



Metrics for minimising environmental impacts while maximising circularity in biobased products: The case of lignin-based asphalt

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ABSTRACT

Achieving a circular economy (CE) is seen by society and policymakers as crucial to achieving a sustainable, resource-efficient, renewable and competitive economy. Given the current threat of climate change, we must develop new products that not only maximise the circularity of resources but also minimise climate change impacts. While these two goals are usually aligned, trade-offs exist. For instance, recycling biobased asphalt is a better end-of-life option than landfilling from a resource efficiency perspective. However, landfilling of biogenic non-biodegradable material leads to permanent carbon storage and, therefore, climate benefits. To fully understand the potential benefits and impacts of biobased circular innovations, we need metrics to capture their complexity from both a circular and climate point of view. This study explores the use of different circularity and sustainability metrics to understand the impacts and trade-offs of lignin-based versus bitumen-based asphalts. The analysis is done by calculating the Material Circularity Index (MCI) and two newly developed indicators quantifying the biogenic carbon storage (BCS) of products (BCS₁₀₀ and c-BCS) while following the CE principles. In addition, the impacts regarding climate change, life cycle costs and ECI (environmental costs indicator) are also provided. Based on the MCI, it can be concluded that lignin-based asphalt roads have slightly higher material circularity than their bitumen-based counterparts. The BCS analysis indicated that the least circular lignin-based alternative sequesters the highest amount of carbon in the long term due to permanent storage in foundations. Despite these trade-offs, the results from the newly developed BCS indicators allowed to align both climate and circularity goals, guiding policymakers and industry actors to implement circular biobased strategies where the value of biobased materials is optimised. Finally, this article discusses the use of different circularity and environmental metrics for decision making in the context of a circular biobased economy.

1. Introduction

Over the last decade, the concept of “circular economy” (CE) has gained popularity and recognition among companies, policymakers, and civil organisations. The CE builds on the well-known and established concepts of waste management, ecoefficiency, and resource value retention (Reike et al., 2018), providing a renovated view of how our society should sustainably produce and consume goods and services. Through this renewed lens, products and services should reduce resource use and waste generation as much as possible by following different strategies labelled the R-strategies (Reike et al., 2018). The CE’s conceptualisation and definition have been the subject of much debate and diverse interpretations (Kirchherr et al., 2017; Korhonen

et al., 2018; Reike et al., 2018). Nevertheless, the shift from a “take, make, consume and waste” linear economy to a CE is recognised by society and policymakers as an essential step to achieving a sustainable, resource-efficient, renewable and competitive economy (European Commission, 2015).

Biomass as a renewable resource can play an essential role in the circular economy. Recently, the term circular bioeconomy has been introduced to intertwine the bioeconomy and circular economy concepts (Stegmann et al., 2020). As indicated by the Dutch government in their CE vision, when the use of new natural resources is unavoidable, renewable and abundant natural resources should be used to substitute critical abiotic resources (IM, 2016).

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Netherlands - eXtra Lignin (CHAPLIN XL) aims to achieve GHG emissions reduction at an industrial scale in asphalt pavement by substituting bitumen (fossil origin) with lignin (biomass origin). Lignin can be obtained as a by-product of pulp and paper industries or biorefineries, and has recently received attention as a renewable raw material to substitute petrochemical resources. It has been used to substitute, for example, urea-formaldehyde in adhesives or polyacrylonitrile in carbon fibers, typically resulting in a reduction of carbon emissions per kg of material (Moretti et al., 2021). The substitution of bitumen with lignin has a double aim in the fight against climate change: it avoids using fossil resources (namely, bitumen) and allows for the sequestration of biogenic carbon. Given the current threats of climate change, it is paramount to develop new technologies that not only maximise the circularity of resources but also minimise the impacts in climate change. While these two goals are usually aligned, some cases, like biobased asphalt applications, present particular challenges. Using biogenic resources for material production is, as explained above, considered a CE strategy provided that the raw materials have been produced in a sustainable approach and the conversion processes are efficient (Razza et al., 2020). Under the CE logic, these materials are renewable and should be either returned to nature (e.g. through biodegradation) or recirculated in the technosphere as long as possible. Lignin asphalt does not biodegrade; therefore, under the CE logic, this material should be recirculated into new roads for as long as possible. However, from a climate change perspective, the most desirable outcome for this biogenic material is landfilling or other forms of permanent storage since it would indefinitely remove CO₂ from the atmosphere without the CO₂ losses due to the recycling loop activities (i.e. when warming up the asphalt mixture or when reused in roads). To fully understand the potential benefits, impacts, and trade-offs of biobased circular innovations, we need metrics that are able to capture their complexity from both a circular and climate point of view.

1.1. Assessing the impacts of circular biobased products

Life Cycle Assessment (LCA) is widely accepted as the best method to quantify environmental impacts and make science-based decisions. It has also been identified as the most used methodology to assess the effect of circular strategies on products and services (Corona et al., 2019). However, the nature of circular and biobased supply chains presents several methodological challenges, whose normative nature leads to low consensus on how to solve them. This is evidenced by the variability of approaches encountered in the literature, leading to methodological uncertainty in the interpretation of LCA results (Guest et al., 2013; Tonini et al., 2021). The methodological challenges are mainly related to three aspects of biobased circular products (Tonini et al., 2021): (1) biobased materials have the potential to sequester CO₂, (2) biobased and circular supply chains are multifunctional, leading to allocation issues, and (3) biobased materials originated from agricultural products are related to direct and indirect Land Use Changes (LUC). Value chains with lignin produced from pulp and paper industries or from biorefineries do not have a direct link with transformation or land (van der Hilst et al., 2019). Land use change impacts are therefore not further discussed in this section.

The fact that biobased materials can sequester CO₂ leads to high variability in how biogenic carbon uptake, release and storage is accounted. When accounting for the biogenic carbon in bio-based materials, two principal approaches are dominant in the literature (Pawelzik et al., 2013): (i) the CO₂ incorporated in biomass during the growth phase and the corresponding emissions throughout the entire life cycle are either not modelled or are both characterised with a characterisation factor (CF, measured in kg CO₂-eq/kg CO₂) of zero (biogenic CO₂ is considered to be "carbon neutral"); or (ii) the CO₂ incorporated in biomass during the growth phase is inventoried as an uptake during the cultivation/growth phase, and as emission when it is released at end-of-life or throughout the life cycle. The CF for CO₂ uptake is then set

equal to -1 , while emissions of biogenic CO₂ emissions have a CF of 1.

