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Full length article

Prospective LCA of valorizing end-of-life tires in asphalt mixtures with emerging pretreatment technologies of crumb rubber

Jin Li[a,b], João Santos[b,*], Andrea Vargas-Farias[b], Daniel Castro-Fresno[c], Feipeng Xiao[a,*]

[a] Key Laboratory of Road and Traffic Engineering of Ministry of Education, Tongji University, Shanghai, China
[b] Department of Civil Engineering & Management (CEM), Faculty of Engineering Technology, University of Twente, Enschede, The Netherlands
[c] Technical School of Civil Channels and Ports Engineering, University of Cantabria, Santander, Spain

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ABSTRACT

Despite numerous research studies devoted to quantify the environmental impacts associated with the valorization of crumb rubber (CR) derived from end-of-life tires (ELTs) in asphalt pavement construction, the number of life cycle assessment (LCA) studies scrutinizing the implementation of emerging CR pretreatment technologies remains scarce. Therefore, this study aims to conduct a prospective LCA study on the ELTs valorization in asphalt mixtures, with a focus on the application of two emerging supercritical pretreatment technologies of CR, namely the supercritical swelling pretreatment and the supercritical de-crosslinking pretreatment. The findings show that the supercritical swelling pretreated crumb rubber modified asphalt (CRMA) materials with a 40 % dosage obtained the lowest environmental impact score in the majority of the impact categories. Further, the supercritical pretreatment process of CR played a critical role in driving the environmental impacts of pretreated CRMA materials. Desipite this, the environmental impacts of supercritical pretreated CRMA materials exhibited relatively higher uncertainty. Finally, the analysis of scaled-up scenarios highlights the effectiveness of reusing carbon dioxide as reaction medium and reducing the use of dry ice and chemical reagent in minimizing the overall environmental impacts.

1. Introduction

In the recent decades, the surge in the global adoption of on-road and off-road vehicles as well as motorcycles has led to a substantial demand for tire production. When these tires reach their end-of-life, they are classified as end-of-life tires (ELTs), and invariably pose several environmental concerns and disposal challenges (Dong et al., 2021). Therefore, substantial endeavors have been dedicated to addressing the generation of ELTs (Martínez et al., 2013; Sienkiewicz et al., 2012).

One primary approach for the disposal of ELTs involves the recycling of their materials by incorporating them as elastomer additives, known as crumb rubber (CR) (Dondi et al., 2014). In the field of pavement engineering, a practical application of CR involves its utilization as an elastomer modifier in asphalt material, leading to the production of crumb rubber modified asphalt (CRMA) materials (Bressi et al., 2019). There are two main production methods of CRMA materials, namely the wet process and the dry process. In the wet process, fine CR derived from ELTs is mixed with neat asphalt at an elevated temperature for a short time period before mixing with hot aggregates. In the dry process, CR particles are blended with hot aggregates prior to mixing with neat asphalt. That mixing process occurs typically at elevated temperatures to ensure proper dispersion and adhesion of CR within the asphalt matrix. Overall, CRMA is widely acknowledged as a potentially sustainable pavement material with several technical benefits, namely the extension of pavement longevity and the mitigation of reflective cracking (Ibrahim et al., 2013). However, the use of CRMA is not devoid of concerns, many of which related to issues such as poor storage stability, high viscosity, and increased hazardous emissions during pavement construction (Li et al., 2022, 2021a).

Further, the key problem among these concerns is the incompatibility between CR and asphalt matrix (Li et al., 2021b; Liang et al., 2022; Ren et al., 2021). Consequently, appropriate pretreatments of CR that modify its chemical and/or physical properties are necessary to enhance the compatibility with asphalt. Currently, the primary CR pretreatment methods aimed at addressing the aforementioned issues can be classified as surface activation and internal de-crosslinking (Colom et al., 2016; Lei, 2021). The surface activation aims to improve or restore the surface activity of CR by means of pre-reaction,

* Corresponding authors.
E-mail addresses: j.m.oliveiradossantos@utwente.nl (J. Santos), fpxiao@tongji.edu.cn (F. Xiao).

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surface oxidation, grafting, polymer coating, plasma treatment and solution soaking. On the other hand, the internal de-crosslinking selectively breaks the crosslinking bonds inside vulcanized CR without affecting the main rubber chains using physical radiation, mechanical extrusion, biological and chemical means (Farina et al., 2024; Kazemi et al., 2021; Kazemi and Fini, 2022).

The state-of-the-art pretreatment methods in each category and their mechanisms have been summarized in the previous studies (Li et al., 2022, 2021a). However, as mentioned in these studies, the existing surface activation methods generally cannot effectively modify the critical bulk properties of CR, and the internal de-crosslinking methods are typically not able to achieve the selective scission of crosslinking bonds. Given that, two emerging CR pretreatment technologies, namely the supercritical swelling pretreatment and the supercritical de-crosslinking pretreatment, have been proposed to address these limitations. Among them, the supercritical swelling can be seen as a kind of surface activation method, and the supercritical de-crosslinking belongs to the category of internal de-crosslinking.

2. Literature review on the valorization of ELTs in asphalt pavements

In addition to the body of literature devoted to the study of the pavement performance of CRMA, the scientific community has also

Table 1
Summary of relevant LCA studies on valorizing ELTs in asphalt pavements.

Reference	Location	CRMA production process	CR pretreatment method	Percentage	System boundary	Allocation rule in CR co-production	LCIA method	Main results
(Bressi et al., 2021)	Italy	Dry	Devulcanization	1.5 %–2.0 % of total mixture mass	Cradle-to-gate	Physical allocation	ReCiPe (CC) & CED	Disadvantageous (vs neat asphalt)
(Polo-Mendoza et al., 2022)	Colombia	Dry	Info not provided	Up to 6 % of total mixture mass	Cradle-to-gate	Physical allocation	BEES+	Not applicable
(Bartolozzi et al., 2012)	Italy	Wet	Info not provided	1.6 % of total mixture mass	Cradle-to-grave*	Not mentioned	CML	Advantageous (vs neat asphalt)
(Mattinzioli et al., 2021)	Spain	Wet & Dry	Info not provided	20 % of total binder mass & 1.5 % of total mixture mass	Cradle-to-gate & Cradle-to-grave	Not mentioned	IPCC	Advantageous (vs neat asphalt)
(Rangelov et al., 2022)	Puerto Rico	Wet	Info not provided	19.5 % of total binder mass	Cradle-to-laid	System expansion	TRACI	Disadvantageous (vs neat asphalt)
(Feraldi et al., 2013)	USA	Wet	Info not provided	4.5 % of total binder mass	Cradle-to-gate	System expansion	IPCC	Not applicable
(Farina et al., 2020)	USA	Wet & Dry	Devulcanization & Polymer coating	0.4 %–1.0 % of total mixture mass	Cradle-to-gate	Not mentioned	IPCC	Advantageous (vs SBS asphalt)
(Farina et al., 2023)	USA	Dry	Polymer coating	0.5 % and 1.0 % of total mixture mass	Cradle-to-gate	System expansion	TRACI	Advantageous (vs SBS asphalt)
(Cao et al., 2019)	Hong Kong	Wet	Info not provided	18 % of total binder mass	Cradle-to-grave	Not mentioned	CED	Not applicable
(Ortíz-Rodríguez et al., 2017)	Colombia	Wet	Info not provided	20 % of total binder mass	Cradle-to-gate	Not mentioned	CML	Not applicable
(Yu et al., 2014)	China	Wet	Info not provided	20 % of total binder mass	Cradle-to-gate	Not mentioned	IPCC & CED	Advantageous (vs SBS asphalt)
(Riekstins, 2022)	Latvia	Wet	Info not provided	15 %–25 % of total binder mass	Cradle-to-grave	Not mentioned	IPCC	Advantageous (vs neat asphalt)
(Praticò et al., 2020)	Italy	Wet	Info not provided	10 % of total binder mass	Cradle-to-grave*	Not mentioned	ILCD midpoint	Advantageous (vs neat asphalt)
(Puccini et al., 2019)	Italy	Wet & Dry	Info not provided	20 % of total binder mass & 2.9 % of total mixture mass	Cradle-to-grave*	Not mentioned	Ecological scarcity	Advantageous (vs SBS asphalt)
(Farina et al., 2014)	Italy	Wet & Dry	Info not provided	18 % of total binder mass & 1 % of total mixture mass	Cradle-to-grave*	System expansion	ReCiPe	Advantageous (vs neat asphalt)
(Gamboa et al., 2021)	USA & Colombia	Wet	Info not provided	10.7 % of total binder mass	Cradle-to-grave	Not mentioned	ReCiPe	Advantageous (vs neat asphalt)
(Piao et al., 2022)	Switzerland	Dry	Info not provided	0.7 %–1.0 % of total mixture mass	Cradle-to-grave*	Not mentioned	IPCC & CED	Advantageous (vs neat asphalt)
(Zhu et al., 2014)	China	Wet	Info not provided	18 %–20 % of total binder mass	Cradle-to-gate	Not mentioned	IPCC	Advantageous (vs SBS asphalt)
(Siverio Lima et al., 2022)	Austria	Dry	Info not provided	0.3 %–0.34 % of total mixture mass	Cradle-to-grave*	Not mentioned	IPCC & CED	Depending on pavement service life
(Tushar et al., 2022)	Australia	Wet	Info not provided	5 %–20 % of total binder mass	Cradle-to-gate	System expansion	ReCiPe	Disadvantageous (vs neat asphalt)
(Wang et al., 2020)	China	Wet	Info not provided	18 %–25 % of total binder mass	Cradle-to-gate	Not mentioned	Not mentioned	Advantageous (vs SBS asphalt)
(Fiksel et al., 2011)	USA	Wet	Info not provided	Not mentioned	Cradle-to-gate	Physical allocation	TRACI	Not applicable
(Chiu et al., 2008)	Taiwan	Wet	Info not provided	20 % of total binder mass	Cradle-to-grave*	Not mentioned	Eco-indicator 99	Advantageous (vs neat asphalt)

Key: BEES= Building for Environmental and Economic Sustainability; CED= Cumulative Energy Demand; CC= Climate Change; CML= Centre for Environmental Studies (Centrum voor Milieuwetenschappen); CR= Crumb rubber; CRMA= Crumb rubber modified asphalt; ILCD= International Life Cycle Data System; IPCC= International Panel for Climate Change; LCIA= Life cycle impact assessment; ReCiPe= RIVM and Radboud University, CML, and PRé Consultants; TRACI= Tool for Reduction and Assessment of Chemical and Other Environmental Impacts.
Notes: *Cradle-to-grave refers cradle-to-grave excluding the use phase.

scrutinized the potential environmental benefits resulting from the valorization of ELTs in asphalt pavements with the life cycle assessment (LCA) methodology. This is evident from a literature review conducted on this research topic using the Scopus database with the following search query: (TITLE-ABS-KEY("life cycle assessment" OR LCA) AND TITLE-ABS-KEY(valorization OR recycling OR reuse) AND TITLE-ABS-KEY("end-of-life tires" OR ELTs OR "waste tires" OR "scrap tires") AND TITLE-ABS-KEY("asphalt pavements" OR "asphalt concrete" OR "road construction")). The retrieved literature was posteriorly subjected to a secondary screening based on the authors' knowledge, and any relevant missing articles were added to the resulting list. The complete literature search resulted in the list of articles summarized in Table 1.

