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1 TITLE: Comparison of cellulose vs. plastic cigarette filter decomposition under distinct
2 disposal environments

3

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21

22 ABSTRACT

23 It is estimated that 4.5 trillion cigarette butts are discarded annually, making them numerically
24 the most common type of litter on Earth. To accelerate their disappearance after disposal, a
25 new type of cigarette filters made of cellulose, a readily biodegradable compound, has been
26 introduced in the market. Yet, the advantage of these cellulose filters over the conventional
27 plastic ones (cellulose acetate) for decomposition, remains unknown. Here, we compared the
28 decomposition of cellulose and plastic cigarettes filters, either intact or smoked, on the soil
29 surface or within a composting bin over a six-month field decomposition experiment. Within
30 the compost, cellulose filters decomposed faster than plastic filters, but this advantage was
31 strongly reduced when filters had been used for smoking. This indicates that the accumulation
32 of tars and other chemicals during filter use can strongly affect its subsequent decomposition.
33 Strikingly, on the soil surface, we observed no difference in mass loss between cellulose and
34 plastic filters throughout the incubation. Using a first order kinetic model for mass loss of for
35 used filters over the short period of our experiment, we estimated that conventional plastic
36 filters take 7.5 to 14 years to disappear, in the compost and on the soil surface, respectively. In
37 contrast, we estimated that cellulose filters take 2.3 to 13 years to disappear, in the compost
38 and on the soil surface, respectively. Our data clearly showed that disposal environments and
39 the use of cellulose filters must be considered when assessing their advantage over plastic
40 filters. In light of our results, we advocate that the shift to cellulose filters should not exempt
41 users from disposing their waste in appropriate collection systems.

42

43 KEYWORDS

44 Cigarette butt – Compostable – Municipal solid waste – Biodegradable

45

46 INTRODUCTION

47 With an estimated 4.5 trillion cigarettes discarded every year in the environment, cigarette
48 butts are the most common type of litter on earth (Novotny et al., 2009) and are typically
49 found in many ecosystems from urban and peri-urban areas to beaches and oceans (Ariza et
50 al., 2008). Aside from being unsightly, they represent a serious threat to organisms and
51 ecosystems as they are toxic to microbes, insects, fish and mammals (Novotny et al., 2011).
52 Since these filters are made of plasticized cellulose-acetate inaccessible to microbes for
53 biological decomposition (Zugenmaier, 2004), they likely accumulate and the environmental
54 issue they cause keeps rising. Consequently, the tobacco-industry has developed in the last
55 decade an environmentally-friendly alternative to conventional plastic filters, consisting of
56 filters made of pure cellulose, i.e. a molecule that is entirely biodegradable by soil and aquatic
57 microbial communities (Berg and McLaugherty, 2008). However, the relative advantage of
58 these filters for decomposition remains unknown.

59 In the only peer-reviewed publication that assessed the decomposition of conventional
60 cigarettes filters, Bonanomi et al. (2015) reported that while the paper wrapped around the
61 filter was readily decomposed, the plastic part was mostly unaffected after two years of
62 decomposition. In turn, the OCB® brand for instance, that sells filters for hand-rolling
63 cigarettes, advertises an almost complete decomposition of cellulose filters in 28 days.
64 However, these results, coming from a test made by an independent laboratory following the
65 301B biodegradability protocol of the Organization for Economic Cooperation and
66 Development (OECD), have not been published, and do not compare with the decomposition
67 of conventional plastic filters, making it impossible to evaluate the advantage of cellulose
68 filters over the plastic ones. Particularly, given the predominant control of environmental
69 conditions on biotic litter decomposition (Berg and McLaugherty, 2008), the decomposition
70 of the cellulose filters is likely to vary widely depending on their disposal environment. In
71 contrast, environmental conditions were shown to have no effect on decomposition of plastic

