© 2020. This manuscript version is made available under the CC-BY-NC-ND 4.0 license http://creativecommons.org/licenses/by-nc-nd/4.0/

# Metrics for quantifying the circularity of bioplastics: the case of bio-based and biodegradable mulch films

Francesco Razza<sup>a</sup>, Cristiana Briani<sup>b</sup>, Tony Breton<sup>c</sup>, Diego Marazza<sup>d</sup>

<sup>a</sup> Novamont S.p.A. - Ecology of Product and Environmental Communication, Piazz.le Donegani 4, 05100 Terni, Italy

<sup>b</sup> CIRSA Centro Interdipartimentale di Ricerca per le Scienze Ambientali, Via S. Alberto 163, 48123 Ravenna, Italy

<sup>c</sup>Novamont S.p.A. – Via Fauser 8, 28100 Novara, Italy

<sup>d</sup>Department of Physics, University of Bologna, Viale B. Pichat 6/2, 40127 Bologna, Italy

## 15 Abstract

3

4

11

12 13

14

16 The concept of circularity and its quantification through the Material Circularity Indicator 17 (MCI) is well established for traditional plastic products. In this paper a methodological approach for calculating the circularity of bio-based and biodegradable (BB) products is 18 19 proposed and applied to BB mulch films. BB products are different from traditional 20 products in as much as they are sourced and regenerated (recycled) not through technical 21 cycles but the biological loop. The suggested method is an adaptation of the MCI where 22 two major changes were made: (i) the mass of the bio-based component corresponds to the 23 recycled material in input and (ii) the mass of the bio-based component leaving the system 24 through composting or biodegradation in soil is accounted as recycled. The modified MCI supports the Eco-design of innovative BB products and allows for the comparison of their 25 26 circularity taking into account the biological source and the expected end of life process 27 such as biodegradation. To demonstrate the adaptation, the method has been applied to BB 28 mulch film. Results showed that the MCI of a biodegradable mulch film, characterized by 29 an average bio-based feedstock content of 30% is  $0.37 \pm 0.04$  in a 0-1 scale. For BB mulch 30 film, the amount of bio-based feedstock is the most sensitive factor and controls linearly 31 the value of the MCI.

32

*Keywords:* circularity indicators, circular economy, bioplastics, biodegradable
 mulch film, bio-based product, biodegradation

36	1 Introduction
37	1.1 The case study of mulch films
38	1.2 Goal of the paper7
39	2 Materials and Methods7
40	2.1 MCI accounting according to the EMF methodology7
41	2.2 MCI accounting for bio-based and biodegradable (BB) products 10
42	2.3 MCI calculation for mulch films: scope, inventory and assumptions 14
43	2.4 Sensitivity analysis
44	3 Results
45	3.1 Sensitivity analysis
46	4 Discussion
47	5 Conclusions
48 49	

Abbreviations		
BB	Biodegradable and bio-based	
CE	Circular Economy	
d.m.	Dry matter	
EMF	Ellen MacArthur Foundation	
LCA	Life Cycle Assessment	
LDPE	Low-Density Poly-Ethylene	
MCI	Material Circularity Indicator	
NRF	Non-Restorative Flows	
PBAT	Polybutylene adipate terephthalate	
PE	Poly-Ethylene	
PLA	Polylactic acid	

#### 51 **1 Introduction**

52 To overcome today's unsustainable model of 'take-make-dispose' and its related risks 53 such as hikes in raw material prices, pressures on the environment, shortage of global 54 resources and waste sinks, a circular approach needs to be applied. It is a new regenerative 55 economic view, based on a balance between economy, environment and society, a total 56 resource efficiency and a Zero Emission Strategy that aims to maximize products value 57 with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with 58 structural changes in environmental legislation, new logistics, technologies and sharing 59 schemes, the Circular Economy (CE) approach which is regenerative by design, aims at 60 closing materials loops, *i.e.* at reducing virgin materials input and waste output.

61 In December 2015, the European Commission developed an Action Plan for Circular 62 Economy (European Commission, 2015), where plastic was considered a priority to be 63 tackled. In January 2018, an EU Plastic Strategy (European Commission, 2018) was 64 adopted, in order to react to the increasing environmental problems concerning plastic 65 production, consumption, use and disposal along the same lines of the CE approach. Two 66 fundamental steps to increase the circularity of different plastic products are (i) the 67 abandonment of fossil fuels, *i.e.* currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development 68 69 of easily recyclable products which are recycled. Today, in EU the share of plastics 70 collected for recycling is 30% while the use of recycled plastics is just 6% (European 71 Commission, 2018).

Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and principles. This is true as long as the supply of renewable raw materials, generally from agriculture, is based on a sustainable approach and the conversion processes along the supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA)
perspective (EPLCA – European Platform on LCA). While traditional plastics can be
mechanically recycled or incinerated with energy recovery, BB plastic products offer new
recycling routes in waste management, due to their biodegradability. Organic recycling
(through composting or anaerobic digestion) or in the case of specific applications such as
agricultural mulch films, biodegradation in the environment, offer additional recovery
options resulting in less wastes.

83 Nevertheless, the research and development of innovative products, such as the BB 84 products, implies the development of methodologies and metrics capable of measuring 85 their circularity. Without this it is not possible to achieve measurable results and 86 improving actions, as well as provide unequivocal references for comparisons of products 87 of the same type/category. In 2015 the Material Circularity Indicator (MCI) was 88 developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify 89 the regeneration of a product's material flow and is considered one of the few, among 90 sixteen CE indexes suiting a micro-scale assessment of circularity at product or company level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled 91 92 materials. Furthermore, recovery and recycling through the biological cycle offered by 93 industrial composting, anaerobic digestion or biodegradation in natural environments are 94 not considered as end of life options. In order to apply the MCI system to BB plastic 95 products, the development of an enhanced methodology is necessary.

96 The approach proposed by the authors allows to quantify the circularity of BB plastic 97 products (*e.g.* starch-based bioplastics) and to make comparisons with equivalent 98 traditional plastic products. To demonstrate the applicability of the proposed method a 99 computational example for mulch film products is provided. In so doing so, the paper 100 aims at contributing to the Eco-design of these innovative products.

#### 101 1.1 The case study of mulch films

102 Plastic mulch films represent an important agronomical technique well established for the 103 production of many crops thanks to numerous agronomical advantages such as: increased 104 yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and 105 reduced use of pesticides; early crop production and reduced soil moisture loss 106 (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has 107 increased year-by-year, reaching a current global market estimated at 1.4 Mt, mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017)  $\,$ , and covering 80,000 km<sup>2</sup> 108 109 of agricultural surface (0.6% of the global arable land). The mulch film market in Europe 110 is estimated by Agriculture Plastic & Environment and by the European Bioplastic 111 Associations at 76-80 kt. The most used raw material is Poly-Ethylene (PE) in its different 112 forms, due to its processability, chemical resistance, high durability and flexibility 113 (Kasirajan and Ngouajio, 2012).

114 Despite these benefits, manifold environmental and agronomic problems have been 115 pointed out. After its useful life – which in general does not exceed 1 to 3 months – the 116 mulch film has to be removed and properly disposed of, a time-consuming and costly 117 procedure. The recovered film is usually heavily contaminated with soil and organic 118 residues, making mechanical recycling technically difficult and not a cost-efficient 119 solution (Briassoulis et al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most 120 common end of life of collected films in Europe is still landfilling (about 50%), followed 121 by energy recovering and finally mechanical recycling (Le Moine, 2014). Recent Chinese 122 prohibition (January 2018) to import different types of wastes is heavily impacting the 123 European agricultural plastic waste management, highlighting the difficulty in properly 124 recycling this type of plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but disposed of by burning in the field or by uncontrolled landfilling or left 125

directly in the (agricultural) soils, causing serious environmental concerns. An example is
the "White pollution" phenomena described in the Xinjiang Autonomous Region (China),
in which the residual plastic film can reach 200 kg/ha in the top soil with detrimental
effects on soils' quality, health and fertility (Liu, He, & Yan, 2014; Gao *et al.*, 2019;
Steinmetz *et al.*, 2016).

131 As a reaction, there has been significant research into novel materials especially related to 132 biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation 133 in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). The term "bio-mulch film" brings together several types of both bio-based and fossil oil-134 135 based biodegradable polymers and blends of them, such as polylactic acid (PLA), 136 polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or 137 copolymers. They biodegrade when exposed to bioactive environments such as soil and 138 compost (Kasirajan et al., 2012) which means that they can be left in situ to be fully 139 biodegraded after being used. However, their biodegradability must be proved by 140 accredited certification bodies and standardized procedures.

141 The EN 17033:2018 is a new European Norm (standard) concerning "Plastics -142 Biodegradable mulch films for use in agriculture and horticulture - Requirements and test 143 methods", which sets the necessary tests and limits to define biodegradability, 144 performances and environmental impacts of BB much films. The material is considered 145 completely biodegradable if it achieves a complete biodegradation (absolute or relative to 146 the reference material) in a test period no longer than 24 months (mineralization into 147 CO<sub>2</sub>). Additionally, a control of constituents (such as metals) and eco-toxicity testing 148 (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test 149 with soil microorganisms) were required. A certified mulch film guarantees that the 150 product will completely biodegrade in the soil without adversely impacting on the 151 environment.

#### 152 *1.2 Goal of the paper*

The goal of the paper is to provide a general and common metric to measure the circularity of a bio-based and biodegradable (BB) product and to apply the methodology at product level to a category of products, namely bio-based and biodegradable mulch films.

156 2 Materials and Methods

#### 157 2.1 MCI accounting according to the EMF methodology

158 The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation 159 (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number 160 that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production 161 provides for the exclusive use of virgin raw materials that turn into waste at the end of the 162 use phase of the product. Vice-versa, pure circularity includes the use of recycled 163 materials and does not produce wastes (regenerative streams). Circularity can be achieved 164 in different ways: as for the purpose of this paper, only recycling will be considered since 165 reuse is not an option for thin biodegradable mulch films. Since the method considers only 166 mass flows, the recycling corresponds to the recovery of materials for the original purpose 167 or for other purposes and excludes energy recovery, considered as a loss of materials equal 168 to landfill disposal. The materials recovered feed back into the process as recycled 169 feedstock.

The MCI methodology differentiates 'technical cycles' from 'biological cycles', modelling only the former. The first contains products and materials re-entering into the system (market) with the highest possible qualities and for as long as possible (thanks to reuse, repair, refurbishment and recycling) and the latter includes biological materials used

174 in cascade until their restoration into the biosphere and the re-constitution of natural 175 resources.

176 The material flows associated to the production of a generic technical cycle from non-177 renewable sources are summarized in Figure 1. The dashed lines indicate that recycled feedstock does not have to be sourced from the same product but can be acquired on the 178 179 market. With reference to Figure 1, the list of the parameters used in the EMF 180 methodology is reported in Table 1, while the equations relevant for the analysis carried 181 out in this paper are described in the following sections (Table 2, Chapter 2.2).







184 product (modified from Ellen MacArthur Foundation & Granta Design, 2015).

185

186 *Table 1:* Parameters and relative definitions used in the EMF methodology.

Parameter	Definition

М	Total mass of the product
Fn	Fraction of mass of a product's feedstock from
- <u>x</u>	recycled sources
<b>F</b>	Fraction of mass of a product's feedstock from
	reused sources
V	Mass of virgin feedstock used in a product
C	Fraction of mass of a product being collected to go
$C_R$	into a recycling process
C	Fraction of mass of a product going into
	component reuse
<b>F</b>	Efficiency of the recycling process used for the
	portion collected for recycling
<i>F</i> _	Efficiency of the recycling process used to produce
LF	recycled feedstock for a product
W	Total mass of unrecoverable waste associated with
,, , , , , , , , , , , , , , , , , , ,	a product
	Mass of unrecoverable waste (landfill, waste to
$W_0$	energy and any other type of process where the
	materials are no longer recoverable)
Wc	Mass of unrecoverable waste generated in the
<i>"</i> (	process of recycling parts of a product (after use)
WE	Mass of unrecoverable waste generated when
'' F	producing recycled feedstock for a product
X	Utility of a product, calculated as $X =$
<b>A</b>	$(L/L_{av})(U/U_{av})$

L	Actual average lifetime of a product
	Actual average lifetime of an industry-average
	product of the same type
	Actual average number of functional units
	achieved during the use phase of a product
	Actual average number of functional units
$U_{av}$	achieved during the use phase of an industry-
	average product of the same type

188 The Material Circularity Indicator is determined as follows:

189 where *LFI* is the Linear Flow Index measuring the flows of virgin materials and190 unrecoverable wastes associated to the examined product.

191 A function of the utility, -, is used to correct the *LFI*. The function *F* is chosen in 192 such a way that improvements of the utility of a product (e.g., by using it longer) have the 193 same impact on its MCI as a reuse of components, leading to the same amount of 194 reduction of virgin material use and unrecoverable waste. Setting a = 0.9, MCI takes, by 195 convention, the value 0.1 for a fully linear product (*i.e.*, *LFI* = 1) whose utility equals the 196 industry average (*i.e.*, X = 1). This leaves some margin to distinguish between processes 197 with a high linearity but different utilities.

198 2.2 MCI accounting for bio-based and biodegradable (BB) products

199 To apply the EMF methodology to BB products, formulas and flows (Figure 1 and Figure200 2) are adapted as it follows:

201 1. The fraction of the recycled feedstock,  $F_R$ , corresponds to the share of the bio-202 based feedstock content in the final BB product,  $F_{R(i)}$ . It is the ratio of the d.m. amount of bio-based feedstock per d.m. amount of the total mass of BB
product (EN 16785-2:2016).

- 205 2. The fraction of restorative mass going into a recycling process,  $C_R$ , corresponds 206 to the share of bio-based feedstock content in the BB product biologically 207 recovered (*e.g.* through composting) or biodegraded in the natural 208 environment, as it happens for specific applications (e.g. biodegradable mulch 209 film, *etc.*). It is the ratio of the d.m. amount of bio-based feedstock per d.m. 210 amount of the total mass of BB product that is biologically recycled.
- The modified scheme is shown in Figure 2. Table 2 lists the formulas as adapted to BBproducts.
- 213214
- **Table 2:** List of formulas as developed by EMF methodology compared to theproposed adaptation to BB products.

EMF methodology	Adaptation to BB products

The mass of fossil-based feedstock which may be contained in BB products (V) is obtained as a difference of the total mass (M) minus the bio-based fraction; in this case the  $F_R$  in the EMF methodology corresponds to the sum of the fractions of all the biobased feedstock/s used in manufacturing the BB product. Therefore, is the total bio-based feedstock mass in the product. In single-use products, such as mulch films, reuse is not considered for BB products, so that  $F_U = C_U = 0$ .

222  $W_F$  is the total amount of unrecoverable waste associated to the production of bio-based 223 feedstock used to produce BB products (i.e. the amount of uncoverable waste per unit of 224 BB product). Bio-based feedstocks such as starch and PLA generate non-restorative flows 225 which can be quantified. Such unrecoverable waste correspond to  $R_{(i)}$ , the specific amount 226 of waste generated within cradle-to-gate boundaries per unit of bio-based feedstock going 227 into manufacturing, and it is estimated through LCA studies. Thus all inputs from growth 228 and harvesting phases and the related wastes generated by fertilisers and pesticides are 229 here accounted.  $R_{(i)}$  can be easily found in specific literature or life cycle inventories 230 (LCI) present in LCA databases. In the calculation of  $W_F$ , also the efficiency of 231 manufacturing process of BB products  $E_P$  is considered, as the ratio of the overall bio-232 based feedstock content in the final BB product to the bio-based feedstock in input to the 233 manufacturing process.

The material flows associated to the production of a generic BB product are summarizedin Figure 2.



237 Figure 2: Description of material flows adaptation to BB products; in this paper, 238 the reuse flow is out of scope ( $C_U = F_U = 0$ ).

239 The biodegradation of bio-based feedstock does not imply the generation of waste  $W_C$  as it occurs in a standard mechanical recycling process. This implies that  $C_R$  and  $E_C$  (i.e. the 240 241 efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to 242 biological treatment (composting) or biodegraded in a natural environment, is fully 243 transformed in its chemical elements (C, H and O mainly) derived from the decomposition 244 of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et 245 al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites, 246 2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the 247 environment and are then available in the respective biogeochemical cycles. The (biodegradable) fossil portion behaves as well; consequently,  $W_C = 0$ . 248

Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular feedstock, since it derives from carbon stored for millions of years and extracted by man, not being part of the active and fast biogeochemical carbon cycle. This is accounted in the quantification of  $W_0$ , the mass of unrecoverable waste from use (*i.e.* the linear stream going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total amount of fossil-based feedstock.

Since  $W_F$  and  $W_C$  are associated to complete different processes and  $W_C$  is always equal zero, the double counting issue does not occur and the quantification of W and *LFI* is modified as reported in Table 2.

#### 258 2.3 MCI calculation for mulch films: scope, inventory and assumptions

The new formulas reported in Table 2 were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.

In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film is assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch,  $F_{(S)}$ , and 7% of a bio-based plasticizer,  $F_{(BP)}$ ), while the rest was assumed to consist of fossil feedstock (Figure 3). Since a generalized approach was used and no primary data were implemented, the information were extrapolated from literature; the main characteristics of the two examined products are presented in Table 3.





Figure 3: Examples of starch-based polymers; in this paper, the first option on the left (starch blend 30/70) has been chosen as representative of a BB mulch film. The figure considers a 100%-efficiency in every phase of production, so that the residues are equal to zero; the same assumption is done in this paper. \*TPS (Thermoplastic starch), starch content 75%; \*\*Ratio TPS/Polymer; modified from Institute of Bioplastics and Biocomposites, 2018.

## 279 *Table 3*: Key features representative of the BB mulch films.

	BB mulch film
Material	30% bio-based feedstock (23% starch + 7% bio- based plasticizer) + 70% fossil-based feedstock
Thickness (µm)	12
<b>Density</b> (g/cm <sup>3</sup> )	1.25
Weight (g/m <sup>2</sup> )	15.2
<b>Functional unit</b> (the covering of the agricultural land)	6000 m <sup>2</sup> /ha (the actual mulched soil in a hectare is generally equal to the 60% of the total area; Malinconico, 2017)

282 In the calculation of MCI for the BB mulch film, the adapted formulas were used together 283 with assumptions. As stated before, BB mulch films are blends of bio-based and fossil 284 based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE 285 mulch film that has to be removed and disposed of, the BB mulch film is left in soil where 286 it undergoes an ultimate biodegradation (so that  $C_R = 1$ ) with no waste (so that  $E_C = 1$ ), in 287 respect of the specific standard EN 17033:2018. As a result of polymers' decomposition, 288 the derived (biogenic) C, H and O finally return into biosphere (atmosphere, 289 microorganism biomass, organic material pool), and back into biogeochemical cycles in a 290 relatively short time ("Biogenic elements accounted as recycled" in Figure 2), with the 291 exception of humified compounds. Actually, also C, H and O deriving from fossil-based 292 sources undergo biodegradation but they are not considered as a regenerative flow 293 ("Waste from non-restorative flow" in Figure 2) and their "wastes" are indeed calculated 294 in  $W_0$ .

295 Applying a conservative approach,  $W_F$ , the waste generated by the production of each bio-296 based feedstock, is quantified considering a "cradle to gate" LCA study. The estimated 297 solid wastes  $R_{(i)}$  for the presented case study are related to the production of starch (F<sub>(S)</sub>), with an amount  $R_{(S)}$  of 0.014 kg of waste per kg of renewable feedstock (source: personal 298 299 communication A. Novelli), and to the production of the bio-based plasticizer ( $F_{(BP)}$ ), with 300  $R_{(BP)}$  equals to 0.025 kg waste/kg renewable feedstock, (source: US-LCI database 301 "Polylactide biopolymer resin at plant kg/RNA"). As assumed in Figure 3, the production 302 efficiency of BB product  $E_P$  (how much bio-based feedstock is needed for every unit of 303 BB product) is estimated equal to 1 and no unrecoverable wastes are generated by the 304 process.

In addition, an explorative sensitivity analysis has been performed regarding exclusively the amount of bio-based feedstock content of the BB mulch film, *i.e.* (*i.e.*,  $F_{(S)}$  +  $F_{(BP)}$ ), as shown in Figure 4 (Chapter 3).

308

#### 2.4 Sensitivity analysis

309 A sensitivity analysis was conducted for BB mulch film to examine the effects of 310 changing the main variables. Given a non-linear dependence of results on parameter 311 values, a Monte Carlo approach (see, e.g., Lloyd and Ries, 2008) has been adopted. The 312 model has been implemented using specifically written routines in the C++ programming 313 language. The model was run with 100,000 events for BB mulch film, where the value of 314 each parameter has been randomly chosen following a Gaussian distribution with a 315 standard deviation within a range of possible and realistic values (Table 5 and Error! 316 Reference source not found.; Figure 5 and Figure 6).

#### 317 **3 Results**

Considering the characteristics of the films (weight,  $g/m^2$ , or thickness,  $\mu m$ , and density, g/cm<sup>3</sup>) and the relative functional unit (6000 m<sup>2</sup>/ha, Table 3), it is possible to calculate a mass, *M*, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas reported in Table 2 (Chapter 2.2) are applied. Results are shown in Table 4.

Figure 4 shows how the value of the MCI varies according to the percentage variation ofthe bio-based feedstock in the total mass of the product.

- 324
- 325

Table 4: Resulting parameters in the calculation of MCI for BB mulch film.

Parameter	BB mulch film




**Figure 4:** MCI as a function of  $\Sigma F_{R(i)}$ , the percentage of all the bio-based 330 feedstock/s of the mulch film on mass basis (X-axis).

#### 332 3.1 Sensitivity analysis

The results of the sensitivity analysis are presented in the followings Table 5 and Figure 5 and Figure 6. The accuracy band is a fraction of the average and corresponds to a probability of 95%. It has been chosen in order to be representative of the variability of the product category, the BB mulch films. The simulation can thus be regarded as a system composed by a high number of companies, each producing films with different characteristics, that are accounted for in the accuracy band.

339 **Table 5:** Parameters used for the sensitivity analysis of the BB mulch film. (\*\*) The

340 *Accuracy Band is defined as twice the standard deviation of the distribution.* 

Variable name	Average	Accuracy Band (**)	Unit
М	1000.00	0%	kg
$F_{(S)}/F_{(BP)}$	3.29	10%	fraction
$F_{(S)} + F_{(BP)}$	0.30	30%	fraction
F <sub>U</sub>	0.00	0%	fraction
C <sub>U</sub>	0.00	0%	fraction
R <sub>(S)</sub>	0.014	100%	fraction
R <sub>(BP)</sub>	0.025	100%	fraction
Ec	1	0%	fraction
E <sub>P</sub>	0.95	10%	fraction
C <sub>R</sub>	1.00	0%	fraction

341

342





Figure 6: The most sensitive and relevant parameters in the calculation of the MCI of the BB mulch films.

#### 349 4 Discussion

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

(1) fossil-based feedstocks or component materials embodied in the BB products whateveris the final disposal (even biological recycling) shall be considered as non-restorative;

(2) bio-based component materials embodied in the BB product that go to biological
recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be
considered restorative as long as they flow through the biosphere safely, without any harm
to the environment (e.g. no toxicity effects).

361 (3) bio-based component materials embodied in the BB product that go to incineration and362 landfill shall be considered as non-restorative;

363 The justification of these rules is described in the following.

364 Fossil-based component materials in the product derive from deposits where they 365 remained stocked for a geological time scale. Once the product is mineralised, its fossil-366 based portion will be accounted as non-regenerative and therefore linear, due to its origin 367 (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological 368 cycles, like CO<sub>2</sub> in the atmosphere and other streams, since both fossil-based and bio-369 based component materials will physically and chemically behave the same, once 370 biodegraded. However, the source of the bio-based carbon was circular before its use 371 (concept of "carbon neutrality", equilibrium between the biogenic carbon released and the 372 carbon absorbed by plants) and will maintain its circularity provided that the carbon is released into the atmosphere at the same rate. The reason has its origin in the EMF general 373

provisions stating that "biologically sourced materials can only be considered part of a Circular Economy if materials are not used faster than they can be restored naturally" (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the bio-based components are still considered linear, maintaining consistency with EMF principles. Basically, a complete circularity for a BB product is satisfied when its renewable components are 100% bio-based and they go 100% to biological recycling or biodegraded in the environment (for specific application like mulch film).

381 As for provision (3), a material health rule has its origin in manyfold normative 382 definitions of the CE. In addition, the EMF definition of biological cycles is that of non-383 toxic materials which are restored into the biosphere and the CE is defined as such if it can 384 "eliminate the use of toxic chemicals". The need of a safety clause has been reviewed 385 under many aspects by Verberne (2016) and can be put as a postulate of the restoration 386 principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the 387 REACH Regulation (EC 1907/2006). In the specific case, the material complies with the 388 standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism 389 groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important 390 ecological processes maintaining soil functions, c) all relevant exposure pathways as soil 391 pore water, soil pore air and soil material.

A comprehensive approach for MCI calculation should also include non-restorative flows generated at upstream level like biomass growth, in the specific case corn, and biomass conversion processes like starch extraction and refining. Specifically these non-restorative flows correspond to the overall non-recyclable wastes associated to the bio-based feedstock supply thus non-recyclable waste from fertilizer and pesticide production, nonrecyclable scraps from conversion processes, etc. In this study such flows of nonrestorative waste coming from upstream manufacturing operations were included for the bio-based feedstocks ( $R_{(i)}$ ) used in manufacturing the BB mulch film applying "cradle to gate" LCA methodology. However, we observed that the inclusion of upstream unrecoverable waste does not significantly influence the MCI results in the chosen case study, since the respective amounts are small. The specific unrecoverable waste for starch and bio-based plasticizer (i.e. kg of waste/kg of bio-based feedstock) were estimated at 0.014 and 0.025, respectively.

405

The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation y = 0.89x + 0.1, where y is the MCI and x is the bio-based feedstock content, therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input is decisive.

411 Apart from the specific application analysed in this paper, the proposed MCI method can
412 be easily applied and calculated for any kind of BB product as long as the following
413 information are available:

The bio-based feedstock content, determined according to the standard EN 167852:2016, if the composition is known, or directly provided by the BB product manufacturer.

• The End of Life scenario of the studied BB product (real or hypothetical).

The amount of un-recoverable waste associated to the production of bio-based
feedstock contained in the BB product. They can be derived from LCA databases or other
specific sources.

#### 420 **5** Conclusions

421 Bioplastic market is steadily increasing. The value proposition of bio-based and422 biodegradable products is linked to:

423 1. the use of renewable feedstock (like starch and its derivates) instead of fossil oil or
424 natural gas;

425

426

2. the waste recovery through biological recycling, thanks to their ability to biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

427 The Material Circularity Indicator (MCI), developed by the EMF, is a metric for 428 quantifying "how much" a product is circular (MCI = 0, fully-linear product; MCI = 1, 429 completely circular product) thus it represents a valuable tool for product eco-design 430 purposes. However, it focuses solely on technical materials, mechanically recycled or 431 reused, leaving out bio-based feedstocks and related biological treatments such as 432 composting. Without common metrics it is not possible to pursue concrete actions, to 433 achieve measurable results and to provide unequivocal references for all products. This 434 research work aims at filling this gap through the development of a methodology coherent 435 with EMF MCI methodology but able to catch the specificities of bio-based and 436 biodegradable products and provide metrics for those innovative products. Direct uses are: 437 (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI 438 of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation:  $MCI_{(BB mulch})$ mulch = 0.89\*bio-based feedstock + 0.1.

The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles. Bioeconomy, thus also BB products, can provide valuable insights in transforming the current (linear) economy in a more circular one, however, the way the biomass is produced, processed and BB products are produced are

448 fundamental aspects to be properly assessed and monitored. This can be done using 449 specific methodologies like LCA. Within this context the proposed MCI has to be seen as 450 a complementary (quantitative) tool for further qualifying the sustainability of BB 451 products.

- 452
- 453

### Declaration of interest

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

457

#### 458 Acknowledgements

The authors thanks prof. Andrea Contin for the fruitful discussion and contribution to the sensitivity analysis, Francesco Degli Innocenti for providing valuable comments and feedback on the topics addressed by the paper and Alessandra Novelli for the general support in the MCI elaboration.

