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

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ABSTRACT

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

KEYWORDS
Cigarette butt – Compostable – Municipal solid waste – Biodegradable

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

In the only peer-reviewed publication that assessed the decomposition of conventional cigarettes filters, Bonanomi et al. (2015) reported that while the paper wrapped around the filter was readily decomposed, the plastic part was mostly unaffected after two years of decomposition. In turn, the OCB® brand for instance, that sells filters for hand-rolling cigarettes, advertises an almost complete decomposition of cellulose filters in 28 days. However, these results, coming from a test made by an independent laboratory following the 301B biodegradability protocol of the Organization for Economic Cooperation and Development (OECD), have not been published, and do not compare with the decomposition of conventional plastic filters, making it impossible to evaluate the advantage of cellulose filters over the plastic ones. Particularly, given the predominant control of environmental conditions on biotic litter decomposition (Berg and McClougherty, 2008), the decomposition of the cellulose filters is likely to vary widely depending on their disposal environment. In contrast, environmental conditions were shown to have no effect on decomposition of plastic
cigarette filters (Bonanomi et al., 2015). Consequently, in composts, where environmental conditions are prone to microbial activity, the relative advantage of cellulose filters over the plastic ones may be reinforced. Moreover, the goal of the OECD protocol is to evaluate the biodegradability of the substance out of which the product is made without necessarily taking into account its previous use. Such potential decoupling of the test from realistic conditions could importantly limit the validity of the results. Indeed, once the cigarette is smoked, the filter gets charged with a large variety of compounds including tars, carcinogenic compounds and numerous metals (Hoffmann, 1997; Moerman and Potts, 2011), which leads to an increased toxicity of filters for wildlife (Dieng et al., 2013; Slaughter et al., 2011; Suárez-Rodríguez et al., 2013) as well as microorganisms (Micevska et al., 2006). Consequently, the microbial decomposition of cellulose filters is likely to be substantially decreased for smoked filters, decreasing the relative advantage of cellulose filters over plastic ones.

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

*Filters*

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

*Experimental design*

Litterbags containing all types of filters were placed to decompose in the experimental field of the Center of Evolutionary and Functional Ecology, on February 21, 2014, under two conditions, either directly on the soil surface of a Mediterranean old-field, or buried in a plastic container containing compost. The compost consisted in a mixture of green manure made of ramial chipped wood and mature compost to ensure microbial inoculation. The first condition corresponds to the scenario where butts are thrown on the soil and remain there to decompose, while the second condition corresponds to the scenario where butts would be collected and composted with other organic waste. The climatic conditions at the study site are typically Mediterranean, with a mean annual temperature of 15°C and a mean annual precipitation of 570 mm (average of the 1981-2010 period). Over the 5.4 months of the experiment, cumulated precipitation was 124 mm, with an average temperature of 17.4°C. The experimental design included four factors: filter type (plastic vs cellulose), use (smoked...
vs unsmoked), soil conditions (soil vs compost) and length of incubation (five harvests). As all factors were crossed, we obtained 40 treatment combinations. For each combination, six replicates were placed in six separate blocks and litterbags were randomized within each block. The six replicates of the smoked filters consisted of three filters from each smoker to allow testing for the smoker effect. To ensure the start of microbial decomposition both on the soil surface and in the compost, all blocks were watered at the beginning of the experiment, with additions of 20 mm precipitation pulses. Additionally, to ensure optimal conditions for microbial decomposition in the compost, the plastic containers were rewetted every month with additions of 10 mm precipitation pulses. Litterbags were harvested at five different times (2, 4, 8, 16, 32 weeks) after the start of the experiment. At each harvest, filters were cleaned to remove soil particles, dried at 60 °C for 48 h and weighed to determine the mass loss. In order to assess the amount of mass loss due to leaching for all filter treatments (plastic and cellulose filters, both smoked and unsmoked), we ran an additional leaching experiment. To do so, 10 filters of each filter treatments were dried at 60°C for 48h, weighed and placed separately in a Falcon® tube with 15 ml of deionized water placed on a rotator spinning at 8 rpm for 24 h (Joly et al., 2016). Filters were then dried at 60°C for 48h and weighed to determine mass loss. For both experiments, mass loss was expressed in percentage of initial litter oven-dry weight.

