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#### EXPLORING THE FACTORS INFLUENCING ENERGY EFFICIENCY IN THE GREEK HOTEL SECTOR Christos Tourkolias<sup>\*</sup>

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#### Abstract

The current paper aims at enhancing the existing literature of studies discussing the parameters, which formulate the phenomenon of the energy efficiency gap in hotels. Specifically, the obtained outcomes, as resulted by the conduction of a stated preference survey are presented and discussed. The survey was carried out on a representative sample of hotels in Greece in the context of the "Consumer Energy Efficiency gap, such as the impact of EU labelling scheme, the linkage of the pro-environmental behaviour with the willingness to invest in more energy-efficient technologies and equipment, the lack of information about electricity prices and the imperfect understanding of energy operating costs. The findings of the survey can be used to explain the role of the behaviour in the decision-making procedures for the further promotion of energy efficiency. Theoretical models can be developed with the collected data to examine the energy efficiency gap phenomenon and to quantify the contribution of different factors to its formulation including the calculation of the implicit discount rate. Finally, the design of more effective policies can be supported with the findings of the paper maximizing the private and social benefits, which are associated with the purchase of energy-efficient technologies and equipment.

#### Keywords

energy labels; energy efficiency; energy behaviour; hotel industry; implicit discount rate

#### Introduction

In Greece, the role of tourism is vital for the whole economy. Specifically, the hotel sector contributed approximately 30.9% to the national GDP in 2018, while the number of employees in the hotel sector amounted to almost 1 million representing 25.9% of the total workforce [1]. The national revenues were equal to  $\leq$ 15.6 billion resulted mainly from approximately 30 million tourists. Considering the current status of the infrastructure, more than 10 thousand hotels are available with a total capacity of 800 thousand beds.

The energy consumption of hotels is considerably high, affecting the competitiveness of the whole sector. According to Figure 1, the per unit primary energy consumption - calculated on the basis of the issued Energy Performance Certificates - of both rooms to let and hotels are higher than many other types of buildings in the tertiary sector. Moreover, the demand for the coverage of space cooling with specific comfort levels is crucial for the hotel sector in Greece, while the seasonality of the hotels creates an additional obstacle in the penetration

of energy-efficient technologies, as their payback period is not considered as attractive enough for the hotel owners/managers.

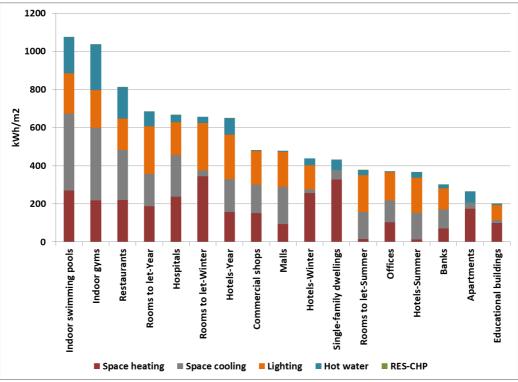


Fig. 1. Primary energy consumption of various types of buildings in the tertiary sector. Source: [2].

The final energy consumption of the hotels is affected by different types of parameters [3]. The physical parameters are related mainly with the characteristics of the building envelope, the climatic location conditions, the type of the installed equipment and technologies including the implemented operation and maintenance procedures and the utilised energy and water sources both in physical quantities and in cost. Furthermore, the operational parameters include: (i) the schedules for the operation of the different functionalities, (ii) the number of the provided facilities and services, (iii) the evolution of the occupancy, (iv) the perceived attitudes of the customers in relation with the indoor comfort levels, (v) the implemented energy efficiency actions and (vi) the level of awareness and knowledge of both the personnel and customers in regards to the improvement of the energy efficiency.

Various scientific papers have analysed and assessed the relation of the hotels with the use and consumption of energy. Initially, the analysis and modelling of the energy consumption were prioritised including the identification of the parameters, which affect its evolution. For instance, Sheng et al. [4] developed both a model for analysing the energy use and specific energy benchmarks of five-star hotel sector in China. Idahosa et al. [5] conducted a panel analysis to understand the factors that drive the energy consumption of hotels in South Africa and Oluseyi et al. [6] explored the carbon footprint of hotels in Nigeria; Parpairi [7] presented the use of successful practises and policies in Greece. Cingoski & Petrevska [8] discussed the obtained outcomes from an online survey among managers of different types hotels for evaluating the potential implementation of energy efficiency technologies in hotels and for identifying the factors, which affect the energy use and consumption. Also, several studies aimed at evaluating the energy and environmental performance of the hotels through the calculation of specific energy indicators [9-12] and the identification of the potential for the extensive penetration of the most effective energy efficiency technologies [13]. Also, emphasis has been put on the examination of customers' perceptions and behaviour towards the application of environmental practises by hotels [14–20]. Limited focus though has been placed on the behavioural aspects of the hotel owners/managers towards the promotion of energy efficiency. More specifically, Leroux and Pupion [21] examined the reasons for the adoption of the eco-label by the hoteliers. A non-parametric quantitative methodology was performed in 182 hotels in France in order to analyse the main factors leading to this choice.

According to the obtained results, the adoption of the eco-label is affected by the risk perceptions, the adoption of eco-label and institutional issues. The regulatory and imitative behaviour and specific entrepreneurial characteristics, such as like risk-taking and bureaucracy, are considered as the main factors for the case of non-

certified hotels, while the difficulties for obtaining the certification and the positive attitudes for the protection of the environment lead to the promotion of eco-labels in certified hotels. Asadi et al. [22] examined the potential effects of green innovation on sustainable performance in the hotel sector. Specifically, the factors influencing the adoption of green innovation were analysed resulting that the environmental and economic performances have the strongest influence for the promotion of green innovation in hotels. Finally, specific studies have examined the role of energy labelling on the formulation of the different perceptions and attitudes mainly for the residential appliances [23–25]. No study though was identified regarding the perceptions and attitudes of hotels owners/managers for the impact of energy labels in the equipment utilised in their premises. The limitations revealed by the literature review are critical because the majority of the countries have already set ambitious targets for increasing energy efficiency. Nevertheless, the "energy efficiency gap" or "energy paradox" [26] creates a serious problem as the businesses and the consumers in reality do not support the acceleration of the energy efficiency equipment or appliances even if the triggered benefits are much higher than the required costs (Damigos et al., 2020). According to Damigos et al. [23] various market and non-market failures contributed to the problem of the energy-efficiency gap, while the energy labels can be confronted by the intensive utilization of energy labels.

Aiming to contribute to knowledge and existing literature, this paper provides an insight into the factors, which are responsible for the energy efficiency gap in the hotel sector. Specifically, the results of a stated preference survey are presented as collected by approximately 100 Greek hotels owners/managers via the method of the computer-assisted web interviewing (CAWI) within the framework of the "Consumer Energy Efficiency Decision making (CONSEED)" project. The survey was designed to collect information about the factors affecting the decision for the purchase of energy-efficient heating and cooling systems. More specifically, the present research aims at:

- assessing the role of energy efficiency in the decisions, which are made by the hotel owners and managers compared with other operational attributes of the heating and cooling technologies ;
- studying specific market and behavioural barriers (such as the provision of incomplete and inadequate information, the imperfect understanding of the triggered energy operating costs and liquidity constraints) and hidden costs and benefits, which hinder the hotel owners and managers from potential investments in energy-efficient technologies and equipment;
- examining the role of various factors (socio-economic characteristics, attitudes, beliefs and perceptions) which affect the decision of the hotel owners/managers for further investments in the field of energy efficiency;
- evaluating the impact of the existing EU labelling scheme on energy-efficient investments and to test
  if the use of monetary labels would foster the willingness of hotel owners/managers to purchase more
  efficient heating and cooling systems;
- calculating the implicit discount rate used by the hotels owners/managers to compare the upfront costs of the energy-efficient technologies and equipment to the discounted value of future savings in energy expenditures during their decision to purchase an energy-efficient technology and equipment.

According to the authors' best knowledge, the above-mentioned issues have not been examined in the literature so far, and thus the current paper provides a solid basis for further discussions in the respective scientific questions. The structure of the paper consists of the Sections 2 and 3, which presents the utilised materials and methods and the obtained results correspondingly. Section 4 analyses the main outcomes of the conducted survey, while Section 5 summarises the main conclusions of the paper.

#### **Materials and methods**

#### Survey design

The survey was carried out among hotels owners/managers using a structured questionnaire. This approach allowed us to elicit specific information on particular variables and study the cause-effect relationships and connections as a function of the socio-economic and operational characteristics of the participated hotels into the survey [27,28]. The questionnaire was designed following the recommendations of the relative literature [29–33] to achieve the specific objectives of the research. After preparing the first version of the

questionnaire, a pilot survey was conducted in order to test it and identify vague and problematic questions, which could lead to biased answers. The final version of the questionnaire comprised a set of specific, concise and standardised questions divided into seven parts. The first part aimed at examining the type of the installed heating and cooling technologies (either central systems or autonomous units), the intention of the participant to install a new heating and system (central or not) in period of five years and the degree of involvement of the participant in the process of purchasing the heating and cooling equipment. The second part investigated the role of the improvement of energy efficiency with other attributes of the heating and cooling technologies such as the price, the years of warranty, manufacturer's reliability, annual electricity consumption, after-sales service, the installed capacity of the heating and cooling technologies, the annual operating costs and the expected annual CO<sub>2</sub> emissions. The discounting question was included in the third part of the questionnaire to elicit the expected return on the potential purchase of the energy-efficient heating and cooling system and to calculate the implicit discount rate. Specifically, the respondents considered the following question:

"Suppose you could buy an energy-efficient cooling/heating system, which would reduce the operating costs of your hotel. The new cooling/heating system will cost {500, 1000, 1500} more and is expected to operate for 10 years. How much would you have to save in your energy bill per year during the next 10 years to pay for the additional {500, 1000, 1500} Euros?"

The fourth part of the questionnaire investigated attitudes and perceptions of the participants concerning the perceived costs and benefits emphasising the various barriers that hinder the further promotion of energy-efficient technologies and equipment. The fifth part focused on the factors, which foster the utilisation of the current energy label and the proposed monetary label for the heating and cooling systems. Firstly, the hotel owners/managers declared their familiarity with the current energy label, while it was examined if selection and purchase of a new energy-efficient system has been mobilised by the energy label. Moreover, the participants evaluated if the energy label is understandable, reliable, used by sellers for the promotion of a specific type of technologies, affecting the decision for buying a specific unit and facilitating the potential buyer to realise her energy consumption and calculate the operating costs of the heating and cooling system. Participants were informed that the current energy label is planned to be substituted by an alternative, which provides information about the total energy cost on annual basis in addition to the annual energy consumption, as follows:

"Imagine the use of energy labels in heating and cooling systems is mandatory. These tags will provide the prospective buyer with information on the annual energy consumption of the heating and cooling systems in kWh and in addition the annual energy cost in euro. For example, "the annual energy cost of this device amounts to  $\notin$  Y annually". This information will be based on the model's cooling/heating power, its average energy efficiency and an average electricity price."

Then, they were asked to evaluate the various options assessed for the case of the existing energy label. The sixth part of the questionnaire explored the environmental perceptions of the participants and the potential installation of specific energy and environmental management systems including the renovation of the building envelope. Finally, the seventh part of the questionnaire information about the operational characteristics and the ownership of the hotel were collected.

The survey was carried out in December 2017 via the CAWI method by a specialised market research and opinion polling company in Greece. Specific quality assurance procedures were applied in compliance with the Data Collection Quality Control rules of the Association of Greek Market & Opinion Research Companies. The target group was owners/managers of Greek hotels and 'rooms to let'. The sample derived by an online research panel maintained by the Opinion Research Company. Furthermore, the sample was selected to be representative in relation to the geographic dispersion and the main characteristics of hotels in Greece. Each hotel owner or manager was notified and received by email an electronic link to the survey questionnaire without the provision of any incentive to participate in the survey. Totally, 102 hotel owners/managers completed the developed questionnaire successfully.

#### Data analysis

The data were analyzed using univariate, bivariate and multivariate statistics. Univariate (i.e. descriptive) statistics was employed to summarize the variables relating to the perceptions, beliefs and characteristics of the

participants. As far as the estimation of the implicit discount rate (IDR) is concerned, the Kaplan–Meier nonparametric estimator (Kaplan & Meier 1958) was utilised to estimate the respective IDR value for those who answered the discounting question. With regards to the bivariate analysis, chi-square tests were conducted to examine potential associations between energy- and environmental-related perceptions and the demographic and attitudinal characteristics of the respondents. Finally, the data were also analysed using multivariate regression models (i.e. binary and ordered logistic ones) following the relevant literature [34–38], to explore which attitudinal and demographic factors influence energy-efficiency decision making.

#### Results

#### Description of the general characteristics of the hotels

According to the collected data, 42% of the hotel units operate as vocational hotels while 31% of them are located in cities covering different multi-purposes travels. Most of the examined hotels are privately owned (90%); 46% of the hotels operate on continuous basis (12 months) and 35% of the hotels have seasonal operation (June to September). During high season, the occupancy rate exceeds 90% for the vast majority of the respondents (70% of the sample). The average hotel occupancy rate is equal to 57% while the average capacity amounts to 99.4 bed-places. The average hotel rating is equal to 2.55 stars while the average permanent and seasonal personnel is equal to 7.6 and 13.7 employees, correspondingly.

As regards the heating and cooling system, 25.5% of the hotels have in operation a central system, 63.7% a distributed system and the rest (i.e. 10.8%) both systems. According to the obtained responses, the average energy cost is equal to  $\notin$ 4,412 on monthly basis (min:  $\notin$ 100 max:  $\notin$ 90,000), while is strongly related to the hotel rating and capacity. More specifically, the average monthly cost for the 1-star hotels is  $\notin$ 860, for the 2-star hotels is  $\notin$ 3,100, for the 3-star hotels is  $\notin$ 1,700, for the 4-star hotels is  $\notin$ 5,400 and for the 5-star hotels is  $\notin$ 34,500. Moreover, the average energy cost per month is  $\notin$ 140 for 0-20 bed-places,  $\notin$ 2,100 for 21-50 bed-places,  $\notin$ 3,800 for 51-100 bed-places,  $\notin$ 4,600 for 101-300 bed places and  $\notin$ 90,000 for more than 1,000 bed-places. In total, the average monthly energy costs per bed-place range from  $\notin$ 35 to  $\notin$ 3,300 (average:  $\notin$ 3,300; median:  $\notin$ 210). Finally, the average energy cost is equal to  $\notin$ 2,700 and to  $\notin$ 3,000 on monthly basis for the case of hotels with central heating and cooling system and distributed system respectively. The operation of a mixed system leads to considerably highest energy cost ( $\notin$ 16,300 per month).

According to Figure 2, 30% of the participants stated that a Building Energy Management System has already been installed in their hotel. The percentage of the hotels, which have been, renovated amounts to 68% indicating the importance of energy efficiency in the operation of the hotel industry. The main reasons for the non-initiation of the required efficiency interventions are related to the relatively young age of the buildings and the difficulty to attain the appropriate funds for the energy upgrade of the buildings and the other facilities. Further, environmental interventions have been initiated. Explicitly, 41% of the sample has installed a waste reduction system, 44% a waste recycling system and 47% a water-saving system, demonstrating the emphasis given in addressing the environmental challenges triggered by the operation of the hotels.

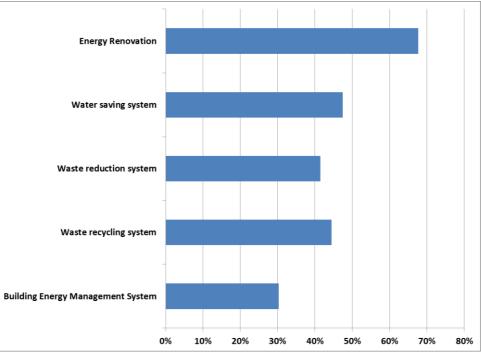


Fig. 2. Implemented energy and environmental interventions in the examined hotels

# Factors affecting the purchase of heating and cooling systems

The factors that affect the purchase of heating and cooling systems were examined (Figure 3). The analysis showed that the energy consumption and the manufacturer's reliability (89.2%) are considered as the most essential aspects from the hotel owners/managers during the purchase of the energy efficient heating and cooling systems. Moreover, the annual energy cost and the after-sales service are also considered important (81.4%), followed by the capacity of the system, the years of warranty and the price (79.4%, 78.4% and 74.5%, respectively). The CO<sub>2</sub> emissions are not considered as a crucial parameter for the selection of the heating/cooling system as only 66.7% of the respondents highlighted its importance. The factors that are linked with the performance of the heating and cooling systems and the comfort levels to ensure customers' satisfaction are considered as more important during the selection of a new heating and cooling system. Finally, 20% of the respondents plan to install a new heating and cooling system - either central or individual - within the next five years, while the majority of the participants (57%) claimed that his involvement in the decision for selecting a new heating and cooling system is essential and definitive.

