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Kraft lignin as a bio-based ingredient for Dutch asphalts: An attributional LCA

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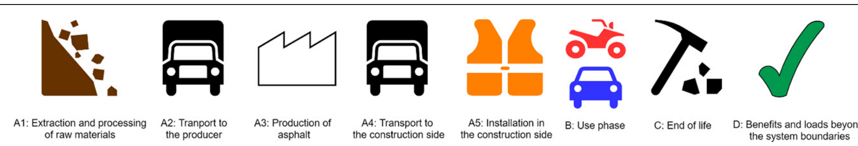
HIGHLIGHTS

- An environmental life cycle assessment of Dutch bio-based asphalts is presented.
- Kraft lignin was considered as a bio-based ingredient for asphalts.
- A comparison was conducted for various types of asphalts and kraft lignins.
- Climate change benefits can be achieved using lignin-based asphalts to replace current Dutch asphalts.

GRAPHICAL ABSTRACT



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ABSTRACT

In the last decade, lignin has received much attention as a feedstock to produce bio-based products. This study investigates the potential benefits of using lignin to mitigate the environmental impact of the road construction sector. An environmental life cycle assessment (LCA) of various top-layer bio-based asphalts using kraft lignin was conducted. From a cradle-to-grave perspective, lignin-based asphalts were compared with conventional asphalts.

The results of the LCA revealed that the climate change impact of lignin-based asphalts could be 30–75% lower than conventional asphalts. For the other ten impact categories, trade-offs were observed. Overall, two key factors to make the environmental impact of lignin-based asphalts lower than conventional asphalts are 1) increasing the amount of bitumen-substituted and 2) using low-grade biomass fuels for process steam in the pulp mill. The substitution of weak filler with lignin was beneficial only for climate change and could lead to a worse overall environmental performance than conventional asphalts. Similarly, higher environmental impacts for lignin-based asphalts could be obtained if the pulp mill consumed natural gas to complete the energy balance to replace the part of the black liquor from which lignin is extracted.

This study also includes an in-depth discussion on methodological choices such as the allocation methods for lignin, functional units, and asphalt layers considered. We believe that such a methodological discussion could be helpful to support future Product Category Rules for asphalt mixtures.

1. Introduction

10% of the GHG emissions of the transportation sector are caused by the construction of roads (Sollazzo et al., 2020). This amount corresponds to more than 5% of the total greenhouse gas (GHG) emissions

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generated in the European Union (EU) (Sustain Euro Road, 2021). Picturing, this amount is twice as much as the impact contributed by the aviation sector (Sustain Euro Road, 2021). To contribute to the EU climate change reduction targets, the Netherlands aims to make this sector carbon-neutral by 2030 (Bizarro et al., 2021). Some circular economy practices, such as the recovery and recycling of deteriorated asphalt, are already well-established all around Europe (Giani et al., 2015; Pantini et al., 2018). However, such practices alone are not sufficient to achieve carbon neutrality. To achieve such a goal, it is necessary to couple asphalt recycling with alternative construction and production methods as well as alternative (e.g. bio-based) materials and renewable energy sources (Giani et al., 2015; Tokede et al., 2020).

Projects in the construction sector are often assigned via public tenders. The environmental aspects are becoming more often part of the public tenders in the EU member states (Ecochain, 2019; Moretti et al., 2017b). With this purpose, the results of environmental Life Cycle Assessments (LCAs) have recently been increasingly used in public tenders of infrastructure projects such as roads, airports and railways (Ecochain, 2019; Moretti et al., 2017b). LCA, which is standardized by ISO (ISO, 2006a, 2006b), is acknowledged as a rigorous methodology and should provide unbiased and comparable environmental calculations. Hence, combining properly the environmental impacts calculated using LCA with project costs should avoid that low-cost materials with high environmental burdens are selected in investments using public funding (Moretti et al., 2017b). With this purpose, tenders in the Dutch construction sector include an environmental cost indicator that simplifies and unites various environmental impacts into a single monetary value score representing the avoided damage cost or shadow cost (Ecochain, 2019; Schwarz et al., 2020).

Bitumen production is one of the most important environmental burdens of asphalts (Schwarz et al., 2020; Tokede et al., 2020). For this reason, some renewable alternatives to bitumen such as plant-based binders, municipal wastes, and live-stock manures are currently under investigation (He et al., 2019; Tokede et al., 2020). Lignin, a bio-based by-product of pulp mills and lignocellulosic biorefineries, has received attention as a renewable binder to replace bitumen in asphalt mixtures (Moretti et al., 2021; Tokede et al., 2020). Several roads using lignin-based asphalts have been paved in the Netherlands in the last five years (Biobaseddelta, 2020). However, there is a lack of clear evidence supporting the environmental benefits of producing lignin-based asphalts. To our knowledge, there is only one peer-reviewed LCA (Tokede et al., 2020) of lignin-based asphalts. The results of this LCA suggest that lignin-based asphalts with 25% replacement of the bitumen binder with lignin allow almost a 6% reduction in asphalt production GHG emissions (Tokede et al., 2020). However, this LCA study (Tokede et al., 2020) presents some research gaps and leaves space for further research. For instance, the use- and end of life- phases were not considered in the study, and it only focused on climate change impacts. Moreover, the life cycle inventory of lignin production lacks transparency for what concerns data, allocation methods and biogenic carbon accounting, which are all crucial aspects to interpret the results of an LCA of a lignin-based application (Moretti et al., 2021).

Our study presents the cradle-to-grave LCA of various top-layer lignin-based asphalts using kraft lignin, assessing 11 impact categories together with an environmental cost indicator. The cradle-to-gate (pulp mill gate) environmental impact of kraft lignin is also discussed. Since the Netherlands is the geographic scope of this LCA, beyond ISO 14040 and ISO 14044 (ISO, 2006a, 2006b) and EN 15804 (CEN, 2013), the Dutch Product Category Rules (NL-PCRs) (Keijzer et al., 2020), SBK bepalingsmethode 3.0 (Bouwkwaliteit, 2019) and Dutch LCA asphalt sector report (Schwarz et al., 2020) were used as reference documents to conduct the LCA. Nevertheless, even following the best LCA practices recommended by these methodological documents, multiple sources of methodological uncertainty exist and the results of LCAs should be carefully interpreted (Cherubini et al., 2018; Moretti et al., 2020a; Reap et al., 2008). For this reason, the effect of the allocation methods, functional units and product systems' definition was broadly discussed through sensitivity analyses.

2. Materials and methods

2.1. Goal and scope definition

This study aims to assess and compare the environmental impact of lignin-based asphalts with conventional asphalts. The intended audience of this study is made of technology developers and researchers working on lignin-based applications. Given the valuable research conducted concerning LCA methodological implications presented in the Discussion section, we believe that this study can be relevant for the LCA community involved in the construction sector. Moreover, the results of this study might be of interest for stakeholders that look for options to reduce the carbon footprint of road construction, e.g. policymakers, the road construction sector. The geographic scope is the Netherlands, and the temporal scope is 5–10 years from now.

In comparative LCAs, a proper functional unit, i.e. the reference unit of comparison, should be carefully selected. Hence, how well a product system fulfills a specific function should be accounted for in the functional unit. In particular, asphalts are required to maintain multiple rheological performances, especially in terms of stiffness/deformation resistance but also workability, durability, permeability and resistance to possible damages caused by moisture or fuels and oils (Giustozzi et al., 2015; Martinez-Arguelles et al., 2015; Tokede et al., 2020). Moreover, one of the key mechanisms of failure of asphalts affecting their lifetimes is the occurrence of fatigue cracks (predominately) within the binder of asphalt (Borghi et al., 2017; Brovelli et al., 2014; Tokede et al., 2020). For this reason, various compositions for lignin-based asphalts were tested via standardized methodologies in the laboratory by Asfalt Kennis Centrum (project partner) to guarantee similar functional properties and performances to conventional asphalts. Empirical and functional tests were conducted according to respective documents of EN 13108 series (1 and 20) and EN 12697 series (8-12-23-24-25-26-31-33-35). The empirical tests included run on gyrator samples, hollow space percentage and water sensitivity and were conducted for all asphalt types investigated. The functional tests included performing on gyrator test pieces and rolled test plates, tri-axial test for rutting resistance, stiffness on test beam sawn from the rolled test plates, fatigue on the same beams. The functional tests were applicable only to asphalt concretes. Using these compositions, based on the goal of the study, a functional unit of 1 tonne (t) of top layer asphalt was selected in line with the reference unit used by the Dutch LCA asphalt sector report (Schwarz et al., 2020). Adopting a different functional unit representing the whole asphalt product made of several layers is also considered in Section 4.2. The baseline calculations refer to stone mastic asphalts (SMAs). Asphalt concretes (ACs) and porous asphalts (ZOABs with the Dutch acronym) were also assessed as alternative product systems. The LCA has a cradle to grave scope. Fig. 1 illustrates the sub-division into life cycle stages with coding according to the Dutch reference documents (Keijzer et al., 2020; Schwarz et al., 2020). As indicated for environmental product declarations based on the European standard EN 15804 used in the construction sector (Durão et al., 2020), this LCA is conducted using attributional modeling. By attributional modeling, all the environmental burdens of all processes involved in the life cycle were accounted for with representative average data (Moretti et al., 2020a; Pelletier et al., 2015). As mentioned in the introduction, the cradle-to-gate environmental impact of kraft lignin is also discussed in this study. For such a product, a functional unit of 1 kg of dry lignin was adopted.

