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Circular economy strategy of reusing olive mill wastewater in the ceramic industry: How the plant location can benefit environmental and economic performance

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ABSTRACT

The olive mill wastewater (OMWW) is one of the highly pollutant residue in olive oil producing countries around the Mediterranean Sea, due to its large organic load and the presence of a variety range of contaminants. The valorization and the recycling of this by-product can represent a successful strategy for the implementation of circular economy models in the agri-food industry. For this purpose, the incorporation of OMWW in the brickmaking process represents a promising solution, which not only can avoid the impacts due to its disposal but also reduce the heat required during the ceramic production process, resulting in lower greenhouse gas emissions and economic benefits. However, many factors should be considered, including the transport activity of the OMWW from the mill to the factory that can affect both the environmental and economic benefits. The present study aims to assess the performance of an open-loop circular economy system based on the reuse of the OMWW in the fired clay brick production, in terms of technical feasibility, environmental and economic sustainability. To evaluate environmental impacts, a comparative Life Cycle Assessment is performed between a conventional production and a system in which OMWW is directly integrated in the brick-making process, considering scenarios with different transport distances. Furthermore, an economic sensitivity analysis was implemented, taking into account energy savings and the additional costs due to transport. The results showed that the overall Global Warming Potential (GWP) decreases up to 3.1% for OMWW-based bricks with respect to conventional ones, as well as the Abiotic Depletion of fossil fuels is reduced by 4.3%. On the other hand, no significant variations were observed for the toxicity impact category, that ranges from -1.1% to 0.7%. Furthermore, the water consumption increases for OMWW-based brick production up to 7.8%. Finally, in terms of GWP, it has been found that to make the benefits persist, the oil mill should be placed in a distance of less than 150 km from the brick factory, indicating a more restrictive constraint than the economic one, corresponding to a distance of 207 km.

1. Introduction

The olive oil industry is one of the most important food industries in many Mediterranean countries (El Gnaoui et al., 2020; Salomone and Ioppolo, 2012). In 2018, the total of productions from Spain, Italy, France, Morocco, and Greece achieved 1.8 million tons of olive oil (Kashiwagi et al., 2020; "The International Olive Council, 2021), resulting steadily increasing (Caputo et al., 2013). During the last two decades, the manufacture of olive oil has been undergone a profound

change in equipment and processes used for separating oil from remaining parts (Borja et al., 2006).

Modern extraction processes of virgin olive oil consist in the defoliation of olives, washing, crushing, malaxation and the final centrifugation phase (Souilem et al., 2017). The two-phase and three-phase methods differ for the way in which the centrifugation process is conducted. In the three-phase systems a large amount of hot water is added to enhance the extraction of olive oil from the olive pulp, producing three distinct fractions, which are a solid fraction (pomace) and two

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liquid fractions (oil and aqueous fraction, wastewater) (Vlyssides et al., 2004). On the other hand, in the two-phase systems, the use of a more effective centrifugation technology allows a lower consumption of the total water (only washing water). At the end of the process, the system produces two different fractions, that are a liquid phase (oil) and a semisolid phase (wet pomace).

Today, the 'two-phase system' is an improved extraction method for olive oil (Khdair and Abu-Rumman, 2020) and represents the current 'ecological' process compared to the traditional 'three-phase system', consuming less water and energy, and producing a olive mill wastewater (OMWW) with a lower pollutant load (Azbar et al., 2004). This system is broadly used in Spain with the 98% of the total oil mills, while its use is constantly increasing in other countries of Mediterranean, such as Portugal, Greece and Italy (Sygouni et al., 2019). In particular, in Italy it has gradually caught on because of the difficulties initially raised by residues treatments facilities, which had to manage residues with high level of moisture which largely differ from those derived from conventional three-phases systems. Today, according to Agronotizie (2020), approximately 80% of the total oil mills in Italy have adopted the two-phases process system.

In the agri-food industry, a successful strategy for the implementation of the circular economy is represented by the valorization of agrifood by-products (Cozma et al., 2020; Gebremikael et al., 2020).

The aim of this study is twofold: first to conduct a comparative cradle-to-gate Life Cycle Assessment (LCA) (ISO 14040:2006; ISO 14044:2006/Amd 1:2017) in order to compare environmental impacts of conventional and OMWW-based ceramic bricks manufacturing; and second to evaluate the differences in terms of costs between the two production processes linked to the different energy used and the transport cost of OMWW. In particular, the LCA has been conducted taking into account only OMWW obtained from two-phase mills, representing not only the current ecological solution but also the reference system for the future production of oil in Europe.

To the best of authors' knowledge, this is the first study providing insights on potential beneficial effects of the use of olive-oil by-product in ceramic industry in a life cycle and open-loop circular economy perspective, taking into account the economic implications, in addition to the environmental benefits.

The authors look at how awareness of a successful valorization of olive oil by-products strategy can enables incisive circular economy business, especially in countries like Italy that are leader in olive oil production. Furthermore, among major producing countries of olive oil in the Mediterranean basin, Italy also absorbs a wide portion of bricks and roof tiles production (BAT, 2007). Indeed, together with Germany, France, and UK, Italy represents the 70% of the total European production (CERAME-UNIE, 2019; Egenhofer et al., 2014) and they have shown a gain of 8.5%, corresponding to approximately ϵ 184 million, only in 2018. As a consequence, the LCA and economics analysis has been modeled assuming the location of the bricks manufacturing site to be in Italy and therefore considering the costs of natural gas and transport for the Italian market.

2. Literature background

The olive oil production generates a significant number of by-products, which includes aqueous solid residues and OMWW. Several methods and processes have been proposed for the valorization of OMWW, the most recent are: the solar distillation (Sklavos et al., 2015), the dark fermentation (Ghimire et al., 2015), the solid biofuels production (Jeguirim et al., 2012; Kraiem et al., 2014) and the fired clay brick production (de la Casa et al., 2009). In particular, the valorization of the OMWW by its incorporation in ceramic clay bricks has been previously studied in literature for both three-phase (Mekki et al., 2006, 2008) and the two-phase olive oil mills (de la Casa et al., 2009), where the authors found out how the introduction of OMWW is able to significantly reduce heating requirements (de la Casa et al., 2009) with

the same or improved technological properties (de la Casa et al., 2009; Mekki et al., 2008). Table 1 summarizes some of the methods and processes currently used for the valorization of OMWW, as well as main advantages and disadvantages.