Chi-square tests revealed that women focus mainly on the manufacturer's reliability (chi2(1) = 4.1839, p = 0.041) and the capacity of the system (chi2(1) = 3.7503, p = 0.053). Furthermore, it seems that older people are more concerned about the price of the system (chi2(3) = 9.1127, p = 0.028), the energy consumption (chi2(3) =13.2082, p = 0.004), the after sales service (chi2(3) = 9.0016, p = 0.029), the annual energy cost (chi2(3) =18.5170, p = 0.000), and the CO<sub>2</sub> emissions (chi2(6) = 13.4253, p = 0.037). Finally, in comparison to franchised hotels, privately owned hotels consider more important the years of guarantee (chi2(2) = 12.6018, p = 0.002), the manufacturer's reliability (chi2(1) = 4.2549, p = 0.039), the energy consumption (chi2(2) = 11.4807, p = 0.003), the capacity of the system (chi2(1) = 10.5332, p = 0.001), the annual energy costs (chi2(2) = 7.2091, p = 0.027), and the CO<sub>2</sub> emissions (chi2(2) = 12.1957, p = 0.002).

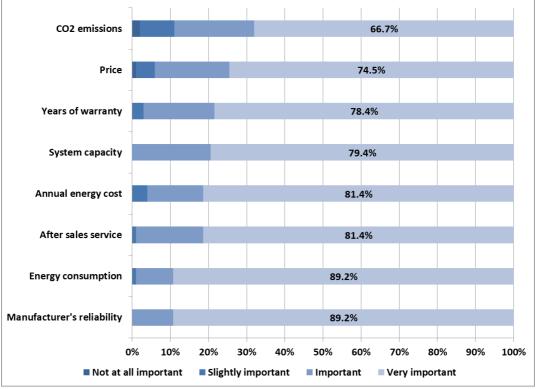


Fig. 3. Importance of factors affecting the purchase of heating and cooling systems at hotels.

# Perceptions towards energy efficiency and environment

The analysis of the perceptions for the promotion of energy efficiency in hotels (Figure 4) demonstrates that the vast majority of the participants link strongly the energy efficiency technologies with the reduction of the energy consumption and the triggered environmental impacts (93.1% and 91.2% of the sample, respectively). Furthermore, more than 80% of the responders stated that the energy upgrade of the heating and cooling equipment would be proved beneficial to them as they will improve the existing comfort levels and will lead to an increase of the current value of the enterprise (84.3% and 82.4% of the sample, respectively).

The majority of the participants initially said that have adequate knowledge of the issues about energy use and consumption and the energy prices. Specifically, the hotel owners/managers claimed that they are informed about the energy prices (83.3% of the sample), understand the heating and cooling system's energy consumption (81.4% of the sample) and are certain about the economic benefit triggered by the more energy-efficient system (77.5% of the sample). Nevertheless, approximately 90% of the participants admitted that they are not informed of the existing energy consumption of the hotel, and about 47% of the respondents were neither able nor willing to mention the required energy costs for the operation of their hotels.

The obstacle of the limited financial incentives was also highlighted as 53.9 % of the participants claimed that it is not possible to finance the upgrade to a more energy-efficient cooling/heating system, while the limited financial incentives was mentioned by 66.7% of the sample as the most important barrier towards making more energy-efficient choices.

Furthermore, a problem was pinpointed regarding the identification of the energy-efficient heating and cooling systems. Specifically, 39.2% the sample are not capable of recognizing the energy-efficiency heating and cooling systems demonstrating the need to provide more accurate information, while 28.4% support the statement that all the new heating and cooling technologies have identical energy performance levels. Yet, the majority of the participants seem to trust the technical reliability of the energy-efficient heating and cooling systems; less than 10% strongly believe that more energy-efficient heating and cooling systems are less reliable. Finally, the vast majority of the participants expressed their concern about the main problems of the environment such as pollution, climate change etc. Specifically, 76% and 20% of the sample are very worried and worried correspondingly about the current status of the environment; only 4% downgrades the severity of the environmental problems.

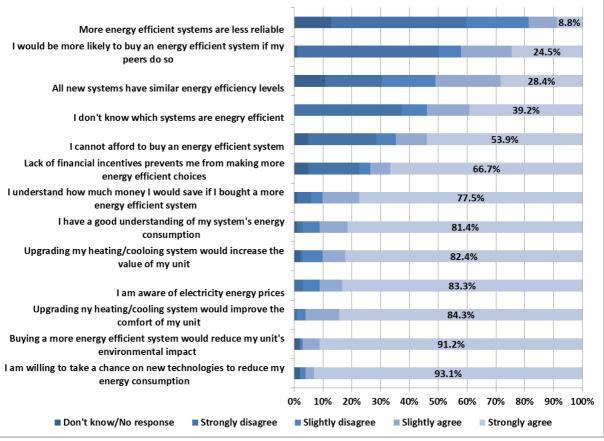


Fig. 4. Agreement with specific statements about energy efficient heating and cooling systems at hotels.

Chi-square tests lead to the conclusion that women face more significant barriers due to the limited financial incentives, which are available (chi2(2) = 16.0959, p = 0.000). Older people support the exploitation of new technologies to reduce their energy consumption (chi2(6) = 14.0135, p = 0.029), face more significant barriers due to the lack of the financial incentives (chi2(6) = 22.5244, p = 0.001), and are more willing to buy and install a more energy-efficient cooling/heating system if their peers do so (chi2(6) = 15.4456, p = 0.017). Finally, private owners face more significant barriers due to the lack of the financial incentives (chi2(2) = 9.2056, p = 0.010), and are less likely to buy and install a more energy-efficient cooling/heating system if their peers do so (chi2(2) = 9.2056, p = 0.010), and are less likely to buy and install a more energy-efficient cooling/heating system if their peers do so (chi2(2) = 9.2056, p = 0.010), and are less likely to buy and install a more energy-efficient cooling/heating system if their peers do so (chi2(2) = 9.2056, p = 0.010), and are less likely to buy and install a more energy-efficient cooling/heating system if their peers do so (chi2(2) = 9.004).

# Understanding, opinions and beliefs about the different version of the energy labels

About two-thirds of the respondents have been informed about the current energy labels, while approximately 70% claimed that the energy labels affected their decision to buy the already installed heating and cooling system. Further, a comparison was initiated between the current energy label and the alternatively proposed based on the provided monetary information. According to Figure 5, the participants confirmed that the current energy labels are understandable (74%), trustworthy (54%), can affect the consumer's decision (79%) and facilitates the better understanding of system's energy consumption and the easiest calculation of the operating costs (70%). Moreover, they claimed that the current energy labels are manipulated and utilised by the sellers (66%) so as to foster the decisions of the potential consumers. The proposed monetary labels manage to improve the understanding (86%) and the trustworthiness (56%) compared to the current energy labels. Furthermore, the realization of the system's energy consumption and the calculation of the operating costs are performed more effectively according to the statement of 90% of the respondents. Nevertheless, the proposed monetary label seems to influence negatively the purchase decision (74% of the sample strongly agree) since the sellers will use it as a more effective tool for manipulating potential consumers (77% of the sample strongly agree). It should be noted that no statistical significance was resulted by the analysis of the differences between the two examined types of labels.

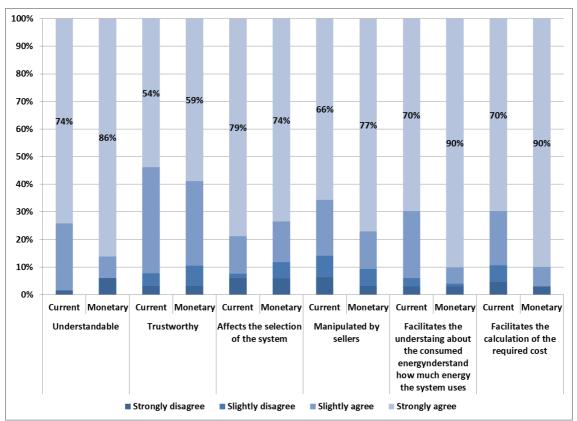


Fig. 5. Comparison of the alternative version of the energy labels.

# Exploring the factors affecting the valuation of energy efficiency

#### A. Importance of energy consumption in choosing energy-efficient heating and cooling systems

The exploration of the factors, which constitutes energy consumption as the main determinant during the selection of energy-efficient heating and cooling systems in hotel sector was implemented through an ordered logistic model (dependent variable coded as 1 = Not at all important, 2 = Not very important; 3 = Fairly important and 4 = Very important). The obtained outcomes in Table 1 revealed that the energy efficiency is considered as more important for those who believe that the role of the installed capacity of the heating and cooling systems (coefficient = 3.163) and the annual operating cost of the enterprise (coefficient = 1.791) is the most important and are willing to use new technologies for reducing the operating costs (coefficient = 2.779). Furthermore, the importance of energy efficiency varies by the hotel rating (coefficient = -0.810). The odds of mentioning the energy consumption as more important are 23.6 times greater for those who perceive as crucial the role of the installed capacity of the selected heating and cooling systems under the prerequisite that all of the other variables in the model are retained constant. Similarly, the importance is higher for those who also consider important the annual operating costs (16.1 times). Finally, the importance of energy efficiency decreases by almost 45% as the hotel rating increases.

Variables (Coding in parentheses)	Coefficient	Standard error
Importance of the installed capacity of the heating and cooling systems (1 = Not at all important to 4 = Very important)	3.163**	1.461
Importance of the annual operating cost (1 = Not at all important to 4 = Very important)	1.791***	0.698
Willingness to use new technologies for reducing the operating costs (1 = Fully disagree to 4 = Fully agree)	2.779**	1.180
Hotel rating (1 = one star to 4 = 5 stars)	-0.810**	0.395
Constant cut1	20.103	10.250
Constant cut2	23.443	10.866
Observations = 100; Log likelihood = - 37.74; Pseudo R <sup>2</sup> = 0.496		·

Table 1. Factors of the role of energy consumption during the selection of energy-efficient heating and cooling systems

(\*\*\* p<0.01, \*\* p<0.05, \* p<0.1)

#### B. Awareness of the existing heating and cooling systems energy label

The examination of the factors, which affects the awareness of the existing energy labels of the various heating and cooling systems (dependent variable coded as 1 = Yes; 0 = No) was carried out by a binary logistic model. According to the obtained results (Table 2), those who don't state that 'it is likely to purchase an energy-efficiency heating and cooling system in the case that other enterprises proceed with the same decision' (coefficient = -0.524), identify the energy-efficient heating and cooling systems (coefficient = -0.592) and have already installed a water conservation system (coefficient = 1.315) are more probable to be aware of the existing energy label for the heating and cooling systems. The odds of being informed about the energy label are 3.7 times greater for those who have decided to install a water conservation system under the prerequisite that all of the other variables in the model are retained constant. Finally, the odds are 10% lower for those who disagree with the statement 'it is likely to purchase an energy-efficiency heating and cooling systems. The codes of being and cooling system in the case that other enterprises proceed with the same decision' and who identify the energy-efficient heating and cooling systems.

Variables (Coding)	Coefficient	Standard error
It is likely to purchase an energy-efficiency heating and cooling system in the case that other enterprises proceed with the same decision (1 = Fully disagree to 4 = Fully agree)	-0.524***	0.202
It is not feasible to identify the energy-efficient heating and cooling systems (1 = Fully disagree to 4 = Fully agree)	-0.592***	0.530
A water conservation system has been installed in the enterprise (1 = Yes and 0 = No)	1.315**	0.207
Constant	3.034*	0.806
Observations = 99; Log likelihood = - 46.8; Pseudo R <sup>2</sup> = 0.248		

(\*\*\* p<0.01, \*\* p<0.05, \* p<0.1)

#### C. Influence of the existing energy label in choosing heating and cooling systems

The role of the energy labels into the formulation of consumers' decision (dependent variable coded as 1 =Yes; 0 =No), was examined through a binary logistic model (Table 3). According to the obtained results, those support the statement that all the new heating and cooling systems have similar levels of energy efficiency (coefficient = -0.734), believe that the lack of financial incentives hinders the investments in energy efficiency (coefficient = -0.595), are willing to invest in energy efficiency under the precondition that other enterprises proceed with the same decision (coefficient = -0.427) and their enterprise does not have a Building Energy Management System (coefficient = 0.991), are less probable to be affected by the energy label when selecting a heating and cooling systems. Specifically, those who state that all the new heating and cooling systems have similar levels of energy performance, who face financial constraints and are affected by their peers, are less probable to be affected by the energy label by almost 48%, 55% and 65%, respectively. On the other hand, those who have and operate a Building Energy Management System are 2.7 times more probable to take into consideration the energy labels

when buying heating and cooling systems for their enterprises.

Table 3. Determinants of the energy label's role in choosing heating and cooling systems

Variables (Coding)	Coefficient	Standard error
All new heating and cooling systems have similar levels of energy efficiency (1 = Fully disagree to 4 = Fully agree)	-0.734***	0.264
Lack of financial incentives is a barrier for making more energy-efficient choices (1 = Fully disagree to 4 = Fully agree)	-0.595**	0.306
It is likely to purchase an energy-efficiency heating and cooling system in the case that other enterprises proceed with the same decision (1 = Fully disagree to 4 = Fully agree)	-0.427**	0.218
Existence of Building Energy Management System (1 = yes and 0 = no)	0.991***	0.636
Constant	5.713	1.512
Observations = 83; Log likelihood = - 21.6; Pseudo R <sup>2</sup> = 0.212		

(\*\*\* p<0.01, \*\* p<0.05, \* p<0.1)

#### Calculation of implicit discount rates

The implicit discount rate (IDR) is estimated using the following equation as the internal rate of return (IRR) of the investment on the energy-efficient cooling and heating system:

$$C_0 = \sum_{t=1}^{10} \frac{AES_t}{(1 + IDR)^t}$$

where  $C_0$  is the investment of 500, 1000 or 1500 Euros and  $AES_t$  is the annual energy savings requested by the respondent

In total, only 39% of the participants responded to the IDR questions. The vast majority (i.e. more than 90%) of those who didn't answer the question said that they didn't know how to estimate future savings. Those who stated an amount for the required annual savings were asked the reason why they declared the specific amount. Practically all respondents justified their answer by calling on the payback period of the investment (i.e. *"I want to get my money back in .... years"*). This finding is an indication that respondents may lack the knowledge required to do the financial calculations required and, thus, rely on simple rules of thumb, as other scholars have also noticed [39–41]. For this reason, the payback period was also estimated, besides the IRR, based on the responses provided by the participants.

The minimum IDR was equal to 0% and the maximum 480%, respectively. The non-parametric Kaplan-Meir statistics (i.e. mean and median values and the respective 95% confidence intervals) are presented in Table 4. The minimum payback period is less than 3 months and the maximum is 10 years. The non-parametric mean and median values, together with the lower and upper 95% bounds were also estimated using the Kaplan-Meier estimator and the results are given in Table 5. Figure 6 illustrates the 'survival' curves for the IDR and the payback period, accordingly.

		Mean			Me	edian	
Estimate	Std. Error	95% Confid	ence Interval	Estimate	Std. Error	95% Confiden	ce Interval
		Lower Bound - Upper Bound				Lower Bound - L	Jpper Bound
65.5%	15.8%	34.5%	96.5%	31.1%	1.2%	28.7%	33.5%

#### Table 4. Kaplan-Meir estimates for the IDR (%)

# Table 5. Kaplan-Meir estimates for the payback period (years)

		Mean			Μ	ledian	
Estimate	Std. Error	95% Confid	ence Interval	Estimate	Std. Error	95% Confider	nce Interval
		Lower Bound	- Upper Bound			Lower Bound -	Upper Bound
3.5	0.45	2.6	4.3	3.0	0.2	2.7	3.4

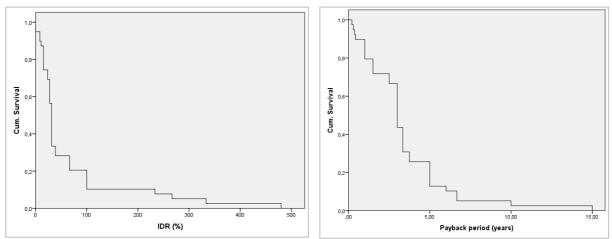


Fig. 6. IDR (left) and payback period (right) 'survival' curves

Finally, the factors affecting the IDR of the hotel owners/managers were explored through a log-linear regression model. According to the results, the value of the IDR increases for the women (coefficient = 0.730) and for those who are not risk adverse, in general (coefficient = 0.103) and decreases with the increase in the capacity of the hotel (coefficient = -0.150). According to the coefficients, and given the log-linear form of the model, the IDR for the women is more than twice as high as that for men. Further, a one-unit increase in riskiness increases the IDR by almost 11%, while a one-unit change in the capacity of the hotel changes the IDR by 14%.