The impact categories and respective weighting factors (see Table 1) were selected based on the calculation rules of SBK bepalingsmethode 3.0, which is based on the recommendations from EN15804 + A2 (2019) (CEN, 2013). In particular, the weighting factors used in the Netherlands are the so-called MKI weighs (Bouwkwaliteit, 2019),

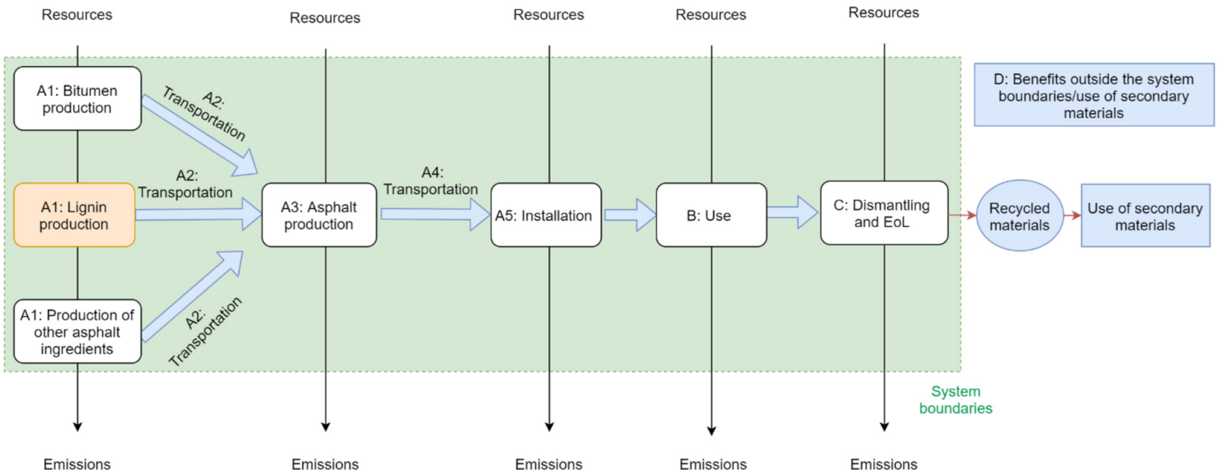

Fig. 1. Flow chart of lignin-based asphalt with cradle-to-grave life cycle stages based on (Schwarz et al., 2020). The unit process named "A1:Lignin production" starts from the cradle to the gate of the pulp mill where lignin is delivered.

which are based on the shadow price method (internationally often referred to as environmental cost indicator). The shadow price corresponds to the cost of the preventive measures for the government to avoid that environmental impact (Schwarz et al., 2020).

2.2. Life cycle inventory

2.2.1. Extraction and processing of raw materials (A1)

The compositions of the asphalts compared were determined with the objective to have similar functionality performances. For this reason, the same type of asphalt (SMAs, ACs or ZOABs) with lignin or without lignin was assumed to have the same lifetime. The compositions of lignin-based and conventional asphalts are reported in Table 2. From this table, it is possible to notice that lignin replaces bitumen and a fraction of weak filler. For ZOABs, lignin replaces only weak filler and not bitumen. Table 2 also reports the background inventory data. As for the Dutch LCA asphalt sector report, the background data were retrieved from the ecoinvent library named "Allocation cut-off by classification" and the Dutch National Environmental Database (Milieudatabase, 2020), which contains various datasets representing the environmental performance of buildings in the Dutch context. In particular, the ecoinvent system model "Allocation, cut-off by classification" is an attributional database where the primary (first) production of materials is always allocated to the primary user of a material (ecoinvent, 2021). Accordingly, if "a material is recycled, the primary producer does not receive any credit for the provision of any recyclable materials", and the recyclable materials are available burden-free to recycling processes and bear only the impacts of the recycling processes (ecoinvent, 2021). However, in the Dutch asphalt sector, the credits generated outside the system boundaries are allocated to the final environmental impact of the asphalt (see Section 2.2.6).

Table 1
Environmental impact categories and weighting factors (Bouwkwaliteit, 2019).

Impact category	Unit	Weighting factor
Abiotic depletion	kg Sb eq	0.16 €/kg
Abiotic depletion (fossil fuels)	MJ	7.7E−05 €/MJ
Global warming (GWP100a)	kg CO_2 eq	0.05 €/kg
Ozone layer depletion (ODP)	kg CFC-11 eq	30.0 €/kg
Human toxicity	kg 1,4-DB eq	0.09 €/kg
Freshwater aquatic ecotoxicity	kg 1,4-DB eq	0.03 €/kg
Marine aquatic ecotoxicity	kg 1,4-DB eq	0.0001 €/kg
Terrestrial ecotoxicity	kg 1,4-DB eq	0.06 €/kg
Photochemical oxidation	kg C_2H_4 eq	2.0 €/kg
Acidification	kg SO_2 eq	4.0 €/kg
Eutrophication	kg PO_4--- eq	9.0 €/kg

2.2.1.1. Lignins. There are various technologies and feedstocks from which lignin can be derived from pulp mills or lignocellulosic biorefineries (Moretti et al., 2021). The kraft process is the dominant process of the pulping industry (Bajwa et al., 2019). With the kraft process, wood is converted into wood pulp, which is used in the production of paper. Black liquor is a by-product of the wood pulping process in a pulp mill. The major component of black liquor is lignin. The black liquor is usually burnt in a recovery boiler to produce internal process energy (Bernier et al., 2013). Alternatively, kraft lignin can be obtained from black liquor through precipitation and separation processes; for example by the LignoBoost process (Tomani, 2010).

In our LCA model, kraft lignin is separated from the black liquor and used in asphalts. For the production of kraft lignin, two studies were considered as main data sources given the unavailability of primary data: Culbertson et al. (2016) & Bernier et al. (2013). The background data can be found in the Appendix. In these two studies, the precipitation step is carried out by injecting liquid carbon dioxide in the black liquor together with sulfuric acid (Bernier et al., 2013; Culbertson et al., 2016). Three lignin production "scenarios" were modeled, one based on Culbertson et al. (2016) and two based on Bernier et al. (2013):

- "kraft1-BIOM" was modeled based on the inventory data from Culbertson et al. (2016). The pulp mill modeled by Culbertson et al. is equipped with a hog fuel boiler, has an internal power plant that sells an electricity surplus to the electric grid and uses natural gas only for the lignin extraction process.
- "kraft2-NG" was modeled based on the pulp mill modeled by (Bernier et al., 2013). The electricity is instead supplied to the pulp mill by the grid and natural gas is used to produce the part of the energy that is no longer produced from burning black liquor (due to the extracting of lignin). Since the consumption of natural gas (NG) to produce the kraft lignin is much higher in Bernier et al. (2013) than Culbertson et al. (2016), the lignin modeled based on Bernier et al. (2013) will be referred to as "kraft2-NG".
- "kraft2-BIOM" was modeled as well based on the pulp mill modeled by Bernier et al. (2013). According to Bernier et al., the same pulp mill could also potentially use hog fuel (chips of wood bark) instead of natural gas to compensate for the loss of steam production from the recovery boiler. Since the heat source is a key factor affecting the environmental impact of lignin (Moretti et al., 2021), this third

Table 2
Composition of 1 t of top layer asphalt based on industrial partner's primary data and background inventory datasets used.

Raw material (amounts expressed in kg)	B-SMA	C-SMA	B-AC	C-AC	B-ZOAB	C-ZOAB	Inventory dataset
Recycled content	0	0	288	300	0	0	Burden-free
Cellulose fiber	3	3	0	0	0	0	SBK_Cellulosevezels (Milieudatabase, 2020)
Bitumen	44.5	65	21.1	40	43	43	ESU NL-PCR bitumen (Schwarz et al., 2020). The selection of the bitumen dataset is discussed in Section 4.1.2
Crusher sand	88	75	154	171	106	106	Sand {CH}| gravel and quarry operation | Cut-off from Ecoinvent 3.6
Natural sand	88	75	76	57	0	0	SBK 296 Industriezand (Milieudatabase, 2020)
Crushed stone (Morene)	0	0	411	410	811	811	Gravel, crushed {CH}| production | Cut-off from Ecoinvent 3.6
Crushed stone (Porfier)	700	710	0	0	0	0	Gravel, crushed {CH}| production | Cut-off from Ecoinvent 3.6
Weak filler	35	72	23	22	0	40	Limestone, crushed, washed {CH}| production | Cut-off from Ecoinvent 3.6
Lignin	40	0	24	0	40	0	Modeled based on literature sources (see Section 2.2.1.1)
Linseed oil	1.5	0	2.9	0	0	0	Linseed seed, at farm {CH}| linseed seed production, at farm | Cut-off from Ecoinvent 3.6

Lifetimes = 15 years for SMAs and ACs and 12 years for ZOABs. B = bio-based, C = conventional.

scenario represents the use of hog fuel instead of natural gas to compensate the loss of steam from the recovery boiler.

Details on what and how inventory data retrieved from the studies mentioned above can be found in Appendix. The lack of recent primary data is a limitation of our LCA study.

2.2.1.1.1. Allocating the environmental burdens to lignin. Since the production of kraft lignin is the result of a multifunctional process, it is necessary to apply an allocation method to apportion the environmental burdens of the pulp mill between lignin and pulp. In the LCA literature, it is acknowledged that the environmental impact of kraft lignin is importantly affected by the allocation method applied between pulp and lignin (Hermansson et al., 2020; Moretti et al., 2021). For this reason, how the allocation method was selected based on the reference documents is here extensively illustrated. The Dutch Product Category Rules (Keijzer et al., 2020) recommends following the directions from EN 15804 (CEN, 2013).

EN 15804:2012+A2:2019 specifies that the allocation between the co-products should be avoided by subdivision every time possible. The other typical option to avoid allocation i.e. system expansion is not allowed by EN 15804:2012+A2:2019. This is a common statement in rules for environmental product declarations to avoid that a substitution approach (one of the methods to perform system expansion) is used as a system expansion method in attributional LCA modeling (EPD, 2019, 2016). In fact, the use of the substitution as a system expansion method in attributional LCAs is considered misconduct by many LCA practitioners and often leads to misleading interpretations or erroneous results (Majeau-Bettez et al., 2018; Pelletier et al., 2015; Sandin et al., 2015). If subdivision is not possible or data for sub-processes are not available, the allocation can be based on the underlying physical causality relationship i.e. reflecting "the way in which the inputs and outputs are changed by quantitative changes in the products or functions delivered by the system" (ISO, 2006b).