On the other hand, the LCA has been widely applied in literature to investigate environmental impacts of ceramic industry (e.g., Ibáñez-Forés et al., 2011; Ye et al., 2018). Koroneos and Dompros (2007) adopted the LCA for providing a quantitative basis to assess potential improvements in environmental performance of brick manufacturing in Greece. De Souza et al. (2016) assessed environmental impacts of three different options for exterior walls that include ceramic bricks, while Lozano-Miralles et al. (2018) conducted an LCA study for comparing bricks made with conventional and innovative raw materials mixture, the latter includes organic waste. Similarly, Joglekar et al. (2018) assessed environmental performance of waste-based ceramic bricks through the LCA methodology. The possibility to recycle concrete and ceramic for substituting a portion of the clay soil for the manufacturing of unfired bricks has been debated by Seco et al., in 2018. The use of recycled materials as the alternative to the traditional ones for the production of ceramic bricks were also studied by other authors (e.g., Marcelino-Sadaba et al., 2017; Mohajerani et al., 2018).

3. Materials and methods

3.1. Life Cycle Assessment: Goal and scope definition

This study aims to achieve a benchmarking of manufacturing processes to obtaining conventional and OMWW-based ceramic bricks, applying the LCA methodology (ISO 14040:2006; ISO 14044:2006/Amd 1:2017). With this regard, this study is a comparative analysis that involves an opened-loop recycling system, in which the OMWW derived from the olive oil extraction process is used instead of fresh water for brick manufacturing. The OMWW can significantly contribute to the heat requirements during the firing stage, reducing the heating up to 7.3% (de la Casa et al., 2009). In fact, the organic content of the OMWW allows to obtain a higher heating value of 73 kJ per kg of clay, which can be saved during the fired stage. As a consequence, end products will have different characteristics. However, according to de la Casa et al. (2009) and Mekki et al. (2008), the technological properties either do not change or improve. A summarization and comparison of main properties are summarized in Table 2. Furthermore, according to Mekki et al. (2006), it is possible to expand the described brick making process from laboratory to factory scale. Basing on results obtained in these studies and previous authors' works (Silvestri et al., 2020, 2021), it has been possible to setup and assess a whole manufacturing chain that involves the use of specific OMWW streams obtainable from the two-phase oil extraction, as well as the energetic advantages derived from the firing stage (Paragraph 3.2.1). One of the aims of this study is to evaluate the trade-off that may occur between the environmental benefits due to the reduction of thermal energy required in the firing stage are achieved and the supplementary environmental load due to the OMWW transportation from the oil mill to the brick factory. Furthermore, the environmental gain achieved by the re-use of OMWW has also been assessed. In particular, for the water footprint was assessed for recognizing the amount of water consumed manufacturing processes.

The results of this LCA are intended to be used to encourage the recycling of OMWW, developing an efficient circular economy model in the agri-food sector and a greener industrial production of bricks.

The proposed methodology for the comparative environmental assessment includes several parts. As first, recent scientific literature, Environmental Product Declaration (EPD) items (ISO 14025:2006) and the European Best Available Techniques (BAT) was investigated for modeling the semi-wet preparation process of ceramic bricks. Finally, the composition of conventional and OMWW-based ceramic bricks and all the production processes involved were modeled according to literature. Finally, manufacturing scenarios were designed and compared

Table 1
Current techniques and products for the valorization of the olive mill wastewater (OMWW). Source: Souilem et al. (2017) and Authors' elaboration.

Technique	Product	Advantages	Disadvantages	Reference
Continuous-flow adsorption and	Obtainment of	Crucial step toward the development of an	The desorption step should be	Frascari et al.
desorption process	phenolic compounds	industrial-scale process	improved	(2016)
Extraction using hydrophobic ionic liquids	Obtainment of tyrosol	Less expensive process	The purication and drying of the recovered tyrosol required additional studies	Larriba et al. (2016)
Integrated process including fermentation, spray drying, and encapsulation technologies	Olive pasteOlive powderEncapsulated phenols	The income from byproduct exploitation could pay for the cost of depollution	Not applicable in small olive mills	Goula and Lazarides (2015)
Solar distillation	Dried OMWWObtainment of phenolic compounds	A low-cost and eco-friendy process for OMWW treatment	Process should be further optimed	Sklavos et al. (2015)
Dark fermentation	Biohydrogen	The method is applicable to different agro- industrial waste	OMWW is less suited for biohydrogen production if compared to typical agro-industrial waste	Ghimire et al. (2015)
Enzymatic hydrolysis and microfiltration-ultrafiltration	Obtainment of natural hydroxytyrosol	The method is applicable to different water streams from olive mills	Processes adapted to pilot-scale applications	Hamza and Sayadi (2015)
Impregnation of OMWW by on dry biomasses	Solid biofuel	Energy content and reactivity of OMWW impregnated samples are increased	An increase in particles emissions is observed	Kraiem et al. (2014)
Dyebath for dyeing wool	Natural dyes for textile industry	Economic and eco-friendly process Considerable color fastness properties for acrylic fibre	-	Haddar et al. (2014)
Fired clay brick production	Materials for ceramic building	Significantly reduce heating requirements OMWW-based products are comparable to traditional ones in terms of extrusion performance and technological properties	-	de la Casa et al. (2009)
Brick-making process	Building bricks	The samples containing OMWW are comparable in forming and extrusion performance to a control product that used fresh water Processes can be expanded to a full-scale factory	-	Mekki et al. (2008, 2006)

Table 2Comparation of average properties of bricks made with fresh water and olive mill wastewater (OMWW).

	Firing linear shrinkage (%)	Mass loss on firing (%)	Water absorption (%)	Fired bulk density (kg/m³)	Fired-bending strength (N/mm²)	Tensile strength (MPa)	Plasticity index after 3 days
Source	de la Casa et al. (2009)	de la Casa et al. (2009)	de la Casa et al. (2009)	de la Casa et al. (2009)	de la Casa et al. (2009)	Mekki et al. (2008)	Mekki et al. (2008)
Conventional bricks	1.3	7.9	7.8	1988	18.7	10.2	0.84
OMWW-based bricks	0.55	9.08	10.5	1880	17.6	10.6	0.9

through the Life Cycle Inventory (LCI) analysis and the Life Cycle Impact Assessment (LCIA) in a cradle-to-gate perspective. Particular importance was given to saving in terms of consumption of fresh water, analyzing the AWARE and identifying the hot spots for this impact category.