Table 6. Determinants of the IDR of the hotel owners/managers

Variables (Coding)	Coefficient	Standard error
Gender (1 = Male and 2 = Female)	0.730**	0.316
Willingness to take risk (1 = not at all; 10 = totally willing)	0.103**	0.061
Capacity of the hotel in beds (1= 1-20, 2=21-50, 3=51-100, 4=101-150, 5=151-200, 6=201-300, 7=301-500, 8=501-1000, 9>1000)	-0.150**	0.069
Constant	2.440***	0.591
Observations = 35; Adj. R-squared =0.206		

Note: The dependent variable is the ln(IDR); \*\*\* p<0.01, \*\* p<0.05, \* p<0.1

#### Impacts

Worldwide, governments adopt policies to improve energy efficiency in end-use sectors. However, energyefficient technologies are not usually adopted despite the fact that it is strictly required for businesses in accordance with the private costs and the delivered benefits. The "under-investment" in energy-efficient technologies, is known as "energy efficiency gap" or "energy paradox" [42] and has been high on the priority list for at least three decades. Previous research suggests that energy paradox is relate among others to the market and non-market failures, biased beliefs, bounded rationality and heuristic decision-making [43].

The main objective of the current paper is the enhancement of the energy policy and research agendas by exploring the factors affecting energy efficiency decisions in the hotel industry. To this end, the paper illustrates the results of a stated preference survey conducted in a sample of Greek owners/managers of collective accommodation facilities. To the authors' best knowledge, this is the first attempt to gather and analyse information on the main factors, which are associated with the energy efficiency gap in the Greek hotel sector. More explicitly, the survey explored issues such as the effectiveness of energy labels and the role of pro-environmental behaviour on the decision-making procedures applied by the hoteliers, the impact of misinformation about the energy consumption and cost on the energy efficiency gap, the significance of the energy-efficiency with other attributes of heating and cooling equipment, etc.

The concluded outcomes by the survey indicate that the hotel industry is not inattentive to energy efficiency. The energy consumption, together with the reliability of the manufacturer, dominates the purchase decision of a heating/cooling system. These factors are linked with the effort of the industry to ensure customers' satisfaction. The majority of the respondents seem to be sufficiently informed about the energy labels (approximately 70% of the sample). It should be highlighted that the vast majority of the participants link the energy efficiency technologies and equipment with the reduction of the energy consumption and the related environmental impacts, suggesting the importance of pro-environmental behaviour in the transition towards more sustainable tourism industry. Moreover, the survey revealed several important types of barriers and biases (both market and behavioural). For instance, more than half of the respondents argue that they are not able to pay for the installation of energy efficiency technologies and nearly two-thirds indicated the limited financial incentives, which are available, as the most important barrier hindering their energy-efficient decisions. Further, there is evidence of bounded rationality provided that the majority of the respondents said is not been informed about the actual their energy consumption of the hotel, 60% didn't reply to the discounting question claiming that the cannot estimate future energy savings, half of them was not able or willing to state their actual energy costs and almost 40% admitted that they are not capable of recognizing the energy-efficiency heating and cooling systems. Finally, the estimated mean IDR is high (i.e. 65.5%), although it lies within the range reported by other empirical studies [44]. The high IDRs and, consequently, the very short payback periods are not uncommon in corporate decision-making and are related to "...irrational 'safety margin' without any further economic rationale..." [44]. Yet, this could hinder the adoption of energy-efficient technologies, since, in real investments, such requirements are rarely satisfied.

The above-mentioned remarks could provide the evidence base for policy-development for the improvement of the energy efficiency in the hotel industry and bring significant environmental and socio-economic benefits. According to previous studies, the energy saving potential in hotels ranges from 10% to 25% of the energy they consume depending on the geographic region and other parameters [7]. The same conclusion is confirmed by several European studies, which presents estimated savings of 15-20% for heating, 40-70% for produced hot water, 7-60% for lighting and 5-30% for cooling [3]. Reducing the energy consumption would result in economic benefits given that energy costs typically account for 3-6% of overall operational costs of hotels [45]. For instance, on average hotels, a comprehensive energy management plan could reduce total energy demand by 742 MWh per year [46]. Additional economic benefits could result from tourists who place importance on hotels implementing green practices [14,15]. Environmental benefits would result from the reduction of CO<sub>2</sub> and air pollutants associated with energy generation. For instance, the standard emission factor for electricity consumption, in EU-27, is 0.460 t CO2 per MWh<sub>e</sub> [47]. Finally, improving energy efficiency in hotels may have social benefits. The economic robustness of the sector could be strengthened and, thus, the employment could be maintained or even further increased.

#### Conclusions

This main objective of the study was the enhancement of the existing literature in regards to the energy efficiency gap in the hotel sector. The outcomes of the survey provide some new insights for policy purposes. For instance, policy-makers should support the development and evaluation of new promising policies, which could alleviate barriers and biases, e.g. the lack of financial support, the lack of access to loans, the limited realization of the energy use and consumption and the energy prices, the reduction of the 'discounting gap', i.e. the difference between market rates and implicit discount rates, etc. Further, they should 'nudge' the hoteliers in the direction of energy efficiency by promoting the environmental, social and economic benefits for their businesses (e.g. the increasing demand for green hotel accommodation worldwide). Yet, any policy recommendations should be treated taking into consideration the relatively small sample size and the fact that the reliability of the received answers depends on participants' capability to provide the appropriate responses. Finally, the outcomes and the limitations of the survey may serve as starting points for future research, especially as regards the role of behavioural biases and the factors affecting the 'discounting gap' and the energy-related financial illiteracy of the hoteliers. To this direction, future surveys should employ larger sample sizes and more complex designs (e.g. split-sample techniques, choice experiments, etc.) for exploring the hoteliers' energy-related investment behaviour and its interrelationship with the energy paradox.

#### **Conflict of interest**

The authors declare that they have no conflict of interest.

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#### ASSESSING THE VISUAL IMPACTS OF SURFACE MINING: A SYSTEMATIC REVIEW

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#### Abstract

This paper provides a systematic review of the methodologies used to evaluate the visual impacts of surface mining. The main objectives are: (a) to analyse the scientific literature and identify the most important issues and the methods and tools used; (b) to conduct an analysis using descriptive of statistical methods and qualitative interpretation; and (c) to evaluate the state of knowledge on this particular topic and identify gaps in the literature, to suggest future research directions. The findings of the analysis suggest that there is no single method capable of integrating all dimensions of the landscape and, thus, future research should put more emphasis on incorporating as many factors contributing to the visual impact of mining as possible towards developing holistic approaches.

#### Keywords

visual impacts; landscape change; mining; quarrying; systematic review

#### Introduction

Surface mining causes dramatic changes in the landscape [1,2]. These changes are in several cases permanent, obvious and intense and, therefore, constitute a principal cause of serious public opposition against mining [3]. The severity of the problem led to continuous interest, worldwide, for the development of methods that could assess the magnitude of the visual impact induced by surface mines and quarries. In the '70s and for almost thirty decades, the primary tool used has been the visual quality assessment of the landscape [2] via semi-quantitative methodologies [4–6]. Later on, the development of Geographic Information Systems and other technologies, such as video-imaging, opened new possibilities for the quantification of surface mining-related changes in the landscape. As a result, several methods and approaches were introduced towards measuring alterations on the topography, chromatic contrast of the excavated surface with the surroundings or land cover/land use changes [3,7–11].

There is no doubt that the necessity to evaluate and quantify the visual impacts of surface mining is reflected in the scientific literature. Nevertheless, there is no systematic review article on this topic. Systematic literature review (SLR) is referred to a more or less systematic search of previous studies, which aims to synthesize previous research findings through a replicable, scientific and transparent process [12–16]. SLR can be used for evaluating the state-of-the-art knowledge on a certain issue or research problem to discuss a particular matter or identify knowledge gaps, to examine the validity or accuracy of a theory, to specify a research question, and to move forward the existing knowledge [14–17]. Hence, broadly speaking, SLR may serve as background for an empirical study (e.g. to identify a gap in the literature) [18] or stand-alone attempts [19]. Depending on the primary goal, the method of the SLR may vary [15]. Over the last decades, an increasing number of systematic reviews have been conducting about research on environmental topics in general [20–30] and landscape management in particular [31–34]. Yet, to the best of our knowledge, a systematic review of the visual impacts of surface mining activities upon the landscape has not been carried out, so far, as mentioned.

Within this context, this paper aims to fill this gap in the literature by conducting an SLR about the adverse effects of surface mines and quarries on the landscape quality, based on peer-reviewed scientific publications. More explicitly, the paper has three primary objectives, namely: (a) to analyse the scientific literature and identify the most important issues and the methods and tools implemented towards assessing the impacts of mining projects upon the landscape; (b) to conduct an analysis using descriptive of statistical methods and qualitative interpretation; and (c) to evaluate the state of knowledge on this particular topic and identify gaps in the literature, to suggest future research directions. The rest of the paper is organised as follows: Section 2 presents the methodology implemented to conduct the systematic literature review; Section 3 illustrates the results of the analysis; Section 4 discusses the main findings of the results, and Section 5 summarises the conclusions drawn by this work and suggests future research directions in the topic of interest.

#### Methodological approach

Several guidance documents on how to perform systematic reviews have been published for various science fields [12,14,15,35–40], including environmental sciences [14,39,41–43]. This review process in this work has been performed following the general steps of the guidelines for systematic reviews in environmental management [39,43], which have been adopted in similar applications [34].

The first step towards planning the review is to set the research question. The review is concerned about the impacts of surface mining on the landscape. For the review, the term 'landscape' is defined as "*a geographical area, characterised by its content of observable, natural and human-induced, landscape elements*", because this definition "*encompasses the physical content of areas without necessarily excluding human perception, and allow for a broad inter-disciplinary comparison among approaches*" [34]. In this context, the two main research questions addressed by the review are the following:

- a. What are the main landscape characteristics studied when assessing the impacts of mining projects upon the landscape?
- b. What are the main methods and tools implemented towards assessing the impacts of mining projects upon the landscape?

Besides the research questions, the review planning decided upon the data collection strategy (i.e. search strings and relevant databases to collect the appropriate information) and the inclusion or exclusion criteria.

As regards the data collection strategy, the terms used to search the title, abstract and keywords were: "visual impacts", "visual intrusion", "visual pollution", "chromatic contrast", "landscape alteration", "landscape change", "mining" and "quarrying". Further, the Scopus database was selected because it includes over 75 million records and more than 24,600 titles in the areas of science, technology, medicine, social sciences, art and humanities<sup>2</sup>. Scopus was preferred over Web of Science (WoS) because Scopus includes most of the journals indexed in WoS and has a larger number of exclusive journals than WoS in all fields [44]. The Google Scholar was not originally searched since the search target comprises peer-reviewed articles only and not in publications such as grey literature, presentations, keynotes, extended abstracts, etc.

The initial Scopus search process started with a broad scoping of articles related to the impacts of mining activities on the landscape, using the following string: TITLE-ABS-KEY ("visual impacts" OR "visual intrusion" OR "visual pollution" OR "chromatic contrast" OR "landscape alteration" OR "landscape change" AND mining OR quarrying). A total of 346 records was originally returned. These records were screened, according to the following criteria:

- a. Papers published over the past 30 years, i.e. 1990-2020
- b. Papers published in peer-reviewed scientific journals
- c. Papers published in English
- d. Papers applying or developing methods or tools for assessing the impacts of mining works on the landscape, including land-use/land-cover changes, topographic alteration, chromatic contrast, etc.
- e. Papers describing the application of visual impact assessment methods and tools on specific case studies

The first three filtering criteria were applied through the Scopus search. After removing publications before 1990, conference papers, book chapters, etc., and articles not written in English, the number of articles fulfilling the criteria for abstract reading were 194. After reading the article abstract, 43 publications were selected and downloaded for full-text screening. As shown in Fig. 1, 33 articles fulfilled all the criteria and were used in the analysis at the final stage. The studies are listed in Table 1.

<sup>&</sup>lt;sup>1</sup> https://www.elsevier.com/ data/assets/pdf file/0017/114533/Scopus GlobalResearch Factsheet2019 FINAL WEB.pdf

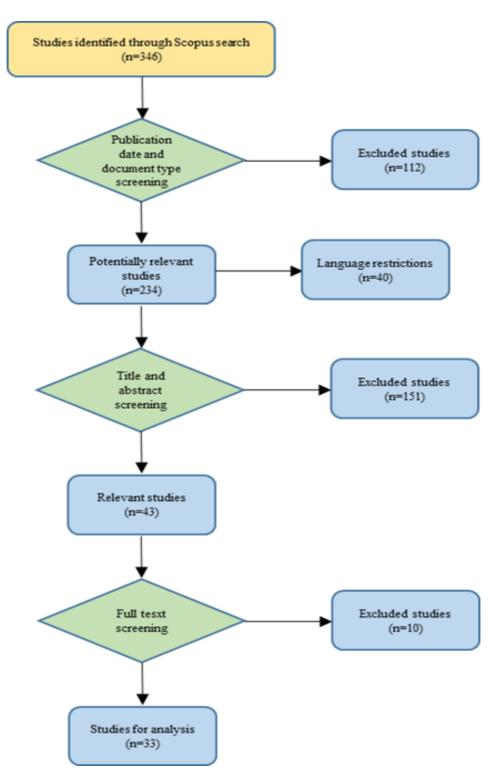


Fig. 1. Filtering of literature

Table 1. The dataset articles	Table	1. The	dataset	articles
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ID	Title	Reference
1	A methodology to evaluate the topographic visual alteration on surface mining	[45]
2	An analysis of 200-year-long changes in a landscape affected by large-scale surface coal mining: History;	[46]
	present and future	
3	Applicability of landscape metrics for the monitoring of landscape change: Issues of scale; resolution and	[47]
	interpretability	
4	Application of a multi-stage method to assess the landscape alteration induced by quarrying sites: A comparative analysis	[48]
5	Assessing the chromatic contrast in open surface excavations: a comparative study between subjective and quantitative approaches	[3]
6	Assessment of the visual impact of marble quarry expansion (1984-2000) on the landscape of Thasos island; NE Greece	[49]
7	Assessment of visual impact induced by surface mining with reference to a case study located in Sardinia (Italy)	[50]
8	Detecting landscape changes pre-and post surface coal mining in Indiana; USA	[51]
9	Dynamic changes in landscape pattern in a large-scale opencast coal mine area from 1986 to 2015: A	[52]
5	complex network approach	[32]
10	Evaluating mining landscape: A step forward	[2]
11	Exploring the perceived intrusion of mining into the landscape using the fuzzy cognitive mapping	[53]
	approach	
12	Exploring the visual impact from open pit mines applying eye movement analyses on mining landscape photographs	[54]
13	Fragmented landscapes of east Bokaro coalfields: A remote sensing based approach highlighting	[55]
	forestland dynamics	[00]
14	Functional differentiation of landscapes in the area of deep coal mining downsizing in the Ostrava region	[56]
15	Image analysis applied to quantitative evaluation of chromatic impact generated by open-pit quarries	[11]
	and mines	
16	Impact of gold mining on Middle Siberian taiga landscapes from Landsat 7 data	[57]
17	Impacts of coal mining subsidence on the surface landscape in Longkou city; Shandong Province of China	[58]
18	Landscape analysis as a tool for surface mining design	[10]
19	Landscape changes due to quarrying activities as a project parameter for urban planning	[9]
20	Landscape metrics for assessment of landscape destruction and rehabilitation	[59]
21	Landscape pattern changes at a county scale: A case study in Fengqiu; Henan Province; China from 1990 to 2013	[60]
22	Mapping the cumulative impacts of long-term mining disturbance and progressive rehabilitation on ecosystem services	[61]
23	Multitemporal aerial image analysis for the monitoring of the processes in the landscape affected by deep coal mining	[62]
24	Quantitative Assessment of Landscape Load Caused by Mining Activity	[63]
25	Setting rehabilitation priorities for abandoned mines of similar characteristics according to their visual impact: The case of Milos Island; Greece	[64]
26	Spatial distribution of the impact of surface mining on the landscape ecological health of semi-arid grasslands	[65]
27	The dynamics of landscape pattern changes in mining areas: The case study of the Adamów-Kozmin Lignite Basin	[8]
28	The mining landscape of the Ostrava-Karviná coalfield: Processes of landscape change from the 1830s to the beginning of the 21st century	[66]
29	Time-varying elevation change at the Centralia coal mine in Centralia; Washington (USA); constrained	[67]
30	with InSAR; ASTER; and optical imagery Visibility of surface mining and impact perception	[68]
30	Visibility of surface mining and impact perception Visual impact evaluation of mines and quarries: the updated Lvi method	[68]
31	Visual impact from quarrying activities: A case study for planning the residential development of	[69]
22	surrounding areas	[7]
33	Visual impact of quarrying in the Polish Carpathians	[7]

In order to conduct the analysis, eighteen variables were defined during the reading of the articles included in the final set. The variables are described in Table 2.