72 cigarette filters (Bonanomi et al., 2015). Consequently, in composts, where environmental
73 conditions are prone to microbial activity, the relative advantage of cellulose filters over the
74 plastic ones may be reinforced. Moreover, the goal of the OECD protocol is to evaluate the
75 biodegradability of the substance out of which the product is made without necessarily taking
76 into account its previous use. Such potential decoupling of the test from realistic conditions
77 could importantly limit the validity of the results. Indeed, once the cigarette is smoked, the
78 filter gets charged with a large variety of compounds including tars, carcinogenic compounds
79 and numerous metals (Hoffmann, 1997; Moerman and Potts, 2011), which leads to an
80 increased toxicity of filters for wildlife (Dieng et al., 2013; Slaughter et al., 2011; Suárez-
81 Rodríguez et al., 2013) as well as microorganisms (Micevska et al., 2006). Consequently, the
82 microbial decomposition of cellulose filters is likely to be substantially decreased for smoked
83 filters, decreasing the relative advantage of cellulose filters over plastic ones.

84 In this study, we aimed at providing some very first robust scientific data assessing
85 how much faster cellulose filters decompose compared to their plastic equivalents. During a
86 six-month incubation under field conditions (Mediterranean old-field), we compared the
87 decomposition of cigarettes filters made out of cellulose (and so-called hereafter) and
88 cellulose acetate (called ‘plastic’ hereafter). To determine the advantage of composting over
89 simple discarding, we compared decomposition on the soil surface to that within a
90 composting bin (referred to as ‘compost’ hereafter). Finally, to evaluate the importance of
91 filter use on their decomposition, we compared the decomposition of smoked and new filters.
92 We hypothesized that (i) cellulose filters would decompose considerably faster than plastic
93 filters, that (ii) smoked filters would decompose more slowly compared to new filters, and
94 that (iii) these effects would be more pronounced in a compost where decomposition would
95 be hastened.

96

97 METHODS

98 *Filters*

99 Cigarette filters of the OCB® brand, made for hand-rolling cigarettes, were purchased in
100 2013. We selected slim filters (length x diameter: 15 x 6 mm) of two different qualities, one
101 made of cellulose acetate (plastic), and one made of cellulose (cellulose). To study the effect
102 of smoking on the subsequent decomposition of filters, cigarette butts were collected from
103 voluntary smokers that collected their own cigarette butts in portable ashtray, and used filters
104 of both plastic and cellulose filter from the same aforementioned brand. Filters were then
105 retrieved from the cigarette butts. All types of filters were then dried at 60°C for 48 h,
106 weighed and placed in a 25 x 25 mm litterbags made of polyethylene (mesh size: 0.6 x 0.5
107 mm).

108

109 *Experimental design*

110 Litterbags containing all types of filters were placed to decompose in the experimental field of
111 the Center of Evolutionary and Functional Ecology, on February 21, 2014, under two
112 conditions, either directly on the soil surface of a Mediterranean old-field, or buried in a
113 plastic container containing compost. The compost consisted in a mixture of green manure
114 made of ramial chipped wood and mature compost to ensure microbial inoculation. The first
115 condition corresponds to the scenario where butts are thrown on the soil and remain there to
116 decompose, while the second condition corresponds to the scenario where butts would be
117 collected and composted with other organic waste. The climatic conditions at the study site
118 are typically Mediterranean, with a mean annual temperature of 15°C and a mean annual
119 precipitation of 570 mm (average of the 1981-2010 period). Over the 5.4 months of the
120 experiment, cumulated precipitation was 124 mm, with an average temperature of 17.4°C.
121 The experimental design included four factors: filter type (plastic vs cellulose), use (smoked