The LCA was carried out by using SimaPro® (PRé-Consultants, 2014) software and records available in Ecoinvent 3.5 (2018) database.

To perform the comparison, the following scenarios were designed:

Scenario 1: Conventional ceramic bricks, where bricks are produced using fresh water in a semi-wet preparation process.

Scenario 2: OMWW-based ceramic bricks, where the semi-wet preparation process of ceramic bricks includes only OMWW effluents derived from the two-phase olive oil extraction process. Furthermore, Scenario 2 was followed up by a sensitivity analysis in order to determine how variations in the distance traveled by trucks during the OMWW transportation determine different environmental impacts.

Results will let the authors to find the maximum distance that should exist between the oil mill and the factory for maintaining environmental benefits. For this purpose, the Global Warming Potential has been considered as the representative key indicator for comparing results.

Building bricks are part of the *bricks and roof tiles* ceramic sector and they include products such as clay blocks, facing bricks, klinker bricks

and lightweight bricks (BAT, 2007). The ceramic bricks considered in this LCA consist in engobed facing bricks (BAT, 2007; Silvestri et al., 2021), which are generally used to provide protection from external exposure of building walls and, when no plaster is applied, producing an attractive aspect. Both conventional and OMWW-based ceramic bricks are compared on the basis of the following *functional unit* (FU): "Complete production of 1 ton of engobed fired facing bricks through a semi-wet preparation process". This FU allows for a comparison between ceramic bricks made with water from different sources.

The manufacturing processes have been divided in two different subprocesses according to Scenario 1 and Scenario 2. This solution is generally adopted to avoid the allocation procedure (ISO 14044:2006/Amd 1:2017, 2006) and is achieved sub-dividing the process into minor units and, thus, solving the multifunctional unit process (Fig. 1). Fig. 1 shows the system boundary and the considered process subdivision. The system boundary includes foreground activities, which correspond to the gate-to-gate chain within the bricks factory, and background activities, consisting in extraction and transportation of raw materials and in electricity and thermal energy supply, as well as in the production of OMWW within the oil mill. Foreground and background activities are presented in Paragraph 3.2.

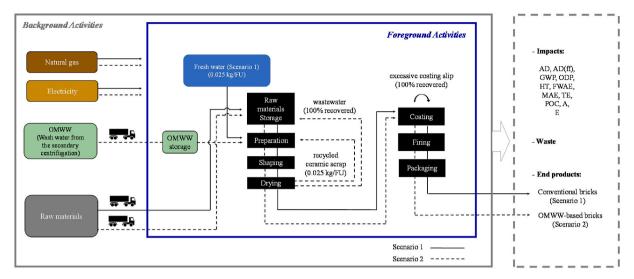


Fig. 1. System boundary considered for Life Cycle Assessment of the production of conventional (Scenario 1) and olive mill wastewater (OMWW)-based bricks (Scenario 2).

3.2. Life cycle inventory analysis

3.2.1. Manufacture of ceramic bricks

In this study, the manufacturing stages and energy inputs required for producing ceramic bricks have been modeled according to on BATs (2007) and recent literature (de la Casa et al., 2009; Silvestri et al., 2021). Conventional and OMWW-based bricks share the same manufacturing activities, differing only for the source of the water used in the brick body. The main steps are shown in Fig. 1 and include the storage and mixing of raw materials, shaping, drying, coating, firing, and packaging phase.

Raw materials required for the realization of the brick body include clay, sand, water, and calcium carbonate. On the other hand, for the coating, it is assumed the use of a mix made of white-burning clay, fluxing agents, water, and colouring oxides. Cut-off criteria were chosen basing on the "Option III" in Almeida et al. (2015) (p. 210), which exclude "inputs and outputs that represent less than 0.5% of the mass of the ceramic brick ready to be sold".

Raw materials are pretreated by grinding and milling operations and finally collected in feeders, sheds or silos at ambient conditions. The preparation process consists in the semi-wet mixing of raw materials. Proportioning of raw materials is realized through the use of large volume feeders that allow the bulk load and the proportion of different raw materials. The mixing water is incorporated to the batch at the end of the feeder up to approximately 20%, corresponding to 196.5 kg to produce 1 Ton of bricks. For the conventional bricks (Scenario 1), fresh tap water has been considered in the semi-wet mix, while OMWW-based bricks (Scenario 2) include water derived from the two-phase olive oil process. More in detail, in the two-phase process, the primary separation of the olive oil portion from the vegetable solid mass and vegetation water is obtained through the action of a horizontally mounted centrifuge. In a second step, the resultant olive oil is first washed for eliminating residual impurities and then separated from the wash water in a vertical centrifuge (Borja et al., 2006). Three separated water streams are identifiable (Raposo et al., 2003), that are: 1) the water used for the fruit cleaning in the initial stage; 2) the aqueous solid residues derived from the primary centrifugation, that includes the vegetation water; 3) the wash water derived from the secondary centrifugation mixed with the wash waters produced when the virgin olive oil has been purified (Borja et al., 2006). The latter water stream is the OMWW considered in this study, that is able to substitute the fresh water in the brick production (de la Casa et al., 2009). It represents the 25% of the total water effluents and its production is around 0.25 l/kg of olives processed (Alba et al.,

2001). Concerning the main physico-chemical features of the OMWW, the pH is 5.17, the acidity to pH = $8.3 \, (\text{mg CaCO}_3/\text{dm}^3)$ is 680, density is $1.012 \, \text{kg/dm}^3$, dry residue 2.52% and COD 7840 mg/dm³.

