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Using lignin from local biorefineries for asphalts: LCA case study for the Netherlands

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ABSTRACT

The production of bio-based asphalt utilizing lignin from amongst others local biorefineries is currently under development in the Netherlands. In this study, life cycle assessment (LCA) methodology was applied to investigate the environmental implications of replacing conventional asphalts with lignin-based asphalts. Inventory data for lignin were collected from two Dutch biorefining industries (Avantium and Vertoro). Eleven impact categories were considered, along with a single-score environmental cost indicator. For the comparative assessment, both kraft lignin asphalts and conventional asphalts were used as benchmarks. The effect of a change of allocation method was discussed.

Process steam, chemicals (mainly hydrochloric acid or methanol) and electricity were identified as the main environmental hotspots of the two biorefineries. Comparing on the basis of the same steam source, both biorefinery lignins showed lower climate change (up to 45% lower) and environmental cost (up to 60%) than kraft lignin. Top-layer asphalts using biorefinery lignins showed a 35–70% lower climate change impact than conventional asphalts. For base-layer asphalts, 25–50% reduction of climate change was calculated compared to conventional asphalts. On an environmental cost-weighted basis, besides climate change, other relevant environmental impacts are marine aquatic ecotoxicity, human toxicity, eutrophication and acidification. Using mass allocation instead of economic allocation showed that the environmental impact of lignin can increase, decrease or remain unchanged depending on the production system and co-products of lignin.

1. Introduction

Next to cellulose, lignin is the second most abundant bio-based polymer on the earth (Chen et al., 2021; Khan et al., 2019). Traditionally, lignin is produced as a by-product of the pulp and paper industries and used for internal energy supply (Benali et al., 2016; Lask et al., 2021; Pola et al., 2019). Worldwide, the paper and pulp industries produce 50–70 million tonnes (Mt) of lignin per year (Bajwa et al., 2019). Lignin can also be produced by lignocellulosic biorefineries and used for similar purposes (Paone et al., 2020; Ragauskas et al., 2014). With the expected growth of the number of lignocellulosic biorefineries in the next decade, the total volume of worldwide lignin production could grow up to 225 Mt per year, generating an excess of lignin supply (Bajwa et al., 2019). An effective utilization of the lignin excess can play an important role in the commercial success of lignocellulosic biorefineries.

The Netherlands has a relatively large and energy-intense (petrochemical) industry sector and an extensive infrastructure network. The Netherlands is also one of the leading European countries producing bio-based chemicals and fuels (Parisi, 2020). Dutch multi-output biorefineries can play a central role in the EU (bio)energy transition and circular economy strategies (Parisi, 2020). With the increasing production of bio-based chemicals and fuels, more lignin is expected to become available in the Netherlands. Such lignin could be marketed for various chemical, energy and material applications to replace petrochemical ingredients (Bajwa et al., 2019). For example, lignin can be used to replace formaldehyde for adhesive (McDevitt and Grigsby, 2014), bitumen for asphalts (van Vliet et al., 2017) or crude oil for the production of liquid fuels (Obydenkova et al., 2017).

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Since lignin derives from biomass, using lignin to replace petrochemical ingredients could reduce the climate change impact and the depletion of fossil fuels of various products (Montazeri et al., 2016; Moretti et al., 2021b). Moreover, the high biogenic carbon content (≈60%) of lignin could potentially be stored in durable bio-based products like asphalt with further climate change benefits (Souto et al., 2018). However, previous literature highlighted that a promising environmental performance for lignin-based products compared to petrochemical products is strictly linked with the specific lignin considered and respective production process (Montazeri et al., 2016; Moretti et al., 2021b).

One of the main tools used to evaluate the environmental performance of lignin-based products and bio-based products more generally is the environmental life cycle assessment (LCA) methodology (Dahiya et al., 2020; Karka et al., 2019; Martin et al., 2018). In the last two decades, LCA methodology, which is standardized by ISO 14040:2006 and ISO 14044:2006 (ISO, 2006a; 2006b), has become a reference tool for supporting policymakers in the implementation of bio-economies to reach sustainability goals and secure energy supplies (European Commission2019; Martin et al., 2018; Sala et al., 2021).

This study presents two environmental life cycle assessments (LCAs) with different scopes. The first LCA focuses on lignins from two innovative biorefineries (Avantium and Vertoro) at a pilot scale in the Netherlands. For the LCA model of these two lignins, primary data were collected. The environmental impact of the lignins produced from these two biorefineries was compared with that of kraft lignin retrieved from our earlier work (Moretti et al., 2021a). The second LCA aims to evaluate the potential environmental benefits of using such lignins in bio-based asphalts in the Netherlands, with "asphalt" used to refer to "asphalt mixture" hereafter. Since the energy source used to produce the process steam has a significant effect on the climate change impact of lignin (Moretti et al., 2021b), the effect of alternative energy sources was investigated.

While the importance of LCA as a tool for environmental evaluations is not questioned, the way LCAs should be conducted is challenged by the development of innovative bio-based products (Martin et al., 2018; Moretti et al., 2020). Even using a standardized method, it is challenging to compare bio-based products with conventional products from an environmental perspective in a fair and incontestable way (Guest et al., 2013; Moretti et al., 2020; Pawelzik et al., 2013). Several LCA modeling methodological choices can strongly affect the environmental impact of lignins. In particular, several authors (Hermansson et al., 2020; Montazeri et al., 2016; Moretti et al., 2021b; Obydenkova et al., 2021) emphasized the effect of selecting a different method of dealing with multifunctionality. For this reason, the influence of the allocation method on the environmental impact of the two biorefinery lignins was discussed in detail.

2. Method

2.1. Goal and scope definition

This study aims to evaluate the environmental impact of lignins from two different biorefineries (Avantium and Vertoro) and their use for bio-based asphalts. The analysis was conducted with the aid of the SimaPro 9.1.1 software. The targeted audiences of these LCAs are both biorefining and asphalt industries and EU policymakers interested in innovative bio-based products. In particular, for lignin production, it is critical to understand that lignin is currently used predominantly as fuel for internal energy in the biorefinery (or pulp mill). This fuel must be substituted by an alternative, and this decision can have far-reaching consequences (Bernier et al., 2013; Moretti et al., 2021b). The two possible scenarios assessed for both biorefineries were natural gas and hog fuel. While natural gas is usually economically favorable and allows reductions in local atmospheric emissions (Bernier et al., 2013; Moretti et al., 2021b), low-value biomass, such as hog fuel (e.g. bark chips), allows lower climate change impacts (Bernier et al., 2013; Moretti et al., 2021b). This option can be considered when hog fuel is locally available and/or there are either no critical issues on local atmospheric emissions in the region of the biorefinery, or it is possible to reduce them via state-of-art treatment methods (Bernier et al., 2013).

The LCA focusing on lignin production from biorefineries has a cradle-to-gate scope with a functional unit defined as 1 kg of dry lignin. The second LCA focuses on asphalts using this lignin to replace conventional ingredients such as bitumen and has a cradle-to-grave scope, i.e. the use and end of life of the asphalt are included in the assessment. Both top layer and base layer asphalts were considered as product systems, and the functional unit was defined as 1 tonne (t) of asphalt used in a specific layer (top or base layer). The geographic scope is the Netherlands. The temporal scope is the year 2030 when an Nth plant for lignin-asphalt production is assumed to be operating.

Both LCAs were conducted following an attributional approach, i.e. cut-off rules and allocation approaches are used to isolate the environmental impact of the investigated product from the global environmental impact (Pelletier et al., 2015). The coding of the life cycle stages of asphalts was based on the Dutch reference documents i.e. 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). Besides ISO 14040 and ISO 14044 (ISO, 2006a; 2006b), these reference documents are based on EN 15804 (CEN, 2013). The process flow diagram including such coding is shown in Fig. 1.

Based on the Dutch LCA PCR approach for asphalts (Bouwkwaliteit, 2019), the following impact categories were considered: abiotic depletion, abiotic depletion (fossil fuels), global warming potential (GWP), ozone layer depletion (ODP), human toxicity, freshwater aquatic ecotoxicity, marine aquatic ecotoxicity, terrestrial ecotoxicity, photochemical oxidation, acidification and eutrophication. Accordingly, the impact assessment models and weighting factors were also based on Bouwkwaliteit (2019). In particular, the weighting factors were used to calculate the so-called "MKI score" (Bouwkwaliteit, 2019), which is based on the shadow price method (internationally often referred to as environmental cost indicator). The applied weighting factors can be found in Table 1.

2.2. Life cycle inventory analysis

2.2.1. Lignin production

In this study, two biorefineries were considered for the production of lignin. The first biorefinery (Avantium Dawn Technology™), hereafter called "AVT", is an improved Bergius-Rheinau process (Bergius, 1937) that transforms wood chips into industrial sugars and lignin via concentrated acid hydrolysis using hydrochloric acid. The production of lignin was tested with wood chips from pine. Wood chips are sourced from the Netherlands, Germany and Belgium. An average transportation distance of 50 km was assumed. The inventory data per 1 t of dry lignin from AVT biorefinery with steam produced from either natural gas (product system named AVT-NG) or hog fuel (product system called AVT-BIOM) can be found in Table 2. It should be remarked that in the case of AVT biorefinery, data were available only for a first of its kind plant.

The second biorefinery is based on the Vertoro (VRT)'s acid methanolysis process (Kouris et al., 2021). Table 3 shows the inventory data for the VRT biorefinery to produce 1 t of dry lignin. In the biorefinery, the ground and dried sawdust, after being mixed with methanol and sulfuric acid, is preheated and subjected to mild solvolysis. Methanol is applied as a solvolytic medium. Solvolysis allows to convert lignin into soluble lignin oligomers, while cellulose remains as solid, and hemicellulose is converted to methylated C5 sugars. The obtained oil is first flashed, which allows recovering about 10% of methanol, which is recycled back to the process, while non-condensable gases are released to the atmosphere. After neutralising sulfuric acid, solid cellulose is filtrated with an excessive amount of methanol to ensure the required separation efficiency. The filtrate is subjected to distillation, allowing to

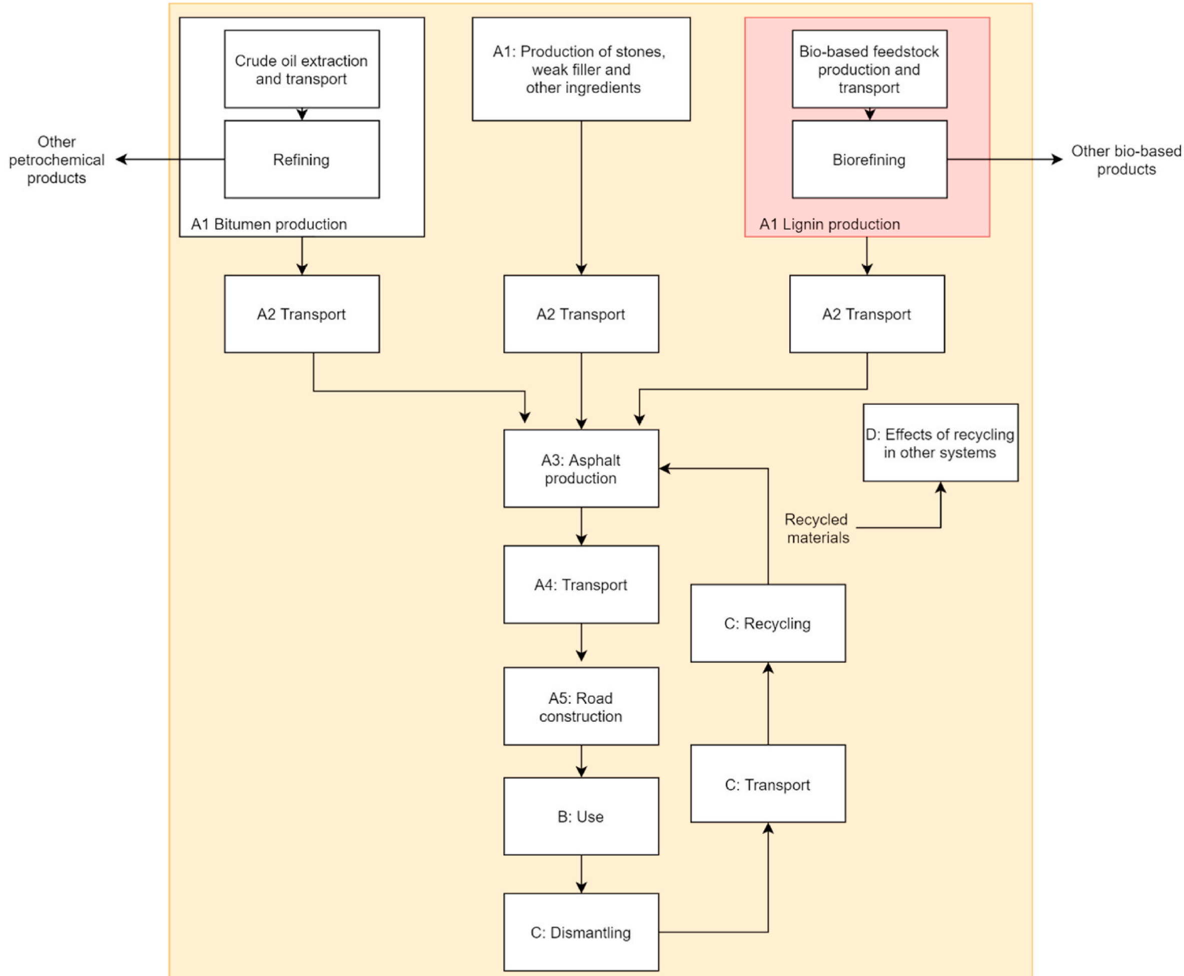

Fig. 1. Process flow diagram of lignin production and lignin-based asphalts. In red, cradle-to-gate boundaries of lignin production. In yellow, cradle-to-grave boundaries of the asphalt product.