As can be seen in Table 1, pavement LCA studies on this topic have been conducted worldwide, covering both dry and wet mixing processes. Different system boundary and life cycle impact assessment (LCIA) methods were respectively selected and employed, while the allocation issue in the CR co-production process was frequently overlooked. Further, the use of CR in asphalt mixtures typically contributes to a decrease in most impact categories, compared with the neat asphalt mixture and SBS modified asphalt mixture. These environmental benefits are guaranteed only if CRMA mixtures are properly designed and laid, with the corresponding possibility of extending the pavement durability and reducing the surface course thickness. Finally, despite the extensive investigations into the environmental impacts of conventional pavements with CRMA, Table 1 makes it clear that there is a dearth of studies exploring the application of emerging CR pretreatment technologies in pavements.

Undoubtedly, the LCA methodology can play a useful role in filling in this knowledge gap. However, the conventional LCA methodology (i.e., post-ante), owing to its retrospective nature, is not suitable for modeling the environmental impacts of emerging technologies and future systems, which may involve the upscaling of lab or pilot-scale data to large-scale production in the foreground system (Adrianto and Pfister, 2022). On the contrary, prospective LCA consists of conducting an LCA study on a new (emerging) technology before its commercial implementation, guiding research and development decisions to ensure the new technology's environmental competitiveness compared to existing technologies (Arvidsson et al., 2018). This approach entails a shift from ex-post to ex-ante environmental assessment, and comes with an additional challenge associated with the epistemological uncertainty (due to the lack of case-specific data) which is a distinctive feature of future systems and/or models (Georgiades et al., 2023). Addressing these challenges may involve the use of proxy data and analysis of multiple likely scenarios for future industrial production (Tsoy et al., 2020).

Motivated by the limited number of pavement LCA studies assessing the impacts of emerging CR pretreatment, their knowledge gaps, and the merits of the prospective LCA methodology in assessing the potential environmental impacts of emerging technologies at lab-scale, the present study aims to conduct a prospective LCA on the valorization of ELTs in asphalt mixtures. A specific focus was placed on two emerging supercritical pretreatment technologies for CR, namely the supercritical swelling pretreatment and the supercritical de-crosslinking pretreatment.

3. Methodology

3.1. Goal and scope definition

3.1.1. Goal and scenarios

As shown in Table 1, the CRMA is generally recognized as a more environmentally sustainable paving material than neat asphalt and SBS modified asphalt. However, there is a need to ascertain whether the CRMA with pre-treated CR still holds the environmental advantage when compared with the conventional counterparts. Thus, by adopting a process-based, attributional and prospective LCA methodology the primary goals of this study are as follows:

1) Quantifying the environmental burden of CRMA materials with two emerging supercritical pretreatment technologies for CR;
2) Comparing the environmental performance of pretreated CRMA materials with conventional asphalt materials;
3) Clarifying the uncertainty of prospective pavement LCA results involving emerging CR pretreatment technologies.

It is worth mentioning that it is not a goal of this study to perform a system level analysis of ELTs waste management. In other words, this study does not aim to ascertain the extent to which producing and using CR from ELTs in asphalt pavements is environmentally preferable than the conventional alternatives, which are usually landfilling (if legally allowed) and energy recovery in cement kilns (i.e., tire-derived fuel), or even the use of CR in other applications (e.g., filling for artificial turf football fields, vibration absorption systems for railway structures, etc.). To achieve the goals outlined above, several alternative scenarios involving asphalt mixtures were considered. They are the following:

1) Neat asphalt mixture, referred to as NA;
2) SBS modified asphalt mixture, referred to as SBSMA;
3) Crumb rubber modified asphalt mixture with 20 % dosage of unpretreated CR in binder, referred to as CRMA-20 %UPT, which represents the conventional valorization scenario of ELTs in asphalt mixtures;
4) Crumb rubber modified asphalt mixture with 20 % dosage of pretreated CR in binder via supercritical swelling, referred to as CRMA-20 %SCS;
5) Crumb rubber modified asphalt mixture with 40 % dosage of pretreated CR in binder via supercritical swelling, referred to as CRMA-40 %SCS;
6) Crumb rubber modified asphalt mixture with 20 % dosage of pretreated CR in binder via supercritical de-crosslinking, referred to as CRMA-20 %SCD;
7) Crumb rubber modified asphalt mixture with 40 % dosage of pretreated CR in binder via supercritical de-crosslinking, referred to as CRMA-40 %SCD. Note that scenarios 4–7 all represent the emerging valorization scenarios of ELTs in asphalt mixtures but with different settings.

It should also be noted that the two dosages of CR considered (i.e., 20 % and 40 %) were defined according to the existing research works on conventional and high-dosage CRMA (see Table 1). Further, the percentages mentioned above are all in relation to the weight of asphalt binder instead of the total asphalt mixtures.

In addition to the scenarios outlined above related to the mixtures composition, several scale-up scenarios will be developed based on the process contribution and hotspots analysis in view of the improvement of the environmental performance of the technologies. To scale-up and generate future scenarios, a set of predictions will be made on the condition of several specific improvements of the emerging CR pretreatment process.

3.1.2. System boundary and functional unit

Due to the unavailability of reliable data for the innovative pavement materials considered in the present study, the construction and use phase, which may be of premium relevance in pavement LCA studies, were not included. Instead, a cradle-to-gate LCA was performed with the system boundary depicted in Fig. 1.

The functional unit was defined as the production of l ton of alternative asphalt mixtures for road pavements located in China, including the NA, SBSMA and CRMA. The specific ingredients and respective quantities considered in the functional unit of the different alternative scenarios are presented in Table 2. Note that the binder content in asphalt mixture and the SBS modifier in asphalt binder were respectively selected as 5 % and 4.5 %, in accordance with the common practice in road pavement engineering projects.

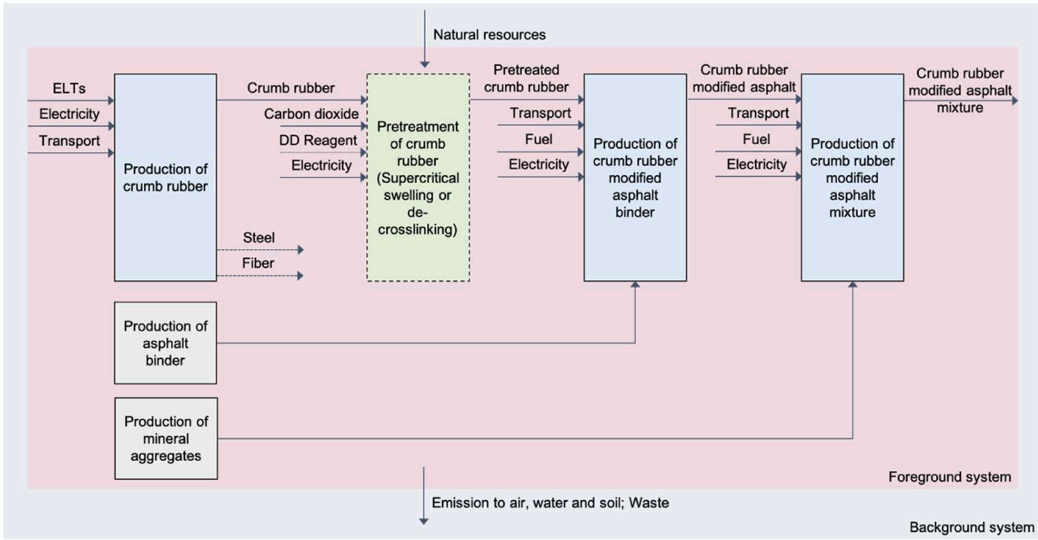

Fig. 1. System boundary of CRMA (with/without supercritical pretreatments of CR).

Table 2
Ingredients and quantities considered in the functional unit (i.e., 1 ton of asphalt mixture) of different alternative scenarios.

Asphalt mixture ID	Asphalt mixture name	Asphalt mixture component	Proportion (%)	Mass (kg)
M1	NA	Neat asphalt	5 %	50
		Mineral aggregate	95 %	950
M2	SBSMA	SBS modifier	5 % × 4.5% % 0.225 %	2.25
		Neat asphalt	5 % × 95.5% % 4.775 %	47.75
		Mineral aggregate	95 %	950
M3	CRMA-20 %UPT	CR modifier without pretreatment	5 % × 20% % 1 %	10
		Neat asphalt	5 % × 80% % 4 %	40
		Mineral aggregate	95 %	950
M4	CRMA-20 %SCS	Pretreated CR modifier via supercritical swelling	5 % × 20% % 1 %	10
		Neat asphalt	5 % × 80 % = 4 %	40
		Mineral aggregate	95 %	950
M5	CRMA-40 %SCS	Pretreated CR modifier via supercritical swelling	5 % × 40 % = 2 %	20
		Neat asphalt	5 % × 60 % = 3 %	30
		Mineral aggregate	95 %	950
M6	CRMA-20 %SCD	Pretreated CR modifier via supercritical de-crosslinking	5 % × 20 % = 1 %	10
		Neat asphalt	5 % × 80 % = 4 %	40
		Mineral aggregate	95 %	950
M7	CRMA-40 %SCD	Pretreated CR modifier via supercritical de-crosslinking	5 % × 40 % = 2 %	20
		Neat asphalt	5 % × 60 % = 3 %	30
		Mineral aggregate	95 %	950

3.2. *Life cycle inventory (LCI) analysis*

LCI analysis involves the systematic gathering of primary and secondary data to quantify the physical inputs and outputs associated with processes occurring within the defined system boundary of the entire product system. The subsequent subsections provide comprehensive information regarding the operational data and underlying assumptions utilized for modeling the ELTs recycling. The primary sources and corresponding acquisition approaches of LCI data used in this study include public databases, enterprise data, field surveys, and literature research.

