA & DeTow^{TM} smoked cigarette filter (Control) and the intermediate cigarette filter sample were studied using JEOL SEM-25 scanning electron microscope (SEM). This microscope uses a concentrated electron beam to create a detailed picture of the filter's surface, allowing for an evaluation of its structural changes.

RESULT AND DISCUSSION

Rate of Biodegradation of Different Types of Cigarette Filters

Cage Method (Natural Environment)

The assessment of biodegradation of cigarette filters in natural environment confirmed unambiguously that cellulose acetate used in cigarette filters degrade completely in active microbial environment i.e., undergoes 100% mass loss in 30 months. However, degree of biodegradation of different types of CA based cigarette filters were found to be dependent on factors such as i) site variation, presumably due to the difference in the climatic and soil conditions (12) and ii) degree of substitution and filter composition which reflect the binding force among molecules and might impact the accessibility of the microbial enzymes responsible for degradation to the target sites.

Mass loss of cigarette filters follows a three-step decomposition pattern. In the first three months, ~17 to 23% of the initial mass was rapidly lost which can be attributed to rapid degradation of the external cellulose layer of cigarette filter overwrap (Figure 6). Subsequently, cigarette filter overwrap completely disappeared within 15 months. Finally, after 21 & 30 months of continuation of the study for CMF & CA cigarette filters respectively, no trace of sample was observed indicating complete decomposition in soil (Figure 6).

Table 5: Cigarette Filter Degradation by Cage Method in Terms of Mass Loss

Cigarette Filter	Months	Mass Loss (%)
CMF Filter	21	100
CA Filter	33	100

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Figure 6: Cigarette Filter Biodegradation Pattern in Cage method

Note: Table 5 illustrates the mass loss of cigarette filters over a specified number of days, while Figure 6 depicts the biodegradation pattern of cigarette filters in cage method.

Figure 7 shows the graphical representation of the rate of biodegradation of smoked filter of two different brands i.e., Capstan Pilot which is a CA filter and Flake Goldcrest which is a CMF. CA filter with 2.4 DS exhibited slower biodegradation rate (complete biodegradation in 30 months) whereas CMF with 2.1 DS exhibited complete mass loss in 21 months (Table 5). The biodegradation of CA cigarette filters involves two steps and biofilm formation of at least two different types of microbes. Bacillus subtilis and Acinetobacter releases acetyl esterase enzyme, which removes the acetyl groups from CA and Pseudomonas which releases cellulase enzymes required for breaking down the cellulose ring (6). CMF is composed of a combination of cellulose acetate as well as paper made of cellulose. The cellulose portion can be easily degraded in a single step by cellulase enzyme which is likely reflected in a slightly faster rate of biodegradation. However, it should be noted that both CMF and CA cigarette filters are completely biodegraded in natural soil environment.

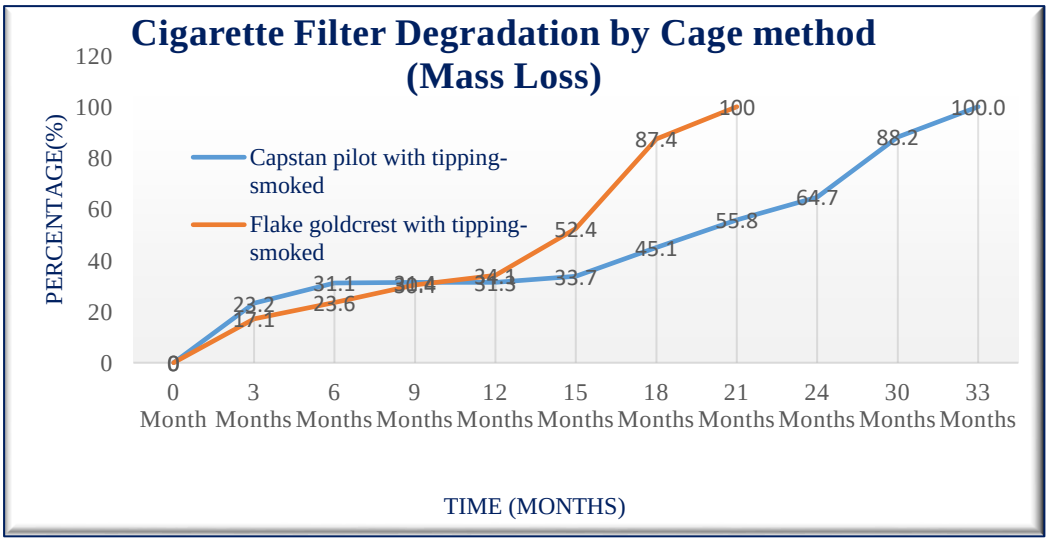

Figure 7: Graphical Representation of Cigarette Filter Degradation Study of CA & CMF Filter under Ambient Condition

Note: The graphical representation in Figure 7 shows the degradation pattern of the cigarette filter (CA, CMF) by Cage method as it progresses over time.

