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Environmental Performance of Road Asphalts Modified with End-of-Life Hard Plastics and Graphene: Strategies for Improving Sustainability

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Abstract: Road construction takes a heavy toll on the environment. Therefore, innovative strategies to improve the environmental performances of this sector are needed, and the use of recycled materials (e.g., plastic) has been recently pursued to achieve this goal. The present work aims to (i) assess the environmental benefits deriving from the use of recycled hard plastics in combination with graphene to generate a new bitumen modifier and related asphalt mixture (AM) formulations (ii) to compare the performance of the bitumen modified using this new modifier with the bitumen modified using a traditional polymer (Styrene-Butadiene-Styrene, SBS) and the non-modified bitumen. A detailed Life Cycle Assessment (LCA) study was performed according to a cradle-to-cradle approach. Different scenarios were compared, including the variability of the pavement's layers thickness and the amount of reclaimed asphalt pavement during the road maintenance cycles. The results demonstrated that the addition of the innovative modifier enhanced the structural performance of AMs, which turns into pavement extended durability, reduced maintenance cycles as well as into reduction in raw material use. The innovative asphalt modifier also creates a synergistic effect by offering a valuable alternative to hard plastic incineration by using it as a secondary raw material. This analysis allowed us to indicate the new-modified AM as the solution with the least environmental burden in all impact categories, suggesting its significant role in implementing new strategies to improve the environmental sustainability of road pavements.

Keywords: LCA; sustainability; end-of-life plastics; hard plastics; environmental performance; circular economy; road pavement

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1. Introduction

Road construction and road paving constitute one of the main activities in Europe that support transports and logistical infrastructures. In 2015 in Europe, plus Norway and Turkey, 280 million tons of hot and warm asphalt mixture for road paving have been produced, with continuous increasing rate reaching about 300 million tons produced in 2018 [1].

Construction industry gives a great contribution in terms of environmental impacts on a worldwide scale, in particular road construction, in which greenhouse gas (GHG) emissions represent 5–10% of total GHG emissions produced by the transport sector; but they are growing rapidly, especially in developing countries [2,3]. Significant quantities of GHG are emitted in producing materials for the construction, maintenance, rehabilitation of highway infrastructure [4].

The most common types of materials used for road pavements are bitumen and aggregates, especially natural gravel; this indicates that large quantities of natural resources are employed for construction and maintenance of road infrastructure and that large amount of asphalt waste are generated during decommissioning of roads. There are several strategies rendering this techno-sphere process more environmentally sustainable. To point in that direction, it is necessary to consider the road pavement through its life cycle and to consider its technical performance. In the road industry, innovative technologies have been developed, such as the use of recycled asphalt and/or of additives/modifiers that extend the pavements service life.

The asphalt mixture (AM) removed from an existing road pavement (reclaimed asphalt pavement- RAP) consists of valuable non-renewable resources, which can be re-used in new AMs since it is a fully recyclable material for construction [5]. This can help reducing the demand for virgin aggregates and bitumen as well as mitigating their overall cost [5,6]. Although reclaimed asphalt is currently added to a new asphalt mixture up to 15–20% by weight due to technological limitations, the latest advances in asphalt plants, best practices and cutting-edge technologies demonstrate the possibility of increasing the amount of reclaimed asphalt in the production of AMs to high (over 40%) or to very high (up to 100%) RAP content [7–11]; thus further enhancing the circularity scheme and reducing the impacts [12–14]. The encouragement of its use reflects the worldwide trend to face the existing environmental issues by trying to increase the efficient use of resources and reduce carbon emission.

The employment of bitumen modifiers may also contribute to the reduction in the impacts of asphalt pavements, since it may allow improvements of the pavement technical performance and resistance, thus providing an extended and safer pavement service life. Common modifiers include polymers of different nature, which improve the rheological properties of asphalt [15–17]. Other alternative/additional materials used to enhance road performance are for example rubber [18,19] and steel [20]. In recent years, nanomaterials (NMs), with their unique properties, such as "small size effect", "surface and boundary effect" and "quantum size effect", have become the most popular research topics in many fields [21]. NMs have attracted a lot of attention from pavement researchers. Research results have shown that specific NMs, such as graphene nanoplatelets (GNPs) [22], graphene oxide (GO) [23], and graphene nanoribbons (GNRs) [24,25] can greatly improve the mechanical properties, aging resistance, fatigue resistance, and adhesion properties of asphalt binders. For this reason, it would be desirable to implement a further level of circularity in the roads paving life cycle, also including polymers and NMs.

In this context, within the ECOPAVE project (funded by the European Union and the Italian Lombardy region, where the project was carried out) [26] a new modifier has been attained by developing a new hybrid combination of end-of-life hard plastics and graphene to obtain a high performance-AM with extended service life. Various types of waste plastics are used as modifiers [27], but since hard plastics are generally rarely recovered, this solution enables employing them as secondary raw material, and introducing them in virtually infinite number of use cycles, as alternative option to incineration. The proposed hard plastics circularity model is an interesting "case study" in accordance with the European Plastics Strategy [28], which defines waste hierarchy as follows: prevention, reuse, recycling, energy recovery and landfilling, as the last option. The modified AM functional performance may be further enhanced by the use of graphene, which is another component of the innovative hard plastics-based innovative modifier. In line with the approach of nanotechnological solutions, a small amount of NMs may strongly enhance the functional performance of products.

Indeed, the incorporation of the obtained granulated graphene-added polyolefin blend, in the AM has been seen enhancing the asphalt performance both in terms of thermal and mechanical resistance as well as of durability [26].

Within the framework of circularity, this study aimed at assessing the potential environmental comparative impact throughout the life cycle of standard road pavements (with neat bitumen), pavements containing a traditional polymer-based modifier (styrene-butadiene-styrene (SBS)) and pavements containing the innovative hard-plastics and graphene-based modifier. This study is meant as a support for the decision-making process concerning the choice of the best available materials and processes for new or to-be-renewed sustainable road pavements. The analysis was performed by means of LCA methodology considering the pavement whole life cycle. The comparison has been defined for two case studies. The first one allows for the extension of the service life for each layer (base, binder and wear) with respect to a benchmark pavement without modifier; the second one considers asphalt layers reduced thickness having the same AM performance of the standard pavement. An equally important aim was the assessment of the environmental performance of AM containing different shares of RAP, dealt with in the third case study.

2. Materials and Methods

The analysis of the impacts was performed using the LCA methodology according to ISO 14040 and 14044 [29,30] which consists of four interconnected phases: goal and scope definition, inventory analysis, impact assessment and interpretation. The methodological details for each phase are described in the following subsections.

2.1. Goal and Scope Definition

This study was performed referring to the Italian context, with a focus on the Northern area of the country (Lombardy Region), and to extra-urban roads. The main goal was to conduct a comparative analysis to determine the potential environmental impacts of roads paved with a modified AM (innovative MAM) containing a modifier made of recycled hard plastics and graphene. The comparative terms of this analysis are: (1) roads paved with MAM containing the conventional modifier Styrene-Butadiene-Styrene (SBS-MAM), and (2) roads paved with standard non-modified AM (SAM).

Three case studies were analyzed referring to different: (a) production schemes, (b) paving solutions, and (c) system model circularity index values.

In all considered cases, the road paving analysis has been limited to the three layers containing bitumen: wearing course, binder, and base.

The study has also been carried out with two different LCA software in order to cross check the consistency of the quantitative outcomes as well as to assess the computational tool-independence of the results obtained.