122 vs unsmoked), soil conditions (soil vs compost) and length of incubation (five harvests). As
123 all factors were crossed, we obtained 40 treatment combinations. For each combination, six
124 replicates were placed in six separate blocks and litterbags were randomized within each
125 block. The six replicates of the smoked filters consisted of three filters from each smoker to
126 allow testing for the smoker effect. To ensure the start of microbial decomposition both on the
127 soil surface and in the compost, all blocks were watered at the beginning of the experiment,
128 with additions of 20 mm precipitation pulses. Additionally, to ensure optimal conditions for
129 microbial decomposition in the compost, the plastic containers were rewetted every month
130 with additions of 10 mm precipitation pulses. Litterbags were harvested at five different times
131 (2, 4, 8, 16, 32 weeks) after the start of the experiment. At each harvest, filters were cleaned
132 to remove soil particles, dried at 60 °C for 48 h and weighed to determine the mass loss. In
133 order to assess the amount of mass loss due to leaching for all filter treatments (plastic and
134 cellulose filters, both smoked and unsmoked), we ran an additional leaching experiment. To
135 do so, 10 filters of each filter treatments were dried at 60°C for 48h, weighed and placed
136 separately in a Falcon® tube with 15 ml of deionized water placed on a rotator spinning at 8
137 rpm for 24 h (Joly et al., 2016). Filters were then dried at 60°C for 48h and weighed to
138 determine mass loss. For both experiments, mass loss was expressed in percentage of initial
139 litter oven-dry weight.

140

141 *Data analysis*

142 First, to ensure that the decomposition process was not affected by the identity of the smoker,
143 the smoker effect (n = 3 per smoker) was evaluated separately using a one-way ANOVA and
144 then with the others factor using a complete ANOVA model. As it was not significant in any
145 case ($p > 0.05$), this factor was finally not taken into account for the final analysis. Then, at
146 each harvest time, mass loss was compared across treatments using ANOVA model for split-

147 plot design (Logan, 2011). Soil conditions (soil vs compost) was the main between-block
148 factor whereas type of filter (plastic vs cellulose) and use (smoked vs unsmoked) were the
149 within-block factors, and block was included as a random factor. For the additional leaching
150 experiment, mass loss by leaching was compared across treatments (filter types and use) using
151 a two-way ANOVA model. All data was checked for normal distribution and
152 homoscedasticity of residuals. As both assumptions were met, analyses were made on non-
153 transformed data. Finally, a first order kinetic decay model ($R_t = R_0 \times e^{-kt}$), in which R_t is
154 the remaining mass at time t and k (d^{-1}) the decomposition constant, was fitted to the
155 experimental data. The estimation of equation parameters was used to estimate the half-life of
156 filters ($T_{50\%}$) and their total decomposition time ($T_{99\%}$). All statistical analyses were
157 performed using the R software, version 2.14.1 (R Core Team, 2014).

158

159 RESULTS

160 *Effect of soil conditions*

161 The decomposition of cigarette filters was strongly affected by soil conditions. At the end of
162 the experiment, 92% of initial mass was remaining when filters decomposed on the soil
163 surface, compared to 58% in the compost, on average across all other treatments. The effect
164 of soil condition was strongly significant ($p < 0.001$) and explained the largest part of the
165 variability in the dataset as indicated by the high mean squares values (Table 1).

166

167 *Effect of filter type*

168 There was a strong effect of filter type on decomposition (Table 1), with cellulose filters
169 decomposing significantly faster than plastic filters. The effect of filter type on decomposition
170 depended on soil conditions as indicated by the significant interaction term (Table 1). Indeed,
171 on the soil surface, filter decomposition was lower and the differences between filter types

172 were not significant. However, in the compost, cellulose filters decomposed clearly more
173 rapidly than plastic filters, with a remaining mass of 33.5% and 83.1% after 157 days for
174 cellulose and plastic filters, respectively, across all filter use treatments.

175

176 *Effect of filter use*

177 Whether filters had been previously smoked or not had no direct effect on decomposition but
178 filter use interacted with other experimental factors. On the soil surface, both filter types
179 decomposed faster when smoked, with 89.1% of mass remaining for smoked filters, compared
180 to 95.4% for unsmoked filters, on average across both filter types (Fig. 1). Conversely, in the
181 compost, smoked filters decomposed more slowly than unsmoked filters, especially for
182 cellulose filters that had a remaining mass of 16.1% for unsmoked filters compared to 50.8%
183 when filters were previously smoked (Fig. 1).