According to a recent review performed (Bishop et al., 2021) on biobased plastics LCAs, the most typical approach is to assume carbon neutrality. Carbon neutrality considers that the amount of CO₂ absorbed during plant growth compensates for the CO₂ released at the EoL, independently of the time scale of the analysis, which in the case of bioplastics is typically short. However, this approach has been criticised for being misleading (Tonini et al., 2021), as the carbon cycle dynamics in the short/medium term can significantly vary when considering the difference in biomass growth rates. According to Bishop et al., the best practice is to explicitly model biogenic carbon uptake, storage and release over the extended biobased plastic life cycle. Relatively complex approaches have been proposed in the LCA literature to tackle this issue, including advanced dynamic modelling of storage time and biomass growth rates (Cherubini et al., 2011; Guest et al., 2013; Levasseur et al., 2013). These methodological approaches address the issue of time differences between uptake and release of carbon. However, there is no well-established approach to deal with the modelling of biogenic carbon that is stored in the product (Finkbeiner et al., 2013), i.e. that does not get released over the lifetime but gets stored in the subsequent products utilising the recycled biogenic material.

Besides biogenic carbon accounting, the modelling of multifunctional circular supply chains is another challenge that presents high variability in the LCA literature, especially for cascades of biobased products (Tonini et al., 2021). This challenge is related to how the waste or residual input is modelled (i.e. is it burden-free, or should it come with some impacts from the previous life cycle) and how the benefits of recycling are allocated at the end-of-life (EoL) between the primary and recycled material (e.g., the avoided burden approach, the recycled content approach, or the partitioning approach). It has been proven that even small differences in EoL modelling formulas can lead to different results when comparing biobased and fossil-based products e.g. plastics (Tonini et al., 2021). When we combine this issue with the accounting of biogenic carbon storage, the modelling becomes even more challenging, leading to very different conclusions from LCA studies (Finkbeiner et al., 2013). A common approach to deal with multifunctionality and carbon dynamics of biobased materials is to allocate the removal of CO₂ (i.e. credits) to the first product, leading to incentivising the use of biogenic materials, but not their recycling (Finkbeiner et al., 2013). This happens because the removal of CO₂ is allocated to the product using the biobased material, while the recycled product does not usually get a share of the credits from this removal. In addition, applying allocation rules that do not respect the physical flows (e.g., economic allocation) leads to incorrect accounting of biogenic carbon content since these rules do not always respect the biogenic carbon share of the products. Therefore, separate accounting of biogenic and non-biogenic carbon emissions, removals, and storage is advisable, as required by standards such as the GHG Protocol (GHG Protocol, 2011) and the updated EU product environmental footprint method (Zampori and Pant, 2019).

1.2. Goal of the study

Through the development of a straightforward LCA-based indicator, this study aims at answering the following main research question: What metrics can we use to promote products/technologies that maximise both biogenic carbon sequestration and resource circularity? Such indicator should allow us to understand the biogenic carbon content dynamics of circular products while minimising normative methodological challenges related to the LCA method. In addition, through a case study of biobased and conventional asphalt, this study answers the question: What is the potential and added value of combining LCA studies with material circularity metrics to provide a holistic assessment of a product system's sustainability and circularity? As explained in this introduction, the case of biobased asphalt entails environmental trade-offs regarding circularity and climate change impacts, but also methodological challenges regarding carbon storage and multifunctionality

processes. This makes bio-based asphalt a comprehensive and at the same time challenging case study from a methodological perspective. In addition, the case study provides first insights into the circularity and environmental trade-offs of lignin-based ashpalts.

The analysis is made by applying a combination of metrics, including a circularity and biogenic carbon storage (BCS) assessment, to different types of lignin-based and bitumen-based ashpalts. Based on the results obtained, recommendations on how to increase the circularity of asphalt roads and insights into the future of biobased circular asphalt are also provided.

The rest of the article is organised as follows. Section 2 includes the methodology used for the analysis of the case study, the explanation of the newly developed metrics, and the description of the asphalt system under study. Section 3 includes the results of the circularity assessment, the biogenic carbon storage assessment and the comparison with other environmental and economic metrics. The discussion of the findings are included in Section 4, providing the methodological insights and limitations from the case study and the use of circularity metrics for sustainable decision making. Finally, section 5 includes the conclusions of the study.

2. Methodology

This study assesses the environmental, economic and circularity implications of replacing bitumen with lignin in asphalt roads when considering a life cycle perspective (from the extraction of raw materials to the disposal of the road) and different sets of metrics.

The circularity assessment was conducted by identifying the main material flows of the system and calculating the Material Circularity Index (MCI) for each asphalt mixture. The description of the MCI calculation can be found in section 2.1. Two new indicators were developed to assess the biogenic carbon storage of the road system; the methodology for these new indicators is described in section 2.2. In addition, the results from these analyses were compared with the results of applying LCA and LCC methodologies. The choice of indicators for comparison are described in section 2.3.

The study's geographical scope is assumed to be the Netherlands, where lignin would be produced, asphalt manufactured, and the road constructed and used. Netherlands was chosen because it is the place where the CHAPLIN project has been developed. In addition, the inventory data for lignin production was obtained from Dutch biorefineries. The temporal scope is medium to long term since the technology is currently reaching a high technology readiness level. The functional unit of analysis used to compare results is defined as "1 m² of road used over 1 year".¹ The main characteristics of the system under study and the analysed scenarios are included in section 2.3.

2.1. Material circularity indicator (MCI)

The MCI, developed by the Ellen MacArthur Foundation, is an indicator expressing to what extent linear flows are minimised and restorative flows are maximised (EMF, 2015). The indicator aims to measure how effectively a company (through its products) transitions from linear to circular models. According to the developers, the MCI measures the circularity level of a product from 0 (fully linear) to 1 (fully circular) (EMF, 2019). This indicator was chosen since it is increasingly gaining popularity in both grey and scientific literature. Additionally, it is considered one of the most complete material circularity indicators (Corona et al., 2019). The method used in this study follows the latest published version of the index, which allows to include materials of biobased origin and material losses during manufacturing (EMF, 2019).