This type of allocation is often referred to as "physical causality allocation (Ahlgren et al., 2015; Mackenzie et al., 2017; Moretti et al., 2020a). A physical causality allocation relies on the mathematical modeling of the changes in operating conditions of the process under investigation to establish a physical causality relationship between functional units.[1] Accordingly, the PCR 2019:14 v.1.0 for construction products (EPD, 2019) and ISO 21930:2017 (ISO, 2017) i.e. the ISO core rules for environmental product declarations of construction products remark that physical causality can be established only if "each of the co-products can be produced without the other(s) or the ratio of the co-products typically varies in normal production", which is the only case when allocation by physical causality can be modeled (Azapagic and Clift, 2000, 1999; Mackenzie et al., 2017; Moretti et al., 2020a). According to EN 15804, allocation based on simple physical properties as mass can be used as a proxy for physical causality only if the difference in revenue from the co-products is low (defined as max. 25% difference) (CEN, 2013). In all other cases, the allocation shall be based on economic values (CEN, 2013).

Accordingly, the pulp mill modeled by Culbertson et al. (2016) generating kraft1-BIOM was subdivided as much as possible. In this way, the lignin extraction process itself was not allocated to the pulp and bleaching was not allocated to lignin. For the other processes, keeping the pulp output constant and extracting kraft lignin leads to a change in the ratio between pulp and the surplus of electricity output produced (and a minor change in the soap output). So, allocation by physical causality cannot be established. Moreover, since the difference in revenue from the co-products is not "low", economic allocation was used. To calculate the economic allocation share for lignin, the following prices were used: 535 €/t for kraft lignin (Moretti et al., 2021), 788 €/t for pulp (Indexmundi, 2020), 111 €/t for soap and 0.08 €/kWh for the surplus of electricity (EUROSTAT, 2020). By using these prices, an allocation factor (A_L) of 7.3% to lignin was calculated. Two sensitivity analyses varying the price of kraft lignin and using mass allocation can be found in Section 4.1.1.

In the case of the kraft2-NG lignin and kraft2-BIOM, electricity is supplied from the grid. Hence, a physical causality allocation was applied for this pulp mill. This type of allocation is based on the "physical causal relationships between burdens and functional outputs, which in turn requires a model of the system behavior" (Azapagic and Clift, 1998). Taking the words of Azapagic and Clift, who are the authors that more than others have emphasized the importance of this allocation method (Moretti et al., 2020a), "when the causal relationships are represented by a model which describes the real behavior of the product system, the model can be used to allocate burdens between different functions by exploring how the burdens change when the quantity of one function is changed with the quantities of all the other functions kept constant" (Azapagic and Clift, 1998). In order of magnitude, there are three types of changes that can be modeled, i.e. marginal-, incremental- and average (Azapagic and Clift, 1998; Moretti et al., 2020a). Based on the approach adopted by the data source (Bernier et al., 2013), average changes were used as the base for the allocation. Average changes are substantial changes such as eliminating or adding a functional output (Azapagic and Clift, 1998; Moretti et al., 2020a). In Bernier et al. (2013), this type of allocation is "performed by comparing emissions with and without lignin recovery for the same mill and assigning the differences to lignin".

Both allocation methods preserve the additivity principle of environmental impacts, one of the key aspects of attributional modeling (Majeau-Bettez et al., 2018; Moretti et al., 2020a). Such a principle for the two allocation methods applied is expressed by Eq. (1).

[1] Sometimes, such a type of relationship can be reflected by a simple physical parameter for volume allocation in a truck transporting empty packaging or mass allocation in a truck transporting full packaging (ISO, 2012).

$$I_t = I_p + I_L = I_t * A_{pp} + I_t * A_{Lp} = I_t * A_{pe} + I_t * A_{le} \quad (1)$$

where:

- I_t is the total impact of the pulp mill to produce both lignin and pulp
- I_p is the impact of the pulp mill if producing only pulp and not lignin
- I_L is the additional impact generated by the pulp mill when lignin is extracted compared to when pulp only is produced
- A_{pp} is the physical causality allocation factor for pulp ($=I_p/I_t$)
- A_{Lp} is the physical causality allocation factor for lignin ($=I_L/I_t$)
- A_{pe} is the economic allocation factor for pulp
- A_{le} is the economic allocation factor for lignin.

2.2.2. Transport to the producer (A2) and production of asphalt (A3)

The transportation distances were mostly based on primary information from the asphalt industry. For the transportation of other materials for which primary information was not available, standard values from the Dutch LCA asphalt sector report (Schwarz et al., 2020) were used. A summary can be found in Table 3.

Based on primary data collected from one industrial producer (Roelofs Groep), natural gas consumption during the production phase differs between lignin-based and conventional asphalts. Each t of lignin-based asphalts require on average 5.3 Nm^3 of natural gas (NG), while conventional asphalts require on average 7.5 Nm^3 of natural gas. According to Schwarz et al. (2020), asphalts with recycled material (PR) use more natural gas due to the overheating stage. The overheating stage is responsible for 6% of the total natural gas use of asphalt production, and the overheating of 1 kg of PR requires 0.0015 Nm^3 of natural gas (Schwarz et al., 2020). Therefore, the natural gas usage of each asphalt type can be calculated by using Eq. (2):

$$\text{NG usage}\left[Nm^3\right] = \text{average NG usage}\left[Nm^3\right] * (1\text{–}0.06) + 0.0015 * \text{PR [kg]} \quad (2)$$

Filling Eq. (2), the natural gas requirement is calculated as 5.0 Nm^3 for bio-based SMA and ZOAB and 5.4 Nm^3 for AC. Conventional SMAs and ZOABs require 7.1 $Nm^{3,}$ while ACs require 7.5 Nm^3. Moreover, based on primary data from the same industrial producer, per each tonne of asphalt, on average, 5.6 kWh of electricity and 0.17 l of diesel are needed during the production phase. For electricity, the dataset *Electricity, medium voltage {RER}| market group for | Cut-off* from ecoinvent 3.6 was used updating the shares of electricity per source based on the 2030 EU reference scenario for the Netherlands (Carpos et al., 2016).

In the Dutch LCA asphalt sector report (Schwarz et al., 2020), for conventional asphalts, natural gas and electricity consumptions are 5–15% higher than assumed in our study based on primary data, while diesel consumption is 40% lower. A sensitivity analysis considering such a relatively reasonable variation of data (probably linked to differences in the plant configurations or younger/older plant) was not conducted. On the other hand, in the Results section, the LCA results of lignin-based asphalts were compared with the calculated impact for conventional asphalts and the impacts for that type of asphalt reported in the Dutch LCA asphalt sector report. In this way, not only the possible differences in utility consumptions but also the ones in the assumed compositions (and transportation distances) were considered.

2.2.3. Transport to the construction site (A4) and installation (A5)

The distance for the transportation to the construction site was taken to be 50 km by lorry (*Transport, freight, lorry 16–32 metric ton, EURO4 {RER}| transport, freight, lorry 16–32 metric ton, EURO4 | Cut-off*) based on (Schwarz et al., 2020). During the installation of the product, 0.26 l of diesel (Schwarz et al., 2020) are consumed (*Diesel, burned in building machine {GLO}| market for | Cut-off* from ecoinvent 3.6).

2.2.4. Use phase (B)

The service lifetime has currently no influence on the environmental impact calculations per tonne of asphalt conducted following the Dutch NL-PCR (Schwarz et al., 2020). Based on the Dutch LCA asphalt sector report (Schwarz et al., 2020), the only process from the use phase included in this LCA is the leaching of substances from top layers to the soil under the influence of precipitation, which mostly happens in the first years after construction. For this reason, such an effect is considered independently of the lifetime of the asphalt (Schwarz et al., 2020). The leaching emissions of various types of conventional asphalts determined with laboratory tests of factory samples were retrieved from Schwarz et al. (2020). Based on industrial partners (Asfalt Kennis Centrum and Holding de Vier Ambachten-H4A), there is a piece of the first evidence that the leaching emissions of lignin-based asphalts are in line with the ones from conventional asphalts, but further research is needed. So, for lignin-based asphalts, the same inventory data for leaching were used.

Other environmental burdens during the use phase, such as the ones due to the maintenance of the road surface and repairs, are not included (Schwarz et al., 2020).

The NL-PCR Asphalt (Keijzer et al., 2020) also considers a 10% mass loss of bitumen in top layers due to the effects of erosion (Schwarz et al., 2020). This material loss occurs in nature as an inert material (Schwarz et al., 2020). Similarly to the bitumen binder, it is assumed that a 10% loss due to erosion is also present in lignin and linseed oil.

2.2.5. End of life (C)

The end of life phase of asphalts is made up of several stages. The first stage is the demolition i.e. the removal of the asphalt, which requires an average of 23.0 MJ of diesel (Schwarz et al., 2020). As background data for such a diesel consumption, the process *Diesel, burned in building machine {GLO}| market for | Cut-off* from ecoinvent 3.6 was used. Then, the recovered asphalt is transported for 50 km to the processing site to make road construction mixtures with a percentage of recycled asphalt. For the modeling of this transport step, the process *Transport, freight, lorry 16–32 metric ton, EURO4 {RER}| transport, freight, lorry 16–32 metric ton, EURO4 | Cut-off* from ecoinvent 3.6 was used. At the asphalt plant, the recovered asphalt is processed via breaking followed by blending/mixing, which requires 13.4 MJ of diesel per tonne of asphalt (Schwarz et al., 2020). As background data, also for such a diesel consumption, the process *Diesel, burned in building machine {GLO}| market for | Cut-off* from ecoinvent 3.6 was used. Based on industrial partner

Table 3
Transportation distances assumed for the asphalt mix's ingredients.

	km by lorry truck	km inland shipping	km transoceanic shipping	Data source
Cellulose fiber	177	0	0	Standard value from the Dutch LCA asphalt sector report (Schwarz et al., 2020)
Bitumen	64	0	0	Primary information from the asphalt industry
Crusher sand	7	0	0	Primary information from the asphalt industry
Natural sand	86	0	0	Primary information from the asphalt industry
Crushed stone (Morene)	25	660	0	Standard value from the Dutch LCA asphalt sector report (Schwarz et al., 2020)
Crushed stone (Porfier)	25	53	933	Standard value from the Dutch LCA asphalt sector report (Schwarz et al., 2020)
Weak filler	7	0	0	Primary information from the asphalt industry
Lignin	146	0	1822	Primary information from the asphalt industry
Linseed oil	150	0	0	Primary information from the asphalt industry

information (Holding de Vier Ambachten-H4A), only 1% of the recovered asphalt (at the net of material losses described in Section 2.2.6) ends in a landfill (*Waste concrete {Europe without Switzerland}| treatment of waste concrete, inert material landfill | Cut-off* from ecoinvent 3.6). Consistently with the selection of the ecoinvent library named “Allocation cut-off by classification”, the entire environmental impact of the end of life (C) is apportioned to the primary asphalt. So when an asphalt uses recycled content (secondary material), this material is free of environmental burden in module A1.