Interestingly, unsmoked CA filter used as control sample also demonstrated mass loss in 30 months indicating that smoking has no impact on the degree of biodegradation of the cigarette filter. Furthermore, figure 4 shows growth of vegetation in the experimental site thereby confirming that CA cigarette filter does not have any negative impact on plant growth and does not exhibit any environmental ecotoxicity.

Further confirmation of biodegradation was obtained by studying cellulose acetate content, volume and CDV which arrived at the same conclusion as discussed above.

Environmental Impact on Biodegradation of CA Cigarette Filter

The monthly minimum and maximum temperatures of the experimental sites ranged from 15.5^{o}C to 22.0^{o}C, and 27.0^{o}C to 34^{o}C, respectively. Total rainfall during the experimental period ranged from 0 to 2662 mm. Effective temperature is considered in the field of agricultural meteorology to estimate crop growth and the apparent amount of nitrogen mineralized in submerged paddy soils (Uchijima 1962, Dei and Yamazaki, 1979). mineralization and immobilization of nitrogen proceeded to a similar extent. The temperature, precipitation as well as the total rainfall during the current study is listed in Table 3. Correlation between the degree of biodegradation of the cigarette filter and the summation of effective temperatures and the total rainfall during the study were observed to be proportional.

Table 6: Soil Characteristics

Parameters	Result
pH	7.8
Soil texture	Sandy Clay
Nitrogen (%)	1.2
Carbon (%)	15.8

Note. Table 6 presents the key soil characteristics that were utilized in the study.

Mass loss of cigarette filters was significantly affected by interactive effects of both abiotic and biotic degradation conditions such as soil type, pH of soil, soil texture and weather conditions (temperature, humidity and rainfall) of the decomposing site. Interestingly nitrogenous soil facilitated the degradation of cigarette filters which might indicate that microorganisms thrive in nitrogen rich soils (Table 6). Biodegradation was found to be correlated positively with the amount of soil organic matter, and the total nitrogen content (Briassoulis and Mistriotis, 2018).

Litter Bag Method (Controlled Simulated)

The rate of biodegradation of the cigarette filters in the controlled simulated conditions show a similar trend to the natural environmental study. In case of CA filter and cellulose acetate-based cigarette filter with alkaline additives (DeTow^{TM} filter), rapid mass loss of ~24 to 27% is observed in the first three months along with the complete disappearance of the wrapping layer (Figure 9). This is attributed to the cellulose-based cigarette filter tipping paper. Followed by this rapid degradation, decomposition proceeds relatively slowly with only an additional ~10.0% mass loss from 3rd month to 6th month. The slowed decomposition can be owed to two main factors: intrinsic resistance to biodegradation of cellulose acetate due to a higher DS

of ~2.45, and limited nitrogen availability inside the cigarette filter eventually leading to microbial starvation. It is well established that soil microbiota capable of decomposing N-poor plant tissues (i.e., wood and coarse root), can actively attain inorganic N from the surrounding soil to sustain their biological requirements. Since this study was conducted under simulated laboratory conditions, availability of inorganic nitrogen is limited in this case. CA filters achieved total mass loss of 99.0% and DeTow^{TM} filters showed total mass loss of 100% at the end of 27 months of incubation post which no trace of sample was observed.

In case of CMF, mass loss is considerably faster and ~50% of initial mass was lost in the first three months of incubation. However, as decomposition proceeded the rate of mass loss dramatically declined, with a mass loss of only ~54% after 6 months of incubation. The decomposition rate then increased linearly from the 6th month and complete mass loss was observed within 15 months of incubation (Table 7). Since the combined material filter is constituted of cellulose acetate and paper from highly biodegradable cellulose in combination, the mass loss is accelerated in comparison to cellulose acetate and DeTow^{TM} filter.