3.2.1. *CR production process*

The input and output flows associated with the CR production process are presented in Table S1 in the Supplementary Information (SI). Further, the “cut-off ” or zero-burden waste approach was applied to the collection of ELTs and the production of CR. Further, in addition to the vulcanized rubber, ELTs typically contain steel wires and tire fibers. Therefore, in the production process of CR, not only the main/target product (i.e., CR) is obtained but also two major additional products (i. e., steel and fibers) are involved. That raises the issue of allocating the environmental burdens among multiple products.

At present, several approaches have been proposed to deal with this issue based on different rationales, including portioning, often simply referred to as “allocation” and where partitioning criterion such as the relative economic value or the relative mass of the co-products are used, and “system expansion”. Considering the significant mismatch between the physical (i.e., mass) and economic attributes of the multiple products in the CR production process considered in this study (i.e., the highest output mass of CR but the lowest economic value), the “system expansion” method was adopted to address the allocation problem in this process. This method treats the co-products in the production process as “avoided products” and directly avoids the allocation problem of multiple products. Thus, the environmental burdens that should have been allocated to the remaining processes can be subtracted from the result of this production process (Cherubini et al., 2011).

3.2.2. *CR pretreatment process*

Two novel (emerging) CR pretreatment methods were considered in this study, namely the supercritical swelling and supercritical de-crosslinking, to enhance the compatibility between CR and asphalt matrix in CRMA. Both CR pretreatments were performed at high temperature and high pressure, in the environment of supercritical carbon dioxide ($ScCO_2$) fluid. $ScCO_2$ is a state of carbon dioxide that exists at temperatures and pressures above its critical point (Brunner, 2010). In the supercritical state, carbon dioxide displays unique properties that make it useful in various industrial applications. It combines the characteristics of both gases and liquids, exhibiting high density like a liquid while maintaining the diffusivity and low viscosity of a gas. These properties can be controlled by adjusting the temperature and pressure, allowing for a wide range of applications.

Therefore, this study adopted the $ScCO_2$ to achieve the supercritical pretreatment of CR. For the supercritical swelling pretreatment, the CR can be effectively swelled by $ScCO_2$ at high temperature (i.e., approximately 200 °C) and high pressure (i.e., 10 MPa) for 2 hours. This is because the $ScCO_2$ can easily penetrate into the internal crosslinking network of CR and make the crosslinking bonds become loose. This physical swelling process is also a dominant interaction between CR and asphalt matrix. With the looser crosslinking structure after pretreatment, there will be less resistance to the CR/asphalt interactions.

For the supercritical de-crosslinking pretreatment, the temperature and pressure of $ScCO_2$ are the same as those of the supercritical swelling. Besides, a chemical de-crosslinking reagent (i.e., diphenyl disulfide) is employed for the targeted attack of crosslinking bonds inside CR, while avoiding as much as possible the undesirable breakage of rubber main chains. This chemical process was also assisted by the $ScCO_2$, which cannot only dissolve the de-crosslinking reagent molecule but can also carry the reagent into the internal structure of CR for the subsequent de-crosslinking reaction. On the basis of supercritical swelling, the supercritical de-crosslinking can further destroy the crosslinking network of CR and enable its fully dissolution in the asphalt like other polymer modifiers (i.e., SBS).

Although technically both supercritical pretreatments of CR can be performed at the industrial scale, currently they are only carried out at the laboratory scale. This means that there is no specific and reliable foreground data about the CR pretreatment process for the pavement LCA study involving the use of pretreated CR. Consequently, this data gap issue must be addressed.

In general, for the prospective LCA studies with a focus on emerging technologies, the missing data is an inevitable problem when the investigated technology has not yet been applied to the industrial production. Considering this, a proxy can be employed as a means to approximate data by utilizing information from an already existing technology that exhibits a strong resemblance to the emerging technology under consideration (Tsoy et al., 2020). In this context, the proxy denotes an existing technology that bears the closest resemblance to the hypothetical upscaled technology being studied. In this way, this study explored similar CR pretreatment technologies with reliable LCI data to the emerging supercritical pretreatment technologies.

Li et al. conducted a LCA study on ground rubber production from scrap tires with foreground data collected in a typical Chinese factory, which included a step of tire rubber dynamic devulcanization (Li et al., 2014). In this rubber devulcanization process, the desulfurization tank was kept at a high temperature of around 230 °C under the relatively high pressure of 2.2 MPa. Besides, some chemical reagents were added to improve plasticity of CR. Similarly, the goal of rubber devulcanization process in Li's study was also to destroy the crosslinking network of vulcanized rubber. Li and Xu also conducted a similar study on environmental impact assessment of ELTs utilization in China, which also involved the dynamic devulcanization process (Li et al., 2010; Li and Xu, 2010). Overall, this existing technology can be seen as a similar technology to the emerging supercritical de-crosslinking pretreatment technology considered in the present study, although the former might lead to more severe breakage of rubber main chains. Therefore, the LCI data of rubber devulcanization process in the studies above was used as proxy data for supercritical pretreatment process of CR in the current study. Combined with the lab-scale data, the supercritical pretreatment process of CR can be scaled from the laboratory scale to the industrial scale. The entire input and output flows associated with two CR supercritical pretreatment processes are presented in Tables 3 and 4.

3.2.3. Modified asphalt binder and mixture production

The input and output flows associated with the asphalt binder production process as well as the corresponding data sources are presented in Table S2 and Table S3 (SI), including the SBS modified asphalt binder and the CR modified asphalt binder through wet process. Besides, it is worth mentioning that the LCI data of neat asphalt production was directly obtained from the ecoinvent database. The input and output flows associated with the asphalt mixture production processes, namely the NA, SBSMA, and CRMA, as well as the corresponding data sources are presented in Table S4, Table S5, and Table S6 in the SI.

Table 3
Input and output flows associated with the CR supercritical de-crosslinking pretreatment process.

Input/output	Amount	Unit	Data source
Product			(Li et al., 2014, 2010; Li and Xu, 2010)
Supercritical de-crosslinking pretreated CR	1	kg	
Input			
CR	1	kg	
Dry ice	1.067	kg	
Water	0.01	kg	
De-crosslinking agent	0.1	kg	
Transportation (CR)	0.05	tkm	
Electricity	0.030825	kWh	
Coal	0.10236	kg	
Outputs			
Carbon dioxide	1408	g	
Particulate matter	1.65	g	
Sulfur dioxide	0.45	g	
Nitrogen oxide	0.7	g	

Table 4
Input and output flows associated with the CR supercritical swelling pretreatment process.

Input/output	Amount	Unit	Data source
Product			(Li et al., 2014, 2010; Li and Xu, 2010)
Supercritical swelling pretreated CR	1	kg	
Input			
CR	1	kg	
Dry ice	1.067	kg	
Water	0.01	kg	
Transportation (CR)	0.05	tkm	
Electricity	0.030825	kWh	
Coal	0.10236	kg	
Outputs			
Carbon dioxide	1408	g	
Particulate matter	1.65	g	

3.2.4. Transportation process

The ELTs, asphalt binder, aggregates, asphalt modifiers, and the subsequently produced CR need to be transported between the different production facilities. In this study, it was assumed that all the materials were hauled from quarries and plants/production facilities by truck. Therefore, the environmental impacts resulting from the transportation of materials were due to the emissions released by the fuel combustion of vehicles. The specific transportation distance of the various materials is listed in Table S7 in the SI.

3.3. Life cycle impact assessment (LCIA) method

When performing a LCA study, it is crucial to select an appropriate LCIA method to determine the environmental impacts of the studied product, service, or process, as it plays a significant role in the results. Below are some of the factors that governed the choice of the LCIA methods adopted in this study:

1) Goal and scope: The geographical scope of this study is set as China. In the absence of LCIA methods specifically tailored to China, it is preferable to prioritize methods that are globally applicable;
2) Data availability and quality: Some LCIA methods require extensive data, while others require less data. It is important to ensure the data

quality and availability of the selected methods in order to obtain accurate and reliable results. Additionally, impact assessment results generally have lower uncertainty when the selected impact indicators are closer to the midpoint level (i.e., midpoint indicators), as the calculation process only requires modeling a few environmental pathways (Bulle et al., 2019). Considering the relatively high data uncertainty of the prospective LCA, it is preferable to prioritize the use of LCIA methods that include midpoint indicators.

Therefore, two widely applied LCIA methods, namely the Cumulative Energy Demand (CED) V1.11 and ReCiPe 2016 Midpoint (H) V1.07, were used to quantify the environmental burdens of pavement materials.

3.4. Scenario and uncertainty analysis

As mentioned in the sub-section 3.2.1, the system extension method was selected in this study to deal with the co-production characterizing the ELTs management from which CR is obtained. To ascertain the extent to which the environmental impact results are affected by the methodology choice of the allocation method, a scenario analysis was performed where physical allocation and economic allocation were considered as an alternative to system expansion. The concepts of physical allocation and economic allocation involve partitioning the total environmental burden of a process that produces multiple products proportionally to the individual production of each product, primarily based on the characteristics and attributes of these products. The calculated mass and economic allocation coefficients of each product are presented in Table 5, and the specific calculation procedures can be found in the SI. Moreover, in the system expansion scenario, the quality of the co-products obtained from the ELTs recycling was considered to be equal to that of the products whose primary production was displaced (avoided).