463

#### 464 **References**

- BASF, 2018. Biodegradable mulch film clarification of polymer fate in soil. CIPA Congress, Bordeaux/Arcachon, France, May 2018.
- Briassoulis, D., Giannoulis, A., 2018. Evaluation of the functionality of bio-based plastic mulching films. Polym. Test. 67, 99–109. https://doi.org/10.1016/j.polymertesting.2018.02.019
- De Lèpinau, P. and Arbenz, A., 2016. Economic and environmental impact of soil contamination in mulching film, Plasticulture, N° 136, 28-48.
- Ellen MacArthur Foundation & Granta Design, 2015. Circularity Indicators An approach to measure circularity Methodology. https://www.ellenmacarthurfoundation.org/assets/downloads/insight/Circularity-Indicators\_Methodology\_May2015.pdf.

- Ellen MacArthur Foundation, 2017. The New Plastic Economy: Rethinking the future of plastic & catalysing action.
- EN 16785-2:2016 Bio-based products Bio-based content Part 2: Determination of the bio-based content using the material balance method.
- EN 17033:2018 Plastics Biodegradable mulch films for use in agriculture and horticulture Requirements and test methods.
- EPLCA European Platform on LCA. https://eplca.jrc.ec.europa.eu/?page\_id=86
- Eubeler, J., Bernhanrd, M., Knepper, T., 2010. Environmental biodegradation of synthetic polymers II. Biodegradation of different polymer groups. Trends in Analytical Chemistry. 29, 1, 84-100
- European Commission, 2015. Closing the loop An EU action plan for the Circular Economy. COM(2015) 614 final. Brussels, 2.12.2015
- European Commission, 2018. A European Strategy for Plastics in a Circular Economy. COM(2018) 28 final. Brussels, 16.1.2018
- Figuier, B., 2016. Plasticulture in Europe, Plasticulture, N° 136, 20-28
- Gao, H., Yan, C., Liu, Q., Ding, W., Chen, B., Li, Z., 2019. Effects of plastic mulching and plastic residue on agricultural production: A meta-analysis. Sci. Total Environ. 651, 484–492. https://doi.org/10.1016/J.SCITOTENV.2018.09.105
- Ghisellini, P., Cialani, C., Ulgiati, S., 2016. A review on circular economy: The expected transition to a balanced interplay of environmental and economic systems. J. Clean. Prod. https://doi.org/10.1016/j.jclepro.2015.09.007
- Institute of Bioplastics and Biocomposites, 2018. Biopolymers Facts and Statistics. Hochschule Hannover, University of Applied Sciences and Arts. Edition 5, ISSN 2510-3431.
- Joos, F., Roth, R., Fuglestvedt, J. S., Peters, G. P., Enting, I. G., Bloh, W. V., ... and Friedrich, T., 2013. Carbon dioxide and climate impulse response functions for the computation of greenhouse gas metrics: a multi-model analysis. Atmospheric Chemistry and Physics, 13(5), 2793-2825.
- Kasirajan, S., Ngouajio, M., 2012. Polyethylene and biodegradable mulches for agricultural applications: a review. Agron. Sustain. Dev. 32, 501–529. https://doi.org/10.1007/s13593-011-0068-3
- Le Moine, B., 2014. Agri-plastics waste management: a voluntary commitment from the industry. Presented at: Agricultural Film 2014 International Conference on silage, mulch, greenhouse and tunnel films used in agriculture (15-17 September, Barcelona, Spain).

- Liu, E. K., He, W. Q., & Yan, C. R., 2014. 'White revolution' to 'white pollution' agricultural plastic film mulch in China. *Environmental Research Letters*, 9(9), 091001.
- Lloyd, S. M., & Ries, R., 2007. Characterizing, propagating, and analyzing uncertainty in life cycle assessment: A survey of quantitative approaches. *Journal of Industrial Ecology*, 11(1), 161-179.
- Lonca, G., Muggéo, R., Tétreault-Imbeault, H., Bernard, S., & Margni, M., 2018. A Bidimensional Assessment to Measure the Performance of Circular Economy: A Case Study of Tires End-of-Life Management. In *Designing Sustainable Technologies*, *Products and Policies* (pp. 33-42). Springer, Cham.
- Malinconico, M., 2017. Soil Degradable Bioplastics for a Sustainable Modern Agriculture. Green Chemistry and Sustainable Technology. Springer.
- Marten, E., Muller, R., and Deckwer W., 2003. Studies on the enzymatic hydrolysis of polyesters I. Low molecular mass model esters and aliphatic polyesters. Polymer Degradation and Stability, 80, 3, 485-501.
- Moreno, M. M., González-Mora, S., Villena, J., Campos, J. A., & Moreno, C., 2017. Deterioration pattern of six biodegradable, potentially low-environmental impact mulches in field conditions. *Journal of environmental management*, 200, 490-501.
- Mormile, P., Stahl, N., Malinconico, M., 2017. The World of Plasticulture, in: Malinconico, M. (Ed.), Soil Degradable Bioplastics for a Sustainable Modern Agriculture. pp. 1–21. https://doi.org/10.1007/978-3-662-54130-2 1
- OWS, 2018. Accumulation of (bio)degradable plastics in soil. CIPA Congress 2018, Archacon, May 29.
- Steinmetz, Z., Wollmann, C., Schaefer, M., Buchmann, C., David, J., Tröger, J., Muñoz, K., Frör, O., Schaumann, G.E., 2016. Plastic mulching in agriculture. Trading shortterm agronomic benefits for long-term soil degradation? Sci. Total Environ. 550, 690–705. https://doi.org/10.1016/J.SCITOTENV.2016.01.153
- Tamma, P., 2018. China's trash ban forces Europe to confront its waste problem, Politico, 2/21/2018.
- Touchaleaume, F., Martin-Closas, L., Angellier-Coussy, H., Chevillard, A., Cesar, G., Gontard, N., Gastaldi, E., 2016. Performance and environmental impact of biodegradable polymers as agricultural mulching films. Chemosphere 144, 433–439. https://doi.org/10.1016/j.chemosphere.2015.09.006
- Verberne, J.J.H., 2016. Building circularity indicators. Eindhoven University of Technology.

- Witt, U., Einig, T., Yamamoto, M., Kleeberg, I., Deckwer, W., Muller, R., 2001. Biodegradation of aliphatic-aromatic copolyesters: evaluation of the final biodegradability and ecotoxicological impact of degradation intermediates. Chemosphere. 44, 289-299.
- Zumstein, M., Schintlmeister, A., Nelson, T., Baumgartner, R., Wagner, M., Sander, M., McNeill, K., Woebken, D., Kohler, H., 2018. Biodegradation of synthetic polymers in soils: Tracking carbon into CO<sub>2</sub> and microbial biomass. Science Advances, 4, 7.

Dear Reviewers,

The table below provides the requested clarifications and the description of the changes made on the paper for each raised point. Many thanks to both of you for your valuable comments and suggestions. We did our best to improve the paper in the light of the received feedback.

n	Reviewers' comments	Revisions made in the paper
	Reviewer #1	• •
1	Reviewer #1 This paper presented a methodological approach for calculating the circularity of bio-based and biodegradable products (mulch films). This research aims at filling this gap through the development of a methodology coherent with EMF MCI methodology but able to catch the specificities of bio-based and biodegradable products and provide metrics for those innovative products. It is a topic of interest to the researchers in the related areas. However, the yield and application range of degradable plastics are important factors affecting their recycling. The whole paper should be reconstructed to make this paper more logically. A major revision is essential before acceptance. The followings are the specific comments.	Many thanks. EU economy has begun taking steps towards a low carbon future (e.g. renewable energy, electric vehicles) and more circular. Bio-based and biodegradable/compostable plastics are seen with interest in all those application where mechanical recycling of traditional plastics is hard to perform. For example in reference to the plastic mulching film the EU market accounts for about 80,000 t/y where >90% is represented by polyethylene (PE) mulch films. The use of PE film < 25 µm is responsible for about 15,000 t/y of microplastics which remain in the soil and about 30,000 t/y of agricultural plastic waste (i.e. PE mulch film) which are dumped or burned in the soil (1). Looking at these figure the great potentialities of developing alternative products results quite evident. However, due to space constraints it is not possible to extensively address these important aspects such as applications of biodegradable plastics, market perspective etc. as suggested by the reviewer. We instead performed some changes in the paper and added two very relevant on-line sources where it is possible to download EU documents, specific reports, case study etc able to direct the reader towards the topics raised by the reviewer. These are: <u>https://bbia.org.uk/reports/</u> (1) Revision of the Fertilisers Regulation – benefits of biodegradable mulch films Kristy-Barbara Lange, European Bioplastics, 12 October 2016
	Highlights: All of them are exceed the word	a%20Lange%20EUBP%20PPT2.pdf
2	limits for highlights (less than 85 characters). Please refer to the Guide for Authors.	The highlights have been reduced (see related attach)
3	Table: All tables should be three-line tables in the manuscript.	The tables have been adjusted
4	Line 79 - 82 reference needed	A reference has been added
5	Line 107 Mt, The first appearance should be slightly explained.	The term "Mt" has been expressed as "millions of tonnes"
6	Line 111 - 113 some new references are	New references have been added.

	needed. Please refer to " <i>Recent advances</i> <i>in toxicological research of nanoplastics in</i> <i>the environment: A review. Environmental</i> <i>Pollution, 2019, 252: 511-521; Microplastic</i> <i>pollution in surface sediments of urban</i> <i>water areas in Changsha, China:</i> <i>Abundance, composition, surface textures.</i> <i>Marine Pollution Bulletin 2018, 136: 414- 423.</i> "	22
7	Line 115 - 117 reference needed	The text has been integrated with the requested time for removing plastic mulch film from the soil and the related reference added
8	Line 137 - 140 Biodegradable polymers are capable of undergoing biological anaerobic or aerobic degradation. A major problem with these plastics is that they have the potential to be biodegraded, but this process requires suitable conditions and microorganisms that are not always reliable in environmental conditions (in situ). The author should explain this point in the article. Please refer to "Analysis and Prevention of Microplastics Pollution in Water: Current Perspectives and Future Directions. ACS Omega 4(4): 6709-6719".	The text has been integrated highlighting the importance of the environment's characteristics on the biodegradation rate of biodegradable bioplastics and the related reference added.
9	Line 284 - 287 reference needed. Although BB mulch films can undergo an ultimate biodegradation with no waste in the soil environment, the biodegradation processes and rate are the keys.	Reference added
10	Line 287 - 291 reference needed.	Reference added
11	Figure 4 should be further revised.	The figure caption has been improved and integrated
12	Line 437 - 438 The authors are encouraged to provide more information and discussion on the eco-design of innovative bio-based products.	The text has been integrated
	Reviewer #2	
13	This manuscript addresses an important topic - how to measure the circularity for a future circular bioeconomy. The suggested approach is novel and it is very good that the approach was demonstrated by the case study of mulch films.	Many thanks
14	It should be recognised that a circularity indicator like MCI is based on material	Absolutely agree. In the paper we only addressed the MCI of bio-based and biodegradable products as additional

	flow analysis only. Thus it does not provide a full picture of sustainability: mass efficiency is not a guarantee of many important sustainability issues like climate change, land use, water use and other resources depletion. This needs to be better elaborated in the paper	metric for further qualifying and assessing bio-based products. This aspect has been further highlighted in the conclusions (line 458-468)
15	The author addressed the toxicity as one of the sustainability aspects but it is: 1) not covered by MCI by definition, and 2) not about life cycle toxicity, which is also an important aspects for biobaesd production (especially in the agricultural phase).	The absence of toxicity is a <i>sine qua non</i> condition of the MCI methodology (line 375). It means that if a BB product causes toxicity effects the MCI does not apply since a fundamental principle (i.e. product safety) is not met. Translating this principle into biodegradable mulch film case study we recalled its compliance with the ISO 17033 standard since it encompasses the criteria regarding toxicity aspects beyond other requirements. That said if a BB mulch film is certified according to the ISO 17033 we can consider it safe for the environment.
16	The authors imply to re-define 'waste' (i.e. a material stream that cannot be recovered/biodegraded, or a material stream from a fossil-based source, see lines 285-293). This definition of 'waste' is very different from the definition of EU waste directive. This deviation should be brought into discussion. For example, the authors define that the stream goes to a landfill should be considered not recoverable. Use the case study of BB mulch films - will they biodegrade in a landfill? If yes, why should they be considered waste in this study? This is a very vague line that could practically hinder the application of a new metric.	It is not a re-definition of the term "waste". We have just defined the conditions for judging if a material stream is regenerative or not according to the proposed methodology. MacArthur methodology defines all material streams that go into incinerator or landfill "not regenerative" (i.e. no circular). Similarly we assumed that all BB product streams that go to landfill or incinerator are not regenerative with an exception: the "fossil part" that may constitute a BB product, even if it goes to biological recycling, it is still considered "not regenerative" since its origin is not biogenic. This methodological choice guarantees that a BB products gets a MCI =1 (complete circularity) only if it satisfies at the same time the following conditions: 1) the BB product is 100% made of renewable raw materials and 2) its end of life is represented by 100% biological recycling (composting or AD) or biodegradation in the environment depending on the BB application. Always according to this choice even if a 100% renewable BB product goes to incinerator or landfil thus it emits biogenic CO2 that goes into the atmosphere and biomass following a circular cycle, this is not considered a regenerative stream since the end of life option does not correspond to that a compostable product has been conceived for (i.e biological recycling). For this reasons MCI will be <1. This is the rationale of the MCI methodology. That said it is not our intention to modify or distort the current definition of "waste".
17	In the case study, the life cycle 'waste' streams from potato/corn/wheat cultivation are not clearly given. The mass balances shown in Figure 3 do not	The Figure 3 has been improved by removing all figures which were not useful for the calculation example. We are sorry for the trouble. In reference to your question about the amounts of agricultural feedstocks they have to be

	added up well: for example, in the case of $30/70$ starch blend, the total biomass required is $0.32t$ corn + 1.26t potato + 0.49t wheat = 2.07t (is this dry mass or	interpreted as 0.32 kg of corn or 1.26 kg of potato or 0.49 kg of wheat. They are the amounts needed to obtain 0.23 kg of starch (dry matter) which goes into the formulation. All the reported amounts of Figure 3 on starch, plasticizer
	green mass?), this gives 0.23t of starch. What is the 2.07-0.23 = 1.84t of the loss? The explanation in line 298 of R(s) of 0.014kg waste per kg renewable feedstock does not seem justified by the	and polymer refer to dry matter. Now the figure 3 should be clearer. In reference to 0.014 kg of not recoverable wastes per kg of renewable feedstock they refer to the "cradle to gate" LCA boundaries of starch. In the calculation we considered $W_F$ associated to the starch as follows 0.22 * 0.014 = 0.0022 kg/kg PB product
	numbers in Figure 3.	Tonows 0.23 * 0.014 = 0.0032 kg/kg BB product.
	Similar to the comment above: the case	In this specific case study the production of BB product (i.e. mulch film) yield is very close to 1 (nossible scraps are
18	mass loss of the production of fossil-based	internally reused in a closed loop) however, the proposed
10	biodegradable polymer.	formula for $W_{\rm F}$ encompasses the mass losses since the
		process yield is at the denominator of the formula.
	The effort of a monte carlo simulation is	A global sensitivity analysis can reveal the effect of the co-
	appreciated but is rather over complicated for the conclusion that F(s) + F(BP) is the most sensitive factor - it can be easily derived from a much simpler method like a regular sensitivity analysis.	variation of all parameters, showing how the variance cancels out or add to the specific variation of a factor; the analysis showed to what extent the value of 0.37 can be considered robust, in consideration of all possible variation in defined ranges. The analysis showed that, all possible variations accounted, the standard deviation is 0.041, meaning that 95% of observation would range between 0.29 and 0.45.
19		Not all parameters have a linear effect here. Ep, in particular, as it is placed in the denominator, might have had a relevant effect; its effect here is relatively small and negligible due to its small variation.
		A sensitivity analysis OAT (one factor at the time), also known as local sensitivity analysis, or an error propagation would suit this case and indicate which are the most sensitive factors. However, as this paper aims at clarifying the meaning and the robustness of the measure, we opted for a thorough analysis
	The sensitivity analysis should discuss	The uncertainty here is measured when assigning all the
	the influence of the missing data (see	factors an accuracy band. R(s) vas assigned a variation of
20	comments 4 and 5 above) or input data	100% thus largely covering possible changes in the
	that are highly uncertain	explanation relative to point 4.
	The discussion section should reflect on	Conclusions have been improved pointing out that MCI is
21	the limitation of this new metric.	just a further metric for characterizing BB products.
	The case study demonstrated a blend	For BB products that contain both biogenic and not
	material. How would it work for a	biogenic feedstocks, like in the calculation example (Figure
	copolymer which has partially biobased	3), only the amount of biogenic feedstock can be
22	content, such as 30% biobased PET? or	considered regenerative. The complementary amount does
	partially biobased PBAT (from biobased	not. The determination of the regenerative amount thus its
	succinic acid). There should be a clear	complementary not regenerative one is described in the
	definition of biobased content (mass),	recalled standard EN 16785-2:2016 (line 245)
	especially for the non-carbon elements	

	such as H, O and even N.	
23	Section 2.4, line 314: justify why a Gaussian distribution is chosen.	All values represent a realisation of industrial processes . The law of large numbers applies here. There is no reason to suspect that a given value would have a different distribution
24	the first para under section 3 Results should be shifted to methodology.	The first para has been moved under methodology section.
25	figure 3: what is the purpose of showing land use and what are the values in cubic meters?	The figure has been adjusted removing the information not needed for the paper purposes. Sorry for the trouble.
26	figure 6: is an illustration needed for the message in the figure?	The figure shows the percent change in the MCI when changing the indicated parameters of + 1%. So, as an example, Fs/F(BP) 3.29 a 1% change (+ 0.03) does not change the MCI; while a change of 1% of Rs (0.014 + 0.0001) yields a change of 0.7% in the MCI
27	- In lines 442-442 in the conclusions section, a relation of MCI of BB mulch films is given. This relation is only based on three data points, which is insufficient to draw a generic conclusion	Actually it is just a graphic representation of the MCI values obtainable through the application of the formulas reported in table 2. The three points represent the three different hypothetical compositions of the BB mulch film (i.e. renewable content equal to 30%, 50% and 70% respectively). For equal end of life (i.e. 100% biodegradation in soil) the MCI increases in function of renewable feedstock content.

#### Metrics for quantifying the circularity of bioplastics: the 1 case of bio-based and biodegradable mulch films 2 3 Francesco Razza<sup>a</sup>, Cristiana Briani<sup>b</sup>, Tony Breton<sup>c</sup>, Diego Marazza<sup>d</sup> 4 5 6 7 8 9 10 <sup>a</sup> Novamont S.p.A. - Ecology of Product and Environmental Communication, Piazz.le Donegani 4, 05100 Terni, Italy <sup>b</sup> CIRSA Centro Interdipartimentale di Ricerca per le Scienze Ambientali, Via S. Alberto 163, 48123 Ravenna, Italy ° Novamont S.p.A. – Via Fauser 8, 28100 Novara, Italy 12 13 <sup>d</sup>Department of Physics, University of Bologna, Viale B. Pichat 6/2, 40127 Bologna, Italy 14 15 Abstract The concept of circularity and its quantification through the Material Circularity Indicator

16 17 (MCI) is well established for traditional plastic products. In this paper a methodological 18 approach for calculating the circularity of bio-based and biodegradable (BB) products is 19 proposed and applied to BB mulch films. BB products are different from traditional 20 products in as much as they are sourced and regenerated (recycled) not through technical 21 cycles but the biological loop. The suggested method is an adaptation of the MCI where two major changes were made: (i) the mass of the bio-based component corresponds to the 22 23 recycled material in input and (ii) the mass of the bio-based component leaving the system 24 through composting or biodegradation in soil is accounted as recycled. The modified MCI supports the Eco-design of innovative BB products and allows for the comparison of their 25 26 circularity taking into account the biological source and the expected end of life process 27 such as biodegradation. To demonstrate the adaptation, the method has been applied to BB mulch film. Results showed that the MCI of a biodegradable mulch film, characterized by 28 29 an average bio-based feedstock content of 30% is  $0.37 \pm 0.04$  in a 0-1 scale. For BB mulch 30 film, the amount of bio-based feedstock is the most sensitive factor and controls linearly 31 the value of the MCI.

32

*Keywords:* circularity indicators, circular economy, bioplastics, biodegradable
 mulch film, bio-based product, biodegradation

35

36	1 Introduction
37	1.1 The case study of mulch films
38	1.2 Goal of the paper
39	2 Materials and Methods <u>8</u> 7
40	2.1 MCI accounting according to the EMF methodology <u>8</u> 7
41	2.2 MCI accounting for bio-based and biodegradable (BB) products <u>1340</u>
42	2.3 MCI calculation for mulch films: scope, inventory and assumptions <u>17</u> 14
43	2.4 Sensitivity analysis
44	3 Results
45	3.1 Sensitivity analysis
46	4 Discussion
47	5 Conclusions

Abbreviations	
BB	Biodegradable and bio-based
CE	Circular Economy
<del>d.m.</del>	Dry matter
EMF	Ellen MacArthur Foundation
LCA	Life Cycle Assessment
LDPE	Low-Density Poly-Ethylene
MCI	Material Circularity Indicator
NRF	Non-Restorative Flows
PBAT	Polybutylene adipate terephthalate
₽E	Poly-Ethylene
PLA	Polylaetie acid
Abbreviations	

BB	Biodegradable and bio-based
<u>CE</u>	Circular Economy
<u>d.m.</u>	Dry matter
EMF	Ellen MacArthur Foundation
LCA	Life Cycle Assessment
LDPE	Low-Density Poly-Ethylene
MCI	Material Circularity Indicator
NRF	Non-Restorative Flows
<b>PBAT</b>	Polybutylene adipate terephthalate

<u>PE</u>	Poly-Ethylene
<b>PLA</b>	Polylactic acid
PHB	Poly hydroxy butyrate
#### 51 **1 Introduction**

To overcome today's unsustainable model of <u>'takeof 'take</u>-make-dispose' and its related 52 53 risks such as hikes in raw material prices, pressures on the environment, shortage of global 54 resources and waste sinks, a circular approach needs to be applied. It is a new regenerative 55 economic view, based on a balance between economy, environment and society, a total 56 resource efficiency and a Zero Emission Strategy that aims to maximize products value 57 with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with 58 structural changes in environmental legislation, new logistics, technologies and sharing 59 schemes, the Circular Economy (CE) approach which is regenerative by design, aims at closing materials loops, i.e. at reducing virgin materials input and waste output. 60

61 In December 2015, the European Commission developed an Action Plan for Circular 62 Economy (European Commission, 2015), where plastic was considered a priority to be 63 tackled. In January 2018, an EU Plastic Strategy (European Commission, 2018) was 64 adopted, in order to react to the increasing environmental problems concerning plastic 65 production, consumption, use and disposal along the same lines of the CE approach. Two fundamental steps to increase the circularity of different plastic products are (i) the 66 67 abandonment of fossil fuels, i.e. currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development 68 of easily recyclable products which are recycled. Today, in EU the share of plastics 69 collected for recycling is 30% while the use of recycled plastics is just 6% (European 70 Commission, 2018). 71

72 Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for 73 Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and 74 principles. This is true as long as the supply of renewable raw materials, generally from 75 agriculture, is based on a sustainable approach and the conversion processes along the

76 supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA) 77 perspective (EPLCA - European Platform on LCA). While traditional plastics can be 78 mechanically recycled or incinerated with energy recovery, BB plastic products offer new 79 recycling routes in waste management, due to their biodegradability. Organic recycling (through composting or anaerobic digestion) or in the case of specific applications such as 80 81 agricultural mulch films, biodegradation in the environment, offer additional recovery 82 options resulting in less wastes and less contamination of soil by plastic residues (Razza et al., 2012; Lange, B., 2016). An extensive literature review about the potentialities and 83 84 benefits of renewable and compostable bioplastics, encompassing market perspective, applications, economic effects etc. can be found here: (BBIA; European Bioplastics). 85 Nevertheless, the research and development of innovative products, such as the BB 86

87 products, implies the development of methodologies and metrics capable of measuring 88 their circularity. Without this it is not possible to achieve measurable results and 89 improving actions, as well as provide unequivocal references for comparisons of products 90 of the same type/category. In 2015 the Material Circularity Indicator (MCI) was developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify 91 the regeneration of a product's material flow and is considered one of the few, among 92 93 sixteen CE indexes suiting a micro-scale assessment of circularity at product or company 94 level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled 95 materials. Furthermore, recovery and recycling through the biological cycle offered by 96 industrial composting, anaerobic digestion or biodegradation in natural environments are 97 not considered as end of life options. In order to apply the MCI system to BB plastic 98 products, the development of an enhanced methodology is necessary.

99 The approach proposed by the authors allows to quantify the circularity of BB plastic
100 products (*e.g.* starch based bioplastics) and to make comparisons with equivalent

traditional plastic products. To demonstrate the applicability of the proposed method a
computational example for mulch film products is provided. In so doing so, the paper
aims at contributing to the Eco-design of these innovative products.

#### 104 1.1 The case study of mulch films

105 Plastic mulch films represent an important agronomical technique well established for the 106 production of many crops thanks to numerous agronomical advantages such as: increased 107 yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and 108 reduced use of pesticides; early crop production and reduced soil moisture loss 109 (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has 110 increased year-by-year, reaching a current global market estimated at 1.4 millions of tonnes Mt, mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017), and 111 112 covering 80,000 km<sup>2</sup> of agricultural surface (0.6% of the global arable land). The mulch 113 film market in Europe is estimated by Agriculture Plastic & Environment and by the European Bioplastic Associations at 76-80 kt. The most used raw material is Poly-114 115 Ethylene (PE) in its different forms, due to its processability, chemical resistance, high 116 durability and flexibility (Kasirajan and Ngouajio, 2012: Plasticulture, 2016 and 2018; 117 Shen, M. et al., 2019; Wen, X. et al., 2018).

118 Despite these benefits, manifold environmental and agronomic problems have been 119 pointed out. After its useful life - which in general does not exceed 1 to 3 months - the 120 mulch film has to be removed and properly disposed of, a time-consuming (about 16 hours per hectare) and costly procedure (Scaringelli, M., 2016; Briassoulis, D., 2013). The 121 122 recovered film is usually heavily contaminated with soil and organic residues, making 123 mechanical recycling technically difficult and not a cost-efficient solution (Briassoulis et 124 al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most common end of life of collected films in Europe is still landfilling (about 50%), followed by energy recovering 125

and finally mechanical recycling (Le Moine, 2014). Recent Chinese prohibition (January 126 127 2018) to import different types of wastes is heavily impacting the European agricultural 128 plastic waste management, highlighting the difficulty in properly recycling this type of plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but 129 130 disposed of by burning in the field or by uncontrolled landfilling or left directly in the 131 (agricultural) soils, causing serious environmental concerns. An example is the "White 132 pollution" phenomena described in the Xinjiang Autonomous Region (China), in which 133 the residual plastic film can reach 200 kg/ha in the top soil with detrimental effects on 134 soils' quality, health and fertility (Liu, He, & Yan, 2014; Gao et al., 2019; Steinmetz et 135 al., 2016).

136 As a reaction, there has been significant research into novel materials especially related to 137 biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation 138 in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). 139 The term "bio-mulch film" brings together several types of both bio-based and fossil oil-140 based biodegradable polymers and blends of them, such as polylactic acid (PLA), 141 polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or 142 copolymers. They biodegrade when exposed to bioactive environments such as soil and 143 compost (Kasirajan et al., 2012) which means that they can be left in situ to be fully 144 biodegraded after being used. Clearly the biodegradation rate of biodegradable bioplastics 145 is influenced by the environmental conditions such as the types of available bacteria, fungi 146 thus specific enzymes namely native microflora (Pico, Y. et al., 2019). However their intrinsic biodegradability must be proved by accredited certification bodies and 147 standardized procedures allow the complete biodegradation with times similar to natural 148 149 polymers such as cellulose used as reference by the relevant standards and certification 150 schemes.

151 The EN 17033:2018 is a new European Norm (standard) concerning "Plastics -Biodegradable mulch films for use in agriculture and horticulture - Requirements and test 152 153 methods", which sets the necessary tests and limits to define biodegradability, performances and environmental impacts of BB much films. The material is considered 154 completely biodegradable if it achieves a complete biodegradation (absolute or relative to 155 the reference material) in a test period no longer than 24 months (mineralization into 156 157 CO<sub>2</sub>). Additionally, a control of constituents (such as metals) and eco-toxicity testing 158 (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test 159 with soil microorganisms) were required. A certified mulch film guarantees that the 160 product will completely biodegrade in the soil without adversely impacting on the 161 environment.

#### 162 1.2 Goal of the paper

163 The goal of the paper is to provide a general and common metric to measure the 164 circularity of a bio-based and biodegradable (BB) product and to apply the methodology at 165 product level to a category of products, namely bio-based and biodegradable mulch films.