Table 1
Weighting factors to calculate the "MKI score" (Bouwkwaliteit, 2019).

Indicator with unit per category	Weighting applied
Abiotic depletion (kg Sb eq)	0.16 €/kg
Abiotic depletion of fossil fuels (MJ)	7.7E-05 €/MJ
Global warming 100a (kg CO_2 eq)	0.05 €/kg
Ozone layer depletion (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

recycle about 78% of the methanol used in the solvolysis. The obtained at the distillation bottom oil undergoes the second filtration step, with water added prior to and during the filtration to ensure lignin precipitation and its efficient removal from the filtration area. The precipitated wet lignin oligomers are dried to obtain 97–98% (w.b.) lignin oligomers (Goldilocks®). The methanol in the filtrate is separated in the second distillation unit. Steam is used to preheat the sawdust slurry, as well as for the distillation and drying processes. Also, for the VRT biorefinery, two product systems differentiated by the energy source for steam production were assessed: VRT-NG, whose process steam is produced from natural gas, and VRT-BIOM, whose process steam is produced from low-value biomass (hog fuel).

Since biorefineries are multi-output products, an allocation method is necessary to apportion environmental impact to each of the co-products. In this LCA, based on the recommendations from EN 15804 (CEN, 2013), an economic allocation was adopted. Moreover, economic allocation represents the causality of the system, i.e. the reason to operate the biorefinery is to produce the products with a higher value on the market.

The prices, allocation shares and amounts of the biorefinery co-products are shown in Table 4. While kraft lignin is commercially available, there are no established markets yet for biorefinery lignin (this also partially applies to the second generation glucose/mixed sugars from wood). As they are expected to compete in similar markets, prices of kraft lignin were assumed to be representative for biorefinery lignin. A sensitivity analysis varying the lignin price and using a different allocation method (i.e. mass allocation) was carried out. The results from this sensitivity analysis are described in the discussion section.

2.2.2. The asphalts' life cycle

The compositions of both asphalts with and without lignin fulfilling these requirements can be found in Table 5. In particular, asphalt concrete was considered since this type of asphalt is used for both top and base layers. To guarantee a meaningful comparison, both lignin-based and conventional asphalts have to provide similar rheological characteristics and fulfil fatigue performance requirements (Brovelli et al., 2014; Martinez-Arguelles et al., 2015). Various empirical and functional

Table 2
Life cycle inventory data for the production of 1 t of dry AVT lignin with either steam produced from natural gas (AVT-NG) or from hog fuel (AVT-BIOM).

Inventory flow	AVT-NG	AVT-BIOM	Unit	Background
Wood chips	3.0	3.0	t	Wood chips, wet, measured as dry mass {DE}| softwood forestry, pine, sustainable forest management | Cut-off
Hydrochloric acid	0.3	0.3	t	Hydrochloric acid, without water, in 30% solution state {RER}| market for | Cut-off
Evaporation media	5.0	5.0	kg	Base oil {GLO}| market for base oil | Cut-off
Process water	24.6	24.6	t	Tap water {Europe without Switzerland}| market for | Cut-off
Sodium hydroxide	0.1	0.1	t	Sodium hydroxide, chlor-alkali production mix, at plant/RER
Active carbon	0.5	0.5	kg	Charcoal {GLO}| market for | Cut-off
Sugar workup resin	0.2	0.2	kg	Polystyrene granulate (PS)/EU-27 from PlasticsEurope
Electricity	0.9	0.9	MWh	2030 EU reference scenario for the Netherlands (Carpos et al., 2016)
Natural gas for heat and/or steam	52267	3850	MJ	Natural gas, burned in industrial furnace >100kW/RER from ecoinvent
Hog fuel production	0	3041	kg	Bark chips, wet, measured as dry mass {CH}| bark chips production, softwood, at sawmill | Cut-off
Hog fuel combustion	0	43576	MJ	Retrieved from Heat, central or small-scale, other than natural gas {CH}| heat production, softwood chips from forest, at furnace 50 kW, state-of-the-art 2014 | Cut-off. Background data for the electricity input for the operation of the furnace updated using 2030 EU reference scenario for the Netherlands (Carpos et al., 2016)
Cooling energy	21.6	21.6	MJ	Cooling energy {CH}| from natural gas, at cogen unit with absorption chiller 100 kW | Cut-off
Wastewater to be treated	15.0	15.0	m3	Wastewater, average {Europe without Switzerland}| market for wastewater, average | Cut-off

Table 3
Life cycle inventory data for the production of 1 t of dry VRT lignin with either steam produced from natural gas (VRT-NG) or hog fuel (VRT-BIOM).

Inventory flow	VRT-NG	VRT-BIOM	Unit	Background data
Direct emissions of fossil carbon dioxide (from non condensable gases)	2.3	2.3	kg	
Sawdust	4.4	4.4	t	Saw dust, wet, measured as dry mass {GLO}| market for | Cut-off
Methanol	0.2	0.2	t	Methanol {GLO}| market for | Cut-off
Sulfuric acid	21.5	21.5	kg	Sulfuric acid {RER}| market for sulfuric acid | Cut-off
Caustic soda	1.8	1.8	kg	Sodium hydroxide, chlor-alkali production mix, at plant/RER
Electricity	409	409	kWh	2030 EU reference scenario for the Netherlands (Carpos et al., 2016) with background data from ecoinvent
Heat or process steam from natural gas	84551	1886	MJ	Natural gas, burned in industrial furnace >100kW/RER
Hog fuel production	0	5728	kg	Bark chips, wet, measured as dry mass {CH}| bark chips production, softwood, at sawmill | Cut-off
Hog fuel combustion	0	82665	MJ	Retrieved from Heat, central or small-scale, other than natural gas {CH}| heat production, softwood chips from forest, at furnace 50 kW, state-of-the-art 2014 | Cut-off. Background data for the electricity input for the operation of the furnace updated using 2030 EU reference scenario for the Netherlands (Carpos et al., 2016)
Cooling water	1180	1180	t	The inventory for cooling water production via industrial cooling tower was taken from (Schulze et al., 2019). For the water consumption, the dataset was retrieved from ecoinvent (Water, decarbonised {DE}| water production, decarbonised | Cut-off). For the electricity input, the electricity mix updated using 2030 EU reference scenario (Carpos et al., 2016) for the Netherlands was used.
Water for precipitation	8.3	8.3	t	Water, decarbonised {DE}| water production, decarbonised | Cut-off

tests were conducted according to European standards EN 13108 series (1 and 20) and EN 12697 series (8-12-23-24-25-26-31-33-35) to guarantee functional performances and lifetimes in line with those of conventional asphalts. The empirical tests included run on gyrator samples, hollow space percentage and water sensitivity, while the functional tests included gyrator test pieces and rolled test plates, tri-axial test for rutting resistance, stiffness on test beam sawn from the rolled test plates, and fatigue on the same beams.

The background data used to model all asphalt ingredients are also provided in this table.

Given the Dutch geographic scope, the life cycle stages (and respective coding) to be included in the calculations were based on the Dutch Product Category Rules (NL-PCRs) (Keijzer et al., 2020) and the Dutch LCA asphalt sector report (Schwarz et al., 2020). The distances for the transportation of each of the asphalt's ingredients were mainly based on primary data from the asphalt industry. Standard values from the Dutch LCA asphalt sector report (Schwarz et al., 2020) were used only if primary data was not available. Details regarding the transportation distances and transport modes can also be found in previous publication (Moretti et al., 2021a). The transportation distance between the biorefineries and the asphalt processing plant was assumed to be 50 km. The consumption of utilities (natural gas, electricity and diesel) for asphalts production was provided by another industrial producer. Details regarding the data for such utilities consumption can be found in Moretti et al. (2021a). The transportation distances of the asphalt product to the construction site and the diesel consumed during the installation of the product were retrieved from the Dutch LCA asphalt sector report (Schwarz et al., 2020). Based on the Dutch LCA asphalt sector report

Table 4
Co-products from the biorefineries per 1 t of dry lignin and economic allocation shares. All prices are inflation corrected into euro 01–2021.

	Price (€/t dry)	Amount (t dry/t dry lignin)	Economic allocation share
AVT biorefinery			
Lignin	535 (Moretti et al., 2021b)	1	**44.0%**
Glucose	439 (Michels, 2014)	1.2	42.3%
Mixed sugars	219.5 (Michels, 2014)	0.6	10.9%
Biomass fines	71.5. Assumed to have 90% of the value of the wet biomass feedstock.	0.4	2.5%
Extractives/oil streams	37.6. Based on the difference in lower heating value between extractives/oil streams (10 MJ/kg) and biomass fines (19 MJ/kg).	0.1	0.3%
VRT biorefinery			
Lignin	535 (Moretti et al., 2021b)	1	**19.7%**
Pulp	788 (Indexmundi, 2020)	2.5	71.6%
Mixed sugars (mainly C5)	219.5 (Michels, 2014)	1.1	8.7%

(Schwarz et al., 2020), the leaching of substances from top layers to the soil under the influence of precipitation was the only activity included in the impact of the use phase of the asphalt. These leaching emissions were retrieved from Schwarz et al. (2020). Based on current industry knowledge, no difference in leaching emissions between lignin-based and conventional asphalts is expected, but further research is needed (Moretti et al., 2021a). Based on the method described in the NL-PCR Asphalt (Keijzer et al., 2020), asphalts used in repairs during maintenance should be considered as another asphalt product, for which a separate LCA is carried out. Accordingly, road surface maintenance was not considered part of the environmental impact of the investigated asphalt. However, the positive outcome of the empirical and functional tests on the investigated lignin-based asphalt mixture should guarantee similar service performance to conventional asphalt, leading to similar timing and magnitude of maintenance interventions.

Based on the guidelines from the NL-PCR Asphalt (Keijzer et al., 2020), 10% of the binder (bitumen or lignin) of top-layer asphalts is assumed to be lost due to erosion in the use phase. At the end of the lifetime, the asphalt is recycled, and the amounts of the utilities for asphalt recycling were retrieved from the NL-PCR Asphalt (Keijzer et al., 2020). The entire environmental impact of the recycling process was apportioned to the primary asphalt. Accordingly, the recycled material used in a new asphalt is considered free of environmental burden. Based on the NL-PCR and Dutch LCA asphalt sector report (Keijzer et al., 2020; Schwarz et al., 2020), the environmental benefits resulting from recycling primary raw materials are included in the calculation of the environmental performance of the asphalt in the so-called "module D". Such benefits are accounted for with a specific formula reported in both the NL-PCR Asphalt (Keijzer et al., 2020) and our other work (Moretti et al., 2021a) that includes detailed information and discussion on background data, methodology, and assumptions made.