In case of the control wood stick samples, the rate of the mass loss increased linearly from 0 month to 12 months of incubation followed by a sharp increase in mass loss finally achieving 45% decomposition in 27 months (Figure 9). This provides us significant insight that cigarette filters which are considered one of the most notorious litter to the environment, degrades significantly faster than known biodegradable lignin-rich wood sample.

Table 7: Mass Loss of Cigarette Filters by Litterbag Method

Time (Months)	Control (Saal Wood)	DT Filter	CMF Filter	CA Filter
3	3.0	27	50	24
6	6.0	39	54	35
9	9.0	52	67	41
12	12.0	65	82	49
15	30.6	75	100	67
18	39.2	90	100	86
21	39.0	92	100	89
24	42.9	97	100	95
27	45.0	100	100	99.9

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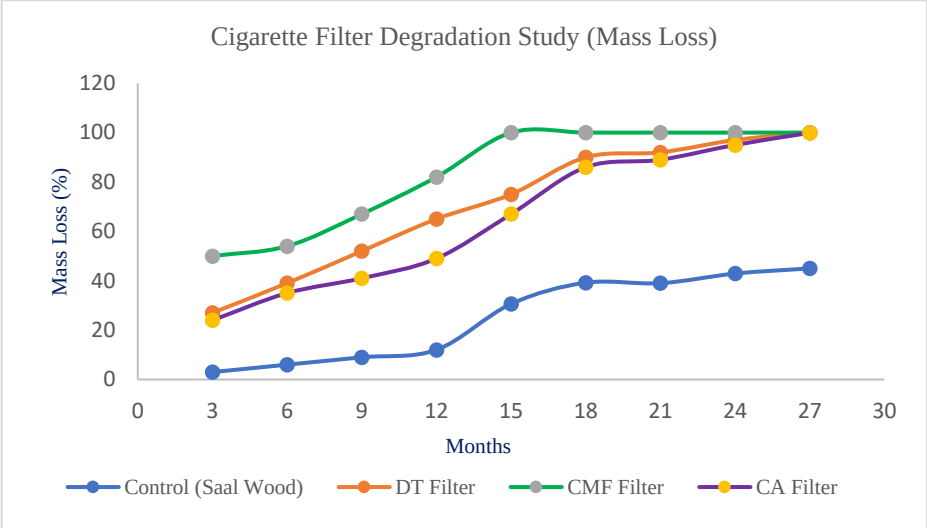

Figure 9: Graphical Representation of Mass Loss by Litterbag Method

Note. Table 7 and Figure 9 depict the mass loss of cigarette filters using the Litterbag method, along with its graphical representation.

After 27 months of study, the samples were not available for sampling indicating complete decomposition in soil (Figure 10).

Figure 10: Mass Loss Pattern of Saal Wood, CA Filter, CMF Filter, DeTow™ Filter and Respective Sample at the End of 27 Months by Litterbag Method

Note. In Figure 10, the time-dependent degradation of cigarette filter samples and their respective control sample is presented, utilizing the Litterbag method for analysis.

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Evaluation of Biomass Post-Degradation of Cigarette Filters

Evaluation by Nuclear Magnetic Resonance Spectroscopy (NMR)

13 CPMAS NMR spectroscopic studies were conducted to gain insight into the presence of cellulose acetate in the biomass post-biodegradation. In this context, the NMR spectra of control cellulose acetate filter (not subjected to degradation) and the resulting biomass after 27 months of decomposition was analyzed. The disappearance of the 13C signals at 1.95 and 1.18 ppm (Figure 11) which are characteristic of acetyl carbon in the post-biodegradation sample indicate that no residual cellulose acetate remains in the biomass after biodegradation. This is most likely a reflection of microbial attack on the sample during two years of decomposition further confirming that cigarette filters can act as a source of carbon for microbes resulting is complete change in chemical nature of the cigarette filters.