¹ According to the LCA terminology, the functional unit is defined as the unit of analysis that quantifies the function of the product allowing for fair comparison among products.

The MCI is calculated by applying equation [1] (EMF, 2019):

$$MCI = 1 - LFI \cdot F(X) \quad [1]$$

Where:

- *Linear Flow Index (LFI)* = Proportion of material sourced from virgin materials and ending up as unrecoverable waste. $0 \leq LFI \leq 1$. Calculated with equation (2) (EMF, 2019),

$$LFI = \frac{V + W}{2M + \frac{W_F - W_C}{2}} \quad [2]$$

where:

V = Mass of virgin feedstock used in a product (this amount excludes any biological material which originates from sustainable production).

W = Total mass of unrecoverable waste associated with a product, without including losses in manufacturing.

M = Total mass of the product.

W_F = Mass of unrecoverable waste generated when producing recycled feedstock for a product.

W_C = Mass of unrecoverable waste generated in the process of recycling parts of a product (after use).

- *Utility factor $F(X)$* = $0.9/X$

Where:

X = The intensity and length of product use, calculated with equation (3). The analysis assumes that the average lifetime of roads (L_{av}) is 30 years (assuming the replacement of the top layer at least once). As proposed by Mantalovas and Di Mino (2019), the utility for asphalt can be represented by the average number of loading cycles before failure in terms of fatigue or rutting. The corresponding test performed in the project tests indicated similar performance for lignin-based and bitumen-based roads, therefore, $U = U_{av}$ (EMF, 2019).

$$X = \left(\frac{L}{L_{av}} \right) \times \left(\frac{U}{U_{av}} \right) \quad [3]$$

2.2. 2.2 Biogenic carbon storage analysis

Several indicators were developed to understand the biogenic carbon content dynamics of circular products. The indicators are proposed as complementary to the LCA climate change impact indicator and thus align with the LCA method. It also complies with international standards, such as the GHG protocol, that requires reporting biogenic carbon stored separated from GW results. The indicators follow the biogenic accounting approach chosen for the LCA performed in this project, i.e. the Dutch product category rules for asphalt, as defined in EN 15804:2012. In this approach, biogenic carbon is assumed permanently stored if the storage time is at least 100 years. The chosen time horizon of 100 years corresponds with the most common time horizon for the assessment period and characterisation models of GHG emissions used in LCA. Although subjective and debated, it could be considered a balanced choice. A short time horizon, e.g. 20 years, would be helpful to assess short term impacts, but would overestimate the benefits of short term carbon storage. A long term time horizon, example.g. 500 years, would reduce the urgency of climate change mitigation relative to potential tipping points that could be exceeded within 100 years without effective mitigation policies (Brandão et al., 2013). The assumption leads to the following axioms for the indicators developed in Equations (1) and (2):

- Carbon can be stored in products during use or after disposal.
- Carbon is considered permanently stored when it has not been released into the air for at least 100 years.

- iii. The carbon contained in products that are landfilled or used for building foundations is considered permanently stored.

The new biogenic carbon storage (BCS) indicators are: (a) BCS₁₀₀, and (b) circular-BCS. These indicators focus on accounting for biogenic carbon stored by each product and do not consider other GHG emissions and removals over the entire life cycle. The latter is accounted for by the climate change result of the LCA.

b) BCS₁₀₀

The BCS₁₀₀ indicator measures the total amount of biogenic carbon stored over a period of 100 years, in kg C, assuming successive loops of product recycling. This indicator avoids the multifunctionality issue by expanding the system to include the previous and subsequent cycles. This means that the result is not provided per functional unit (as defined in the LCA), but per a unit of time defined as 100 years.

Equation (1) describes the calculation of the BCS₁₀₀ indicator. The carbon content of each asphalt mixture and scenario is defined by considering the initial biogenic carbon content of each asphalt ingredient (CC_i) minus the carbon losses along the lifetime of the road due to erosion and the recycling process (LU_i and LR_i), plus the carbon permanently stored due to their use in foundations (or landfilling in some countries) in all future product loops taking place up to a 100-years time horizon (PS_i • (100/LT - 1)).

$$BCS_{100} \text{ (kg C)} = \sum_i \left[CC_i - LU_i - LR_i + PS_i \cdot \left(\frac{100}{LT} - 1 \right) \right] \quad [1]$$

where:

- CC_i is the biogenic carbon content of each material *i* contained in the product (including recycled material input), in kg per FU
- LU_i is the loss of biogenic carbon in each material *i* due to the use of the product, in kg per FU
- LR_i is the loss of biogenic carbon in each material *i* due to the recycling or disposal process of the product, in kg per FU
- PS_i is the biogenic carbon in each material *i* that gets permanently stored at the EoL of the product, in kg per FU
- LT is the product lifetime in years. The maximum value for LT is 100 years, i.e.:

$$IF \text{ } LT \leq 100 \rightarrow LT = LT$$

$$IF \text{ } LT > 100 \rightarrow LT = 100$$

The biogenic carbon content of the product materials needs to be stored for 100 years in order to be considered permanently stored. This formula assumes that a product can permanently store carbon provided that the same production conditions are maintained over several closed-loop cycles in a time span of 100 years. Independently on the EoL treatment, the product's virgin materials store carbon for as long as the product's lifetime. If the product is successively recycled for 100 years (e.g. a product with a 20 years lifetime is recycled five times in 100 years), permanent storage is only considered for the materials recycled in the first product cycle. Materials used (and recycled) in successive product cycles cannot be considered permanently stored under our defined time horizon since they would be stored for a maximum of 100 minus LT years. The amount of carbon stored would thus equal the amount of carbon remaining in the recycled material at the end of one cycle of the product, hence the term (CC_i - LU_i - LR_i - PS_i) of Equation (1). However, the materials that are landfilled (or stored in foundations) at the EoL of each cycle are considered to be permanently stored, and therefore, the corresponding biogenic carbon would be stored as many times as product cycles taking place within the 100-years time horizon, hence the term PS_i • (100/LT) of Equation (1). For instance, if a product has a lifetime of 20 years (LT = 20 years), BCS₁₀₀ includes the

biogenic carbon stored by the first lifetime of the product (CC_i - LU_i - PS_i) plus the biogenic carbon permanently stored by the four successive lifetimes (= 100/LT - 1) taking place in the 100-years time horizon.

c) Circular BCS (c-BCS)

C-BCS accounts for the share of carbon content that is circular, e.g., recycled or reused, after one recycling cycle (instead of a 100-years time horizon). The c-BCS indicates to what extent biogenic carbon is stored while following the CE principles, avoiding the choice of alternatives that promote an inefficient use of carbon resources. The change in temporal scope (1 cycle vs. 100 years) keeps the scope in line with usual product-based metrics, such as the MCI and the LCA and LCC indicators. When considering a one-cycle scope, the permanent storage of biogenic carbon at the EoL happens only once (since only one cycle is accounted), and the rest of the materials are assumed to be partly stored by using an allocation factor that equals the share of storage time (i.e. lifetime of the product) over a 100-years time horizon (i.e., LT/100). This partial allocation of carbon sequestration according to storage time was also considered for the LCA of the system under study, performed by Moretti et al. (2022).