Variable	Description	Туре
Year of publication	Publication year of the article	Discrete
Country of origin	Country of the case studied	Nominal
Branches of science	The scientific discipline of the affiliation of the first author	Nominal
Journal	Title of the journal	Nominal
Method	'Tailor-made': methodology developed by the authors specifically for assessing visual impacts of mining projects; 'Landscape metrics': common indicators used to measure various landscape changes related to land cover/land uses; 'Other': e.g. questionnaire survey and EC decision 272	Nominal
Input	DEM (Digital Elevation Model) files, aerial or satellite images, photographs, topographic maps	Nominal
Tools	Computational tools or software used	Nominal
Mining activity	Type of exploitation such as quarry, metal mine, coal mine, etc.	Nominal
Spatial resolution	The scale addressed in the study, i.e. mine site, mine site and surroundings, and region	Nominal
Topographic alteration	The analysis accounts for topographic relief changes	Binary
Chromatic contrast	The analysis assesses impacts related to chromatic contrast	Binary
Texture/land cover change	The analysis measures changes in the texture or land cover of the landscape	Binary
Land-use change	The analysis measures changes in the land-use	Binary
Viewshed	The visibility analysis is based on the viewshed	Binary
Viewpoints	The visibility analysis is taken from specific viewpoints	Binary
Type of analysis	The analysis provides quantitative results	Binary
Total area	The total area studied in km <sup>2</sup>	Continuous
Time periods	Number of periods used to study landscape changes	Discrete

#### Table 2. Variables used for the analysis

Each study was abstracted and coded, using the variables presented in Table2. However, it was not possible to combine the results using meta-analysis because the included studies follow different methodological approaches and do not share common statistical measures. Therefore, besides simple descriptive statistics, a more qualitative discussion was followed to assess the main findings of the surveys and to compare the results. This process is also known as a qualitative systematic review [15] and refers to using a systematic review process to collect articles, and then a qualitative approach to assess them [71].

#### Results

#### Year of publication, journal title and affiliation

As illustrated in Fig, 2, only one paper was published between 1990 and 2000 by [45]. About one-third of the papers were published between 2000 and 2010, and the rest, i.e. around 65%, were published during the last ten years. Not surprisingly, almost one-fourth of the research studies have been published by scholars and faculty members from mining departments and another 20% from environmental and other engineering departments (Fig.3). Also, about 20% of the articles have been published by authors from departments of geography and environmental sciences, respectively. The rest of the authors of the papers are affiliated with earth sciences, forestry and natural resources departments. Concerning the affiliation of the authors, the papers are published mainly in mining, geography and environmental journals (Table 3). All the journals but the "International Journal of Mining; Reclamation and Environment" which publishes research on mining and environmental engineering, are interdisciplinary in character according to their mission statement. Further, only two of them, namely "Catena" and "Ecological indicators" include the term "landscape" in their statement mission.

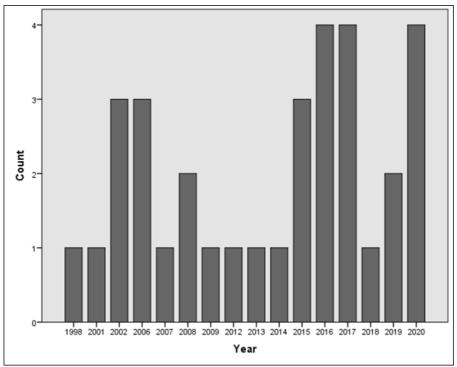


Fig.2. Number of articles published during the period under consideration

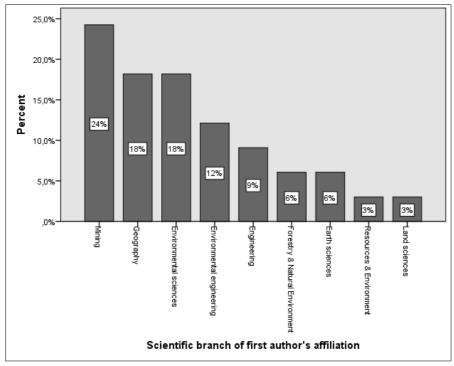


Fig.3. Percentage of articles published according to the first author's affiliation

Journal title	Frequency
Environmental Earth Sciences	3
International Journal of Mining; Reclamation and Environment	3
Applied Geography	2
Catena	2
Ecological Engineering	2
Ecological Indicators	2

Table 3. Titles of journals with two or more papers published

More than 70% of the papers have been published by authors affiliated with European universities (Fig.4) and more than half of them (i.e. about 40%) have been published by Greek and Italian scholars and faculty members. China follows with 12% (practically four studies) and the USA with 6% (two studies).

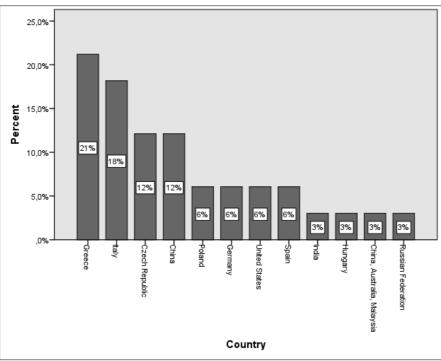


Fig.4. Percentage of articles published according to the authors' country of affiliation

# Study area characteristics

Towards examining the characteristics of the study area, two variables were used that classified the type of exploitation involved in the publication (e.g. quarry, coal mine, etc.) and the scale addressed in the study (i.e. mine site, mine site and surroundings, and region).

About 45.5% of the publications are related to landscape impacts caused by coal mining activities and about 35% from quarrying projects. The rest of the cases are referred wither to metal mines or surface mining, in general. Almost half of the research efforts evaluate the visual impacts of surface mining upon the landscape at a local scale, i.e. at the mine site and its adjacent surroundings, while the rest take into consideration larger regions.

#### Landscape characteristics, methods and tools used

As mentioned in Section 2, the two key research questions aim to identify the main landscape characteristics studied when assessing the impacts of mining projects upon the landscape and the methods and tools used for this purpose. These two issues are closely interrelated and, thus, they are discussed in an integrated manner. As regards the landscape impacts, four group variables were formed, i.e. "Topographic alteration", "Chromatic

contrast", "Texture/land cover change" and "Land use change". The analysis reveals that there is a clear distinction in the related literature. Almost half of the articles (i.e. 48.5%) study the alteration of the original topographic relief (24.4%) and the colour (24.4%) and the rest (i.e. 51.5%) are concerned with the land cover or land-use changes (practically these two terms are used interchangeably). Interestingly, there is no publication studying the topographic and chromatic alterations or land cover/land use changes induced by mining works. Only one publication by Quanyuan et al. [58] involves both elevation and land-use changes. Nevertheless, the differences in the original topography are only used to locate subsidence areas, which in turn are used to analyse the landscape impacts before and after the subsidence through changes in arable land, garden land, forest land, etc. Further, two publications do not fall into any of these categories. Misthos et al. [53] recruited a team of experts in mining and landscape engineering and developed a conceptual model of the "mining-landscapesociety" system using Fuzzy Cognitive Mapping (FCM). The model involves twelve mining-, socioeconomic- and landscape-driven factors affecting the perceived nuisance caused by mining projects on the landscape. The authors argue that the model can be helpful to policy-makers and mining practitioners, as it offers the ability to study the impact of each of the factors that give rise to differences in the degree of nuisance employing dynamic simulations or scenario analyses. Nevertheless, it does not provide evidence on the degree of landscape changes. Finally, Misthos et al. [54] used, for the very first time, an eye-tracking experiment to investigate viewing patterns and behaviours of mining landscape photographs. The research focused specifically on how the relative positioning and apparent size of open-pit mines attract the observer's visual attention. The findings of the survey suggest that the lower-left and the centre placement of the quarry in the photograph attract the attention of the viewer. Besides, they found that if the apparent size of the excavated surface decreases so does the clustered visual attention. According to the authors, however, further work is required to explore these issues, combining eye-tracking with subjective methods (e.g. questionnaire surveys).

Almost half of the studies use landscape metrics for assessing mainly changes related to texture, land cover and land uses evolution. About one-third of the studies have used a tailor-made methodology developed by the authors for the assessment of alterations in topography and of chromatic contrast between the mined land and the surrounding area. Further, three publications used the EC decision 272/02 [72] and, more specifically, the visual impact indicator (*x*), while the rest implemented different techniques, e.g. eye-tracking [54]. The analyses are based primarily on Digital Elevation Model (DEM) files (around 27%), satellite or aerial images (about 40%), and photographs (about 24%). Also, two studies have made use of available land cover databases and one study has involved experts through in-depth interviews. As regards the visibility analysis, five studies (15%) study landscape changes from certain observer viewpoints and eleven (33%) conduct a viewshed analysis to find the areas from which the mining works are visible. It is noted that the visibility analysis does not concern studies using landscape metrics. A more detailed analysis is provided hereinafter.

#### • <u>Topographic alteration</u>

In total, seven studies analyse the visual impacts of mining through the evaluation of the topographic alteration. Gutierrez Del Alamo & Chacon [45] developed a methodology to study topography before and after the excavation of the mine area from specific viewpoints using DEM files. In more detail, they evaluate the projections of the original and the altered landscape using three spherical coordinates, namely the real distance between the vertical angle between the observer's visual line with the horizontal plane and the horizontal angle between the observer's visual and the observer-alteration line. Further, they use the fourth indicator to account for the complexity of the contours. The variation of the four parameters for each observer is valued using four  $\chi^2$  functions and the estimated values can be used to estimate local and global appraisals. Menegaki & Kaliampakos [10] developed an approach to evaluate the destruction of the original topographic relief due to surface mining activities. The analysis is conducted via GIS and is based on DEM files representing the original contour and the final contour, i.e. the mine topography. The change in elevation and terrain is estimated via five indices that measure the correlation of the original and final contour, the vertical change, the slope difference and the aspect deviation. Later the authors expanded their methodology to assess how this change is perceived [2]. They proposed an approach named LETOPID, which measures: (i) the change in the topographic relief and (ii) the sensitivity of viewing conditions. Viewshed analysis is the tool used to estimate the viewing sensitivity considering the surrounding area, up to a distance of 8 km. The analysis considers the degree of excavation exposure, the distance of the observation point, the surrounding land uses and the observers' sensitivity. Quanyuan et al. [58] used DEM data to identify subsidence areas in a coal-mining region. Nevertheless, their primary goal was to analyse the influence of mining-related subsidence on the landscape and, thus, this study is further discussed in Section 3.3.3. Lippiello et al. [9] proposed an assessment procedure for the visual impacts of extractive activities, which is divided into three steps and takes into account: (a) the visible area of the quarry onto the plane view, which varies according to the position of the observer; (b) the surrounding area from which the quarried surface is visible, which is obtained graphically from polar diagrams; and (c) the critical angle above which the excavated area is no longer visible to the observer. The same approach was used by Lippiello et al. [48] in two different case studies. Prush & Lohman [67] studied elevation changes from coal mines using DEMs that were created by satellite and optical images. The proposed approach, however, does not provide any indicators or parameters to evaluate the degree of impact.

#### <u>Chromatic contrast</u>

Seven studies have applied quantitative approaches for evaluating the chromatic contrast generated by openpit excavations. Pinto et al. [11] developed an image treatment model for the evaluation of the visual contrast between the excavated area and the surrounding landscape. The analysis is based on digital photographs, which are processed with a replicable, objective and automatic procedure. A similar approach, namely the Lvi method, was proposed by Dentoni et al. [73] and was used in four studies which are included in the dataset [7,50,68,70]. The Lvi method uses digital photographs and image analysis software and estimates the chromatic contrast between the excavation and the surrounding area and the extent of the change in the natural landscape. The analysis is conducted from the most significant viewpoints. An updated version of the Lvi method was proposed quite recently by Dentoni et al. [69]. The updated version eliminates some drawbacks of the original method and improves the repeatability of the evaluation by incorporating two image segmentation algorithms. Menegaki et al. [3] made use of the CIEL\*a\*b\* colour space, to calculate the Euclidean distance between a quarry and its surrounding environment, using digital photographs and image analysis software. The authors tested three different calculation procedures. In the first procedure, the chromatic contrast is estimated between the rock exposed in the quarry face and the dominant landscape elements in the photograph (e.g. sky). In the second procedure, the chromatic contrast is estimated between the quarry and the immediately adjacent surrounding landscape. In the third procedure, the chromatic contrast is estimated between the quarry face and the foreground and the background landscape elements depicted in the image. To explore the effectiveness of each calculation process, a survey was conducted through personal interviews. The sample consisted of 200 undergraduate and postgraduate students, administrative staff and faculty members. The participants were shown a series of photographs and were asked to score the chromatic contrast between the quarry and its surrounding environment. The survey showed that the second approach was closer to the subjective estimates of the survey participants.

#### • <u>Texture, land cover and land use changes</u>

Almost half of the publications, as already mentioned, have studied the impacts of mining operations on the landscape measuring changes in the land cover or land uses through geometry-based landscape metrics. Herzog et al. [59] explored the landscape impacts induced by agricultural and surface mining activities (more specifically the Espenhain open coal mine) in a 75 km<sup>2</sup> study area in Saxony, eastern Germany. To this end, they created digital maps for four periods (i.e. 1912, 1944, 1973, 1989) and adopted eight landscape metrics (e.g. number of patches, mean size of patches, landscape shape index, Simpson's diversity index, etc.) to identify changes in natural landscape units using the FRAGSTATS software. Lausch & Herzog [47] also investigated the land-use changes in the Espenhain coal mine (as a test area) and in a 700 km<sup>2</sup> region in eastern Germany (i.e. Leipzig South). The investigation was conducted based on a time series of digital maps, namely 1912, 1944, 1973 and 1989 for the Espenhain mine, and 1990, 1994, 1996 and 2020 for the Leipzig South region, respectively. Using the FRAGSTATS software, 16 and 27 landscape metrics were calculated in total for the Espenhain area and Leipzig South region, accordingly. Kharuk et al. [57] used Landsat satellite images and field data to assess the impacts of gold mining on the Middle Siberian taiga landscapes. The images were analyzed using Erdas Imagine software and the ratio of the modified territory to the total area was introduced as a means to quantify the impacts of human activities, mining among them, on the landscape. Mouflis et al. (2008) [49] assessed the landscape impacts of marble quarrying on the island of Thasos, Greece, using two Landsat images for the years 1984 and 2000, respectively. The classification of the landscape elements was performed with Erdas Imagine software and the landscape metrics of quarry patches were estimated via the Patch Analyst extension. In addition to the landscape metrics, the authors conducted a viewshed analysis for the two time periods to quantify the amount of visibility load, i.e. the number of visible quarry perimeter pixels from each location of the island.

Yang [51] used three Landsat images for the years 1989, 2000 and 2006 to analyze land use/cover changes following reclamation on surface coal mines in southwestern Indiana, USA. The analysis was based on the "from-to" changes among five land use classes and several landscape metrics, calculated through FRAGSTATS.

Quanyuan et al. [58] combined DEM files and remote sensing images for five periods, i.e. 1978, 1984, 1996, 2000, and 2004, to assess the degree of landscape destruction due to subsidence phenomena in the coal mining area of Longkou in Shandong, China. Using GIS, four landscape metrics (i.e. largest patch index, landscape shape index, patch cohesion index, and distribution index) were calculated to analyze the landscape changes before and after subsidence. Hendrychová & Kabrna [46] examined landscape transformations in the North Bohemian Basin, Czech Republic, in a coal mining area of 228.48 km<sup>2</sup>. The analysis was carried out six different time periods (i.e. 1845, 1954, 1975, 1989, 2010 and 2050 based on planned future conditions). Twelve basic land use classes were identified (e.g. water bodies, forest lands, built-up areas, mining areas, etc.) and two specific indices, i.e the coefficient of ecological stability and the landscape diversity index, were analyzed in a GIS environment. Fan & Ding [60] used Landsat images for four periods (1990, 2002, 2009 and 2013) of Fengqiu County, China, to investigate landscape pattern changes and their driving forces. Five different land uses were identified (i.e. cultivated land, forest land, water bodies, settlements and mining sites and unused land). The landscape pattern changes between the time periods were analyzed utilizing five main landscape indices using FRAGSTATS and introduced the "Entropy model" to evaluate the whole land-use change.

Popelková & Mulková [62] used a series of aerial images combined with the EU CORINE Land Cover to study the landscape impacts of coal mining at the Ostrava-Karviná region, Czech Republic, in an area of 197.16 km<sup>2</sup>. The changes in the land cover were assessed using visual photointerpretation for seven different processes (i.e. urbanization, intensification of agriculture, afforestation, deforestation, flooding, abandonment and drainage) and three different time periods between 1947-2009. Using the same approach enriched by cadastral maps and historical aerial photographs, the authors extended the analysis over a longer period of time and evaluated temporal-spatial land cover changes for three time periods, i.e. 1836-1947, 1947-1971 and 1971-2009 [66].

Upgupta & Singh [55] studied land cover changes in the East Bokaro coalfield region, in India, using Landsat images of three time periods (i.e. 1972, 2001 and 2016). The analysis was performed for an area of 259 km<sup>2</sup> in a GIS environment using FRAGSTATS and, in total, twelve landscape metrics were calculated. Further, temporal changes for six different land cover classes, including mining, were analyzed. Csüllög et al. [63] assessed the landscape impacts of mining using a landscape load index as a proxy. The analysis was carried out in a GIS environment using a database of mining claims and deposits of mining waste. The proposed method offers the means to conduct comparative analyses between different areas about problems and landscape-use conflicts caused by mining. Nevertheless, it does not measure land use or land cover changes in absolute terms.