Further, as mentioned in sub-Section 3.2.2, the prospective LCA of emerging CR pretreatment technologies performed in this study was associated with various challenges, including the upscaling of the LCI data. For the LCI data of CR supercritical pretreatment process, this study used proxy data to overcome this challenge. This methodological decision inevitably led to additional uncertainty in the results. Thus, to account for the uncertainty inherent to data quality, the "Pedigree method" was adopted. It enables the calculation of data quality indicators (DQIs) based on a scoring system (scale 1–5), which assesses data sources on five independent characteristics, namely completeness, reliability, temporal correlation, geographic correlation, and technological correlation (Weidema and Wesnæs, 1996). These indicator scores were used to generate probability density functions (PDFs) using the log-normal distribution. Monte Carlo (MC) simulation was subsequently used to randomly sample these PDFs 10,000 times.

3.5. Modelling software and database

The SimaPro software version 9.4.0.1 with ecoinvent database version 3.8 was used to calculate the environmental impacts of the alternative asphalt mixtures. The ecoinvent datasets used can be found in Table S9 in the SI.

Table 5
Mass and economic allocation coefficient of main product and co-products.

Product type	Mass allocation coefficient	Economic allocation coefficient
CR (Main product)	68.97 %	31.79 %
Steel (Co-product)	20.00 %	28.97 %
Fiber (Co-product)	11.03 %	39.24 %

4. Results and discussion

4.1. Environmental impact analysis of alternative asphalt mixtures

The relative life cycle environmental impact results of the alternative asphalt mixtures are presented in Figs. 2 and 3, including the midpoint and endpoint impact indicators as well as the overall score. The relative scores were calculated in relation to the score obtained for the asphalt mixture with the highest score in the impact category under consideration.

In terms of midpoint impact indicators, the supercritical swelling pretreated CRMA with 40 % dosage (M5: CRMA-40 %SCS) generally obtained the lowest impact scores. In most cases (except for the global warming), its environmental performance was superior to that of the neat asphalt (M1: NA) and the unpretreated CRMA (M3: CRMA-20 % UPT) mixtures. However, it should be noted that its performance is poorer at lower CR dosages (i.e., M4: CRMA-20 %SCS). This can be attributed to the fact that the increase of the incorporation rate of CR was beneficial to the environmental impact of asphalt mixtures. On the other hand, the supercritical de-crosslinking pretreated CRMA with 40 % dosage (M7: CRMA-40 %SCD) and SBS modified asphalt (M2: SBSMA) mixtures obtained the highest scores among all midpoint indicators, indicating their relatively high environmental impact.

Another result worth mentioning is the fact that all CRMA mixtures showed negative scores in the impact category Mineral resources scarcity. Similar results were observed for the mixtures CRMA-40 %SCS and CRMA-40 %SCD in the impact category Human carcinogenic toxicity, and the mixture CRMA-40 %SCS in the impact category Land use. These results arise from the fact that the ELTs recycling and repurposing related to the production of CRMA reduce the abovementioned impact indicators scores, when the system expansion method is adopted as the allocation rule in the CR co-production process. It can be further inferred that incorporation of CR as a modifier at the high-dosage reduces the consumption of neat asphalt and avoids burdens from recovered materials (i.e., steel and fibers) displacing primary production as a result of the CR production process.

In terms of endpoint impact indicators, both unpretreated and supercritical swelling pretreated CRMA mixtures showed lower environmental burdens than NA and SBSMA. However, the supercritical de-crosslinking pretreated CRMA still obtained the highest environmental impact scores in the categories of Human health and Ecosystems. On the contrary, in the Resources category, regardless of whether they involved supercritical pretreatment, the relative impact scores of all CRMA materials were considerably lower than the two reference mixtures (i.e., NA and SBSMA). Again, this result shows that the utilization of ELTs by means of CR production and application in the production of asphalt mixtures reduces the consumption and dependence on natural resources in asphalt paving materials production processes.

When analyzing the overall environmental impact scores obtained with the ReCiPe method according to the predetermined baselines and weighting values (Fig. 3), it can be observed that supercritical de-crosslinking pretreated CRMA mixtures (including 20 % and 40 % dosages) showed the highest life cycle environmental burdens. The two reference mixtures (i.e., NA and SBSMA) denoted an intermediate performance, and the supercritical swelling pretreated CRMA mixtures (including the 20 % and 40 % dosages) exhibited similar environmental impacts to those of the unpretreated CRMA, both at the lowest level. One should bear in mind that the only difference between the LCI of supercritical swelling and supercritical de-crosslinking process is the use of chemical de-crosslinking reagent (i.e., diphenyl disulfide) in the case of the latter. Therefore, it can be deduced that the adoption of chemical reagent significantly increases the overall environmental burden of CRMA mixtures, and therefore it is natural that the reagent dosage plays a dominant role in this outcome. Finally, it should be noted that these overall scores are calculated based on various assumptions, selected baselines, and pre-allocated weights, and the modeling process involves

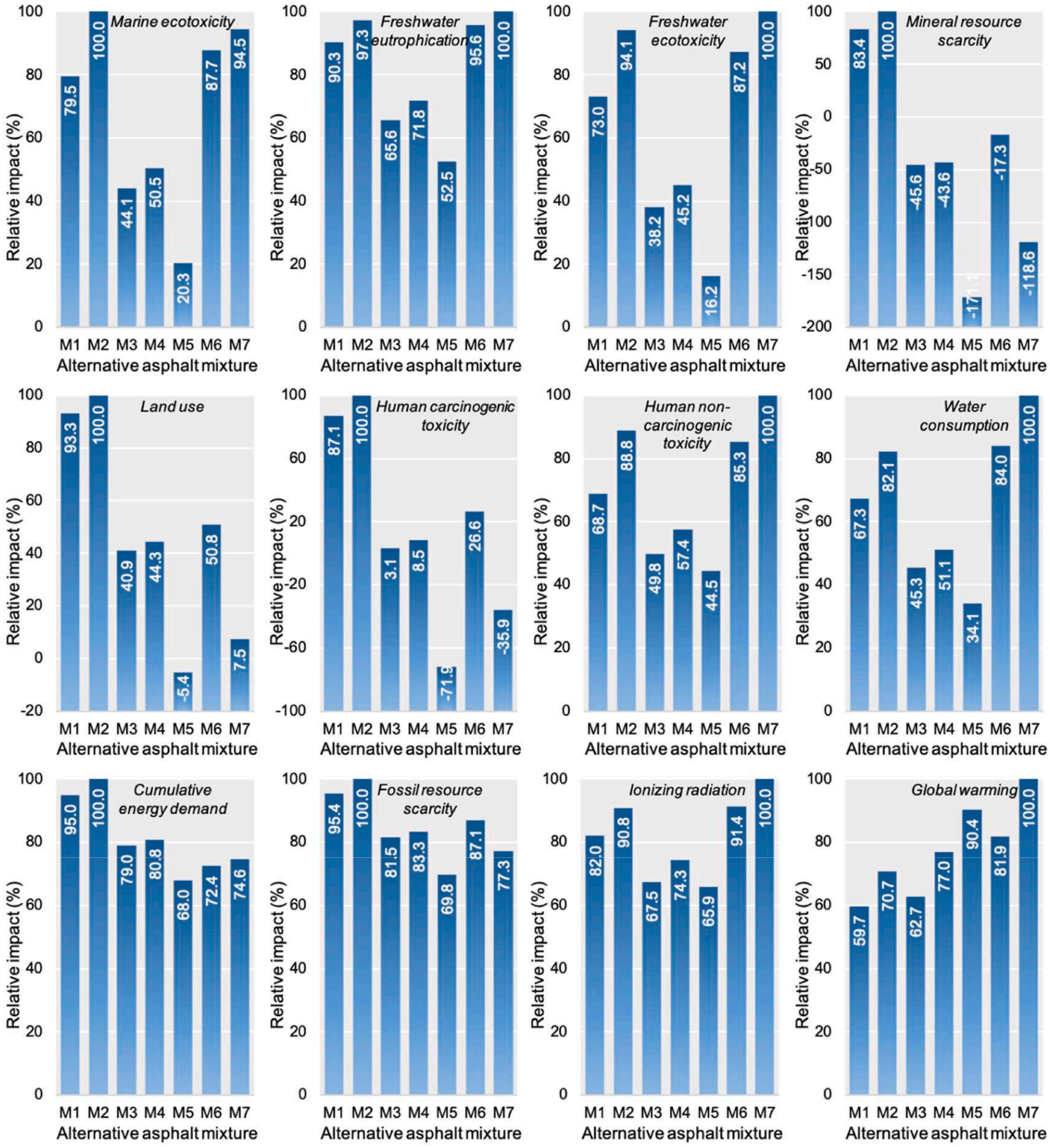

Fig. 2. Relative impact scores of midpoint indicators for different alternative asphalt mixtures. Key: M1 = NA; M2 = SBSMA; M3= CRMA-20 %UPT; M4= CRMA-20 %SCS; M5= CRMA-40 %SCS; M6= CRMA-20 %SCD; M7= CRMA-40 %SCD.

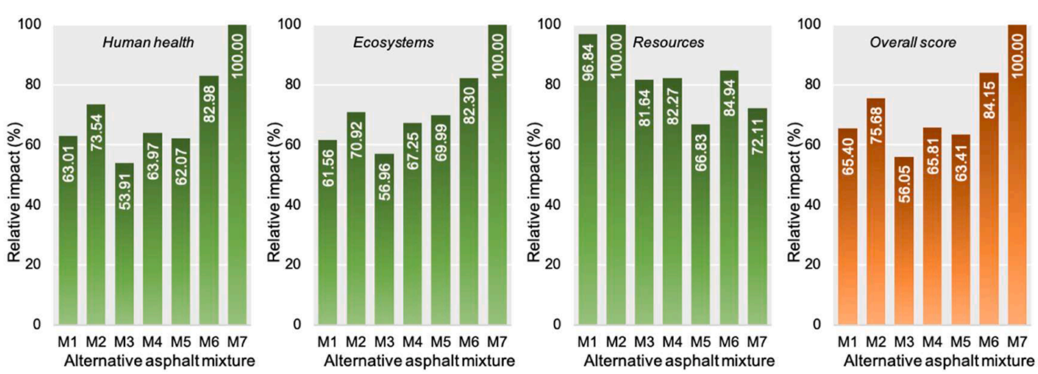

Fig. 3. Relative impact scores of endpoint indicators and overall score for different alternative asphalt mixtures. Key: M1= NA; M2= SBSMA; M3= CRMA-20 %UPT; M4= CRMA-20 %SCS; M5= CRMA-40 %SCS; M6= CRMA-20 %SCD; M7= CRMA-40 %SCD.

subjectivity, uncertainty, and geographical limitations. This is also the main reason why such overall indicators and endpoint indicators are not as widely used as midpoint indicators in various LCA studies (Bare et al., 2000; Dong and Ng, 2014).