The c-BCS indicator avoids the multifunctionality issue by expanding the system to include the previous and subsequent cycle. This means that the result is not strictly provided per functional unit (as defined in the LCA) but includes the biogenic carbon that is stored by both the product providing the recycled material (previous cycle) and the product using the recycled material (subsequent cycle).

As defined in Equation (2), c-BCS considers the biogenic carbon content of the recycled input (through CCr_i) and the recycled output (through rr_i), over a time period defined as 2LT, i.e., the current cycle and the previous (for the recycled input) or the current cycle plus the subsequent (for the recycled output) cycles.

$$c - BCS \text{ (kg C)} = \sum_i \left[rr_i (CC_i - LU_i - LR_i - PS_i) \cdot \frac{2LT}{100} \right] + CCr_i \cdot \frac{2LT}{100} \quad [2]$$

where:

- rr_i is the recycling rate of each material *i* at the end of life
- CC_i is the biogenic carbon content of each material *i* contained in the product (including recycled material input), in kg per FU
- LU_i is the loss of biogenic carbon in each material *i* due to the use of the product, in kg per FU
- LR_i is the loss of biogenic carbon in each material *i* due to the recycling process of the product, in kg per FU
- PS_i is the biogenic carbon in each material *i* that gets permanently (and linearly, e.g., landfill) stored at the EoL of the product, in kg per FU
- LT is the product lifetime in years
- CCr_i is the biogenic carbon contained in the recycled material input, in kg per FU

2.3. Indicators from LCA and LCC studies

The results obtained by applying the MCI, BCS100 and c-BCS were compared to those obtained by applying the LCA and LCC methodologies. The results and detailed methodology for the LCA and LCC of the asphalt mixtures under study are already available in the publications from Moretti et al. (2022) and van Veen et al. (2021), respectively. The LCA environmental indicators chosen for comparison are: the impact in climate change (in kg CO₂/m²/yr) and in abiotic depletion (i.e. depletion of metals and minerals, in kg Sb eq/m²/yr). Both indicators were calculated with the CML baseline V3.06 method as implemented in Simapro 9. The climate change impact includes credits from biogenic carbon sequestration at the EoL of the road. For a better discussion and

interpretation of the climate change impact and the effect of biogenic carbon storage, the result in climate change is here provided both with and without biogenic carbon credits (the latter is labelled in the results Table 5 as *Climate change w/o BC*). The abiotic depletion indicator from the CML method captures materials' scarcity by considering both the extraction rates and the reserves of each resource and is recommended as the best LCA characterisation method to measure the impact in resource depletion (EC-JRC, 2011). It is, therefore, a good indicator to estimate the impacts of material use from a circular perspective. The economic indicators chosen for comparison are: total life cycle costs (in €/m²/yr) and the environmental costs indicator (ECI, in €/m²/yr). The latter indicator includes the environmental externalities per functional unit (also called as "shadow prices") and was calculated by Moretti et al. (2022) according to Stichting Bouwkwalityteit, 2019.

2.4. Scenarios and data inputs

The effect of replacing bitumen with lignin in asphalt production was calculated for different types of roads and corresponding asphalt compositions. The lignin-based roads were also compared to conventional roads made entirely with fossil-based bitumen. The analysis is performed for three different types of asphalt mixtures: SMA (stone mastic asphalt), AC (asphalt concrete) and ZOAB (porous asphalt, from Dutch *zeer open asfaltbeton*), and two scenarios: lignin-based or bitumen-based. These three types of asphalt contain different ingredient recipes allowing for different properties and road applications. SMA is designed for heavily trafficked roads, airfields, industrial and harbour areas, while AC is used for general roads and ZOAB for highway roads where good water drainage is needed. It should be noted that the composition of the ZOAB mixture is based on a recipe used in the past (till the year 2007). It is highly unlikely that ZOAB roads will use this mixture in the future. However, the mixture was included in this study as data was available and provided interesting methodological insights as a case study. The different asphalt compositions for each asphalt type and scenario are described in Table 1. The quantities are given per ton of mixed asphalt, including the three layers of the road (top, bind, base). The lignin-based asphalt mixtures provide similar performance and functionality to the bitumen references, as confirmed by the road tests done during the CHAPLIN XL project. The lifetime of the road differs per asphalt type, amounting to 30 years for the SMA and ZOAB mixtures, and 24 years for the AC mixture. The quantities provided include the extra asphalt needed to replace the top layer of the road once (after 15 years for SMA and ZOAB, and after 12 years for AC). It is assumed that lignin is produced from sustainable feedstock using the Goldilocks® process. In this

Table 1

Asphalt mixture compositions (per ton of asphalt) and lifetimes of the SMA, AC and ZOAB roads in both lignin-based and bitumen-based scenarios.

	Lignin-based scenarios			Bitumen-based scenarios		
	SMA	AC	ZOAB	SMA	AC	ZOAB
<i>Anti-drip fabric, kg</i>	1.22	0	0	1.22	0	0
<i>Asphalt granulates (recycled), kg</i>	398.4	515.9	380.0	398.2	520.9	380.0
<i>Bitumen 40/60, kg</i>	20.8	11.5	2.57	14.3	31.0	34.3
<i>Bitumen 70/100, kg</i>	5.52	5.47	25.9	26.5	0.0	0
<i>Crushed sand, kg</i>	224.8	248.3	197.0	220.3	253.6	202.9
<i>Gravel, kg</i>	503.6	389.5	598.3	501.7	382.5	592.2
<i>Filler, kg</i>	26.18	21.5	11.4	41.3	21.1	30.6
<i>Lignin, kg</i>	21.16	14.91	23.88	0	0	0
<i>Undisclosed^b, kg</i>	1.59	3.07	0.94	0	0	0
<i>TOTAL^a, kg</i>	1203.2	1210.2	1240.0	1203.6	1209.3	1240.0
<i>t/m²</i>	0.388	0.390	0.406	0.388	0.390	0.406
<i>LIFETIME top/base layers, years</i>	15/30	15/30	12/24	15/30	15/30	12/24

^a Note that the total amount of materials weights more than 1 ton because of the extra materials needed to replace the top layer (once per lifetime).