Fagiewicz & Łowicki [8] examined land-use changes in the Adamów-Koźmin Lignite Basin using maps and orthophotomaps. The landscape pattern analysis was carried out in a total area of 152.56 km<sup>2</sup>, for two periods, that is 1940 (the period preceding the lignite mining) and 2011. Seven land cover classes following the EU CORINE Land Cover and ten landscape metrics were considered.

Wang et al. [61] mapped and quantified changes in an area of 76.62 km<sup>2</sup> which is part of one of the largest surface coal mines of Australia, namely the Curragh mine. The study focused on land-cover changes associated with four ecosystem services (water yield, air quality regulation, soil conservation and carbon sequestration) in four time periods (1989, 1997, 2005 and 2013). Totally, 56 Landsat images were used between 1988 and 2015. Four

landscape metrics were analyzed in FRAGSTATS and topographic changes were studied using DEM files. Wu et al. [65] evaluated the impact of surface mining in Shengli surface mines, China, based on six Landsat images for the years 2002, 2005, 2008, 2011, 2014 and 2017. The methodology includes the development of a new algorithm, namely "the Modified Landscape Disturbance Index (MLDI)", which is calculated by three other measurements, i.e. the Landscape Disturbance Index, the Landscape Ecological Health and the Distance to the Surface Mining Landscape. Zhang et al. [52] analyzed land-use changes in an area of 517.48 km<sup>2</sup> at the Pingshuo coal mine region, based on remote sensing images for six periods (i.e. 1986, 1996, 2000, 2009, 2013 and 2015). Three first-level and eleven second-level land types were identified and two indices, namely the landscape change index and the patch level selection index, were calculated using ten and six measurements, respectively.

#### Impact

The review reveals a pattern of three substantially different groups of methodological approaches. The first group includes studies associated with changes in the topographic relief. With two exceptions [58,67], these studies involve computational processes and estimate quantitative indicators related to elevation changes between the original and altered landscape and the visibility of the mining operations. However, none of these approaches includes landscape elements nor they account for aesthetic, cultural or ecological values. The methodologies of [9], [45] and [48] conduct the analysis from specific viewpoints and, thus, are prone to subjective bias, while the methodology of [2] takes into account the total surrounding zone (up to a distance of 8 km). The second group of studies [3,7,11,50,68–70] focuses exclusively on the evaluation of the visual contrast between the mine/quarry face and the surrounding environment. All the proposed approaches are based on digital photographs processed with image analysis software. Although these methods suggest a replicable, objective and automatic procedure that is easy to implement, some limitations exist. The perceived visual contrast (and, consequently, the findings of the analysis) is affected by the atmospheric conditions and the season of the year [11,68] as well as by the diversity of landscape features, the distance from mountain ridges, the existence of other man-made activities, etc. [53]. In other words, there exists subjectivity (e.g. when

selecting the viewpoints from which the analysis shall be made), variability (e.g. due to the illumination conditions or the season of the year) and, most importantly, heterogeneity amongst the observers. So it is not surprising, as research efforts have shown [3], that only a small part of the observers' beliefs may be explained through objective methods. The third group of studies incorporates research efforts that emphasize the use of landscape metrics. The impacts of mining operations (mainly coal mining) are evaluated solely through the prism of land cover and/or land-use changes (these two terms are usually used interchangeably). Critical aspects of the visual impacts of mining, such as the destruction of the original topography and the visibility of the mining operations from the surrounding area are neglected. Only [58] evaluated topographic alterations but only as a means to analyze landscape changes in mine subsidence areas and [49] conducted a viewshed analysis to quantify the visibility load of quarrying without connecting, however, the two elements (i.e. the land-use change and the visibility) directly.

Also, a very clear connection is identified between these three groups and the scientific disciplines of the authors. More analytically, five out of seven publications in topographic alteration and six out of seven publications in chromatic contrast have been published by authors who work in engineering (mining or environmental) departments, while fifteen out of seventeen studies investigating land cover/land use changes via landscape metrics have been published by scholars who work in departments of geography and earth or environmental sciences. This trend is related not only to the scientific background of the authors but also to the scope of the research efforts. Scholars with engineering background aim mainly to come up with quantitative indicators that directly measure and compare variations in principal landscape characteristics, such as topography. This approach allows them to identify the most efficient mine exploitation or rehabilitation plans in terms of visual impact management and to design, if needed, appropriate preventive or mitigation measures. On the other hand, geographers and environmental scientists are primarily concerned with the environmental footprint of mining. Thus, they evaluate impacts related to spatial mosaics of interacting biophysical and socioeconomic landscape components. These approaches adopt a biophysical concept for the landscape and are closer to 'mapping' than 'assessment' [34]. They are combined with GIS-overlay techniques, which are widely used in biophysical approaches, and can be easily automated [74]. Yet, they are based on a priori selection of land use types and indicators that hinders subjectivity for the characterisation process.

Based on the above remarks, it becomes apparent that every method has advantages, disadvantages and constraints. Therefore, no single method is capable of integrating all dimensions of the landscape without compromises. Further, it could be argued that the existing methodological approaches are rather complementary than competitive and the selection of the method will depend on the characteristics, purposes and specific needs of each application. In this direction, future research efforts should put more emphasis on incorporating as many factors contributing to the visual impact of mining as possible towards developing holistic approaches.

#### Conclusions

Assessing the impacts of surface mining activities upon the landscape is far from being a simple and straightforward process because of the fuzzy, complex, subjective and multidimensional character of the landscape itself [53]. As Olwig et al. [75] mention: "...in any discussion of landscape characterization, the elephant

*in the room is the question of just what is landscape..."* and this raises the question of "...whether, under what conditions ..... and to what degree the presence of an open pit causes visual nuisance..." (cited by [53]).

The findings of this survey suggest that the main landscape characteristics considered when assessing the visual impacts of mining projects and the methods used for this purpose depend on their scientific rooting, just like in the case of methods used for landscape characterization and mapping [34]. Specifically, three different categories of landscape impacts were identified, namely "Topographic alterations", "Chromatic contrast", and "Texture/land cover or land-use change" and an equal number of approaches were recognized, i.e. (a) approaches aiming to quantify topographic alterations and the visibility of the mining operations; (b) approaches aiming to evaluate the chromatic contrast between the excavation and the surrounding landscape; and (c) approaches aiming to map landscape elements and to identify disturbances by mining activities using landscape metrics and statistical analyses. The first two categories are rooted in engineering disciplines and the latter is established primarily in disciplines of geography and ecology.

All things considered, future research efforts should incorporate as many factors as possible towards developing holistic approaches capable of evaluating the visual impacts of mining projects. In the same direction, they should consider establishing quantitative criteria and/or thresholds against which the visual impacts from mining projects can be rated.

# **Conflict of interest**

There are no conflicts to declare.

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#### DECOLORIZATION OF DYES FROM TEXTILE WASTEWATER USING BIOCHAR: A REVIEW

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#### Abstract

The textile industry is one of the largest in many low and middle-income countries, especially in Asia, second only to agriculture. Textile wastewater is discharged into the environment due to the lack of affordable and sustainable solutions to adsorb or remove the dye from the water. Biochar is generated by pyrolysis of organic material from plant waste in low-oxygen conditions, and is considered carbon-negative. Biochar for dye adsorption in textile wastewater effluent was proven to be highly effective. However, adsorption efficiency varies with experimental parameters, therefore there is a gap in application especially in small dye houses. Efforts should be made to find innovative and affordable solution to make the textile industry more sustainable, by developing methods for collection and reuse, recycle and upcycle of textile waste, by reducing the consumption of water, energy and chemicals and by developing methods for treatment of the textile wastewater.

#### Keywords

textile dye effluent; biochar; wastewater; sustainability; contamination; sorption

#### Introduction

Climate change, population growth, rising standards of living and uneven distribution of water are the main causes for competition over water resources, water scarcity, poor water quality and variability of hydrological events. Water is the core of sustainable development and unfortunately, water is not equally available and in many areas around the world clean water is out of reach [1]. The environmental stress on water bodies is evident in terms of not only quantity, but also quality. Globalization of industrialization has resulted in high pollution of water resources worldwide. The major industries responsible for pollution are the dyeing industries, paper industries, tanneries, metal-plating industry, mining operations, fertilizer industries, agricultural waste and pesticides. The demand for water in the industrial sector is expected to increase by 283% during the first half of the 21<sup>st</sup> century [2], resulting in increasing industrial wastewater discharge.

#### Discussion

The textile industry is one of the largest in many low and middle-income countries, especially in Asia [3]. The textile industry is one of the cornerstones in economies of many countries [4]. For example, the Indian textile industry according to the India Brand Equity Foundation (IBEF), it is the second largest industry, after agriculture, providing employment to over 45 million people directly and 60 million people indirectly, and it contributes 14% of the Indian total industrial production [5]. Humans are aesthetically interested in dyed textile, therefore the use of dyes in textile will not be abandoned. Different types of dyes are used in a variety of industries including the food industry, textile, tanneries, plastics and pharmaceuticals. Many products in those industries contain

several dyes from different chemical classes resulting in a complex wastewater [6]. Industrial dyes, in particular used in the textile industry, have complex molecular structures, synthetic in origin and recalcitrant [7].

Most of the chemicals are added in the dyeing process where a color is added to the dye baths, the fabric is immersed in the dye baths until the dye is fixed. In some cases, there is a need to add salt to the bath in addition to the color in the coloring processes to increases the affinity of the color to the fabric [8]. Over  $7 \times 10^5$  tons of synthetic dye are produced annually, with 10–15% of it not ending up in the final product [4] thus eventually further contaminating the environment.

Therefore the effluent also contains a large amount of recalcitrant unfixed dyes (as acid dyes, basic dyes, sulfur dyes, chrome dyes, optical/fluorescent brightener and azoic dyes) as the dyes are not totally fixed to the fiber of different textiles during the dyeing process (fibers as wool and nylon, cotton and viscose, polyester and acrylic). The textile industry requires a large amount of water for the production process, and is also one of the major producers of wastewater that can have carcinogenic and mutagenic compounds [9]. Wastewater from the textile industry is frequently discharged directly into lakes and rivers without any proper treatment and often these water sources are used by locals domestically [10,11]. Since the dyeing process uses a significant amount of water, recycling used in the dyeing process can conserve water, however it requires treatment whether recycled or discharged to the environment. Reference values for water reuse in textile industry, included COD between 60-80 mg/L, conductivity of 1000  $\mu$ S/cm and dissolved solids up to 500 mg/L [11]. In addition to dyes, the textile industry's wastewater contains also salts, acids and alkalis, oxidizers and recyclers, heavy metals, lubricating oils and fibers [11].

Dyes cannot be removed through conventional treatment unit operations due to the complex characteristics of the wastewater as high solubility, non-degradable nature, diversity and often changing speciation in water. When industrial wastewater is discharged into natural water bodies it can result in hazardous effects on the living systems because of the carcinogenic, mutagenic, allergenic and toxic nature of dyes [6]. This is a paradox as current conventional and advanced methods for the removal or degradation of persistent and emerging textile contaminants are limited, since they often involve intensive capital, lack of adaptive technological tools, social barriers and emphasis on centralized systems. Consequently, to close the gap, there is a dire need for innovative solutions and for widespread decentralized systems in the textile industry suitable for rural areas and capital-challenged countries.

There are many treatment methods when dealing with textile wastewater. The different techniques can be based on physical (sedimentation, filtration, floatation, coagulation, reverse osmosis, solvent extraction, adsorption, incineration, and distillation), chemical (neutralization, reduction, oxidation, catalysis, ion exchange, electrolysis) and biological (stabilization, aerated lagoons, trickling filters, activated sludge, anaerobic digestion) [12]. Physical methods are very common methods in textile wastewater treatment due to their high color removal efficiency, especially adsorption, filtration and membrane filtration. Other treatment processes as reverse osmosis, nanofiltration and multiple effect evaporators are effective but expensive, while the common treatment methods are non-destructive, lower in cost, time-consuming and less efficient [13].

Adsorption by porous materials is one of the most promising and affordable techniques for the removal of dissolved pollutants, serving as an alternative to energy-intensive technologies [14]. Activated carbon is a widely used adsorbent, but biochar, which is inexpensive, abundant and may have comparable adsorption capacity, can be used as an affordable alternative [15-17]. However, the efficiency, local manufacturing, availability and costs must be examined. Biochar is the result of low-temperature pyrolysis of carbon-rich biomass (from agricultural and forestry wastes) under low-oxygen conditions [18]. As can be seen in Fig. 1, there is an increase in the number of publications using the keywords: dye sorption, textile wastewater and biochar (by google scholar).

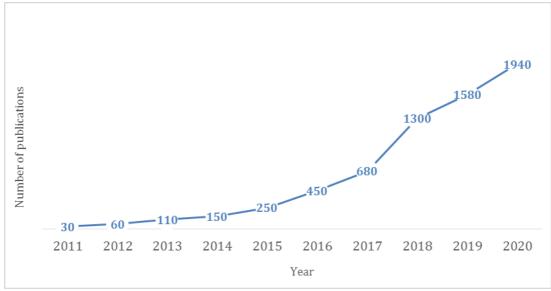


Fig. 1. Yearly number of publications using the keywords: dye sorption, textile wastewater and biochar *(by google scholar)* 

The biochar created is a stable carbon black solid that is highly porous with large surface area (Fig. 2.). More than 70 percent of its composition is carbon [19]. The biochar's chemical composition varies with feedstocks used to make it and methods used to heat the biomass. The pyrolysis process varies as it can be done with different conditions as burning temperature and burning time, reactor volume, other gasses and materials in the reactor [20]. However, different thermochemical processes can be also used for biochar production [18]. Compared to activated carbon, biochar can be created from various types of biomass, requires less energy in the production process and consequently can provide a solution in the treatment of textile wastewater in poor income countries.



Fig. 2. Biochar particles image (left) and scanning electron microscopy (right, taken at Tel-Aviv University)

The porosity and high surface area of biochar make it an excellent adsorbent of organic contaminants and heavy metals in wastewater [17]. This has led to a growing interest in using biochar for water treatment, although most research today focuses on its abilities for soil fertilization [15]. Adsorption of textile dyes was examined with various biochar types [15,21,22]. For each type of dye, the biochar type, process parameters (temperature, pH, agitation time) and wastewater quality influence the efficiency of the dye removal from the wastewater. Most studies have focused on the removal of pure, highly concentrated dye solutions that do not represent the actual effluent from real dye houses. Real effluent may contain lower dye concentrations and additional substances as previously mentioned, such as salts, detergents, solids, and fiber residuals, which have a tremendous effect on the biochar's sorption capacity.

There are numerous types of dyes and biochars – and therefore the biochar type and dose should match the dye type. The major anionic dyes are the acid and reactive dyes, and the most problematic ones are the brightly colored, water-soluble acid dyes [23]. Acid dyes are a sub-group of anionic dyes and are called so, because they are usually applied to the fibers in acid solutions [24]. Cationic dyes are dyes that can be dissociated into positively charged ions in the aqueous solution, and the cationic dye dyes the fiber through the binding of its cation ion [7,23]. Basic dye, are highly visible and have high brilliance and intensity of colors [25].

As mentioned above, the textile industry uses different colors, each with its own properties and characteristics. Moreover, the biochar itself can vary based on the initial biomass from which it is produced. In order to find the best biochar-dye combination, a summary of articles examining different combinations was made [26-43]. In these articles, the biochar efficiency in removing the dye from a mixture was calculated using mass balance equation for dye adsorption on biochar [37].

Table 1 summarizes different characteristics from 18 articles that present the use of biochar in treatment of textile wastewater [26-43]. The articles are catagorized to biochar properties, dye properties, experimental conditions, dye adsorption experiments compared to a common biochar and modified biochar. All the experiments presented were batch experiment in a controlled environment. From the table it is evident that there are some differences between the treatment requirements for cationic dyes and anionic dyes. The adsorption of the dye to the biochar depend on the characteristics of the solution, the type of the biochar and the conditions in which the experiment was performed.

One of the critical conditions is the solutions pH, where the adsorption of cationic dyes will be maximized when the pH of the solution is basic and anionic dyes in acidic pH, with preferred pH at 3-4. The pH of the solution has a significant effect on the interaction of the dyes with the biochar in the adsorption process. For example, the effects of the solutions pH were examined on the adsorption of three dyes, Methyl Orange, an anionic dye and Malachite green & Methylene blue, both cationic dyes and three types of biochar, each produced with a different solvent (acetone, ethanol and methanol) [30]. The adsorption of the cationic dye Malachite Green onto the biochar increased with the increase in pH. The solution pH influenced the adsorption of anionic dye Reactive Red 141 onto a Pecan Nutshell based biochar occured mostly in an acidic environment with a low pH [32]. Under acidic conditions, the biochar surface is positively charged whereas the Reactive Red 141 has several sulfonated groups, which are negatively charged that are attracted to the biochar surface, increasing the dye removal percentage.