#### 166 2 Materials and Methods

#### 167 2.1 MCI accounting according to the EMF methodology

The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production provides for the exclusive use of virgin raw materials that turn into waste at the end of the use phase of the product. Vice-versa, pure circularity includes the use of recycled materials and does not produce wastes (regenerative streams). Circularity can be achieved in different ways: as for the purpose of this paper, only recycling will be considered since

175	reuse is not an option for thin biodegradable mulch films. Since the method considers only		
176	mass flows, the recycling corresponds to the recovery of materials for the original purpose		
177	or for other purposes and excludes energy recovery, considered as a loss of materials equal		
178	to landfill disposal. The materials recovered feed back into the process as recycled		
179	feedstock.		
180	The MCI methodology differentiates 'technical cycles' from 'biological cycles',		
181	modelling only the former. The first contains products and materials re-entering into the		
182	system (market) with the highest possible qualities and for as long as possible (thanks to		
183	reuse, repair, refurbishment and recycling) and the latter includes biological materials used		
184	in cascade until their restoration into the biosphere and the re-constitution of natural		
185	resources.		
186	The material flows associated to the production of a generic technical cycle from non-		
187	renewable sources are summarized in Figure 1Figure 1. The dashed lines indicate that	 Formatted: Font:	Not Italic
188	recycled feedstock does not have to be sourced from the same product but can be acquired		
189	on the market. With reference to Figure 1Figure 1, the list of the parameters used in the	 Formatted: Font:	Not Italic
190	EMF methodology is reported in <u>Table 1</u> , while the equations relevant for the	 Formatted: Font:	Not Italic
191	analysis carried out in this paper are described in the following sections ( <u>Table 2</u> Table 2,	 Formatted: Font:	Not Italic
192	Chapter 2.2).		

Formatted: Font: Not Italic
Formatted: Font: Not Italic
Formatted: Font: Not Italic



Figure 1: Diagram of material flows and associated variables of a generic

195 product (modified from Ellen MacArthur Foundation & Granta Design, 2015).

Table 1: Parameters and relative definitions used in the EMF methodology.

Parameter	Definition
	Total mass of the product
<b></b>	Fraction of mass of a product's feedstock from
<i>r</i> <sub><i>R</i></sub>	recycled sources
E	Fraction of mass of a product's feedstock from
τų.	reused sources
¥	Mass of virgin feedstock used in a product
6	Fraction of mass of a product being collected to go
€ <u>₽</u>	into a recycling process

<b>C</b> n	Fraction of mass of a product going into
	component reuse
E	Efficiency of the recycling process used for the
₽¢	portion collected for recycling
E	Efficiency of the recycling process used to produce
	recycled feedstock for a product
W	Total mass of unrecoverable waste associated with
<b>**</b>	a product
	Mass of unrecoverable waste (landfill, waste to
₩ <sub>a</sub>	energy and any other type of process where the
	materials are no longer recoverable)
₩c	Mass of unrecoverable waste generated in the
	process of recycling parts of a product (after use)
Wr	Mass of unrecoverable waste generated when
	producing recycled feedstock for a product
¥	Utility of a product, calculated as X=
	$\frac{(L/L_{av})(U/U_{av})}{(U/U_{av})}$
Ŀ	Actual average lifetime of a product
L	Actual average lifetime of an industry average
	product of the same type
Ľ	Actual average number of functional units
	achieved during the use phase of a product
	Actual average number of functional units
<del>U</del> ar	achieved during the use phase of an industry-
	average product of the same type

Parameter	Definition
<u>M</u>	Total mass of the product
<u><i>F</i></u> <sub><i>R</i></sub>	Fraction of mass of a product's feedstock from recycled sources
<u><b>F</b></u> <sub>U</sub>	Fraction of mass of a product's feedstock from reused sources
<u>V</u>	Mass of virgin feedstock used in a product
<u><i>C</i></u> <sub><i>R</i></sub>	Fraction of mass of a product being collected to go into a recycling process
<u><b>C</b></u> <u>U</u>	Fraction of mass of a product going into component reuse
<u><i>E<sub>C</sub></i></u>	Efficiency of the recycling process used for the portion collected for recycling
<u><i>E<sub>F</sub></i></u>	Efficiency of the recycling process used to produce recycled feedstock for a
	product
<u>W</u>	Total mass of unrecoverable waste associated with a product
<u>W_0</u>	Mass of unrecoverable waste (landfill, waste to energy and any other type of
	process where the materials are no longer recoverable)
<u>W</u> <sub>C</sub>	Mass of unrecoverable waste generated in the process of recycling parts of a
	product (after use)
<u><i>W<sub>F</sub></i></u>	Mass of unrecoverable waste generated when producing recycled feedstock for
	a product
<u>X</u>	<u>Utility of a product, calculated as <math>X = (L/L_{av})(U/U_{av})</math></u>
<u>L</u>	Actual average lifetime of a product
<u>Lav</u>	Actual average lifetime of an industry-average product of the same type
<u>U</u>	Actual average number of functional units achieved during the use phase of a
	product
<u>Uav</u>	Actual average number of functional units achieved during the use phase of an
	industry-average product of the same type

199 The Material Circularity Indicator is determined as follows:

200 where LFI is the Linear Flow Index measuring the flows of virgin materials and

201 unrecoverable wastes associated to the examined product.

A function of the utility, -, is used to correct the *LFI*. The function *F* is chosen in such a way that improvements of the utility of a product (e.g., by using it longer) have the same impact on its MCI as a reuse of components, leading to the same amount of reduction of virgin material use and unrecoverable waste. Setting a = 0.9, MCI takes, by convention, the value 0.1 for a fully linear product (*i.e.*, *LFI* = 1) whose utility equals the industry average (*i.e.*, X = 1). This leaves some margin to distinguish between processes with a high linearity but different utilities.

#### 209 2.2 MCI accounting for bio-based and biodegradable (BB) products

210To apply the EMF methodology to BB products, formulas and flows (Figure 1Figure 1211and Figure 2Figure 2) are adapted as it follows:2121. The fraction of the recycled feedstock,  $F_R$ , corresponds to the share of the bio-213based feedstock content in the final BB product,  $F_{R(i)}$ . It is the ratio of the d.m.214amount of bio-based feedstock per d.m. amount of the total mass of BB215product (EN 16785-2:2016).

216 2. The fraction of restorative mass going into a recycling process,  $C_R$ , corresponds 217 to the share of bio-based feedstock content in the BB product biologically 218 recovered (*e.g.* through composting) or biodegraded in the natural 219 environment, as it happens for specific applications (e.g. biodegradable mulch 220 film, *etc.*). It is the ratio of the d.m. amount of bio-based feedstock per d.m. 221 amount of the total mass of BB product that is biologically recycled.

222 The modified scheme is shown in Figure 2. Table 2. Table 2 lists the formulas as

adapted to BB products.

13

Formatted: Font: Not Italic

Formatted: Font: Not Italic

Formatted: Font: Not Italic Formatted: Font: Not Italic

#### Table 2: List of formulas as developed by EMF methodology compared to the

#### 225

224

proposed adaptation to BB products.



227 The mass of fossil-based feedstock which may be contained in BB products (V) is 228 obtained as a difference of the total mass (M) minus the bio-based fraction; in this case the 229  $F_R$  in the EMF methodology corresponds to the sum of the fractions of all the bio-230 based feedstock/s used in manufacturing the BB product. Therefore, is the 231 total bio-based feedstock mass in the product. In single-use products, such as mulch films, reuse is not considered for BB products, so that  $F_U = C_U = 0$ . 232 233  $W_F$  is the total amount of unrecoverable waste associated to the production of bio-based 234 feedstock used to produce BB products (i.e. the amount of uncoverable waste per unit of 235 BB product). Bio-based feedstocks such as starch, -and-PLA, -PHB etc. generate non-236 restorative flows which can be quantified. Such unrecoverable waste correspond to  $R_{(i)}$ , 237 the specific amount of waste generated within cradle-to-gate boundaries per unit of bio-238 based feedstock going into manufacturing, and it is estimated through LCA studies. Thus 239 all inputs from growth and harvesting phases and the related wastes generated by 240 fertilisers and pesticides are here accounted.  $R_{(i)}$  can be easily found in specific literature 241 or life cycle inventories (LCI) present in LCA databases. In the calculation of  $W_F$ , also the

226

- 242 efficiency of manufacturing process of BB products  $E_P$  is considered, as the ratio of the
- 243 overall bio-based feedstock content in the final BB product to the bio-based feedstock in
- 244 input to the manufacturing process.
- 245 The material flows associated to the production of a generic BB product are summarized

#### 246 in Figure 2 Figure 2.



247

#### 16

#### Formatted: Font: Not Italic

<sup>248</sup> Figure 2: Description of material flows adaptation to BB products; in this paper, 249 the reuse flow is out of scope ( $C_U = F_U = 0$ ).

The biodegradation of bio-based feedstock does not imply the generation of waste  $W_C$  as it occurs in a standard mechanical recycling process. This implies that  $C_R$  and  $E_C$  (*i.e.* the efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to biological treatment (composting) or biodegraded in a natural environment, is fully transformed in its chemical elements (C, H and O mainly) derived from the decomposition

255	of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et	
256	al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites,	
257	2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the	
258	environment and are then available in the respective biogeochemical cycles. The	
259	(biodegradable) fossil portion behaves as well; consequently, $W_C = 0$ .	
260	Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular	
261	feedstock, since it derives from carbon stored for millions of years and extracted by man,	
262	not being part of the active and fast biogeochemical carbon cycle. This is accounted in the	
263	quantification of $W_0$ , the mass of unrecoverable waste from use ( <i>i.e.</i> the linear stream	
264	going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total	
265	amount of fossil-based feedstock.	
266	Since $W_F$ and $W_C$ are associated to complete different processes and $W_{C-}$ is always equal	
267	zero, the double counting issue does not occur and the quantification of $W$ and $LFI$ is	
268	modified as reported in <u>Table 2</u> Table 2.	
268 269	modified as reported in <u>Table 2</u> .         2.3 MCI calculation for mulch films: scope, inventory and assumptions	
268 269 270	modified as reported in <u>Table 2</u> <u>Table 2</u> .         2.3 MCI calculation for mulch films: scope, inventory and assumptions         The new formulas reported in <u>Table 2</u> <u>Table 2</u> were applied to a single use product namely	
268 269 270 271	<ul> <li>modified as reported in <u>Table 2Table 2</u>.</li> <li>2.3 MCI calculation for mulch films: scope, inventory and assumptions</li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of BB</li> </ul>	
268 269 270 271 272	<ul> <li>modified as reported in <u>Table 2Table 2</u>.</li> <li>2.3 MCI calculation for mulch films: scope, inventory and assumptions</li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of BB</li> <li>materials into the final products (i.e. white mulch films) takes place without any</li> </ul>	
<ul> <li>268</li> <li>269</li> <li>270</li> <li>271</li> <li>272</li> <li>273</li> </ul>	<ul> <li>modified as reported in <u>Table 2Table 2</u>.</li> <li>2.3 MCI calculation for mulch films: scope, inventory and assumptions</li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of- BB</li> <li>materials into the final products (i.e. white mulch films) takes place without any</li> <li>modification of the bio-based feedstock content and the process yield is close to 1.</li> </ul>	
268 269 270 271 272 273 274	<ul> <li>modified as reported in <u>Table 2Table 2</u>.</li> <li>2.3 MCI calculation for mulch films: scope, inventory and assumptions</li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of– BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.</li> <li>In the global market, there are several branded BB mulch films (Moreno et al., 2017), both</li> </ul>	
<ul> <li>268</li> <li>269</li> <li>270</li> <li>271</li> <li>272</li> <li>273</li> <li>274</li> <li>275</li> </ul>	<ul> <li>modified as reported in <u>Table 2Table 2</u>.</li> <li>2.3 MCI calculation for mulch films: scope, inventory and assumptions</li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.</li> <li>In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the -BB film has been arbitrarily is</li> </ul>	
<ul> <li>268</li> <li>269</li> <li>270</li> <li>271</li> <li>272</li> <li>273</li> <li>274</li> <li>275</li> <li>276</li> </ul>	modified as reported in <u>Table 2Table 2</u> . <b>2.3</b> <i>MCI calculation for mulch films: scope, inventory and assumptions</i> The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of– BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1. In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the -BB film <u>has been arbitrarily is</u> assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e.	
<ul> <li>268</li> <li>269</li> <li>270</li> <li>271</li> <li>272</li> <li>273</li> <li>274</li> <li>275</li> <li>276</li> <li>277</li> </ul>	<ul> <li>modified as reported in <u>Table 2Table 2</u>.</li> <li>2.3 MCI calculation for mulch films: scope, inventory and assumptions</li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of- BB</li> <li>materials into the final products (i.e. white mulch films) takes place without any</li> <li>modification of the bio-based feedstock content and the process yield is close to 1.</li> <li>In the global market, there are several branded BB mulch films (Moreno et al., 2017), both</li> <li>starch-based or blends of polyesters. In the following, the -BB film has been arbitrarily is</li> <li>assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e.</li> <li>23% of starch, F<sub>(S)</sub>, and 7% of a bio-based plastieizeradditive, F<sub>(BPA)</sub>), while the rest was</li> </ul>	
268 269 270 271 272 273 274 275 276 277 278	<ul> <li>modified as reported in <u>Table 2Table 2</u>.</li> <li>2.3 MCI calculation for mulch films: scope, inventory and assumptions</li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of- BB</li> <li>materials into the final products (i.e. white mulch films) takes place without any</li> <li>modification of the bio-based feedstock content and the process yield is close to 1.</li> <li>In the global market, there are several branded BB mulch films (Moreno et al., 2017), both</li> <li>starch-based or blends of polyesters. In the following, the -BB film has been arbitrarily is</li> <li>assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e.</li> <li>23% of starch, F<sub>(S)</sub>, and 7% of a bio-based plasticizeradditive, F<sub>(BPA)</sub>), while the rest was</li> </ul>	
268 269 270 271 272 273 274 275 276 277 278 279	<ul> <li>modified as reported in <u>Table 2Table 2</u>.</li> <li>2.3 MCI calculation for mulch films: scope, inventory and assumptions</li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of- BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.</li> <li>In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the -BB film has been arbitrarily is assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch, F<sub>(S)</sub>, and 7% of a bio-based plastieizeradditive, F<sub>(BPA)</sub>), while the rest was assumed to consist of fossil feedstock (</li> </ul>	

Formatted: Font: Not Italic

Formatted: Font: Not Italic

Formatted: Font: Not Italic





297	Figure 3: Examples of <u>hypothetical starehbio</u> -based polymers; in this paper, the
298	first option on the left (starch blend 30/70) has been chosenas representative of a BB
299	mulch film for carrying out the numerical MCI calculation (working hypothesis). The
300	figure considers a 100%-efficiency in every phase of production, so that the residues are
301	equal to zero; the same assumption is done in this paper. *TPS (Thermoplastic starch),
302	starch content 75%; **Ratio TPS/Polymer; modified from Institute of Bioplastics and
303	Biocomposites, 2018.

## Table 3: Key features representative of the BB mulch films.

	BB mulch film		-(	Formatted: Font: 11 pt	
Material	30% bio based feedstock (23% starch + 7% bio-		-1	Formatted: Font: 11 pt	
	based plasticizer) + 70% fossil based feedstock		C		
<del>Thickness (µm)</del>	12	1	-(	Formatted: Font: 11 pt	
Density (g/cm <sup>3</sup> )	1.25		-(	Formatted: Font: 11 pt	
Weight (g/m <sup>2</sup> )	15.2		1	Formatted: Font: 11 pt	
			Ľ	Formatted: Font: 11 pt	
Eurotional unit	6000 m <sup>2</sup> /ha (the actual mulched soil in a hectare is		Ľ	Formatted: Font: 11 pt	
(4	generally equal to the 60% of the total area;		Y	Formatted: Font: 11 pt	
(the covering of the agricultural land)	Malinconico, 2017)				
1	BB mulch film	-			
Material	30% bio-based feedstock (23% starch + 7% bio-				
	based additive) + 70% fossil feedstock				
<u>Thickness (μm)</u>	<u>12</u>				
Density (g/cm <sup>3</sup> )	<u>1.25</u>				
Weight (g/m <sup>2</sup> )	<u>15.2</u>				
Functional unit	6000 m <sup>2</sup> /ha (the actual mulched soil in a hectare				
(the covering of the agricultural land)	is generally equal to the 60% of the total area;				
	Malinconico, 2017)				

308	In the calculation of MCI for the BB mulch film, the adapted formulas were used together
309	with assumptions. As stated before, BB mulch films are blends of bio-based and fossil
310	based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE
311	mulch film that has to be removed and disposed of, the BB mulch film is left in soil where
312	it undergoes an ultimate biodegradation (so that $C_R = 1$ ) with no waste (so that $E_C = 1$ ), in
313	respect of the specific standard EN 17033:2018. As a result of polymers' decomposition,
314	the derived (biogenic) C, H and O finally return into biosphere (atmosphere,
315	microorganism biomass, organic material pool) (OWS, 2018), and back into
316	biogeochemical cycles in a relatively short time ("Biogenic elements accounted as
317	recycled" in Figure 2Figure 2), with the exception of -humified compounds. Actually, also
318	C, H and O deriving from fossil-based sources undergo biodegradation (Zumstein, M.T.,
319	2018) but they are not considered as a regenerative flow ("Waste from non-restorative
320	flow" in <u>Figure 2</u> and their "wastes" are indeed calculated in $W_0$ .
321	Applying a conservative approach, $W_F$ , the waste generated by the production of each bio-
322	based feedstock, is quantified considering a "cradle to gate" LCA study. The estimated
323	solid wastes $R_{(i)}$ for the presented case study are related to the production of starch (F <sub>(S)</sub> ),
324	with an amount $R_{(S)}$ of 0.014 kg of waste per kg of renewable feedstock (source: personal
325	communication A. Novelli), and to the production of the bio-based additive plasticizer
326	$(F_{(B\underline{AP})})$ , with $R_{(B\underline{P}\underline{A})}$ equals to 0.025 kg waste/kg renewable feedstock (US-LCI database),
327	(source: US LCI database "Polylactide biopolymer resin at plant kg/RNA"). As assumed
328	in
329	
226	
330	<u>Figure 3</u> , the production efficiency of BB product $E_P$ (how much bio-based

feedstock is needed for every unit of BB product) is estimated equal to 1 and nounrecoverable wastes are generated by the process.

20

Formatted: Font: Not Italic

Formatted: Font: Not Italic

333	In addition, an explorative sensitivity analysis has been performed regarding exclusively	
334	the amount of bio-based feedstock content of the BB mulch film, <i>i.e.</i> $(i.e., F_{(S)} +$	
335	$F_{(BPA)}$ ), as shown in <u>Figure 4</u> (Chapter 3). <u>Considering the characteristics of the</u>	Formatted: Font: Not Italic
336	films (weight, g/m <sup>2</sup> , or thickness, µm, and density, g/cm <sup>3</sup> ) and the relative functional unit	
337	(6000 m <sup>2</sup> /ha, <u>Table 3</u> , it is possible to calculate a mass, <i>M</i> , that is 90 kg/ha for the	Formatted: Font: Not Italic
		Formatted: Font: Bold, Not Italic
338	<u>BB one. Once calculated the masses, the formulas reported in Table 2 Table 2 (Chapter</u>	Formatted: Font: Not Italic
220	2.2) are applied Results are shown in Table (Table 4	Formatted: Font: Not Italic
559		Formatted: Font: Not Italic
<ul> <li>341</li> <li>342</li> <li>343</li> <li>344</li> <li>345</li> <li>346</li> <li>347</li> </ul>	<b>2.4</b> Sensitivity analysis A sensitivity analysis was conducted for BB mulch film to examine the effects of changing the main variables. Given a non-linear dependence of results on parameter values, a Monte Carlo approach (see, <i>e.g.</i> , Lloyd and Ries, 2008) has been adopted. The model has been implemented using specifically written routines in the C++ programming language. The model was run with 100,000 events for BB mulch film, where the value of each parameter has been randomly chosen following a Gaussian distribution with a	
348	standard deviation within a range of possible and realistic values (Table STable 5 and	Formatted: Font: Not Bold Not Italic
510	sandard deviation within a range of possible and realistic values (14010 5 14010 5 and	
349	Error! Reference source not found. Table 6; Figure 5 Figure 5 and Figure 6 Figure 6).	Formatted: Font: Not Italic
		Formatted: Font: Not Italic
350	3 Results	
351	Considering the characteristics of the films (weight, g/m <sup>2</sup> , or thickness, µm, and density,	
352	g/cm <sup>3</sup> ) and the relative functional unit (6000 m <sup>2</sup> /ha, Table 3), it is possible to calculate a	Field Code Changed
353	mass, M, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas	
354	reported in Table 2 (Chapter 2.2) are applied. Results are shown in Table 4.	Field Code Changed
355	Figure 4 <del>Figure 4</del> shows how the value of the MCI varies according to the percentage	Field Code Changed
555	right in the value of the works according to the percentage	Formatted: Font: Not Italic
356	variation of the bio-based feedstock in the total mass of the product.	

Parameter BB mulch film

# Table 4: Resulting parameters in the calculation of MCI for BB mulch film.



Formatted: Font: Not Bold, Not Italic

Formatted: Font: Not Italic
Formatted: Font: Not Italic

- 372 regarded as a system composed by a high number of companies, each producing films
- 373 with different characteristics, that are accounted for in the accuracy band.
- 374 Table 5: Parameters used for the sensitivity analysis of the BB mulch film. (\*\*) The
- 375 Accuracy Band is defined as twice the standard deviation of the distribution.

Variable name	Average	Accuracy Band (**)	Unit
M	1000.00	0%	kg
₽ <sub>(S)</sub> /₽ <sub>(BP)</sub>	3.29	<del>10%</del>	fraction
$F_{(S)} + F_{(BP)}$	0.30	30%	fraction
₽u	0.00	0%	fraction
e <sub>u</sub>	0.00	0%	fraction
₽ <sub>(S)</sub>	0.014	<del>100%</del>	fraction
₽ <sub>(8P)</sub>	0.025	<del>100%</del>	fraction
ŧ	±	0%	fraction
€₽	0.95	10%	fraction
€ <sub>R</sub>	1.00	0%	fraction
Variable name	Average	Accuracy Band (**)	<u>Unit</u>
M	1000.00	<u>0%</u>	kg
<u>F(s)/F(BPA)</u>	<u>3.29</u>	<u>10%</u>	fraction
$\underline{\mathbf{F}}_{(S)} + \underline{\mathbf{F}}_{(BP\underline{\mathbf{A}})}$	<u>0.30</u>	<u>30%</u>	fraction
<u><b>E</b></u> <u>u</u>	<u>0.00</u>	<u>0%</u>	fraction
<u><b>C</b></u> <u>U</u>	<u>0.00</u>	<u>0%</u>	fraction
<u><b>R</b>(S)</u>	<u>0.014</u>	<u>100%</u>	fraction
<u><b>R</b>(BAP)</u>	<u>0.025</u>	<u>100%</u>	fraction
<u>E</u>	<u>1</u>	<u>0%</u>	fraction
<u>E</u> P	<u>0.95</u>	<u>10%</u>	fraction



Figure 5: Resulting distribution of MCI values for BB mulch film.



382 Figure 6: The most sensitive and relevant parameters in the calculation of the
383 MCI of the BB mulch films.

#### 384 4 Discussion

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

- 390 (1) fossil-based feedstocks or component materials embodied in the BB products whatever
- 391 is the final disposal (even biological recycling) shall be considered as non-restorative;
- 392 (2) bio-based component materials embodied in the BB product that go to biological
- 393 recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be

394 considered restorative as long as they flow through the biosphere safely, without any harm

395 to the environment (e.g. no toxicity effects).

396 (3) bio-based component materials embodied in the BB product that go to incineration and

397 landfill shall be considered as non-restorative;

398 The justification of these rules is described in the following.

Fossil-based component materials in the product derive from deposits where they 399 400 remained stocked for a geological time scale. Once the product is mineralised, its fossil-401 based portion will be accounted as non-regenerative and therefore linear, due to its origin 402 (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological 403 cycles, like CO2 in the atmosphere and other streams, since both fossil-based and bio-404 based component materials will physically and chemically behave the same, once 405 biodegraded. However, the source of the bio-based carbon was circular before its use 406 (concept of "carbon neutrality", equilibrium between the biogenic carbon released and the 407 carbon absorbed by plants) and will maintain its circularity provided that the carbon is 408 released into the atmosphere at the same rate. The reason has its origin in the EMF general 409 provisions stating that "biologically sourced materials can only be considered part of a 410 Circular Economy if materials are not used faster than they can be restored naturally" 411 (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the 412 bio-based components are still considered linear, maintaining consistency with EMF 413 principles. Basically, a complete circularity for a BB product is satisfied when its 414 renewable components are 100% bio-based and they go 100% to biological recycling or 415 biodegraded in the environment (for specific application like mulch film).

416 As for provision (3),- a material health rule has its origin in manyfold normative
417 definitions of the CE. In addition, the EMF definition of biological cycles is that of non418 toxic materials which are restored into the biosphere and the CE is defined as such if it can

"eliminate the use of toxic chemicals". The need of a safety clause has been reviewed 419 420 under many aspects by Verberne (2016) and can be put as a postulate of the restoration 421 principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the REACH Regulation (EC 1907/2006). In the specific case, the material complies with the 422 standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism 423 424 groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important 425 ecological processes maintaining soil functions, c) all relevant exposure pathways as soil 426 pore water, soil pore air and soil material.

427 A comprehensive approach for MCI calculation should also include non-restorative flows 428 generated at upstream level like biomass growth, in the specific case corn, and biomass 429 conversion processes like starch extraction and refining. Specifically these non-restorative 430 flows correspond to the overall non-recyclable wastes associated to the bio-based 431 feedstock supply thus non-recyclable waste from fertilizer and pesticide production, non-432 recyclable scraps from conversion processes, etc. In this study such flows of nonrestorative waste coming from upstream manufacturing operations were included for the 433 434 bio-based feedstocks  $(R_{(i)})$  used in manufacturing the BB mulch film applying "cradle to gate" LCA methodology. However, we observed that the inclusion of upstream 435 436 unrecoverable waste does not significantly influence the MCI results in the chosen case 437 study, since the respective amounts are small. The specific unrecoverable waste for starch 438 and bio-based additive plasticizer (i.e. kg of waste/kg of bio-based feedstock) were 439 estimated at 0.014 and 0.025, respectively.

440

# The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation y = 0.89x + 0.1, where y is the MCI and x is the bio-based feedstock content,

therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input isdecisive.

446 Apart from the specific application analysed in this paper, the proposed MCI method can
447 be easily applied and calculated for any kind of BB product as long as the following
448 information are available:

The bio-based feedstock content, determined according to the standard EN 167852:2016, if the composition is known, or directly provided by the BB product manufacturer.
The End of Life scenario of the studied BB product (real or hypothetical).

The amount of un-recoverable waste associated to the production of bio-based
 feedstock contained in the BB product. They can be derived from LCA databases or other
 specific sources.

#### 455 5 Conclusions

456 Bioplastic market is steadily increasing. The value proposition of bio-based and457 biodegradable products is linked to:

the use of renewable feedstock (like starch and its derivates) instead of fossil oil ornatural gas;

460 2. the waste recovery through biological recycling, thanks to their ability to461 biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

The Material Circularity Indicator (MCI), developed by the EMF, is a metric for quantifying "how much" a product is circular (MCI = 0, fully-linear product; MCI = 1, completely circular product) thus it represents a valuable tool for product eco-design purposes. However, it focuses solely on technical materials, mechanically recycled or reused, leaving out bio-based feedstocks and related biological treatments such as composting. Without common metrics it is not possible to pursue concrete actions, to 468 achieve measurable results and to provide unequivocal references for all products. This 469 research work aims at filling this gap through the development of a methodology coherent 470 with EMF MCI methodology but able to catch the specificities of bio-based and 471 biodegradable products and provide metrics for those innovative products. Direct uses are: 472 (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI 473 of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation:  $MCI_{(BB mulch})$ mulch = 0.89\*bio-based feedstock + 0.1.

479 The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles like the use of renewable materials and the reduction of 480 481 the amount of not recoverable waste. MCI will support the development of innovative 482 products just based on these two important characteristics specific for each BB product/application and end of life scenario- Bioeconomy, thus also BB products, can 483 484 provide valuable insights in transforming the current (linear) economy in a more circular one, however, the way the biomass is produced, processed and BB products are produced 485 486 are fundamental aspects to be properly assessed and monitored. This can be done using 487 specific methodologies like LCA. Within this context the proposed MCI has to be seen as 488 a complementary (quantitative) tool for further qualifying the sustainability of BB 489 products and not as a substitute tool.