2.2.6. Benefits and loads beyond the boundaries (D)

Based on the NL-PCR and Dutch LCA asphalt sector report (Keijzer et al., 2020; Schwarz et al., 2020), module D is “outside the system boundaries” but included in the calculation of the environmental performance of the asphalt. In module D, the benefits resulting from recycling primary raw materials and the loads resulting from the loss of secondary raw materials are accounted. Therefore, module D includes the benefits generated by the avoidance of the extraction (A1) and transport of to the asphalt plant (A2) but also the loads of material that was obtained via recycling from the previous cycle but does not get recycled in the current cycle.

$$m_i\text{credited} = (m_i(1-L_{1i})(1-L_{2i})(1-Q_{1i}-Q_{2i}))-m_R M_{Ri} L_{1i}(1-L_{2i})(1-Q_{1i}-Q_{2i}) \quad (3)$$

Eq. (3) from NL-PCR (Keijzer et al., 2020) shows the mass of each raw material *i* that obtains credits or loads for their A1 and A2 impacts. In particular, the loads are represented only by the second part of the equation where m_R is the mass of recycled content and M_{Ri} is the fraction of raw material *i* in the recycled content. As a derogation from NL-PCR (Keijzer et al., 2020), the mass of the recycled content was assumed to be the same as the one of the top layer asphalt. In particular, based on the compositions assumed (see Table 2), ACs only have recycled content and therefore a small percentage of loads (minor compared to the benefits) using Eq. (3).

According to the NL-PCR (Keijzer et al., 2020), the amount of materials recovered during recycling is calculated considering two types of material losses. The first loss is due to the fraction of asphalt that leaves the asphalt system with no further use (e.g. a small fraction goes to landfill) or reused in a low-value function other than asphalt e.g. towards foundation layers. In the NL-PCR (Keijzer et al., 2020), this fraction accounts for 29% (L_1). Such a figure was based on the results of the investigation conducted by EAPA (2017) and applies to all asphalt layers. The second material loss (L_2) is represented by the 10% loss of bitumen (and lignin and linseed oil by assumption) due to erosion in the use phase of top layers described in Section 2.2.4. Moreover, based on the NL-PCR (Keijzer et al., 2020), two quality losses were also accounted respectively for bitumen quality (4% quality loss Q_1), which was also applied to lignin, and the quality loss of crushed stones (Q_2 = 10% in the top layer and 5% for sub-layers). With respect to anti-drip inhibitors, they remain in asphalt but no longer fulfill any function (Keijzer et al., 2020). So, they do not receive any credit in module D.

2.2.7. Biogenic carbon

Lignin-based asphalts contain two bio-based ingredients, which are lignin and linseed oil. The biogenic carbon content of kraft lignin was measured in the lab as 2.4 kg CO_2eq/kg. The biogenic carbon content of linseed oil was assumed as equivalent to 2.9 kg CO_2eq/kg based on (Peterson and Hustrulid, 1998). One of the main updates to EN 15804:2012 (A2:2019) is that, in the new version, it is specified that “the biogenic carbon content shall not be allocated but always reflect the physical flow” (Durão et al., 2020). Additionally, the NL-PCR (Keijzer et al., 2020) specify that “if the submitter can demonstrate based on tests and other technical evidence that the CO_2 is permanently stored in the asphalt product, the CO_2 stored could be included in the calculation of the CO_2 impacts”.

The biogenic carbon from the bio-based inputs that is lost as an inert material (or ends to landfill) is assumed not to biodegrade since lignin does not biodegrade under landfill anaerobic conditions (EREF, 2014; Khan and Ahring, 2019) and is mixed with bitumen in the asphalt matrix. So, it can be considered permanently stored during the life cycle due to losses/landfill since it is expected to remain stored for 100 years. Moreover, the lignin and linseed oil contained in the asphalt that is recycled (the part that is not lost) will go to sub-layers and over a time horizon of 100 years, it will be permanently stored in other asphalt products or e.g. lost as inert material in foundations in the following life cycles. The top layer asphalt contributed to store such an amount of biogenic carbon for a percentage of time, and after 100 years, it will be permanently stored. Hence, the top layer should be entitled to a percentage of the credit for the permanent storage of biogenic carbon over 100 years, which is therefore accounted (for SMAs and ACs, 15% i.e. 15 years lifetime over 100 years permanent storage, for ZOABs 12% i.e. 12 years lifetime over 100 years permanent storage). This credit was included in module D.

3. Results

3.1. Kraft lignins

The cradle-to-gate climate change impact of the three kraft lignins assessed is shown in Fig. 2.

As highlighted by the authors of the LCAs from which the data were retrieved (Bernier et al., 2013; Culbertson et al., 2016), the climate change impacts of kraft lignin production are mainly caused by the consumption of natural gas (mainly emissions from combustion i.e. 83% of climate change impact) and the production of liquid carbon dioxide. For kraft1-BIOM, other relevant impacts are direct emissions from the calcination reaction (carbon dioxide) and from the combustion of hog fuel (direct emissions of dinitrogen monoxide and biogenic methane and electricity to operate the furnace) and the impact (mainly emissions of biogenic methane) from the treatment of the solid waste generated by the pulp mill. For kraft2 (both -NG and -BIOM), the additional sodium hydroxide, which is necessary to make up for sodium losses caused by lignin extraction, is also important. In particular, beyond natural gas, the differences in impacts are also linked to the different allocation methods. For example, the direct emissions from the calcination reaction are allocated via economic value to lignin in kraft1-BIOM while they were not allocated to kraft 2 using physical causality allocation (since their amount does not change extracting or not lignin). Similarly, the impact of sodium hydroxide is much higher for kraft2 (both -NG and -BIOM) than for kraft1-BIOM since the additional sodium hydroxide that is necessary due to lignin extraction was entirely apportioned to lignin via physical causality allocation. Conversely, given the impossibility to subdivide the process in which it is used, this additional sodium hydroxide was also allocated to the pulp and only a negligible fraction to lignin (via economic allocation see Section 2.2.1.1.1).

Fig. 3 shows the cradle-to-gate environmental impact of kraft lignin expressed in terms of MKI score. The environmental impact per each impact category can be found in Appendix.

From Fig. 3, it is possible to observe that the difference of impact between the kraft lignins assessed is much lower than in the case of climate change (see Fig. 2). For all lignins, the reason is that the impacts of carbon dioxide and sulfuric acid production are much higher in terms of MKI scores than for climate change. The reasons (see Fig. 3 and details for these categories in Fig. 4) can be found in their marine aquatic ecotoxicity and human toxicity impacts. In particular, for liquid carbon dioxide production, the main causes are the materials (mainly copper and steel) used to construct the chemical plant and the production of MEA. Similarly, for sulfuric acid production, the main impact is caused by the materials consumed for the construction of the chemical

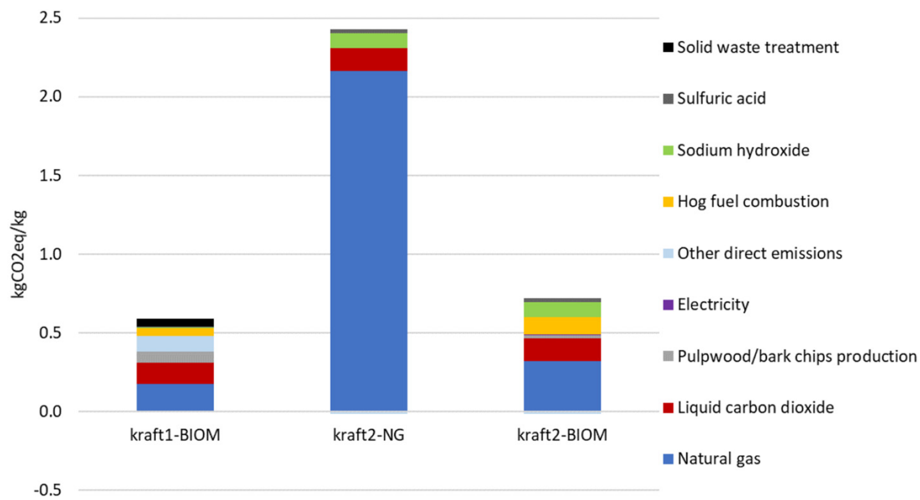

Fig. 2. Cradle-to-gate climate change impact (excl. biogenic carbon removal) of 1 kg of kraft lignin. Biogenic carbon content expressed in terms of carbon dioxide equivalent: 2.4 kg CO_2eq/kg.

plant. An important contribution to the impact in these categories is also caused by hog fuel combustion for kraft1-BIOM and kraft2-BIOM. For human toxicity, the main source of impact of hog fuel combustion is made from direct emissions (mainly benzene to air). For marine aquatic ecotoxicity, the electricity for operation of the furnace and materials for the construction of the furnace are the main impacts. For kraft2-NG and