184

185 *Filter mass loss through leaching*

186 The percentage of mass lost through leaching was affected by the type of filters ($p < 0.001$),
187 with greater leaching for plastic than cellulose filters. Filter use also had a significant effect (p
188 < 0.001), with more leaching for smoked than unsmoked filters (Fig. 2). The interaction
189 between filter types and use was also significant ($p < 0.001$), with a 22-fold increase in
190 leaching for cellulose filters when smoked, increasing from 0.4% to 8.9% of initial mass lost,
191 while the increase was less than two-fold for plastic filters, increasing from 6.6% to 11% of
192 initial mass lost (Fig. 2).

193

194 *First order kinetic decay model for filter decomposition*

195 The first order kinetic decay models fitted to the remaining mass of smoked filters showed
196 that cellulose filters in the compost had the shortest half-life ($T_{50\%}$) with a $T_{50\%}$ of 0.4 year,

197 compared to 2 years for both cellulose and plastic filters decomposing on the soil surface
198 (Table 2). The estimation of the total decomposition time ($T_{99\%}$) suggests that cellulose filters
199 would take 2.8 years to be entirely decomposed in a compost, compared to 7.5 years for
200 plastic filters. On the soil surface, the estimated total decomposition time was 13.3 and 14
201 years for cellulose and plastic filters respectively.

202

203 DISCUSSION

204 *Importance of disposal environments*

205 According to our first hypothesis, filter decomposition varied depending on filter type, with
206 cellulose filters decomposing significantly faster on average than plastic ones. This advantage
207 of cellulose filters over the plastic ones for decomposition was expected given the resistance
208 of plastic to microbial decomposition (Zugenmaier, 2004) while cellulose molecules are
209 known to be readily metabolized by microbial enzymes (Berg and McLaugherty, 2008).
210 However, this advantage of cellulose over plastic filters for decomposition largely depended
211 on the decomposition location. Indeed, when disposed within the compost, cellulose filters
212 decomposed much more rapidly than the plastic ones, but this advantage was absent when
213 filters were decomposing on the soil surface. Such faster decomposition in the compost was
214 expected as litter decay is typically increased by litter burial (Coulis et al., 2016; Joly et al.,
215 2017; Withington and Sanford, 2007), which favors the moisture conditions, and by the
216 higher nutrient availability (Berg and McLaugherty, 2008), which permits nitrogen
217 immobilization from the decomposition environment to the decaying litter (Bonanomi et al.,
218 2017, 2015). In turn, while the limited decomposition observed on the soil surface was
219 expected given the lower nutrient availability and harsher climatic conditions, the complete
220 lack of difference in decomposition between cellulose and plastic filters on the soil surface is
221 unexpected and noteworthy. This context-dependency lies in the fact that cellulose filters

222 decomposed much more slowly on the soil surface, while plastic filter decomposition was
223 hardly affected by the disposal environment. This limited context-dependence for plastic
224 filters was previously documented by (Bonanomi et al., 2015) who reported no difference in
225 plastic filter decomposition among different incubation sites varying from sand to grassland.
226 Although this equal decomposition of cellulose and plastic items might be an extreme case
227 given the rather dry conditions at this Mediterranean site during the decomposition period,
228 limiting the microbial activity, and may not last at later stages of decomposition, it still
229 highlights the context-dependency of the advantage of cellulose items for waste
230 decomposition. In addition, such harsh conditions for microbial decomposers are quite
231 common in places where cigarette butts accumulate such as roadsides and beaches. In view of
232 our results, the shift from plastic to cellulose filters, should not exempt citizens from
233 collecting and disposing their waste in appropriate collecting systems.

234

235 *Intact versus used material*

236 In line with our third hypothesis, the decomposition of both filter types differed when filters
237 had been used in a cigarette prior to decomposition, and this effect interacted with filter type
238 and disposal environments. When filters decomposed in a compost, prior use of cellulose
239 filters reduced their decomposition by 41.4%. In contrast, decomposition of plastic filters did
240 not differ between used and new filters. This suggests that filter-use, charging the filter with
241 tar and chemical compounds, increases the recalcitrance of the waste and limit microbial
242 decomposition. However, this microbial inhibition was not visible on the soil surface, where
243 mass losses were higher for used filters of both filter types. However, given the low
244 decomposition on the soil surface and the fact that both filter types were similarly affected, it
245 is unlikely that the use of filter favored microbial activity under these conditions. Instead, this
246 increased mass loss may be due to the fact that the compounds charged on the filters after use

247 could be readily lost through leaching. This hypothesis is supported by our additional leaching
248 experiment for which we observed substantial mass losses of undecomposed filters, due to
249 leaching, that were significantly higher for used filters (Figure 2). The ecological impact of
250 these cigarette butt leachates has already been considered for aquatic organisms (Dieng et al.,
251 2013). However, attention must be paid to the impact of these leachates on soil organisms,
252 and particularly those involved in organic matter decomposition, as their abundance and
253 activity may be altered by leachate quality (Joly et al., 2016).