The three presented case studies are referred to three different paving solutions and are organized as follows:

(1) The first comparative case study is based on the assumption that the pavement stratification dimensional specifications cannot be changed, whereas the composition of the AM used may be changed towards improved performance. Thus, this case study considers roads with equal pavements stratification characterized by the same thickness for wearing course, binder and base layer, produced with three different AMs (Tables 1 and 2).
(2) The second comparative case study releases the pavement stratification design constraints allowing for modification of the layers thickness as well as for a more rigorous assumption of functional unit, which requires getting the same AM layers stiffness and durability (pavement functionality) among the corresponding layers of the investigated solutions (Tables 1 and 3).
(3) The third case study compares the impacts of three different system model circularity indexes determined by different amounts of RAP employed in the production of AM (Tables 1 and 4).

The first two comparative cases studies considered have a twofold objective: (a) to both assess the comparative environmental impacts of the different solutions proposed for unmodified and modified asphalt mixture, and (b) to assess the reproducibility of LCA computations and any possible dependence of the obtained impact assessment results due to different LCA software and related databases as computational tools.

Table 1. Summary of the three case studies analyzed.

Case Study	Variable in the Case Study	Description
First case study	Production schemes	Comparison of innovative MAM, SBS-MAM and SAM: same thickness for wearing course, binder and base across all AMs thus different time spans according to the AM performance (see Table 2).
Second case study	Paving solutions	Comparison of innovative MAM, SBS-MAM and SAM: same time span for all AMs, thus reduced thickness of the layers containing the innovative MAM and SBS-MAM (see Table 3).
Third case study	System model circularity indexes	Comparison of AMs containing different shares of RAP (see Table 4).

Table 2. First case study. Characteristics and performance of different asphalt pavements with same thickness and different service life for each layer determined by pavement layers technical performance (AM stiffness and durability).

Type of Asphalt Mixture (AM)	Layer	Thickness [cm]	AM Amount [t] for L = 1 km, W = 15 m	Service Life According to Maintenance Limits [Years]
Standard AM (SAM)	Wearing	5	1785	5
	Binder	6	2142	10
	Base	14	4998	20
SBS modified AM (SBS-MAM)	Wearing	5	1785	8
	Binder	6	2142	16
	Base	14	4998	32
Innovative modified AM (MAM)	Wearing	5	1785	15
	Binder	6	2142	30
	Base	14	4998	90

Table 3. Second case study. Characteristics and performance of different asphalt pavements with different thickness and equal AM stiffness and durability for each layer.

Type of Asphalt Mixtures (AM)	Layer	Thickness [cm]	AM Amount [t] for per L = 1 km, W = 15 m	Service Life According to Maintenance [Years]
Standard AM (SAM)	Wearing	5	1785.0	5
	Binder	6	2142.0	10
	Base	14	4998.0	20
SBS modified AM (SBS-MAM)	Wearing	4.3	1535.1	5
	Binder	5.5	1693.5	10
	Base	12.9	4605.3	20
Innovative modified AM (MAM)	Wearing	3.8	1356.6	5
	Binder	4.8	1713.6	10
	Base	11	3927.0	20

Table 4. Third case study. Share of RAP pavement considered in each scenario: Reference scenario (also applied to the first two case studies), first scenario with increased recycled RAP rate (enhanced circularity index), second scenario with decreased recycled RAP rate (lowest circularity index).

Layer	Reference	Scenario 1	Scenario 2
Wearing	20%	30%	10%
Binder	30%	50%	20%
Base	30%	50%	20%

2.1.1. Functional Unit

The functional unit (FU) is a measure of the performance of the analysed product system and defines a common basis for performing comparisons among the environmental performance of the considered optional case studies. The FU for road pavements is defined herein by their geometry (physical dimensions), service life, and levels of traffic supported [31]. To compare the different pavements, the length and the width of the road is the same for all alternatives, whereas the pavement thickness can vary and is determined by conventional pavement design methods; so that all analyzed solutions comply with the functional specifications, requiring withstanding the same traffic load and intensity within the road service life.

Given the different perspectives considered in the three case studies, suitable FUs have been proposed for each case study, involving pavement technical performance, and service specifications to run fair comparisons among the three paving solutions investigated.

The functional unit for case study one and three is defined through physical and time parameters: a kilometer of pre-existing extra-urban road (width: 15 m) used for 20 years with a traffic intensity of 4,000,000 vehicles per year and including the ordinary maintenance. The FU refers to the first three layers of the road (those containing bitumen). Twenty years was chosen as time reference because it is the useful life of the standard pavement. In case study one, all pavement types considered have the same thickness (4 cm wearing course, 6 cm binder and 14 cm base) and therefore different maintenance frequencies according to their performance (see Table 2). In case study two, the thickness of the layers of each pavement type is reduced in order to obtain the same maintenance frequency for all pavements (Table 3).

The road stratification design and slabs (layers) thickness are determined by the structural number (SN), which—in turn—is derived by the structural performance of the asphalt mixture, defined by the W18 index, accounting for a specific number of axels at specified load the road can stand without degradation.

Road characteristics with SN and W18 are presented in Supplementary Materials (S1). SN and W18 are strictly implied in the definition of the FU considered in this work for the second case study analyzed.

In the third case the analysis of the impacts changes due to the increase or reduction in the RAP used is assessed. For this purpose, the FU considered is 30 tons of AM produced, corresponding to the amount of obtained AM in one processing batch lasting 45 minutes.

2.1.2. System Boundaries

For the first and second case studies the system boundaries of the analyzed systems encompassed all the life cycle phases of the asphalt mixtures in a “from-cradle-to-cradle” perspective, considering a circular scheme in which RAP (obtained by modified or non-modified AM) enters the AM production process in subsequent cycles. The Functional Systems (FS) compared in the different case studies were treated according to a modular framework, which allowed for the required modelling flexibility to consider different scenarios. The processes considered are shown in Figure 1. Processes common to the three systems under assessment (SAM, SBS-MAM, innovative-MAM): raw material extraction and processing, production of AM, road paving, road maintenance and demolition, collection and processing of RAP. Processes that differ among the three system under assessment (in the dashed boxes of Figure 1): production of the modifiers and fate of end-of-life plastics.