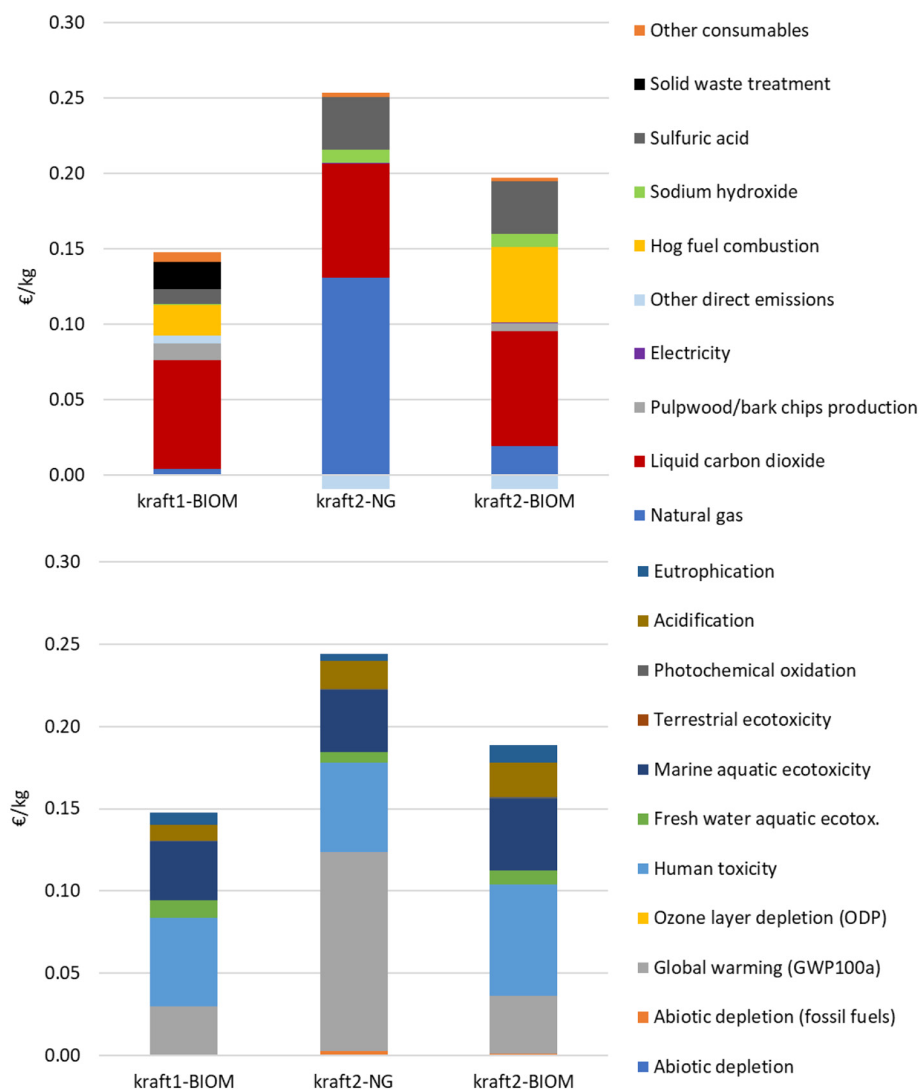

Fig. 3. Cradle-to-gate MKI score of 1 kg of kraft lignin. Breakdown per process contribution (top bars) and impact category (bottom bars). In this figure, the MKI score is without biogenic carbon removal, which accounts for 0.12 €/kg.

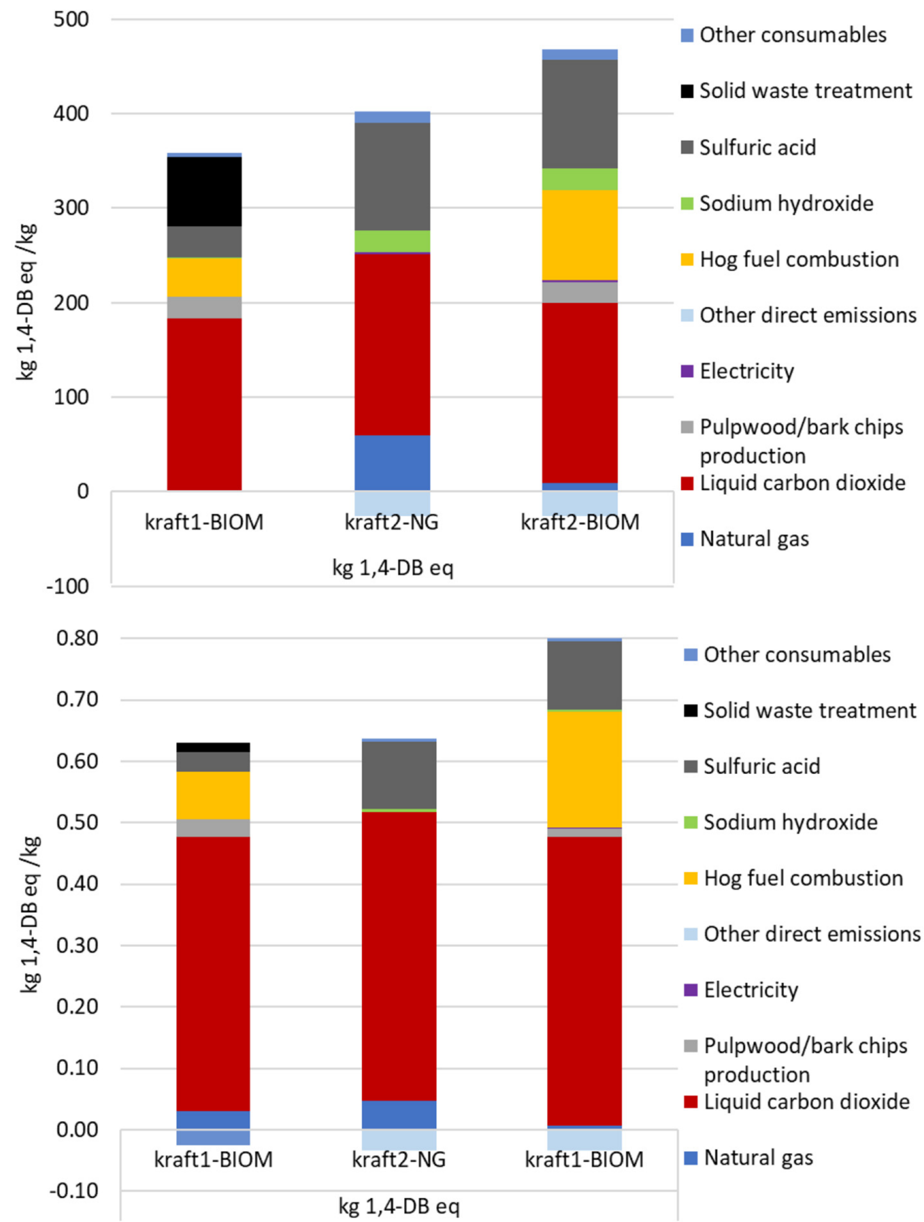

Fig. 4. Cradle-to-gate impacts of 1 kg of kraft lignin in marine aquatic ecotoxicity (top bars) and human toxicity (bottom bars).

kraft2-BIOM, a small negative impact is generated by the reduction of direct emissions from the combustion of black liquor (see Appendix for further information about the meaning of negative emissions using physical causality allocation). Regarding kraft2-NG lignin, the important impact of natural gas is mainly caused by its combustion (76%).

3.2. SMAs

Fig. 5 shows the climate change impact and environmental profile (in MKI score) for SMAs divided into life cycle stages.

In all SMAs investigated, the extraction and processing of raw materials (A1) make the largest contribution to the total climate change impact and MKI score. What is immediately noticeable is that the environmental impact of the asphalt components (other than lignin) is smaller when lignin is used. The main reason is the replacement of a fraction of bitumen (and for a minor fraction by the biogenic carbon content of linseed oil, which is higher than the climate change impact generated by the production of linseed oil). For climate change, the impact of lignin is negative when hog fuel is used for steam production (kraft1-BIOM and kraft2-BIOM) since the biogenic carbon stored in lignin outperforms the climate change impacts caused by their production. Conversely, in the case of kraft2-NG lignin, the biogenic carbon content of lignin does not outperform the climate change impact caused by the production of lignin but it is almost entirely compensated (almost 95%). Regarding the climate change benefits of accounting module D, these benefits are important for kraft2-NG lignin and conventional SMAs since bitumen (and other materials for a minor fraction) is recycled. However, in the case of kraft1-BIOM lignin and kraft2-BIOM, module D represents a small positive impact since the climate change benefits of recycling bitumen and lignin (and other materials) are lower than the biogenic carbon removal that from the lignin in the assessed asphalt will be incorporated in the recycled asphalt in the following cycle. Overall, all lignin-based SMAs offer a reduction of climate change environmental impacts i.e. 78% using kraft1-BIOM lignin, 34% using kraft2-NG lignin and 75% for kraft2-BIOM.

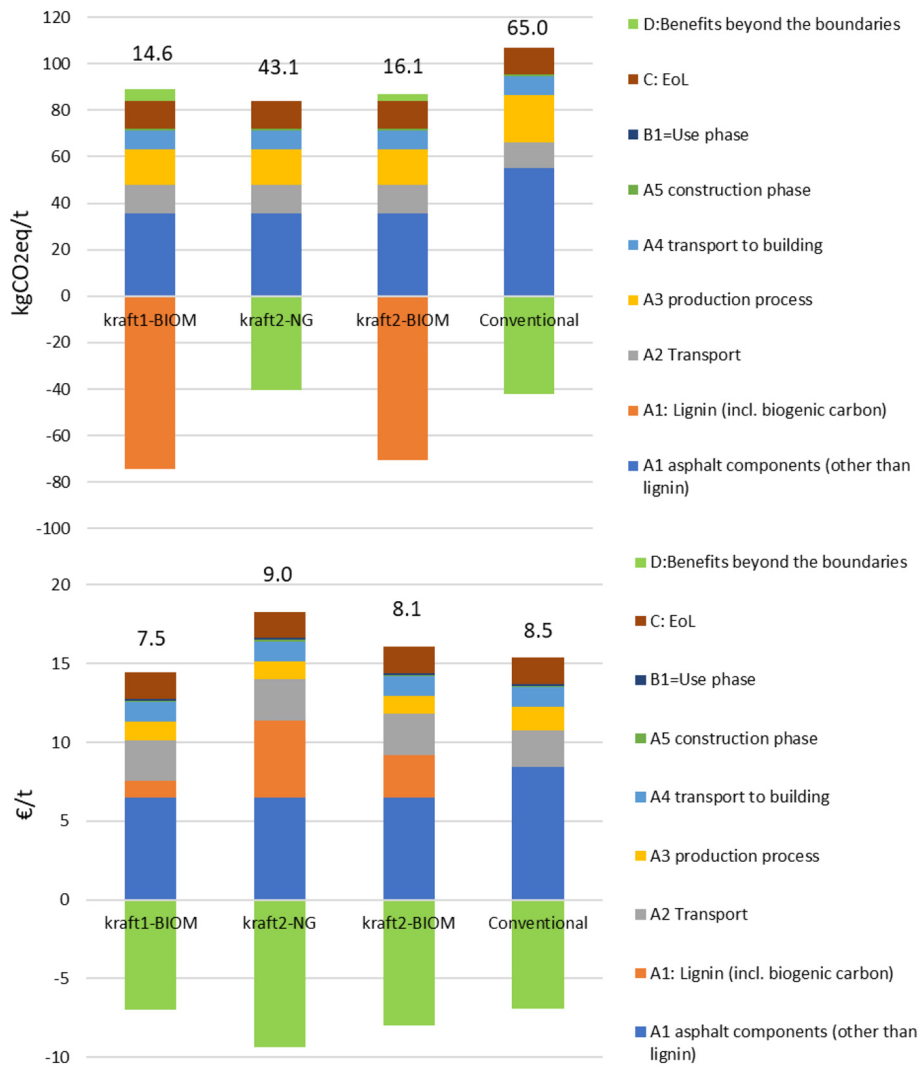

Fig. 5. Cradle-to-grave climate change impact (top bars) and MKI score (bottom bars) of 1 t of SMA asphalt. Breakdown per life cycle stage.