^b For confidentiality issues, the name of this biobased material is undisclosed.

process, developed by the biotechnology company Vertoro, sawdust is converted into pulp, lignin and cellulosic sugars.

Table 2 includes the biogenic carbon content of each material integrating the different asphalt mixtures.

The description of the material flows in the system per life cycle stage (asphalt production, road construction and use, and road demolition and waste management) is included in the following sections. Data inputs were collected from asphalt and lignin producers (partners in the CHAPLIN XL), and the assumptions are aligned with the inventory analysis of the previously performed LCA (Moretti et al., 2022).

2.4.1. Asphalt production and construction

The materials contained in the asphalt mixtures are virgin and fossil except for the biobased materials and the asphalt granulates that come from reclaimed asphalt. The lignin and the undisclosed biobased materials are mainly used to substitute bitumen in the top layer, although in the SMA and ZOAB mixtures, these materials are also used for the base layer. The reclaimed asphalt granulates are integrated in the bind and bottom layers of the SMA and ZOAB roads, contributing to 50% of the layer mass. In the case of AC roads, reclaimed asphalt is also used for the top layer, contributing to 29% of the layer mass. A summary of the lignin content and recycled content of each asphalt mixture per road layer is included in Table 3. Bitumen-based asphalt mixtures have the same recycled content as the lignin-based asphalts.

There are material losses during the manufacturing of asphalt granulate (1%), bitumen (5%), and sand and gravel (4%). The material losses are assumed to be lost or landfilled, except for the bitumen losses that are used for energy purposes in the refinery.

2.4.2. Road use

During use, top layers are exposed to climate conditions and oxygen from air which leads to the partial oxidation and an increase in stiffness of binders. Following the Dutch Product Category Rules for asphalt (as defined in EN 15804:2012), it was assumed that road use leads to a loss of 10% of the bind materials (for both bitumen and biobased) contained in the top layer due to road abrasion. For the carbon storage calculation, it is assumed that the carbon contained in these losses is permanently stored (i.e., encapsulated in bitumen particles released to the environment). Binder aging during use also leads to emissions to air in the form of volatile organic compounds (VOC), and intermediate/semivolatle organic compounds (I/SVOCs), in particular during hot and sunny days (Khare et al., 2020). The mechanisms behind these emissions are still poorly understood and uncertain, and were not included in the analysis. However, these emissions are mainly a concern to human health effects and carbon losses are likely small.

2.4.3. Road demolition and waste management

73% of the road is recycled at the end of life. It is assumed that the recycling process leads to material losses of 10% and 5% in weight of crushed stones in the top layer and the other layers, respectively. Additionally, it is assumed that both bitumen and lignin will suffer a decrease in quality, which is modelled through a reduction of 4% in weight of the recovered material. These losses are assumed to be oxidised and emitted as CO₂ when calculating the biogenic carbon storage.

The LCA model considers that 27% of the road materials are used for low value applications such as filling materials. However, using the recovered asphalt for foundations leads to a total loss of bitumen's and lignin's functionality; only the gravel is being used for its filling properties. Nevertheless, since these applications would probably store the biogenic carbon for more than 100 years, this carbon is assumed to be permanently stored.

Table 2
Biogenic carbon content of each asphalt mixture per asphalt type and scenario at factory gate.

Asphalt ingredients (in kg)	Biogenic carbon content in ingredient (%)	Lignin-based scenarios			Bitumen-based scenarios		
		SMA kg C/t	AC kg C/t	SMA kg C/t	SMA kg C/t	AC kg C/t	SMA kg C/t
Anti-drip fabric	46%	0.56	0	0	0.56	0	0
Reclaimed asphalt ^a	1.0%	5.06	5.08	4.88	0	0	0
Lignin	61.5%	13.0	9.17	14.7	0	0	0
Undisclosed	79%	1.26	2.43	0.74	0	0	0
TOTAL biogenic content (kg C/t asphalt):		19.89	16.67	19.97	0.56	0	0

^a This share of biogenic carbon content was calculated considering the remaining carbon content in reclaimed asphalt at the end of one road cycle made with virgin lignin-based asphalt. †For confidentiality issues, the name of this biobased material is undisclosed.

Table 3
Lignin content and recycled content of each lignin-based asphalt mixture (per road layer).

	SMA mixture	AC mixture	ZOAB mixture
LIGNIN CONTENT %wt top/bind/base layers	4%/0%/1%	2%/0%/1%	4%/0%/1%
RECYCLED CONTENT %wt top/bind/base layers	0/50%/50%	30%/50%/50%	0/50%/50%

3. Results

3.1. Circularity assessment

Figs. 1 and 2 show the material flow diagrams (in wt%) for each lignin-based asphalt mixture. These diagrams include the loop of the reclaimed asphalt at the EoL that is used as input in the next road cycle. The recirculated asphalt amounts to 33% of the initial mass for SMA, 31% for ZOAB roads, and 43% for AC roads. The amount of recirculated material is higher in AC roads because this application allows for a higher recycled content than in the other applications (i.e. 30% of the top layer mass).

The amount of recycled asphalt available at the EoL of the road is higher than the amount of reclaimed asphalt used in the mixture, therefore, some reclaimed asphalt (i.e. 31% of the initial asphalt mass in the case of SMA, 23% for AC and 33% for ZOAB) gets recirculated into other applications (such as recreation, agricultural or industrial grounds), without totally closing the material loop. Additionally, there are approximately 35% of material losses during manufacturing (3%), use (4%) and waste management (29%).