The dye to biochar interaction depends on the temperature of the mixture, the interaction time, the steering speed and more. Almost all the experiments showed that a temperature of 25-30°C is the optimal temperature for the dye's adsorptions. A higher or lower temperature can affect the dye particles' velocity in the mixture and will reduce the adsorption [27]. This result is significant because it means the treatment will not require any major heating or cooling systems and the treatment can be effective in real wastewater. The interaction time is also a crucial parameter. With short interaction time, the dye will not be able to sufficiently interact with the biochar, while with long interaction time, the binding between the dye and the biochar could become loose, and the biochar can undergo modifications that can damage its functionality and even release material and pollutants to the water and cause more damage [27,30,34]. Each biochar-dye combination has its own optimal interaction time depending on the biochar and dye's structure and concentrations and on the other conditions regulating

the environment. Mubarak et. al. showed an increase in removal effecincy with contact time of methylene blue (cationic) and orange-G (anionic) onto Empty fruit bunch based biochar [36]. Another factor that can affect the interaction is the rotation speed of the mixture. A high rotation speed can lead to a high shear stress that can break the bindings between the biochar and the dye and can even damage the biochar particles and thus lower its efficiency. The most common rotation speed as presented in Table 1, is ~130-150 rpm.

Several studies attempted to increase an existing biochar activity by performing different modifications on it as Iron impregnation biochar, addition of cationic surfactant to the biochar surface, inserting magnetic formation to the biochar and more. These new characteristics provide the biochar with a stronger affinity and interaction between the biochar and the dye. One example of the benefits of these modifications used Rice Husk derived biochar mixed with a solution containing an aqueous phase reduction of ferrous iron (FeSO<sub>4</sub>×7H<sub>2</sub>O) called nZVI, resulting in attachment of nanoscale zero-valent iron particles to the biochar creating a modified biochar (nZVI/BC) to adsorb the anionic dye Methyl Orange. In this experiment, three types of biochar various theoretical mass ratios of nZVI/BC at 1:3, 1:5, and 1:7 [28]. The nZVI/BC 1:5 adsorbed almost 100% of the dye in the mixture compared to the other modified biochars that peaked at 90% and the non-modified biochar that reached only 10%. Although there may be an advantage in using modified biochar, there might be a problem after extended usage. For example, high concentrations of oxygen can convert the Fe<sub>0</sub> molecules into ferrous or ferric oxide leading to a passivation layer forming on the nZVI surface.

Table 1. A summary of articles presenting biochar source, postproduction modifications, dye type, optimal conditions and
experimental results

Biochar	Biochar	Postproduction	Dye	Optimal Condition					Results	Refere
Source	preparation	Modifications		рН	Temp <sup>(o</sup> C)	RPM	Contact time (min)			nce
Spent mushroom substrate (SMS)	Pyrolysis at 450℃ for 4 hours	200 cm <sup>3</sup> STP/min of N <sub>2</sub> was fed into the reactor, and steam was used as the activation agent at 800 °C for 2 h. Modification goals: Enhancement of the biochar's textural properties.	Congo Red (CR) (anionic) Crystal Violet (CV) (cationic)	CR:4 CV:6	30	150	CR:750 CV:1000	✓ ✓	Color and COD removal efficiencies up to 99.6% and 67.7% for CV. Color and COD removal efficiencies up to 10.3% and 23.7% for CR.	[41]
Rice husk (RHB) and Coir pith (CPB)	Pyrolysis at 700℃ for 5 hours	RHB or CPB were added to a solution containing Fe (NO <sub>3</sub> ) <sub>3</sub> ·9H <sub>2</sub> O dissolved in water. The mixture was oven dried at 105 °C for one day, followed by calcination at 500 °C for 4 h. Modification goals: Provide the biochar with oxidizing properties.	Acid Red 1 (AR1)	3	30-50	150	120	✓ ✓	Maximum dye removal for Fe- RHB was 97.6%. Maximum dye removal for Fe- CPB was 99.1%	[27]
Rice husk	Pyrolysis at 500°C	Biochar was mixed with HCl for demineralization then mixed with different mass ratios of nZVI.	Methyl orange (anionic)	4	25	-	15	✓ ✓	Maximum dye removal for nZVI at 5:1 was 98.5%. Removal capacity of 97.8,	[28]

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		Modification goals: increase dye adsorption by transforming it to low molecular weight products through destruction of its N=N bonds.						<ul> <li>✓ 306.7, 605.0, and 709.1 mg/g for initial concentrations of 60, 200, 400, and 600 mg/L, respectively.</li> </ul>	
Bael shell (BS)	Pyrolysis at 500°C for 3 hours	The biochar did not undergo any special modifications	Patent blue (PB) (Anionic)	2.7	-	110	60	<ul> <li>✓ Maximum dye removal was 74% (3.7 mg/g)</li> </ul>	[21]
Carboxym- ethyl cellulose (CM) from raw chicken manure	Pyrolysis at 600°C for 2 hours	The biochar did not undergo any special modifications	Methyl orange (anionic)	6.5	25	150	30	Almost 100% dye removal	[29]
Sewage sludge (SS) with acetone as the solvent	Liquefaction at 260-280 °C	The biochar did not undergo any special modifications	Methyl orange (MO) (anionic) Malachite green (MG) (cationic) Methylene blue (MB) (cationic)	7	30	150	60	<ul> <li>Acetone based solution gave 53.12% removal.</li> <li>The bio-chars were only effective on cationic MG and MB with removal of 10– 40 mg/g and 15–45 mg/g</li> </ul>	[30]
Eichhornia crassipes- molasses	Pyrolysis at 400°C for 5 hours	The biochar did not undergo any special modifications	Methylene blue (MB) (cationic)	8	25	125	30	<ul> <li>✓ Maximum adsorption capacity of 44.13 mg/g</li> </ul>	[31]
Pecan nutshell	Pyrolysis at 800°C for 1 hour	The biochar did not undergo any special modifications	Reactive Red 141 (RR-141) (anionic)	2-3	25	250	80	<ul> <li>✓ Increased initial dye concentration provided an increase adsorption from 40 to 130 mg/g</li> <li>✓ 85% dye removal. 80% of saturation was attained within 10 min</li> </ul>	[32]
Palm Kernel Shell (PKS)	Pyrolysis at 350oC in a rotary kiln for 20 min	The biochar did not undergo any special modifications	Crystal Violet (CV) (cationic)	6	25	100	30	<ul> <li>✓ maximum adsorption capacity of 24.45 mg/g</li> </ul>	[33]

Switchgrass Biochar modified by cationic surfactant (SB-TTAB)	Pyrolysis at 450°C for 20 minutes	Biochar added to a solution of tetradecyltrimethyl ammonium bromide (TTAB) in ethanol. Modification goals: binding a cationic surfactant to the biochar to form micelle like structures which can solubilize dye within this structure and increase the biochar capabilities.	Reactive Red (RR- 195A) (anionic)	5	25	150	40	<ul> <li>✓ Dye removal from different contaminated solutions: conc.10/30/50 mg/L)</li> <li>✓ Tap water %: 10 - 96.61</li> <li>30 - 98.82</li> <li>50 - 98.76</li> <li>✓ Raw water %:</li> <li>10 - 98.56</li> <li>30 - 97.79</li> <li>50 - 99.26</li> <li>✓ Wastewater %:</li> <li>10 - 100.00</li> <li>30 - 94.83</li> <li>50 - 94.24</li> <li>✓ Sea water %:</li> <li>10 - 92.96</li> <li>30 - 92.60</li> <li>50 - 90.98</li> </ul>	[34]
Kappaphy- cus alvarezii seaweed	Pyrolysis at 350°C for 2 hours	The biochar did not undergo any special modifications	Reactive blue 4 (RB4) (anionic) Reactive orange 16 (RO16) (anionic)	2-3	30	180	60	<ul> <li>Around 90% of total reactive dye sorption occurred within the first 60 min of contact</li> <li>uptake of 0.324 mmol/g for RB4</li> <li>uptake of 0.140 mmol/g for RO16</li> </ul>	[35]
Empty fruit bunch (EFB)	microwave at 800W for 30 min.	EFB particles were treated chemically by (FeCl <sub>3</sub> ) before pyrolysis. Flow of nitrogen gas provided iron oxide magnetite formation to the chemical treated EFB. Modification goals: biochar with magnetic features enable the dye to be separated by magnetic separation techniques	Methylene blue (MB) (cationic) Orange-G (OG) (anionic)	MB: Both 2&10 OG:2	25	120	120	<ul> <li>Maximum adsorption capacity of 96.68% (31.25 mg/g) for MB</li> <li>Maximum adsorption capacity of 90.76% (32.36) mg/g for Orange-G</li> </ul>	[36]
Empty fruit bunch (EFB)	Pyrolysis at 400°C	The biochar did not undergo any special modifications	Methylene blue (MB) (cationic)	-	30	150	250	<ul> <li>✓ Dye removal of 91%, 90%, 49% and 36% for 50, 100, 200 and 300 mg/L respectively</li> <li>✓ The EFB biochar has a maximum sorption of 55.25 mg/g.</li> </ul>	[37]

Spirulina	Pyrolysis at	The biochar did not	Congo red	2-7	30	120	15	✓	75-80% dye	[38]
platensis algae	450°C for 2 hours	undergo any special modifications	dye (CR) (anionic)	2-1	3	120		✓ ✓	removal For different initial dye concentrations. : 30,50,70,90,20 Omg/l	[20]
Chicken bones (CBB) after modificatio n-MCBB	Pyrolysis at 500°C for 2 hours	Powdered CBB subjected to co- precipitation with a mixture of Fe <sup>3+</sup> and Fe <sup>2+</sup> salts. The CBB was added into a solution containing FeSO <sub>4</sub> ×7H <sub>2</sub> O and FeCl <sub>3</sub> ×6H <sub>2</sub> O. Modification goals: biochar with magnetic properties for rapid sorption and a convenient recovery.	Rhodamin e-B (RB) (basic dye)	10	50	150	120	✓ ✓	88.5% dye removal for 40mgL-1. (36.2 mg/g) after 120 minutes. Approximately 96.5 mg/g of RB was adsorbed at pH 10 within 180 min and reduced to 68.5 mg/g in the presence of 0.5 g NaCl.	[39]
Pulp and paper sludge (PPS)	Pyrolysis at 108°C for 2 hours	PPS soaked in a FeCl <sub>3</sub> .6H <sub>2</sub> O solution and dried before being pyrolyzed. Modification goals: Reduce porosity and decrease in pore volume as a result of nanoparticle impregnation that will lead to a rapid dye diffusion into the active sites when the particles organically detach from the biochar into the solution	Methylene blue (MB) (cationic)	12	-	-	40	✓ ✓	Impregnating PPS with Fe <sub>2</sub> O <sub>3</sub> increased maximum adsorption capacity of the adsorbent by more than 50% saturation points for BC (97%) and NC (98%) both occurred at 5 g/L adsorbent The maximum adsorption capacities calculated 33 and 50 mg/g for BC and NC	[40]
Korean cabbage (KC)	Pyrolysis at 500°C for 1 hour	The biochar did not undergo any special modifications	Congo red (CR) (anionic) crystal violet (CV) (cationic)	CV:11 CR:7	30	150	1400	<b>v</b>	maximum adsorption values: CR on KC: 95.81mg/g CV on KC: 1304mg/g	[41]
Sugarcane bagasse (SCB)	Pyrolysis at 400,600,800 °C for 1 hour. Best results for the 800 °C biochar	The biochar did not undergo any special modifications	malachite green (MG) (anionic)	7.5	60	10,000	51.89	✓	100% removal of dye (conc.:500 mg/L) for SCB prepared at 800°C	[42,43]

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## Impact

The sustainability of the textile industry should be addressed across all sectors from fashion designers, manufacturers, product developers and the consumers. The process residuals as waste and wastewater generated requires innovative and affordable technologies and processes for collection and reuse, recycle and upcycle of textile waste (clothing and other textiles), for reducing the consumption of water, energy and chemicals and for treatment of the textile wastewater both in treatment plants and in small dye house industries. The environmental law for the dyeing effluent is in many cases very stringent and this necessitates the need for efficient treatment methods that must follow Zero Liquid Discharge (ZLD) either in common or in non-common treatment plants; however in practice due to the treatment costs it is not always practiced. In addition, even when plants are set for the ZLD, they are still generating hundred tons of hazardous solid waste per day as sludge (residual dyes and waste salts). Efforts should be made also on recovery of the dyes, and other organics from the wastewater before their discharge on to the soil and water bodies, in addition to efficient water treatment by combining novel hybrid membranes and nanotechnologies.

Tamil Nadu is in southern part of India and is engaged in textile processes especially the cotton textile industry. Real textile wastewater effluent from dye houses located in Coimbatore, Tamil Nadu, southern India, was examined for acid-dye removal from wastewater generated when dyeing silk filaments for production of soft silk sarees. The used dye solution from the dye houses is often discharged to the drainage or into the environment due to lack of affordable solutions. These dyes can potentially cause serious environmental damage and health. In our study, optimal conditions were demonstrated for filtration followed by high dye adsorption onto pine derived biochar (both in batch and column studies), and recommendations were suggested for reuse of the water back to the dye houses and for recovery of the biochar post use.

## Conclusions

Different types of biochar were effective in adsorption of dyes from the textile wastewater effluent. Parameters that affect the process are temperature, rotation and mixing speed of the biochar with the dye in batch tests, and reaction time. Another important parameter is the pH of the biochar-dye suspension. Basic environment (pH higher than 7) was proven to be ideal for cationic dyes, where acidic environment (pH below 7) was proven to be ideal for anionic and acid dyes, with the optimal pH being between 3-4. Different post-production modifications to the biochar can increase the efficiency of the adsorption process and thus improve the entire treatment process; however, the long-term use and reuse of the modified biochar should be monitored.

#### Conflict of interest '

There are no conflicts to declare'.

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#### THE ROLE OF BIOREFINERING RESEARCH IN THE DEVELOPMENT OF A MODERN BIOECONOMY

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#### Abstract

The current economy system is based in an intensive consumption of fossil fuels in a way that severely compromise future of the planet due to the severe consequences in climate change. In this scenario, the development of flexible and integrated biorefineries to produce biofuels and bioproducts from renewable biomass sources represent a key tool to perform the transition from a petroleum-based economy to a novel bioeconomy that looks for a more efficient and sustainable global development. This article analyses: the significance of biomass sources (such as agricultural and woody crops and residues, agro-food and wood processing industries residues and urban wastes) as feedstocks in the biorefinery, the most relevant biorefinering process technologies of the biochemical and thermochemical conversion pathways that are nowadays under development, and the need of further research and innovation effort to eventually achieve the commercialization and application in the market of the different biorefinery products.

#### Keywords

biomass; bioenergy; bioproduct; circular economy

#### Introduction

Bioeconomy is a relatively recent concept that arises as a reaction to the intensive development in the presentday society of an economy based on fossil fuels, in a way that severely compromises an efficient and sustainable use of natural resources. As a concept still under development, there is no consensus on the definition of a bioeconomy and several meanings can be found in the literature. For example, according to OECD [1], bioeconomy should involve three major elements: biotechnological knowledge, renewable biomass and integration across applications. In this definition, the second element refers to the use of renewable biomass and efficient bioprocesses to achieve sustainable production, with renewable feedstocks being obtained from primary biomass sources or wastes derived from household, industrial and agricultural activities. According to the European Commission, "bioeconomy comprises those parts of the economy that use renewable biological resources from land and sea – such as crops, forest, fish, animals, and micro-organisms – to produce food, feed, fibre, bio-based materials and energy", as stated in the European Bioeconomy Strategy launched in 2012 [2]. In the same line, the US Department of Energy [3] defines bioeconomy as "the global industrial transition of sustainably utilizing renewable aquatic and terrestrial biomass resources in energy, intermediate, and final products for economic, environmental, social, and national security benefits". In the opinion of the US National Academies of Sciences, Engineering and Medicine [4], a fundamental challenge in defining a bioeconomy is that it is not a single economic sector or grouping of sectors. A general consensus exists on bioeconomy contributes to the overall economy sustainability by implying a great number of industries and services in this new economic model.

It is an unquestionable fact that natural resources available for world's population are finite. The society is currently facing a series of environmental and social challenges such as the growth of world population, the climate change and the degradation of ecosystems that require the definition of new production and consumption pathways that are sustainable from the economic, social and environmental point of view. In this context, bioeconomy can provide a clear response by contributing to an equitable supply and distribution of food, the mitigation of climatic change effects and the reduction of the use of fossil fuels. Moreover, it can open new opportunities for economy development and employment.