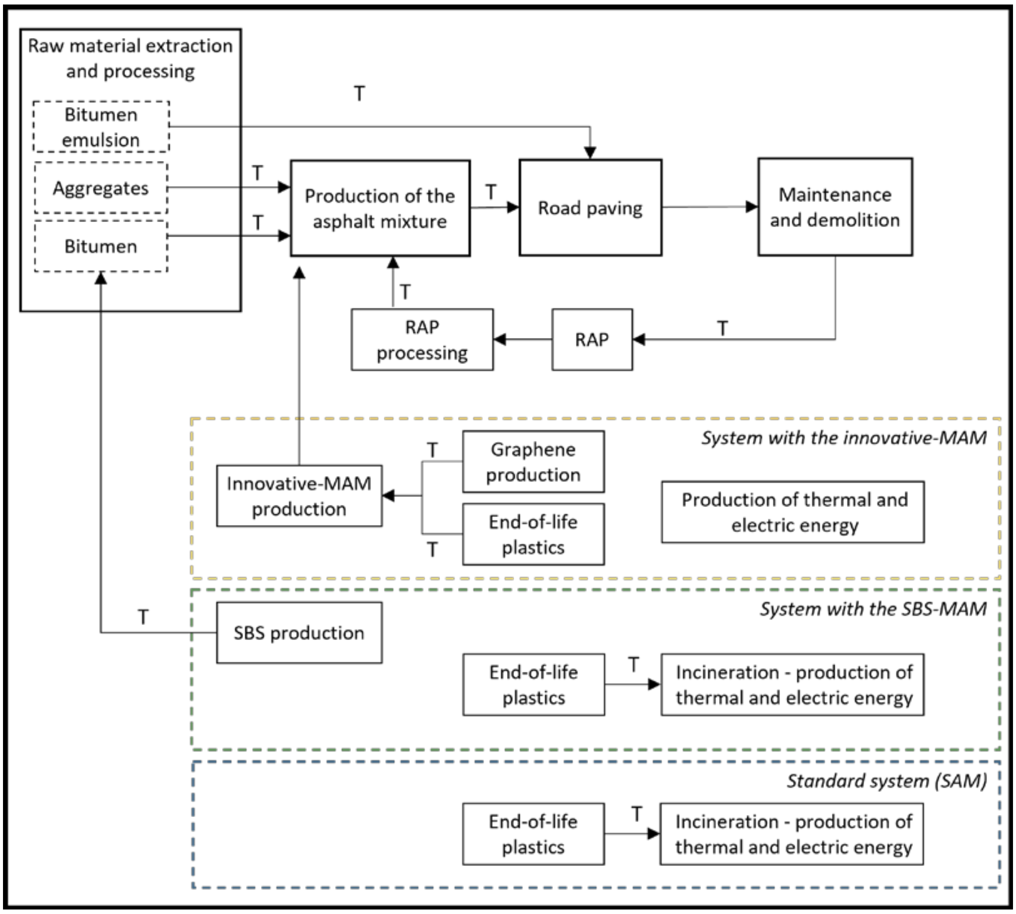

Figure 1. System boundaries of the analysis. The three dashed boxes correspond to the three systems under comparison. Yellow: system with the innovative-MAM; Green: System with the SBS-MAM; Blue: Standard system SAM. T stands for transport processes.

Concerning this last point, a fair comparison among the solutions was assured by the inclusion of the fate of the end-of-life hard plastics for all systems. Indeed, the option of reusing hard plastics side streams implies avoiding its conventional incineration. Therefore, the system boundaries also include the parallel production of the equivalent amount of thermal and electrical energy through national energy mix that would be obtained by incinerating the amount hard plastics used for the innovative-MAM (see yellow dashed box in Figure 1). For a coherent analysis, an extension of the conventional functional system for producing the standard AM and the SBS-MAM was implemented to consider the incineration of the amount hard plastics diversely used in the innovative modifier. This way the hard plastic end-of-life options are included in a balanced way among the different investigated solutions (see green and blue dashed boxes in Figure 1).

For the third case study referred to the amount of RAP employed as secondary raw material in the AM production process, the functional system considered includes: raw material extraction and processing; innovative modifier production (for the MAM solution only); asphalt mixture production; pavement reclaiming process (maintenance and demolition) considered as a (secondary) raw material sourcing process; transport of raw materials from the sourcing sites to the production site.

Considering the end of life, as described in the previous paragraph, part of the material removed during demolition operation (RAP) re-enters the supply chain. The fate of the milled material that is not closed-loop recycled varies according to the context. The scope of this study is the Norther Italy and, as stated in a dedicated sector report [32] in the

Lombardy Region less than 1% of demolition waste is headed to landfill, since most of it is recycled or stored. Therefore, with the same approach of Giani et al. [33] and of the PCR (Product category rules), the current analysis does not include the environmental impact of the milled material after the storage phase, since it belongs to the downstream supply chain, where it will be treated as secondary raw material.

2.1.3. Data Quality, Representativeness and Assumptions

When not otherwise indicated, the core processes (production of the AM, road construction and demolition, production of the innovative modifier) of the described supply chain are covered by primary data, provided by both the experts who are developing the modifier, since they work closely with the pavement industry, and their partners. For the background processes, whenever primary data were not available, literature data (from sector reports and scientific papers cited as references) and data from database have been used (namely, the database of GaBi and of OpenLCA).

Once external conditions have been fixed (e.g., climatic conditions, characteristics of the foundations, etc.), the lifespan of asphalt pavements is function of its structural performance and functionality throughout the time. Different layers and different AMs have different lifespans. The lifespans were quantified by experts via functionality tests on samples of pavements according to international standards (AASHTO Road Test). Asphalt pavements last many years, hence assessments of field data for the mixture containing the modifier would not have been possible at this stage.

The production of the modifier has not been scaled up yet, so data available on the industrial production process (namely consumption of energy and water) come from the design phase (in order also to allow the results of the environmental assessment to be included in the final decisions).

No direct measurements of emissions to air from the BM production plant were carried out during the study. Nevertheless, data from measurements done in 2015 on a standard plant were made available by sector experts and could be used as a proxy.

As far as the representativeness is concerned, we refer to (i) technology, (ii) geographic and (iii) temporal representativeness. (i), (ii) Technology and geographic location of the core industry processes—related to pavements production and demolition, modifier production and incineration—represents the current technology in use in the area of the assessment. Data of background processes mainly refer to national and European averages, or, in few cases, to global averages when other options are not available. (iii) The first paving cycle included in the analysis is assumed to take place in the current period and the available data cover this time frame; therefore, we are assuming that these values and settings are still valid throughout the subsequent maintenance cycles.

2.2. Inventory Analysis

The LCA analysis required a large amount of input data, mainly coming from companies involved in the ECOPAVE project, with additional minor assumptions. The next paragraphs provide a summary description for all life cycle phases.

2.2.1. Raw Materials Extraction Collection and Processing

Extraction collection and processing of raw materials consists of extraction of limestone from quarries to produce aggregates and production of bitumen via refining of crude oil. Aggregates share in the AM represent from 94% to 96% of the final product, whereas bitumen 6–4%, respectively. In SBS-modified bitumen, SBS amounts to 3% of the bitumen's weight; in the asphalt mixture containing the innovative modifier, the modifier amounts to 5% of the bitumen's weight. Data associated with the production of aggregates were obtained from the Gabi and OpenLCA database: Limestone, crushed gravel, grain size 0–2 mm. The databases were also used to model bitumen production.

The system includes the production of the bituminous emulsion whose composition is: bitumen 55%, water (from public supply) 44.5%, ethyl-diamine 0.3%, hydrochloric acid 0.25% [34].

2.2.2. Asphalt Mixture Production

The production of the AM takes place in a dedicated plant, where aggregates are dried prior to be processed in the mixing tower, where aggregates, bitumen (and modifiers) are mixed. Temperatures vary from 160 °C (for SAM) to 170 °C (for the innovative modifier), according to the aggregate type. Aggregates and bitumen are mixed in the mixing tower. The SBS is added prior to bitumen, whereas the innovative modifier is added in the mixing tower. At the production site batches of three tons (3 t) are processed every 45 s. It was considered that 1 ton of AM production requires the consumption of 9 kWh/t of electricity and 8 m^3/t of methane, according to the data provided by the project's partners.

Besides virgin materials, a variable share (according to case study) of RAP coming from the demolition and pavement removal (maintenance) operations of the pre-existing road layers, enters in the production process.

All production internal logistics material transfer is managed by the wheel loader, whose fuel consumption has been accounted for. The reference fuel consumption and emissions by the wheel loader are referred to the Swiss database *Nonroad* [35]. Total emissions of the AM production are the result of measurements by the manufacturing companies.

2.2.3. Road Paving

Once ready, the asphalt mixture and the bituminous emulsion are carried from the AM production site to the construction site where the different layers (wear, binder and base) are paved. Bitumen emulsion is sprayed between the pavement layers to create a stabilizing and adhesive effect.

The paving process for the three layers consists of multiple operations:

(a) Bituminous emulsion sprinkling is supplied at 0.6 kg/m^2 for each layer.
(b) The paver, assisted by the dump truck for a continuous asphalt supply, lays out the asphalt mixture.
(c) The roller compacts the pavement layers. The number of passes depends on the AM type: 6–8 passes and 8–10 are accounted, respectively, for the SAM and the MAM with the innovative modifier.