Regarding the biogenic carbon, its content for both lignins was measured in the lab and found to be 61.1% for AVT lignin and 61.5% for VRT lignin. Such biogenic carbon is assumed to not biodegrade in aerobic and anaerobic conditions typical of the ambient environment (and landfills). The biogenic carbon contained in virgin lignin was considered as a negative physical flow if entering the asphalt product system (module A1) and as a positive physical flow (i.e. an impact) if lignin leaves the boundaries of the product system i.e. when credited in module D. The biogenic carbon of the virgin lignin leaving the system boundaries is either recycled to new asphalts becoming permanently stored or lost as inert material (in foundations) in the following life cycles. So, the asphalt containing virgin lignin contributed to the permanent storage of biogenic carbon by storing such carbon for a specific time. There is no shared consensus on how to account for temporary storage (delayed emissions) of biogenic carbon that becomes permanently stored over several life cycles (Bishop et al., 2021; Tonini et al., 2021). In fact, in this case, various bio-based products have contributed to reaching that goal by storing such carbon for a specific time. There are several methods for such dynamic accounting and allocation of the stored biogenic carbon to each life cycle (Tonini et al., 2021). In our study, a simplified linear time-based credit was applied. The top layer was assumed to be entitled to a percentage of the credit (15%) for the permanent storage of biogenic carbon over 100 years, having stored such lignin for 15 years (average lifetime of a top layer asphalt concrete). This percentage becomes 30% for the base layer since the lifetime is 30 years. According to the NL-PCR Asphalt, the recycled content is free of the environmental burden in module A1 (see Fig. 1). So, module A1 does not incorporate the biogenic carbon removal of the recycled content. Nonetheless, the recycled content could potentially be derived from asphalt with a lignin percentage. Consequently, the recycled content does not lead to any environmental benefits in module D. However, in module D, the asphalts are charged with a fraction of environmental burdens resulting from physical and quality losses generated in this cycle that lead to the delivery of less material (or of lower quality) in the following cycle than before utilization. Further details can be found in the NL-PCR Asphalt (Keijzer et al., 2020).

2.2.3. Comparison: data sources

The environmental impacts of kraft lignins were retrieved from our other work (Moretti et al., 2021a), namely kraft1-BIOM, kraft2-NG and kraft2-BIOM. In particular, kraft1-BIOM refers to the lignin produced by a specific pulp mill whose data were retrieved from Culbertson et al. (2016). The other two kraft lignins were based on the pulp mill modelled by Bernier et al. (2013). Please refer to the background data and assumptions used in the LCA modeling of the kraft lignins (Moretti et al.,

Table 5
Composition of 1 t of asphalt concretes used in top and base layer with and without lignin based on (Moretti et al., 2021a).

Composition	Top layer with lignin	Base layer with lignin	Top Layer (conventional)	Base layer (conventional)	Inventory dataset
Recycled content (kg) obtained from older asphalts	288	500	300	500	Burden-free according to the guidelines from the NL-PCR Asphalt (Keijzer et al., 2020)
Bitumen (kg)	21.1	5.5	40	18	ESU NL-PCR bitumen (Schwarz et al., 2020).
Crusher sand (kg)	154	0	171	0	Sand {CH}| gravel and quarry operation | Cut-off from ecoinvent.
Natural sand (kg)	76	187.5	57	200	SBK 296 Industriezand (Milieudatabase, 2020)
Crushed stones (kg)	411	280	410	267	Gravel, crushed {CH}| production | Cut-off from ecoinvent
Weak filler (kg)	23	15	22	15	Limestone, crushed, washed {CH}| production | Cut-off from ecoinvent
Lignin (kg)	24	10	0	0	Modelled as illustrated in section 2.2.1
Linseed oil (kg)	2.9	2	0	0	Linseed seed, at farm {CH}| linseed seed production, at farm | Cut-off from ecoinvent

2021a). The environmental impacts of lignin-based top-layer asphalts using these kraft lignins were retrieved from our other work (Moretti et al., 2021a). For asphalts with kraft lignin used in base layers, the environmental impacts of kraft lignin were described in previous publication (Moretti et al., 2021a). The environmental impacts of base-layer asphalts with kraft lignin are provided in this article for the first time. The environmental impact of conventional asphalts was retrieved from our other work (Moretti et al., 2021a).

3. Results

Table 6 shows the numerical values of the characterized environmental impact per each impact category for the biorefinery lignins assessed.

3.1. AVT lignin

Fig. 2 shows the contributions to the cradle-to-gate climate change impact of AVT lignin with steam produced either from natural gas (AVT-NG) or hog fuel (AVT-BIOM).

Regarding the climate change impact of AVT lignin, natural gas, electricity, and hydrochloric acid emerge as the leading climate change hotspots of this technology. When process steam is produced using natural gas, the climate change impact of AVT lignin (AVT-NG) is dominated by the production of steam from natural gas. The climate change impact of natural gas is mainly (for 83%) due to its combustion. Other important environmental hotspots for AVT-NG and AVT-BIOM lignins are the production of wood chips (assumed from pine softwood) and sodium hydroxide. For AVT-BIOM lignin, steam production from low-value biomass is also a relatively substantial impact. The climate change impact of steam production from hog fuel is mainly broken down into 16% production of bark chips, 40% electricity to operate the furnace and 27% direct emissions (mainly nitrous oxide). Overall, AVT-BIOM lignin performs much better than AVT-NG lignin on climate change because of biogenic carbon dioxide emissions released burning hog fuel (net zero effect on the calculated climate change impact).

Fig. 3 shows the cradle-to-gate environmental impact of AVT lignin with steam produced either from natural gas (AVT-NG) or hog fuel (AVT-BIOM), expressed by the MKI score.

As for climate change, the main environmental hotspots of AVT-NG are natural gas (76% due to the combustion phase and 24% due to upstream emissions), the production of hydrochloric acid and the production of electricity. In particular, compared to the shares of climate change impact of each of these consumables, the production of hydrochloric acid becomes a more critical impact (28.5% in MKI score versus 4.5% in climate change). Fig. 3 shows that, on a weighted basis, human

Table 6
Characterized environmental impact of the lignins assessed (CML-IA baseline V3.06). Results per 1 kg of dry lignin.

Impact category	AVT-NG	AVT-BIOM	VRT-NG	VRT-BIOM
Abiotic depletion	6.30E-06	7.10E-06	9.03E-07	1.59E-06
Abiotic depletion (fossil fuels)	3.00E+01	8.13E+00	2.01E+01	3.33E+00
Global warming (GWP100a)	1.96E+00	5.95E-01	1.26E+00	2.13E-01
Ozone layer depletion (ODP)	3.31E-07	1.28E-07	1.77E-07	2.19E-08
Human toxicity	3.11E-01	4.15E-01	8.00E-02	1.71E-01
Fresh water aquatic ecotox.	2.02E-01	2.49E-01	5.60E-02	9.72E-02
Marine aquatic ecotoxicity	4.07E+02	4.45E+02	1.24E+02	1.61E+02
Terrestrial ecotoxicity	2.54E-03	2.75E-03	5.23E-04	7.43E-04
Photochemical oxidation	2.72E-04	5.21E-04	1.00E-04	3.22E-04
Acidification	2.59E-03	3.14E-03	1.20E-03	1.76E-03
Eutrophication	8.56E-04	1.31E-03	2.98E-04	6.99E-04

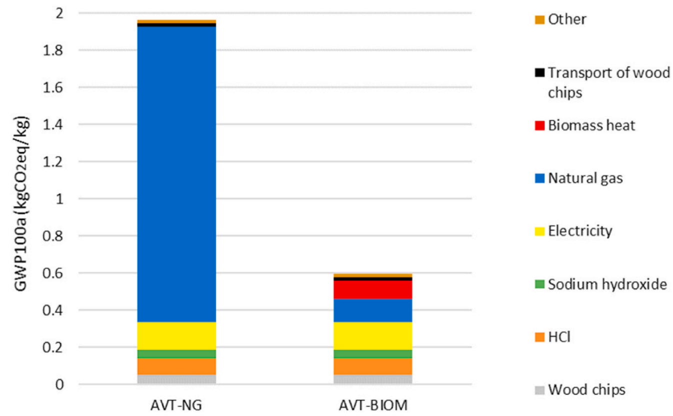

Fig. 2. Cradle-to-gate climate change impact (excl. biogenic carbon removal) of 1 kg of AVT lignin.

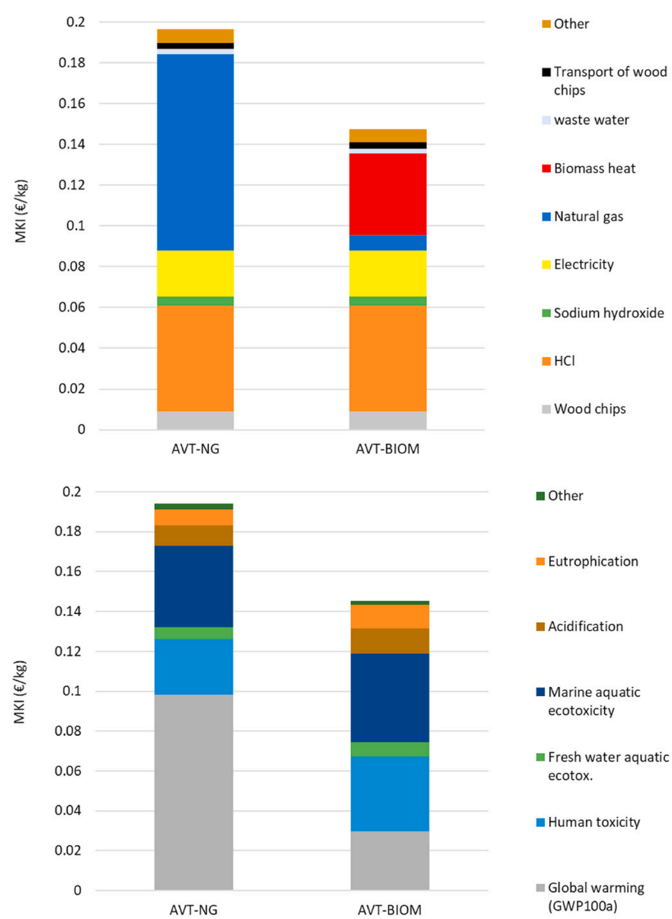

Fig. 3. Cradle-to-gate MKI score of 1 kg of AVT lignin. Breakdown per process contribution (top bars) and impact category (bottom bars). In this figure, the MKI score is without biogenic carbon removal.

toxicity and marine aquatic ecotoxicity are the other two main environmental impacts in addition to climate change. In particular, the production of hydrochloric acid has a significant impact in these two categories (see Fig. 4). The leading causes of said environmental impact are the production of chemicals (mainly liquid chlorine and sodium chloride) and materials (primarily copper) used in the construction of the chemical plant.

Regarding AVT-BIOM lignin, the environmental impact of biomass heat (from hog fuel) is more significant in terms of MKI score than

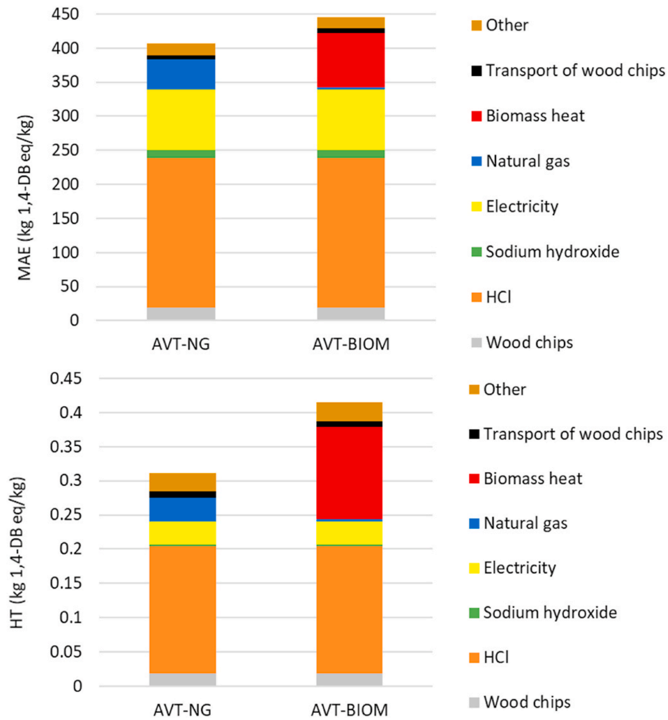

Fig. 4. Cradle-to-gate impacts of 1 kg of AVT lignin in terms of marine aquatic ecotoxicity (MAE) in the top graph and human toxicity (HT) in the bottom graph.

climate change. The reason can be found in the impact of biomass heat on marine aquatic ecotoxicity and human toxicity (see Fig. 4). For human toxicity, the impact of biomass heat is mainly caused by the combustion of hog fuel (94%). The human toxicity impact caused by the combustion of hog fuel is mainly (52%) due to direct combustion emissions. Regarding marine aquatic ecotoxicity, the impact of hog fuel combustion is mainly due to the production of the materials (especially steel and copper) used for the furnace and the electricity for the operation of the furnace. However, given the important climate change benefits from using process steam from hog fuel, AVT-BIOM lignin has a 25% lower MKI score than AVT-NG lignin.