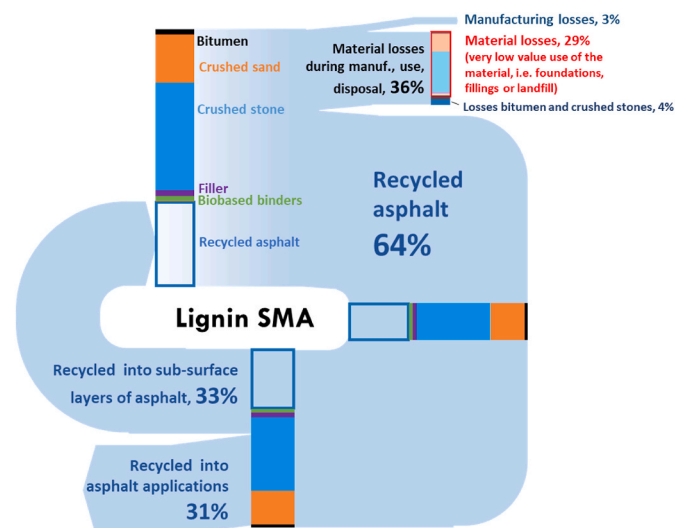


Fig. 1. Material flows diagram for lignin-based SMA road.

Fig. 3 show the results of the MCI calculation for each asphalt type and scenario. The MCI values for lignin-based scenarios are slightly better (around 1% better) than the bitumen-based scenarios due to the biobased mass included in the lignin-based scenarios, which is relatively low.

Independently on the scenario, the circularity of the AC roads is the highest, followed by ZOAB roads. AC roads have a higher recycled content in the top layer, leading to 0.13 and 0.18 higher MCI than SMA and ZOAB roads, respectively. The MCI for ZOAB roads is the lowest due mainly to the shorter lifetime (assumed to be 24 years). Although SMA roads do not have recycled content in the top layers, their circularity is higher than for ZOAB roads due to a relatively long lifetime (30 years) and a higher content of the biobased undisclosed binder and anti-drip fabric (made of cellulose).

3.2. Biogenic carbon storage assessment

The results for the BCS₁₀₀ (i.e. over a 100-years time horizon) and c-BCS (only circular biogenic carbon) for the lignin-based asphalts are depicted in Fig. 4. The values calculated for the terms defined in Equations (1) and (2) are described in Table 4. The BCS values for the bitumen-based roads are almost zero, with SMA roads having a small share of biogenic carbon storage (0.44 kg C/m² over 100 years) because of the cellulose material used as anti-drip filter. As shown in the figure, the highest BCS₁₀₀ is achieved by the application of lignin into ZOAB roads, amounting to 22.5 kg C per ton of asphalt. This higher value is due to the shorter lifetime, that leads to more asphalt being produced and stored in foundations over a 100-years period. SMA roads have the second highest BCS₁₀₀ value (18.7 kg C/m²), due to the higher lignin content of the top layer. The value for AC is the lowest (14.5 kg C/m²), since it has a long lifetime and not as much lignin content as SMA or ZOAB (whose top layer is thicker than in the other asphalts). Conversely, the c-BCS values are highest for lignin SMA (3.71 kg C/m²), followed by lignin AC (3.63 kg C/m²). Since c-BCS accounts only for the circular carbon storage, lignin ZOAB is no longer preferable, since a significant part of the biogenic carbon was stored in low value applications, which has no value in a CE. For this reason, lignin AC with its higher recycled content scores better in c-BCS than in BCS₁₀₀. Nevertheless, the higher biogenic content of lignin SMA leads to a higher c-BCS value.

3.3. Comparison of results for the circularity, environmental and economic analyses

A summary of the results for each indicator calculated in the circularity analysis and the LCC and LCA analyses performed in previous studies (Moretti et al., 2022; van Veen et al., 2021) are included in Table 5. The table includes a first section (in blue) with indicators representing desirable attributes, i.e. the higher the better. These indicators are the MCI, and the BCS series. The rest of the indicators are marked with a red background and represent undesirable impacts, i.e. the higher the worse. These are: climate change impact (in kg CO₂/m²/yr) with and without biogenic carbon credits, abiotic depletion (i.e. depletion of minerals, in kg Sb eq/m²/yr) and the ECI (in €/m²/yr). Since the three

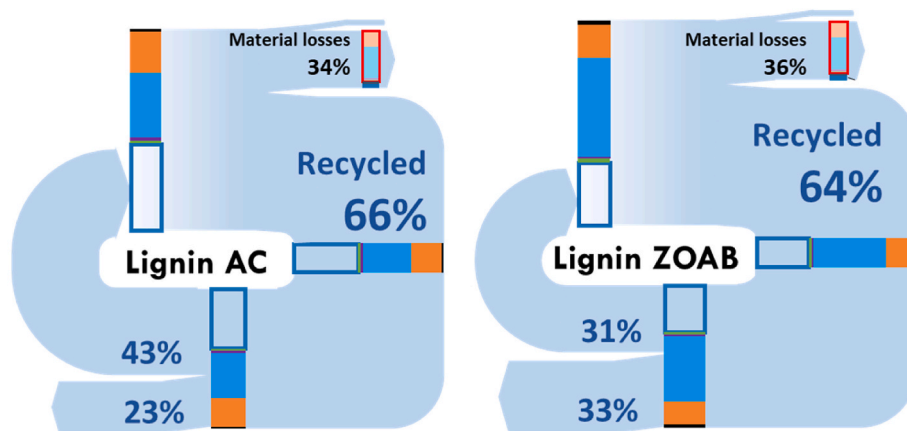


Fig. 2. Material flows diagram for lignin-based AC and ZOAB roads.

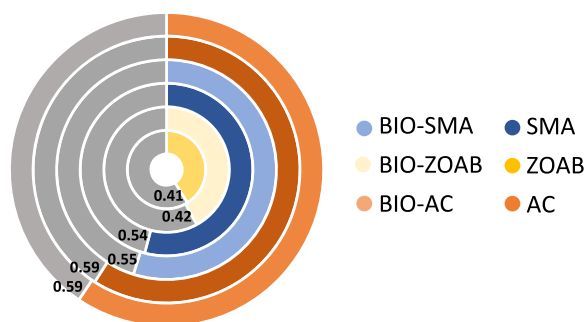


Fig. 3. Comparison of MCI results per road type and scenario.