As presented in Table 1, the literature on pavement LCA comprises several studies intended to determine the environmental impacts associated with the production of asphalt mixtures incorporating CR. To infer the plausibility of the results obtained in this study, Fig. 4 compares them with those reported by relevant literature. However, it should be noted that the aim of this comparison is not to demonstrate the environmental superiority of the alternative asphalt mixtures investigated in this study, as they are not directly comparable due to differences in methodological choices, scenario setting and data sources used in the LCA modeling. Thus, this comparison should be seen as a benchmarking exercise. For that purpose, the global warming indicator was taken as an example, since it is an indicator common to all studies. Overall, Fig. 4 indicates that the environmental burdens of the asphalt mixtures incorporating CR in our study are within the ranges of values reported in the literature.

4.2. Process contribution and hotspot analysis

To quantify the contribution and identify hotspots of different processes, the life cycle of asphalt mixture production was divided into the production processes of asphalt binders and aggregates as well as the plant mixing process of asphalt mixture. Subsequently, the cumulative energy demand (CED) and global warming (GW), as representative environmental impact indicators in pavement LCA studies, were selected to ascertain the contribution of the main processes to overall environmental impacts, as shown in Fig. 5.

From the analysis of Fig. 5a, it is evident that in terms of CED, the majority (approximately 80 %) of energy demand in various asphalt mixture production processes originates from the production of asphalt binder, while the contribution of aggregate production process is minimal. It is worth acknowledging that the mass ratio of aggregate in asphalt mixture is around 95 %, while the asphalt binder content is only 5 % of the total mass. This discrepancy in quantity indicates that the production process of petroleum asphalt for road paving derived from crude oil extraction and refining is a high energy-intensive process. This outcome is well aligned with the findings reported in the pavement LCA literature.

In terms of GW (Fig. 5b), the plant mixing process of asphalt mixture and the production process of asphalt binder are generally at similar levels, although exhibiting differentiated characteristics depending on the type of asphalt mixture. Specifically, for neat asphalt and unpretreated CRMA (CRMA-20 %UPT) mixtures, the CO_2-eq emissions from the plant mixing process of asphalt mixtures account for over 60 % of the overall emissions. However, in the production processes of SBS modified asphalt (SBSMA) and all supercritical pretreated CRMA mixtures, the production process of modified asphalt binder gradually replaces the plant mixing process as the dominant contributor. In extreme cases, the CO_2-eq emissions from the production process of 40 % supercritical decrosslinking pretreated CRMA (CRMA-40 %SCD) binder account for 60 % of the overall emissions. Based on this result, it can be stated that for the production of supercritical pretreated CRMA mixtures, the production process of CRMA binder (including the production and pretreatment of CR) is a significant contributor to the overall CO_2-eq emissions associated with the production of such asphalt mixtures.

To further scrutinize the provenience of the CO_2-eq emissions resulting from the supercritical pretreatment process of CR, the production process of asphalt binder (including neat asphalt, SBS modified asphalt, and CR modified asphalt) was subjected to a deeper analysis. Using the production of 1 ton of asphalt binder as the reference, the contribution of the unit processes, such as modifier production (i.e., SBS and CR) and modifier pretreatment (i.e., supercritical pretreatment of CR), to the CO_2-eq emissions were calculated and shown in Fig. 6.

As shown in Fig. 6, in all production processes of modified asphalt binders, the CO_2-eq emissions resulting from the neat asphalt production decrease with the increase of modifiers. This was due to the substitution effect of modifiers as asphalt substitute. In other words, the addition of modifier in modified asphalt avoided the production of neat asphalt in the same quantity. In terms of modifiers production, SBS modifier outperformed the remaining as the most carbon-intensive modifier production process. This result contrasts with those related to the production of CR modifier (unpretreated), which exhibited a negative effect on CO_2-eq emissions. Finally, in the production processes of the four types of supercritical pretreated CRMA binders, the CR pretreatment process considerably increased the overall CO_2-eq emissions, and even stood out as the main source of emissions. In other words, this can be considered as a CO_2 emissions hotspot in the supercritical pretreatment process of CR and even in the production process of supercritical pretreated CRMA materials. Among them, the supercritical decrosslinking pretreatment process, which involved the use of chemical reagents, emitted an even higher quantity of CO_2-eq than the purely physical supercritical swelling pretreatment process. At the lab-scale, the cooled carbon dioxide (originating from the added dry ice in the recipe) is directly emitted to the air after CR pretreatment and accounted into the process inventory. This result emphasizes the need of paying special attention to the post-collection and reuse of CO_2 that only acts as a reaction medium in the supercritical pretreatment process of CR.

4.3. Scenario analysis on allocation issue

The calculation of the environmental burdens of the CR production process and the entire CRMA mixtures production process for the different allocation scenarios was performed according to the conditions

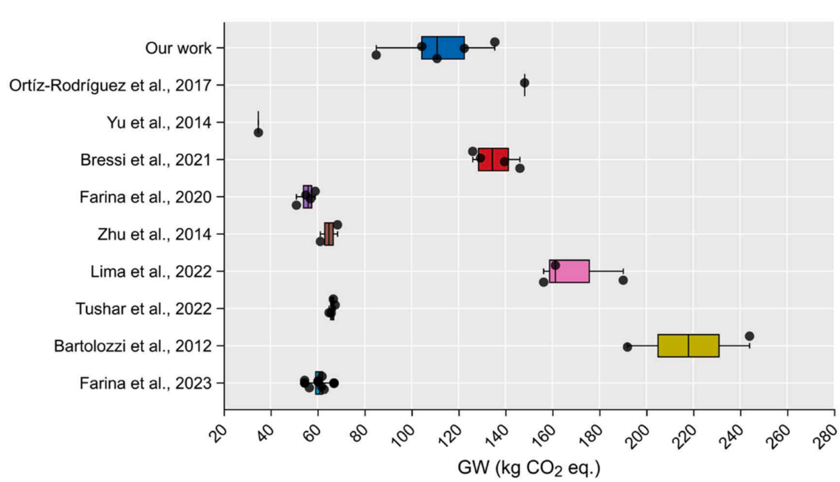

Fig. 4. Environmental impact (global warming) comparison of valorizing ELTs in asphalt mixtures between our study and other relevant studies (Bartolozzi et al., 2012; Bressi et al., 2021; Farina et al., 2023, 2014; Ortíz-Rodríguez et al., 2017; Siverio Lima et al., 2022; Tushar et al., 2022; Yu et al., 2014; Zhu et al., 2014).

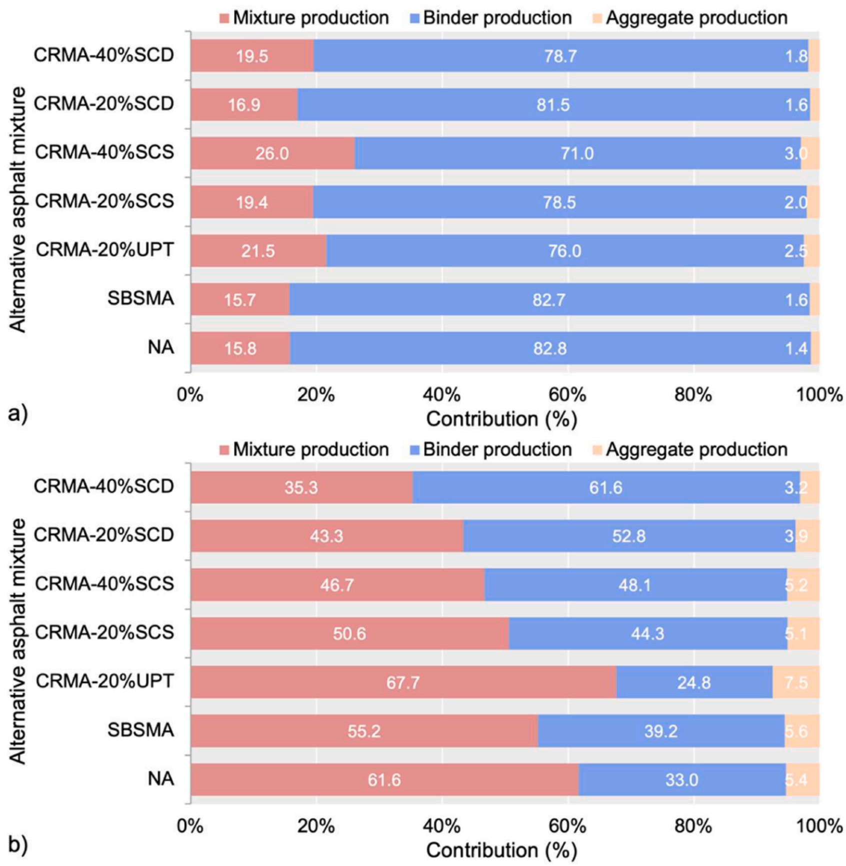

Fig. 5. Process contribution to the overall environmental impacts of producing 1 ton of asphalt mixture: (a) Cumulative energy demand; (b) Global warming.