3.2. VRT lignin

Fig. 5 shows the cradle-to-gate climate change impact of VRT lignin with steam produced either from natural gas (VRT-NG) or hog fuel (VRT-BIOM).

As for AVT lignin, if process steam is obtained from natural gas, its combustion (and production) dominates the climate change impact of 1 kg of VRT-lignin at the biorefinery gate. For VRT lignin, other (small) climate change impacts are methanol production, electricity, cooling water and sawdust. At the same time, the impact of the production of sulfuric acid and sodium hydroxide is negligible. For VRT-BIOM, the climate change impact of biomass heat is also important but minor compared to heat from natural gas in VRT-NG. The climate change impact of biomass (hog fuel) heat is mainly due to combustion emissions (mainly nitrous oxides) and electricity to operate the furnace.

In terms of MKI scores, the environmental impact of VRT lignin is dominated by the production of steam for both VRT-NG and VRT-BIOM lignins (see Fig. 6). Besides climate change, other significant environmental impacts are marine aquatic ecotoxicity and human toxicity (and, to a certain extent, eutrophication and acidification). Given the important climate change benefits of steam from hog fuel over steam from natural gas, the MKI score of VRT-BIOM has a 37% lower MKI score. Other important impacts are the production of sawdust, methanol, electricity and cooling water used by VRT biorefinery. As for AVT-lignin, the fraction of the MKI score of VRT lignin caused by biomass heat is affected mainly by its impact on marine aquatic ecotoxicity and human toxicity (see Figs. 6 and 7). In these two categories, methanol, sawdust, cooling water and electricity are also important, as shown by Fig. 7. For methanol production, the environmental impact in these two categories is mainly caused by materials production (such as molybdenum and copper), natural gas and electricity production. For sawdust, the human toxicity impact is dominated by transportation and electricity production, while the marine aquatic ecotoxicity is mainly influenced by electricity production (e.g. used for aspiration of sawdust). For cooling water, the electricity consumption dominates the marine aquatic ecotoxicity impact and is the main environmental impact of human toxicity (accounting for 31% of the human toxicity impact of cooling water). For human toxicity, the production of the pumps and the steel used to build the cooling tower are also important human toxicity sources.

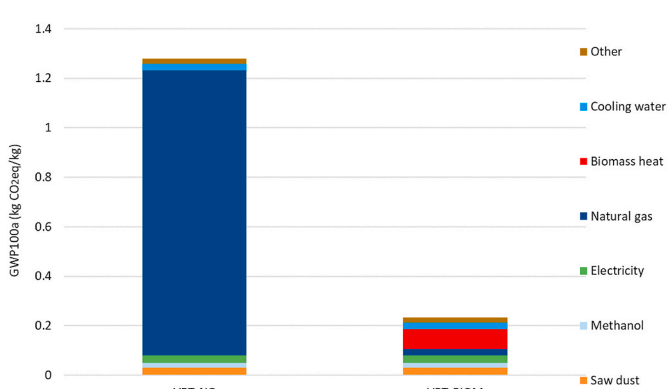

Fig. 5. Cradle-to-gate climate change impact (excl. biogenic carbon removal) of 1 kg of VRT lignin.

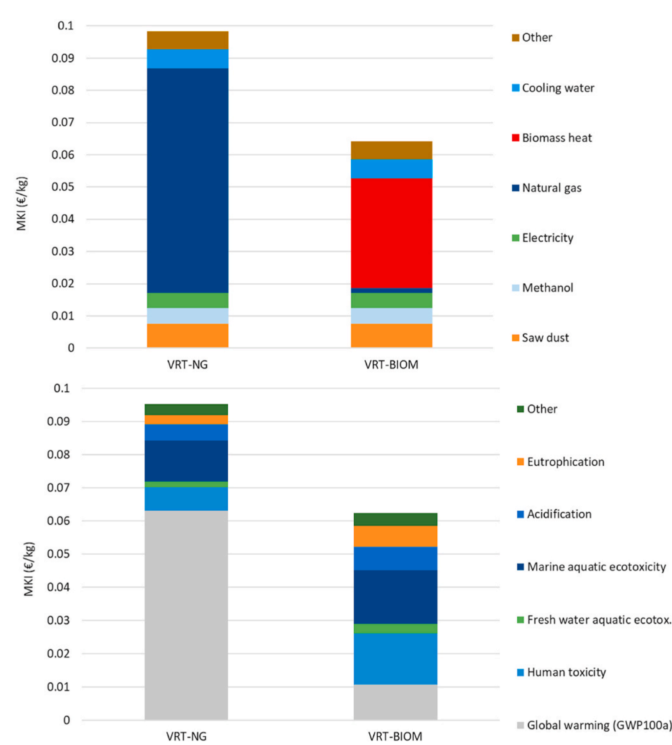

Fig. 6. Cradle-to-gate MKI score of 1 kg of VRT lignin. Breakdown per process contribution (top bars) and impact category (bottom bars). In this figure, the MKI score is without biogenic carbon removal.

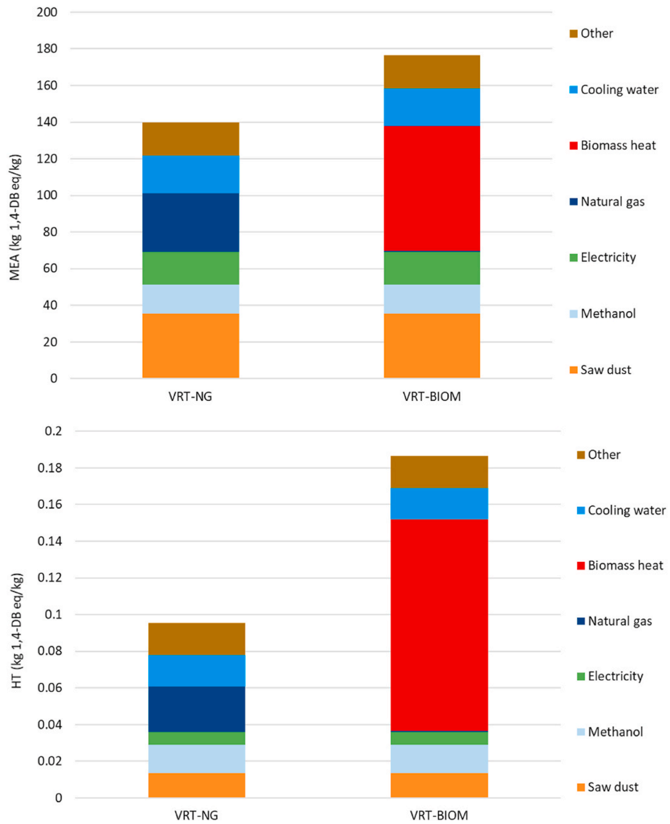

Fig. 7. Cradle-to-gate impacts of 1 kg of VRT lignin in terms of marine aquatic ecotoxicity (MAE) in the top graph and human toxicity (HT) in the bottom graph.

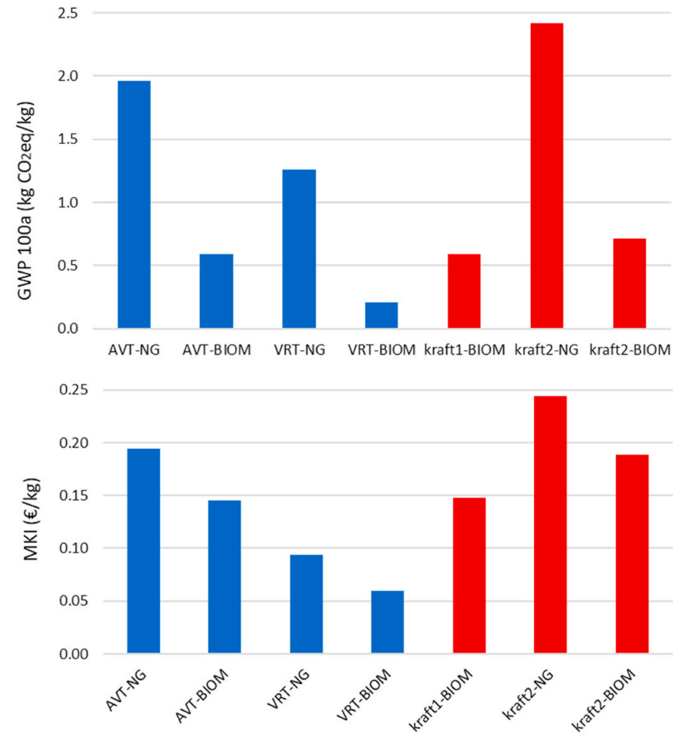

Fig. 8. Comparing the cradle-to-gate impacts of 1 kg of biorefinery lignins (blue) versus 1 kg of kraft lignins (red) in terms of climate change (top) and MKI score (bottom). Environmental impacts of kraft lignins retrieved from our other work (Moretti et al., 2021a).

3.3. Comparing lignins from biorefineries and kraft mills

Fig. 8 shows the climate change impact and environmental impact expressed in terms of the MKI score of the biorefinery lignins assessed in this study compared to kraft lignins.

If steam for lignin production is produced from natural gas, both AVT lignin and VRT lignin have lower climate change and MKI scores compared to kraft lignin. In all these cases, the impact is dominated by the combustion and production of natural gas used for steam. However, another major factor is the (economic) allocation factor that allocates the environmental burden between lignin and other outputs of the biorefinery or pulp mill and which depends on the type and amounts of co-products produced. For example, the AVT biorefinery uses a 43.5% lower amount of natural gas per kg of lignin produced than VRT biorefinery. However, the climate change impact of AVT-NG lignin is higher than the one of VRT lignin. The reason is that the economic allocation factor of AVT-lignin was 44%, while the one of VRT lignin was 20%.

These results are attained using the assumption that the price of biorefinery lignins is the same as kraft lignins. However, the market of biorefinery lignins is still emerging. Generally, the price of lignin depends on its quality, which is mainly based on the percentage of impurities present in the lignin and whether it is used in niche or bulk markets. So, assuming the same price means that the quality of the two lignins is comparable, and an asphalt producer would not prefer one lignin to the other. Given the broad price range of lignins on the market for various quality levels, a sensitivity analysis on the price assumed for lignin and its effect on the allocation method was conducted in section 4. Moreover, other allocation methods might be applicable based on the reference document used to perform the LCA. For this reason, an allocation based on a physical property was also considered (mass allocation) in the same sensitivity analysis. Other methodological and data uncertainties are broadly discussed in section 5.

Regarding lignins where hog fuel is used for steam production, AVT-BIOM lignin showed a climate change impact in line with the one of kraft-lignin and a much lower MKI score than kraft-lignin. VRT-BIOM lignin performed significantly better than kraft lignin from climate change and MKI score perspectives.

3.4. Top layer asphalts with AVT and VRT lignins

Fig. 9 shows the climate change impact and environmental profile (in MKI score) for bio-based asphalts used in top layers using AVT and VRT lignins divided into life cycle stages.