- 490
- 491

÷

492

#### 493 Declaration of interest

494	The author declares that the research was conducted in the absence of any
495	commercial or financial relationships that could be construed as a potential conflict of
496	interest.
497	
498	Acknowledgements
499	The authors thanks prof. Andrea Contin for the fruitful discussion and contribution
500	to the sensitivity analysis, Francesco Degli Innocenti for providing valuable comments
501	and feedback on the topics addressed by the paper and Alessandra Novelli for the general
502	support in the MCI elaboration.
503	
504	
505	References
506	
	BASF, 2018. Biodegradable mulch film – clarification of polymer fate in soil. CIPA Congress, Bordeaux/Arcachon, France, May 2018.
	Bio-Based and Biodegradable Industries Association. BBIA reports. https://bbia.org.uk/reports/ (accessed 28 November 2019)
	<ul> <li>Briassoulis, D., Giannoulis, A., 2018. Evaluation of the functionality of bio-based plastic mulching films. Polym. Test. 67, 99–109. https://doi.org/10.1016/j.polymertesting.2018.02.019</li> </ul>
	Briassoulis D., Hiskakis, M., Babou, E., 2013. Technical specifications for mechanical recycling of agricultural plastic waste. Waste Management, Volume 33, issue 6, pages 1516-1530, ISNN 0956-053X. https://doi.org/10.1016/j.wasman.2013.03.004
	De Lèpinau, P. and Arbenz, A., 2016. Economic and environmental impact of soil

Ellen MacArthur Foundation & Granta Design, 2015. Circularity Indicators – An approach to measure circularity – Methodology.

contamination in mulching film, Plasticulture, N° 136, 28-48.

https://www.ellenmacarthurfoundation.org/assets/downloads/insight/Circularity-Indicators Methodology May2015.pdf.

- Ellen MacArthur Foundation, 2017. The New Plastic Economy: Rethinking the future of plastic & catalysing action.
- EN 16785-2:2016 Bio-based products Bio-based content Part 2: Determination of the bio-based content using the material balance method.
- EN 17033:2018 Plastics Biodegradable mulch films for use in agriculture and horticulture Requirements and test methods.
- EPLCA European Platform on LCA. https://eplca.jrc.ec.europa.eu/?page\_id=86
- Eubeler, J., Bernhanrd, M., Knepper, T., 2010. Environmental biodegradation of synthetic polymers II. Biodegradation of different polymer groups. Trends in Analytical Chemistry. 29, 1, 84-100
- European Commission, 2015. Closing the loop An EU action plan for the Circular Economy. COM(2015) 614 final. Brussels, 2.12.2015
- European Commission, 2018. A European Strategy for Plastics in a Circular Economy. COM(2018) 28 final. Brussels, 16.1.2018
- European Bioplastics. European Bioplastic publications. https://www.europeanbioplastics.org/news/publications/ (accessed 28 November 2019)
- Figuier, B., 2016. Plasticulture in Europe, Plasticulture, N° 136, 20-28
- Gao, H., Yan, C., Liu, Q., Ding, W., Chen, B., Li, Z., 2019. Effects of plastic mulching and plastic residue on agricultural production: A meta-analysis. Sci. Total Environ. 651, 484–492. https://doi.org/10.1016/J.SCITOTENV.2018.09.105
- Ghisellini, P., Cialani, C., Ulgiati, S., 2016. A review on circular economy: The expected transition to a balanced interplay of environmental and economic systems. J. Clean. Prod. https://doi.org/10.1016/j.jclepro.2015.09.007
- Institute of Bioplastics and Biocomposites, 2018. Biopolymers Facts and Statistics. Hochschule Hannover, University of Applied Sciences and Arts. Edition 5, ISSN 2510-3431.
- Joos, F., Roth, R., Fuglestvedt, J. S., Peters, G. P., Enting, I. G., Bloh, W. V., ... and Friedrich, T., 2013. Carbon dioxide and climate impulse response functions for the computation of greenhouse gas metrics: a multi-model analysis. Atmospheric Chemistry and Physics, 13(5), 2793-2825.

Kasirajan, S., Ngouajio, M., 2012. Polyethylene and biodegradable mulches for agricultural applications: a review. Agron. Sustain. Dev. 32, 501–529. https://doi.org/10.1007/s13593-011-0068-3

Lange, B.K., 2016. Revision of the fertilisers regulation – benefits of biodegradable mulch films. European Bioplastics. http://www.europarl.europa.eu/cmsdata/108931/Kristy%20Barbara%20Lange%20E UBP%20PPT2.pdf (accessed 28 November 2019)

- Le Moine, B., 2014. Agri-plastics waste management: a voluntary commitment from the industry. Presented at: Agricultural Film 2014 – International Conference on silage, mulch, greenhouse and tunnel films used in agriculture (15-17 September, Barcelona, Spain).
- Liu, E. K., He, W. Q., & Yan, C. R., 2014. 'White revolution' to 'white pollution' agricultural plastic film mulch in China. *Environmental Research Letters*, 9(9), 091001.
- Lloyd, S. M., & Ries, R., 2007. Characterizing, propagating, and analyzing uncertainty in life cycle assessment: A survey of quantitative approaches. *Journal of Industrial Ecology*, 11(1), 161-179.
- Lonca, G., Muggéo, R., Tétreault-Imbeault, H., Bernard, S., & Margni, M., 2018. A Bidimensional Assessment to Measure the Performance of Circular Economy: A Case Study of Tires End-of-Life Management. In *Designing Sustainable Technologies*, *Products and Policies* (pp. 33-42). Springer, Cham.
- Malinconico, M., 2017. Soil Degradable Bioplastics for a Sustainable Modern Agriculture. Green Chemistry and Sustainable Technology. Springer.
- Marten, E., Muller, R., and Deckwer W., 2003. Studies on the enzymatic hydrolysis of polyesters I. Low molecular mass model esters and aliphatic polyesters. Polymer Degradation and Stability, 80, 3, 485-501.
- Moreno, M. M., González-Mora, S., Villena, J., Campos, J. A., & Moreno, C., 2017. Deterioration pattern of six biodegradable, potentially low-environmental impact mulches in field conditions. *Journal of environmental management*, 200, 490-501.
- Mormile, P., Stahl, N., Malinconico, M., 2017. The World of Plasticulture, in: Malinconico, M. (Ed.), Soil Degradable Bioplastics for a Sustainable Modern Agriculture. pp. 1–21. https://doi.org/10.1007/978-3-662-54130-2\_1
- OWS, 2018. Accumulation of (bio)degradable plastics in soil. CIPA Congress 2018, Archacon, May 29.

Pico Y., Barcelò, D., 2019. Analysis and prevention of microplastics pollution in water: current perspectives and future directions. <u>ACS Omega</u>, 4, 6709-6719.

Formatted: Italian (Italy)

Plasticulture catalogues, 2018. http://plasticulture.qualif.e-catalogues.info (accessed 28 November 2019)

http://plasticulture.qualif.e catalogues.info

- <u>Razza, F., Degli Innocenti, F., 2012. Bioplastics from renewable resources: the benefits of biodegradability. Asia-Pacific Journal of Chemical Engineering</u>, 7 (Suppl. 3): S301– S309. https://doi.org/10.1002/apj.1648
- Scaringelli, M., Giannoccaro, G., prosperi, M., Lopolito, A., 2016. Adoption of biodegradable mulching films in agriculture: is there a negative prejudice towards materials derived from organic waste? *Italian Journal of agronomy*, 11:92.
- Shen M., Zhang, Y., Zhu, Y., Song, B., Zeng, G., Hu, D., Wen, Y., Ren, X., 2019. Recent advances in toxicological research of nanoplastics in the environment: A review. *Environmental Pollution*, 252: 511-521. https://doi.org/10.1016/j.envpol.2019.05.102
- Steinmetz, Z., Wollmann, C., Schaefer, M., Buchmann, C., David, J., Tröger, J., Muñoz, K., Frör, O., Schaumann, G.E., 2016. Plastic mulching in agriculture. Trading shortterm agronomic benefits for long-term soil degradation? Sci. Total Environ. 550, 690–705. https://doi.org/10.1016/J.SCITOTENV.2016.01.153
- Tamma, P., 2018. China's trash ban forces Europe to confront its waste problem, Politico, 2/21/2018.
- Touchaleaume, F., Martin-Closas, L., Angellier-Coussy, H., Chevillard, A., Cesar, G., Gontard, N., Gastaldi, E., 2016. Performance and environmental impact of biodegradable polymers as agricultural mulching films. Chemosphere 144, 433–439. https://doi.org/10.1016/j.chemosphere.2015.09.006
- <u>US-LCI</u> database. "Polylactide biopolymer resin at plant kg/RNA". <u>https://www.nrel.gov/lci/ (accessed 9 December 2019)</u>
- Verberne, J.J.H., 2016. Building circularity indicators. Eindhoven University of Technology.
- Wen, X., Du, C., Xu, P., zeng, G., Huang, D., Yin, L., Yin, Q., Hu, L., Wan, J., Zhang, J., Tan, S., Deng, R., 2018. Microplastic pollution in surface sediments of urban water areas in Changsha, China: Abundance, composition, surface textures. *Marine Pollution Bulletin*, 136: 414-423. https://doi.org/10.1016/j.marpolbul.2018.09.043.
- Witt, U., Einig, T., Yamamoto, M., Kleeberg, I., Deckwer, W., Muller, R., 2001. Biodegradation of aliphatic-aromatic copolyesters: evaluation of the final biodegradability and ecotoxicological impact of degradation intermediates. Chemosphere. 44, 289-299.

Field Code Changed

Formatted: Italian (Italy)
Formatted: Italian (Italy)

Zumstein, M., Schintlmeister, A., Nelson, T., Baumgartner, R., Wagner, M., Sander, M., McNeill, K., Woebken, D., Kohler, H., 2018. Biodegradation of synthetic polymers in soils: Tracking carbon into CO<sub>2</sub> and microbial biomass. Science Advances, 4, 7.

# Metrics for quantifying the circularity of bioplastics: the case of bio-based and biodegradable mulch films

3

4

11

12 13

14

Francesco Razza<sup>a</sup>, Cristiana Briani<sup>b</sup>, Tony Breton<sup>c</sup>, Diego Marazza<sup>d</sup>

<sup>a</sup> Novamont S.p.A. - Ecology of Product and Environmental Communication, Piazz.le Donegani 4, 05100 Terni, Italy

<sup>b</sup> CIRSA Centro Interdipartimentale di Ricerca per le Scienze Ambientali, Via S. Alberto 163, 48123 Ravenna, Italy

<sup>c</sup>Novamont S.p.A. – Via Fauser 8, 28100 Novara, Italy

<sup>d</sup>Department of Physics, University of Bologna, Viale B. Pichat 6/2, 40127 Bologna, Italy

# 15 Abstract

16 The concept of circularity and its quantification through the Material Circularity Indicator 17 (MCI) is well established for traditional plastic products. In this paper a methodological approach for calculating the circularity of bio-based and biodegradable (BB) products is 18 19 proposed and applied to BB mulch films. BB products are different from traditional 20 products in as much as they are sourced and regenerated (recycled) not through technical 21 cycles but the biological loop. The suggested method is an adaptation of the MCI where 22 two major changes were made: (i) the mass of the bio-based component corresponds to the 23 recycled material in input and (ii) the mass of the bio-based component leaving the system 24 through composting or biodegradation in soil is accounted as recycled. The modified MCI 25 supports the Eco-design of innovative BB products and allows for the comparison of their 26 circularity taking into account the biological source and the expected end of life process 27 such as biodegradation. To demonstrate the adaptation, the method has been applied to BB 28 mulch film. Results showed that the MCI of a biodegradable mulch film, characterized by 29 an average bio-based feedstock content of 30% is  $0.37 \pm 0.04$  in a 0-1 scale. For BB mulch 30 film, the amount of bio-based feedstock is the most sensitive factor and controls linearly 31 the value of the MCI.

32

*Keywords:* circularity indicators, circular economy, bioplastics, biodegradable
 mulch film, bio-based product, biodegradation

36	1 Introduction	3
37	1.1 The case study of mulch films	5
38	1.2 Goal of the paper	7
39	2 Materials and Methods	7
40	2.1 MCI accounting according to the EMF methodology	7
41	2.2 MCI accounting for bio-based and biodegradable (BB) products	l
42	2.3 MCI calculation for mulch films: scope, inventory and assumptions 14	1
43	2.4 Sensitivity analysis	7
44	3 Results	7
45	3.1 Sensitivity analysis	)
46	4 Discussion	2
47	5 Conclusions	1
48 49		

	Abbreviations
BB	Biodegradable and bio-based
CE	Circular Economy
d.m.	Dry matter
EMF	Ellen MacArthur Foundation
LCA	Life Cycle Assessment
LDPE	Low-Density Poly-Ethylene
MCI	Material Circularity Indicator
NRF	Non-Restorative Flows
PBAT	Polybutylene adipate terephthalate
PE	Poly-Ethylene
PLA	Polylactic acid
PHB	Poly hydroxy butyrate

## 51 **1 Introduction**

52 To overcome today's unsustainable model of 'take-make-dispose' and its related risks 53 such as hikes in raw material prices, pressures on the environment, shortage of global 54 resources and waste sinks, a circular approach needs to be applied. It is a new regenerative 55 economic view, based on a balance between economy, environment and society, a total 56 resource efficiency and a Zero Emission Strategy that aims to maximize products value 57 with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with 58 structural changes in environmental legislation, new logistics, technologies and sharing 59 schemes, the Circular Economy (CE) approach which is regenerative by design, aims at 60 closing materials loops, *i.e.* at reducing virgin materials input and waste output.

61 In December 2015, the European Commission developed an Action Plan for Circular 62 Economy (European Commission, 2015), where plastic was considered a priority to be 63 tackled. In January 2018, an EU Plastic Strategy (European Commission, 2018) was 64 adopted, in order to react to the increasing environmental problems concerning plastic 65 production, consumption, use and disposal along the same lines of the CE approach. Two 66 fundamental steps to increase the circularity of different plastic products are (i) the 67 abandonment of fossil fuels, *i.e.* currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development 68 69 of easily recyclable products which are recycled. Today, in EU the share of plastics 70 collected for recycling is 30% while the use of recycled plastics is just 6% (European 71 Commission, 2018).

Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and principles. This is true as long as the supply of renewable raw materials, generally from agriculture, is based on a sustainable approach and the conversion processes along the

supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA) 76 77 perspective (EPLCA - European Platform on LCA). While traditional plastics can be 78 mechanically recycled or incinerated with energy recovery, BB plastic products offer new 79 recycling routes in waste management, due to their biodegradability. Organic recycling 80 (through composting or anaerobic digestion) or in the case of specific applications such as 81 agricultural mulch films, biodegradation in the environment, offer additional recovery 82 options resulting in less wastes and less contamination of soil by plastic residues (Razza et 83 al., 2012; Lange, B., 2016). An extensive literature review about the potentialities and 84 benefits of renewable and compostable bioplastics, encompassing market perspective, 85 applications, economic effects etc. can be found here: (BBIA; European Bioplastics).

86 Nevertheless, the research and development of innovative products, such as the BB 87 products, implies the development of methodologies and metrics capable of measuring 88 their circularity. Without this it is not possible to achieve measurable results and 89 improving actions, as well as provide unequivocal references for comparisons of products 90 of the same type/category. In 2015 the Material Circularity Indicator (MCI) was 91 developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify 92 the regeneration of a product's material flow and is considered one of the few, among 93 sixteen CE indexes suiting a micro-scale assessment of circularity at product or company 94 level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled 95 materials. Furthermore, recovery and recycling through the biological cycle offered by 96 industrial composting, anaerobic digestion or biodegradation in natural environments are 97 not considered as end of life options. In order to apply the MCI system to BB plastic 98 products, the development of an enhanced methodology is necessary.

99 The approach proposed by the authors allows to quantify the circularity of BB plastic 100 products and to make comparisons with equivalent traditional plastic products. To
demonstrate the applicability of the proposed method a computational example for mulch
film products is provided. In so doing so, the paper aims at contributing to the Eco-design
of these innovative products.

104

## 1.1 The case study of mulch films

105 Plastic mulch films represent an important agronomical technique well established for the 106 production of many crops thanks to numerous agronomical advantages such as: increased 107 yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and 108 reduced use of pesticides; early crop production and reduced soil moisture loss 109 (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has 110 increased year-by-year, reaching a current global market estimated at 1.4 millions of 111 tonnes, mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017), and 112 covering  $80,000 \text{ km}^2$  of agricultural surface (0.6% of the global arable land). The mulch 113 film market in Europe is estimated by Agriculture Plastic & Environment and by the 114 European Bioplastic Associations at 76-80 kt. The most used raw material is Poly-115 Ethylene (PE) in its different forms, due to its processability, chemical resistance, high 116 durability and flexibility (Kasirajan and Ngouajio, 2012; Plasticulture, 2016 and 2018; 117 Shen, M. et al., 2019; Wen, X. et al., 2018).

118 Despite these benefits, manifold environmental and agronomic problems have been 119 pointed out. After its useful life - which in general does not exceed 1 to 3 months - the 120 mulch film has to be removed and properly disposed of, a time-consuming (about 16 hours 121 per hectare) and costly procedure (Scaringelli, M., 2016; Briassoulis, D., 2013). The 122 recovered film is usually heavily contaminated with soil and organic residues, making 123 mechanical recycling technically difficult and not a cost-efficient solution (Briassoulis et 124 al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most common end of life of 125 collected films in Europe is still landfilling (about 50%), followed by energy recovering

126 and finally mechanical recycling (Le Moine, 2014). Recent Chinese prohibition (January 127 2018) to import different types of wastes is heavily impacting the European agricultural 128 plastic waste management, highlighting the difficulty in properly recycling this type of 129 plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but 130 disposed of by burning in the field or by uncontrolled landfilling or left directly in the 131 (agricultural) soils, causing serious environmental concerns. An example is the "White 132 pollution" phenomena described in the Xinjiang Autonomous Region (China), in which 133 the residual plastic film can reach 200 kg/ha in the top soil with detrimental effects on 134 soils' quality, health and fertility (Liu, He, & Yan, 2014; Gao et al., 2019; Steinmetz et 135 al., 2016).

136 As a reaction, there has been significant research into novel materials especially related to 137 biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation 138 in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). The term "bio-mulch film" brings together several types of both bio-based and fossil oil-139 140 based biodegradable polymers and blends of them, such as polylactic acid (PLA), 141 polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or 142 copolymers. They biodegrade when exposed to bioactive environments such as soil and 143 compost (Kasirajan et al., 2012) which means that they can be left in situ to be fully 144 biodegraded after being used. Clearly the biodegradation rate of biodegradable bioplastics 145 is influenced by the environmental conditions such as the types of available bacteria, fungi 146 thus specific enzymes namely native microflora (Pico, Y. et al., 2019). However their 147 intrinsic biodegradability allow the complete biodegradation with times similar to natural 148 polymers such as cellulose used as reference by the relevant standards and certification 149 schemes.

150 The EN 17033:2018 is a new European Norm (standard) concerning "Plastics -151 Biodegradable mulch films for use in agriculture and horticulture - Requirements and test 152 methods", which sets the necessary tests and limits to define biodegradability, 153 performances and environmental impacts of BB much films. The material is considered 154 completely biodegradable if it achieves a complete biodegradation (absolute or relative to 155 the reference material) in a test period no longer than 24 months (mineralization into 156 CO<sub>2</sub>). Additionally, a control of constituents (such as metals) and eco-toxicity testing 157 (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test 158 with soil microorganisms) were required. A certified mulch film guarantees that the 159 product will completely biodegrade in the soil without adversely impacting on the 160 environment.

161 *1.2 Goal of the paper* 

162 The goal of the paper is to provide a general and common metric to measure the 163 circularity of a bio-based and biodegradable (BB) product and to apply the methodology at 164 product level to a category of products, namely bio-based and biodegradable mulch films.

165 **2** Materials and Methods

#### 166 2.1 MCI accounting according to the EMF methodology

167 The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation 168 (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number 169 that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production 170 provides for the exclusive use of virgin raw materials that turn into waste at the end of the 171 use phase of the product. Vice-versa, pure circularity includes the use of recycled 172 materials and does not produce wastes (regenerative streams). Circularity can be achieved 173 in different ways: as for the purpose of this paper, only recycling will be considered since 174 reuse is not an option for thin biodegradable mulch films. Since the method considers only 175 mass flows, the recycling corresponds to the recovery of materials for the original purpose 176 or for other purposes and excludes energy recovery, considered as a loss of materials equal 177 to landfill disposal. The materials recovered feed back into the process as recycled 178 feedstock.

The MCI methodology differentiates 'technical cycles' from 'biological cycles', modelling only the former. The first contains products and materials re-entering into the system (market) with the highest possible qualities and for as long as possible (thanks to reuse, repair, refurbishment and recycling) and the latter includes biological materials used in cascade until their restoration into the biosphere and the re-constitution of natural resources.

The material flows associated to the production of a generic technical cycle from nonrenewable sources are summarized in Figure 1. The dashed lines indicate that recycled feedstock does not have to be sourced from the same product but can be acquired on the market. With reference to Figure 1, the list of the parameters used in the EMF methodology is reported in Table 1, while the equations relevant for the analysis carried out in this paper are described in the following sections (Table 2, Chapter 2.2).





192 Figure 1: Diagram of material flows and associated variables of a generic

193 product (modified from Ellen MacArthur Foundation & Granta Design, 2015).

194

 Table 1: Parameters and relative definitions used in the EMF methodology.

Parameter	Definition
М	Total mass of the product
$F_R$	Fraction of mass of a product's feedstock from recycled sources
$F_U$	Fraction of mass of a product's feedstock from reused sources
V	Mass of virgin feedstock used in a product
$C_R$	Fraction of mass of a product being collected to go into a recycling process
$C_U$	Fraction of mass of a product going into component reuse
$E_{C}$	Efficiency of the recycling process used for the portion collected for recycling
$E_F$	Efficiency of the recycling process used to produce recycled feedstock for a
	product

W	Total mass of unrecoverable waste associated with a product	
W <sub>0</sub>	Mass of unrecoverable waste (landfill, waste to energy and any other type of	
	process where the materials are no longer recoverable)	
W <sub>C</sub>	Mass of unrecoverable waste generated in the process of recycling parts of a	
	product (after use)	
$W_F$	Mass of unrecoverable waste generated when producing recycled feedstock for	
	a product	
X	Utility of a product, calculated as $X = (L/L_{av})(U/U_{av})$	
L	Actual average lifetime of a product	
$L_{av}$	Actual average lifetime of an industry-average product of the same type	
U	Actual average number of functional units achieved during the use phase of a	
	product	
$U_{av}$	Actual average number of functional units achieved during the use phase of an	
	industry-average product of the same type	

196 The Material Circularity Indicator is determined as follows:

where *LFI* is the Linear Flow Index measuring the flows of virgin materials andunrecoverable wastes associated to the examined product.

A function of the utility, -, is used to correct the *LFI*. The function *F* is chosen in such a way that improvements of the utility of a product (e.g., by using it longer) have the same impact on its MCI as a reuse of components, leading to the same amount of reduction of virgin material use and unrecoverable waste. Setting a = 0.9, MCI takes, by convention, the value 0.1 for a fully linear product (*i.e.*, *LFI* = 1) whose utility equals the industry average (*i.e.*, X = 1). This leaves some margin to distinguish between processes with a high linearity but different utilities.

## 206 2.2 MCI accounting for bio-based and biodegradable (BB) products

207 To apply the EMF methodology to BB products, formulas and flows (Figure 1 and Figure208 2) are adapted as it follows:

- 209 1. The fraction of the recycled feedstock,  $F_R$ , corresponds to the share of the bio-210 based feedstock content in the final BB product,  $F_{R(i)}$ . It is the ratio of the d.m. 211 amount of bio-based feedstock per d.m. amount of the total mass of BB 212 product (EN 16785-2:2016).
- 213 2. The fraction of restorative mass going into a recycling process,  $C_R$ , corresponds 214 to the share of bio-based feedstock content in the BB product biologically 215 recovered (*e.g.* through composting) or biodegraded in the natural 216 environment, as it happens for specific applications (e.g. biodegradable mulch 217 film, *etc.*). It is the ratio of the d.m. amount of bio-based feedstock per d.m. 218 amount of the total mass of BB product that is biologically recycled.

The modified scheme is shown in Figure 2. Table 2 lists the formulas as adapted to BBproducts.

221

222

**Table 2:** List of formulas as developed by EMF methodology compared to the proposed adaptation to BB products.

# EMF methodology

## Adaptation to BB products

The mass of fossil-based feedstock which may be contained in BB products (V) is obtained as a difference of the total mass (M) minus the bio-based fraction; in this case the  $F_R$  in the EMF methodology corresponds to the sum of the fractions of all the biobased feedstock/s used in manufacturing the BB product. Therefore, is the total bio-based feedstock mass in the product. In single-use products, such as mulch films, reuse is not considered for BB products, so that  $F_U = C_U = 0$ .

223

 $W_F$  is the total amount of unrecoverable waste associated to the production of bio-based 230 231 feedstock used to produce BB products (i.e. the amount of uncoverable waste per unit of 232 BB product). Bio-based feedstocks such as starch, PLA, PHB etc. generate non-restorative 233 flows which can be quantified. Such unrecoverable waste correspond to  $R_{(i)}$ , the specific 234 amount of waste generated within cradle-to-gate boundaries per unit of bio-based 235 feedstock going into manufacturing, and it is estimated through LCA studies. Thus all 236 inputs from growth and harvesting phases and the related wastes generated by fertilisers and pesticides are here accounted.  $R_{(i)}$  can be easily found in specific literature or life 237 238 cycle inventories (LCI) present in LCA databases. In the calculation of  $W_F$ , also the 239 efficiency of manufacturing process of BB products  $E_P$  is considered, as the ratio of the

- 240 overall bio-based feedstock content in the final BB product to the bio-based feedstock in
- 241 input to the manufacturing process.
- 242 The material flows associated to the production of a generic BB product are summarized
- in Figure 2.



Figure 2: Description of material flows adaptation to BB products; in this paper, the reuse flow is out of scope ( $C_U = F_U = 0$ ).

The biodegradation of bio-based feedstock does not imply the generation of waste  $W_C$  as it occurs in a standard mechanical recycling process. This implies that  $C_R$  and  $E_C$  (*i.e.* the efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to biological treatment (composting) or biodegraded in a natural environment, is fully transformed in its chemical elements (C, H and O mainly) derived from the decomposition of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites, 254 2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the 255 environment and are then available in the respective biogeochemical cycles. The 256 (biodegradable) fossil portion behaves as well; consequently,  $W_C = 0$ .

Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular feedstock, since it derives from carbon stored for millions of years and extracted by man, not being part of the active and fast biogeochemical carbon cycle. This is accounted in the quantification of  $W_0$ , the mass of unrecoverable waste from use (*i.e.* the linear stream going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total amount of fossil-based feedstock.

Since  $W_F$  and  $W_C$  are associated to complete different processes and  $W_C$  is always equal zero, the double counting issue does not occur and the quantification of W and *LFI* is modified as reported in Table 2.

### 266 2.3 MCI calculation for mulch films: scope, inventory and assumptions

The new formulas reported in Table 2 were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the biobased feedstock content and the process yield is close to 1.

In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch,  $F_{(S)}$ , and 7% of a bio-based additive,  $F_{(BA)}$ ), while the rest was assumed to consist of fossil feedstock (



- 281
- 282
- 283



Figure 3: Examples of hypothetical bio-based polymers; in this paper, the first option on the left (starch blend 30/70) has been chosen for carrying out the numerical MCI calculation (working hypothesis). The figure considers a 100%-efficiency in every phase of production, so that the residues are equal to zero; the same assumption is done in this paper. \*TPS (Thermoplastic starch), starch content 75%; \*\*Ratio TPS/Polymer; modified from Institute of Bioplastics and Biocomposites, 2018.