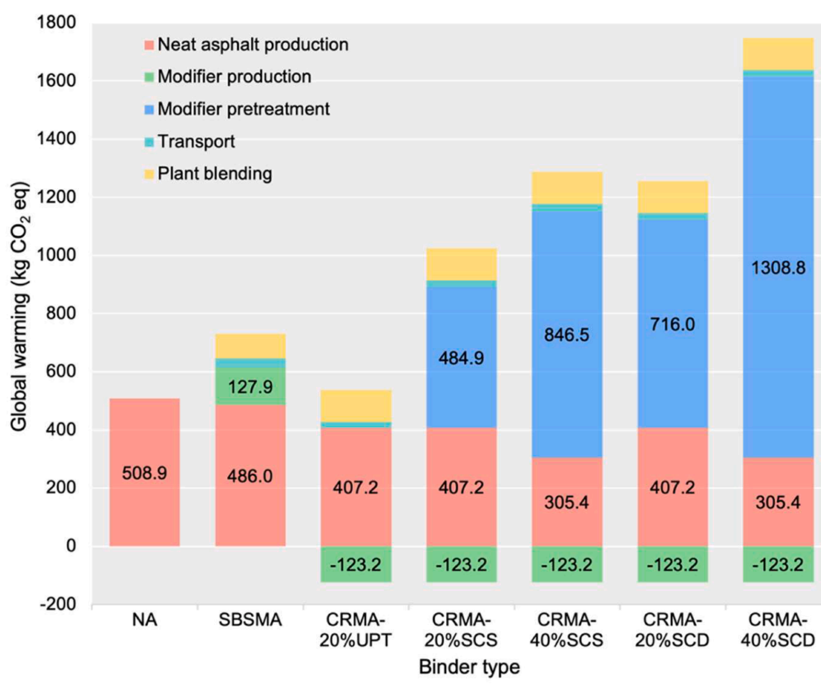

Fig. 6. Process contribution to environmental impacts of producing 1 ton of asphalt binder.

described in sub-section 3.4, while keeping the values of all parameters and remaining LCA methodological choices constant. Fig. 7 presents the results of the scenario analysis using the global warming indicator as the representative indicator. The comparison reveals that the environmental impact of the CR production process is highly sensitive to the choice of allocation method. When the chosen method shifted from the system expansion to the two allocation methods, the environmental benefits resulting from the "avoided production" of the two co-products no

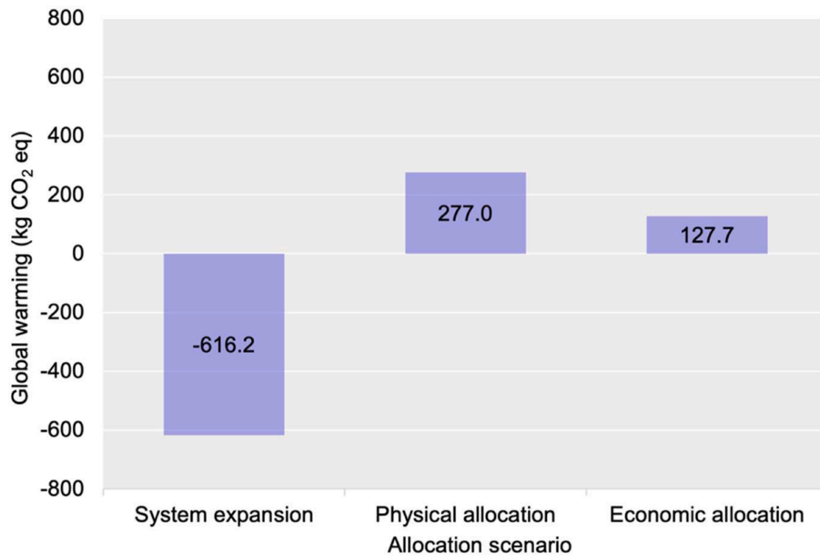

Fig. 7. Scenario analysis on producing 1 ton of CR considering different allocation scenarios.

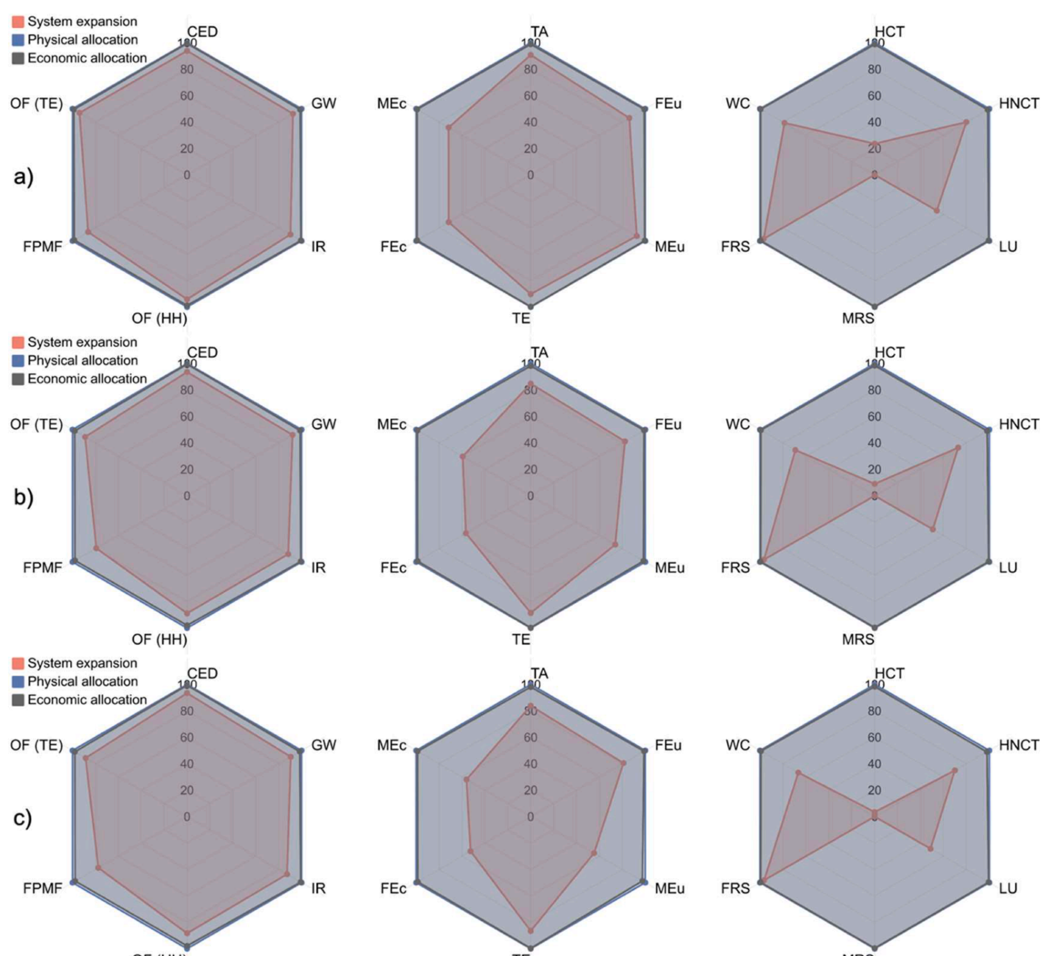

Fig. 8. Sensivity analysis results on allocation issue of CR production process, (a) CRMA-20 %UPT, (b) CRMA-20 %SCS, (c) CRMA-20 %SCD.
Key: CED= Cumulative Energy Demand; GW= Global Warming; IR= Ionizing Radiation; OF (HH)= Ozone Formation, Human Health; FPMF= Fine Particulate Matter Formation; OF (TE)= Ozone Formation, Terrestrial Ecosystems; TA= Terrestrial Acidification; FEu= Freshwater Eutrophication; MEu= Marine Eutrophication; TE= Terrestrial Ecotoxicity; FEc= Freshwater Ecotoxicity; MEc= Marine Ecotoxicity; HCT= Human Carcinogenic Toxicity; HNCT= Human Non-Carcinogenic Toxicity; LU= Land Use; MRS= Mineral Resource Scarcity; FRS= Fossil Resource Scarcity; WC= Water Consumption.

longer exists. Instead, the total environmental burden of the process is proportionally allocated among the three products using specific ratios. This led to a significant increase in the environmental impact of CR production process when using the allocation methods because the sign of the impacts in these scenarios is positive. Besides, the environmental impact of the CR production process is considerably higher under the physical allocation method compared to the economic allocation method, with the former being more than twice as high as the latter. This is because in the physical allocation method, CR, which is of high mass but low economic value, bears a larger share of the environmental burden.

The analysis above described was expanded to the asphalt mixtures. Specifically, unpretreated CRMA (CRMA-20 %UPT), supercritical swelling pretreated CRMA (CRMA-20 %SCS), and supercritical de-crosslinking pretreated CRMA (CRMA-20 %SCD) were considered to illustrate the effects of the choice of the allocation method in the environmental impact results. The analysis results are presented in Fig. 8, and the environmental impacts of each mixture are all relative scores to the highest score in the corresponding impact category. Meanwhile, the detailed environmental impacts results for the production of these asphalt mixtures under different allocation scenarios can be found in Table S10, Table S11, and Table S12, respectively, in the SI. It can be observed that for most of the impact indicators, the environmental burdens under the "system expansion" condition were considerably lower than those in the other two allocation methods, regardless of the presence of the supercritical pretreatment process of CR. This reflects the high effect of allocation issues in the CR production process on the overall environmental impacts. The difference between physical allocation and economic allocation methods was greatly reduced at the level of CRMA mixture production, where only minimal differences were observed. This indicates that when the production system of the studied materials became more complex, the overall results are not highly sensitive to the choice of attributes for the calculation of allocation coefficients.

Additionally, the effect of the selection of the allocation method varied for different environmental impact indicators among the different asphalt mixtures. Taking the GW and CED indicators as examples, the ranking of the asphalt mixtures in terms of GW score remained unchanged when transitioning from the system expansion method to physical allocation or economic allocation, while slight changes occurred with respect to CED. Therefore, although the variation in allocation methods might affect the absolute values of the environmental impact scores for individual asphalt mixtures, it does not necessarily lead to changes in the relative values (ranking) of environmental impact scores for all asphalt mixtures being compared.

4.4. Uncertainty analysis on emerging technology process

The data quality evaluation and specific uncertainty quantification

Table 6
Data quality evaluation and uncertainty quantification of CR supercritical de-crosslinking pretreatment process.