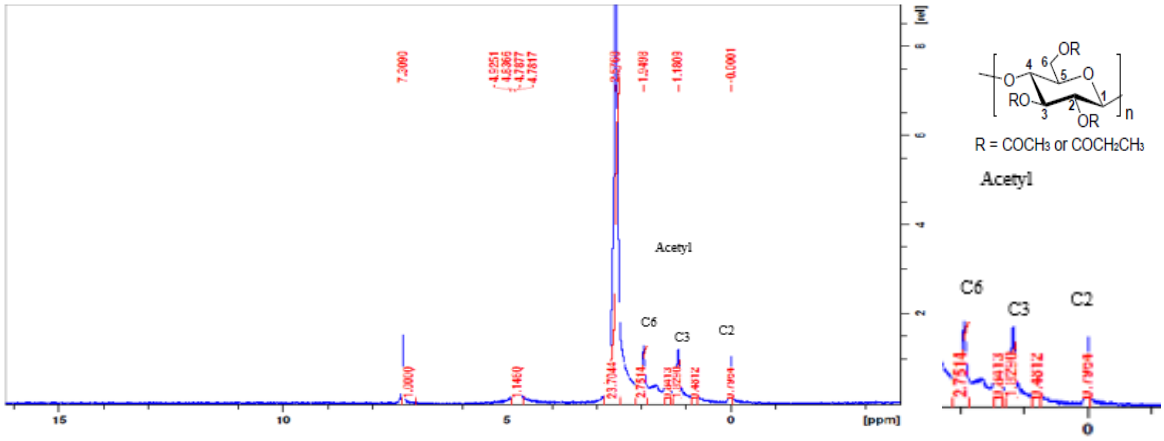
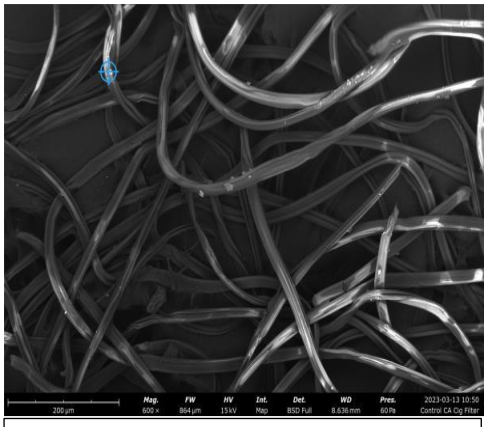


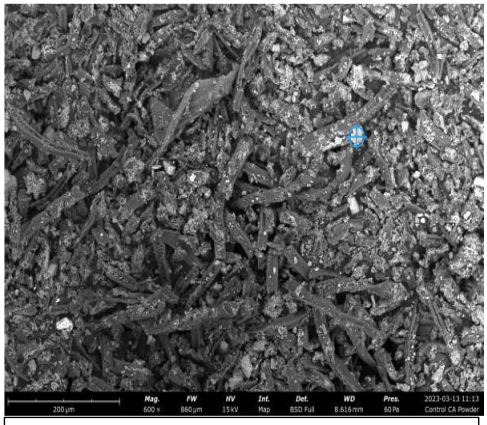
Figure 11: 13C CPMAS NMR spectra of undecomposed cigarette filter

Scanning Electron Microscopy study (SEM)

SEM image of undecomposed (control) and decomposed (sample) CA Filter and DeTow™ Filter are shown in Figure 12 and 13. Before the study, the degraded secondary cellulose acetate-based cigarette filters were dried under sputter-coated gold.

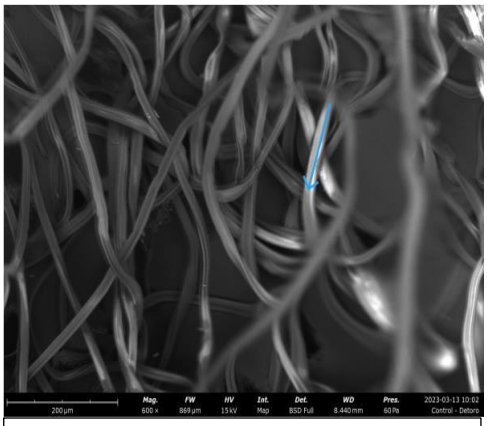


CA Control

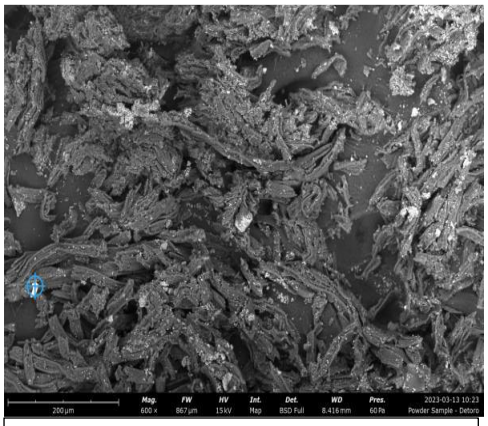


CA Sample

Figure 12: Scanning Electron Microscope image of CA Control and CA Sample at the End of 21 Months



DeTow™ **Control**



DeTow™ **Sample**

Figure 13: Scanning Electron Microscope Image of DeTow™ Control and DeTow™ Sample at the End of 21 Months

Figure 12 and Figure 13 of the cigarette filters, both in their initial biodegradation state and during processing, illustrate the structural alterations occurring in the cellulose acetate fibres.