	BB mulch film
Material	30% bio-based feedstock (23% starch + 7% bio-
	based additive) + 70% fossil feedstock
Thickness (μm)	12
Density (g/cm <sup>3</sup> )	1.25
Weight (g/m <sup>2</sup> )	15.2
Functional unit	$6000 \text{ m}^2/\text{ha}$ (the actual mulched soil in a hectare
(the covering of the agricultural land)	is generally equal to the 60% of the total area;
	Malinconico, 2017)

302 In the calculation of MCI for the BB mulch film, the adapted formulas were used together 303 with assumptions. As stated before, BB mulch films are blends of bio-based and fossil 304 based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE 305 mulch film that has to be removed and disposed of, the BB mulch film is left in soil where 306 it undergoes an ultimate biodegradation (so that  $C_R = 1$ ) with no waste (so that  $E_C = 1$ ), in 307 respect of the specific standard EN 17033:2018. As a result of polymers' decomposition, 308 the derived (biogenic) C, H and O finally return into biosphere (atmosphere, 309 microorganism biomass, organic material pool) (OWS, 2018), and back into 310 biogeochemical cycles in a relatively short time ("Biogenic elements accounted as 311 recycled" in Figure 2), with the exception of humified compounds. Actually, also C, H 312 and O deriving from fossil-based sources undergo biodegradation (Zumstein, M.T., 2018) 313 but they are not considered as a regenerative flow ("Waste from non-restorative flow" in 314 *Figure 2*) and their "wastes" are indeed calculated in  $W_0$ .

Applying a conservative approach,  $W_F$ , the waste generated by the production of each biobased feedstock, is quantified considering a "cradle to gate" LCA study. The estimated solid wastes  $R_{(i)}$  for the presented case study are related to the production of starch ( $F_{(S)}$ ), with an amount  $R_{(S)}$  of 0.014 kg of waste per kg of renewable feedstock (source: personal communication A. Novelli), and to the production of the bio-based additive ( $F_{(BA)}$ ), with  $R_{(BA)}$  equals to 0.025 kg waste/kg renewable feedstock (US-LCI database). As assumed in

321

Figure 3, the production efficiency of BB product  $E_P$  (how much bio-based feedstock is needed for every unit of BB product) is estimated equal to 1 and no unrecoverable wastes are generated by the process.

In addition, an explorative sensitivity analysis has been performed regarding exclusively the amount of bio-based feedstock content of the BB mulch film, *i.e.* (*i.e.*,  $F_{(S)}$  +  $F_{(BA)}$ ), as shown in Figure 4 (Chapter 3). Considering the characteristics of the films (weight, g/m<sup>2</sup>, or thickness, µm, and density, g/cm<sup>3</sup>) and the relative functional unit (6000 m<sup>2</sup>/ha, Table 3), it is possible to calculate a mass, *M*, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas reported in Table 2 (Chapter 2.2) are applied. Results are shown in Table 4.

332

333 2.4 Sensitivity analysis

334 A sensitivity analysis was conducted for BB mulch film to examine the effects of 335 changing the main variables. Given a non-linear dependence of results on parameter 336 values, a Monte Carlo approach (see, e.g., Lloyd and Ries, 2008) has been adopted. The 337 model has been implemented using specifically written routines in the C++ programming 338 language. The model was run with 100,000 events for BB mulch film, where the value of 339 each parameter has been randomly chosen following a Gaussian distribution with a 340 standard deviation within a range of possible and realistic values (Table 5 and Error! 341 Reference source not found.; Figure 5 and Figure 6).

## **342 3 Results**

- 343 Figure 4 shows how the value of the MCI varies according to the percentage variation of
- 344 the bio-based feedstock in the total mass of the product.
- 345
- 346

**Table 4:** Resulting parameters in the calculation of MCI for BB mulch film.

Parameter BB mulch film



Figure 4: MCI as a function of the amount of bio-based feedstock/s in the BB mulch film  $\Sigma F_{R(i)}$ , expressed as the percentage of all the bio-based feedstock/s of the mulch film on dry mass basis (X-axis). The dots correspond to the three different hypothetical bioplastic compositions of Figure 3.

#### 355 3.1 Sensitivity analysis

The results of the sensitivity analysis are presented in the followings Table 5 and Figure 5 and Figure 6. The accuracy band is a fraction of the average and corresponds to a probability of 95%. It has been chosen in order to be representative of the variability of the product category, the BB mulch films. The simulation can thus be regarded as a system composed by a high number of companies, each producing films with different characteristics, that are accounted for in the accuracy band.

362 *Table 5:* Parameters used for the sensitivity analysis of the BB mulch film. (\*\*) The
363 Accuracy Band is defined as twice the standard deviation of the distribution.

Variable name	Average	Accuracy Band (**)	Unit
Μ	1000.00	0%	kg
$F_{(S)}/F_{(BA)}$	3.29	10%	fraction
$\mathbf{F}_{(S)} + \mathbf{F}_{(BA)}$	0.30	30%	fraction
$\mathbf{F}_{\mathbf{U}}$	0.00	0%	fraction
C <sub>U</sub>	0.00	0%	fraction
R <sub>(S)</sub>	0.014	100%	fraction
R <sub>(BA)</sub>	0.025	100%	fraction
E <sub>C</sub>	1	0%	fraction
E <sub>P</sub>	0.95	10%	fraction
C <sub>R</sub>	1.00	0%	fraction





Figure 6: The most sensitive and relevant parameters in the calculation of the

F(S)+F(BP)

R<sub>(S)</sub>

R<sub>(BP)</sub>

Ep

371 MCI of the BB mulch films.

F(s/F(BP)

#### **372 4 Discussion**

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

378 (1) fossil-based feedstocks or component materials embodied in the BB products whatever379 is the final disposal (even biological recycling) shall be considered as non-restorative;

(2) bio-based component materials embodied in the BB product that go to biological
recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be
considered restorative as long as they flow through the biosphere safely, without any harm
to the environment (e.g. no toxicity effects).

(3) bio-based component materials embodied in the BB product that go to incineration andlandfill shall be considered as non-restorative;

386 The justification of these rules is described in the following.

387 Fossil-based component materials in the product derive from deposits where they 388 remained stocked for a geological time scale. Once the product is mineralised, its fossil-389 based portion will be accounted as non-regenerative and therefore linear, due to its origin 390 (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological 391 cycles, like CO<sub>2</sub> in the atmosphere and other streams, since both fossil-based and bio-392 based component materials will physically and chemically behave the same, once 393 biodegraded. However, the source of the bio-based carbon was circular before its use 394 (concept of "carbon neutrality", equilibrium between the biogenic carbon released and the 395 carbon absorbed by plants) and will maintain its circularity provided that the carbon is 396 released into the atmosphere at the same rate. The reason has its origin in the EMF general 397 provisions stating that "biologically sourced materials can only be considered part of a 398 Circular Economy if materials are not used faster than they can be restored naturally" 399 (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the 400 bio-based components are still considered linear, maintaining consistency with EMF 401 principles. Basically, a complete circularity for a BB product is satisfied when its 402 renewable components are 100% bio-based and they go 100% to biological recycling or 403 biodegraded in the environment (for specific application like mulch film).

404 As for provision (3), a material health rule has its origin in manyfold normative definitions 405 of the CE. In addition, the EMF definition of biological cycles is that of non-toxic 406 materials which are restored into the biosphere and the CE is defined as such if it can 407 "eliminate the use of toxic chemicals". The need of a safety clause has been reviewed 408 under many aspects by Verberne (2016) and can be put as a postulate of the restoration 409 principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the 410 REACH Regulation (EC 1907/2006). In the specific case, the material complies with the 411 standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism 412 groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important 413 ecological processes maintaining soil functions, c) all relevant exposure pathways as soil 414 pore water, soil pore air and soil material.

A comprehensive approach for MCI calculation should also include non-restorative flows generated at upstream level like biomass growth, in the specific case corn, and biomass conversion processes like starch extraction and refining. Specifically these non-restorative flows correspond to the overall non-recyclable wastes associated to the bio-based feedstock supply thus non-recyclable waste from fertilizer and pesticide production, nonrecyclable scraps from conversion processes, etc. In this study such flows of nonrestorative waste coming from upstream manufacturing operations were included for the bio-based feedstocks ( $R_{(i)}$ ) used in manufacturing the BB mulch film applying "cradle to gate" LCA methodology. However, we observed that the inclusion of upstream unrecoverable waste does not significantly influence the MCI results in the chosen case study, since the respective amounts are small. The specific unrecoverable waste for starch and bio-based additive (i.e. kg of waste/kg of bio-based feedstock) were estimated at 0.014 and 0.025, respectively.

428

The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation y = 0.89x + 0.1, where y is the MCI and x is the bio-based feedstock content, therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input is decisive.

434 Apart from the specific application analysed in this paper, the proposed MCI method can
435 be easily applied and calculated for any kind of BB product as long as the following
436 information are available:

The bio-based feedstock content, determined according to the standard EN 167852:2016, if the composition is known, or directly provided by the BB product manufacturer.

• The End of Life scenario of the studied BB product (real or hypothetical).

The amount of un-recoverable waste associated to the production of bio-based
feedstock contained in the BB product. They can be derived from LCA databases or other
specific sources.

## 443 **5** Conclusions

444 Bioplastic market is steadily increasing. The value proposition of bio-based and445 biodegradable products is linked to:

the use of renewable feedstock (like starch and its derivates) instead of fossil oil or
natural gas;

448

449

2. the waste recovery through biological recycling, thanks to their ability to biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

450 The Material Circularity Indicator (MCI), developed by the EMF, is a metric for 451 quantifying "how much" a product is circular (MCI = 0, fully-linear product; MCI = 1, 452 completely circular product) thus it represents a valuable tool for product eco-design 453 purposes. However, it focuses solely on technical materials, mechanically recycled or 454 reused, leaving out bio-based feedstocks and related biological treatments such as 455 composting. Without common metrics it is not possible to pursue concrete actions, to 456 achieve measurable results and to provide unequivocal references for all products. This 457 research work aims at filling this gap through the development of a methodology coherent 458 with EMF MCI methodology but able to catch the specificities of bio-based and 459 biodegradable products and provide metrics for those innovative products. Direct uses are: 460 (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI 461 of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation:  $MCI_{(BB mulch})$ mulchmulch = 0.89\*bio-based feedstock + 0.1.

The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles like the use of renewable materials and the reduction of the amount of not recoverable waste. MCI will support the development of innovative products just based on these two important characteristics specific for each BB

471	product/application and end of life scenario Bioeconomy, thus also BB products, can
472	provide valuable insights in transforming the current (linear) economy in a more circular
473	one, however, the way the biomass is produced, processed and BB products are produced
474	are fundamental aspects to be properly assessed and monitored. This can be done using
475	specific methodologies like LCA. Within this context the proposed MCI has to be seen as
476	a complementary (quantitative) tool for further qualifying the sustainability of BB
477	products and not as a substitute tool.
478	
479	
480	
481	Declaration of interest
482	The author declares that the research was conducted in the absence of any
483	commercial or financial relationships that could be construed as a potential conflict of
484	interest.
485	
486	Acknowledgements
487	The authors thanks prof. Andrea Contin for the fruitful discussion and contribution
488	to the sensitivity analysis, Francesco Degli Innocenti for providing valuable comments
489	and feedback on the topics addressed by the paper and Alessandra Novelli for the general
490	support in the MCI elaboration.
491	
492	References
493	
	BASF, 2018. Biodegradable mulch film – clarification of polymer fate in soil. CIPA Congress, Bordeaux/Arcachon, France, May 2018.

- Bio-Based and Biodegradable Industries Association. BBIA reports. <u>https://bbia.org.uk/reports/</u> (accessed 28 November 2019)
- Briassoulis, D., Giannoulis, A., 2018. Evaluation of the functionality of bio-based plastic mulching films. Polym. Test. 67, 99–109. https://doi.org/10.1016/j.polymertesting.2018.02.019
- Briassoulis D., Hiskakis, M., Babou, E., 2013. Technical specifications for mechanical recycling of agricultural plastic waste. Waste Management, Volume 33, issue 6, pages 1516-1530, ISNN 0956-053X. <u>https://doi.org/10.1016/j.wasman.2013.03.004</u>
  De Lèpinau, P. and Arbenz, A., 2016. Economic and environmental impact of soil contamination in mulching film, Plasticulture, N° 136, 28-48.
- Ellen MacArthur Foundation & Granta Design, 2015. Circularity Indicators An approach to measure circularity Methodology. https://www.ellenmacarthurfoundation.org/assets/downloads/insight/Circularity-Indicators\_Methodology\_May2015.pdf.
- Ellen MacArthur Foundation, 2017. The New Plastic Economy: Rethinking the future of plastic & catalysing action.
- EN 16785-2:2016 Bio-based products Bio-based content Part 2: Determination of the bio-based content using the material balance method.
- EN 17033:2018 Plastics Biodegradable mulch films for use in agriculture and horticulture Requirements and test methods.
- EPLCA European Platform on LCA. https://eplca.jrc.ec.europa.eu/?page\_id=86
- Eubeler, J., Bernhanrd, M., Knepper, T., 2010. Environmental biodegradation of synthetic polymers II. Biodegradation of different polymer groups. Trends in Analytical Chemistry. 29, 1, 84-100
- European Commission, 2015. Closing the loop An EU action plan for the Circular Economy. COM(2015) 614 final. Brussels, 2.12.2015
- European Commission, 2018. A European Strategy for Plastics in a Circular Economy. COM(2018) 28 final. Brussels, 16.1.2018
- European Bioplastics. European Bioplastic publications. https://www.europeanbioplastics.org/news/publications/ (accessed 28 November 2019)
- Figuier, B., 2016. Plasticulture in Europe, Plasticulture, N° 136, 20-28
- Gao, H., Yan, C., Liu, Q., Ding, W., Chen, B., Li, Z., 2019. Effects of plastic mulching and plastic residue on agricultural production: A meta-analysis. Sci. Total Environ. 651, 484–492. https://doi.org/10.1016/J.SCITOTENV.2018.09.105

- Ghisellini, P., Cialani, C., Ulgiati, S., 2016. A review on circular economy: The expected transition to a balanced interplay of environmental and economic systems. J. Clean. Prod. https://doi.org/10.1016/j.jclepro.2015.09.007
- Institute of Bioplastics and Biocomposites, 2018. Biopolymers Facts and Statistics. Hochschule Hannover, University of Applied Sciences and Arts. Edition 5, ISSN 2510-3431.
- Joos, F., Roth, R., Fuglestvedt, J. S., Peters, G. P., Enting, I. G., Bloh, W. V., ... and Friedrich, T., 2013. Carbon dioxide and climate impulse response functions for the computation of greenhouse gas metrics: a multi-model analysis. Atmospheric Chemistry and Physics, 13(5), 2793-2825.
- Kasirajan, S., Ngouajio, M., 2012. Polyethylene and biodegradable mulches for agricultural applications: a review. Agron. Sustain. Dev. 32, 501–529. https://doi.org/10.1007/s13593-011-0068-3
- Lange, B.K., 2016. Revision of the fertilisers regulation benefits of biodegradable mulch films. European Bioplastics. http://www.europarl.europa.eu/cmsdata/108931/Kristy%20Barbara%20Lange%20E UBP%20PPT2.pdf (accessed 28 November 2019)
- Le Moine, B., 2014. Agri-plastics waste management: a voluntary commitment from the industry. Presented at: Agricultural Film 2014 International Conference on silage, mulch, greenhouse and tunnel films used in agriculture (15-17 September, Barcelona, Spain).
- Liu, E. K., He, W. Q., & Yan, C. R., 2014. 'White revolution' to 'white pollution' agricultural plastic film mulch in China. *Environmental Research Letters*, 9(9), 091001.
- Lloyd, S. M., & Ries, R., 2007. Characterizing, propagating, and analyzing uncertainty in life cycle assessment: A survey of quantitative approaches. *Journal of Industrial Ecology*, 11(1), 161-179.
- Lonca, G., Muggéo, R., Tétreault-Imbeault, H., Bernard, S., & Margni, M., 2018. A Bidimensional Assessment to Measure the Performance of Circular Economy: A Case Study of Tires End-of-Life Management. In *Designing Sustainable Technologies, Products and Policies* (pp. 33-42). Springer, Cham.
- Malinconico, M., 2017. Soil Degradable Bioplastics for a Sustainable Modern Agriculture. Green Chemistry and Sustainable Technology. Springer.
- Marten, E., Muller, R., and Deckwer W., 2003. Studies on the enzymatic hydrolysis of polyesters I. Low molecular mass model esters and aliphatic polyesters. Polymer Degradation and Stability, 80, 3, 485-501.

- Moreno, M. M., González-Mora, S., Villena, J., Campos, J. A., & Moreno, C., 2017. Deterioration pattern of six biodegradable, potentially low-environmental impact mulches in field conditions. *Journal of environmental management*, 200, 490-501.
- Mormile, P., Stahl, N., Malinconico, M., 2017. The World of Plasticulture, in: Malinconico, M. (Ed.), Soil Degradable Bioplastics for a Sustainable Modern Agriculture. pp. 1–21. https://doi.org/10.1007/978-3-662-54130-2 1
- OWS, 2018. Accumulation of (bio)degradable plastics in soil. CIPA Congress 2018, Archacon, May 29.
- Pico Y., Barcelò, D., 2019. Analysis and prevention of microplastics pollution in water: current perspectives and future directions. *ACS Omega*, 4, 6709-6719.
- Plasticulture catalogues, 2018. http://plasticulture.qualif.e-catalogues.info (accessed 28 November 2019)
- Razza, F., Degli Innocenti, F., 2012. Bioplastics from renewable resources: the benefits of biodegradability. *Asia-Pacific Journal of Chemical Engineering*, 7 (Suppl. 3): S301– S309. https://doi.org/10.1002/apj.1648
- Scaringelli, M., Giannoccaro, G., prosperi, M., Lopolito, A., 2016. Adoption of biodegradable mulching films in agriculture: is there a negative prejudice towards materials derived from organic waste? *Italian Journal of agronomy*, 11:92.
- Shen M., Zhang, Y., Zhu, Y., Song, B., Zeng, G., Hu, D., Wen, Y., Ren, X., 2019. Recent advances in toxicological research of nanoplastics in the environment: A review. *Environmental Pollution*, 252: 511-521. https://doi.org/10.1016/j.envpol.2019.05.102
- Steinmetz, Z., Wollmann, C., Schaefer, M., Buchmann, C., David, J., Tröger, J., Muñoz, K., Frör, O., Schaumann, G.E., 2016. Plastic mulching in agriculture. Trading shortterm agronomic benefits for long-term soil degradation? Sci. Total Environ. 550, 690–705. https://doi.org/10.1016/J.SCITOTENV.2016.01.153
- Tamma, P., 2018. China's trash ban forces Europe to confront its waste problem, Politico, 2/21/2018.
- Touchaleaume, F., Martin-Closas, L., Angellier-Coussy, H., Chevillard, A., Cesar, G., Gontard, N., Gastaldi, E., 2016. Performance and environmental impact of biodegradable polymers as agricultural mulching films. Chemosphere 144, 433–439. https://doi.org/10.1016/j.chemosphere.2015.09.006
- US-LCI database. "Polylactide biopolymer resin at plant kg/RNA". <u>https://www.nrel.gov/lci/</u> (accessed 9 December 2019)
- Verberne, J.J.H., 2016. Building circularity indicators. Eindhoven University of Technology.

- Wen, X., Du, C., Xu, P., zeng, G., Huang, D., Yin, L., Yin, Q., Hu, L., Wan, J., Zhang, J., Tan, S., Deng, R., 2018. Microplastic pollution in surface sediments of urban water areas in Changsha, China: Abundance, composition, surface textures. *Marine Pollution Bulletin*, 136: 414-423. https://doi.org/10.1016/j.marpolbul.2018.09.043.
- Witt, U., Einig, T., Yamamoto, M., Kleeberg, I., Deckwer, W., Muller, R., 2001. Biodegradation of aliphatic-aromatic copolyesters: evaluation of the final biodegradability and ecotoxicological impact of degradation intermediates. Chemosphere. 44, 289-299.
- Zumstein, M., Schintlmeister, A., Nelson, T., Baumgartner, R., Wagner, M., Sander, M., McNeill, K., Woebken, D., Kohler, H., 2018. Biodegradation of synthetic polymers in soils: Tracking carbon into CO<sub>2</sub> and microbial biomass. Science Advances, 4, 7.

# HIGHLIGHTS

- A modification of the MacArthur methodology on product circularity (i.e. Material Circularity Indicator MCI) has been developed to make it applicable to bio-based and biodegradable (BB) products.
- 2. The proposed metric has been applied to a specific case study: the bio-based and biodegradable mulch film.
- 3. Results show that a biodegradable mulch film with a 30% of bio-based feedstock content is characterized by a MCI of  $0.37 \pm 0.04$  in a 0-1 scale.
- 4. For a BB mulch film the amount of bio-based feedstock is the most sensitive factor and controls linearly the value of the MCI.

# **REVISED HIGHLIGHTS**

- 5. A MCI methodology suitable for Bio-based and Biodegradable (BB) products has been developed.
- 6. The proposed metric has been applied to a specific case study: BB mulch film.
- 7. BB mulch film with a 30% of renewable feedstock is characterized by a MCI of  $0.37 \pm 0.04$  in a 0-1 scale.
- 8. The amount of renewable feedstock is the most sensitive factor of the MCI

## **Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

□The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

**Francesco Razza:** Conceptualization, Methodology, Writing - original draft, Writing - Review & Editing, Data Curation, Investigation, Validation, Supervision

Cristiana Briani: Writing - Original Draft, Validation

Tony Breton: Writing - Original Draft, Supervision

Diego Marazza: Writing - Original Draft, Data curation, Validation

Dear Reviewers,

Many thanks for your time. The table below provides our replies to your further comments and the description of the changes made on the paper for each raised point. Many thanks again to all of you for the valuable comments and suggestions that allow us to further improve the work.

n	Reviewers' comments	Revisions made in the paper
	Reviewer #1	
	All the issues mentioned by the reviewers have been	
1	addressed, and the paper quality has been greatly	Many thanks
	improved. Now, the manuscript may be considered	Many thanks
	for acceptance.	
	Reviewer #3	
	The authors have prepared an extensive revision of	Many thanks
	the original manuscript and addressed the	
2	reviewers' comments in a satisfactory manner. I	
2	have one major and one minor comment at this	
	stage, and recommend acceptance of the work. I do	
	not need to see a possible further revision.	
	Major comment:	Actually, we fully agree with you. The MCI here
	I am not convinced that complex material	proposed is meaningful for judging how much
	interactions (fossil carbon biodegrading, harmful	circular a bio-based and biodegradable product is
	organic waste, bio-based material recycling, etc.) can	only if the bio-based material/product does not
	be meaningfully represented in a single indicator	cause toxic concerns or issues. This is our
3	such as the MCI or its derivatives. But that is	postulate reported in R396-406.
	something that the community should decide and	
	not the reviewers, by taking up your work or not.	That said we have further pointed out this very
	But I ask you to add a short remark on the critique of	important aspect in the conclusion and made an
	the general usefulness of this indicator in the	addition in R468-471.
	discussion section.	
	One minor comment remains:	It stands for dry matter. On page 1 under
4	+ L202 and other places: The abbreviation d.m. is not	"abbrevations" section is reported
	clear to me. Please spell out! Dry mass?	d.m. = Dry matter.
	Reviewer #4	
	The authors have satisfactorily addressed the	Many thanks
5	comments raised by the previous reviewers and	
	appropriately modified the manuscript.	
	This work attempts to augment the MCI proposed by	Many thanks for this comment. Following your
	EMF. Although the need for the work is clear,	hint we have found that the EMF methodology
	however recently (in 2019) EMF has already	has been recently changed
	proposed MCI for biological products. Hence,	https://www.ellenmacarthurfoundation.org/asse
	authors need to compare and contrast the MCI	ts/downloads/ce100/MCI-SC-28Nov-2019-
	proposed in this work with EMF MCI for bio	Master-MB-4.pdf however we would like to point
	products.	out that our work started <u>long before</u> the
6		changes of MCI and in an complete
		independently way. For the sake of clarity we
		here report the (documented) main stages of our
		original work followed by our proposal for
		handling this issue.
		Story of our paper.
		<b>2017</b> : preliminary idea of the methodology
		<b>2018</b> : the beta version of the methodology is
		presented within the third working group of the

Italian Circular Economy Stakeholder Platform (ICESP <u>www.icesp.it</u>). On page 38 of the ICESP report (dated December 2018) here available https://www.icesp.it/landing/docs/gdl/gdl3/REP ORT GdL3%20Strumenti%20per%20la%20misura zione%20dell%E2%80%99economia%20circolare. pdf a brief description of the - not finalmethodology is provided. Please note that the report is dated December 2018 and it was developed in the last four month period of 2018. 2019: within StarBioPro project http://www.starprobio.eu/ thanks to the collaboration between Novamont and the University of Bologna (PhD D. Marazza and Prof. A. Contin) the methodology was further developed and improved till the present version. The first submission of the paper occurred the 31<sup>st</sup> of April 2019. At that time we were not aware about the EMF initiative about biological products so we wrote our paper blissfully unaware.

That said, we have seen that some consideration of the recast EMF methodology are very close to what we proposed.

As an example,

- a principle "ensuring biological materials remain uncontaminated and biologically accessible" has been added
- virgin material now considers the biological materials fraction in its formula
- all formulas now include the contribution of biological materials
- composting has been added as an end-of-life option.

However, the recast MCI differs now from our proposal because it accounts for energy recovery of biological materials which can make the MCI of a BB product higher than what we propose. Other points are still open such as the demonstration that the feedstock has been extracted from "Sustained Production".

To compare and defend our choices against the recast MCI would require to re-write almost completely sections 2 and 3, all figures, tables and formulas included. Section 4 ought to be extended and oriented to a comparison of our methodological proposal versus the recast MCI. We believe this makes the case for an additional, different paper, while the purpose of this paper is still justified. Indeed,

we would like to remark that the new MCI does not provide any specific guidance on practical

	cases as we did for the biodegradable mulch film. For these reasons we believe our paper can give an important scientific contribution to the debate.
	We decided to add an addendum in the paper reciting as follows:
	While this paper was undergoing peer review the authors became aware that the EMF published an update of the MCI methodology (Ellen MacArthur Foundation & Granta Design, 2019) including the extension of it to include the treatment of biological materials. This update introduces new definitions and formulas. The authors believe that most of the changes regarding accounting are in the direction here proposed and that this study can contribute as an illustration on how the material circularity of a biological based material can be addressed in a real case study. Furthermore the authors would like to highlight that the proposed methodology started long before the EMF changes: specifically the original idea dated back to 2017 and a beta version of it - not as it is now - was presented in the middle of 2018 at the Italian Circular Economy Stakeholder Platform (ICESP www.icesp.it).
	Beyond the integrations described above we have further integrated the section "Acknowledgements" with the following text since, as described above, the final development and refinement of the methodology has been carried out within StarProBio project along with the project partner University of Bologna (PhD Diego Marazza).
	Added text The contents of the paper are part of the findings of the project STAR-ProBio. STAR-ProBio has received funding from the European Union's Horizon 2020 program research and innovation programme under grant agreement No. 727740

#### Metrics for quantifying the circularity of bioplastics: the 1 case of bio-based and biodegradable mulch films 2 3 Francesco Razza<sup>a</sup>, Cristiana Briani<sup>b</sup>, Tony Breton<sup>c</sup>, Diego Marazza<sup>d</sup> 4 5 6 7 8 9 10 11 <sup>a</sup> Novamont S.p.A. - Ecology of Product and Environmental Communication, Piazz.le Donegani 4, 05100 Terni, Italy <sup>b</sup> CIRSA Centro Interdipartimentale di Ricerca per le Scienze Ambientali, Via S. Alberto 163, 48123 Ravenna, Italy <sup>c</sup>Novamont S.p.A. – Via Fauser 8, 28100 Novara, Italy 12 13 <sup>d</sup>Department of Physics, University of Bologna, Viale B. Pichat 6/2, 40127 Bologna, Italy 14 15 Abstract The concept of circularity and its quantification through the Material Circularity Indicator 16 (MCI) is well established for traditional plastic products. In this paper a methodological 17 18 approach for calculating the circularity of bio-based and biodegradable (BB) products is 19 proposed and applied to BB mulch films. BB products are different from traditional 20 products in as much as they are sourced and regenerated (recycled) not through technical cycles but the biological loop. The suggested method is an adaptation of the MCI where 21 22 two major changes were made: (i) the mass of the bio-based component corresponds to the 23 recycled material in input and (ii) the mass of the bio-based component leaving the system through composting or biodegradation in soil is accounted as recycled. The modified MCI 24 25 supports the eEco-design of innovative BB products and allows for the comparison of their circularity taking into account the biological source and the expected end of life 26 process such as biodegradation. To demonstrate the adaptation, the method has been 27 28 applied to BB mulch films. Results showed that the MCI of a biodegradable mulch film, 29 characterized by an average bio-based feedstock content of 30% is 0.37 $\pm$ 0.04 in a 0-1 scale. For BB mulch film, the amount of bio-based feedstock is the most sensitive factor 30 31 and controls linearly the value of the MCI.