During the subsequent shaping phase, an extrusion press shapes the clay mixture body under an average pressure of 1.05 MPa. The resulting column is finally cut by means of a wire cutter that realizes the individual bricks. After the shaping process, bricks are dried for extracting the excessive moisture from soft bricks. This process is realized in tunnel dryers, where they need up to 72 h and an average temperature of around 82 °C. The energy source required for the drying stage is represented by excess heat available from the kiln during firing stage. The brick coating has been assumed required only for aesthetic purposes and not for other performance parameters, such as the improvement of the thermal insulation properties, as well as strength or water absorption. To this purpose, the coating is performed through the so-called 'engobing' technology, which involves the application of a thick slip to the dried brick (Silvestri et al., 2021). The engobing has been considered in a mixture compatible with other materials used for the bricks production, according to current BAT and literature (Hashmi, 2014; NPCS, 2007). The engobe layer has a paint-like consistency and its amount, in reference to the considered FU, has been computed according to the standard size of bricks (ASTM C216-19). In particular, a prismatic unit sizes 210 mm \times 100 mm x 65 mm has been selected. The engobe amount is made of 46% of fresh water and is applied in a rate of 1% in mass (Table 3). The firing stage is performed with tunnel kiln fed by natural gas, where bricks, arranged in kiln cars, move through to complete the firing phase. The last step consists in the packaging phase, which has been modeled according to the Ecoinvent record "Packing, clay product {RoW}| processing | APOS, S". Table 3 shows detailed amounts of raw materials assumed in this study, as well as electricity and thermal energy flows demanded during the manufacturing process for both Scenario 1 and Scenario 2. According to de la Casa et al. (2009), the thermal energy required for the Scenario 2 reduces in a range that varies from 2.4 to 7.3% in respect to conventional standards (Scenario 1). In this study, the saving percentage has been assumed to be 7.3%, in order to investigate the maximum benefits that is possible to achieve through this manufacturing option. The eco-profile of electricity is referred to the Italian electricity mix, according to the bricks factory location which is assumed to be in Italy.

Waste generation during the ceramic process has been modeled according to BAT (2007) and Hashmi (2014). Waste mainly consists in unfired ceramic scrap containing non-hazardous compounds, which is possible to be fully reintroduced within the manufacturing process. On

Table 3Extrapolation of the inventory provided by Life Cycle Inventory Analysis for main emissions and for raw materials and energy flows within production plant, where S1 and S2 stand for Scenario 1 and Scenario 2, respectively. Values are referred to the Functional Unit.

	Unit	S1	S2 - 50 km	S2 - 150 km	S2 – 500 km
Energy flows within the	ceramic	productio	nn		
Heat for Firing/Drying (Natural gas)	MJ	2300	2132.2	2132.2	2132.2
Electrical energy (DE mix)	MJ	270	270	270	270
Raw materials for brick	body				
Clay	kg	877.4	877.4	877.4	877.4
Sawdust	kg	22.6	22.6	22.6	22.6
Sand	kg	53.1	53.1	53.1	53.1
Calcium Carbonate	kg	29.5	29.5	29.5	29.5
Fresh water	kg	194.2	_	_	_
OMWW	kg		196.5	196.5	196.5
Engobe	Кδ		170.5	170.5	170.5
Clay	kg	5.81	5.81	5.81	5.81
Calcium Carbonate	kg	1.45	1.45	1.45	1.45
Silica Sand	kg	0.48	0.48	0.48	0.48
Feldspar		0.48	0.48	0.43	0.48
•	kg		0.97	0.97	0.97
Pigments (Copper Oxide)	kg	0.97	0.97	0.97	0.97
Water	L	8.09	8.09	8.09	8.09
Emissions to air					
Cadmium	mg	106.71	106.93	107.42	109.11
CO	g	365.93	372.35	394.43	471.54
CO_2	kg	309.55	300.84	310.75	345.34
Chlorine	mg	106.39	109.63	117.57	145.27
Fluorene	μg	30.31	28.14	28.15	28.15
Nickel	mg	644.69	646.66	651.00	666.13
NO_x	g	574.64	588.83	648.41	856.48
Partic. (<2.5 μm)	g	67.57	69.20	73.28	87.50
Partic. (>2.5 μ m, < 10 μ m)	g	47.78	49.08	52.15	62.85
SO ₂	g	646.55	642.75	657.63	709.59
VOC	g	6.03	6.03	6.03	6.03
Emissions to water	δ	0.03	0.03	0.03	0.03
NH ₃	mg	156.21	147.68	147.68	147.68
Arsenic	g	1.86	1.87	1.88	1.93
BOD ₅	g	307.91	322.54	353.39	461.12
Cadmium		1.09	1.09	1.10	1.12
Chromium	g mg	19.13	19.41	20.43	23.99
COD	kg	0.87	0.89	0.93	1.06
Pb	-	1.11	1.12	1.13	1.19
	g	11.36	11.43	11.67	12.50
Mercury Nickel	mg	5.42	5.45	5.55	5.91
	g	3.27	3.03	3.03	3.03
Nitrogen	mg	69.26		3.03 71.78	3.03 77.85
Phosphorus Sulfate	mg ka	8.85	70.04 8.89	71.78 8.97	77.85 9.27
	kg				
Suspended solids	g	405.66	394.60	399.80	417.94
TOC VOC	g mg	683.52 223.66	689.60 248.54	704.43 300.07	756.24 480.02

the other hand, fired scrap can be estimated as about 30 kg per ton of ceramic bricks and can be recovered and reintroduced in the same system in a percentage of 40%.

An additional amount of water, which should not be confused with water incorporated within the brick body, is represented by water employed for cleaning and reducing fugitive substances generation during the transportation of materials from the open-air storage within the plant interior. This water can be totally recovered after specific treatments. However, hazardous materials can be present within sludges generated during such wastewater treatments.

Waste that cannot be internally recovered, such as oils and other materials derived from maintenance operations (i.e., used filters or exhausted batteries) can be re-used in different industries or directly sent to disposal facilities. These data are generally unavailable and difficult to determine, thus, potential treatments of hazardous waste have not taken into account in this study.

3.2.2. Materials transportation

Materials transportation is required in the following stages:

- Raw materials transportation from quarries to brick factory (Scenario 1 and Scenario 2);
- Waste transportation from brick factory to disposal facility (Scenario 1 and Scenario 2);
- OMWW transportation from olive oil mill to brick factory (Scenario 2).

Raw materials (Table 3) extracted from quarries are transported assuming an average distance between the quarry and the brick factory of 200 km. Materials from quarries to the factory are transported by Euro 3 heavy trucks, that corresponds to Ecoinvent record "Transport, freight, lorry >32 metric ton, EURO3 {RER}| transport, freight, lorry >32 metric ton, EURO3 | APOS, S". On the other hand, coating materials and waste are both assumed to be transported with medium trucks "Transport, freight, lorry 3.5-7.5 metric ton, EURO3 {RER} | transport, freight, lorry > 32 metric ton, EURO3 | APOS, S" for a distance of 150 km. Waste is assumed to be transported from the bricks factory to the disposal facility for a distance of 50 km, by means of the same trucks considered for the raw materials transportation. Finally, for the Scenario 2, OMWW from olive oil mill to brick factory is assumed to be transported with tank trucks (15,000-30,000 L) for a distance that ranges according to different hypothetical locations of olive oil mills that may potentially represent an OMWW source. More in detail, a sensitivity analysis was performed for this distance to obtain a set of respective environmental impacts. The transportation distances are 50 km and 500 km. The set of environmental impacts represents the base for estimating the break-even point (identified for a distance of about 150 km), where environmental benefits due to the thermal energy savings for the manufacturing process of OMWW-based bricks will meet the environmental load due to the transportation. Finally, being the most impactful solution among those available on Ecoinvent, it ensures that there will be no underestimation of road emissions.