Regarding the environmental impacts of bio-based asphalts, the construction phase (A5) and use phase (B1) have both negligible climate change impact and MKI scores. This aspect is typical for the environmental impacts of asphalts assessed following Dutch Product Category Rules (NL-PCRs) as shown in our other work (Moretti et al., 2021a) and by the Dutch LCA asphalt sector report (Schwarz et al., 2020). On the other hand, the climate change impact of phase A1 (asphalts components-other than lignins), typically dominated by the climate change impact of bitumen production, is significantly reduced in bio-based asphalts since lignin partially replaces bitumen. In particular, the four product systems investigated (asphalts using various bio-refinery lignins) differ only for the type of lignin used. For all lignins, the climate change impact is overall negative in phase A1 (A1: lignin-incl. biogenic carbon). This means that the climate change impact generated during lignin production is lower than the equivalent carbon dioxide of the biogenic content of the lignin itself. In the case of VRT-lignin, given the lower MKI score than AVT lignin, this is also reflected in the negative MKI score of the same phase (A1: lignin-incl. biogenic carbon). This means that in the case of VRT-lignin, the climate change benefits of lignin (weighted in MKI score) fully compensate the total environmental impact (expressed in MKI score as

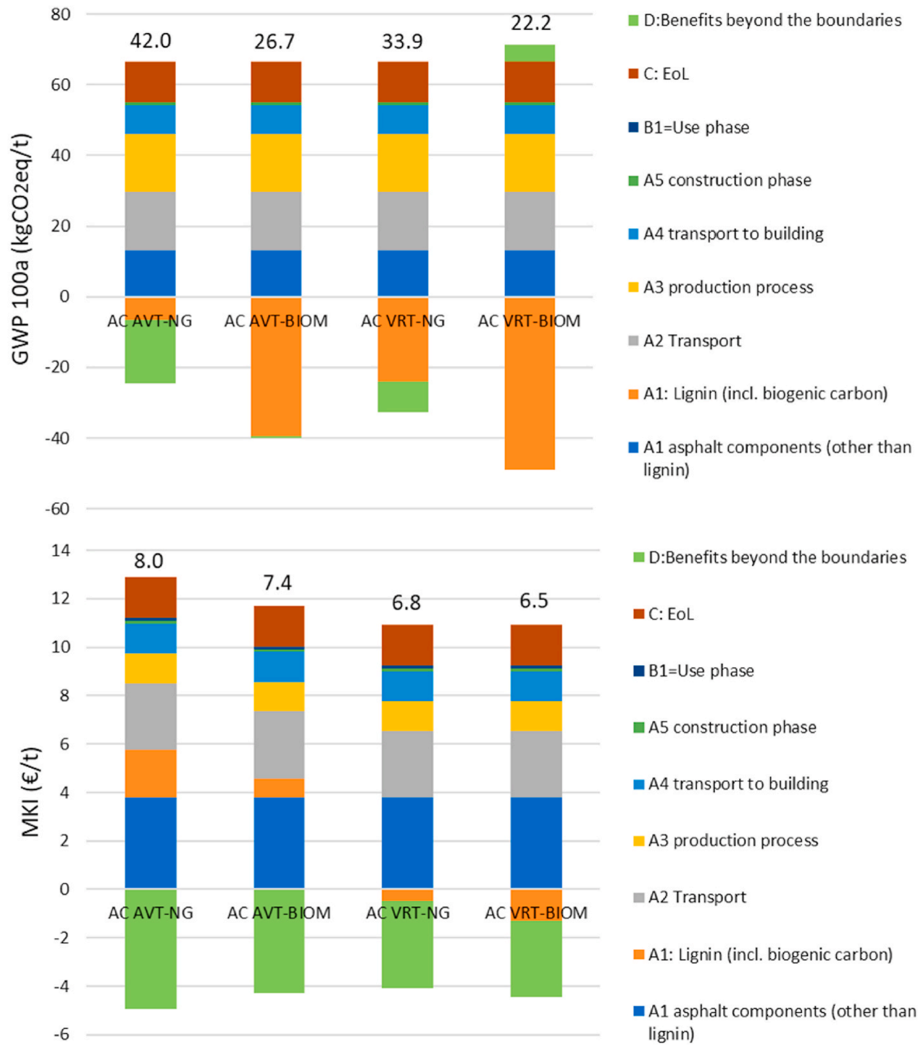

Fig. 9. Cradle-to-grave climate change impact (top) and MKI score (bottom) of 1 t of bio-based asphalt for a top layer using biorefinery lignin. Breakdown per life cycle stage.

well) of lignin production via VRT biorefinery.

Regarding the climate change benefits from recycling primary raw materials (module D), these benefits are significant for the asphalts using AVT-NG and VRT-NG, negligible for AVT-BIOM and become an impact for VRT-BIOM. The relatively small climate change impact of module D for VRT-BIOM means that the benefits from 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. In fact, in module A1 (life cycle stage representing the extraction and processing of the raw materials), the entire biogenic carbon removal was included in the climate change impact of lignin but the top layer asphalt contributes storing only a fraction of such a biogenic carbon during its life cycle. The rest of the biogenic carbon will be stored by the recycled asphalt in the following life cycle. Hence, the benefit for biogenic carbon storage needs to be distributed over various life cycles. For a complete understanding of how the biogenic carbon was accounted for, such modeling can be found in section 2.2.2 and further details in our other work (Moretti et al., 2021a).

Overall, VRT lignin showed a better overall environmental performance (MKI score) than AVT lignin. VRT lignin shows a lower climate change impact than AVT lignin only if the same source for steam production is assumed.

3.5. *Comparing top layer asphalts*

Fig. 10 compares the environmental impacts between three types of top-layer asphalts i.e. the investigated asphalts using biorefinery lignins, asphalts using kraft lignins and conventional asphalts.

This study confirms the findings of our other work (Moretti et al., 2021a) that 1) lignin-based asphalts have a significantly lower climate change impact than conventional asphalts and 2) higher climate change benefits are achieved if low-value biomass (hog fuel) is used for steam. In particular, top-layer asphalts using biorefinery lignins showed possible climate change benefits compared to conventional asphalts estimated between 35% and 70% depending on the biorefinery lignin and conventional asphalt considered. These savings of climate change impact are either similar or higher than those obtained for top-layer asphalts using kraft lignin.

Compared to conventional asphalts (MKI score), the overall environmental benefits are between 6% and 12% for top-layer asphalts using AVT lignin. Top-layer asphalts using VRT lignin show up to a 24% lower MKI score than conventional asphalts. The MKI score of top-layer asphalts using biorefinery lignins is generally lower than for the case of kraft lignins (which is intuitive looking at the comparison shown in section 4.3 for the lignins themselves). For both lignins, lower MKI scores are obtained if hog fuel is used for steam production.

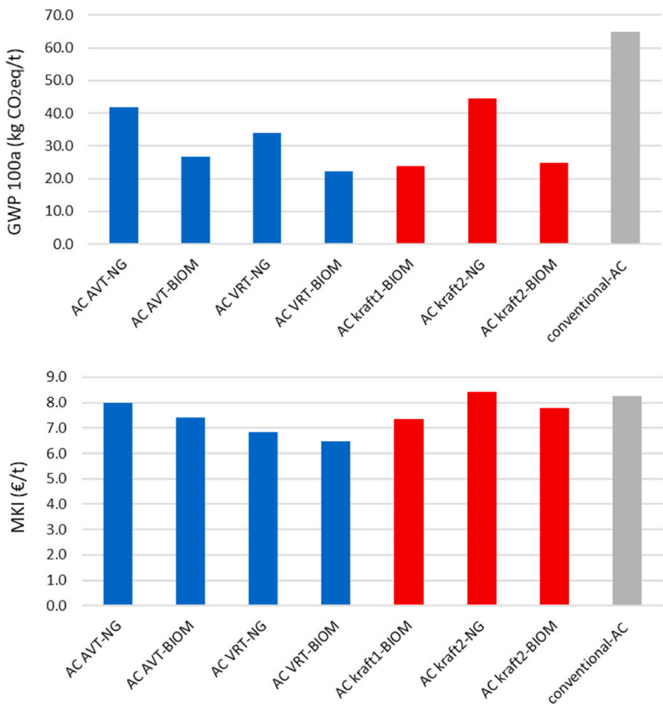

Fig. 10. Comparing the cradle-to-grave impacts of 1 t of top-layer asphalt using biorefinery lignins (blue) using kraft lignins (red) and conventional asphalts (grey). Top graph for climate change. Bottom graph for MKI score (bottom).

3.6. *Base-layer asphalts with AVT and VRT lignins*

Fig. 11 shows the climate change impact and environmental profile (in MKI score) for bio-based asphalts used in base layers using AVT and VRT lignins divided into life cycle stages.

It is seen that the climate change impacts of base-layer asphalts derived from (biorefinery) lignins are comparable (a bit higher or lower) to the impact of top-layer asphalts, while the MKI scores are lower than those for top-layer asphalts using the same lignins. The reason is that the increase in burden-free recycled content reduces both the climate change and MKI score. Conversely, the lower amount of virgin lignin in base layers than top layers leads to lower net-climate change benefits from the biogenic carbon removal.

3.7. *Comparing base-layer asphalts*

Fig. 12 shows the comparison of the environmental impacts between three types of base-layer asphalts i.e. the investigated asphalts using biorefinery lignins, asphalts using kraft lignins and conventional asphalts.

Asphalts with biorefinery lignins were confirmed to have a much lower climate change impact than conventional asphalts of the order of 25–50%. For base-layer asphalts, the differences in climate change impacts between biorefinery lignins and kraft lignins are lower than for top-layer asphalts. The main reason is the lower amount of virgin lignin (and bitumen) in the base layer compared to the top layer. For the same reason, the mitigation of climate change impact of bottom layer asphalts by using lignin showed to be lower than for top-layer asphalts (25–50% versus 35–70%). As for top-layer asphalts, the MKI of asphalts using biorefinery lignins is generally lower than for the case of kraft lignins and conventional asphalts. Compared to base-layer conventional asphalts, using biorefinery lignins can lead to savings up to 15% (MKI score).

4. Sensitivity analysis

Since kraft lignin has a more established market and can be considered the main competitor of biorefinery lignins for asphalt application, an average market price of 535 €/t was assumed based on the current price of kraft lignin with medium quality. However, the price of lignins of adequate quality for asphalt application can range between 370 and 700 €/t (Moretti et al., 2021b). A sensitivity analysis was conducted considering these two prices, i.e. 370 and 700 €/t. In particular, the economic allocation share of AVT lignin using this range varies between 35% and 51% (44% used in the baseline calculations) while the one of VRT lignin between 15% and 24% (20% used in the baseline calculations). The results with a different price assumed for lignin were compared with the ones that would have been obtained using mass allocation. The mass allocation share of AVT lignin (30%) is lower than the shares using economic allocation. For VRT lignin, the mass allocation factor (22%) is between the two economic allocation shares with the highest and lowest price.

Sensitivity results were compared with the results of kraft1-BIOM lignin using the same assumptions. For kraft1-BIOM, mass allocation assigns more impact to lignin than economic allocation (with both highest or lowest price assumed).

The sensitivity analysis results on the environmental impact of lignins can be found in Fig. 13. Kraft2-NG and kraft2-BIOM were also kept in the comparison graphs of this sensitivity analysis, but their allocation method remained unchanged due to the lack of data. A physical causality allocation was applied to these two kraft lignins; for more details, see Moretti et al. (2021a).

This sensitivity analysis shows that increasing the price of lignin used for calculating the allocation share favors kraft lignin over biorefinery lignins. In contrast, a lower price of lignin favors biorefinery lignins over kraft lignins. For example, this means that VRT-BIOM has a 70% lower climate change impact than kraft1-BIOM assuming the lowest price for lignin, and 66% taking the highest price for lignin. As mentioned above, what happens to the environmental burdens using mass allocation compared to using economic value is strongly dependent on the specific lignin considered. We observed that for VRT lignin, using mass allocation provides environmental impacts in line with the baseline results given the similar allocation share (22% with mass allocation versus 20% with economic allocation based on average price). Conversely, mass allocation decreases the environmental impact of AVT lignin significantly compared to the baseline environmental impact (from 44% of the impact allocated to lignin to 30%). As a result, if a mass allocation is applied to both lignins, the environmental impact of the two lignins becomes closer (see Fig. 13). This is also due to the difference in the biomass feedstock that influences the amount of lignin that can be produced and also has an influence on the impact allocated to each kg of product with the economic allocation share.