**Data analysis**

First, to ensure that the decomposition process was not affected by the identity of the smoker, the smoker effect (n = 3 per smoker) was evaluated separately using a one-way ANOVA and then with the others factor using a complete ANOVA model. As it was not significant in any case (p > 0.05), this factor was finally not taken into account for the final analysis. Then, at each harvest time, mass loss was compared across treatments using ANOVA model for split-
plot design (Logan, 2011). Soil conditions (soil vs compost) was the main between-block factor whereas type of filter (plastic vs cellulose) and use (smoked vs unsmoked) were the within-block factors, and block was included as a random factor. For the additional leaching experiment, mass loss by leaching was compared across treatments (filter types and use) using a two-way ANOVA model. All data was checked for normal distribution and homoscedasticity of residuals. As both assumptions were met, analyses were made on non-transformed data. Finally, a first order kinetic decay model \( R_t = R_0 \times e^{-kt} \), in which \( R_t \) is the remaining mass at time \( t \) and \( k \) (d\(^{-1}\)) the decomposition constant, was fitted to the experimental data. The estimation of equation parameters was used to estimate the half-life of filters (\( T_{50\%} \)) and their total decomposition time (\( T_{99\%} \)). All statistical analyses were performed using the R software, version 2.14.1 (R Core Team, 2014).

RESULTS

Effect of soil conditions

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

Effect of filter type

There was a strong effect of filter type on decomposition (Table 1), with cellulose filters decomposing significantly faster than plastic filters. The effect of filter type on decomposition depended on soil conditions as indicated by the significant interaction term (Table 1). Indeed, on the soil surface, filter decomposition was lower and the differences between filter types
were not significant. However, in the compost, cellulose filters decomposed clearly more rapidly than plastic filters, with a remaining mass of 33.5% and 83.1% after 157 days for cellulose and plastic filters, respectively, across all filter use treatments.

Effect of filter use

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

Filter mass loss through leaching

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

First order kinetic decay model for filter decomposition

The first order kinetic decay models fitted to the remaining mass of smoked filters showed that cellulose filters in the compost had the shortest half-life ($T_{50\%}$) with a $T_{50\%}$ of 0.4 year,
compared to 2 years for both cellulose and plastic filters decomposing on the soil surface (Table 2). The estimation of the total decomposition time ($T_{99\%}$) suggests that cellulose filters would take 2.8 years to be entirely decomposed in a compost, compared to 7.5 years for plastic filters. On the soil surface, the estimated total decomposition time was 13.3 and 14 years for cellulose and plastic filters respectively.

DISCUSSION

Importance of disposal environments

According to our first hypothesis, filter decomposition varied depending on filter type, with cellulose filters decomposing significantly faster on average than plastic ones. This advantage of cellulose filters over the plastic ones for decomposition was expected given the resistance of plastic to microbial decomposition (Zugenmaier, 2004) while cellulose molecules are known to be readily metabolized by microbial enzymes (Berg and McClougherty, 2008). However, this advantage of cellulose over plastic filters for decomposition largely depended on the decomposition location. Indeed, when disposed within the compost, cellulose filters decomposed much more rapidly than the plastic ones, but this advantage was absent when filters were decomposing on the soil surface. Such faster decomposition in the compost was expected as litter decay is typically increased by litter burial (Coulis et al., 2016; Joly et al., 2017; Withington and Sanford, 2007), which favors the moisture conditions, and by the higher nutrient availability (Berg and McClougherty, 2008), which permits nitrogen immobilization from the decomposition environment to the decaying litter (Bonanomi et al., 2017, 2015). In turn, while the limited decomposition observed on the soil surface was expected given the lower nutrient availability and harsher climatic conditions, the complete lack of difference in decomposition between cellulose and plastic filters on the soil surface is unexpected and noteworthy. This context-dependency lies in the fact that cellulose filters
decomposed much more slowly on the soil surface, while plastic filter decomposition was hardly affected by the disposal environment. This limited context-dependence for plastic filters was previously documented by (Bonanomi et al., 2015) who reported no difference in plastic filter decomposition among different incubation sites varying from sand to grassland. Although this equal decomposition of cellulose and plastic items might be an extreme case given the rather dry conditions at this Mediterranean site during the decomposition period, limiting the microbial activity, and may not last at later stages of decomposition, it still highlights the context-dependency of the advantage of cellulose items for waste decomposition. In addition, such harsh conditions for microbial decomposers are quite common in places where cigarette butts accumulate such as roadsides and beaches. In view of our results, the shift from plastic to cellulose filters, should not exempt citizens from collecting and disposing their waste in appropriate collecting systems.