Input/Output	DQI vector	Uncertainty	Distribution
Input			
CR	(1, 5, 1, 1, 1)	1.21	Log-normal
Dry ice	(1, 5, 1, 1, 1)	1.21	Log-normal
Water	(1, 5, 3, 1, 3)	1.32	Log-normal
De-crosslinking agent	(1, 5, 3, 1, 3)	1.32	Log-normal
Transportation	(1, 1, 1, 1, 1)	1.05	Log-normal
Electricity	(1, 5, 3, 1, 3)	1.32	Log-normal
Coal	(1, 5, 3, 1, 3)	1.32	Log-normal
Outputs			
Carbon dioxide	(1, 5, 3, 1, 3)	1.32	Log-normal
Particulate matter	(1, 5, 3, 1, 3)	1.32	Log-normal
Sulfur dioxide	(1, 5, 3, 1, 3)	1.32	Log-normal
Nitrogen oxide	(1, 5, 3, 1, 3)	1.32	Log-normal

Table 7
Data quality evaluation and uncertainty quantification of CR supercritical swelling pretreatment process.

Input/Output	DQI vector	Uncertainty	Distribution
Input			
CR	(1, 5, 1, 1, 1)	1.21	Log-normal
Dry ice	(1, 5, 1, 1, 1)	1.21	Log-normal
Water	(1, 5, 3, 1, 3)	1.32	Log-normal
Transportation	(1, 1, 1, 1, 1)	1.05	Log-normal
Electricity	(1, 5, 3, 1, 3)	1.32	Log-normal
Coal	(1, 5, 3, 1, 3)	1.32	Log-normal
Outputs			
Carbon dioxide	(1, 5, 3, 1, 3)	1.32	Log-normal
Particulate matter	(1, 5, 3, 1, 3)	1.32	Log-normal

of two CR supercritical pretreatment processes are presented in Tables 6 and 7. The results of four impact indicators, namely CED, GW, fossil resource scarcity (FRS), and terrestrial acidification (TA) are shown in Figs. 9–12 for the different alternative asphalt mixtures. These indicators were selected due to different reasons. CED and GW are indicators widely used in pavement LCA studies. The selection of the FRS indicator was underpinned by the reduction in the use of petroleum asphalt due to the high-dosage incorporation of supercritical pretreated CR. Finally, the selection of TA, measured in terms of sulfur dioxide emissions, was governed by the presence of sulfur-containing gas emissions in the supercritical de-crosslinking pretreatment process of CR. The complet set of uncertainty analysis results for all mixtures considered in this case study can be found in Table S13 to Table S19 in the SI.

From the analysis of Figs. 9–12, it can be observed that regardless of the impact indicator selected, the uncertainty of the calculated scores for supercritical pretreated CRMA mixtures waere considerably higher than those of the others, as shown by the dispersion of the impact indicator scores. This is primarily due to the high uncertainty and variability of the inventory data used in the supercritical pretreatment process of CR. Among them, supercritical pretreated CRMA mixtures exhibited the highest uncertainty in the GW score, mainly because the CO_2 emissions data in the supercritical pretreatment process were largely originated from the less reliable laboratory data estimations. This is often an unavoidable challenge in prospective LCA with emerging technologies. However, despite the presence of uncertainty, in most cases, the differences among the impact scores of CRMA mixtures and reference asphalt mixtures (i.e., NA and SBSMA) remained clearly visible. Note that the GW was an exception due to the extensive use of CO_2 in the emerging supercritical pretreatment process and the associated relatively high inventory data uncertainty. Therefore, the assertions drawn in the previous sections based on the deterministic LCA remain largely valid.

In addition to CED and GW, some findings can be outlined from the analysis of the distribution scores of the remaining two indicators. Supercritical pretreated CRMA mixtures exhibited lower fossil energy consumption values, with higher CR content resulting in even lower values. Conversely, SBS modified asphalt (SBSMA) mixture exhibited the highest fossil energy consumption, followed by NA mixture. This can be inferred from the considerable reduction in petroleum asphalt consumption achieved through the high-dosage incorporation of supercritical pretreated CR. However, SBS modifier and NA, as typical petroleum and chemical products, had higher fossil energy consumption values. Furthermore, the results of TA also show that supercritical de-crosslinking pretreated CRMA mixtures had the largest score in this impact category, accompanied by high uncertainty. However, this high uncertainty does not overshadow the considerable differences between them and the other mixtures, including supercritical swelling pretreated CRMA mixtures. This emphasizes the need for special attention to the emission and treatment of sulfur-containing waste gas during the supercritical de-crosslinking pretreatment stage of CR in the production

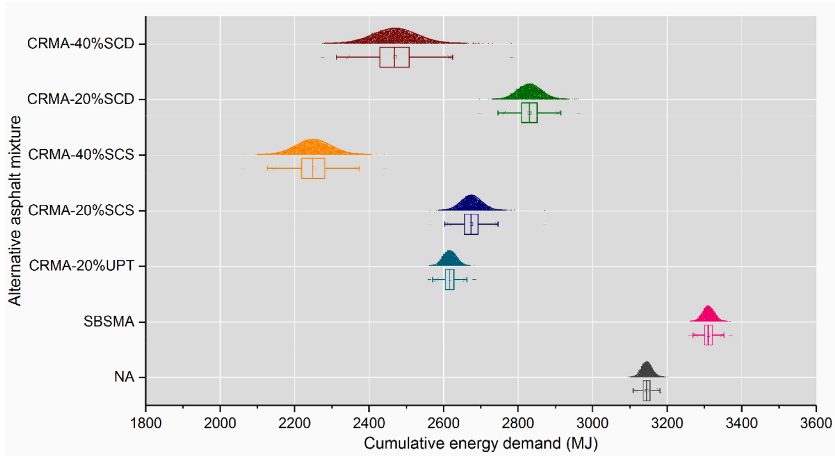

Fig. 9. Uncertainty analysis results expressed in terms of cumulative energy demand score.

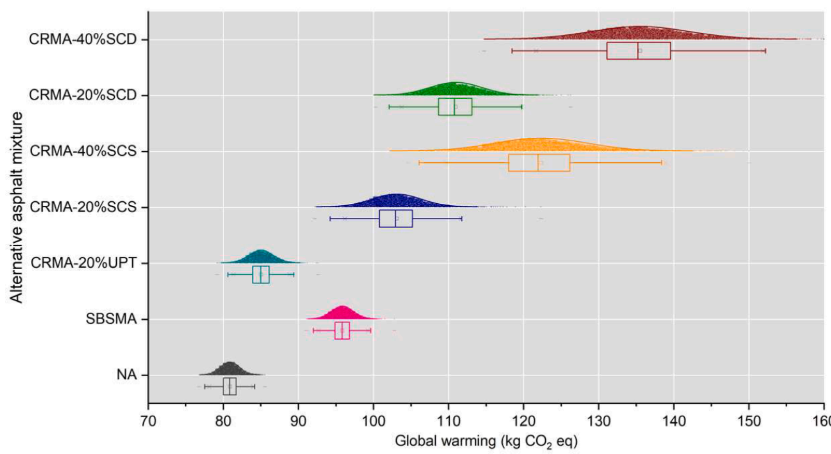

Fig. 10. Uncertainty analysis results expressed in terms of global warming score.

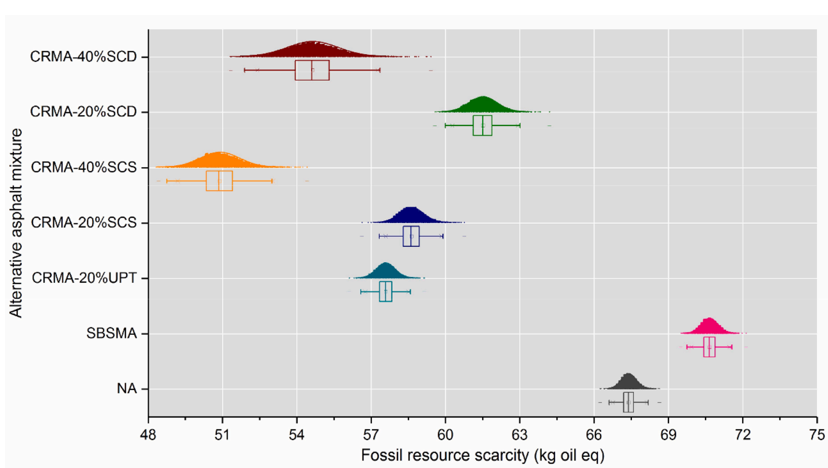

Fig. 11. Uncertainty analysis results expressed in terms of fossil resource scarcity score.

process of such mixtures.

4.5. Scale-up scenarios based on LCA hotspots

Four "what-if" scenarios were formulated based on hotspots identified in the laboratory case. The details of these forthcoming scenarios are explained in Table 8. They specify several potential advancements aimed at upscaling the laboratory-based system, based on the hotspot analysis above. These four scenarios have technically evolved from the emerging CR pretreatment technology over time all the way to the best performing scenario from an environmental perspective. Besides, they are presented in a decreasing order of ease of implementation.

In scenario 1, it is assumed that no change in the production process and materials formulation occurred at the future commercial scale,

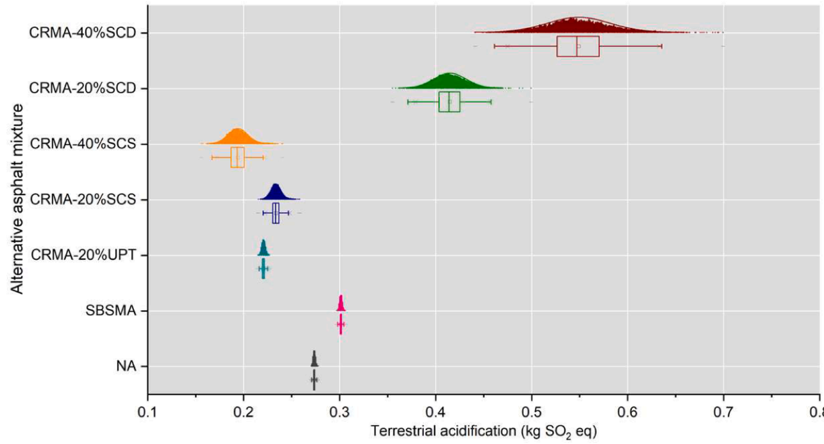

Fig. 12. Uncertainty analysis results expressed in terms of terrestrial acidification score.