Nonetheless, VRT-BIOM lignin still shows a better environmental performance for both climate change and MKI score perspectives. Conversely, the climate change impact of AVT-NG becomes slightly lower than VRT-NG, and the MKI score of AVT-BIOM becomes the same as VRT-NG. Given the minor difference in the biogenic carbon content (see section 2.2.1), this means that the asphalt's climate change mitigation potential is the same for both AVT-NG and VRT-NG when applying mass allocation. Likewise, the MKI score of asphalts using AVT-BIOM becomes similar to the one of asphalt using VRT-NG.

5. Discussion

5.1. *Multifunctionality*

Lignin is always a product resulting from a multi-output process. Such a process can be a biorefining process or a pulp mill process. In both cases, lignin is a co-product obtained from producing another product (energy product or materials in the case of a biorefinery or pulp

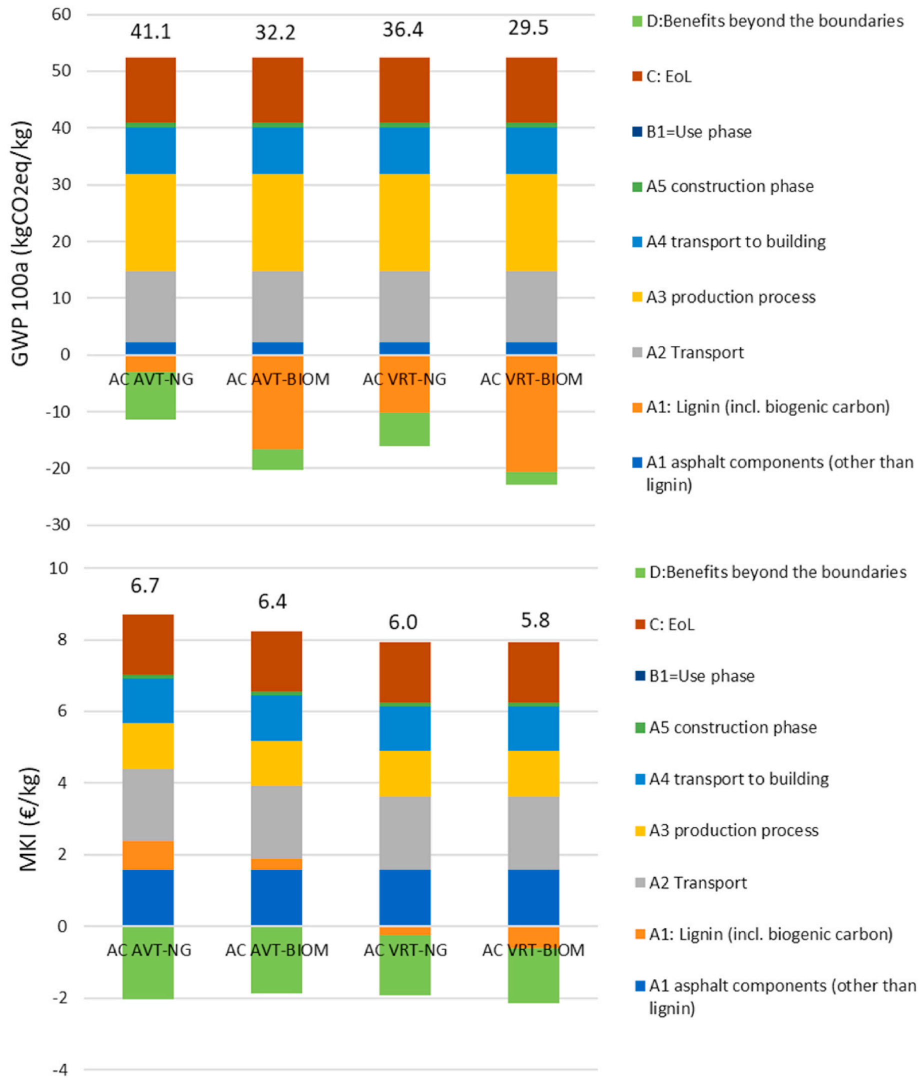

Fig. 11. Cradle-to-grave climate change impact (top) and MKI score (bottom) of 1 t of bio-based asphalt for base layers using biorefinery lignin. Breakdown per life cycle stage.

for pulp mills). Allocating the environmental impact of a multi-output process among the various products produced by the process is a historical challenge in LCA. In fact, applying a different allocation method can lead to significantly different environmental impacts, especially for the by-products like lignin. For this case study, a sensitivity analysis on the allocation method can be found in section 4. A comprehensive investigation of allocation methods that have been applied in the literature to lignin can be found in a recent study by Hermansson et al. (2020).

In our study, an economic allocation was used to apportion the environmental impact of the biorefineries between lignin and its co-products. However, using a method that relies on a market price can be sensitive to the exact market price used for the allocation method. In particular, lignin price depends on quality specifications that vary depending on the level of impurities of that specific lignin. Based on the lignin quality, a broader range of applications for lignin is possible. As there is no established market for biorefinery lignins yet, the assumed price is uncertain. For example, it could be that the market price of the lignin produced by the two biorefineries will be significantly different depending on the purity achieved on real operation at a large scale and consequently on the application for which they will be used, but also other future applications of lignin and subsequent demand. The price of industrial sugars might also increase faster than lignin if the demand for sugar increases due to offsets for oil use, lowering the allocation factor for lignin.

5.2. Biogenic carbon storage and recycling

In the so-called “circular (bio)economy”, it becomes crucial to properly account for the use of secondary raw material and biomass feedstock into recycled bio-based products (Poluzzi et al., 2020; Stegmann et al., 2020). Our study accounted for the environmental benefits of storing biogenic carbon in asphalts. The assumption was that lignin-bitumen mixes do not biodegrade, neither in asphalt nor when lost in the environment due to erosion nor when used in foundations in subsequent life cycles. Biogenic carbon can be considered permanently stored beyond a long and specified time-horizon set by convention. In this study, above 100 years, such biogenic carbon was considered to be permanently stored. Therefore, the part of the carbon contained in the virgin lignin was calculated as a climate change credit since it will be stored permanently over various cycles in the following asphalt products. In particular, the asphalt was entitled to credit only for the virgin lignin’s carbon that it contributed to store. In our study, a simplified time-based credit was applied for allocating biogenic carbon permanently over several life cycles, e.g., if an asphalt has stored a certain amount of carbon that goes to the following life cycle for 15 years, it was

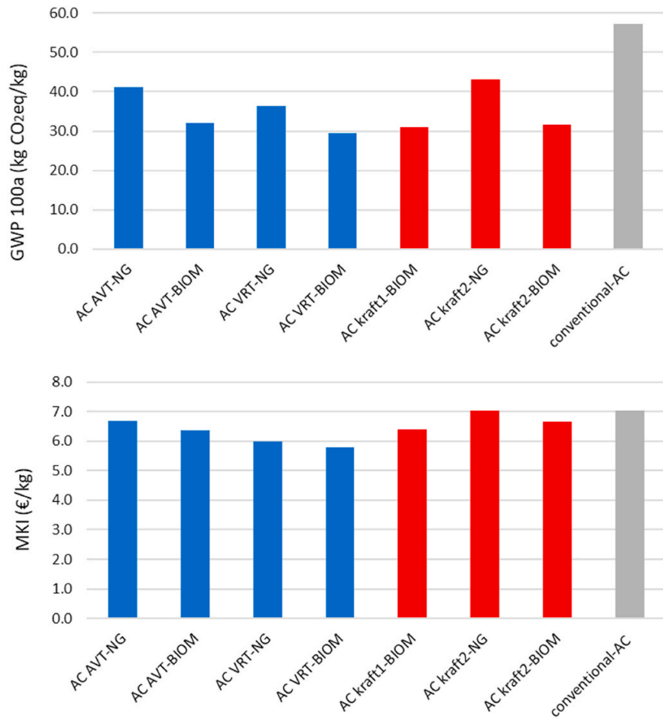

Fig. 12. Comparing the cradle-to-grave impacts of 1 t of base-layer asphalt using biorefinery lignins (blue) with asphalts using kraft lignins (red) and conventional asphalts (grey). Top graph for climate change. Bottom graph for MKI score (bottom).

entitled to 15% of such biogenic carbon permanently storage.

An inconsistency issue arises if the recycled content is derived from asphalt with a lignin percentage. According to the NL-PCR (Keijzer et al., 2020), such recycled content is free of the environmental burden in module A1 and does not lead to any environmental benefits in module D. Hence, since the environmental impact of producing the recycling content is not incorporated in module A1, the biogenic carbon contained in the recycled content should also not be incorporated in module A1. However, in this way, the fate of the remaining part of the permanently stored biogenic carbon is not tracked. This problem arises from accounting for module D that it is inconsistent with the rest of the modeling according to NL-PCR (Keijzer et al., 2020). In fact, the NL-PCR (Keijzer et al., 2020) assumes that the first cycle takes all the burden of producing a material, and such material becomes burden-free in the following cycle. Hence, it is inconsistent with subtracting the credit for the benefits resulting from recycling primary raw materials at the end of the cycle (module D).

5.3. Environmental cost indicator

The environmental cost indicator (the so-called MKI score in the Dutch context) is helpful since it allows merging multiple environmental impacts into a single-score indicator. In this way, LCA results become easy to compare and use in contract requirements for tender purposes. Similar methods may increasingly be used in public tenders of infrastructure projects such as roads, airports and railways (Moretti et al., 2017).

However, using a single score to unite different environmental impacts reduces transparency, increases the uncertainties and introduces subjective weighting of individual impact categories. Using a different weighting factor might lead to a different outcome in the comparison (Reap et al., 2008). For example, there are different monetized weighting leading to the same units (e.g. €). However, even for the same unit, the results cannot be considered comparable or additive, nor can they be combined (Reap et al., 2008).

In particular, the environmental cost indicator used in this study relies on the Centrum voor Milieuwetenschappen in Leiden (CML) impact assessment method (Keijzer et al., 2020). The CML method does

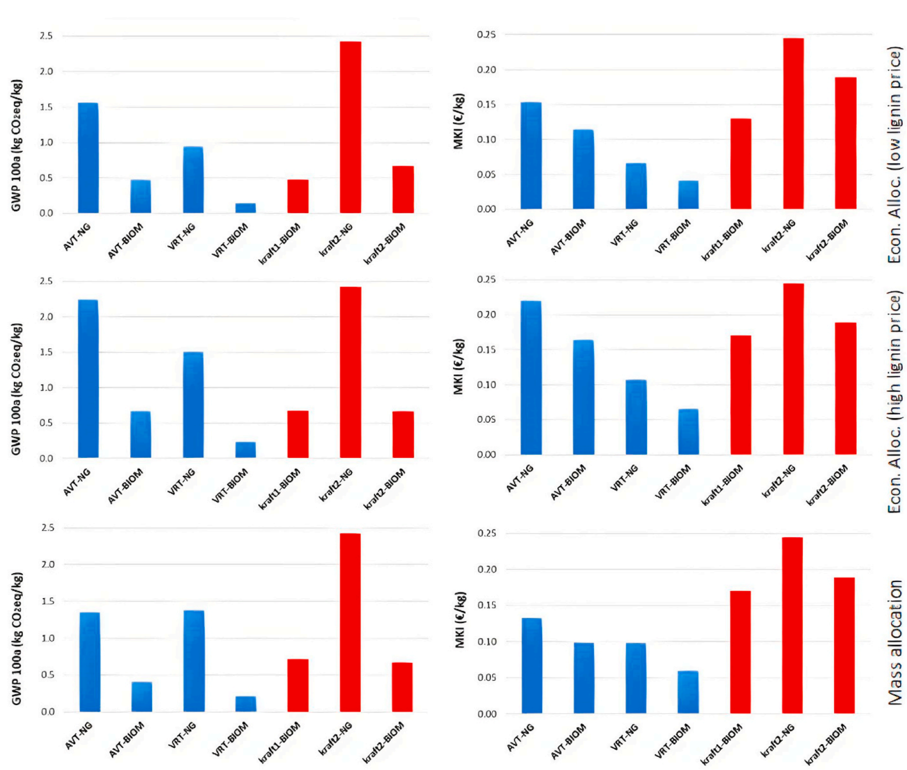

Fig. 13. Comparison of cradle-to-gate impacts of 1 kg of lignin varying the allocation shares. Biorefinery lignins in blue and kraft lignins in red. Environmental impacts of Kraft2-NG and kraft2-BIOM lignins (unchanged due to lack of data) retrieved from Moretti et al. (2021a).

not account for certain impacts included by other methods such as land use or water use. Moreover, the CML method does not distinguish between freshwater and marine eutrophication and between human toxicity with cancer and without cancer effects. Furthermore, in our case study, marine aquatic ecotoxicity and human toxicity were significant environmental impacts on a weighted basis. However, there is an important ongoing effort to standardize and decrease the uncertainty behind the calculation of the environmental impacts for these two categories (Saouter et al., 2018). In fact, there are difficulties in interpreting the differences in the results obtained for similar products from different LCA databases for these two categories (European Commission, 2019; Saouter et al., 2018). For this reason, given their low "reliability", some authors exclude these categories in their comparative assessments (European Commission, 2019; Moretti et al., 2021c).