*Intact versus used material*

In line with our third hypothesis, the decomposition of both filter types differed when filters had been used in a cigarette prior to decomposition, and this effect interacted with filter type and disposal environments. When filters decomposed in a compost, prior use of cellulose filters reduced their decomposition by 41.4%. In contrast, decomposition of plastic filters did not differ between used and new filters. This suggests that filter-use, charging the filter with tar and chemical compounds, increases the recalcitrance of the waste and limit microbial decomposition. However, this microbial inhibition was not visible on the soil surface, where mass losses were higher for used filters of both filter types. However, given the low decomposition on the soil surface and the fact that both filter types were similarly affected, it is unlikely that the use of filter favored microbial activity under these conditions. Instead, this increased mass loss may be due to the fact that the compounds charged on the filters after use
could be readily lost through leaching. This hypothesis is supported by our additional leaching experiment for which we observed substantial mass losses of undecomposed filters, due to leaching, that were significantly higher for used filters (Figure 2). The ecological impact of these cigarette butt leachates has already been considered for aquatic organisms (Dieng et al., 2013). However, attention must be paid to the impact of these leachates on soil organisms, and particularly those involved in organic matter decomposition, as their abundance and activity may be altered by leachate quality (Joly et al., 2016).

Conclusions

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

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REFERENCES


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

<table>
<thead>
<tr>
<th>Source of variance</th>
<th>df</th>
<th>Mean squares</th>
<th>F-value</th>
<th>p-value</th>
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<td><strong>Between blocks</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Disposal environment</td>
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<td>13006</td>
<td>62.0</td>
<td>&lt;0.001</td>
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<td>Residuals</td>
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<td>1887</td>
<td>210.0</td>
<td></td>
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<tr>
<td><strong>Within blocks</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Filter type</td>
<td>1</td>
<td>7427</td>
<td>71.2</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>Use</td>
<td>1</td>
<td>190</td>
<td>1.8</td>
<td>0.187</td>
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<tr>
<td>Disposal environment x Filter type</td>
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<td>7553</td>
<td>72.4</td>
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<tr>
<td>Disposal environment x Use</td>
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<td>1404</td>
<td>13.5</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Filter type x Use</td>
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<td>969</td>
<td>9.3</td>
<td>&lt;0.01</td>
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<tr>
<td>Disposal environment x Filter type x Use</td>
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<td>1090</td>
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<td>&lt;0.01</td>
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<td>Residuals</td>
<td>28</td>
<td>104</td>
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Table 2. Parameters of first order kinetic decay models fitted to mass loss data for the two types of smoked filters under different disposal environments. For each treatment combination, estimations of half-life ($T_{50\%}$) and total decomposition time ($T_{99\%}$) were made from models (n=24).

<table>
<thead>
<tr>
<th>Disposal environments</th>
<th>Filter type</th>
<th>Decomposition constant (1/year)</th>
<th>Standard error of the regression</th>
<th>$T_{50%}$ days/years</th>
<th>$T_{99%}$ days/years</th>
<th>p-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soil</td>
<td>Cellulose (smoked)</td>
<td>0.0009</td>
<td>0.0001</td>
<td>733/2</td>
<td>4871/13</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td>Plastic (smoked)</td>
<td>0.0009</td>
<td>0.0001</td>
<td>772/2</td>
<td>5131/14</td>
<td>&lt;0.001</td>
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<tr>
<td>Compost</td>
<td>Cellulose (smoked)</td>
<td>0.0045</td>
<td>0.0007</td>
<td>154/0.4</td>
<td>1026/2.8</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td>Plastic (smoked)</td>
<td>0.0017</td>
<td>0.0002</td>
<td>410/1.1</td>
<td>2726/7.5</td>
<td>&lt;0.001</td>
</tr>
</tbody>
</table>
Fig. 1: Decomposition dynamic of cigarette filters on the soil surface (left) and in the compost (right). The cellulose (circle) and plastic (square) filters were either smoked (filled symbols) or unsmoked (empty symbols) before the decomposition experiment. Different letters indicate significant differences within each date (Tukey HSD test).

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