3.3. Life Cycle Impact Assessment

The Life Cycle Impact Assessment (LCIA) has been performed according to the current international standards ("ISO 14044:2006/Amd 1:2017" 2006, "UNI EN ISO 14040:2006" 2006). The AWARE, and CML-IA Baseline v.3.5 were used to evaluate environmental burdens into twelve impact categories. In particular, the AWARE, that is a midpoint method for evaluating the water footprint (Boulay et al., 2018; Willet et al., 2019), was selected in order to recognize the amount of water consumption in the manufacturing processes. This method has been already applied for the assessment of green water flows (Quinteiro et al., 2018), freshwater scarcity in production processes (Northey et al., 2018) and agri-food chains (Villanueva-Rey et al., 2018), and then it has been considered highly pertinent within the aim of this research. On the other hand, the CML-IA is a midpoint-oriented method developed by the Center of Environmental Science of Leiden University (CML) (Guinée et al., 2001a, 2001b, 2001b). This method is not only widely used in literature for investigating environmental performance of ceramic products and construction materials (Almeida et al., 2015; Traverso et al., 2010), but it can be also considered as the reference method for the LCA of bricks for hotspots identification, as well as for comparative purposes (Huarachi et al., 2020; Joglekar et al., 2018).

Moreover, CML is a required method for the evaluation of environmental impacts in construction processes (Durão et al., 2020; EN 15804:2012+A2:2019). The following impact categories were included: water footprint (W), Acidification potential (A), Depletion of abiotic resources - elements, ultimate reserves (AD), Depletion of abiotic resources - fossil fuels (AD(ff)), Global Warming Potential (GWP), Eutrophication (E), Fresh water aquatic ecotoxicity (FWAE), Human toxicity (HT), Marine aquatic ecotoxicity (MAE), Ozone layer depletion (ODP),

Photochemical oxidation (POC), Terrestrial ecotoxicity (TE).

3.4. Economic implications

The scenario considered in the LCA analysis, were analyzed in terms of costs differences. In detail the savings deriving from the lower thermal energy used and the burden due the transport of the OMWW were evaluated. Also, from an economic point of view, a sensitivity analysis was performed to estimate the economic break-even point distance, where thermal energy savings for the manufacturing process of OMWW-based bricks will meet the increase in cost due to the transportation.

As previously mentioned, the analysis was carried out with reference to the Italian productive context. Consequently, the cost of natural gas was valued equal to $0.54~\rm e/Sm^3$, according to ARERA (the Italian regulatory authority for energy). The cost of tank transport was assumed according to the most recent data provided by the Italian Ministry of Transport shown in Table 4, assuming to use tanks with a maximum capacity of 30,000 L.

4. Results and discussion

4.1. Life Cycle Impact Assessment results

Results obtained through the CML-IA method are summarized in Tables 3 and 5. For the S2 - 50 km scenario, the variations for the impact categories analyzed are always less than 5% in comparison to Scenario 1. In addition to a 3.1% decrease in the GWP, due to the lower thermal energy required in the process (BAT, 2013), a 4.9% decrease is observed for the same reason in the AD(ff). The reduction of emissions into the atmosphere also has positive effects on ODP, which decreased by 4.3%. The other impact categories considered in this study and in particular the impact category group of toxicity that includes (HT, FWAE, MAE, TE), show less significant variations, ranging from -1.1% to 0.7%. It should be noted that, even if in S1 no OMWW is employed, the level of BOD5 remains high. In fact, according to Vega-Coloma et al. (2011), most of the BOD5 loads can be associated with the fuel extraction and refining, especially from Diesel based plants. Therefore, such emissions can be attributed to the transport activity. Generally, raw materials used in the brick body, that mainly consist in clay, sawdust, and sand, produce a very low contribution to environmental impacts. On the other hand, according to Silvestri et al. (2021), small quantities of engobe are responsible for larger contribute, being some of the required raw materials both toxic and soluble.

For a distance of 150 km (S2 - 150 km), a break-even distance is obtained with Scenario 1 in terms of GWP. As mentioned, for this distance the impacts due to transport compensate for the saving of natural gas. Benefits still remain for the AD (2.8% decrease) and ODP (1.6% decrease) categories compared to the baseline scenario, while all the other impact categories worsen, ranging from 1.8% to 4.9%.

By increasing the distance to 500 km (S2 - 500 km) the negative effects of transport are considerably more significant than the advantages deriving from the use of the OMWW. All the impact categories analyzed increase, in comparison to the process with the use of fresh water. In detail, a 10.2% increase in GWP is observed and due to the high incidence of emissions resulting from transport, AD(ff) and ODP increase by 4.5% and 10.2% respectively. In this scenario, transport exhibits high

Table 4Cost of tank transport data from Italian Ministry of Transport.

Cost of transport (€/km)	Range of distances (km)
2.031	up to 150
1.880	151-250
1.714	251-350
1.533	351-500
1.355	over 500

Table 5Life Cycle Impact Assessment results for the considered Scenarios (CML-IA method), where S1 and S2 stand for Scenario 1 and Scenario 2, respectively. Values are referred to the Functional Unit.