In terms of MKI score, only SMAs using kratf2-NG shows higher MKI scores than conventional SMAs. The reason can be found in the high environmental impact of the production and combustion of natural gas for kraft1-NG (see Section 3.1).

The MKI score of conventional asphalt was calculated as 8.5 €/t, which is 0.3 €/t lower than calculated in the Dutch LCA asphalt sector report (Schwarz et al., 2020) assuming a different composition for conventional SMA and supply chains. Taking this value 8.8 €/t as a reference would further improve the environmental competitiveness of lignin-based SMAs.

3.3. ACs

Fig. 6 shows the climate change and environmental profile (in MKI score) for ACs divided into life cycle stages.

Compared to SMAs, the extraction and processing of raw materials (A1) make a lower contribution to the total climate change impact and MKI score since 1) the amount of bitumen in ACs is lower than in SMAs, 2) ACs have a fraction of recycled component (i.e. burdens free) and 3) the amount of lignin is as much lower compared to SMAs (see Section 2.2.1). Comparing lignin-based and conventional ACs, similar trends to SMAs are observed for climate change and MKI scores. The MKI score of conventional AC was calculated as 8.3 €/t, which is in line with the range 7.4–8.8 €/t reported for several compositions of ACs in the Dutch LCA asphalt sector report (Schwarz et al., 2020). However, considering this broader range instead of 8.3 €/t, it is not possible to claim what asphalt between lignin-based and conventional ACs is better except for the AC using kraft1-BIOM.

3.4. ZOABs

Fig. 7 shows the climate change (top) and environmental profile (bottom, in MKI score) for ZOABs divided into life cycle stages.

In the case of ZOABs, lignin substituted weak filler instead of bitumen. This means that this type of asphalt has benefits related to the biogenic carbon content of lignin, which is beneficial for climate change (and consecutively for the MKI score). However, for the same amount in mass, the impact of weak filler is negligible compared to bitumen. For this reason, ZOABs show a good performance in terms of reductions of climate change impact (up to 60%) but MKI scores are in line or higher than conventional ZOAB. The MKI score of conventional ZOAB was calculated as 9.2 €/t, which is higher than 7.3–8.5 €/t reported in the Dutch LCA asphalt sector report (Schwarz et al., 2020). So, taking this range, it can be concluded that lignin-based ZOABs substituting weak filler with lignin leads to a (slightly) higher MKI score than conventional ZOABs.

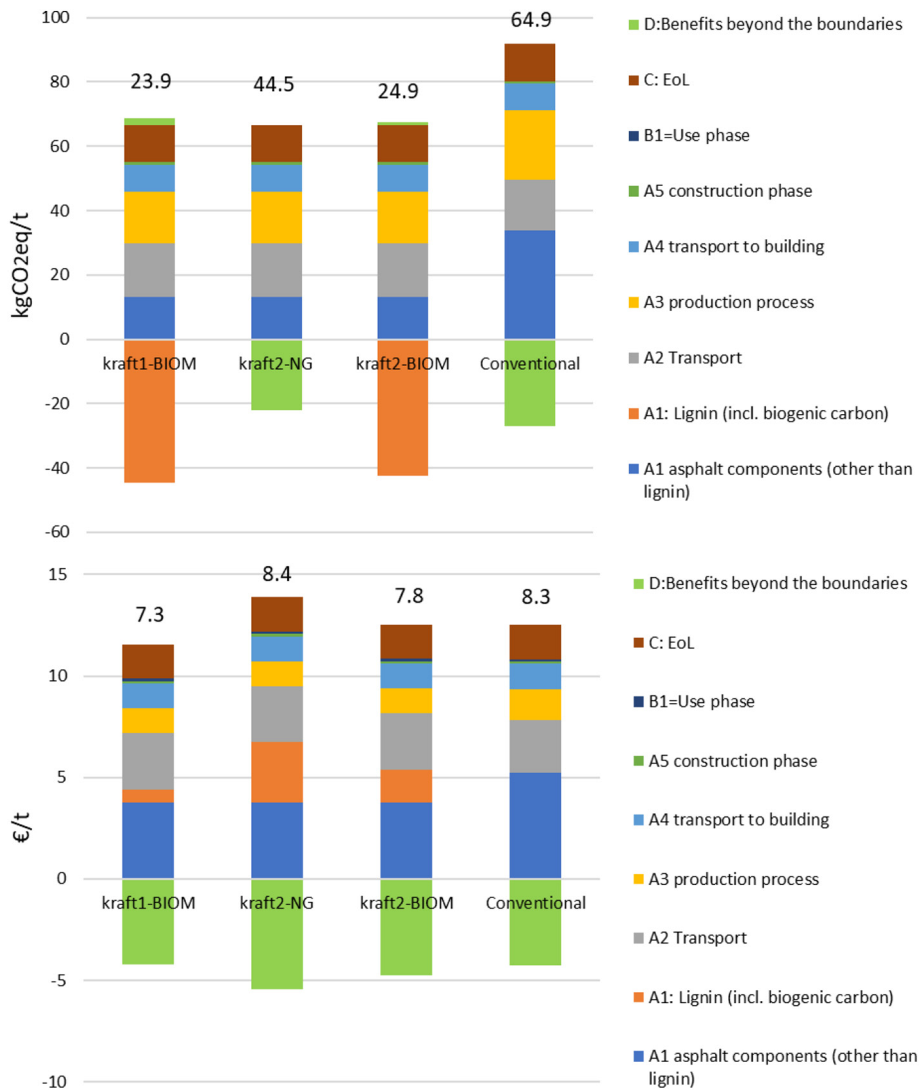

Fig. 6. Cradle-to-grave climate change impact (top) and MKI score (bottom) of 1 t of AC asphalt. Breakdown per life cycle stage.

4. Discussion

4.1. Multifunctionality

4.1.1. Lignin allocation

Previous literature showed that by-products like lignin are more affected than other products by the LCA issue of multifunctionality due to their lower physical/economic significance than the main product (Moretti et al., 2021, 2020b; Sandin et al., 2015). The price of lignin with the same quality specifications (mainly measured in terms of impurities) has been quite stable over time. However, lignin prices can vary significantly depending on its quality specifications (mainly impurities) (Hodásová et al., 2015; Moretti et al., 2021).

In our calculations, an average market price of 535 €/t was assumed for kraft lignin. This price is in line with \$600/t assumed by Dessbesell et al. (2018) but higher than 250\$/t assumed by Abbati De Assis et al. (2018) and Culbertson et al. (2016). For a kraft lignin that meets the quality requirements to be used in asphalt, a reasonable price range is between 370 and 700 €/t (Moretti et al., 2021). This price range was used for a sensitivity analysis on the economic allocation applied to the pulp mill producing kraft1-BIOM. To assess the impact of allocation methods, mass allocation was also applied. The results of this sensitivity analysis are shown in Table 4. Based on the results shown in Table 4, climate change impact and MKI scores of asphalts using kraft1-BIOM lignin are affected by the allocation method respectively within the order of ±10% and ± 4%. Since the price of pulp in €/t is higher than the lignin price (see Section 2.2.1.1), mass allocation results in a higher environmental impact of lignin compared to economic allocation.

4.1.2. Bitumen allocation and dataset

In the baseline calculations, the prescribed process map of the NL-PCR asphalt i.e. ESU NL-PCR bitumen (Keijzer et al., 2020) was used. This dataset was based on data from Energie-Stoffe-Umwelt (ESU) consultants, which uses energy allocation (Schwarz et al., 2020). However, bitumen, like lignin for biorefineries, is a low-economic/physical significance product of oil refineries and therefore, its environmental impact is affected strongly by allocation (Elgowainy et al., 2014; Moretti et al., 2017a). Moreover, applying energy allocation to bitumen seems in contrast with what is recommended by ISO 14049:2012 (ISO, 2012), which is the ISO technical report illustrating how to apply ISO 14044:2006. In such a report, ISO makes an example of bitumen production for which economic allocation is used since no physical parameter ("mass, feedstock energy, thermal conductivity, viscosity, specific mass, etc.") reflects the underlying physical relationship between bitumen and the other co-products.