32

*Keywords:* circularity indicators, circular economy, bioplastics, biodegradable
 mulch film, bio-based product, biodegradation

35

36	1 Introduction
37	1.1 The case study of mulch films
38	1.2 Goal of the paper
39	2 Materials and Methods
40	2.1 MCI accounting according to the EMF methodology
41	2.2 MCI accounting for bio-based and biodegradable (BB) products <u>11</u> 44
42	2.3 MCI calculation for mulch films: scope, inventory and assumptions 14
43	2.4 Sensitivity analysis
44	3 Results 1'
45	3.1 Sensitivity analysis
46	4 Discussion
47	5 Conclusions

#### Abbreviations

Biodegradable and bio-based
Circular Economy
Dry matter
Ellen MacArthur Foundation
Life Cycle Assessment
Low-Density Poly-Ethylene
Material Circularity Indicator
Non-Restorative Flows
Polybutylene adipate terephthalate
Poly-Ethylene
Polylactic acid
Poly hydroxy butyrate

#### 51 **1 Introduction**

52 To overcome today's unsustainable model of 'take-make-dispose' and its related risks 53 such as hikes in raw material prices, pressures on the environment, shortage of global 54 resources and waste sinks, a circular approach needs to be applied. It is a new regenerative 55 economic view, based on a balance between economy, environment and society, a total 56 resource efficiency and a Zero Emission Strategy that aims to maximize products value 57 with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with 58 structural changes in environmental legislation, new logistics, technologies and sharing 59 schemes, the Circular Economy (CE) approach which is regenerative by design, aims at 60 closing materials loops, *i.e.* at reducing virgin materials input and waste output.

61 In December 2015, the European Commission developed an Action Plan for Circular 62 Economy (European Commission, 2015), where plastic was considered a priority to be 63 tackled. In January 2018, an EU Plastic Strategy (European Commission, 2018) was 64 adopted, in order to react to the increasing environmental problems concerning plastic 65 production, consumption, use and disposal along the same lines of the CE approach. Two fundamental steps to increase the circularity of different plastic products are (i) the 66 67 abandonment of fossil fuels, i.e. currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development 68 69 of easily recyclable products which are recycled. Today, in EU the share of plastics collected for recycling is 30% while the use of recycled plastics is just 6% (European 70 71 Commission, 2018).

72 Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for 73 Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and 74 principles. This is true as long as the supply of renewable raw materials, generally from 75 agriculture, is based on a sustainable approach and the conversion processes along the

76 supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA) 77 perspective (EPLCA - European Platform on LCA). While traditional plastics can be 78 mechanically recycled or incinerated with energy recovery, BB plastic products offer new 79 recycling routes in waste management, due to their biodegradability. Organic recycling 80 (through composting or anaerobic digestion) or in the case of specific applications such as agricultural mulch films, biodegradation in the environment, offer additional recovery 81 options resulting in less wastes and less contamination of soil by plastic residues (Razza et 82 83 al., 2012; Lange, B., 2016). An extensive literature review about the potentialities and 84 benefits of renewable and compostable bioplastics, encompassing market perspective, 85 applications, economic effects etc. can be found here: (BBIA; European Bioplastics). 86 Nevertheless, the research and development of innovative products, such as the BB

87 products, implies the development of methodologies and metrics capable of measuring 88 their circularity. Without this it is not possible to achieve measurable results and 89 improving actions, as well as provide unequivocal references for comparisons of products 90 of the same type/category. In 2015 the Material Circularity Indicator (MCI) was 91 developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify the regeneration of a product's material flow and is considered one of the few, among 92 93 sixteen CE indexes suiting a micro-scale assessment of circularity at product or company 94 level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled 95 materials. Furthermore, recovery and recycling through the biological cycle offered by industrial composting, anaerobic digestion or biodegradation in natural environments are 96 97 not considered as end of life options. In order to apply the MCI system to BB plastic 98 products, the development of an enhanced methodology is necessary.

99 The approach proposed by the authors allows to quantify the circularity of BB plastic 100 products and to make comparisons with equivalent traditional plastic products. To
demonstrate the applicability of the proposed method a computational example for mulch
film products is provided. In so doing so, the paper aims at contributing to the Eco-design
of these innovative products.

## 104 1.1 The case study of mulch films

105 Plastic mulch films represent an important agronomical technique well established for the 106 production of many crops thanks to numerous agronomical advantages such as: increased 107 yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and 108 reduced use of pesticides; early crop production and reduced soil moisture loss 109 (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has 110 increased year-by-year, reaching a current global market estimated at 1.4 millions of 111 tonnes, mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017), and 112 covering 80,000 km<sup>2</sup> of agricultural surface (0.6% of the global arable land). The mulch 113 film market in Europe is estimated by Agriculture Plastic & Environment and by the 114 European Bioplastic Associations at 76-80 kt. The most used raw material is Poly-115 Ethylene (PE) in its different forms, due to its processability, chemical resistance, high 116 durability and flexibility (Kasirajan and Ngouajio, 2012; Plasticulture, 2016 and 2018; 117 Shen, M. et al., 2019; Wen, X. et al., 2018).

118 Despite these benefits, manifold environmental and agronomic problems have been 119 pointed out. After its useful life - which in general does not exceed 1 to 3 months - the 120 mulch film has to be removed and properly disposed of, a time-consuming (about 16 hours 121 per hectare) and costly procedure (Scaringelli, M., 2016; Briassoulis, D., 2013). The 122 recovered film is usually heavily contaminated with soil and organic residues, making 123 mechanical recycling technically difficult and not a cost-efficient solution (Briassoulis et 124 al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most common end of life of 125 collected films in Europe is still landfilling (about 50%), followed by energy recovering

126 and finally mechanical recycling (Le Moine, 2014). Recent Chinese prohibition (January 127 2018) to import different types of wastes is heavily impacting the European agricultural 128 plastic waste management, highlighting the difficulty in properly recycling this type of 129 plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but 130 disposed of by burning in the field or by uncontrolled landfilling or left directly in the 131 (agricultural) soils, causing serious environmental concerns. An example is the "White 132 pollution" phenomena described in the Xinjiang Autonomous Region (China), in which 133 the residual plastic film can reach 200 kg/ha in the top soil with detrimental effects on 134 soils' quality, health and fertility (Liu, He, & Yan, 2014; Gao et al., 2019; Steinmetz et 135 al., 2016).

136 As a reaction, there has been significant research into novel materials especially related to 137 biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation 138 in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). 139 The term "bio-mulch film" brings together several types of both bio-based and fossil oil-140 based biodegradable polymers and blends of them, such as polylactic acid (PLA), 141 polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or 142 copolymers. They biodegrade when exposed to bioactive environments such as soil and 143 compost (Kasirajan et al., 2012) which means that they can be left in situ to be fully 144 biodegraded after being used. Clearly the biodegradation rate of biodegradable bioplastics 145 is influenced by the environmental conditions such as the types of available bacteria, fungi 146 thus specific enzymes namely native microflora (Pico, Y. et al., 2019). However their 147 intrinsic biodegradability allow the complete biodegradation with times similar to natural 148 polymers such as cellulose used as reference by the relevant standards and certification 149 schemes.

150 The EN 17033:2018 is a new European Norm (standard) concerning "Plastics -151 Biodegradable mulch films for use in agriculture and horticulture - Requirements and test 152 methods", which sets the necessary tests and limits to define biodegradability, 153 performances and environmental impacts of BB much films. The material is considered 154 completely biodegradable if it achieves a complete biodegradation (absolute or relative to the reference material) in a test period no longer than 24 months (mineralization into 155 156 CO<sub>2</sub>). Additionally, a control of constituents (such as metals) and eco-toxicity testing 157 (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test 158 with soil microorganisms) were required. A certified mulch film guarantees that the 159 product will completely biodegrade in the soil without adversely impacting on the 160 environment.

## 161 1.2 Goal of the paper

162 The goal of the paper is to provide a general and common metric to measure the 163 circularity of a bio-based and biodegradable (BB) product and to apply the methodology at 164 product level to a category of products, namely bio-based and biodegradable mulch films.

## 165 2 Materials and Methods

## 166 2.1 MCI accounting according to the EMF methodology

167 The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation 168 (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number 169 that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production 170 provides for the exclusive use of virgin raw materials that turn into waste at the end of the 171 use phase of the product. Vice-versa, pure circularity includes the use of recycled 172 materials and does not produce wastes (regenerative streams). Circularity can be achieved 173 in different ways: as for the purpose of this paper, only recycling will be considered since

174	reuse is not an option for thin biodegradable mulch films. Since the method considers only
175	mass flows, the recycling corresponds to the recovery of materials for the original purpose
176	or for other purposes and excludes energy recovery, considered as a loss of materials equal
177	to landfill disposal. The materials recovered feed back into the process as recycled
178	feedstock.
179	The MCI methodology differentiates 'technical cycles' from 'biological cycles',
180	modelling only the former. The first contains products and materials re-entering into the
181	system (market) with the highest possible qualities and for as long as possible (thanks to
182	reuse, repair, refurbishment and recycling) and the latter includes biological materials used

183 in cascade until their restoration into the biosphere and the re-constitution of natural184 resources.

The material flows associated to the production of a generic technical cycle from nonrenewable sources are summarized in <u>Figure 1Figure 1</u>. The dashed lines indicate that recycled feedstock does not have to be sourced from the same product but can be acquired on the market. With reference to <u>Figure 1Figure 1</u>, the list of the parameters used in the EMF methodology is reported in <u>Table 1Table 1</u>, while the equations relevant for the analysis carried out in this paper are described in the following sections (<u>Table 2Table 2</u>,

191 Chapter 2.2).

Formatted: Font: Not Italic
Formatted: Font: Not Italic
Formatted: Font: Not Italic

Formatted: Font: Not Italic





product (modified from Ellen MacArthur Foundation & Granta Design, 2015).

Table 1: Parameters and relative definitions used in the EMF methodology.

Parameter	Definition
М	Total mass of the product
$F_R$	Fraction of mass of a product's feedstock from recycled sources
$F_U$	Fraction of mass of a product's feedstock from reused sources
V	Mass of virgin feedstock used in a product
$C_R$	Fraction of mass of a product being collected to go into a recycling process
$C_U$	Fraction of mass of a product going into component reuse
$E_C$	Efficiency of the recycling process used for the portion collected for recycling
$E_F$	Efficiency of the recycling process used to produce recycled feedstock for a
	product

W	Total mass of unrecoverable waste associated with a product
W <sub>0</sub>	Mass of unrecoverable waste (landfill, waste to energy and any other type of
	process where the materials are no longer recoverable)
W <sub>C</sub>	Mass of unrecoverable waste generated in the process of recycling parts of a
	product (after use)
$W_F$	Mass of unrecoverable waste generated when producing recycled feedstock for
	a product
X	Utility of a product, calculated as $X = (L/L_{av})(U/U_{av})$
L	Actual average lifetime of a product
$L_{av}$	Actual average lifetime of an industry-average product of the same type
U	Actual average number of functional units achieved during the use phase of a
	product
Uav	Actual average number of functional units achieved during the use phase of an
	industry-average product of the same type

197 The Material Circularity Indicator is determined as follows:

where *LFI* is the Linear Flow Index measuring the flows of virgin materials andunrecoverable wastes associated to the examined product.

A function of the utility, -, is used to correct the *LFI*. The function *F* is chosen in such a way that improvements of the utility of a product (e.g., by using it longer) have the same impact on its MCI as a reuse of components, leading to the same amount of reduction of virgin material use and unrecoverable waste. Setting a = 0.9, MCI takes, by convention, the value 0.1 for a fully linear product (*i.e.*, *LFI* = 1) whose utility equals the industry average (*i.e.*, X = 1). This leaves some margin to distinguish between processes with a high linearity but different utilities.

207	2.2 MCI accounting for bio-based and biodegradable (BB) products	
208	To apply the EMF methodology to BB products, formulas and flows (Figure 1Figure 1	 Formatted: Font: Not Italic
209	and <u>Figure 2</u> Figure 2) are adapted as it follows:	Formatted: Font: Not Italic
210	1. The fraction of the recycled feedstock, $F_R$ , corresponds to the share of the bio-	
211	based feedstock content in the final BB product, $F_{R(i)}$ . It is the ratio of the d.m.	
212	amount of bio-based feedstock per d.m. amount of the total mass of BB	
213	product (EN 16785-2:2016).	
214	2. The fraction of restorative mass going into a recycling process, $C_R$ , corresponds	
215	to the share of bio-based feedstock content in the BB product biologically	
216	recovered (e.g. through composting) or biodegraded in the natural	
217	environment, as it happens for specific applications (e.g. biodegradable mulch	
218	film, etc.). It is the ratio of the d.m. amount of bio-based feedstock per d.m.	
219	amount of the total mass of BB product that is biologically recycled.	
220	The modified scheme is shown in Figure 2 Figure 2. Table 2 lists the formulas as	Formatted: Font: Not Italic
221	adapted to BB products.	Formatted: Font: Not Italic
222	Table 2: List of formulas as developed by EMF methodology compared to the	
223	proposed adaptation to BB products.	

EMF methodology

Adaptation to BB products

224	
225	The mass of fossil-based feedstock which may be contained in BB products ( $V$ ) is
226	obtained as a difference of the total mass $(M)$ minus the bio-based fraction; in this case the
227	$F_R$ in the EMF methodology corresponds to the sum of the fractions of all the bio-
228	based feedstock/s used in manufacturing the BB product. Therefore, is the
229	total bio-based feedstock mass in the product. In single-use products, such as mulch films,
230	reuse is not considered for BB products, so that $F_U = C_U = 0$ .
231	$W_F$ is the total amount of unrecoverable waste associated to the production of bio-based
232	feedstock used to produce BB products (i.e. the amount of uncoverable waste per unit of
233	BB product). Bio-based feedstocks such as starch, PLA, PHB etc. generate non-restorative
234	flows which can be quantified. Such unrecoverable waste correspond to $R_{(i)}$ , the specific
235	amount of waste generated within cradle-to-gate boundaries per unit of bio-based
236	feedstock going into manufacturing and it is estimated through LCA studies. Thus all

feedstock going into manufacturing, and it is estimated through LCA studies. Thus all inputs from growth and harvesting phases and the related wastes generated by fertilisers and pesticides are here accounted.  $R_{(i)}$  can be easily found in specific literature or life cycle inventories (LCI) present in LCA databases. In the calculation of  $W_F$ , also the efficiency of manufacturing process of BB products  $E_P$  is considered, as the ratio of the

- 241 overall bio-based feedstock content in the final BB product to the bio-based feedstock in
- 242 input to the manufacturing process.
- The material flows associated to the production of a generic BB product are summarized 243



Formatted: Font: Not Italic



246 Figure 2: Description of material flows adaptation to BB products; in this paper, the reuse flow is out of scope ( $C_U = F_U = 0$ ). 247

248 The biodegradation of bio-based feedstock does not imply the generation of waste  $W_C$  as it 249 occurs in a standard mechanical recycling process. This implies that  $C_R$  and  $E_C$  (i.e. the 250 efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to 251 biological treatment (composting) or biodegraded in a natural environment, is fully 252 transformed in its chemical elements (C, H and O mainly) derived from the decomposition of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et 253

254	al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites,	
255	2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the	
256	environment and are then available in the respective biogeochemical cycles. The	
257	(biodegradable) fossil portion behaves as well; consequently, $W_C = 0$ .	
258	Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular	
259	feedstock, since it derives from carbon stored for millions of years and extracted by man,	
260	not being part of the active and fast biogeochemical carbon cycle. This is accounted in the	
261	quantification of $W_0$ , the mass of unrecoverable waste from use ( <i>i.e.</i> the linear stream	
262	going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total	
263	amount of fossil-based feedstock.	
264	Since $W_F$ and $W_C$ are associated to complete different processes and $W_C$ is always equal	
265	zero, the double counting issue does not occur and the quantification of $W$ and $LFI$ is	
200	modified as reported in Table 2 <del>Table 2</del>	Formatted: Font: Not Italic
266	mounted as reported in <u>resort 2</u> ratio 2.	
266	2.3 MCI calculation for mulch films: scope, inventory and assumptions	
266 267 268	<ul> <li><i>ACL calculation for mulch films: scope, inventory and assumptions</i></li> <li>The new formulas reported in <u>Table 2</u> were applied to a single use product namely</li> </ul>	Formatted: Font: Not Italic
266 267 268 269	<ul> <li><i>ACL calculation for mulch films: scope, inventory and assumptions</i></li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of BB</li> </ul>	Formatted: Font: Not Italic
266 267 268 269 270	<ul> <li><i>ACI calculation for mulch films: scope, inventory and assumptions</i></li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any</li> </ul>	Formatted: Font: Not Italic
<ul> <li>266</li> <li>267</li> <li>268</li> <li>269</li> <li>270</li> <li>271</li> </ul>	<ul> <li><i>ACI calculation for mulch films: scope, inventory and assumptions</i></li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.</li> </ul>	Formatted: Font: Not Italic
<ul> <li>266</li> <li>267</li> <li>268</li> <li>269</li> <li>270</li> <li>271</li> <li>272</li> </ul>	<ul> <li><i>ACI calculation for mulch films: scope, inventory and assumptions</i></li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.</li> <li>In the global market, there are several branded BB mulch films (Moreno et al., 2017), both</li> </ul>	Formatted: Font: Not Italic
<ul> <li>266</li> <li>267</li> <li>268</li> <li>269</li> <li>270</li> <li>271</li> <li>272</li> <li>273</li> </ul>	<ul> <li><i>ACI calculation for mulch films: scope, inventory and assumptions</i></li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.</li> <li>In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily</li> </ul>	Formatted: Font: Not Italic
<ul> <li>266</li> <li>267</li> <li>268</li> <li>269</li> <li>270</li> <li>271</li> <li>272</li> <li>273</li> <li>274</li> </ul>	<ul> <li><i>2.3 MCI calculation for mulch films: scope, inventory and assumptions</i></li> <li>The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely</li> <li>a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1.</li> <li>In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e.</li> </ul>	Formatted: Font: Not Italic
<ul> <li>266</li> <li>267</li> <li>268</li> <li>269</li> <li>270</li> <li>271</li> <li>272</li> <li>273</li> <li>274</li> <li>275</li> </ul>	<b>2.3</b> <i>MCI calculation for mulch films: scope, inventory and assumptions</i> The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1. In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch, $F_{(S)}$ , and 7% of a bio-based additive, $F_{(BA)}$ ), while the rest was assumed to	Formatted: Font: Not Italic
266 267 268 269 270 271 272 273 274 275 276	<b>2.3 MCI calculation for mulch films:</b> scope, inventory and assumptions The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1. In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch, $F_{(S)}$ , and 7% of a bio-based additive, $F_{(BA)}$ ), while the rest was assumed to consist of fossil feedstock (	Formatted: Font: Not Italic Formatted: Font: Not Italic
266 267 268 269 270 271 272 273 274 275 276 277	<b>2.3</b> <i>MCI</i> calculation for mulch films: scope, inventory and assumptions The new formulas reported in <u>Table 2Table 2</u> were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the bio-based feedstock content and the process yield is close to 1. In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch, $F_{(S)}$ , and 7% of a bio-based additive, $F_{(BA)}$ ), while the rest was assumed to consist of fossil feedstock (	Formatted: Font: Not Italic Formatted: Font: Not Italic



Formatted: Font: Not Italic

Figure 3: Examples of hypothetical bio-based polymers; in this paper, the first option on the left (starch blend 30/70) has been chosen for carrying out the numerical MCI calculation (working hypothesis). The figure considers a 100%-efficiency in every phase of production, so that the residues are equal to zero; the same assumption is done in this paper. \*TPS (Thermoplastic starch), starch content 75%; \*\*Ratio TPS/Polymer; modified from Institute of Bioplastics and Biocomposites, 2018.

300

Table 3: Key features representative of the BB mulch films.

	BB mulch film
Material	30% bio-based feedstock (23% starch + 7% bio-
	based additive) + 70% fossil feedstock
Thickness (µm)	12
Density (g/cm <sup>3</sup> )	1.25
Weight (g/m <sup>2</sup> )	15.2
Functional unit	$6000 \text{ m}^2$ /ha (the actual mulched soil in a hectare
(the covering of the agricultural land)	is generally equal to the 60% of the total area;
	Malinconico, 2017)

303 In the calculation of MCI for the BB mulch film, the adapted formulas were used together 304 with assumptions. As stated before, BB mulch films are blends of bio-based and fossil 305 based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE 306 mulch film that has to be removed and disposed of, the BB mulch film is left in soil where 307 it undergoes an ultimate biodegradation (so that  $C_R = 1$ ) with no waste (so that  $E_C = 1$ ), in 308 respect of the specific standard EN 17033:2018. As a result of polymers' decomposition, 309 the derived (biogenic) C, H and O finally return into biosphere (atmosphere, 310 microorganism biomass, organic material pool) (OWS, 2018), and back into 311 biogeochemical cycles in a relatively short time ("Biogenic elements accounted as recycled" in Figure 2Figure 2), with the exception of humified compounds. Actually, also 312 313 C, H and O deriving from fossil-based sources undergo biodegradation (Zumstein, M.T., 314 2018) but they are not considered as a regenerative flow ("Waste from non-restorative flow" in *Figure 2Figure 2*) and their "wastes" are indeed calculated in  $W_0$ . 315 316 Applying a conservative approach,  $W_F$ , the waste generated by the production of each bio-

317 based feedstock, is quantified considering a "cradle to gate" LCA study. The estimated

318 solid wastes  $R_{(i)}$  for the presented case study are related to the production of starch (F<sub>(S)</sub>),

16

Formatted: Font: Not Italic

301

319	with an amount $R_{(S)}$ of 0.014 kg of waste per kg of renewable feedstock (source: personal	
320	communication A. Novelli), and to the production of the bio-based additive $(F_{(BA)})$ , with	
321	$R_{(BA)}$ equals to 0.025 kg waste/kg renewable feedstock (US-LCI database). As assumed in	Formatted: Font: Not Italic
322		
323	<u>Figure 3</u> , the production efficiency of BB product $E_P$ (how much bio-based	
324	feedstock is needed for every unit of BB product) is estimated equal to 1 and no	
325	unrecoverable wastes are generated by the process.	
326	In addition, an explorative sensitivity analysis has been performed regarding exclusively	
327	the amount of bio-based feedstock content of the BB mulch film, <i>i.e.</i> $(i.e., F_{(S)} +$	
328	$F_{(BA)}$ ), as shown in Figure 4Figure 4 (Chapter 3). Considering the characteristics of the	Formatted: Font: Not Italic
329	films (weight, g/m <sup>2</sup> , or thickness, $\mu$ m, and density, g/cm <sup>3</sup> ) and the relative functional unit	
330	(6000 m <sup>2</sup> /ha, <u>Table 3</u> Table 3), it is possible to calculate a mass, <i>M</i> , that is 90 kg/ha for the	Formatted: Font: Not Italic
331	BB one Once calculated the masses, the formulas reported in Table 2 Table 2 (Chapter	Formatted: Font: Bold, Not Italic
551	bb one. Once calculated the masses, the formulas reported in <u>radie 2</u> (Chapter	Formatted: Font: Not Italic
332	2.2) are applied. Results are shown in <u>Table 4</u> <u>Table 4</u> .	Formatted: Font: Not Italic
		Formatted: Font: Not Italic
333		
334	2.4 Sensitivity analysis	

335 A sensitivity analysis was conducted for BB mulch film to examine the effects of changing the main variables. Given a non-linear dependence of results on parameter 336 337 values, a Monte Carlo approach (see, e.g., Lloyd and Ries, 2008) has been adopted. The 338 model has been implemented using specifically written routines in the C++ programming 339 language. The model was run with 100,000 events for BB mulch film, where the value of 340 each parameter has been randomly chosen following a Gaussian distribution with a 341 standard deviation within a range of possible and realistic values (Table 5 Table 5 and Error! Reference source not found. Table 6; Figure 5Figure 5 and Figure 6Figure 6). 342

Formatted: Font: Not Bold, Not Italic

Formatted: Font: Not Italic Formatted: Font: Not Italic

343	3 Results	
344	Figure 4 Figure 4 shows how the value of the MCI varies according to the percentage	Formatted: Font: Not Italic
345	variation of the bio-based feedstock in the total mass of the product.	
346		
347	Table 4: Resulting parameters in the calculation of MCI for BB mulch film.	

Parameter BB mulch film



350

351 Figure 4: MCI as a function of the amount of bio-based feedstock/s in the BB 352 mulch film  $\Sigma F_{R(i)}$ , expressed as the percentage of all the bio-based feedstock/s of the mulch 353 film on dry mass basis (X-axis). The dots correspond to the three different hypothetical 354 bioplastic compositions of Figure 3.

## 356 3.1 Sensitivity analysis

- The results of the sensitivity analysis are presented in the followings <u>Table 5Table 5</u> and <u>Figure 5Figure 5</u> and <u>Figure 6Figure 6</u>. The accuracy band is a fraction of the average and corresponds to a probability of 95%. It has been chosen in order to be representative of the variability of the product category, the BB mulch films. The simulation can thus be regarded as a system composed by a high number of companies, each producing films with different characteristics, that are accounted for in the accuracy band. **Table 5:** Parameters used for the sensitivity analysis of the BB mulch film. (\*\*) The
- 364 Accuracy Band is defined as twice the standard deviation of the distribution.

19

Formatted: Font: Not Bold, Not Italic

Formatted: Font: Not Italic Formatted: Font: Not Italic

Variable name	Average	Accuracy Band (**)	Unit
М	1000.00	0%	kg
F(S)/F(BA)	3.29	10%	fraction
$\mathbf{F}_{(S)} + \mathbf{F}_{(BA)}$	0.30	30%	fraction
$\mathbf{F}_{\mathbf{U}}$	0.00	0%	fraction
C <sub>U</sub>	0.00	0%	fraction
R <sub>(S)</sub>	0.014	100%	fraction
R <sub>(BA)</sub>	0.025	100%	fraction
E <sub>C</sub>	1	0%	fraction
E <sub>P</sub>	0.95	10%	fraction
C <sub>R</sub>	1.00	0%	fraction





371 Figure 6: The most sensitive and relevant parameters in the calculation of the

372 MCI of the BB mulch films.

## 373 4 Discussion

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

(1) fossil-based feedstocks or component materials embodied in the BB products whateveris the final disposal (even biological recycling) shall be considered as non-restorative;

(2) bio-based component materials embodied in the BB product that go to biological
recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be
considered restorative as long as they flow through the biosphere safely, without any harm
to the environment (e.g. no toxicity effects).

(3) bio-based component materials embodied in the BB product that go to incineration andlandfill shall be considered as non-restorative;

387 The justification of these rules is described in the following.

388 Fossil-based component materials in the product derive from deposits where they 389 remained stocked for a geological time scale. Once the product is mineralised, its fossil-390 based portion will be accounted as non-regenerative and therefore linear, due to its origin 391 (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological 392 cycles, like CO<sub>2</sub> in the atmosphere and other streams, since both fossil-based and bio-393 based component materials will physically and chemically behave the same, once 394 biodegraded. However, the source of the bio-based carbon was circular before its use 395 (concept of "carbon neutrality", equilibrium between the biogenic carbon released and the 396 carbon absorbed by plants) and will maintain its circularity provided that the carbon is 397 released into the atmosphere at the same rate. The reason has its origin in the EMF general

398 provisions stating that "biologically sourced materials can only be considered part of a 399 Circular Economy if materials are not used faster than they can be restored naturally" 400 (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the 401 bio-based components are still considered linear, maintaining consistency with EMF 402 principles. Basically, a complete circularity for a BB product is satisfied when its 403 renewable components are 100% bio-based and they go 100% to biological recycling or 404 biodegraded in the environment (for specific application like mulch film).

405 As for provision (3), a material health rule has its origin in maniy fold normative 406 definitions of the CE. In addition, the EMF definition of biological cycles is that of non-407 toxic materials which are restored into the biosphere and the CE is defined as such if it can 408 "eliminate the use of toxic chemicals". The need of a safety clause has been reviewed 409 under many aspects by Verberne (2016) and can be put as a postulate of the restoration 410 principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the 411 REACH Regulation (EC 1907/2006). In the specific case, the material complies with the 412 standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism 413 groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important 414 ecological processes maintaining soil functions, c) all relevant exposure pathways as soil 415 pore water, soil pore air and soil material.