The European Commission approved last year the European Green Deal Action Plan to face the challenges that the fulfilment of the goals assumed by world leaders in the 2030 Agenda for Sustainable Development involve [5]. The plan aims at making the EU's economy sustainable by turning climate and environmental challenges into

opportunities, and making the transition just and inclusive for all. As a key element of that plan, the EC has recently adopted a new Action Plan on Circular Economy [6] with a sustainable products initiative and a particular focus on resource intense sectors such as textiles, construction, electronics and plastics. The new Action Plan has the objective to adapt the economy to a sustainable future and reinforce competitiveness, while protecting the environment and granting new rights to the consumers. According to European Parliament's definition, the circular economy is "a model of production and consumption, which involves sharing, leasing, reusing, repairing, refurbishing and recycling existing materials and products as long as possible". In this way, the life cycle of products is extended. The circular economy in the context of bioeconomy is referred as circular bioeconomy, encompassing the fields of resources, byproducts and residues of organic nature (Fig. 1). Thus, the bioeconomy symbolizes the renewable segment of circular economy.

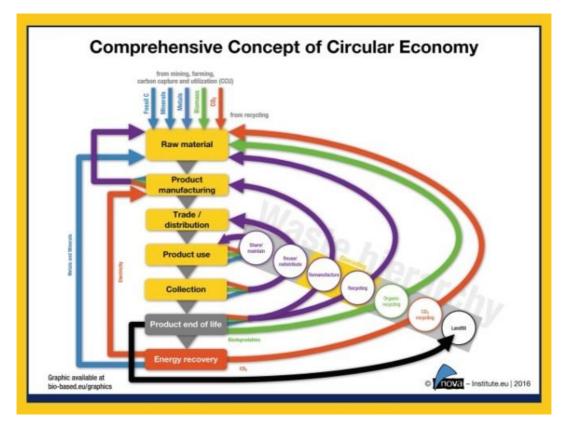


Fig. 1. Comprehensive Concept of Circular Economy. Source: nova-Institut GmbH, 2016-10-06.

One of the key enabling strategies of the circular economy is biorefinering, by closing loops of raw biomass materials (reutilization of residues from forestry, agro, processing and postconsumer activities), minerals, water and carbon. According to IEA (International Energy Agency) Bioenergy organization biorefining, "is an innovative and efficient approach to use available biomass resources for the synergistic coproduction of power, heat and biofuels alongside food and feed ingredients, pharmaceuticals, chemicals, materials, minerals and short-cyclic CO<sub>2</sub>". In the opinion of this organization, biorefining is the optimal strategy for large-scale sustainable use of biomass in the circular bioeconomy.

As a result of biorefinering, the production of food/feed constituents, biobased products and bioenergy will be cost-competitive while optimizing socio-economic and environmental impacts such as an efficient resources use, reduced GHG emissions, etc. Biorefinering strategy has been identified as a key element to boost emerging bioeconomy by offering a wide-range variety of products from an extensive spectrum of biomass sources to satisfy the different demands of the community [7]. In addition, biorefineries are key elements to support the development of a circular bioeconomy by connecting the stream and loops and allowing for the valorisation of multiple side-products.

The aim of the present article is to discuss and analyse the importance of biorefinering research in the development of a real bioeconomy. Firstly, the role of biomass resource as a pillar of bioeconomy development is highlighted. Secondly, relevant process technologies in biorefinering are discussed and examples of existing biorefinery applications are given. Finally, recent innovation advances in the different fields comprised in the biorefinery concept, which are expected to contribute to biorefinery deployment, are briefly examined.

#### Biomass in the core of the bioeconomy

Biomass is the renewable resource that constitutes the basis of bioeconomy development. Biomass features as constant supply, wide availability, easy accessibility and production potential make this renewable feedstock a unique material source for the production of energy, fuels and bioproducts. The relevance of biomass as enewable material can be illustrated by the significance of its contribution worldwide in terms of primary energy, which the International Renewable Energy Agency (IRENA) calculates could range from 97-147 EJ/Yr by 2030 [8]. About 38-45% of the total supply is estimated to come from agricultural residues and waste, the remaining supply potential being shared between energy crops and forest products, including forestry residues.

Biomass is a broad concept that has been described in many different ways, depending on the context. Thus, it is important clearly state the framework of the term used and avoid partial [9]. For instance, in the EU Directive 2018/2001 on the promotion of the use of energy from renewable sources, biomass is explained as "the biodegradable fraction of products, waste and residues from biological origin from agriculture, including vegetal and animal substances, from forestry and related industries, including fisheries and aquaculture, as well as the biodegradable fraction of waste, including industrial and municipal waste of biological origin" [10]. A more general description refers to "all organic matter existing in the biosphere, whether of plant or animal origin, as well as those materials obtained through their natural or artificial transformation" [11].

Biomass sources can be classified in relation to origin, chemical composition or biomass end use and consequently many comprehensive classifications exist [9, 12]. Relevant to the bioeconomy and biorefinery concepts, biomass feedstocks consist of current cultivated crops, such as cereals and oilseeds; dedicated biomass crops, such as woody crops; residues from agriculture (e.g. straw), agro-food or urban waste, lignocellulosic biomass from forest clean-up operations, and residues from the wood processing industry. All these biomass feedstocks have been already considered in a first approach for energy production as bioenergy is, nowadays, the main application for biomass resources worldwide. Nowadays bioeconomy offers many opportunities for integrating bioenergy applications within the biorefinery production models in order to optimize biomass resources use and maximize the generation of bio-based products. This idea is clearly supported by EU research programmes such as the Energy European Research Alliance (EERA) Bioenergy Joint Programme that, when defining the Strategic Research And Innovation Agenda (SIRA) [13], states that a "sustainable biomass feedstock must be developed within the broader framework of bioeconomy and circular economy concepts, where bioenergy is an essential part and may play a fundamental role in its deployment". Also, an interesting issue addressed in the SIRA of EERA is that exploring and making use of the potential of still underused, or even unused, biomass resources are also important aspects to satisfy the increasing future biomass demand. A good example of this type of biomass is biomass produced in shrub lands, which area in Europe has been estimated in 21 Mha [13].

Biomass feedstock, as the starting premise in the development of a bio-based economy, has a decisive role in determining the viability of any biomass processing activity, and the success will only be attained when sustainable supply of biomass can be guaranteed. To this end, it will be crucial to know what type of biomass will be necessary for each particular process application and in what quantity is needed, assessing the technical, economic and environmental viability linked to the biomass supply. Moreover, it will be necessary to identify in each case the current and future market needs to couple biomass demand and supply.

Several challenges in relation to biomass as feedstock for the production of biofuels and bioproducts have been recognised and research needs to face them identified. On the one hand, it is necessary to improve biomass productivity and yield and, for some specific biomasses, the yield of characteristic constituents (e.g. lipids, sugars, and proteins), always considering that these goals have to be achieved under sustainable conditions. To this end, it is crucial to better understand the biosynthesis and structure of the different types of biomass materials and study possibilities for engineering improved yields of polymers, oligomers, monomers and other biomass components [14]. On the other hand, aspects related to cultivation systems need to be studied to enhance yields, decrease feedstock costs and attain a sustainable biomass production. According to the EERA Bioenergy

Programme [13], a research priority in this area would be the design and optimization of innovative systems combining different crops, particularly for agricultural biomass feedstocks.

There are other issues related with biomass availability and supply that also need research effort to reach a successful bio-based economy. Thus, it is necessary to advance in the identification; quantification and geolocation of biomass sources with advanced technologies, such as satellite imaging or remote sensing, to provide a precise biomass inventory that allows an adequate planning of biomass use. Moreover, the development and application of standardized compositional analytical techniques that permit standardization of biomass materials quality, the creation of associated composition databases and the determination of the most adequate transformation pathway, appear as main challenges to tackle in the future. It is also crucial to address aspects related with the development of advanced tools to improve the current biomass logistic models and the optimization of biomass storage and transport systems, since all these aspects will definitely have a great impact in the development of cost efficient and sustainable biomass transformation processes.

Finally, as pointed out at the beginning of this section, the sustainability of integrated biomass production and supply chains systems is essential to secure a satisfactory bio-based economy development. To reach this ambitious goal, studies that sustain well-informed resolutions by landowners, communities, businesses and governments will be needed. A fundamental objective is to intensify the knowledge of the environmental, social and economic impacts of producing and using biomass within a broad bioeconomy scenario. Environmental impacts of biomass production involve water availability and quality, soil and air quality, biodiversity, and climate through the emissions of greenhouse gases and C sequestration in soils. All the different areas on biomass resource discussed above are closely related and the development of one of them also impacts on the others [14].

## Biorefinery: a main element in the framework of the emerging bioeconomy

Biorefinery is not a novel concept and examples of conventional biorefieneries exist for a long time. For instance, industrial development associated with food production and pulp and paper industry represent clear examples of biorefinery applications at commercial scale. However, a broader and more advanced conception has lately emerged around this concept and nowadays biorefineries constitute a key tool for bioeconomy development, aimed at providing a wide range of product portfolio from a broad spectrum of biomass resources to fulfill the different needs of the society [15]. In a wide sense, a biorefinery can be defined as this facility that using different biomass feedstocks generates an ample range of different products (chemicals, fuels and materials). It can be considered somehow analogous to the petrochemical refinery in a "bio" base.

Since the term biorefinery involves several industrial sectors, such as transport, chemical, energy, agricultural and forestry, it seems difficult to establish a single definition and there are different classification criteria for biorefineries proposed by several authors [16-18]. Most biorefinery classification approaches are developed using principles such as their technological implementation status (advanced vs conventional), the type of biomass feedstock used (sugar, starch, lignocellulose or oil containing crops, aquatic biomass and organic residues), the main conversion process pathway (mechanical/physical, chemical, biochemical, thermochemical), the main intermediates produced (C5 and C6 sugars, oil, biogas, syngas, lignin, etc.) that constitute the "platforms" or the products generated (energy or materials) [15, 18].

Due to the complexity of the biorefinery concept and the diversity of feedstocks/process/products applications, it is challenging to describe in short the precise status of each technological pathway. For instance, in the last decades an important focus in the development of biorefinery systems has been the production of energy products such as biofuels, power and heat, in the so-called energy-driven concept of biorefinery [7]. By contrast, the above cases of biorefineries associated to the pulp and mill and food industry would represent examples of product-driven biorefineries, in which the main objective is to generate a set of relatively high added-value bio-based products, in smaller quantities. Another interesting case of this type of biorefinery is the evolving bio-based polymers and plastics industry (e.g., vinyl polymers, polyesters, polyamides, polyurethanes and synthetic rubbers), which shows an enormous growth potential [19].

The energy-driven biorefinery approach, based in the generation of large volumes of relatively low-cost energy from biomass, has lately evolved to the intensification of bioproducts production associated to a main energy carrier, thus enhancing the profitability of the biomass use. Moreover, the development of integrated biorefineries producing energy, biofuels, chemicals and biomaterials will provide a higher flexibility against market fluctuations and society needs. In this context, the emerging concept of bioeconomy offers many

opportunities for integrating bioenergy technologies within the biorefinery production models in order to increase the efficiency, sustainability and viability of the full systems [13].

Within the different bioenergy technologies under development, those for the production of advanced biofuels such as liquid and gaseous biofuels for road transport, aviation and shipping (e.g. hydrogenated vegetable oil, cellulosic ethanol, Fisher Trop diesel, kerosene, alcohols, bio-methane, bio-CN and bio-LNG) represent a clear example of promising bioenergy applications. Supporting this statement, Wenger et al. [15], as a result of a large literature review on biorefineries research work, have recently claimed that biofuels drive the current biorefinery research. While conventional or first-generation biofuels are produced in biorefineries using food crops as feedstocks by mature and well-developed technologies, the second-generation or advanced biofuels production processes use non-edible biomass, such as dedicated energy crops, lignocellulosic residues and wastes as raw materials, and they are still not fully commercial. The challenge is to enlarge the utilization of these alternative biomass resources in a biorefinery based context to develop real advanced and integrated biorefineries, in which biofuels are produced together with a wide range of bioproducts for different markets. An interesting and promising approach to put into effect, in the context of biorefineries integration, is the combination of first- and second-generation (1G&2G) bioethanol technologies by sharing the existing infrastructure and thus increasing the potential for energy optimization and products cost reduction [7]. The objective is to improve the process efficiency and economics of the biomass conversion and to get better economic and environmental performances than with standalone 2G facilities. For instance, corn ethanol industry has recently evaluated the potential of using corn kernel fiber to produce ethanol in existing corn mill ethanol plants in the so-called 1.5G technology and several process strategies are being developed [20]. For instance, the technology company D3MAX, formed by the Public-Private Partnership Bio-based Industry Initiative (BBI, https://www.bbi-europe.eu/about/about-bbi), has developed a process to convert corn fiber and residual starch generated in a corn dry mill ethanol plant into cellulosic ethanol. Figure 2 shows a schematic diagram describing the main stages of this technology, which combines conventional starch and cellulosic ethanol conversion processes [21]. The company D3MAX claims that this technology raises the overall ethanol yield of the plant by 11% and is advancing in the licensing and implementation in EEUU [22]. Another important advance in biorefinery integration is the case of sugarcane integrated biorefinery for the production of bioethanol and energy from sugarcane juice and bagasse, which has been demonstrated to have a high potential of economic viability [23]. The important role that the existing industrial infrastructure plays in the development of integrated biorefineries is highlighted by Hinsgsamer and Jungmeier [7], who state that the analysis of the possibilities for that integration will provide support to investors and decision-makers to invest and integrate resource-efficient biomass uses in a context of bioeconomy development. This opinion is also shared by Wenger and Stern [15], who assert that present agro and forest related industries will substantially contribute to make biorefineries a reality.

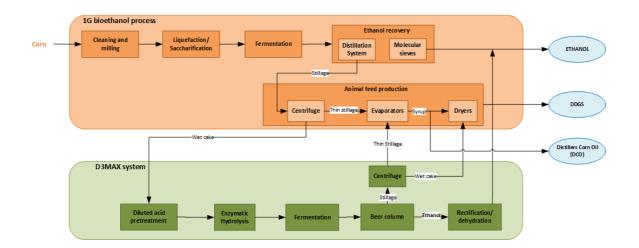


Fig. 2. Schematic diagram of D3MAX technology. Source: [21]

In relation to the biorefineries technologies currently under development for the production of advanced biofuels, they comprise, in a general sorting, thermo-chemical and biochemical processes. Without any doubt, a future perspective of advanced biorefinery systems must consider the combination of one or more of the different approaches included in these two large groups. Thermochemical biomass conversion processes can be generally classified in torrefaction, pyrolysis, gasification and hydrothermal liquefaction, while biochemical pathways are based on the use of microorganisms and enzymes for biomass conversion to chemicals, materials or fuels. Nowadays, different bioenergy/biorefinery technologies are at various stages of development and so, it is difficult to identify the most promising ones. Many features such as i.e., feedstock availability and cost, technology readiness level (TRL), products/intermediates, and situation of emerging or existing markets for the products, among others, influence the real prospects for each one.

Regarding the status and main bottlenecks and barriers of each biorefinery conversion route, several recent papers and reports carried out by relevant organizations in the field have been published and research roadmaps have been defined [13, 14, 24]. It would be difficult to summarize herein this relevant information, so that to illustrate the progress achieved in the development of biorefinery systems and discuss the need for further research effort in this field, it may be interesting to focus on one of the most developed biorefinery systems so far for the production of advanced biofuels, that of lignocellulose-based biorefineries (LB). LBs use biochemical or thermochemical conversion routes, or a combination of both, to generate bioethanol, electricity, heat and bio-based chemicals using biomass from dedicated lignocellulosic crops or lignocellulosic residues.

LBs have been extensively investigated in the last decades and significant progress has been achieved in this field. Particularly in the case of cellulosic ethanol produced by biochemical conversion, the technological advances permitted in the past decade the construction of several pioneer facilities of > 10 million gallons/year, turning cellulosic ethanol into the cellulosic biofuel deployed on the largest scale. [25]. Unfortunately, the global financial crisis started in 2008, the collapse in oil prices in 2014 and some drawbacks related to the expected anticipated technological readiness have somehow levelled off the prospects foreseen for this biofuel [26]. However, in spite of these "globalized" reasons, it is also acknowledged that still key technology-driven challenges need to be overcome to reach a cost- effective production. According to Lynd [26], strong investment in R&D focused on innovation and novel processing models is required to achieve financial viability of cellulosic biofuels, including cellulosic ethanol.

The technical assessment and further development of innovative routes involves testing and demonstration, sustained by R&D&I, to demonstrate their effectiveness and contribute to eventually achieve the commercialization and application in the market. This scientific and technical effort will contribute to advance towards a knowledge-driven bioeconomy in the future and result in the development of new markets and competitiveness in different bioeconomy sectors.

#### Innovation on lignocellulose biorefinering

On the one hand, as biomass feedstock constitutes the starting point of all downstream processes in biorefinery, research and innovation in the field of primary biomass production has been identified as one of the key pillars to develop a feasible sustainable biorefinery-driven bioeconomy [27]. Research efforts need to be directed to the improvement of biomass yields per unit of land, while minimising negative environmental impacts through the optimization of water and nutrients efficiency. To this end, it appears crucial to advance in the knowledge of the metabolism, biochemistry and structure of the plant that will pave the way for the development of new varieties with enhanced processing characteristics. Advanced genetic engineering tools, as well as modern breeding techniques, show enormous potential to maximize biomass productivity and develop novel varieties suited to specific environments.