In the modelling, fuel consumption of all vehicles (paver, dump truck, and roller) has been considered according to the capacity and performance of the specific machinery (Table 5).

Table 5. Machinery parameters.

Machine	Power class	Productivity	Fuel Consumption
Paver	18–37 Kw/h	140 t/h	18.60 kg/h
Dumper and Tippers	<18 kW	-	13.93 kg/h
Roller	<18 kW	40 m^3/h	13.93 kg/h
Emulsion applier	-	0.025 km^2/h	4.24 kg/km

Emissions in the paving process are referred to on field measurements by companies.

2.2.4. Road Maintenance

Maintenance operations are represented by removal of one or more pavement layers, by milling and brushing to obtain RAP.

Milling and brushing and trucks for collecting the RAP are involved in this process. The on-site fuel transport to feed such work vehicles and their consumption were included in the model. Table 6 contains the fuel consumption of the construction vehicles.

Table 6. Machinery parameters.

Machine	Power Class	Productivity	Fuel Consumption
Milling machine	75–130 kW	104 t/h	104.74 kg/h
Dumper and Tippers	<18 kW	-	13.93 kg/h
Sweeper	-	0.038 km^2/h	15 L/h

2.2.5. Reclaimed Asphalt Pavement

Before laying the new AM, the pre-existing pavement layer (or layers, according to the number of layers to be replaced) is removed and part of it re-enters the production process. The percentage of RAP in the AM is defined according to the scenarios described in Section 2.1. Once the materials have been removed, it is transported to the AM production plant, where it is sieved and milled (2 kWh/ton of RAP) to be ready for use.

2.2.6. Innovative Modifier Production Module

The relevant components in the innovative modifier are: recycled hard plastics (mainly constituted by polyolefins, such as polyethylene and polypropylene), polyvinyl butyral (PVB, an adhesion promoter acting on bitumen and aggregates to enhance their adhesion) and graphene to improve the efficiency in mixing phase (see also Table S5).

The modifier production implies two treatment lines, one for the collected hard plastics and one for the treatment of PVB. End-of-life plastics of various origins are collected, separately in municipal collecting sites, manually separated from any metal parts and transported to a plastic recycling plant. The selection of the plastic fraction suitable for the process is performed through a floating process (the water is then internally treated and recycled). Afterwards, the plastic is milled and added to the other components. PVB comes instead from glass recycling plants, being used as plastic film to wrap, e.g., car glasses, and it is treated to remove all glass residues. The data needed to describe these processes are directly sourced from the producer.

Graphene is produced starting from expandable graphite provided by a United States company from natural graphite. The intercalating agents used in the graphite expansion process are sulfuric and nitric acids. Further expansion worked out by graphite thermal expansion and ultrasound exfoliation in acid solution medium and water removal to obtain graphene [36]. At the final stage, plastics, PVB and graphene are mixed to produce the modifier in a granular form, which is packed in big bags (Flexible Intermediate Bulk Containers, FIBC) which are made of polypropylene. The data on the production of polypropylene fibers was extracted by the LCA databases.

2.2.7. Incineration Process

In non-modified asphalt mixture (SAM) and in SBS modified AM, the end of life for hard plastics is incineration. The reference incineration plant considered is A2A located in Brescia, which technical and performance data are available from public reports [37]. The discarded portion of hard plastics and PVB in the production process of the innovative modifier are also sent to incineration, as well as the big bags after their use.

2.2.8. Conventional Modifier: Bitumen Production with SBS for SBS MAM

The formulation of SBS-modified bitumen implies the use of Styrene-Butadiene-Styrene and Sulphur, which are added to the bitumen at the refining plant premises [34] and whose production was modelled with data from the LCA databases. The detailed chemical composition and the energy requirements for the mixing process of bitumen and the other components [37,38] was included.

2.2.9. Transport

The transport module is organized in sub-modules accounting for each unit of transport. The transport processes modelled are:

- Raw material transport. The distance for aggregates and for bitumen was assumed to be, respectively, 20 km and 200 km according to expert judgment), since aggregates are usually supplied from the quarries in the Apennines mountains and bitumen comes from the Romagna region. The truck selected for this transport is transport, freight, lorry 16–32 metric ton, EURO4 from the LCA databases used.
- Transport from AM production site to the paving site. The maximum distance between the production site and the paving site is intrinsically constrained by the AM temperature which determines the viscosity to be met for the paving process. The minimum accepted temperature is 20 °C less than the temperature the AM has in the mixing tower. On average, the transport causes a temperature drop of about 10 °C/h. Therefore, the production site has to be close to the paving site and an average distance of 50 km has been selected as a representative one. The truck used for this transport is transport, freight, lorry 16–32 metric ton, EURO4 from the LCA databases used.
- Transport of RAP. RAP is transported from the paving site to a storage area and then to the AM production site. The average distance considered is comparable to the one for carrying the AM from the production to the paving site. The truck used for this transport is transport, freight, lorry 16–32 metric ton, EURO4 from the LCA databases used.
- Transport of modifier SBS and related items. This (sub-) modules considered the transport of the SBS from its production site to the AM production site. There is not a unique supplier of SBS for the European/Italian market, but different potential countries of origin: Spain, USA and China. This transport sub-module was modelled by ship transport followed by train and/or by truck transport. Four routes were considered: (i) from China (Shanghai) to Rotterdam harbor by ship, from Rotterdam to Milan by train and then from Milan to the refining site by truck; (ii) from China (Shanghai) to Genoa harbor and (iii) from USA to Genoa harbor by ship, then from Genoa to the refining site by truck; (iv) from Madrid directly to the refining site by truck. This transport sub-module also included the transport of the big bags to the incineration site located at 75 km from the refining plant.
- Transport of innovative modifier. This transport module includes the supply of raw materials (hard plastics, PVB and graphene), the transport of end-of-life plastics and the product delivery to the AM production. The municipal collecting sites for end-of-life plastics (plastics side streams) are in the same province of the production plant, with an average distance: 23.4 km; the process selected for this transport was transport, freight, lorry 3.5–7.5 metric ton, EURO6. The glass recycling plants from where PVB is sources are located at about 60 km from the production plant (transport, freight, lorry 3.5–7.5 metric ton, EURO 4). The origin of the big bags used as packaging for the modifier was set to be India: transport has been modelled accordingly. The system included also the transport of the plastics, metal and glass residues discarded in the production process. The graphene supply chain starts from the expanded graphite, which is transported from USA to the production site packed in paper and plastic bags. After its conversion form expanded graphite, the graphene is transported from the production site to the innovative modifier manufacturing plant and, at the end to the production site of AM which is located to 175 km distance.

2.2.10. Raw Materials Requirements

For the case studies investigated different raw materials requirements (including the recycled component) have been considered as input data to the proposed models. Tables 2 and 3 specify the inventoried amounts for each case study referred to the pavement layering design including details of the technical and structural parameters for each layer and for the three compared pavement solutions with standard AM, SBS-modified AM and AM with innovative modifier. The tables provide also the duration of each layer according to the maintenance program (depending on the assessed and estimated useful life), which determines the frequency of the single layer maintenance. Table 4 shows the rate of RAP considered for the third case study.

2.3. Impact Assessment

The impact assessment computations were made by using different computational tools:

- OpenLCA 1.10 software by using Ecoinvent 3.6 database;
- Gabi 9.1 software and database Thinkstep service pack 39

The impact methodology applied is CML-IA baseline (2016 update).