Impact Category	Units	S1	S2 50 km	S2 150 km	S2 500 km
Abiotic depletion	kg Sb eq	2,05E-03	2,08E-03	2,13E-03	2,31E-03
Abiotic depletion (fossil fuels)	MJ	7,14E+03	6,79E+03	6,94E+03	7,45E+03
Global warming (GWP100a)	kg CO2 eq	3,40E+02	3,30E+02	3,40E+02	3,75E+02
Ozone layer depletion (ODP)	kg CFC- 11 eq	6,50E-05	6,22E-05	6,39E-05	7,01E-05
Human toxicity	kg 1,4- DB eq	1,91E+02	1,92E+02	1,95E+02	2,06E+02
Fresh water aquatic ecotox.	kg 1,4- DB eq	8,93E+01	8,98E+01	9,10E+01	9,53E+01
Marine aquatic ecotoxicity	kg 1,4- DB eq	2,57E+05	2,58E+05	2,62E+05	2,74E+05
Terrestrial ecotoxicity	kg 1,4- DB eq	4,40E-01	4,46E-01	4,61E-01	5,17E-01
Photochemical oxidation	kg C2H4 eq	5,90E-02	5,85E-02	6,04E-02	6,69E-02
Acidification	kg SO2 eq	1,12E+00	1,12E+00	1,17E+00	1,33E+00
Eutrophication	kg PO4— eq	4,32E-01	4,35E-01	4,47E-01	4,90E-01

impacts across all the other impact categories considered (increasing between 6.6% and 19.6%), especially in Acidification (A) characterized by the greatest increase, since this impact category is dominated by NOx emissions.

Furthermore, the contribution of the main process phases to the impact categories for each scenario was identified. The results are shown in Appendix. As can be noted, transport contributes to all impact categories, resulting in increasingly worse environmental performances as the distance increases.

With regard to the consumption of fresh water, evaluated through the Available WAter REmaining (AWARE) method, results are shown in Table 6 and stratified for the different contributions. As it is possible to note, the water consumption increases in case of replacing the fresh water with the OMWW and this increase is directly proportional to the transport distance. In particular, the increases for the considered

Table 6Use of fresh water for the considered scenarios evaluated through the Available WAter REmaining (AWARE) method, where S1 and S2 stand for Scenario 1 and Scenario 2, respectively. Values are referred to the Functional Unit.

Impact Contribute	S1	S2 50 km	S2 150 km	S2 500 km
Water use	m^3	m^3	m^3	m^3
OMWW - Electric Energy	26.86	26.86	26.86	26.86
OMWW - Engobe final	5.21	5,21	5.21	5.21
OMWW - Natural gas - production & combustion - S1	1.39	-	-	-
OMWW - Natural gas - prod. & comb S2	_	1.29	1.29	1.29
OMWW - Packaging	2.32	2.32	2.32	2.32
OMWW - Raw materials production	15.77	15.77	15.77	15.77
OMWW - Transport engobe	0.12	0.12	0.12	0.12
OMWW - Transport raw materials	1.67	1.67	1,67	1.67
OMWW - Transport waste	0.01	0.01	0.01	0.01
OMWW - Transport water enriched 50- 150-500 km	-	0.42	1.28	4.26
TOTAL	53.36	53.68	54.54	57.51

scenarios range from 0.6% for a distance of 50 km up to 7.8% estimated at a distance of 500 km. This is due to the consumption of water necessary for the refining phase of fuels, as claimed by Chen (2020).

These results show once again how the greatest criticality in the use of OMWW is linked to transport and therefore to distance. In a future scenario in which the use of electrified transport systems is foreseen, the incidence of transport could be significantly reduced, making the proposed model more efficient.

It should also be noted how the major contributions in water consumption are due to electricity (26.86%) and production of raw materials (15.77%), which remain unchanged for all the scenarios considered in the study. On the other hand, the water saving obtained from the lower use of natural gas determines a negligible variation.

4.2. Validation of Life Cycle Impact Assessment results

In order to validate the consistency of LCIA results, a comparison with recent literature, addressing LCA for the ceramic industry, was performed. Furthermore, also EPD reports have been selected for the comparison, being generally considered objective, credible, and comparable. The literature includes four academic articles and three EPDs (Table 7). According to technical description given in the Paragraph 3.2.1, LCIA results from literature and EPDs were converted to the same FU chosen in this study, that consists in one ton of bricks. In particular, Koroneos and Dompros (2007) approached through a cradle-to-grave analysis the brick production in Greece, while the cradle-to-gate method has been used by Almeida et al. (2010), Lozano-Miralles et al. (2018) and Silvestri et al. (2021). For the comparison with EPDs, only activity included in the cradle-to-gate strategy used in this study were taken into account. These activities are the A1) processing and extraction of raw materials, A2) transport from the production site of raw materials to the bricks plant, and A3) The production of bricks within the plant. Among impact categories, GWP has been selected as the most representative indicator for comparing results. In Table 7, GWP results are shown, as well as the year of each study and the analysis approach used.

As it is summarized in the Table 7, the GWP resulting from the less impactful scenario (S2 - 50 km) is largely greater than GWP from literature studies and EPDs, in a range up to +140%. These variations can be attributed to different characterization models with which the LCA has been performed, as well as different strategies or system boundary. Furthermore, also more performant kilns processes with different efficiency can play a crucial role, as well as the use of different electrical energy mixes. It should be noted that in the considered EPDs, transportation distances are not indicated. Finally, GWP resulted from the S2 - 50 km is slightly lower only to results obtained by Lozano-Miralles

Table 7 Validation of resulting Global Warming Potential (GWP) values with other studies of ceramic bricks production, where S1 and S2 stand for Scenario 1 and Scenario 2, respectively. Activities included in Environmental Product Declaration (EPD) correspond to A1 = Raw material and supply, A2 = Transport, and A3 = Manufacturing. Values are referred to the Functional Unit.

	Year	Approach	GWP (kg CO_2 eq)
S1	2021	Cradle-to-gate	340.0
S2 - 50 km	2021	Cradle-to-gate	330.0
S2 - 150 km	2021	Cradle-to-gate	340.0
S2 - 500 km	2021	Cradle-to-gate	375.0
Literature source			
Koroneos and Dompros	2007	Cradle-to-grave	220.7
Lozano-Miralles et al.	2018	Cradle-to-gate	360.1
Almeida et al.	2010	Cradle-to-gate	141.0
Silvestri et al.	2021	Cradle-to-gate	349.5
EPD source		Stages	
BDA	2015	A1-A2-A3	158.0
Vandersanden	2016	A1-A2-A3	255.3
Wienerberger	2014	A1-A2-A3	277.7

et al. (2018). Indeed, in this study a similar amount of total energy has been considered for foreground activities.

4.3. Economic implications results

A comparative analysis was performed to evaluate the impact of transport on the cost of the OMWW-based brick production process. According to the costs identified in Paragraph 3.4, which refer to the Italian market, the mathematical function of transport cost (C_{tr}) has been defined and the economic value of natural gas saving (ΔC_{en}) has been calculated. As previously stated, the aim of this analysis is to assess the Economic Break-Even Distance (EBED), which allows to balance the cost increasing due to OMWW transport and the energy cost saving.