416 A comprehensive approach for MCI calculation should also include non-restorative flows 417 generated at upstream level like biomass growth, in the specific case corn, and biomass 418 conversion processes like starch extraction and refining. Specifically these non-restorative 419 flows correspond to the overall non-recyclable wastes associated to the bio-based 420 feedstock supply thus non-recyclable waste from fertilizer and pesticide production, non-421 recyclable scraps from conversion processes, etc. In this study such flows of non-422 restorative waste coming from upstream manufacturing operations were included for the

423 bio-based feedstocks ( $R_{(i)}$ ) used in manufacturing the BB mulch film applying "cradle to 424 gate" LCA methodology. However, we observed that the inclusion of upstream 425 unrecoverable waste does not significantly influence the MCI results in the chosen case 426 study, since the respective amounts are small. The specific unrecoverable waste for starch 427 and bio-based additive (*i.e.* kg of waste/kg of bio-based feedstock) were estimated at 428 0.014 and 0.025, respectively.

Formatted: Font: Italic

429

The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation y = 0.89x + 0.1, where y is the MCI and x is the bio-based feedstock content, therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input is decisive.

435 Apart from the specific application analysed in this paper, the proposed MCI method can
436 be easily applied and calculated for any kind of BB product as long as the following
437 information are available:

The bio-based feedstock content, determined according to the standard EN 167852:2016, if the composition is known, or directly provided by the BB product manufacturer.
The Eend of Life scenario of the studied BB product (real or hypothetical).
The amount of un-recoverable waste associated to the production of bio-based

feedstock contained in the BB product. They can be derived from LCA databases or otherspecific sources.

## 444 5 Conclusions

Bioplastic market is steadily increasing. The value proposition of bio-based andbiodegradable products is linked to:

447 1. the use of renewable feedstock (like starch and its derivates) instead of fossil oil or448 natural gas;

449 2. the waste recovery through biological recycling, thanks to their ability to450 biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

451 The Material Circularity Indicator (MCI), developed by the EMF, is a metric for quantifying "how much" a product is circular (MCI = 0, fully-linear product; MCI = 1, 452 453 completely circular product) thus it represents a valuable tool for product eco-design 454 purposes. However, it focuses solely on technical materials, mechanically recycled or 455 reused, leaving out bio-based feedstocks and related biological treatments such as 456 composting. Without common metrics it is not possible to pursue concrete actions, to 457 achieve measurable results and to provide unequivocal references for all products. This 458 research work aims at filling this gap through the development of a methodology coherent 459 with EMF MCI methodology but able to catch the specificities of bio-based and 460 biodegradable products and provide metrics for those innovative products. Direct uses are: 461 (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI 462 of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation:  $MCI_{(BB mulch})$ film) = 0.89\*bio-based feedstock + 0.1.

The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles like the use of renewable materials and the reduction of the amount of not recoverable waste. MCI will support the development of innovative products just based on these two important characteristics specific for each BB

472	product/application and end of life scenario. Bioeconomy, thus also BB products, can
473	provide valuable insights in transforming the current (linear) economy in a more circular
474	one, however, the way the biomass is produced, processed and BB products are produced
475	are fundamental aspects to be properly assessed and monitored. This can be done using
476	specific methodologies like LCA. Within this context the proposed MCI has to be seen as
477	a complementary (quantitative) tool for further qualifying the sustainability of BB
478	products and not as a substitute tool. Furthermore the MCI here proposed is meaningful
479	only if BB products meet health and safety material requirements according to the national
480	and European laws and standards. This is a postulate of the proposed methodology
481	especially for those BB products conceived to biodegrade in the environment like
482	biodegradable mulch film.
483	
484	
485	
486	Declaration of interest
487	The author declares that the research was conducted in the absence of any
488	commercial or financial relationships that could be construed as a potential conflict of
489	interest.
490	
491	Acknowledgements
492	The contents of the paper are part of the findings of the project STAR-ProBio.
493	STAR-ProBio has received funding from the European Union's Horizon 2020 program
494	research and innovation programme under grant agreement No. 727740. The authors
495	thanks prof. Andrea Contin for the fruitful discussion and contribution to the sensitivity
496	analysis, Francesco Degli Innocenti for providing valuable comments and feedback on the

497 topics addressed by the paper and Alessandra Novelli for the general support in the MCI498 elaboration.

499 500

# 501 Addendum

502	While this paper was undergoing peer review the authors became aware that the EMF
503	published an update of the MCI methodology (Ellen MacArthur Foundation & Granta
504	Design, 2019) including the extension of it to include the treatment of biological materials.
505	This update introduces new definitions and formulas. The authors believe that most of the
506	changes regarding accounting are in the direction here proposed and that this study can
507	contribute as an illustration on how the material circularity of a biological based material
508	can be addressed in a real case study. Furthermore the authors would like to highlight that
509	the proposed methodology started long before the EMF changes: specifically the original
510	idea dated back to 2017 and a beta version of it - not as it is now - was presented in the
511	middle of 2018 at the Italian Circular Economy Stakeholder Platform (ICESP
512	www.icesp.it).

513

## 514 **References**

- BASF, 2018. Biodegradable mulch film clarification of polymer fate in soil. CIPA Congress, Bordeaux/Arcachon, France, May 2018.
- Bio-Based and Biodegradable Industries Association. BBIA reports. https://bbia.org.uk/reports/ (accessed 28 November 2019)
- Briassoulis, D., Giannoulis, A., 2018. Evaluation of the functionality of bio-based plastic mulching films. Polym. Test. 67, 99–109. https://doi.org/10.1016/j.polymertesting.2018.02.019
- Briassoulis D., Hiskakis, M., Babou, E., 2013. Technical specifications for mechanical recycling of agricultural plastic waste. Waste Management, Volume 33, issue 6,
  - 27

pages 1516-1530, ISNN 0956-053X. <u>https://doi.org/10.1016/j.wasman.2013.03.004</u> De Lèpinau, P. and Arbenz, A., 2016. Economic and environmental impact of soil contamination in mulching film, Plasticulture, N° 136, 28-48.

- Ellen MacArthur Foundation & Granta Design, 2015. Circularity Indicators An approach to measure circularity Methodology. https://www.ellenmacarthurfoundation.org/assets/downloads/insight/Circularity-Indicators\_Methodology\_May2015.pdf.
- Ellen MacArthur Foundation & Granta Design, 2019. Circularity Indicators An approach to measure circularity Methodology.
- Ellen MacArthur Foundation, 2017. The New Plastic Economy: Rethinking the future of plastic & catalysing action.
- EN 16785-2:2016 Bio-based products Bio-based content Part 2: Determination of the bio-based content using the material balance method.
- EN 17033:2018 Plastics Biodegradable mulch films for use in agriculture and horticulture Requirements and test methods.
- EPLCA European Platform on LCA. https://eplca.jrc.ec.europa.eu/?page\_id=86
- Eubeler, J., Bernhanrd, M., Knepper, T., 2010. Environmental biodegradation of synthetic polymers II. Biodegradation of different polymer groups. Trends in Analytical Chemistry. 29, 1, 84-100
- European Commission, 2015. Closing the loop An EU action plan for the Circular Economy. COM(2015) 614 final. Brussels, 2.12.2015
- European Commission, 2018. A European Strategy for Plastics in a Circular Economy. COM(2018) 28 final. Brussels, 16.1.2018
- European Bioplastics. European Bioplastic publications. https://www.europeanbioplastics.org/news/publications/ (accessed 28 November 2019)
- Figuier, B., 2016. Plasticulture in Europe, Plasticulture, Nº 136, 20-28
- Gao, H., Yan, C., Liu, Q., Ding, W., Chen, B., Li, Z., 2019. Effects of plastic mulching and plastic residue on agricultural production: A meta-analysis. Sci. Total Environ. 651, 484–492. https://doi.org/10.1016/J.SCITOTENV.2018.09.105
- Ghisellini, P., Cialani, C., Ulgiati, S., 2016. A review on circular economy: The expected transition to a balanced interplay of environmental and economic systems. J. Clean. Prod. https://doi.org/10.1016/j.jclepro.2015.09.007

- Institute of Bioplastics and Biocomposites, 2018. Biopolymers Facts and Statistics. Hochschule Hannover, University of Applied Sciences and Arts. Edition 5, ISSN 2510-3431.
- Joos, F., Roth, R., Fuglestvedt, J. S., Peters, G. P., Enting, I. G., Bloh, W. V., ... and Friedrich, T., 2013. Carbon dioxide and climate impulse response functions for the computation of greenhouse gas metrics: a multi-model analysis. Atmospheric Chemistry and Physics, 13(5), 2793-2825.
- Kasirajan, S., Ngouajio, M., 2012. Polyethylene and biodegradable mulches for agricultural applications: a review. Agron. Sustain. Dev. 32, 501–529. https://doi.org/10.1007/s13593-011-0068-3
- Lange, B.K., 2016. Revision of the fertilisers regulation benefits of biodegradable mulch films. European Bioplastics. http://www.europarl.europa.eu/cmsdata/108931/Kristy%20Barbara%20Lange%20E UBP%20PPT2.pdf (accessed 28 November 2019)
- Le Moine, B., 2014. Agri-plastics waste management: a voluntary commitment from the industry. Presented at: Agricultural Film 2014 International Conference on silage, mulch, greenhouse and tunnel films used in agriculture (15-17 September, Barcelona, Spain).
- Liu, E. K., He, W. Q., & Yan, C. R., 2014. 'White revolution' to 'white pollution' agricultural plastic film mulch in China. *Environmental Research Letters*, 9(9), 091001.
- Lloyd, S. M., & Ries, R., 2007. Characterizing, propagating, and analyzing uncertainty in life cycle assessment: A survey of quantitative approaches. *Journal of Industrial Ecology*, 11(1), 161-179.
- Lonca, G., Muggéo, R., Tétreault-Imbeault, H., Bernard, S., & Margni, M., 2018. A Bidimensional Assessment to Measure the Performance of Circular Economy: A Case Study of Tires End-of-Life Management. In *Designing Sustainable Technologies, Products and Policies* (pp. 33-42). Springer, Cham.
- Malinconico, M., 2017. Soil Degradable Bioplastics for a Sustainable Modern Agriculture. Green Chemistry and Sustainable Technology. Springer.
- Marten, E., Muller, R., and Deckwer W., 2003. Studies on the enzymatic hydrolysis of polyesters I. Low molecular mass model esters and aliphatic polyesters. Polymer Degradation and Stability, 80, 3, 485-501.
- Moreno, M. M., González-Mora, S., Villena, J., Campos, J. A., & Moreno, C., 2017. Deterioration pattern of six biodegradable, potentially low-environmental impact mulches in field conditions. *Journal of environmental management*, 200, 490-501.

- Mormile, P., Stahl, N., Malinconico, M., 2017. The World of Plasticulture, in: Malinconico, M. (Ed.), Soil Degradable Bioplastics for a Sustainable Modern Agriculture. pp. 1–21. https://doi.org/10.1007/978-3-662-54130-2\_1
- OWS, 2018. Accumulation of (bio)degradable plastics in soil. CIPA Congress 2018, Archacon, May 29.
- Pico Y., Barcelò, D., 2019. Analysis and prevention of microplastics pollution in water: current perspectives and future directions. *ACS Omega*, 4, 6709-6719.
- Plasticulture catalogues, 2018. http://plasticulture.qualif.e-catalogues.info (accessed 28 November 2019)
- Razza, F., Degli Innocenti, F., 2012. Bioplastics from renewable resources: the benefits of biodegradability. *Asia-Pacific Journal of Chemical Engineering*, 7 (Suppl. 3): S301– S309. https://doi.org/10.1002/apj.1648.
- Scaringelli, M., Giannoccaro, G., prosperi, M., Lopolito, A., 2016. Adoption of biodegradable mulching films in agriculture: is there a negative prejudice towards materials derived from organic waste? *Italian Journal of agronomy*, 11:92.
- Shen M., Zhang, Y., Zhu, Y., Song, B., Zeng, G., Hu, D., Wen, Y., Ren, X., 2019. Recent advances in toxicological research of nanoplastics in the environment: A review. *Environmental Pollution*, 252: 511-521. https://doi.org/10.1016/j.envpol.2019.05.102
- Steinmetz, Z., Wollmann, C., Schaefer, M., Buchmann, C., David, J., Tröger, J., Muñoz, K., Frör, O., Schaumann, G.E., 2016. Plastic mulching in agriculture. Trading shortterm agronomic benefits for long-term soil degradation? Sci. Total Environ. 550, 690–705. https://doi.org/10.1016/J.SCITOTENV.2016.01.153
- Tamma, P., 2018. China's trash ban forces Europe to confront its waste problem, Politico, 2/21/2018.
- Touchaleaume, F., Martin-Closas, L., Angellier-Coussy, H., Chevillard, A., Cesar, G., Gontard, N., Gastaldi, E., 2016. Performance and environmental impact of biodegradable polymers as agricultural mulching films. Chemosphere 144, 433–439. https://doi.org/10.1016/j.chemosphere.2015.09.006
- US-LCI database. "Polylactide biopolymer resin at plant kg/RNA". https://www.nrel.gov/lci/ (accessed 9 December 2019)
- Verberne, J.J.H., 2016. Building circularity indicators. Eindhoven University of Technology.
- Wen, X., Du, C., Xu, P., zeng, G., Huang, D., Yin, L., Yin, Q., Hu, L., Wan, J., Zhang, J., Tan, S., Deng, R., 2018. Microplastic pollution in surface sediments of urban water areas in Changsha, China: Abundance, composition, surface textures. *Marine Pollution Bulletin*, 136: 414-423. https://doi.org/10.1016/j.marpolbul.2018.09.043.

Field Code Changed Formatted: English (United Kingdom)

Formatted: English (United Kingdom)

- Witt, U., Einig, T., Yamamoto, M., Kleeberg, I., Deckwer, W., Muller, R., 2001. Biodegradation of aliphatic-aromatic copolyesters: evaluation of the final biodegradability and ecotoxicological impact of degradation intermediates. Chemosphere. 44, 289-299.
- Zumstein, M., Schintlmeister, A., Nelson, T., Baumgartner, R., Wagner, M., Sander, M., McNeill, K., Woebken, D., Kohler, H., 2018. Biodegradation of synthetic polymers in soils: Tracking carbon into CO<sub>2</sub> and microbial biomass. Science Advances, 4, 7.

# Metrics for quantifying the circularity of bioplastics: the case of bio-based and biodegradable mulch films

Francesco Razza<sup>a</sup>, Cristiana Briani<sup>b</sup>, Tony Breton<sup>c</sup>, Diego Marazza<sup>d</sup>

<sup>a</sup> Novamont S.p.A. - Ecology of Product and Environmental Communication, Piazz.le Donegani 4, 05100 Terni, Italy

<sup>b</sup> CIRSA Centro Interdipartimentale di Ricerca per le Scienze Ambientali, Via S. Alberto 163, 48123 Ravenna, Italy

<sup>c</sup> Novamont S.p.A. – Via Fauser 8, 28100 Novara, Italy

<sup>d</sup>Department of Physics, University of Bologna, Viale B. Pichat 6/2, 40127 Bologna, Italy

# 15 Abstract

3

4

11

12 13

14

16 The concept of circularity and its quantification through the Material Circularity Indicator 17 (MCI) is well established for traditional plastic products. In this paper a methodological approach for calculating the circularity of bio-based and biodegradable (BB) products is 18 19 proposed and applied to BB mulch films. BB products are different from traditional 20 products in as much as they are sourced and regenerated (recycled) not through technical 21 cycles but the biological loop. The suggested method is an adaptation of the MCI where 22 two major changes were made: (i) the mass of the bio-based component corresponds to the 23 recycled material in input and (ii) the mass of the bio-based component leaving the system 24 through composting or biodegradation in soil is accounted as recycled. The modified MCI 25 supports the eco-design of innovative BB products and allows for the comparison of their 26 circularity taking into account the biological source and the expected end of life process 27 such as biodegradation. To demonstrate the adaptation, the method has been applied to BB 28 mulch films. Results showed that the MCI of a biodegradable mulch film, characterized by 29 an average bio-based feedstock content of 30% is  $0.37 \pm 0.04$  in a 0-1 scale. For BB mulch 30 film, the amount of bio-based feedstock is the most sensitive factor and controls linearly 31 the value of the MCI.

32

*Keywords:* circularity indicators, circular economy, bioplastics, biodegradable
 mulch film, bio-based product, biodegradation

36	1 Introduction
37	1.1 The case study of mulch films
38	1.2 Goal of the paper
39	2 Materials and Methods
40	2.1 MCI accounting according to the EMF methodology
41	2.2 MCI accounting for bio-based and biodegradable (BB) products
42	2.3 MCI calculation for mulch films: scope, inventory and assumptions 14
43	2.4 Sensitivity analysis
44	3 Results 17
45	3.1 Sensitivity analysis
46	4 Discussion
47	5 Conclusions
48 49	

Abbreviations	
BB	Biodegradable and bio-based
CE	Circular Economy
d.m.	Dry matter
EMF	Ellen MacArthur Foundation
LCA	Life Cycle Assessment
LDPE	Low-Density Poly-Ethylene
MCI	Material Circularity Indicator
NRF	Non-Restorative Flows
PBAT	Polybutylene adipate terephthalate
PE	Poly-Ethylene
PLA	Polylactic acid
PHB	Poly hydroxy butyrate

## 51 **1 Introduction**

52 To overcome today's unsustainable model of 'take-make-dispose' and its related risks 53 such as hikes in raw material prices, pressures on the environment, shortage of global 54 resources and waste sinks, a circular approach needs to be applied. It is a new regenerative 55 economic view, based on a balance between economy, environment and society, a total 56 resource efficiency and a Zero Emission Strategy that aims to maximize products value 57 with zero, or minimal, environmental impact (Ghisellini et al., 2016) . Together with 58 structural changes in environmental legislation, new logistics, technologies and sharing 59 schemes, the Circular Economy (CE) approach which is regenerative by design, aims at 60 closing materials loops, *i.e.* at reducing virgin materials input and waste output.

61 In December 2015, the European Commission developed an Action Plan for Circular 62 Economy (European Commission, 2015), where plastic was considered a priority to be 63 tackled. In January 2018, an EU Plastic Strategy (European Commission, 2018) was 64 adopted, in order to react to the increasing environmental problems concerning plastic 65 production, consumption, use and disposal along the same lines of the CE approach. Two 66 fundamental steps to increase the circularity of different plastic products are (i) the 67 abandonment of fossil fuels, *i.e.* currently 90% of the plastic is produced by virgin petroleum-based feedstock (Ellen MacArthur Foundation, 2017), and (ii) the development 68 69 of easily recyclable products which are recycled. Today, in EU the share of plastics 70 collected for recycling is 30% while the use of recycled plastics is just 6% (European 71 Commission, 2018).

Biodegradable and bio-based (BB) plastics are spreading across markets (Institute for Bioplastics and Biocomposites, 2018) as a valid contribution to meet CE aims and principles. This is true as long as the supply of renewable raw materials, generally from agriculture, is based on a sustainable approach and the conversion processes along the

supply chain are efficient and highly integrated in a Life Cycle Assessment (LCA) 76 77 perspective (EPLCA - European Platform on LCA). While traditional plastics can be 78 mechanically recycled or incinerated with energy recovery, BB plastic products offer new 79 recycling routes in waste management, due to their biodegradability. Organic recycling 80 (through composting or anaerobic digestion) or in the case of specific applications such as 81 agricultural mulch films, biodegradation in the environment, offer additional recovery 82 options resulting in less wastes and less contamination of soil by plastic residues (Razza et 83 al., 2012; Lange, B., 2016). An extensive literature review about the potentialities and 84 benefits of renewable and compostable bioplastics, encompassing market perspective, 85 applications, economic effects etc. can be found here: (BBIA; European Bioplastics).

86 Nevertheless, the research and development of innovative products, such as the BB 87 products, implies the development of methodologies and metrics capable of measuring 88 their circularity. Without this it is not possible to achieve measurable results and 89 improving actions, as well as provide unequivocal references for comparisons of products 90 of the same type/category. In 2015 the Material Circularity Indicator (MCI) was 91 developed (Ellen MacArthur Foundation & Granta Design, 2015) which aims to quantify 92 the regeneration of a product's material flow and is considered one of the few, among 93 sixteen CE indexes suiting a micro-scale assessment of circularity at product or company 94 level (Lonca et al., 2018). However, it focuses solely on technical cycles and recycled 95 materials. Furthermore, recovery and recycling through the biological cycle offered by 96 industrial composting, anaerobic digestion or biodegradation in natural environments are 97 not considered as end of life options. In order to apply the MCI system to BB plastic 98 products, the development of an enhanced methodology is necessary.

99 The approach proposed by the authors allows to quantify the circularity of BB plastic 100 products and to make comparisons with equivalent traditional plastic products. To

demonstrate the applicability of the proposed method a computational example for mulch
film products is provided. In so doing so, the paper aims at contributing to the Eco-design
of these innovative products.

104

# 1.1 The case study of mulch films

105 Plastic mulch films represent an important agronomical technique well established for the 106 production of many crops thanks to numerous agronomical advantages such as: increased 107 yield and higher quality of productions (Steinmetz et al., 2016) ; weed control and 108 reduced use of pesticides; early crop production and reduced soil moisture loss 109 (Briassoulis and Giannoulis, 2018). As a consequence, the plastic films consumption has 110 increased year-by-year, reaching a current global market estimated at 1.4 millions of 111 tonnes, mainly in Asia (Briassoulis and Giannoulis, 2018; Mormile et al., 2017), and 112 covering  $80,000 \text{ km}^2$  of agricultural surface (0.6% of the global arable land). The mulch 113 film market in Europe is estimated by Agriculture Plastic & Environment and by the 114 European Bioplastic Associations at 76-80 kt. The most used raw material is Poly-115 Ethylene (PE) in its different forms, due to its processability, chemical resistance, high 116 durability and flexibility (Kasirajan and Ngouajio, 2012; Plasticulture, 2016 and 2018; 117 Shen, M. et al., 2019; Wen, X. et al., 2018).

118 Despite these benefits, manifold environmental and agronomic problems have been 119 pointed out. After its useful life - which in general does not exceed 1 to 3 months - the 120 mulch film has to be removed and properly disposed of, a time-consuming (about 16 hours 121 per hectare) and costly procedure (Scaringelli, M., 2016; Briassoulis, D., 2013). The 122 recovered film is usually heavily contaminated with soil and organic residues, making 123 mechanical recycling technically difficult and not a cost-efficient solution (Briassoulis et 124 al., 2018; Figuier, 2016; De Lèpinau, Arbenz, 2016). The most common end of life of 125 collected films in Europe is still landfilling (about 50%), followed by energy recovering

126 and finally mechanical recycling (Le Moine, 2014). Recent Chinese prohibition (January 127 2018) to import different types of wastes is heavily impacting the European agricultural 128 plastic waste management, highlighting the difficulty in properly recycling this type of 129 plastics (Tamma, 2018). Plastic films may not be properly collected and recycled but 130 disposed of by burning in the field or by uncontrolled landfilling or left directly in the 131 (agricultural) soils, causing serious environmental concerns. An example is the "White 132 pollution" phenomena described in the Xinjiang Autonomous Region (China), in which 133 the residual plastic film can reach 200 kg/ha in the top soil with detrimental effects on 134 soils' quality, health and fertility (Liu, He, & Yan, 2014; Gao et al., 2019; Steinmetz et 135 al., 2016).

136 As a reaction, there has been significant research into novel materials especially related to 137 biodegradable and bio-based (BB) mulch films, which enable an effective biodegradation 138 in soil and provide comparable agronomical performances (Touchaleaume et al., 2016). The term "bio-mulch film" brings together several types of both bio-based and fossil oil-139 140 based biodegradable polymers and blends of them, such as polylactic acid (PLA), 141 polybutylene adipate co-terephthalate (PBAT), starch-based polymer blends or 142 copolymers. They biodegrade when exposed to bioactive environments such as soil and 143 compost (Kasirajan et al., 2012) which means that they can be left in situ to be fully 144 biodegraded after being used. Clearly the biodegradation rate of biodegradable bioplastics 145 is influenced by the environmental conditions such as the types of available bacteria, fungi 146 thus specific enzymes namely native microflora (Pico, Y. et al., 2019). However their 147 intrinsic biodegradability allow the complete biodegradation with times similar to natural 148 polymers such as cellulose used as reference by the relevant standards and certification 149 schemes.

150 The EN 17033:2018 is a new European Norm (standard) concerning "Plastics -151 Biodegradable mulch films for use in agriculture and horticulture - Requirements and test 152 methods", which sets the necessary tests and limits to define biodegradability, 153 performances and environmental impacts of BB much films. The material is considered 154 completely biodegradable if it achieves a complete biodegradation (absolute or relative to 155 the reference material) in a test period no longer than 24 months (mineralization into 156 CO<sub>2</sub>). Additionally, a control of constituents (such as metals) and eco-toxicity testing 157 (acute and chronic toxicity tests on plant growth, earthworm; nitrification inhibition test 158 with soil microorganisms) were required. A certified mulch film guarantees that the 159 product will completely biodegrade in the soil without adversely impacting on the 160 environment.

161 *1.2 Goal of the paper* 

162 The goal of the paper is to provide a general and common metric to measure the 163 circularity of a bio-based and biodegradable (BB) product and to apply the methodology at 164 product level to a category of products, namely bio-based and biodegradable mulch films.

165 2 Materials and Methods

## 166 2.1 MCI accounting according to the EMF methodology

167 The Material Circularity Indicator (MCI), according to the Ellen MacArthur Foundation 168 (EMF) methodology (Ellen MacArthur Foundation & Granta Design, 2015), is a number 169 that can range from 0 (pure linearity) to 1 (pure circularity). A purely linear production 170 provides for the exclusive use of virgin raw materials that turn into waste at the end of the 171 use phase of the product. Vice-versa, pure circularity includes the use of recycled 172 materials and does not produce wastes (regenerative streams). Circularity can be achieved 173 in different ways: as for the purpose of this paper, only recycling will be considered since 174 reuse is not an option for thin biodegradable mulch films. Since the method considers only 175 mass flows, the recycling corresponds to the recovery of materials for the original purpose 176 or for other purposes and excludes energy recovery, considered as a loss of materials equal 177 to landfill disposal. The materials recovered feed back into the process as recycled 178 feedstock.

The MCI methodology differentiates 'technical cycles' from 'biological cycles', modelling only the former. The first contains products and materials re-entering into the system (market) with the highest possible qualities and for as long as possible (thanks to reuse, repair, refurbishment and recycling) and the latter includes biological materials used in cascade until their restoration into the biosphere and the re-constitution of natural resources.

The material flows associated to the production of a generic technical cycle from nonrenewable sources are summarized in Figure 1. The dashed lines indicate that recycled feedstock does not have to be sourced from the same product but can be acquired on the market. With reference to Figure 1, the list of the parameters used in the EMF methodology is reported in Table 1, while the equations relevant for the analysis carried out in this paper are described in the following sections (Table 2, Chapter 2.2).





192 Figure 1: Diagram of material flows and associated variables of a generic

193 product (modified from Ellen MacArthur Foundation & Granta Design, 2015).

194

 Table 1: Parameters and relative definitions used in the EMF methodology.

Parameter	Definition
М	Total mass of the product
$F_R$	Fraction of mass of a product's feedstock from recycled sources
$F_U$	Fraction of mass of a product's feedstock from reused sources
V	Mass of virgin feedstock used in a product
$C_R$	Fraction of mass of a product being collected to go into a recycling process
$C_U$	Fraction of mass of a product going into component reuse
$E_C$	Efficiency of the recycling process used for the portion collected for recycling
$E_F$	Efficiency of the recycling process used to produce recycled feedstock for a
	product
W	Total mass of unrecoverable waste associated with a product
----------------	---
W <sub>0</sub>	Mass of unrecoverable waste (landfill, waste to energy and any other type of
	process where the materials are no longer recoverable)
W <sub>C</sub>	Mass of unrecoverable waste generated in the process of recycling parts of a
	product (after use)
$W_F$	Mass of unrecoverable waste generated when producing recycled feedstock for
	a product
X	Utility of a product, calculated as $X = (L/L_{av})(U/U_{av})$
L	Actual average lifetime of a product
$L_{av}$	Actual average lifetime of an industry-average product of the same type
U	Actual average number of functional units achieved during the use phase of a
	product
$U_{av}$	Actual average number of functional units achieved during the use phase of an
	industry-average product of the same type

196 The Material Circularity Indicator is determined as follows:

where *LFI* is the Linear Flow Index measuring the flows of virgin materials andunrecoverable wastes associated to the examined product.

A function of the utility, -, is used to correct the *LFI*. The function *F* is chosen in such a way that improvements of the utility of a product (e.g., by using it longer) have the same impact on its MCI as a reuse of components, leading to the same amount of reduction of virgin material use and unrecoverable waste. Setting a = 0.9, MCI takes, by convention, the value 0.1 for a fully linear product (*i.e.*, *LFI* = 1) whose utility equals the industry average (*i.e.*, X = 1). This leaves some margin to distinguish between processes with a high linearity but different utilities.