The methodology considers the following impact categories with the relative units of measurement (reported in squared brackets):

1. Abiotic Depletion (ADP elements) [kg Sb eq.]
2. Abiotic Depletion (ADP fossil) [MJ]
3. Acidification Potential (AP) [kg SO_2 eq.]
4. Eutrophication Potential (EP) [kg Phosphate eq.]
5. Freshwater Aquatic Ecotoxicity Pot. (FAETP inf.) [kg DCB eq.]
6. Global Warming Potential (GWP 100 years) [kg CO_2 eq.]
7. Human Toxicity Potential (HTP inf.) [kg DCB eq.]
8. Marine Aquatic Ecotoxicity Pot. (MAETP inf.) [kg DCB eq.]
9. Ozone Layer Depletion Potential (ODP, steady state) [kg R11 eq.]
10. Photochemical Ozone Creation Potential (POCP) [kg Ethene eq.]
11. Terrestrial Ecotoxicity Potential (TETP inf.) [kg DCB eq.]

3. Results and Discussion

LCA results referring to the three case studies for the three different pavement solutions are presented and organized as follows:

(1) environmental impacts assessment referred to the first case study (fixed layer thickness for three different AMs); the analyses were operated with both Gabi and OpenLCA software;
(2) environmental impacts assessment referred to the second case study (different layer thickness for three different AM solutions); the analyses were operated with both Gabi and OpenLCA software;
(3) environmental impacts assessment referred to the third case study (different rates of RAP introduced in the SAM and MAM production process determining different circularity indexes); the comparative impact assessments were operated with OpenLCA software only.

3.1. First Case Study: Environmental Impacts Considering the Scenario with Fixed Layer Thickness

The first design case assumes the same pavement stratigraphy. Figure 2 provides a comparative representation of the three AM solutions considered; the correspondent absolute values obtained for the different impact categories are shown in Table 7.

The enhanced impacts reduction provided by the innovative modifier MAM case is proven for all impact categories by reducing on average 70% the SAM impacts and by reducing more than 50% the impacts of the SBS-MAM case.

The life cycle impact assessment results obtained with Gabi and Open-LCA indicate a good reproducibility for the compared cases. Indeed, although providing slightly different values in the comparisons of the MAM (SBS) and MAM (innovative modifier) with respect to the SAM solution, OpenLCA results show a quite reproduced modulation for the eleven impact categories considered. As the matter of fact, only minor fluctuations in the order of few percent may be detected between the Gabi and OpenLCA computations for the first case study. Nevertheless, Thinkstep and Ecoinvent databases show differences in the transport modelling and related impacts per unit of transport. The incineration process needed also a dedicated modeling based on literature and incineration plant data for the OpenLCA computations with Ecoinvent, as it is not an inbuilt process like in the Thinkstep database. The transport and incineration process modeling differences turned into relative impacts differences in the OpenLCA results shown in a mild mitigation of the climate change (GWP)- and photochemical ozone creation (POCP)- impact categories as well as a slightly increased ozone depletion potential (ODP) and toxicological related categories with respect to Gabi results.

It is important to highlight that these minor fluctuations are obtained only when comparing solutions within the same computational tool results. On the contrary a direct comparison between results obtained with the two computational tools for the same solution or comparisons of results referred to different solutions across computation tools would not provide the same comparable outcomes. Therefore, comparisons of results obtained by the same computational tool are recommended.

Given the accordance of the comparative results among the different solutions within Gabi and OpenLCA computations, the enhanced improvement of the environmental performance of the innovative modifier MAM compared to the SBS-MAM and with respect to the reference unmodified SAM performance can be confirmed.

Emissions of CO_2-eq and impacts related to energy carriers obtained from the analysis are in line with the literature belonging to the same context and providing results which refer to a similar functional unit (assessment of an Italian extra-urban road pavements) [33,39].

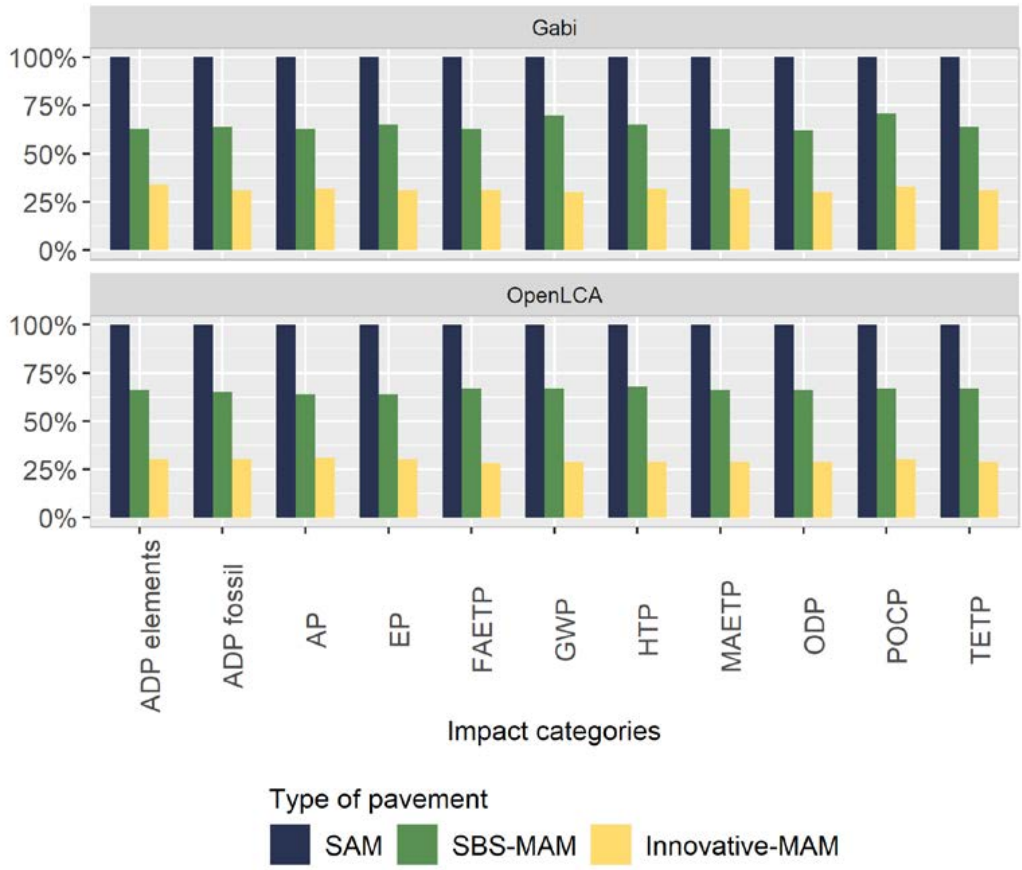

Figure 2. First case study with correspondent layers with fixed thickness. Comparative environmental impact assessment results for all the impact categories for the three cases SAM, SBS-MAM and Innovative MAM considered obtained by the computation with the Gabi (**upper** panel) and Open LCA (**lower** panel) software.

Table 7. Impact results for the three cases, Innovative MAM, MAM (SBS), SAM, with fixed thickness.