5.4. Data quality and uncertainty

LCAs of lignin production are mainly based on laboratory data, process modeling, or literature data (Moretti et al., 2021b). In this study, despite new transparent primary data for lignin production were collected from the industry, they are not based on real operation at a large scale. For this reason, they are more affected by uncertainties than data collected from actual operating plants. In the case of AVT lignin, data were available only for a first of a kind plant. In the future, scaling down for example the steam consumption (proper heat integration) and optimization of the chemicals and further waste valorization could further improve AVT lignin's environmental profile. Moreover, the plant could also operate with a different feedstock in the future. Regarding background data, uncertainty also applies to the datasets retrieved from the ecoinvent database. Despite being one of the most used databases for conducting LCAs, the datasets from ecoinvent have uncertainties, and the assumption of average technology data is not always robust. Ecoinvent datasets contain both model uncertainties (e.g. using linear instead of non-linear modeling) and mistakes made by human errors not caught by the subsequent validation and review (Weidema et al., 2013). Moreover, the assumption that the biogenic carbon of the lignin in the asphalt mixture does not biodegrade should be validated in future research via lab testing on asphalt samples at the end of their life.

5.5. Other lignin-valorization options: climate change mitigation

From a climate change perspective, top-layer asphalts using biorefinery lignins showed a 35–70% lower climate change impact than conventional asphalts. For base-layer asphalts, the climate change benefits were quantified in 25–50%. These percentages are much higher than the 6% mitigation potential calculated by (Tokede et al., 2020). However, the study of Tokede et al. assumed only 25% bitumen replacement and the end of life and module D were not considered. In our study, the replacement of bitumen was 47% for top layers and 70% for base layers. The calculated climate change benefits are in line with the one obtained for lignin-based polypropylene (Liao et al., 2020), adipic acid (Corona et al., 2018), lignin-based transportation fuels (Obydenkova et al., 2017) and phenol (Liao et al., 2020). They are, however, much higher than calculated by the LCA literature for lignin-based adhesives (McDevitt and Grigsby, 2014), catechols (Montazeri and Eckelman, 2016) and wood laminates (Hildebrandt et al., 2019). Nonetheless, a direct comparison of climate change mitigation potentials can be hampered by methodological choices e.g allocation practices and biogenic carbon modeling and data quality and technical aspects e.g. the amount of lignin in the product and the type of energy used (Moretti et al., 2021b). Regarding technical aspects, a higher carbon mitigation potential for lignin-based top-layer asphalt than lignin-based base-layer does not mean that it is better to use lignin in top-layer asphalt than in base-layers.

6. Conclusions

Previous LCAs highlighted that the environmental performance of lignin-based products compared to petrochemical products is strictly linked with the specific lignin considered with respective production process as well as energy sources and chemicals employed. This study investigated the environmental impact of lignins from two different biorefineries (Avantium, named AVT and Vertoro, named VRT) and their potential use as ingredients for bio-based asphalts.

The production of steam, chemicals and electricity was identified as the primary source of environmental impacts of lignin production from biorefineries. The most impacting chemicals were hydrochloric acid for the AVT biorefinery and methanol for the VRT biorefinery. Using steam from low-value biomass (hog fuel) instead of natural gas could significantly reduce climate change impact and, given the importance of such impact category, the overall environmental impact (expressed as environmental cost indicator: so-called MKI score).

Overall, when applying economic allocation and the same steam source is considered for both biorefineries, VRT lignin has lower climate change and overall environmental impact than AVT lignin. Two reasons play a major role in the lower impact of VRT lignin: 1) the lower lignin economic allocation factor and 2) lower impact of methanol production compared to hydrochloric acid. When mass allocation is applied, the difference in the environmental impact becomes smaller.

Moreover, assuming the same steam source, both biorefinery lignins showed lower climate change and MKI scores than kraft lignin. Top-layer asphalts using biorefinery lignins showed a climate change mitigation potential between 35% and 70% replacing conventional asphalts. Depending on the biorefinery and steam source considered, such savings are comparable or higher than those allowed by bio-based asphalts with kraft lignin. Top-layer asphalts with AVT and VRT lignin showed respectively a 6–12% and 20–24% lower MKI score. In both cases, a lower MKI score was obtained if steam production used hog fuel. Hence, this option should be considered by the industry given the priority agenda on climate change. However, trade-offs occur in other categories and could lead to a higher cost compared to natural gas and might conflict with the economic viability of the biorefinery.

Base-layer asphalts have higher recycled content and a lower amount of bitumen and lignin. For this reason, base-layer asphalts have a lower environmental impact than conventional asphalt. This is also the case for the climate change impact of conventional asphalt. However, this is not reflected in the climate change impact of lignin-based asphalts. In fact, base-layer asphalts using lignin have a climate change impact comparable to top-layer asphalts due to the lower amount of virgin lignin that leads to lower climate change benefits from the biogenic carbon removal. Given the decrease of climate change impact of conventional asphalts, the climate change benefits are reduced to 25–50% but remain substantial.

Hence, replacing bitumen in the top and base-layer asphalts with lignin from lignocellulosic biorefineries shows significant climate change benefits. Since a local lignin surplus is projected to become available in the future, this option could contribute to reaching the 2050 Dutch climate change mitigation targets. In the Netherlands, 8.3 Mt of asphalt is produced yearly (Urgenda, 2020). Indicatively, 30 kg CO_2eq per t of average asphalt could be saved using bio-based asphalts with lignin from lignocellulosic biorefineries. If enough lignin is available to replace 1% of Dutch asphalts with lignin-based asphalts, 21 kt of CO_2eq could potentially be saved yearly.

As shown in this study and our earlier study, the environmental performance of lignin-based asphalts compared to conventional asphalts is strictly linked to the specific lignin considered and respective production process as well as the specific composition of the asphalts. While our findings regarding a better environmental performance of lignin-based asphalts cannot be easily generalized to all lignin-based asphalts, we can conclude that lignin-based asphalts with promising environmental performances typically have 1) high rates of bitumen replacement in asphalts per kg of lignin and 2) use renewable process energy to replace lignin as an energy carrier in the lignocellulosic biorefinery as well as low carbon intensity chemicals for lignin extraction. Moreover, the environmental performance of durable bio-based applications like asphalts benefits from biogenic carbon storage accounting. So, LCAs neglecting the accountancy of biogenic carbon storage over multiple cycles could penalize this type of bio-based product.

CRediT authorship contribution statement

Christian Moretti: Conceptualization, Methodology, Software, Data curation, Formal analysis, Investigation, Visualization, Writing – original draft, Writing – review & editing. **Ric Hoefnagels:** Data curation, Formal analysis, Investigation, Writing – review & editing, Project administration, Funding acquisition. **Marco van Veen:** Data curation, Investigation, Methodology, Visualization, Writing – original draft, Writing – review & editing. **Blanca Corona:** Data curation, Investigation, Methodology, Visualization, Writing – original draft, Writing – review & editing. **Svetlana Obydenkova:** Data curation, Investigation, Methodology, Visualization, Writing – review & editing. **Scott Russell:** Data curation, Investigation, Methodology, Visualization, Writing – review & editing. **Anna Jongerius:** Data curation, Investigation, Methodology, Visualization, Writing – review & editing. **Iris Vural-Gürsel:** Conceptualization, Data curation, Investigation, Methodology, Writing – review & editing. **Martin Junginger:** Conceptualization, Methodology, Data curation, 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: Svetlana Obydenkova, Scott Russel and Anna Jongerius are employed by industrial partners that could have financial interests in the technology investigated by this article. 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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References

Bajwa, D.S., Pourhashem, G., Ullah, A.H., Bajwa, S.G., 2019. A concise review of current lignin production, applications, products and their environment impact. Ind. Crop. Prod. 139 https://doi.org/10.1016/j.indcrop.2019.111526.

Benali, M., Ajao, O., Jeaidi, J., Gilani, B., Mansoornejad, B., 2016. Integrated lignin-kraft pulp biorefinery for the production of lignin and its derivatives. economic assessment and LCA-based environmental footprint 379–418. https://doi.org/10.1007/978-981-10-1965-4_13.

Bergius, F., 1937. Conversion of wood to carbohydrates and problems in the industrial use of concentrated hydrochloric acid. Ind. Eng. Chem. 29, 247–253. https://doi.org/10.1021/ie50327a002.

Bernier, E., Lavigne, C., Robidoux, P.Y., 2013. Life cycle assessment of kraft lignin for polymer applications. Int. J. Life Cycle Assess. 18, 520–528. https://doi.org/10.1007/s11367-012-0503-y.

Bishop, G., Styles, D., Lens, P.N.L., 2021. Environmental performance comparison of bioplastics and petrochemical plastics: a review of life cycle assessment (LCA) methodological decisions. Resour. Conserv. Recycl. 168 https://doi.org/10.1016/j.resconrec.2021.105451.

Bouwkwaliteit, S., 2019. Determination Method Environmental performance Buildings and civil engineering works, 31, pp. 1–83.

Brovelli, C., Crispino, M., Pais, J.C., Pereira, P.A.A., 2014. Assessment of fatigue resistance of additivated asphalt concrete incorporating fibers and polymers. J. Mater. Civ. Eng. 26, 554–558. https://doi.org/10.1061/(asce)mt.1943-5533.0000837.

Carpos, P., De Vita, A., Tasios, N., Siskos, P., Kannavou, M., Preopoulos, A., Evangelopoulou, S., Zampara, M., Papadopoulos, D., Nakos, C., Paroussos, L., Fragiadakis, K., Tsani, S., Karkatsoulis, P., Fragkos, P., Kouvaritakis, N., Höglund-Isaksson, L., Winiwarter, W., Purohit, P., Gomez-Sanabria, A., Frank, S., Forsell, N., Gusti, M., HAvlik, P., Oberseiner, M., Witzke, H.P., Kestling, M., 2016. EU reference scenario 2016 - energy, transport and GHG emissions - trends to 2050, energy, transport and GHG emissions - trends to 2050. https://doi.org/10.2833/9127.

CEN, E.C.F.S., 2013. EN 15804:2013 - standards Publication Sustainability of construction works — environmental product declarations — core rules for the product category of construction products. Int. Stand.

Chen, M., Zhang, J., Wang, Y., Tang, Z., Shi, J., Wang, C., Yang, Z., Wang, J., Zhang, H., 2021. Lignin catalytic depolymerization for liquid fuel and phenols by using Mo/sepiolite catalysts calcined at different temperature. J. Environ. Chem. Eng. 9, 105348. https://doi.org/10.1016/j.jece.2021.105348.

Corona, A., Biddy, M.J., Vardon, D.R., Birkved, M., Hauschild, M.Z., Beckham, G.T., 2018. Life cycle assessment of adipic acid production from lignin. Green Chem. 20, 3857–3866. https://doi.org/10.1039/c8gc00868j.

Culbertson, C., Treasure, T., Venditti, R., Jameel, H., Gonzalez, R., 2016. Life cycle assessment of lignin extraction in a softwood Kraft pulp mill. Nord. Pulp Pap. Res. J. https://doi.org/10.3183/npprj-2016-31-01-p030-040.

Dahiya, S., Katakojwala, R., Ramakrishna, S., Mohan, S.V., 2020. Biobased products and life cycle assessment in the context of circular economy and sustainability. Mater. Circ. Econ. 2 https://doi.org/10.1007/s42824-020-00007-x.