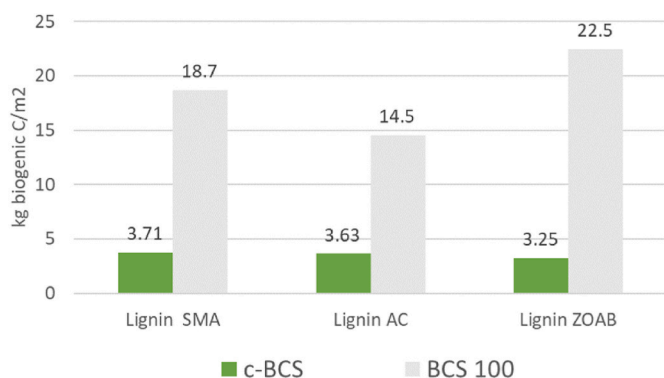


Fig. 4. Results for c-BCS and BCS100 per m² of lignin-based road.

Table 4
Values used for the c-BCS and BCS100 calculations for the lignin-based SMA, AC and ZOAB.

	SMA	AC	ZOAB	Term used by BCS ₁₀₀ indicator	Term used by c-BCS indicator
∑ CCI (kg/m ²)	9.274	7.872	10.06	✓	✓
∑ LUi (kg/m ²)	0	0	0	✓	✓
∑ LRi (kg/m ²)	0	0	0	✓	✓
∑ PSi (kg/m ²)	4.028	2.858	3.915	✓	✓
LT (years)	30	30	24	✓	✓
Rri	73%	73%	73%	✓	✓
∑ CCri (kg/m ²)	2.359	2.397	2.290	✓	✓
BCS ₁₀₀	18.7	14.5	22.5		
c-BCS	3.71	3.63	3.25		

asphalt roads have different lifetimes, the impact results are provided per year of use to allow for a fair comparison (functional unit defined as 1 m² of road used over 1 year).

As observed in the results, the lignin AC road obtains the highest circularity value and lowest environmental impacts, as indicated by every indicator except for both BCS indicators and climate change (including biogenic carbon). From a climate change perspective, the lignin SMA road scores better than the lignin AC road (4% lower impact for SMA than AC) due to the higher lignin content, which leads to higher biogenic carbon storage. Despite having a higher impact in GWP than the lignin SMA road, the lignin AC road still has the lowest ECI score (weighted environmental impact). This is mostly due to lower toxicity impacts (see Moretti et al., 2022 for more information). Concerning life cycle costs, the bitumen AC road is the cheapest one, followed by the lignin AC road. This is also the case when adding the externalities to the internal costs (LCC + ECI).

The LCA results on climate change indicated the best performance for the lignin SMA road (lowest impact in climate change), followed closely by the lignin AC road. This is due to the higher biogenic carbon storage in the SMA road, as evidenced by the results of the c-BCS indicator that points to lignin-SMA as the application with the highest BCS value. When looking at the climate change result that excludes biogenic carbon credits, the lignin AC road has the best result. This is due to the higher recycled content of the AC road, which leads to a lower impact in climate change than the other options.

The lignin ZOAB road is only scoring better when considering the BCS₁₀₀, which implies that when considering multiple recycling loops, the higher losses into foundations of ZOAB would lead to a higher permanent biogenic carbon storage for the non-recycled materials. However, permanent biomass/biogenic carbon storage without further utilisation or value-added goes against the CE principles. An indicator such as the BCS₁₀₀ would be suitable under a scenario where unlimited lignin is available, and not using the lignin would lead to its decay and corresponding GHG emissions. In such a hypothetical scenario, it would be beneficial to use the lignin once and then sequester biogenic C as long as possible. However, that does not correspond with the current situation where lignin is a valuable and restricted resource. For that reason, c-BCS give a better understanding of the combined benefits for climate change and CE. When looking at the c-BCS results, we can see how lignin SMA still scores better than the other applications, followed closely by lignin AC (11% difference).

Concluding, both bio-SMA and bio-AC seem to be promising alternatives to bitumen-based asphalt based on circularity, climate, environmental impacts and overall cost aspects. ZOAB, on the other hand, scores significantly worse in all but the BCS₁₀₀ category. For these reasons, no production of lignin-based asphalt is currently envisioned in the Netherlands.

Table 5

Summary of results for the economic, environmental and circularity assessment of lignin-based (Vertoro lignin, biomass energy) and bitumen-based roads. Blue cells contain positive indicators (the higher the better) and red cells contain negative indicators (the higher the worse).

	SMA road		AC road		ZOAB road	
	Lignin-based	Bitumen-based	Lignin-based	Bitumen-based	Lignin-based	Bitumen-based
Material Circularity Index (MCI)	0.549	0.541	0.594	0.591	0.420	0.410
BCS ₁₀₀ , kg C/m ²	18.7	0.44	14.5	0	22.5	0
c-BCS, kg C/m ²	3.71	0	3.63	0	3.25	0
LCC*, cent€/m ² /yr	89	84	79	74	109	100
Climate change*†, kg CO ₂ eq/ m ² /yr	0.45	0.80	0.47	0.78	0.65	1.05
Climate change w/o BC*†, kg CO ₂ eq/ m ² /yr	1.78	2.07	1.66	2.01	2.44	2.59
Abiotic depletion*†, Kg Sb eq/ m ² /yr	0.003	0.005	0.002	0.004	0.004	0.005
ECI*, cent€/ m ² /yr	9.4	10.1	8.8	9.6	13.2	13.5
LCC + ECI, €/ m ² /yr	0.98	0.94	0.88	0.83	1.22	1.13

*Results are provided for the biomass scenario as defined in (Moretti et al., 2022). Moretti et al. (2022) calculated the ECI value by assuming the climate change results with BC.

†Calculated with the impact assessment method CML-IA baseline V3.06 / EU25

4. Discussion

4.1. Methodological insights from the case study

This study used different metrics to evaluate and measure the circularity of the cases under study to add relevant information to the results obtained through the LCA and LCC. The calculation of circularity indicators is usually faster and easier than performing an LCA. In addition, the indicators here proposed minimise variability in normative choices related to multifunctionality and biogenic carbon accounting. However, they still give no indication on emissions to the environment. In particular, the MCI neither indicates which alternative consumes more resources overall nor considers scarcity or value of resources, but only to what extent the materials are being recirculated.