	Innovative MAM		**MAM (SBS)**		**SAM**	
Impact Category	**GaBi**	**OpenLca**	**GaBi**	**OpenLca**	**GaBi**	**OpenLca**
ADP elements [kg Sb eq.]	4.85×10^{-2}	1.92	9.02×10^{-2}	4.28	1.42×10^{-1}	6.48
ADP fossil [MJ]	1.07×10^{7}	1.19×10^{7}	2.21×10^{7}	2.63×10^{7}	3.45×10^{7}	4.03×10^{7}
AP [kg SO_2 eq.]	5.97×10^{2}	7.12×10^{3}	1.20×10^{3}	1.49×10^{4}	1.89×10^{3}	2.33×10^{4}
EP [kg Phosphate eq.]	8.25×10^{1}	1.62×10^{3}	1.71×10^{2}	3.40×10^{3}	2.63×10^{2}	5.32×10^{3}
FAETP [kg DCB eq.]	3.11×10^{3}	8.33×10^{4}	6.35×10^{3}	1.95×10^{5}	1.01×10^{4}	2.93×10^{5}
GWP [kg CO_2 eq.]	2.39×10^{5}	5.21×10^{5}	5.57×10^{5}	1.22×10^{6}	8.00×10^{5}	1.81×10^{6}
HTTP [kg DCB eq.]	2.35×10^{4}	1.80×10^{5}	4.81×10^{4}	4.21×10^{5}	7.35×10^{4}	6.21×10^{5}
MAETP [kg DCB eq.]	1.39×10^{7}	2.31×10^{8}	2.75×10^{7}	5.20×10^{8}	4.35×10^{7}	7.89×10^{8}
ODP [kg R11 eq.]	2.76×10^{-2}	1.24×10^{-1}	5.76×10^{-2}	2.77×10^{-1}	9.22×10^{-2}	4.22×10^{-1}
POCP [kg Ethene eq.]	2.75×10	1.28×10^{2}	5.91×10	2.88×10^{2}	8.28×10	4.32×10^{2}
TETP [kg DCB eq.]	2.13×10^{2}	6.72×10^{2}	4.34×10^{2}	1.53×10^{3}	6.83×10^{2}	2.29×10^{3}

3.1.1. Main Contributions to Impacts for SAM and MAM with Innovative Modifier in OpenLCA

In order to identify the major sources of impacts, the standard and modified asphalt mixture functional system have been considered. The reference SAM case impact assessment results are reported in Figure 3, referring to the OpenLCA computations.

From this comparative analysis emerges that the different processes have different impact weights according to the impact category analyzed. Nevertheless, the three most contributing processes may be identified in extraction and processing of raw materials, transport, and production of the AM. This is in accordance with previous studies [33,40,41].

These extraction and processing of raw materials with the transport process represent the dominant impacting processes in all categories accounting for 80–90% of the total impacts in each category, except for Acidification and Eutrophication potentials, for which the production process accounts alone for about 70% of total impacts. This is particularly interesting when focusing on the climate change impact category (GWP) in which, regardless the relevant energy demand and emissions of the production process, this represents less than 20% of the equivalent carbon dioxide emitted for the whole AM life cycle.

The remaining processes: paving, incineration, maintenance, and demolition for RAP have negligible impact in the whole system life cycle balance.

Life cycle of innovative modifier, employing the recycling of hard plastics with the introduction of graphene show a similar assessment, as reported in Figure 1.

Comparing SAM and MAM diagrams, it is possible to see that the two AM life cycles show a quite similar environmental profile for all the impact categories. The analysis outcomes highlight that the processes connected to the modifier production, including the transport due to the hard plastic collection, provide a negligible relative impact, although enhancing the AM product functionality in terms of durability and structural (stiffness) properties. This result suggests that the major impact sources are strictly connected to the amount of material employed to provide the functional unit.

3.2. Second Case Study: Environmental Impacts Considering the Scenario with Variable Layer Thickness

This case allows us to design different corresponding layers thickness but requires the same material technical performance in terms of stiffness and durability, regardless the material nature. This implies the same service life for all the considered pavement solutions and thus the same maintenance frequency for all corresponding layers. Given the different structural performance of the various pavement types, different thickness, and thus different amounts of materials, are required for the different solutions. This shifts the focus of the compared solutions exclusively on comparing the amount of material used (and the related processes to obtain it), in order to attain the defined comparable functionality among the systems involving the specified time frame (reference period) for the functional system under comparison.

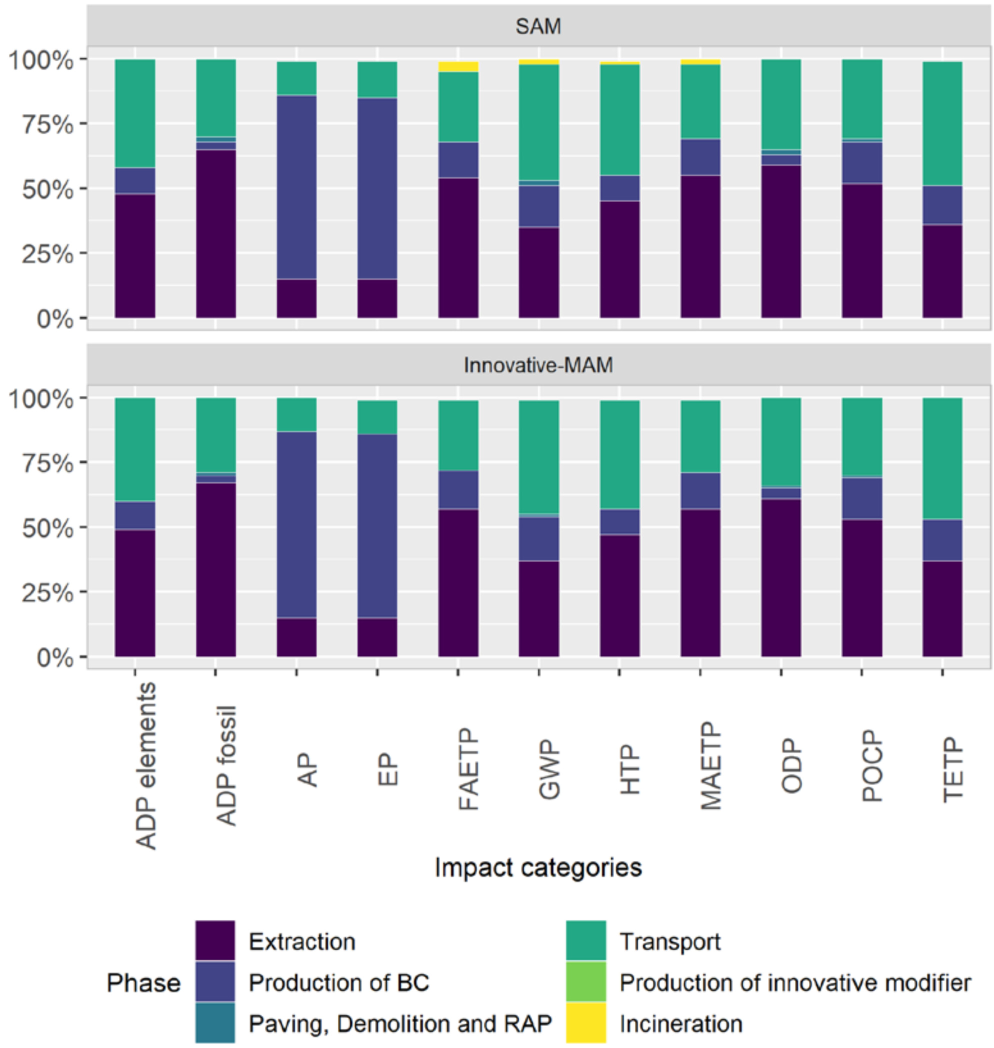

Figure 3. Analysis of the major contributions to environmental impacts provided by different processes referred to the conventional pavement (SAM) life cycle (**upper** panel) and the pavement with the innovative modifier (MAM) life cycle (**lower** panel).

The results of this analysis carried out with the Gabi and OpenLCA computational tools and related database resources are provided in Figure 4 and Table 8.

The results obtained with GaBi confirm the relative improvement of the SBS-MAM solution with respect to the SAM one although the environmental reductions are limited to the 10% range, whereas the innovative modifier MAM system shows a stable and well discriminated reduced environmental impact, with an average decrease of 20% on all the impact categories. The climate change impact category shows an enhanced reduction by 23% with respect to the SAM solution.