The surface of undecomposed cellulose acetate fibre displays defibrillation of the fibrils into individual fibrils of uniform size, whereas surface of sample at the end of 21 months shows complete distortion of fibre structure due to biodegradation. It should be noted that SEM study could not be conducted at the end of 27 months since no trace of samples remained owing to complete mass loss (Table 7).

Evaluation by Gel Permeation Chromatography (GPC)

When assessing the biomass of cigarette filters by GPC, we focused on chacterization of number average peak molecular weight (MP). The chromatograms shown in Figure 12 depict the cellulose acetate filter (CA), Combined Material filter (CMF), DE-Tow™ Filter control, along with their associated biomass in THF, where signal intensity is represented as a function of elution time (in minutes).

Molecular weight test results (average in Daltons) from GPC analysis indicated that completely degraded, as no traces of these materials were found in GPC analysis. In order to confirm these results, control samples of the respective filters were analysed and the molecular weights as follows were confirmed CA: 187876 Daltons, CMF: 185162 Daltons and DE-Tow™ Filter: 90760 Daltons.

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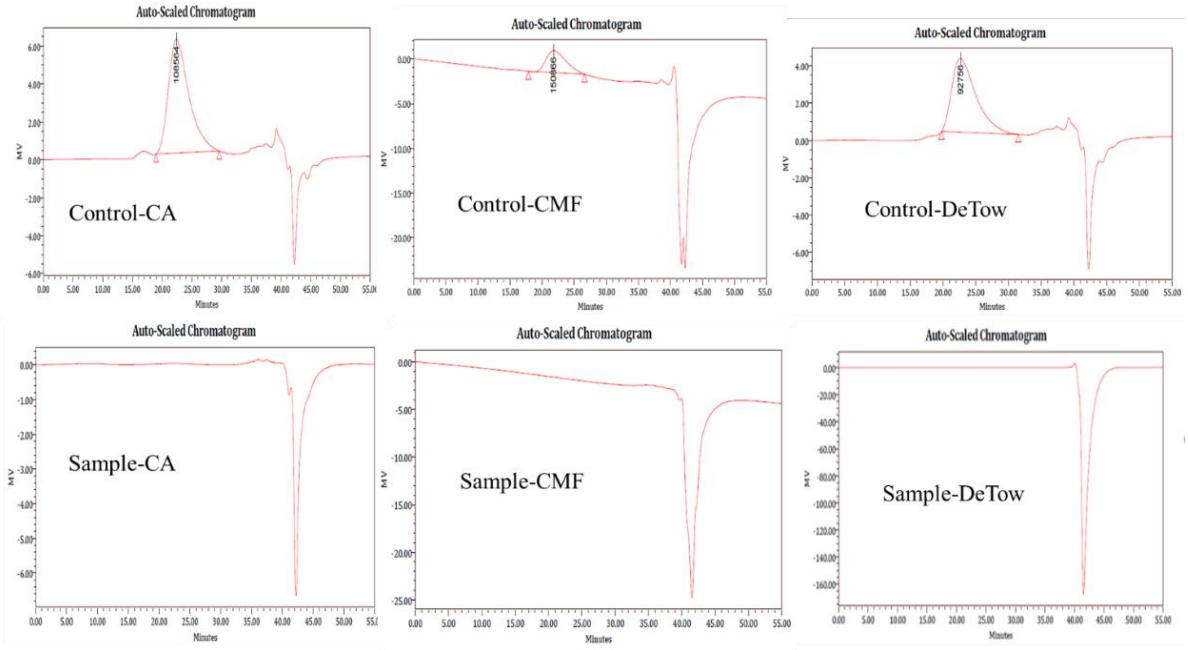

Figure 14: GPC Chromatogram of CA Filter, CMF Filter & DeTow™ Filter – A. Control (Smoked Filter) and B. Sample (Resulting Biomass of Smoked Filter)

Note. The signal intensity of cellulose acetate in the biomass derived from CA, CMF, and DeTow™ cigarette filters, along with their corresponding controls, has been depicted in Figure 12.