## 206 2.2 MCI accounting for bio-based and biodegradable (BB) products

207 To apply the EMF methodology to BB products, formulas and flows (Figure 1 and Figure208 2) are adapted as it follows:

- 209 1. The fraction of the recycled feedstock,  $F_R$ , corresponds to the share of the bio-210 based feedstock content in the final BB product,  $F_{R(i)}$ . It is the ratio of the d.m. 211 amount of bio-based feedstock per d.m. amount of the total mass of BB 212 product (EN 16785-2:2016).
- 213 2. The fraction of restorative mass going into a recycling process,  $C_R$ , corresponds 214 to the share of bio-based feedstock content in the BB product biologically 215 recovered (*e.g.* through composting) or biodegraded in the natural 216 environment, as it happens for specific applications (e.g. biodegradable mulch 217 film, *etc.*). It is the ratio of the d.m. amount of bio-based feedstock per d.m. 218 amount of the total mass of BB product that is biologically recycled.

The modified scheme is shown in Figure 2. Table 2 lists the formulas as adapted to BBproducts.

221

222

**Table 2:** List of formulas as developed by EMF methodology compared to the proposed adaptation to BB products.

# EMF methodology

## Adaptation to BB products

The mass of fossil-based feedstock which may be contained in BB products (V) is obtained as a difference of the total mass (M) minus the bio-based fraction; in this case the  $F_R$  in the EMF methodology corresponds to the sum of the fractions of all the biobased feedstock/s used in manufacturing the BB product. Therefore, is the total bio-based feedstock mass in the product. In single-use products, such as mulch films, reuse is not considered for BB products, so that  $F_U = C_U = 0$ .

223

 $W_F$  is the total amount of unrecoverable waste associated to the production of bio-based 230 231 feedstock used to produce BB products (i.e. the amount of uncoverable waste per unit of 232 BB product). Bio-based feedstocks such as starch, PLA, PHB etc. generate non-restorative 233 flows which can be quantified. Such unrecoverable waste correspond to  $R_{(i)}$ , the specific 234 amount of waste generated within cradle-to-gate boundaries per unit of bio-based 235 feedstock going into manufacturing, and it is estimated through LCA studies. Thus all 236 inputs from growth and harvesting phases and the related wastes generated by fertilisers and pesticides are here accounted.  $R_{(i)}$  can be easily found in specific literature or life 237 238 cycle inventories (LCI) present in LCA databases. In the calculation of  $W_F$ , also the 239 efficiency of manufacturing process of BB products  $E_P$  is considered, as the ratio of the

- 240 overall bio-based feedstock content in the final BB product to the bio-based feedstock in
- 241 input to the manufacturing process.
- 242 The material flows associated to the production of a generic BB product are summarized
- in Figure 2.



Figure 2: Description of material flows adaptation to BB products; in this paper, the reuse flow is out of scope ( $C_U = F_U = 0$ ).

The biodegradation of bio-based feedstock does not imply the generation of waste  $W_C$  as it occurs in a standard mechanical recycling process. This implies that  $C_R$  and  $E_C$  (*i.e.* the efficiency of the biodegradation process) equal to 1. Indeed, a BB raw material, sent to biological treatment (composting) or biodegraded in a natural environment, is fully transformed in its chemical elements (C, H and O mainly) derived from the decomposition of complex molecules (polymers) without the release of waste (Witt et al., 2001; Marten et al., 2003; Eubeler et al., 2010; BASF, 2018; Institute of Bioplastics and Biocomposites, 254 2018; OWS, 2018; Zumstein et al., 2018). These natural elements return into the 255 environment and are then available in the respective biogeochemical cycles. The 256 (biodegradable) fossil portion behaves as well; consequently,  $W_C = 0$ .

Nevertheless, the fossil-based feedstock cannot be considered as a regenerative circular feedstock, since it derives from carbon stored for millions of years and extracted by man, not being part of the active and fast biogeochemical carbon cycle. This is accounted in the quantification of  $W_0$ , the mass of unrecoverable waste from use (*i.e.* the linear stream going to landfill or incineration, the Non-Restorative Flows, NRF), as , the total amount of fossil-based feedstock.

Since  $W_F$  and  $W_C$  are associated to complete different processes and  $W_C$  is always equal zero, the double counting issue does not occur and the quantification of W and *LFI* is modified as reported in Table 2.

### 266 2.3 MCI calculation for mulch films: scope, inventory and assumptions

The new formulas reported in Table 2 were applied to a single use product namely a BB mulch film, to calculate their corresponding MCI. The transformation of BB materials into the final products (i.e. white mulch films) takes place without any modification of the biobased feedstock content and the process yield is close to 1.

In the global market, there are several branded BB mulch films (Moreno et al., 2017), both starch-based or blends of polyesters. In the following, the BB film has been arbitrarily assumed to be a starch-based mulch film with a 30%-portion of bio-based feedstock (i.e. 23% of starch,  $F_{(S)}$ , and 7% of a bio-based additive,  $F_{(BA)}$ ), while the rest was assumed to consist of fossil feedstock (



- 281
- 282
- 283



Figure 3: Examples of hypothetical bio-based polymers; in this paper, the first option on the left (starch blend 30/70) has been chosen for carrying out the numerical MCI calculation (working hypothesis). The figure considers a 100%-efficiency in every phase of production, so that the residues are equal to zero; the same assumption is done in this paper. \*TPS (Thermoplastic starch), starch content 75%; \*\*Ratio TPS/Polymer; modified from Institute of Bioplastics and Biocomposites, 2018.

	BB mulch film	
Material	30% bio-based feedstock (23% starch + 7% bio-	
	based additive) + 70% fossil feedstock	
Thickness (μm)	12	
Density (g/cm <sup>3</sup> )	1.25	
Weight (g/m <sup>2</sup> )	15.2	
Functional unit	$6000 \text{ m}^2$ /ha (the actual mulched soil in a hectare	
(the covering of the agricultural land)	is generally equal to the 60% of the total area;	
	Malinconico, 2017)	

302 In the calculation of MCI for the BB mulch film, the adapted formulas were used together 303 with assumptions. As stated before, BB mulch films are blends of bio-based and fossil 304 based feedstocks (in the specific case, 30% and 70% respectively). Unlike the LDPE 305 mulch film that has to be removed and disposed of, the BB mulch film is left in soil where 306 it undergoes an ultimate biodegradation (so that  $C_R = 1$ ) with no waste (so that  $E_C = 1$ ), in 307 respect of the specific standard EN 17033:2018. As a result of polymers' decomposition, 308 the derived (biogenic) C, H and O finally return into biosphere (atmosphere, 309 microorganism biomass, organic material pool) (OWS, 2018), and back into 310 biogeochemical cycles in a relatively short time ("Biogenic elements accounted as 311 recycled" in Figure 2), with the exception of humified compounds. Actually, also C, H 312 and O deriving from fossil-based sources undergo biodegradation (Zumstein, M.T., 2018) 313 but they are not considered as a regenerative flow ("Waste from non-restorative flow" in 314 *Figure 2*) and their "wastes" are indeed calculated in  $W_0$ .

Applying a conservative approach,  $W_F$ , the waste generated by the production of each biobased feedstock, is quantified considering a "cradle to gate" LCA study. The estimated solid wastes  $R_{(i)}$  for the presented case study are related to the production of starch ( $F_{(S)}$ ), with an amount  $R_{(S)}$  of 0.014 kg of waste per kg of renewable feedstock (source: personal communication A. Novelli), and to the production of the bio-based additive ( $F_{(BA)}$ ), with  $R_{(BA)}$  equals to 0.025 kg waste/kg renewable feedstock (US-LCI database). As assumed in

321

Figure 3, the production efficiency of BB product  $E_P$  (how much bio-based feedstock is needed for every unit of BB product) is estimated equal to 1 and no unrecoverable wastes are generated by the process.

In addition, an explorative sensitivity analysis has been performed regarding exclusively the amount of bio-based feedstock content of the BB mulch film, *i.e.* (*i.e.*,  $F_{(S)}$  +  $F_{(BA)}$ ), as shown in Figure 4 (Chapter 3). Considering the characteristics of the films (weight, g/m<sup>2</sup>, or thickness, µm, and density, g/cm<sup>3</sup>) and the relative functional unit (6000 m<sup>2</sup>/ha, Table 3), it is possible to calculate a mass, *M*, that is 90 kg/ha for the BB one. Once calculated the masses, the formulas reported in Table 2 (Chapter 2.2) are applied. Results are shown in Table 4.

332

333 2.4 Sensitivity analysis

334 A sensitivity analysis was conducted for BB mulch film to examine the effects of 335 changing the main variables. Given a non-linear dependence of results on parameter 336 values, a Monte Carlo approach (see, e.g., Lloyd and Ries, 2008) has been adopted. The 337 model has been implemented using specifically written routines in the C++ programming 338 language. The model was run with 100,000 events for BB mulch film, where the value of 339 each parameter has been randomly chosen following a Gaussian distribution with a 340 standard deviation within a range of possible and realistic values (Table 5 and Error! 341 Reference source not found.; Figure 5 and Figure 6).

## **342 3 Results**

- 343 Figure 4 shows how the value of the MCI varies according to the percentage variation of
- 344 the bio-based feedstock in the total mass of the product.
- 345
- 346

**Table 4:** Resulting parameters in the calculation of MCI for BB mulch film.

Parameter BB mulch film



Figure 4: MCI as a function of the amount of bio-based feedstock/s in the BB mulch film  $\Sigma F_{R(i)}$ , expressed as the percentage of all the bio-based feedstock/s of the mulch film on dry mass basis (X-axis). The dots correspond to the three different hypothetical bioplastic compositions of Figure 3.

#### 355 3.1 Sensitivity analysis

The results of the sensitivity analysis are presented in the followings Table 5 and Figure 5 and Figure 6. The accuracy band is a fraction of the average and corresponds to a probability of 95%. It has been chosen in order to be representative of the variability of the product category, the BB mulch films. The simulation can thus be regarded as a system composed by a high number of companies, each producing films with different characteristics, that are accounted for in the accuracy band.

362 *Table 5:* Parameters used for the sensitivity analysis of the BB mulch film. (\*\*) The
363 Accuracy Band is defined as twice the standard deviation of the distribution.

Variable name	Average	Accuracy Band (**)	Unit
М	1000.00	0%	kg
$F_{(S)}/F_{(BA)}$	3.29	10%	fraction
$\mathbf{F}_{(S)} + \mathbf{F}_{(BA)}$	0.30	30%	fraction
$\mathbf{F}_{\mathbf{U}}$	0.00	0%	fraction
C <sub>U</sub>	0.00	0%	fraction
R <sub>(S)</sub>	0.014	100%	fraction
R <sub>(BA)</sub>	0.025	100%	fraction
E <sub>C</sub>	1	0%	fraction
E <sub>P</sub>	0.95	10%	fraction
C <sub>R</sub>	1.00	0%	fraction





Figure 6: The most sensitive and relevant parameters in the calculation of the

F(S)+F(BP)

R<sub>(S)</sub>

R<sub>(BP)</sub>

Ep

371 MCI of the BB mulch films.

F(s/F(BP)

#### **372 4 Discussion**

This work applies the principles of the EMF methodology into BB products so as to define common metrics for calculating their circularity. By doing so it proposes some substantial changes to the EMF methodology but still coherent with the overall methodological framework. Such changes should be seen as a generalisation of the methodology provided the following rules are applied:

378 (1) fossil-based feedstocks or component materials embodied in the BB products whatever379 is the final disposal (even biological recycling) shall be considered as non-restorative;

(2) bio-based component materials embodied in the BB product that go to biological
recycling like composting, or biodegrade in the environment (i.e. BB mulch film) shall be
considered restorative as long as they flow through the biosphere safely, without any harm
to the environment (e.g. no toxicity effects).

(3) bio-based component materials embodied in the BB product that go to incineration andlandfill shall be considered as non-restorative;

386 The justification of these rules is described in the following.

387 Fossil-based component materials in the product derive from deposits where they 388 remained stocked for a geological time scale. Once the product is mineralised, its fossil-389 based portion will be accounted as non-regenerative and therefore linear, due to its origin 390 (Joos et al., 2013). This is true, even if fossil carbon, for example, will re-enter biological 391 cycles, like CO<sub>2</sub> in the atmosphere and other streams, since both fossil-based and bio-392 based component materials will physically and chemically behave the same, once 393 biodegraded. However, the source of the bio-based carbon was circular before its use 394 (concept of "carbon neutrality", equilibrium between the biogenic carbon released and the 395 carbon absorbed by plants) and will maintain its circularity provided that the carbon is 396 released into the atmosphere at the same rate. The reason has its origin in the EMF general 397 provisions stating that "biologically sourced materials can only be considered part of a 398 Circular Economy if materials are not used faster than they can be restored naturally" 399 (Ellen MacArthur Foundation & Granta Design, 2015). If BB products are incinerated, the 400 bio-based components are still considered linear, maintaining consistency with EMF 401 principles. Basically, a complete circularity for a BB product is satisfied when its 402 renewable components are 100% bio-based and they go 100% to biological recycling or 403 biodegraded in the environment (for specific application like mulch film).

404 As for provision (3), a material health rule has its origin in manifold normative definitions 405 of the CE. In addition, the EMF definition of biological cycles is that of non-toxic 406 materials which are restored into the biosphere and the CE is defined as such if it can 407 "eliminate the use of toxic chemicals". The need of a safety clause has been reviewed 408 under many aspects by Verberne (2016) and can be put as a postulate of the restoration 409 principle: if a flow is toxic it cannot be defined restorative. This is also at the core of the 410 REACH Regulation (EC 1907/2006). In the specific case, the material complies with the 411 standard EN 17033-2018 certifying that no harm is caused to a) all relevant organism 412 groups as plants, invertebrates (e.g. earthworm) and microorganisms, b) important 413 ecological processes maintaining soil functions, c) all relevant exposure pathways as soil 414 pore water, soil pore air and soil material.

A comprehensive approach for MCI calculation should also include non-restorative flows generated at upstream level like biomass growth, in the specific case corn, and biomass conversion processes like starch extraction and refining. Specifically these non-restorative flows correspond to the overall non-recyclable wastes associated to the bio-based feedstock supply thus non-recyclable waste from fertilizer and pesticide production, nonrecyclable scraps from conversion processes, etc. In this study such flows of nonrestorative waste coming from upstream manufacturing operations were included for the bio-based feedstocks ( $R_{(i)}$ ) used in manufacturing the BB mulch film applying "cradle to gate" LCA methodology. However, we observed that the inclusion of upstream unrecoverable waste does not significantly influence the MCI results in the chosen case study, since the respective amounts are small. The specific unrecoverable waste for starch and bio-based additive (*i.e.* kg of waste/kg of bio-based feedstock) were estimated at 0.014 and 0.025, respectively.

428

The resulting MCI for the 30/70 blend of the BB mulch film is equal to 0.37 in a 0-1 scale and its circularity is linearly linked to the amount of bio-based feedstock used according to the equation y = 0.89x + 0.1, where y is the MCI and x is the bio-based feedstock content, therefore the amount of recycled feedstock or (renewable) bio-based feedstock in input is decisive.

434 Apart from the specific application analysed in this paper, the proposed MCI method can
435 be easily applied and calculated for any kind of BB product as long as the following
436 information are available:

The bio-based feedstock content, determined according to the standard EN 167852:2016, if the composition is known, or directly provided by the BB product manufacturer.

• The end of life scenario of the studied BB product (real or hypothetical).

The amount of un-recoverable waste associated to the production of bio-based
feedstock contained in the BB product. They can be derived from LCA databases or other
specific sources.

### 443 **5** Conclusions

444 Bioplastic market is steadily increasing. The value proposition of bio-based and445 biodegradable products is linked to:

the use of renewable feedstock (like starch and its derivates) instead of fossil oil or
natural gas;

448

449

2. the waste recovery through biological recycling, thanks to their ability to biodegrade in composting facilities or in soil (*e.g.* biodegradable mulch film).

450 The Material Circularity Indicator (MCI), developed by the EMF, is a metric for 451 quantifying "how much" a product is circular (MCI = 0, fully-linear product; MCI = 1, 452 completely circular product) thus it represents a valuable tool for product eco-design 453 purposes. However, it focuses solely on technical materials, mechanically recycled or 454 reused, leaving out bio-based feedstocks and related biological treatments such as 455 composting. Without common metrics it is not possible to pursue concrete actions, to 456 achieve measurable results and to provide unequivocal references for all products. This 457 research work aims at filling this gap through the development of a methodology coherent 458 with EMF MCI methodology but able to catch the specificities of bio-based and 459 biodegradable products and provide metrics for those innovative products. Direct uses are: 460 (i) supporting the eco-design of innovative bio-based products and (ii) comparing the MCI 461 of BB products with MCI of traditional products (e.g. fossil based).

The proposed method has been applied to a real case study (*i.e.* biodegradable mulch film) providing quantitative metrics about its circularity. Specifically considering a bio-based feedstock content of 30%, the correspondent MCI is 0.37 in a 0-1 scale and its circularity is heavily linked to the bio-based feedstock content according to this relation:  $MCI_{(BB mulch})$ mulchmulch = 0.89\*bio-based feedstock + 0.1.

The MCI is a key performance indicator to develop more circular products, in line with the Circular Economy principles like the use of renewable materials and the reduction of the amount of not recoverable waste. MCI will support the development of innovative products just based on these two important characteristics specific for each BB 471 product/application and end of life scenario. Bioeconomy, thus also BB products, can 472 provide valuable insights in transforming the current (linear) economy in a more circular 473 one, however, the way the biomass is produced, processed and BB products are produced 474 are fundamental aspects to be properly assessed and monitored. This can be done using 475 specific methodologies like LCA. Within this context the proposed MCI has to be seen as 476 a complementary (quantitative) tool for further qualifying the sustainability of BB 477 products and not as a substitute tool. Furthermore the MCI here proposed is meaningful 478 only if BB products meet health and safety material requirements according to the national 479 and European laws and standards. This is a postulate of the proposed methodology 480 especially for those BB products conceived to biodegrade in the environment like 481 biodegradable mulch film.

482

483

484 **Declaration of interest** 

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

488

## 489 Acknowledgements

The contents of the paper are part of the findings of the project STAR-ProBio. STAR-ProBio has received funding from the European Union's Horizon 2020 program research and innovation programme under grant agreement No. 727740. The authors thanks prof. Andrea Contin for the fruitful discussion and contribution to the sensitivity analysis, Francesco Degli Innocenti for providing valuable comments and feedback on the 495 topics addressed by the paper and Alessandra Novelli for the general support in the MCI496 elaboration.

497

498

### 499 Addendum

500 While this paper was undergoing peer review the authors became aware that the EMF 501 published an update of the MCI methodology (Ellen MacArthur Foundation & Granta 502 Design, 2019) including the extension of it to include the treatment of biological materials. 503 This update introduces new definitions and formulas. The authors believe that most of the 504 changes regarding accounting are in the direction here proposed and that this study can 505 contribute as an illustration on how the material circularity of a biological based material 506 can be addressed in a real case study. Furthermore the authors would like to highlight that 507 the proposed methodology started long before the EMF changes: specifically the original 508 idea dated back to 2017 and a beta version of it - not as it is now - was presented in the 509 middle of 2018 at the Italian Circular Economy Stakeholder Platform (ICESP 510 www.icesp.it).

511

```
512 References
```

- BASF, 2018. Biodegradable mulch film clarification of polymer fate in soil. CIPA Congress, Bordeaux/Arcachon, France, May 2018.
- Bio-Based and Biodegradable Industries Association. BBIA reports. https://bbia.org.uk/reports/ (accessed 28 November 2019)
- Briassoulis, D., Giannoulis, A., 2018. Evaluation of the functionality of bio-based plastic mulching films. Polym. Test. 67, 99–109. https://doi.org/10.1016/j.polymertesting.2018.02.019
- Briassoulis D., Hiskakis, M., Babou, E., 2013. Technical specifications for mechanical recycling of agricultural plastic waste. Waste Management, Volume 33, issue 6,

pages 1516-1530, ISNN 0956-053X. <u>https://doi.org/10.1016/j.wasman.2013.03.004</u> De Lèpinau, P. and Arbenz, A., 2016. Economic and environmental impact of soil contamination in mulching film, Plasticulture, N° 136, 28-48.

- Ellen MacArthur Foundation & Granta Design, 2015. Circularity Indicators An approach to measure circularity Methodology. https://www.ellenmacarthurfoundation.org/assets/downloads/insight/Circularity-Indicators\_Methodology\_May2015.pdf.
- Ellen MacArthur Foundation & Granta Design, 2019. Circularity Indicators An approach to measure circularity Methodology.
- Ellen MacArthur Foundation, 2017. The New Plastic Economy: Rethinking the future of plastic & catalysing action.
- EN 16785-2:2016 Bio-based products Bio-based content Part 2: Determination of the bio-based content using the material balance method.
- EN 17033:2018 Plastics Biodegradable mulch films for use in agriculture and horticulture Requirements and test methods.
- EPLCA European Platform on LCA. https://eplca.jrc.ec.europa.eu/?page\_id=86
- Eubeler, J., Bernhanrd, M., Knepper, T., 2010. Environmental biodegradation of synthetic polymers II. Biodegradation of different polymer groups. Trends in Analytical Chemistry. 29, 1, 84-100
- European Commission, 2015. Closing the loop An EU action plan for the Circular Economy. COM(2015) 614 final. Brussels, 2.12.2015
- European Commission, 2018. A European Strategy for Plastics in a Circular Economy. COM(2018) 28 final. Brussels, 16.1.2018
- European Bioplastics. European Bioplastic publications. https://www.europeanbioplastics.org/news/publications/ (accessed 28 November 2019)
- Figuier, B., 2016. Plasticulture in Europe, Plasticulture, N° 136, 20-28
- Gao, H., Yan, C., Liu, Q., Ding, W., Chen, B., Li, Z., 2019. Effects of plastic mulching and plastic residue on agricultural production: A meta-analysis. Sci. Total Environ. 651, 484–492. https://doi.org/10.1016/J.SCITOTENV.2018.09.105
- Ghisellini, P., Cialani, C., Ulgiati, S., 2016. A review on circular economy: The expected transition to a balanced interplay of environmental and economic systems. J. Clean. Prod. https://doi.org/10.1016/j.jclepro.2015.09.007

- Institute of Bioplastics and Biocomposites, 2018. Biopolymers Facts and Statistics. Hochschule Hannover, University of Applied Sciences and Arts. Edition 5, ISSN 2510-3431.
- Joos, F., Roth, R., Fuglestvedt, J. S., Peters, G. P., Enting, I. G., Bloh, W. V., ... and Friedrich, T., 2013. Carbon dioxide and climate impulse response functions for the computation of greenhouse gas metrics: a multi-model analysis. Atmospheric Chemistry and Physics, 13(5), 2793-2825.
- Kasirajan, S., Ngouajio, M., 2012. Polyethylene and biodegradable mulches for agricultural applications: a review. Agron. Sustain. Dev. 32, 501–529. https://doi.org/10.1007/s13593-011-0068-3
- Lange, B.K., 2016. Revision of the fertilisers regulation benefits of biodegradable mulch films. European Bioplastics. http://www.europarl.europa.eu/cmsdata/108931/Kristy%20Barbara%20Lange%20E UBP%20PPT2.pdf (accessed 28 November 2019)
- Le Moine, B., 2014. Agri-plastics waste management: a voluntary commitment from the industry. Presented at: Agricultural Film 2014 International Conference on silage, mulch, greenhouse and tunnel films used in agriculture (15-17 September, Barcelona, Spain).
- Liu, E. K., He, W. Q., & Yan, C. R., 2014. 'White revolution' to 'white pollution' agricultural plastic film mulch in China. *Environmental Research Letters*, 9(9), 091001.
- Lloyd, S. M., & Ries, R., 2007. Characterizing, propagating, and analyzing uncertainty in life cycle assessment: A survey of quantitative approaches. *Journal of Industrial Ecology*, 11(1), 161-179.
- Lonca, G., Muggéo, R., Tétreault-Imbeault, H., Bernard, S., & Margni, M., 2018. A Bidimensional Assessment to Measure the Performance of Circular Economy: A Case Study of Tires End-of-Life Management. In *Designing Sustainable Technologies*, *Products and Policies* (pp. 33-42). Springer, Cham.
- Malinconico, M., 2017. Soil Degradable Bioplastics for a Sustainable Modern Agriculture. Green Chemistry and Sustainable Technology. Springer.
- Marten, E., Muller, R., and Deckwer W., 2003. Studies on the enzymatic hydrolysis of polyesters I. Low molecular mass model esters and aliphatic polyesters. Polymer Degradation and Stability, 80, 3, 485-501.
- Moreno, M. M., González-Mora, S., Villena, J., Campos, J. A., & Moreno, C., 2017. Deterioration pattern of six biodegradable, potentially low-environmental impact mulches in field conditions. *Journal of environmental management*, 200, 490-501.

- Mormile, P., Stahl, N., Malinconico, M., 2017. The World of Plasticulture, in: Malinconico, M. (Ed.), Soil Degradable Bioplastics for a Sustainable Modern Agriculture. pp. 1–21. https://doi.org/10.1007/978-3-662-54130-2 1
- OWS, 2018. Accumulation of (bio)degradable plastics in soil. CIPA Congress 2018, Archacon, May 29.
- Pico Y., Barcelò, D., 2019. Analysis and prevention of microplastics pollution in water: current perspectives and future directions. *ACS Omega*, 4, 6709-6719.
- Plasticulture catalogues, 2018. http://plasticulture.qualif.e-catalogues.info (accessed 28 November 2019)
- Razza, F., Degli Innocenti, F., 2012. Bioplastics from renewable resources: the benefits of biodegradability. *Asia-Pacific Journal of Chemical Engineering*, 7 (Suppl. 3): S301– S309. https://doi.org/10.1002/apj.1648
- Scaringelli, M., Giannoccaro, G., prosperi, M., Lopolito, A., 2016. Adoption of biodegradable mulching films in agriculture: is there a negative prejudice towards materials derived from organic waste? *Italian Journal of agronomy*, 11:92.
- Shen M., Zhang, Y., Zhu, Y., Song, B., Zeng, G., Hu, D., Wen, Y., Ren, X., 2019. Recent advances in toxicological research of nanoplastics in the environment: A review. *Environmental Pollution*, 252: 511-521. https://doi.org/10.1016/j.envpol.2019.05.102
- Steinmetz, Z., Wollmann, C., Schaefer, M., Buchmann, C., David, J., Tröger, J., Muñoz, K., Frör, O., Schaumann, G.E., 2016. Plastic mulching in agriculture. Trading shortterm agronomic benefits for long-term soil degradation? Sci. Total Environ. 550, 690–705. https://doi.org/10.1016/J.SCITOTENV.2016.01.153
- Tamma, P., 2018. China's trash ban forces Europe to confront its waste problem, Politico, 2/21/2018.
- Touchaleaume, F., Martin-Closas, L., Angellier-Coussy, H., Chevillard, A., Cesar, G., Gontard, N., Gastaldi, E., 2016. Performance and environmental impact of biodegradable polymers as agricultural mulching films. Chemosphere 144, 433–439. https://doi.org/10.1016/j.chemosphere.2015.09.006
- US-LCI database. "Polylactide biopolymer resin at plant kg/RNA". https://www.nrel.gov/lci/ (accessed 9 December 2019)
- Verberne, J.J.H., 2016. Building circularity indicators. Eindhoven University of Technology.
- Wen, X., Du, C., Xu, P., zeng, G., Huang, D., Yin, L., Yin, Q., Hu, L., Wan, J., Zhang, J., Tan, S., Deng, R., 2018. Microplastic pollution in surface sediments of urban water areas in Changsha, China: Abundance, composition, surface textures. *Marine Pollution Bulletin*, 136: 414-423. https://doi.org/10.1016/j.marpolbul.2018.09.043.

- Witt, U., Einig, T., Yamamoto, M., Kleeberg, I., Deckwer, W., Muller, R., 2001. Biodegradation of aliphatic-aromatic copolyesters: evaluation of the final biodegradability and ecotoxicological impact of degradation intermediates. Chemosphere. 44, 289-299.
- Zumstein, M., Schintlmeister, A., Nelson, T., Baumgartner, R., Wagner, M., Sander, M., McNeill, K., Woebken, D., Kohler, H., 2018. Biodegradation of synthetic polymers in soils: Tracking carbon into CO<sub>2</sub> and microbial biomass. Science Advances, 4, 7.