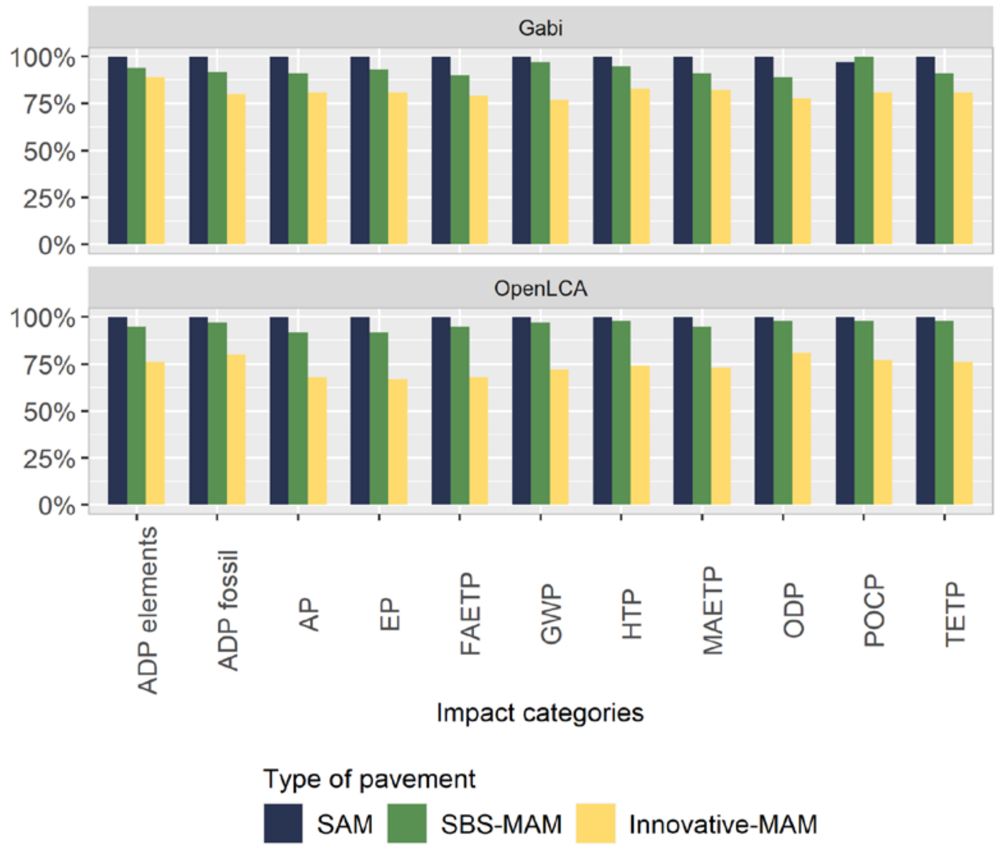

Figure 4. Second case study related to pavements with corresponding layers variable thickness and same stiffness. Comparative environmental impact assessment results for all the impact categories for the three cases SAM, SBS-MAM and innovative -MAM considered obtained by the computation with the GaBi and OpenLCA software.

Table 8. Impact results of three cases, innovative MAM, MAM (SBS), SAM, with variable thickness.

	Innovative MAM		**MAM (SBS)**		**SAM**	
Impact Categories	**GaBi**	**OpenLca**	**GaBi**	**OpenLca**	**GaBi**	**OpenLca**
ADP elements [kg Sb eq.]	1.25×10^{-1}	4.94	1.32×10^{-1}	6.17	1.41×10^{-1}	6.48
ADP fossil [MJ]	2.75×10^{7}	3.23×10^{7}	3.16×10^{7}	3.90×10^{7}	3.44×10^{7}	4.03×10^{7}
AP [kg SO_2 eq.]	1.53×10^{3}	1.58×10^{4}	1.72×10^{3}	2.15×10^{4}	1.88×10^{3}	2.33×10^{4}
EP [kg Phosphate eq.]	2.12×10^{2}	3.59×10^{3}	2.45×10^{2}	4.90×10^{3}	2.63×10^{2}	5.33×10^{3}
FAETP [kg DCB eq.]	7.92×10^{3}	2.12×10^{5}	9.05×10^{3}	2.96×10^{5}	1.01×10^{4}	3.13×10^{5}
GWP [kg CO_2 eq.]	6.16×10^{5}	1.35×10^{6}	7.83×10^{5}	1.81×10^{6}	8.04×10^{5}	1.87×10^{6}
HTTP [kg DCB eq.]	6.05×10^{4}	4.70×10^{5}	6.95×10^{4}	6.20×10^{5}	7.32×10^{4}	6.35×10^{5}
MAETP [kg DCB eq.]	3.56×10^{7}	5.90×10^{8}	3.95×10^{7}	7.68×10^{8}	4.34×10^{7}	8.12×10^{8}
ODP [kg R11 eq.]	7.17×10^{-2}	3.41×10^{-1}	8.24×10^{-2}	4.15×10^{-1}	9.22×10^{-2}	4.23×10^{-1}
POCP [kg Ethene eq.]	6.88×10	3.32×10^{2}	8.50×10	4.21×10^{2}	8.22×10	4.32×10^{2}
TETP [kg DCB eq.]	5.49×10^{2}	1.74×10^{3}	6.21×10^{2}	2.23×10^{3}	6.82×10^{2}	2.29×10^{3}

With OpenLCA tool, the reduction in the environmental impacts for the innovative system are more evident, although modulated for the different impact categories, with minimum reduction of 20% seen for the ADP and ODP impact categories. The acidification

(AP), eutrophication (EP) and fresh aquatic eco-toxicity potential (FAETP) are assessed well above the 30% impact reduction with a GWP reduction of 28%.

In this scenario, greater differences are shown when different computational tools are used. The major discrepancies, as reported above, are due to the incineration process modelling which reflects on the GWP, POCP and the human toxicity- and ecotoxicity-potentials. Although the amplified modulations between the two computational cases with Gabi and OpenLCA, the impact ranking among the different solutions considered is confirmed, proving the enhanced environmental performance of the new solution with innovative modifier with respect to the other modified SBS-MAM and unmodified SAM.

3.3. Third Case Study: SAM and Innovative Modifier MAM with Different RAP Rates

The analysis of main contributions to impacts carried out in Section 3.1.1 showed that the extraction phase is one of the dominant factors affecting the overall environmental burden, while the process to collect RAP can be considered negligible within the overall system balance along the pavement life cycle. Based on this evidence, the analysis has been further focused on the relevance of the circularity index related to the rate of RAP that may be reintroduced into the AM life cycle expected to benefit all functional systems considered. Specifically, the solutions considered in the third case study involve the life cycle of pavements obtained with SAM and MAM with innovative modifier considering different RAP rates, which fix increased and decreased circularity indexes on the functional systems, with respect to reference scenario.

For this purpose, three different scenarios were considered, one which minimizes the amount of RAP, one which replicates the reference values set in the previous analyzed cases and the final one which enhances the reference rates for each layer, as show in Table 4.

The increase in recycled RAP rate implies the increase in process temperature at the AM mixing tower. This was considered in the computations based on the indications of the AM manufacturers and on the calorific power of the AM also found in the literature [42]. No technological change in the production line was considered in the study as a function of the recycled RAP rate. All other processing parameters and inventoried technical data used for AM processing were kept the same as for the reference case.

In the analysis the process to obtain the corresponding RAP rate through road layers demolition and transport of RAP was included in the computations.

The results reported below are referred to innovative modifier and SAM (Figure 5) and compare the impacts of the scenarios where different amounts of RAP are added to the asphalt mixture.