4.3.1. Cost of olive mill wastewater transport

Starting from the density value reported in Paragraph 3.2.1, 194.16 $\,\mathrm{dm}^3$ of OMWW is the volume to produce 1 ton of bricks. Assuming to use tanks with a maximum capacity of 30,000 L, the function describing cost of transport (C_{tr}) for the calculated volume can be defined:

$$C_{tr} = c_i d \frac{194.16}{30000} \tag{1}$$

Where c_i is the cost of OMWW transport (ℓ /km), which varies for different distance ranges and d is the distance between the OMWW and bricks production sites (km).

4.3.2. Energy saving

Starting from the thermal energy consumption in the considered scenarios (Table 3), the thermal energy reduction (Δ TE) is obtained:

$$\Delta TE = TE_{S1} - TE_{S2} = 2300 \left[\frac{MJ}{ton} \right] - 2132 \left[\frac{MJ}{ton} \right] = 168 \left[\frac{MJ}{ton} \right]$$
 (2)

Being the Higher Heating Value (HHV) of natural gas equal to 38.5 MJ/Sm 3 , it is possible to calculate the volume of natural gas saved (ΔV_{NG}) for the considered Functional Unit:

$$\Delta V_{NG} = 4.36 \left[\frac{m^3}{ton} \right] \tag{3}$$

Finally, considering a cost of natural gas equal to 0.54 €/Sm³, the saving due to lower natural gas consumption can be calculated:

$$\Delta C_{en} = 4.36 \left[\frac{m^3}{ton} \right] \cdot 0.54 \left[\frac{\epsilon}{m^3} \right] = 2.35 \left[\frac{\epsilon}{ton} \right]$$
 (4)

4.3.3. Economic Break-Even Distance definition

By equating the cost of transport (C_{tr}) and the saving due to lower natural gas consumption (ΔC_{en}) as in Equation (5), the EBED can be obtained according to Equation (6).

$$2.35 \left[\frac{\epsilon}{ton} \right] = c_i d \frac{194.16}{30000} \tag{5}$$

$$EBED = \frac{391}{c_i} [km] \tag{6}$$

By applying equation (6), for the different transport costs shown in Table 4 it is possible to verify if the EBED mathematically falls within the range of distances to which each cost refers. The results are summarized in Table 8 and as it can be seen, the calculated distance complies with the definition range only for a cost of 1.88 ϵ /km and is approximately 207 km, as shown in Fig. 2. For distances greater than this break-even distance, the use of OMWW for bricks production would not be economically advantageous. On the other hand, considering a cost of 2.031 ϵ /km, the break-even distance would be higher than the referring range; consequently the proposed strategy would be convenient in the whole range of distances considered (0–150 km).

By applying the approach proposed to the considered scenarios, it is

Table 8Economic Break-Even Distance for the different costs of transport, referring to different ranges of distances.

Cost of transport (€/km)	Distance (km)	Range of distances (km)	
2.031	192	up to 150	Out of range EBED Out of range Out of range Out of range
1.880	207	151–250	
1.714	228	251–350	
1.533	255	351–500	
1.355	288	over 500	

possible to calculate the cost of OMWW transport necessary for the production of 1 ton of bricks (Fig. 3) and consequently the economic performance of the proposed circular economy strategy, also taking into account $\Delta C_{en}.$ The results are summarized in Table 9.

In relation to the functional unit of 1 ton of bricks considered in this study, the calculated costs are low. However, it should be noted, to provide a broader view, that the daily production of bricks in a single furnace can exceed 350 tons, corresponding to costs in the order of several thousand euros per year.

The results of the study were achieved using energy and transport costs for the Italian market. The maximum distance that makes the application of the proposed strategy advantageous is clearly influenced by variations in the considered costs. In particular, an increase in the cost of energy leads to an increase in the break-even distance, making the proposed strategy more attractive. On the other hand, the cost of the transport is inversely proportional to the break-even distance and therefore an increase in this cost would be penalizing for the applicability of the circular economy model analyzed.

Regardless of the influence of costs on the results, the developed model allows to immediately evaluate the feasibility of the strategy once the real costs and distances are known.

Furthermore, the comparison between findings of the proposed methodology with similar studies, summarized in Table 1, demonstrates how this research is consistent with the proposal of an open loop circular economy strategy for the valorization of OMWW.

5. Conclusions and prospects

In the present research, the environmental and economic performances of the proposed circular economy strategy were examined, evaluating the possibility of reusing the OMWW in the brick-making process, instead of fresh water. The analysis was carried out in relation to the distance between the OMWW extraction point and the brick production site, since transport has a strong impact in reducing the

economic and environmental benefits enabled by a lower use of natural gas.

In particular, an LCA was performed, evaluating and comparing the impacts associated with the traditional process with those of the OMWW-based process, varying the transport distance between 50 and 500 km.

The results showed that the overall GWP decreases up to 3.1% when bricks are produced using OMWW, as well as the Abiotic Depletion of fossil fuels is reduced by 4.3%. However, no significant variations were observed for the toxicity impact category group, that ranges from -1.1% to 0.7%. Furthermore, the water consumption for OMWW-based brick production increases up to 7.8%.

The break-even distance, where GWP balances the environmental benefits derived from saving natural gas for firing bricks, resulted equal to 150 km. For the same range of distances, an analogous approach was used in order to evaluate cost increases and savings through a sensitivity analysis. In detail, the mathematical function of transport cost was identified and compared with the lower outlay in natural gas, resulting in a break-even distance of approximately 207 km. The results therefore showed that the environmental constraint is more restrictive than the economic one, determining a limit in the implementation of the proposed circular economy strategy which is economically and environmentally advantageous for transport distances of OMWW within 150 km.

The possibility of reusing the OMWW in the brick-making process, instead of fresh water, has economic and environmental implications, both for the brick industry and for the olive oil production one. In particular, for the production of bricks, environmental advantages linked to a lower use of energy and water would be obtained, as well as economic advantages deriving from a cheaper production process. At the same time, the reuse of OMWW would be guaranteed, avoiding the environmental problems related to disposal.