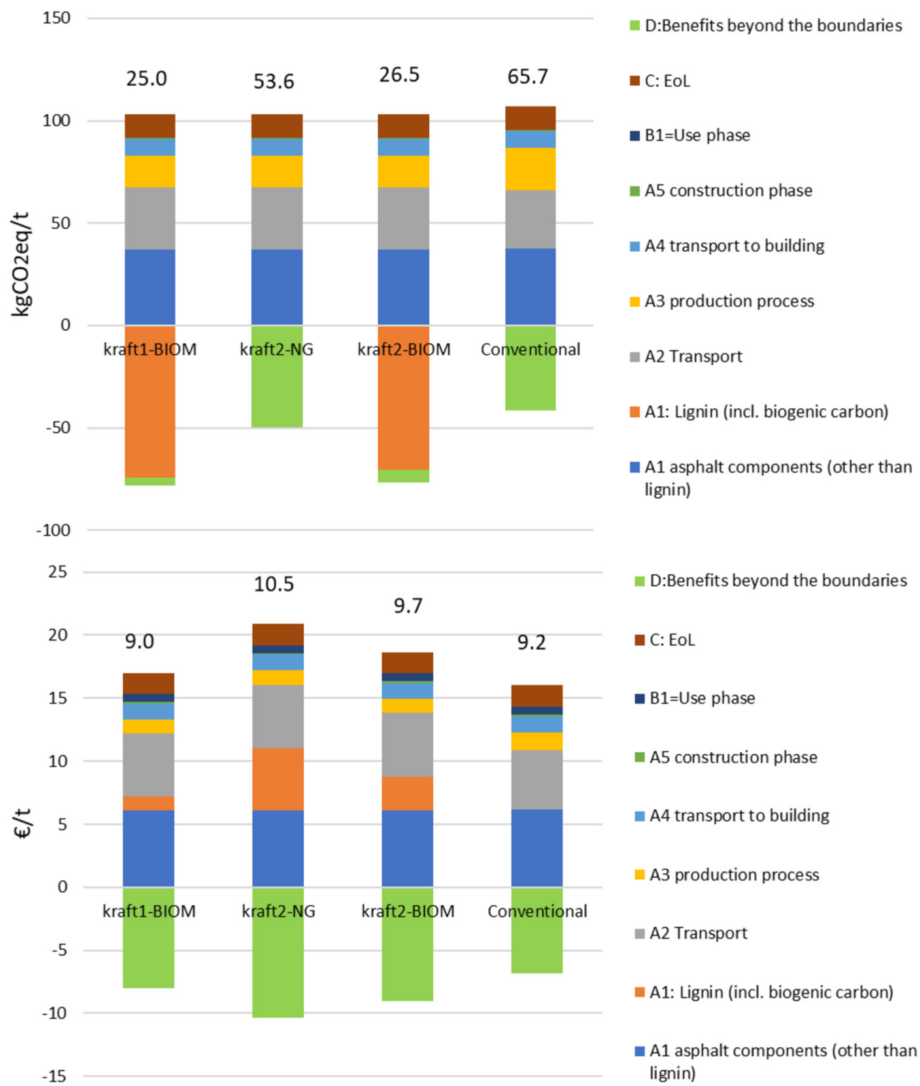

Fig. 7. Cradle-to-grave climate change impact (top bars) and MKI score (bottom bars) of 1 t of ZOAB asphalt. Breakdown per life cycle stage.

In literature, there are some alternatives available that LCA practitioners in the construction sector often use. For example, the previous version of the Dutch LCA asphalt sector report (Vos-Effting et al., 2018) was using a dataset for bitumen (SBK bitumen) that was based on data from Eurobitume and economic allocation was applied (Schwarz et al., 2020). Ecoinvent is also often used to retrieve datasets for petroleum products. For bitumen, two possible datasets are *Bitumen, at refinery/RER* and *Bitumen adhesive compound, hot {RER}| production | Cut-off*. Both these datasets use energy allocation (Schwarz et al., 2020).

Alternatively, a practitioner could follow a simplified approach and imagine the oil refinery as a black box. In literature, this approach is also referred to as allocation at the aggregate refinery level (Elgowainy et al., 2014; Han et al., 2015; Moretti et al., 2017a). By using this method, the whole upstream and oil refinery emissions are allocated to oil products using allocation shares based on the production volumes (mass allocation), or economic value (economic allocation) or their energy value (energy allocation).

Typical efficiencies (kg of products/kg of petroleum oil input) of oil refineries are between 90 and 94% (Han et al., 2015). This means that for 1 kg of petroleum input (from ecoinvent, *Petroleum {GLO}| market for | Cut-off*), 0.92 kg of petroleum products are produced on average (among them gasoline, diesel and bitumen). The remaining 0.08 kg of petroleum input is burned for refining such products (from ecoinvent, *Refinery gas, burned in furnace/kg/RER*). Based on this simplified modeling, the environmental impact generated by burning fuels in the refinery and producing the upstream crude oil can be allocated to the oil

Table 4
Sensitivity analysis on the allocation method applied to kraft1-BIOM.

Sensitivity	1 kg kraft1-BIOM		1 t SMA kraft1-BIOM		1 t AC kraft1-BIOM		1 t ZOAB kraft1-BIOM	
	GWP 100 (kg CO_2eq)	MKI (€)	GWP 100 (kg CO_2eq)	MKI (€)	GWP 100 (kg CO_2eq)	MKI (€)	GWP 100 (kg CO_2eq)	MKI (€)
Min price	0.47	0.13	13.0	7.2	22.7	7.1	23.4	8.7
Baseline (avg. price)	**0.58**	**0.15**	**14.6**	**7.5**	**23.9**	**7.3**	**25.0**	**9.0**
Max price	0.67	0.17	16.1	7.7	25.0	7.5	26.6	9.3
Mass allocation	0.71	0.17	16.8	7.8	25.5	7.6	27.2	9.4

refinery products using the abovementioned allocation shares. In Europe, bitumen represents about 3% of the economic share of oil refineries (Eurobitume, 2019). Mass and energy allocation shares for bitumen were assumed as 4.8% and 5.0% using petroleum coke as a proxy (Moretti et al., 2017a).

Fig. 8 shows the climate change and MKI scores of producing 1 kg of bitumen. From Fig. 8, it is possible to notice that 1) the dataset for bitumen from the NL-ESU PCR (Keijzer et al., 2020) is the one with the highest impact compared to all other alternatives compared, 2) economic allocation is the method that provides the lowest environmental impact for bitumen (in line with the effect of allocation on lignin shown by Table 4) while energy/mass allocation the highest. Moreover, the use of a different dataset and/or a different allocation method can influence the climate change impact up to a factor of 2–3 and the MKI score up to a factor of 4. It also emerges that the difference of data and not only of allocation play an important role in the difference of environmental impacts between the current and previous bitumen datased used by the Dutch LCA asphalt sector report. Fig. 9 shows the effect on the comparative analysis between the environmental performances of lignin-based and conventional asphalts taking the lowest environmental impact for bitumen. Consequently, the environmental impacts of both lignin-based and conventional asphalts would be lower since both contain bitumen. On the other hand, assuming a lower impact for bitumen, while the trends observed for climate change in the baseline analysis are conserved, the MKI scores of SMAs and ACs using kraft2-BIOM lignin becomes worse than the ones of the conventional asphalts while they were better in the baseline calculations. In particular, the impact of ACs is the least affected by the allocation applied to bitumen since its bitumen content was the lowest compared to SMAs and ZOABs.

4.2. Change of functional unit (to 1 m^2) and product system

In this LCA, the functional unit was defined as 1 tonne (t) of top layer asphalt. In the LCA literature, it is acknowledged that different functional units could lead to different results for the same product system (Hischier and Reichart, 2003; Moretti et al., 2021; Reap et al., 2008). Besides a mass-based functional unit, a common functional unit for asphalts is a surface-based functional unit (Giani et al., 2015; Keijzer et al., 2020; Yuan et al., 2018). Taking a functional unit of 1 m^2, it becomes a reasonable option to include in the product system also the middle and base layer asphalts that are under the top layer asphalt. A sensitivity analysis was conducted taking a functional unit of 1 m^2 of asphalt made of three layers. The compositions, lifetimes, densities and thicknesses of each layer can be found in Appendix. Depending on the type of asphalt, the mass of asphalt made of three layers under a surface of 1 m^2 has a mass between 0.39 and 0.40 t.

It is necessary to remark what follows to interpret the results of this sensitivity analysis correctly. Sub-layers have important recycled percentages. For lignin-based asphalts, the recycled part was tested with lignin-based recovered asphalt. The recycled material is free of the environmental burden in module A1 (that for lignin was accounted as environmental impact at the net of biogenic carbon removal). Accordingly, asphalt mixtures that already contain secondary material (recycled asphalt) do not include this percentage as environmental benefits in module D. Conversely, they are charged with a fraction of environmental burdens in module D as a result of the 29% loss of these secondary raw materials leaving the product system that cannot be used in the following cycle (Keijzer et al., 2020). These charges are net of the quality losses that these materials would have had in the following cycle in the sub-layers where they would have been utilized. The distance for the asphalt to be recycled to the transportation to the construction site was assumed as 50 km (Schwarz et al., 2020). Leaching does not take place in sub-layers since they are installed above groundwater level and do not come into contact with precipitation (Schwarz et al., 2020). Virgin lignin used in the middle and bottom layer takes a 30% credit on the biogenic carbon that will be stored in the following cycles and was stored by this product system for 30 years.

Fig. 10 shows the results of changing functional unit and product system. Changing functional unit does not lead to other trends. On the other hand, 1) the total impact is reduced in terms of surface since 1 m^2 contains less than 1 t of asphalt and 2) the differences between lignin-based and conventional asphalts are reduced since middle and base layers are 50% made of recycled asphalt (free of burden) and middle layer does not have any virgin lignin and base layer only 1%.

5. Conclusions

This study contributes to the existing research in the field of environmental sustainability of bio-based asphalt products. Various asphalts (stone mastic asphalt, asphalt concrete and porous asphalt) using kraft lignin were assessed using LCA methodology and compared with their conventional counterparts. The major LCA methodological choices were mostly based on the Dutch Product Category Rules (NL-PCRs) for asphalts. The data for kraft lignin were retrieved from two studies from the literature. The effects of allocation methods and defining a

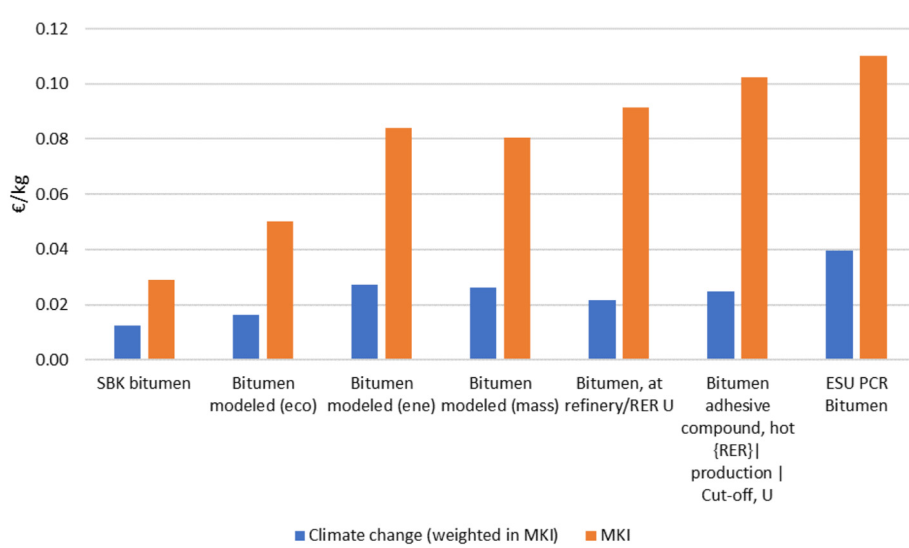

Fig. 8. Climate change impact (weighted in terms of MKI score) and MKI score of 1 kg of bitumen using various data and allocation methods. Eco = economic; ene = energy.