The results obtained for both cases (SAM and innovative modifier) show a strong agreement in the environmental profiles obtained by the modulation of the rate of the recycled RAP. As expected, no relevant differences among scenarios may be detected comparing the two solutions, as the principal factor determining the result is the reduced amount of virgin materials needed to obtain the same FU.

What is quite significant here is the environmental impact reductions that can be obtained by enhancing the amount of RAP to produce AM. The improvement in environmental burden reduction is proportional to the rate of virgin materials reductions, with different values according to the impact categories. This expresses the sensitivity of each impact category to the combined effect achieved by the reduction in virgin materials and the proportional decrease in other operational steps, such as the demolition process and the transport of RAP, which definitely affect each impact category in a different way. These results confirmed what has been recently reported by Oreto and colleagues [43], who estimated a significant reduction in the environmental burden by LCA when recycled aggregates and polymers are used in paving.

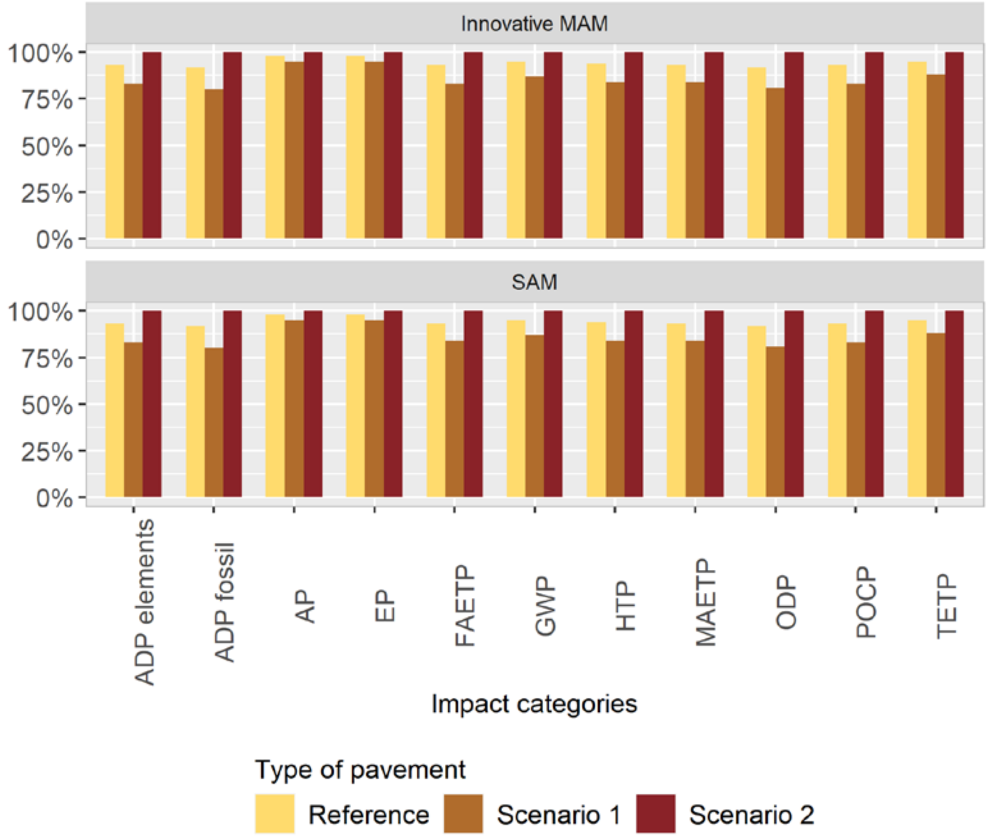

Figure 5. Environmental impact categories referred to the MAM with innovative modifier FU with different amount of recycled RAP rates as reported in the defined scenarios (scenario 1: providing +50% RAP with respect to reference scenario; scenario 2: providing −50% RAP with respect to reference scenario).

Comparing scenario 1 and scenario 2 we can see that, as expected, relevant results in environmental performance may be obtained by the reduction in virgin non-renewable raw materials use obtaining a drop in the in ADP-En of 20%, and >15% reduction in the Human toxicology and eco-toxicology potentials with GHG emissions related potentials (GWP, POCP and ODP) reductions by 16%.

4. Conclusions

A comparative impact assessment through the whole life cycle of three asphalt mixtures was performed through LCA methodology within a "from cradle to cradle" perspective. The systems assessed were the following: standard (unmodified)-, SBS modified- and innovative modified (obtained by recycled hard plastics and graphene) asphalt mixtures (referred as "AM" in the text).

Three case studies have been analyzed: (1) constant dimensional specifications for the pavement, implying comparable amount of material used for road paving and thus different service lifetime frames; (2) dedicated sizing of the layer thickness to obtain equal structural performance and thus life span for all systems; (3) different shares of RAP in the production of the AM.

The AM modified with the innovative modifier showed the best environmental performance thanks to the enhanced properties provided by the new modifier, whereas the standard AM had the largest impacts. In the first case study, the AM with the innovative modifier achieved an average of 70% reduction in all impact categories with respect to the standard AM and an average 50% impact reduction in all impact categories with respect to the SBS modified AM.

In the second case study innovative modified AM provided 30% less impact on average with respect to standard unmodified AM and more than 25% impact average reductions with respect to the SBS-modified AM. Two different computational tools were used with comparable results: Gabi and OpenLCA software, with the Thinkstep and Ecoinvent 3.6 databases, respectively.

The third case study showed that an environmental impact average reduction exceeding 15% can be obtained by increasing the RAP percentage from 20% to 30% in the wearing course, and from 30% to 50% in the binder- and base layers. In this case, the environmental benefits resulted independent from the modifier used in RAPs.

The AM innovative modifier considered in this work proved to have beneficial and synergistic effects for the environment. First of all the new modifier may strongly enhance the structural performance of asphalt pavements and allow for an extension of the pavement layers durability and thus for fewer material requirements over a specified service lifetime frame (first case study). Moreover, according to the second case study perspective, the new modifier may allow for thickness reduction in each pavement layer by decreasing the raw material requirements, while keeping the same structural performances compared to the standard and SBS-modified AM. Second, the use of innovative modifiers embedding recycled polymers also creates a positive effect on the sustainable management of end-of life non-recyclable hard plastic, offering an adequate opportunity to plastic waste prevention as well as a valuable option to hard plastic incineration.

This result confirms what was found in previous LCA studies: (i) the use of recycled polymers in asphalt production as AM modifiers, resulted in a high environmental advantage in respect to the use of virgin materials; (ii) LCA proved to be a powerful tool to perform these analyses [40,42,44,45].

Finally, we can additionally conclude that to achieve strong environmental impact reductions in asphalt pavements a specific focus has to be put on raw materials performance and origin. Indeed, models with enhanced circularity index, obtained by increased rates of reclaimed asphalt used in AM formulations can significantly contribute to the road pavement sustainability due to proportional reduction in non-renewable raw materials of fossil origin use as well as to the reduction in RAP over-storage or landfilling caused by roads standard maintenance.

Nevertheless, a better understanding of the environmental impacts deriving from the use of complex "new" materials should be carefully addressed in the next future, as well as the potential effect of the innovative modifier on the long-term functionality of the pavement or on the performance of RAP containing it [28,46,47].

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/pr10102151/s1. Table S1: Synoptics of the different compared cases with standard thickness, Table S2: Synoptics of the different compared cases with reduced thickness, Table S3: Frequency of maintenance for each layer for the first case (standard thickness), Table S4: Frequency of maintenance for each layer for the second case (reduced thickness), Table S5: composition of modifier, Table S6: composition of graphene.

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