Table 8
Scale-up scenarios regarding the emerging CR pretreatment process (i.e., supercritical pretreatments).

Scenario	Technology change	Description
Scenario 1	Baseline scenario without change	No change in the production processes and material formulation
Scenario 2	Collection and reuse of waste carbon dioxide based on scenario 1	The carbon dioxide is reused for the subsequent batch of CR pretreatment, thus avoiding the emission.
Scenario 3	Reduction of the pressure of CO_2 (i.e., the amount of dry ice) for supercritical fluid environment based on scenario 1	The pressure of CO_2 is reduced from 10 MPa to 7.5 MPa, thus generating subcritical fluid environment.
Scenario 4	Reduction of the dosage of chemical reagent based on scenario 1	The dosage of chemical reagent is reduced from 10 % to 5 %.

compared to the current laboratory scale. This scenario is used as the baseline scenario for comparison. Scenario 2 is considered as the one with the highest viability and likelihood at industrial scale. In this scenario, the aftercooling CO_2 derived from its supercritical state can be collected and used again to generate the supercritical fluid environment for the next batch instead of being directly emitted to the air, after one batch of CR pretreatment. In scenario 3, the amount of dry ice in the recipe to achieve the corresponding pressure of supercritical CO_2 at the target temperature is seen as another feasible improvement on the basis of scenario 1. The pressure of CO_2 is reduced from 10 MPa to 7.5 MPa, with corresponding reduction of material demand of dry ice. Note that the critical pressure of CO_2 is 7.29 MPa, thus the CO_2 under this pressure (i.e., 7.5 MPa) can be categorized yet into the supercritical fluid and possesses similar characteristics to that under 10 MPa in the applications. Scenario 4 explores another possible pathway for commercial use, namely the decrease in dosage of chemical reagent. It was observed that the huge difference between the environmental impacts of supercritical de-crosslinking pretreated CRMA and supercritical swelling pretreated CRMA is caused by the use of chemical de-crosslinking reagent. Besides, the determination of chemical reagent dosage in the laboratory recipe is based on the redundancy design methodology in this study, which means that at this stage the dosage of this reagent in the lab scale is intentionally more than the required amount. Meanwhile, the reagent can be expected to be used more effectively in the industrial applications. Thus, in scenario 4, the recipe is improved by reducing the use of chemical reagent from 10 % to 5 %. It should be mentioned that this reduction in de-crosslinking reagent can be seen as the most challenging technological improvement from the current lab-scale setup, considering its dominant role in the supercritical de-crosslinking pretreatment.

Similarly, four impact indicators, including CED, GW, FRS, and TA, were chosen for the comparison of the environmental performance of the four scale-up scenarios, and the results are displayed in Fig. 13. Firstly, it should be mentioned that the changes forecasted in the scale-up scenarios do not affect the environmental impacts of NA, SBSMA, and unpretreated CRMA (CRMA-20 %UPT) mixtures, since they do not involve the emerging CR pretreatment process. On the contrary, all scenarios led to a reduction of the impact scores of the other three mixtures compared to scenario 1. Compared to the baseline scenario, the collection and reuse of waste CO_2 in scenario 2 can remarkably reduce the GW score of both supercritical de-crosslinking pretreated CRMA and supercritical swelling pretreated CRMA by up to 30 %, whereas a slight decrease (less than 5 %) can also be observed in terms of the other three impact indicators. Besides, this can be seen as the most effective method to reduce the CO_2-eq emissions of these alternative asphalt mixtures. In scenario 3, the decreased use of dry ice for the generation of supercritical fluid medium can further lower the CED, FRS, and TA scores to some extent, comparatively to the scenario 2. Furthermore, although the reduced dosage of chemical reagent does not significantly improve the environmental performance of supercritical swelling pretreated CRMA, this greatly reduces every impact score of supercritical de-crosslinking pretreated CRMA (especially the TA with a decrease of almost 50 %). Finally, it is worth mentioning that a reduction of the impacts scores from scenario 2 to scenario 4 can be seen in all impact category but GW, and this indicates that the collection and reuse of waste CO_2 in scenario 2 plays the most important role in reducing CO_2 emissions.

It should also be noted that there are some limitations with respect to the scenarios development. At this stage, four scale-up scenarios were modeled centered on the foreground system, namely the studied emerging CR pretreatment process, which is the research focus of this study. It is desirable that forthcoming research efforts explore alternative background scenarios (e.g., energy transition), leveraging future available and reliable data resources related to the study's geographical scope.

5. Conclusions

This study conducted a prospective LCA on the valorization of ELTs in asphalt mixtures considering the application of two emerging CR pretreatment technologies, by using proxy data to upscale the critical processes. Further, scenario and uncertainty analysis were also performed, and scale-up scenarios in the future setting were designed. The primary conclusion can be drawn as follows:

For the conditions considered in the case study the supercritical swelling pretreated CRMA with 40 % dosage generally obtained the lowest environmental impacts, among multiple scenarios of valorizing ELTs in asphalt mixtures. Although the increased incorporation rate of

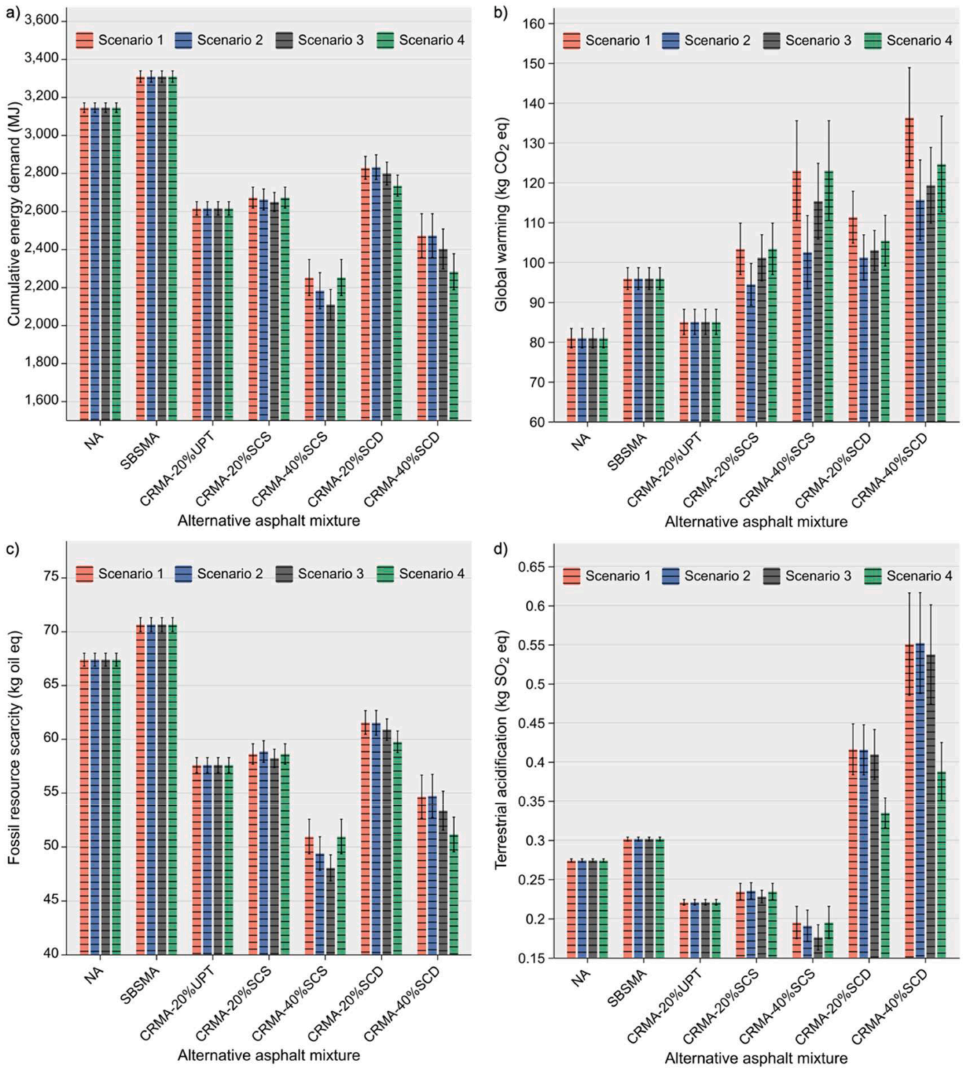

Fig. 13. Comparative environmental impact results of scale-up scenarios: (a) CED, (b) GW, (c) FRS, (d) TA.

CR was beneficial to the environmental performance of asphalt mixtures, the supercritical pretreatment process of CR played a critical role in the overall environmental impact, especially in the GW impact category The use of the system expansion method to deal with co-production of CR resulted in additional environmental benefits compared to the allocation methods, although the overall differences between the two allocation methods were minimal.

Further, an uncertainty analysis was successfully performed with the Pedigree metrix method and Monte Carlo simulation, to account for and propagate the uncertainties into the LCA results. The environmental impact scores of supercritical pretreated CRMA mixtures exhibited relatively high uncertainty due to the data variability of the emerging CR pretreatment process inventory. In most cases, the uncertainty in the environmental impact scores of individual supercritical pretreated CRMA material did not outweigh the differences observed among the compared materials. Finally, scale-up scenarios were designed considering future technology improvement of these emerging CR pretreatment processes. It was found that the reuse of CO_2 as reaction medium and the reduced use of dry ice and chemical reagent in the recipe proved to be effective in reducing the overall environmental impact.

All in all, this study emphasizes the need for special attention to the impacts of data gaps related to emerging technologies and innovative materials in pavement LCA studies. Thus, future efforts should be devoted to collect primary data representative of the production of asphalt mixtures with innovative/alternative materials.

Disclaimer

All authors confirm that there is no conflict of interest in this article.

CRediT authorship contribution statement

Jin Li: Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **João Santos:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Supervision. **Andrea Vargas-Farias:** Writing – review & editing, Software, Data curation. **Daniel Castro-Fresno:** Writing – review & editing. **Feipeng Xiao:** Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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