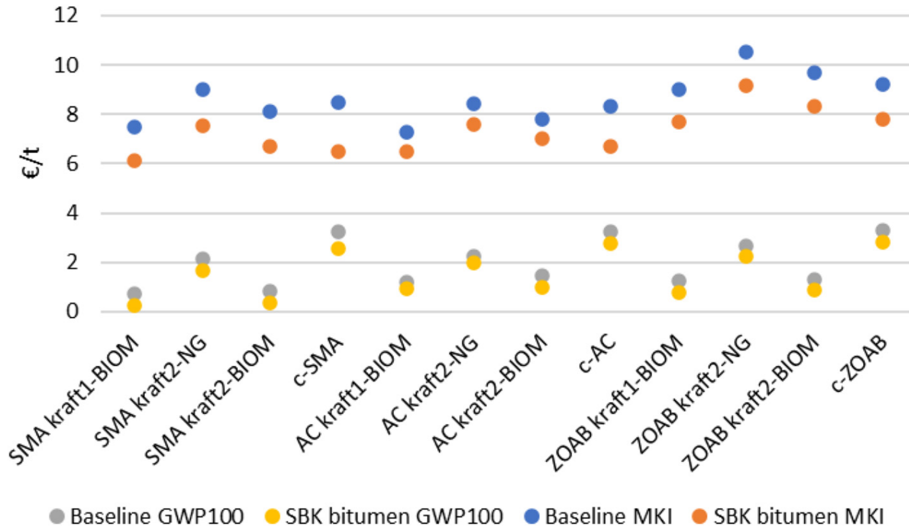

Fig. 9. Climate change impact (weighted in terms of MKI score) and MKI score of 1 t of top layer asphalt using a different dataset for bitumen (SBK bitumen). C = conventional.

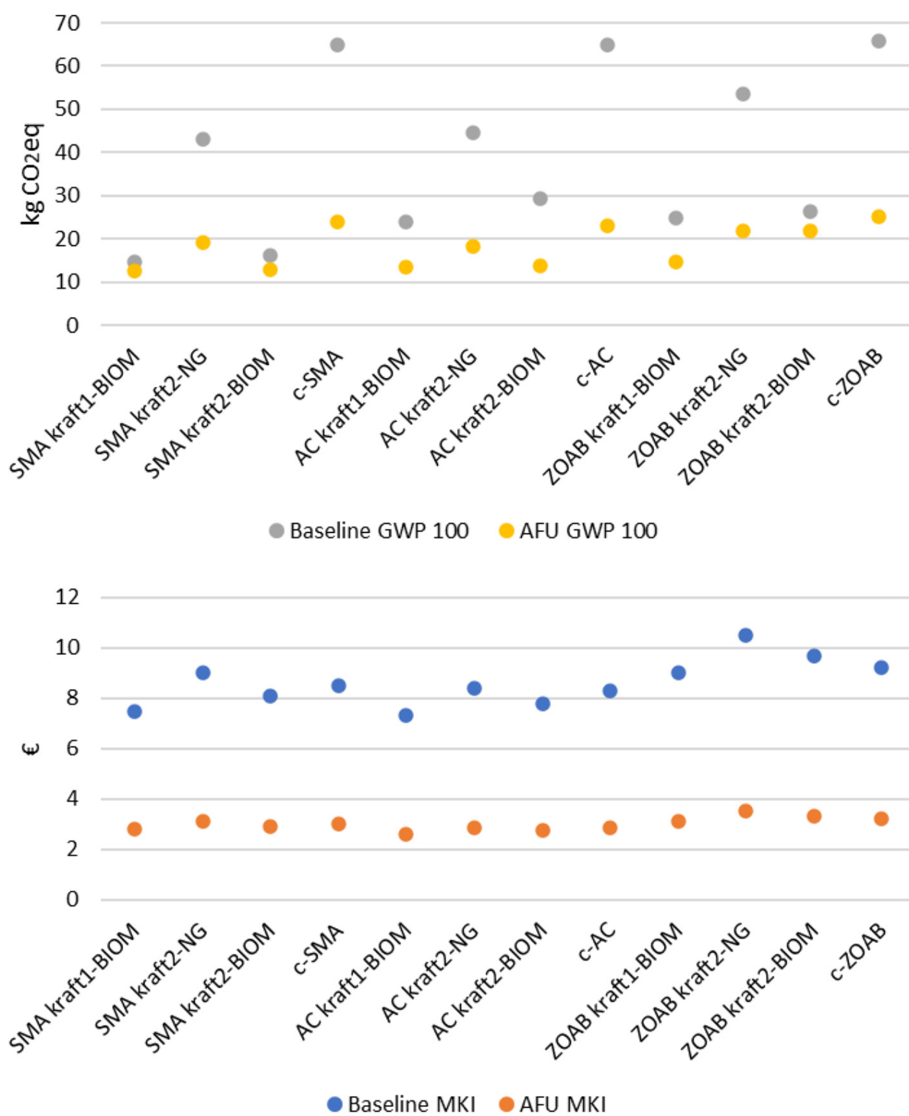

Fig. 10. Cradle-to-grave climate change impact (top graph) and MKI score (bottom graph) of baseline (1 t of top layer asphalt) versus alternative functional unit (AFU) i.e. 1 m^2 of asphalt made of three layers.

different functional unit were broadly discussed as well as the effect of using a different fuel (natural gas or hog fuel) for steam production.

The results of the LCA revealed that using lignin in asphalts could reduce the climate change impact of top-layer asphalt products over their life cycle substantially (order between 30% and 75% depending on the type of asphalt considered). The highest reductions are achieved if the current use of lignin (contained in the black liquor) for process energy is replaced with low-grade biomass fuels, for example hog fuel. Using natural gas to replace the heating value of lignin for energy production leads to a significant/strong reduction in the climate change benefits.

Considering also other impact categories, on a weighted basis expressed in terms of economic cost (MKI scores), the advantages of using hog fuel instead of natural gas in the pulp mill on the impact of the asphalts are in part mitigated since other categories are less affected by such shift of burdens. Nonetheless, given the important advantages in the climate change category, using hog fuel instead of natural gas also showed advantages in total MKI scores.

For this reason, stone mastic asphalt containing lignin produced from a pulp mill where hog fuel is used to replace the part of the energy that is no longer produced from burning black liquor showed environmental advantages also in terms of MKI scores compared to their conventional counterpart. Conversely, if natural gas is used to deliver the required process heat, the MKI score of lignin-based stone mastic asphalts was worse than the conventional counterpart. In other words, the climate change impact (and, to a lesser extent, overall environmental impact) is largely determined by how additional process heat is produced in the pulp mill. This conclusion is in line with previous research on other lignin-based applications (Moretti et al., 2021; Obydenkova et al., 2017; Secchi et al., 2019).

For asphalt concretes, it was not possible to identify an "environmental winner" between lignin-based and conventional asphalts since the differences in impact were in line with the uncertainties. For porous asphalts with lignin used only as weak filler, lignin-based asphalts perform worse than conventional asphalts. The reason is that the impact of limestone filler is minor compared to kraft lignin. To maximize environmental benefits, the maximum substitution of bitumen with lignin should be targeted, whereas the substitute of weak filler should be limited.

Regarding the allocation method, using mass allocation instead of economic allocation would lead to much higher environmental impacts for both lignin and bitumen. Using a physical causality allocation avoids that impacts that are typical of pulp production and are unaffected by extracting or not lignin are allocated also to lignin. On the other hand, once the credits for their recycled are accounted for, the effect of the change in allocation method becomes smaller.

Regarding the sensitivity analysis on the functional unit and product system considered, since middle and base layers have a higher percentage of recycled content and a lower amount of bitumen/lignin, once the mix of the three layers is considered as the product system instead of the top layer only, the MKI score difference between the two asphalt options becomes minor. Conversely, looking at climate change only, the difference is still significant given the importance of the biogenic carbon storage for this category.

CRediT authorship contribution statement

Christian Moretti: Conceptualization, Methodology, Software, Data curation, Formal analysis, Investigation, Visualization, Writing – original draft, Writing – review & editing. **Blanca Corona:** Data curation, Investigation, Conceptualization, Methodology, Visualization, Writing – original draft. **Ric Hoefnagels:** Conceptualization, Methodology, Project administration, Writing – original draft, Writing – review & editing. **Marco van Veen:** Data curation, Investigation, Methodology, Visualization, Writing – original draft. **Iris Vural-Gürsel:** Data curation, Investigation, Visualization, Methodology, Writing – review & editing. **Tobias Strating:** Methodology, Data curation, Formal analysis, Investigation, Writing – review & editing. **Richard Gosselink:** Conceptualization, Data curation, Project administration, Funding acquisition, Resources, Supervision, Writing – review & editing. **Martin Junginger:** Conceptualization, Data curation, Methodology, Supervision, Funding acquisition, Resources, Writing – review & editing.

Declaration of competing interest

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

Tobias Strating is employed by an industrial partner (Roelofsgroep) that might have financial interests in the technology investigated by this article.

Richard Gosselink is the inventor of a patent regarding lignin-based bio-asphalt.

The other authors do not have any known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.150316.

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