Resulting biomass from both the studies i.e Cigarette butt degradability study and Litterbag method has been characterised by SEM, NMR and GPC techniques to confirm nonpersistent of SCA based cigarette filter in the environment (Figure 15).

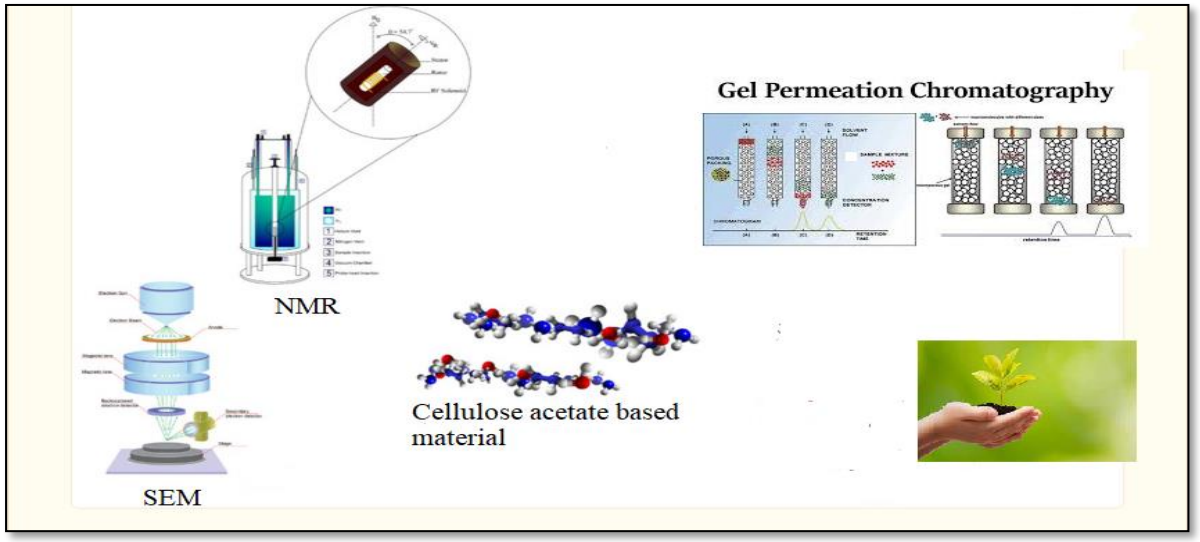

Figure 15: Analytical Techniques Used for the Identification of Nonpersistent of Cellulose Acetate

These studies indicate no adverse impact on ecological surroundings, as during biodegradation process, microbes use SCA as food and thrive on that and produce carbon dioxide, water, new biomass leaving behind no SCA residues. Also, the vegetation surrounded in and around the cage, indicating the new biomass from the cigarette filter is supporting the plant growth and harmless to environment.

CONCLUSION

This first of its kind comprehensive assessment study displays the in-depth understanding of the complicated interplay between structure, environment and biodegradation process of cigarette filter made with cellulose acetate. The study revealed that CA cigarette filters are indeed completely degraded with 100% mass loss well within 33 months both in natural environment and within 27 months in controlled simulated laboratory conditions. These findings are further substantiated by SEM studies which confirms complete structural change in cigarette filter morphology due to biodegradation and GPC & NMR studies which reaffirms that no trace of cellulose acetate remains in the resulting biomass.

The key take-away from this study is that cigarette filters are labile to microbial degradation in soil environments. Unlike most typical synthetic polymer, cigarette filters made with secondary cellulose acetate, breaks down into cellulose and acetic acid by biotic and abiotic factors in the natural environment and the resulting biomass presents no adverse environmental consequences.

The distinctive feature of this study lies in the utilization of a dual methodology, incorporating both natural and laboratory-simulated soil environments, which is further enhanced by comprehensive analytical verifications. Most importantly, this research offers robust scientific justification for all examined cigarette filters, including both traditional and newly formulated cellulose acetate filters, confirming their complete biodegradability and hence absence of any negative impact on environment. However, biodegradability and non-persistence, do not mitigate the immediate adverse effects of careless littering; they do reduce the long-term consequences.

Biodegradation is significantly influenced by climatic factors. Future investigations should concentrate on comprehensive analyses of regional variations in biodegradation rates, as well as the examination of various types of cigarette filters. It is essential to establish a standardized testing protocol to evaluate the biodegradation of cigarette filters across different demographic regions.

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