The abiotic depletion indicator from LCA is based on resource scarcityOF² and absolute consumption notions and, therefore, is a more complete impact indicator than MCI. The case under study shows the lowest resource depletion for the AC road, followed by the SMA road, indicating good alignment with the MCI score. However, LCA's abiotic depletion is more difficult to obtain and subject to methodological variability when dealing with multifunctionality issues. In addition, it does not directly represent to what extent circular strategies are implemented or the consumption of biotic resources.

The new indicator c-BCS is a complementary indicator to climate change LCA's indicator that does not favour linear biobased strategies such as landfilling or "permanent" low-value applications. By providing climate change impact results and a separate indicator for c-BCS, we can consider carbon storage in the decision-making process while avoiding favouring decisions that do not maximise the value of biobased materials. Therefore, it combines notions of climate change and circularity, aligning both goals. However, this approach does not address the issue of assigning credits from biogenic carbon storage when calculating the

impacts in climate change. Whereas it is generally understood that storing biogenic carbon leads to climate benefits, the extent of this benefit and how it relates to the time of emissions is highly debated. For instance, current guidelines for the PEF method disregard credits associated with temporary and permanent carbon storage and delayed emissions due to the absence of expert-based consensus (European Commission, 2021). In that respect, the c-BCS indicator provides an alternative to consider the potential benefits of carbon storage in the decision-making process. Nevertheless, this approach is not suitable for carbon footprint accounting. In such a case, a method to calculate the net balance of biogenic and fossil carbon uptake, storage and emission, remains necessary.

4.2. The role of circularity metrics in decision-making processes

A wide range of circularity metrics has been suggested over the last years in an effort to understand how well circular strategies are and can be implemented in current human activities. Recent reviews on circularity metrics have identified at least 63 different metrics applied at different levels (micro, meso, macro) (Parchomenko et al., 2019). Due to the CE's diverse interpretations, these metrics differ in their primary aim and type of insights provided. Many newly developed metrics are suitable for representing specific aspects of the circular economy, e.g., to what extent a product system closes material loops at the end of life or extends the lifetime of products providing higher value with less resource use.

The Life Cycle Assessment (LCA) methodology has been criticised for being too complex to be fully applied by companies, and at the same time, not being able to model open recycling loops consistently (Corona et al., 2019). However, it is also considered the best framework to quantify the potential environmental impacts (such as climate change impacts) of products and services (European Commission, 2003, 2013) and is currently the most used methodology to assess the impacts of circular products and services (Corona et al., 2019). While LCA does not advocate for specific material efficiency strategies, it provides a scientific assessment framework to understand the sustainability implications

² The indicator is calculated following the CML IA method whose characterization factors are calculated considering ultimate reserves and extraction rates of each mineral with respect to antimony.

of different options or strategies (Peña et al., 2021). On the other hand, newly developed metrics focused on material circularity can indicate to what extent circular strategies are applied (e.g., the MCI) but are unable to indicate the effects on energy consumption, climate change, material scarcity, or other environmental impacts. It has also been argued that making decisions based only on material circularity metrics could lead to burden-shifting and increased environmental impacts (Corona et al., 2019). As shown with the asphalt case study, the option with the highest material circularity is not necessarily the option with less impact on climate change.

The presence of many different metrics in the literature claiming to measure circularity hinders the development of a common and consistent framework for circularity analysis. Nevertheless, such variety has the advantage of allowing for customised analyses adapted to the CE intervention's goal and the product or activity at stake. As evidenced with the asphalt case study, choosing a combination of metrics based on a similar methodological framework (and thus, allowing for consistency within the analysis) but focused on different CE and sustainability aspects can be a comprehensive way to obtain insights and understand trade-offs between different perspectives or dimensions of human activities, leading to truly sustainable decision making.

Combining LCA indicators with material circularity metrics based on life cycle thinking allows the circularity and environmental assessment of a product or service, leading to a better informed sustainable decision making (Lonca et al., 2018; Pauliuk, 2018; Sassanelli et al., 2019). This approach is also followed by recent efforts in the construction sector to define a common circularity measurement framework (Platform CB'23, 2020).

5. Conclusions

The circularity assessment indicated that lignin-based asphalt (as developed in the CHAPLIN XL project) has higher circularity than the bitumen-based counterpart, although the circularity gains are relatively low considering the small share of lignin in the total weight of asphalt. This outcome is also a consequence of the metric used since the MCI assigns the same level of relevance to high-value materials (such as lignin or bitumen) than to low value and abundant materials (such as stone and sand). The BCS analysis indicated that the least circular lignin-based alternative sequesters the highest amount of carbon in the long term due to permanent storage as foundations. Despite this trade-off of impacts, the results from the newly developed indicator allowed to align both climate and circularity goals, guiding policymakers and industry actors to the implementation of circular biobased strategies where the value of biobased materials is optimised.

The assessment also pointed out the most promising strategies to increase the circularity of asphalt roads, independently of their biobased or fossil nature. These involve extending the lifetime of current roads and increasing the recycled content of the asphalt mixture. These strategies would increase not only the circularity but also decrease the environmental impacts of the road system. The biogenic carbon storage assessment and the LCA results indicated that most of the sustainability gains in lignin-based asphalts lay on their lower impact on climate change. This is due to both lower GHG emissions during production and the ability to store carbon (in high-value applications). This ability would be further increased if less asphalt is used for low-value applications (such as foundations) and used for pavements where the properties of lignin are kept.

Eventually, combining or pairing LCA indicators with material or substance circularity metrics based on life cycle thinking has the potential to provide a holistic picture of the circularity and environmental sustainability of a product or service, allowing for an understanding of trade-offs, and thus leading to a better-informed sustainable decision making. Nevertheless, choosing the right combination of metrics for each case study is challenging, and the metrics used for the case here presented may not be comprehensive for other types of products or

technologies. Future research steps should focus on testing the metrics with different case studies.

CRedit authorship contribution statement

B. Corona: Conceptualization, Methodology, Investigation, Writing – original draft. **R. Hoefnagels:** Conceptualization, Funding acquisition, Writing – review & editing. **I. Vural Gürsel:** Conceptualization, Writing – review & editing. **C. Moretti:** Investigation, Writing – review & editing. **M. van Veen:** Investigation. **M. Junginger:** Funding acquisition, Project administration, Writing – review & editing.

Declaration of competing interest

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.

Data availability

Data will be made available on request.

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