European Commission, 2019. Environmental impact assessments of innovative bio-based product. Task 1 of “study on support to R&I policy in the area of bio-based products and services “ - study. https://doi.org/10.2777/251887.

Guest, G., Bright, R.M., Cherubini, F., Strømman, A.H., 2013. Consistent quantification of climate impacts due to biogenic carbon storage across a range of bio-product systems. Environ. Impact Assess. Rev. 43, 21–30. https://doi.org/10.1016/j.eiar.2013.05.002.

Hermansson, F., Janssen, M., Svanström, M., 2020. Allocation in life cycle assessment of lignin. Int. J. Life Cycle Assess. https://doi.org/10.1007/s11367-020-01770-4.

Hildebrandt, J., Budzinski, M., Nitzsche, R., Weber, A., Krombholz, A., Thrän, D., Bezama, A., 2019. Assessing the technical and environmental performance of wood-based fiber laminates with lignin based phenolic resin systems. Resour. Conserv. Recycl. 141, 455–464. https://doi.org/10.1016/j.resconrec.2018.10.029.

Indexmundi, 2020. Wood pulp monthly price - euro per metric ton [WWW Document]. URL. https://www.indexmundi.com/commodities/?commodity=wood-pulp&months=120&currency=eur, 3.10.21.

ISO, 2006a. ISO 14044, Environmental Management — Life Cycle Assessment — Requirements and Guidelines. International Standard Organization. Iso 14044.

ISO, 2006b. ISO 14040: Environmental Management — Life Cycle Assessment — Principles and Framework.

Karka, P., Papadokonstantakis, S., Kokossis, A., 2019. Environmental impact assessment of biomass process chains at early design stages using decision trees. Int. J. Life Cycle Assess. https://doi.org/10.1007/s11367-019-01591-0.

Keijzer, E., Kootstra, L., Schwarz, A., Bizarro, D., Kuling, L., Van Horssen, A., Albers, R., 2020. Product Category Rules voorbitumineuze materialen in verkeersdragers en waterwerken in Nederland (PCR Asfalt) 1–71.

Khan, T.A., Lee, J.H., Kim, H.J., 2019. Lignin-based adhesives and coatings. Lignocellul. Futur. Bioecon. 153–206. https://doi.org/10.1016/B978-0-12-816354-2.00009-8.

Kouris, P.D., Huang, X., Ouyang, X., van Osch, D.J.G.P., Cremers, G.J.W., Boot, M.D., Hensen, E.J.M., 2021. The impact of biomass and acid loading on methanolysis during two-step lignin-first processing of birchwood. Catalysts 11. https://doi.org/10.3390/catal11060750.

Lask, J., Rukavina, S., Zorić, I., Kam, J., Kiesel, A., Lewandowski, I., Wagner, M., 2021. Lignocellulosic ethanol production combined with CCS—a study of GHG reductions and potential environmental trade-offs. GCB Bioenergy 13, 336–347. https://doi.org/10.1111/gcbb.12781.

Liao, Y., Koelewijn, S.F., van den Bossche, G., van Aelst, J., van den Bosch, S., Renders, T., Navare, K., Nicolaï, T., van Aelst, K., Maesen, M., Matsushima, H., Thevelein, J.M., van Acker, K., Lagrain, B., Verboekend, D., Sels, B.F., 2020. A sustainable wood biorefinery for low-carbon footprint chemicals production. Science (80- 367, 1385–1390. https://doi.org/10.1126/science.aau1567.

Martin, M., Røyne, F., Ekvall, T., Moberg, Å., 2018. Life cycle sustainability evaluations of bio-based value chains: reviewing the indicators from a Swedish perspective. Sustain. 10 https://doi.org/10.3390/su10020547.

Martinez-Arguelles, G., Giustozzi, F., Crispino, M., Flintsch, G.W., 2015. Laboratory investigation on mechanical performance of cold foamed bitumen mixes: bitumen source, foaming additive, fiber-reinforcement and cement effect. Construct. Build. Mater. 93, 241–248. https://doi.org/10.1016/j.conbuildmat.2015.05.116.

McDevitt, J.E., Grigsby, W.J., 2014. Life cycle assessment of bio- and petro-chemical adhesives used in fiberboard production. J. Polym. Environ. 22, 537–544. https://doi.org/10.1007/s10924-014-0677-4.

Michels, J., 2014. Lignocellulose Biorefinery—phase 2”—Final Scientific and Technical Report of All Project Partners.

Milieudatabase, 2020. Nationale MilieuDATABASE. MAAKT CIRCULAIR BOUWEN MEETBAR [WWW Document]. URL. https://milieudatabase.nl/milieudata/, 3.10.21.

Montazeri, M., Eckelman, M.J., 2016. Life cycle assessment of catechols from lignin depolymerization. ACS Sustain. Chem. Eng. 4, 708–718. https://doi.org/10.1021/acssuschemeng.5b00550.

Montazeri, M., Zaimes, G.G., Khanna, V., Eckelman, M.J., 2016. Meta-analysis of life cycle energy and greenhouse gas emissions for priority biobased chemicals. ACS Sustain. Chem. Eng. 4, 6443–6454. https://doi.org/10.1021/acssuschemeng.6b01217.

Moretti, C., Corona, B., Edwards, R., Junginger, M., Moro, A., Rocco, M., Shen, L., 2020. Reviewing ISO compliant multifunctionality practices in environmental life cycle modeling. Energies 13, 3579. https://doi.org/10.3390/en13143579.

Moretti, C., Corona, B., Hoefnagels, R., van Veen, M., Vural-Gürsel, I., Strating, T., Gosselink, R., Junginger, M., 2021a. Kraft lignin as a bio-based ingredient for Dutch asphalts: an attributional LCA. Sci. Total Environ. https://doi.org/10.1016/j.scitotenv.2021.150316.

Moretti, C., Corona, B., Hoefnagels, R., Vural-Gürsel, I., Gosselink, R., Junginger, M., 2021b. Review of life cycle assessments of lignin and derived products: lessons learned. Sci. Total Environ. 770, 144656. https://doi.org/10.1016/j.scitotenv.2020.144656.

Moretti, C., Hamelin, L., Jakobsen, L.G., Junginger, M.H., Steingrimsdottir, M.M., Høibye, L., Shen, L., 2021c. Cradle-to-grave life cycle assessment of single-use cups made from PLA, PP and PET. Resour. Conserv. Recycl. 169, 105508.

Moretti, L., Mandrone, V., D'Andrea, A., Caro, S., 2017. Comparative "from cradle to gate" life cycle assessments of Hot Mix Asphalt (HMA) materials. Sustain. 9 https://doi.org/10.3390/su9030400.

Obydenkova, S.V., Kouris, P.D., Hensen, E.J.M., Heeres, H.J., Boot, M.D., 2017. Environmental economics of lignin derived transport fuels. Bioresour. Technol. 243, 589–599. https://doi.org/10.1016/j.biortech.2017.06.157.

Obydenkova, S.V., Kouris, P.D., Smeulders, D.M.J., Boot, M.D., van der Meer, Y., 2021. Modeling life-cycle inventory for multi-product biorefinery: tracking environmental burdens and evaluation of uncertainty caused by allocation procedure. Biofuels Bioprod. Bioref. 15, 1281–1300. https://doi.org/10.1002/bbb.2214.

Paone, E., Tabanelli, T., Mauriello, F., 2020. The rise of lignin biorefinery. Curr. Opin. Green Sustain. Chem. 24, 1–6. https://doi.org/10.1016/j.cogsc.2019.11.004.

Parisi, C., 2020. Distribution of the bio-based industry in the EU. Database and visualization. https://doi.org/10.2760/745867.

Pawelzik, P., Carus, M., Hotchkiss, J., Narayan, R., Selke, S., Wellisch, M., Weiss, M., Wicke, B., Patel, M.K., 2013. Critical aspects in the life cycle assessment (LCA) of bio-based materials - reviewing methodologies and deriving recommendations. Resour. Conserv. Recycl. https://doi.org/10.1016/j.resconrec.2013.02.006.

Pelletier, N., Ardente, F., Brandão, M., De Camillis, C., Pennington, D., 2015. Rationales for and limitations of preferred solutions for multi-functionality problems in LCA: is increased consistency possible? Int. J. Life Cycle Assess. 20, 74–86. https://doi.org/10.1007/s11367-014-0812-4.

Pola, L., Collado, S., Oulego, P., Díaz, M., 2019. Hydrothermal processing of Kraft lignin for carboxylic acid production. J. Environ. Chem. Eng. 7 https://doi.org/10.1016/j.jece.2019.103472.

Poluzzi, A., Guandalini, G., Romano, M.C., 2020. Potential carbon efficiency" as a new index to track the performance of biofuels production processes. Biomass Bioenergy 142. https://doi.org/10.1016/j.biombioe.2020.105618.

Ragauskas, A.J., Beckham, G.T., Biddy, M.J., Chandra, R., Chen, F., Davis, M.F., Davison, B.H., Dixon, R.A., Gilna, P., Keller, M., Langan, P., Naskar, A.K., Saddler, J. N., Tschaplinski, T.J., Tuskan, G.A., Wyman, C.E., 2014. Lignin valorization: improving lignin processing in the biorefinery. Science (80 344. https://doi.org/10.1126/science.1246843.

Reap, J., Roman, F., Duncan, S., Bras, B., 2008. A survey of unresolved problems in life cycle assessment. Part 2: impact assessment and interpretation. Int. J. Life Cycle Assess. 13, 374–388. https://doi.org/10.1007/s11367-008-0009-9.

Sala, S., Amadei, A., Beylot, A., Ardente, F., 2021. The evolution of life cycle assessment in European policies over three decades. Int. J. Life Cycle Assess. https://doi.org/10.1007/s11367-021-01893-2.

Saouter, E., Biganzoli, F., Ceriani, L., Versteeg, D., Crenna, E., Zampori, L., Sala, S., Pant, R., 2018. Environmental Footprint : Update of Life Cycle Impact Assessment Methods: Ecotoxicity Freshwater, Human Toxicity Cancer, and Non-cancer. EUR 29495 EN.

Schulze, C., Thiede, S., Herrmann, C., 2019. Life cycle assessment of industrial cooling towers. Sustain. Prod. Life Cycle Eng. Manag. 135–146. https://doi.org/10.1007/978-3-319-92237-9_15.

Schwarz, A., Overmars, L., Godoi Bizarro, D., Keijzer, E., Kuling, L., van Horssen, A., 2020. LCA Achtergrondrapport Voor Brancherepresentatieve Nederlandse Asfaltmengsels 2020. TNO Rapport 10987.

Souto, F., Calado, V., Pereira, N., 2018. Lignin-based carbon fiber: a current overview. Mater. Res. Express 5. https://doi.org/10.1088/2053-1591/aaba00.

Stegmann, P., Londo, M., Junginger, M., 2020. The circular bioeconomy: its elements and role in European bioeconomy clusters. Resour. Conserv. Recycl. 6 https://doi.org/10.1016/j.rcrx.2019.100029.

Tokede, O.O., Whittaker, A., Mankaa, R., Traverso, M., 2020. Life cycle assessment of asphalt variants in infrastructures: the case of lignin in Australian road pavements. Structures 25, 190–199. https://doi.org/10.1016/j.istruc.2020.02.026.

Tonini, D., Schrijvers, D., Nessi, S., Garcia-Gutierrez, P., Giuntoli, J., 2021. Carbon footprint of plastic from biomass and recycled feedstock: methodological insights. Int. J. Life Cycle Assess. 26, 221–237. https://doi.org/10.1007/s11367-020-01853-2.

Urgenda, 2020. 54 MAATREGELEN VOOR 17 MTON CO2-REDUCTIE [WWW Document]. URL. https://www.urgenda.nl/themas/klimaat-en-energie/40-puntenplan/, 7.2.21.

van Vliet, D., Slaghek, T., Giezen, C., Haaksman, I., 2017. Lignin as a green alternative for bitumen. https://doi.org/10.14311/ee.2016.159.

Weidema, B.P., Bauer, C., Hischier, R., Mutel, C.L., Nemecek, T., Reinhard, J., Vadenbo, C., Wernet, G., 2013. Overview and Methodology: Data Quality Guideline for the Ecoinvent Database Version 3.