254

255 *Conclusions*

256 Our study provides clear evidence that cellulose cigarette filters provide an important
257 advantage over plastic regarding decomposition upon disposal. Using first order kinetic decay
258 models for used filters over the short incubation period of our experiment, we estimated that
259 used plastic filters take 7.5 to 14 years to disappear, in a compost and on the soil surface,
260 respectively. In contrast, we estimated that used cellulose filters take 2.3 to 13 years to
261 disappear, in a compost and at soil surface, respectively. Since mass loss through leaching and
262 decomposition of the paper wrapped around the filter could not be separated from the
263 decomposition of the core filter, these estimations might underestimate the expected residence
264 time of these wastes upon disposal. The advantage of cellulose filters for decomposition
265 greatly varies depending on disposal environments and we advocate that the transition from
266 plastic to cellulose filters should not exempt citizens from collecting and disposing their waste
267 in appropriate collection systems. In addition, our results suggest that composting may not be
268 a potential alternative, as the estimated time for full disappearance of used cellulose filters
269 (2.3 years) is longer than usual composting cycles. This decreased decomposition for used
270 cellulose filters indicates that biodegradability tests should consistently consider the effect of
271 product use on its subsequent decomposition for all types of waste. Complementary studies

272 are needed to evaluate the persistence of compounds accumulating in products before
273 composting can be considered as a viable waste management system.

274
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284

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340

341 **Table 1.** Results of ANOVA testing for the effects of disposal environment, filter type and
 342 their use on mass loss after 157 days of decomposition.

Source of variance	df	Mean squares	F-value	p-value
<i>Between blocks</i>				
Disposal environment	1	13006	62.0	<0.001
Residuals	9	1887	210.0	
<i>Within blocks</i>				
Filter type	1	7427	71.2	<0.001
Use	1	190	1.8	0.187
Disposal environment x Filter type	1	7553	72.4	<0.001
Disposal environment x Use	1	1404	13.5	<0.01
Filter type x Use	1	969	9.3	<0.01
Disposal environment x Filter type x Use	1	1090	10.5	<0.01
Residuals	28	104		

343

344

345 **Table 2.** Parameters of first order kinetic decay models fitted to mass loss data for the two
 346 types of smoked filters under different disposal environments. For each treatment combination,
 347 estimations of half-life ($T_{50\%}$) and total decomposition time ($T_{99\%}$) were made from models
 348 (n=24).

Disposal environments	Filter type	Decomposition constant (1/year)	Standard error of the regression	$T_{50\%}$ days /years	$T_{99\%}$ days/years	<i>p</i> -value
Soil	Cellulose (smoked)	0.0009	0.0001	733 / 2	4871 / 13	<0.001
	Plastic (smoked)	0.0009	0.0001	772 / 2	5131 / 14	<0.001
Compost	Cellulose (smoked)	0.0045	0.0007	154 / 0.4	1026 / 2.8	<0.001
	Plastic (smoked)	0.0017	0.0002	410 / 1.1	2726 / 7.5	<0.001

349

350 **Fig. 1:** Decomposition dynamic of cigarette filters on the soil surface (left) and in the
351 compost (right). The cellulose (circle) and plastic (square) filters were either smoked (filled
352 symbols) or unsmoked (empty symbols) before the decomposition experiment. Different
353 letters indicate significant differences within each date (Tukey HSD test).

354

355 **Fig. 2:** Percentage of filter mass lost through leaching. Different letters indicated significant
356 differences among treatments (Tukey HSD test).