As mentioned, the proposed strategy was designed for the Italian production context, even if not for a specific site. The break-even distance identified is fully compatible with Italian production, since in Italy there are about 80 brick production sites and 5,000 olive oil mills. Similar conditions can be found in other countries such as Greece and Spain, to which this study could be extended, favoring good circular economy practices.

The proposed calculation model allows in any case, to verify the break-even distance and therefore the applicability of the proposed strategy for each specific case once the distances between the sites and energy and transport costs are known.

In order to apply the results of this research in Italy, the natural development consists in an accurate mapping of the sites where it is

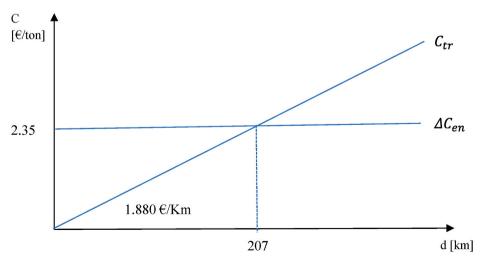


Fig. 2. Economic Break-Even Distance for a cost of transport equal to 1.88 €/km, referring to a range of distances between 151 and 250 Km.

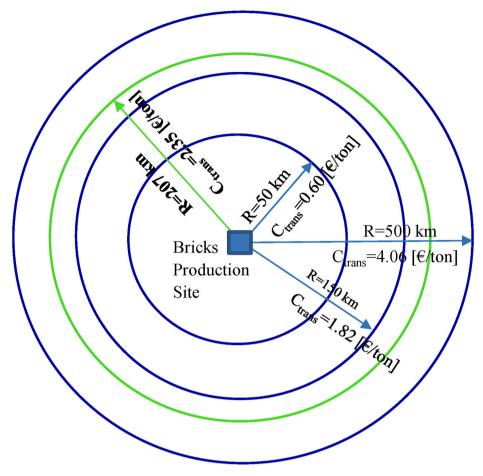


Fig. 3. Variation of olive mill wastewater cost of transport (194.16 dm3) within different ranges of distances.

Table 9Economic performance for the considered scenarios, where S2 stands for Scenario 2.

	S2	S2	S2
	50 km	150 km	500 km
Cost of Transport (ℓ /ton)	0.60	1.82	4.06
$\Delta C_{en}(\ell$ /ton)	2.35	2.35	2.35
Economic performance (ϵ /ton)	+1.75	+0.53	-2.71

possible to find availability of OMWW and brick production sites. This survey would allow to evaluate the real opportunities for applying the proposed circular economy strategy and, at the same time, could represent a location study for the new sites.

The main limitations of the present research, which also represent the further future developments, concern the need to evaluate the environmental and economic implications of different uses of OMWW, such as in fertilizers, and the possibility of extending the analysis also to olive pomace which is the solid by-product of olive oil.

Author contributions

Luca Silvestri: Conceived and designed the analysis, Collected the data, Contributed data or analysis tools, Performed the analysis, Wrote the paper.

Antonio Forcina: Conceived and designed the analysis, Collected the data, Contributed data or analysis tools, Performed the analysis, Wrote the paper.

Gianpaolo Di Bona: Conceived and designed the analysis, Collected the data, Contributed data or analysis tools, Performed the analysis, Wrote the paper.

Cecilia Silvestri: Conceived and designed the analysis, Collected the data, Contributed data or analysis tools, Performed the analysis, Wrote the paper.

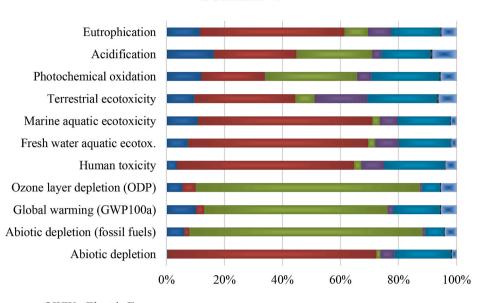
Declaration of competing interest

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.

Appendix

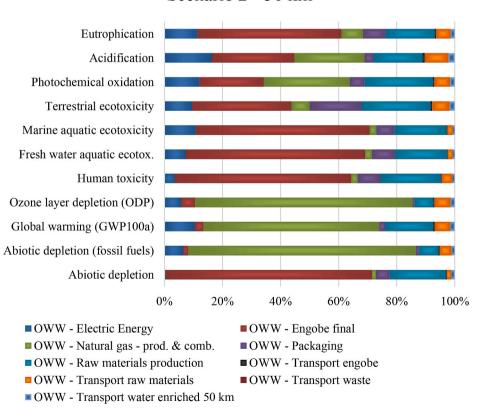
Cradle-to-gate life cycle impacts of facing bricks production in percentages for the considered scenarios. Values are reported per functional unit.

Scenario 1

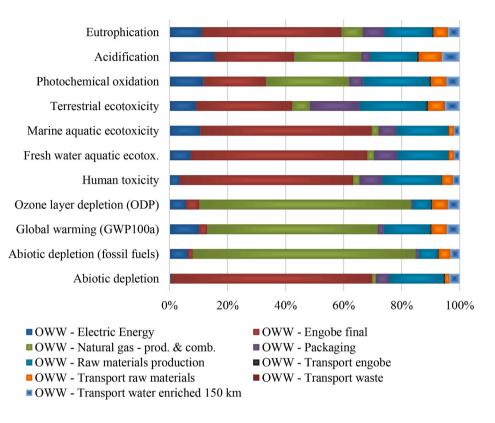


- OWW Electric Energy
- OWW Engobe final
- OWW Natural gas production & combustion S1
- OWW Packaging
- OWW Raw materials production
- OWW Transport engobe
- OWW Transport raw materials
- OWW Transport waste

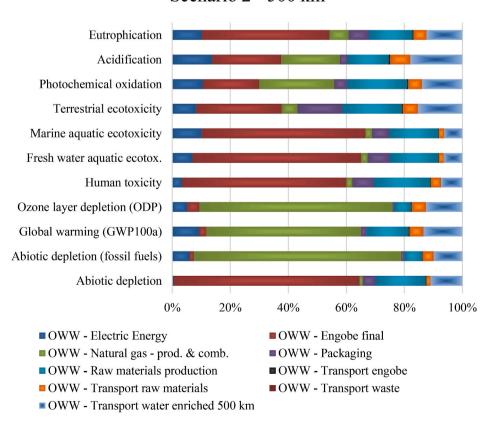
Scenario 2 - 50 km



Scenario 2 - 150 km



Scenario 2 - 500 km



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