Specifically for the lignocellulosic biomass feedstocks, understanding the structure and interactions of the main components of plant cell wall (i.e., cellulose, hemicellulose and lignin), will help to develop and design lignocellulose materials that enable improved and cost-effective fractionation processes, aimed at extracting the largest possible quantity of valuable material. For instance, a modified lignocellulosic biomass composition with increased cellulose content and reduced lignin content is an interesting approach to enhance biomass properties, which in turn can imply a major impact on processing costs by, for example, decreasing the energy needed for pre-treatment and the enzyme requirements for downstream processing [13]. However, although this approach

appears to be a good strategy to overcome biomass structure recalcitrance and it has been explored [27, 28], the molecular mechanisms regulating biomass components biosynthesis are not fully understood yet and further research progress is still required in this field.

On the other hand, lignocellulose biorefining research is needed to introduce innovations involving upstream and downstream technologies in both thermochemical and biochemical conversion pathways to enhance profitability, diminish risks and enlarge the value of co-products and biobased products (26).

In the development of thermochemical production systems, general challenges that hinder commercial deployment such as the lack of large scale demonstration of the technologies, comprising economic, technical and social aspects, and the quality of the end products, have been identified [14]. Specific research needs for each technology pathway are well presented in the document drawn up in the framework of the EERA Bioenergy Joint Programme, aimed at increasing the efficiency, sustainability and cost-competitive production of advanced biofuels and bioenergy carriers from biomass through thermochemical processing [13]. For example, in gasification, the most mature thermochemical process, the research topics identified are focused in broad terms on increasing feedstock flexibility by utilizing low-cost materials, improving gasifier performance, optimising product gas composition for downstream processing and developing innovative gasification processes such as molten bed gasification, reforming gasification and thermal and cold plasma gasification.

In the biochemical conversion pathway, based on the action of enzymes and fermentation processes using microorganisms, also general challenges and more specific R&D needs have been extensively analysed by different research groups and scientific organizations [13,14,24,27,29]. As a first and essential step in biochemical processes, pretreatment for deconstruction and fractionation of biomass components needs further research and innovation work leading to improve the energy and water use efficiencies of the existing methodologies and develop new flexible processes capable of enhancing biological conversion effectiveness and product yield. Advanced and economically feasible extraction methods constitute an important goal in the future to produce new materials and chemicals [14]. Intensive research in process integration is also needed and innovative solutions are necessary for product purification. Moreover, the recovery of process side streams to obtain high value co-products has been highlighted as capital to achieve higher energy efficiencies and cost competitiveness for biofuel production in biorefinery schemes, so boosting the development of a circular economy. An example is the potential revalorization of the sugars and lignin streams generated in forest related- lignocellulosic raw material processing and pretreatment, as feedstocks to obtain a series of high-added value chemicals such as 5-HMF, levulinic acid, furfurals, sugar alcohols, succinic acid, lactic acid and aromatics [30].

Another research and innovation priority in biochemical conversion concerns the search for new biocatalysts and microbial strains with improved features (tolerance, productivity and substrate range) for the development of advanced biochemical processes in biorefineries. Innovation in this field will come from the application of high-throughput screening and metagenomics techniques, genetic engineering, fermentation and biochemical reaction science, as well as the optimization of existing biotechnology tools to develop improved biocatalytic processes and fermentation strategies [13, 14]. Synthetic biology will enable the production of new biocatalysts with improved performance under industrial conditions, while the development of new microbial systems by introducing novel metabolic pathways appears as a very promising tool to reduce the steps or units required in a biological process-based biorefinery. A good example is this last approach for the conversion of lignocellulosic biomass directly into biofuels is the development of Consolidated Bioprocessing, where a single microorganism features enzyme production, hydrolysis of polysaccharides, and fermentation of sugars within a single-unit operation, thus contributing to achieve a cost-efficient production of biomass-based fuels and chemicals [13].

Furthermore, it is important to highlight that to promote the biorefinery application potential in a future bioeconomy scenario, technology and full chain development of multi-stakeholder consortia is still necessary, according to the Biorefinering working group of the IEA [24]. The experts claim that joint international priorities and R&D&I programmes between industry, research institutes, universities, governmental bodies and NGOs are needed to boost innovation in biorefinering. Moreover, and as a result of the evolving character of bioeconomy development, an important issue to contemplate is that further growth in the biorefinery-based bio-economy will depend on the creation and expansion of markets for new co-products [20].

Finally, sustainability assessment of the whole value chain is needed to get an environmentally robust biorefinery-based bioeconomy. It must consider the economic, environmental and societal effects, as well as aspects as nutrient cycles, water management and food-feed-fuel-competition. To perform this analysis, suitable

and practical assessment tools for an integrated sustainability evaluation are required [14]. To this respect, Life Cycle Analysis is a well-recognized and developed methodology that can be utilized to assess the global environmental performance of a product or process throughout its whole life cycle in a biorefinery, considering the economic, environmental and social impacts. Also, for a successful introduction of these new biorefinery-based products, it is essential to get consumers to have a positive perception of the benefits inherent to the biofuels and bioproducts generated in a biorefinery, as a part of a successful change to a real bioeconomy. Besides supporting policies to promote biorefineries from governmental bodies, it will be necessary to perform information and awareness campaigns to make the society understand that an important added-value of biorefinery-derived bioproducts and biofuels is the generation of valuable benefits from a socioeconomic and environmental point of view [31].

#### Impact

The use of biomass feedstocks to generate biofuels, bioproducts and bioenergy in a biorefinery concept will have a great social and economic impact at regional level through maximizing local resources to promote industry development, generate added-value and create employment, which in turn will result in the structuring of the territory. Added-value will be created through the whole value chain: from the biomass resource utilization (agricultural or industrial residues upgrading, development of biotechnology tools for plant breeding, new biomass dedicated crops, etc.), to the processes to produce bioenergy and bioproducts (advanced conversion technologies, use of process residual streams to generate new products in a circular economy, etc.). It means that apart from the value provided by the final product generated (bioenergy/bioproduct); the whole biomass to product productive process will be carried out contributing to connected sectors, i.e., agricultural and industrial sectors.

Finally and from a global perspective, the development of a bio-based economy will definitely help to face the energy, environmental and social challenges that a modern economy that assumes the objectives for a sustainable development (climate change mitigation, energy security, rural development, reindustrialization, poverty alleviation, etc.) involves.

#### Conclusions

Biorefieneries are a key pillar in the development of a future bioeconomy-based society. The conception of generating a large range of biofuels and bioproducts from a vast variety of biomass sources within a flexible and integrated biorefinery, opens the possibility to develop new and more sustainable processes and products that will eventually drive the transition from the current scenario of a mostly petroleum-based economy to a real bioeconomy. Significant progress has been achieved in the last decades in the assessment of biomass feedstocks production and supply as well as in the development of new and optimized technologies for the conversion of biomass materials into bio-based commodities through different technologies. Although some of the technologies have reached a development level very close to full commercialization, there is still a need of further research and innovation effort to overcome the drawbacks and barriers that all technologies still face. Finally, it is crucial to continue improving and developing tools to assure the sustainability of the full value chain, with special emphasis on the assessment of the socio-economic impacts that the biorefinery-based biofuels and bioproducts will have in the society.

#### **Conflict of interest**

There are no conflicts to declare.

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## POTENTIAL CONTRIBUTION OF NANOTECHNOLOGY TO THE CIRCULAR ECONOMY OF PLASTIC MATERIALS

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## Abstract

The problem of plastic accumulation in the environment requires the development of effective strategies to shift the paradigm of used plastics from wastes to resources. In the present contribution, after an overview of the current plastic management strategies, the possible role of nanotechnology to this emerging field is considered. In particular, the challenges related to the use of nano-additives to improve the properties of recycled plastics is discussed based on the fundamental aspects of colloid stabilisation. Finally, the contribution of nanotechnology to the fabrication of effective catalysts for the depolymerisation of plastics into the constituent monomers is outlined.

#### Keywords

nanoparticles; inorganic pigments; polymers; nanocomposites; packaging

#### Introduction

The images of the Great Pacific Garbage Patch have become the symbol of plastic pollution worldwide. An island of floating trash with an area three times larger than France and more than twice the size of Texas is located between California and Hawaii and contains around 79,000 tons of plastic debris [1]. The durability and low cost of plastic has made it a widespread material in everyday life, both in consumer and industrial products. Only packaging and single-use materials constitute 6.3 billion metric tons of plastic generated worldwide [2]. It has been estimated that between 4 and 12 billion of metric tons of plastics enters the ocean annually [2,3]. Traditional plastics do not undergo biodegradation, especially in the oceans, rather sunlight-promoted size reduction processes lead to the formation of so-called microplastics. These are tiny fragments of particles smaller than 5 mm, that due to their small size can easily be ingested by marine living organisms, and enter the biological nutrient cycle. Microplastics have been found in drinking water, kitchen salt and honey, through largely not yet understood mechanisms; remarkably, the effects of microplastics on the human health is not known [4]. Consequently, the European Community has set the ambitious target of ensuring that all plastic packaging is recyclable by 2030 under the roof of the circular economy [4,5] According to this concept, the traditional linear consumption model should be substituted by closed loops, where each product at the end of its life cycle can be re-transformed in the original product or in another useful one. In other words, the design of sustainable products shifted from the cradle-to-grave to cradle-to-cradle model, where ideally every material can be recycled infinite number of times [5].

Before analysing the possible contribution of nanotechnology to the field of circular economy of plastic materials, it is worth to chart a course of the current plastic management strategies.

Currently, plastic wastes undergo three main paths: landfill disposal, incineration and mechanical recycling. The landfill disposal has been recognised as less ideal solution for the waste management, in general, and more specifically for plastics. The accumulation of large amount of non-degradable plastic wastes over a limited area poses serious problems connected with the security of the sites and represents an economical loss of value products with a very short lifetime.

Incineration enables energy recovery from plastic wastes and has certainly some advantages with respect to landfill disposal, such as avoiding littering, leakages and accumulation in landfill. The value of only packaging plastics disposed in landfill and incineration has been estimated between 70 and 105 billion euro that are annually lost [4]. On the other hand, plastic materials are nowadays mainly synthesized starting from fossil fuels, therefore, their incineration to recover energy leads also to the formation of carbon dioxide, besides other gases,

contributing remarkably to the greenhouse gas (GHG) emissions. With the Paris Agreement, the European Community has committed to reduce the GHG emission by 2030 and become carbon neutral by 2050 [6,7]. To meet the challenge of GHG abatement and reduce the amount of plastic waste, the urge to move in the direction of a circular economy arises.

The third strategy of mechanical recycling of polymers is a viable route to reuse plastics over several cycles [8]. Following this approach, plastic materials at the end of their life cycles are envisaged not as wastes but as potential source of valuable products with properties (mechanical, optical) comparable to the pristine material. The mechanical recycling of polymeric materials appeared so far to be a feasible route from an operational and economical perspective. However, this route is at the same time an extremely challenging task: the presence of contaminants, including other polymer types, and the thermo-mechanical degradation of the polymers during the processing affect negatively the properties of the recycled material with respect to the original one. In fact, throughout the process chain from milling, melting and extrusion the properties of the polymeric materials are degrading due to the occurrence of physico-chemical phenomena such as thermo-mechanical oxidation and polymer chain scission under the action of radical formation, or upon hydrolysis. Consequently, a loss of the quality of the original products is observed.

A general strategy undertaken to compensate the loss in mechanical and optical properties of the recycled polymer is to mix it with virgin polymer in suitable proportions, usually not exceeding 10 wt. %. In this way, the new products introduced in the market contain a (relatively small) portion of recycled plastics and match the benchmark properties.

The challenges for the mechanical recycling of plastics are even more severe when the recycling of composite materials is targeted. Composite materials comprise several thin layers of structures closely connected with each other. As an example, aluminium is present in many packaging materials as a thin layer. Considering that bauxite, the mineral from which aluminium is extracted, has been recently enclosed in the list of critical raw materials from the EU community [9], the topic of composite recycling has become even more fundamental. The recycling of packaging materials accounts for only 9 % of the plastic generated worldwide in 2015 [4]. The co-presence of several polymeric materials, various coatings and adhesives, along with additives such as substances used during manufacturing (solvents and non-intentional impurities), oligomers and degradation products are among the technical reasons that limit to overcome this threshold [10].

Besides these traditional approaches of waste management, alternative routes are under developments. In a first instance, several efforts have been undertaken for the development of plastics that are produced from renewable raw materials and are, thus, classified as biobased plastics. Some relevant examples are poly-lactic acid (PLA) and poly-hydroxybutyrate (PHB), both synthesised from the fermentation of starch or glucose. Despite their promising biocompatible properties, these materials have not substituted the traditional plastic materials due to two major pitfalls: the competition with land for producing "crops for plastic" and the fact that the degradation process requires suitable experimental conditions (temperature, amount of oxygen, moisture, microorganisms) achievable only in dedicated composting plants. Therefore, if released into the environment they will not undergo biodegradation. Thus, it will be central in the near future to develop environmentally friendly materials, ideally able to degrade promptly and completely in non-harmful products under natural environmental conditions [11,12].

More recently, the concept of chemical recycling of polymeric materials has emerged as an intriguing approach to transform the polymeric materials into the corresponding monomers and/or into useful products for the chemical industry. In turns, the monomers can be reused to synthesise the virgin polymer with the same characteristics as the original starting material. The co-products besides the monomer shall be useful components to be fed into the market or to be used as chemical feedstock for further transformations in high added-value products. The guiding principle is therefore the atom economy: every atom that constitutes the waste material and the chemical educts used for their transformation into monomers shall be incorporated into useful chemicals, following the principle of green chemistry [13,14]. Therefore, the challenge is the design of materials and processes that enable the recycling of plastics, preserving their properties. While this approach is certainly promising, it requires an intensive research and development effort, especially for the identification of efficient, benign and recyclable catalysts able to realize the cradle-to-cradle transformation of plastic materials [13].

Table 1 summarizes the current strategies of plastic management and highlights advantages and disadvantages of each approach.

Approach	Short description	Advantages	Disadvantages		
Landfill	Wastes are accumulated	Release in the	High costs, risks management is		
disposal	in suitable locations	environment is avoided	required, polymers do not degrade		
			spontaneously		
Incineration	Plastic materials are	Energy production	CO <sub>2</sub> and other GHGs are generated,		
	combusted		hazardous emissions, dependency		
			on fossil fuels		
Mechanical	Comminution, melting	Polymers are reusable	Loss of the mechanical and optical		
recycling	and extrusion lead to	over a certain number of	properties of materials		
	new polymer	cycles			
Biodegradable	Materials derived from	Good potential to GHGs	Requires well-controlled conditions		
polymers	biomass	abatement	for the degradation		
Chemical	Depolymerisation	The properties of the	Under development, requires		
recycling	reactions are carried out	final polymer are	efficient, cheap and reusable		
	to obtain the monomers	maintained	catalyst for the large scale		
	and useful co-products		application		

Table 1. Advantages and disadvantages of current approaches in plastic management

In the end, the problem of plastic management remains and will most probably continue to be a scientific. economic and technological challenge also in the next decades. Besides an undeniable need to reduce the plastic consumption and its release into the environment, effective routes have to be developed to change the paradigm of plastic materials from wastes to resources.

In the current paper, the possible contribution of nanotechnology to the field of circular economy of plastic materials will be outlined. Namely, the use of nano-additives to improve the mechanical and optical properties of recycled materials and the development of nanocatalysts for the depolymerisation of plastics will be considered. Emphasis is given to the challenges related to the use of nanomaterials as plastic additives, considering the intrinsic thermodynamic instability of nanoparticles. Therefore, before considering the application of nanoparticles in this emerging field, it is worth to consider some fundamental concepts of nanoparticulate materials, in particular with respect to their colloidal stability.

# What is nanotechnology?

Nanotechnology is defined as the ensemble of materials and techniques that are able to exploit new properties arising at nanoscale, whereby "nano" refers to the size of 10<sup>-9</sup> meters (nm). Nanomaterials are defined as objects that have at least one dimension smaller than 100 nm. It has been observed that materials in this size regime deploy unique properties, which make them clearly different from their bulk counterpart. Examples of these changes in material properties are the reduced values of melting point, the tuneable optical properties of semiconductors in the quantum size regime and the magnetic properties, to mention only a few.

For the purpose of this contribution, it is worth to focus the attention on three fundamental aspects of nanoparticles: the dispersability, biocompatibility and their catalytic properties.

Particles are surface determined solids where size distribution, shape and surface stabilisers define their properties, functionality and stability over time. The size and shape of particles can be tuned precisely at atomic level using state of the art advanced liquid phase synthesis. However, interfaces play a major role in providing stability over time and functionality. Nanoparticles are kinetically stable objects. Therefore, in the absence of an effective stabilisation their tendency to undergo aggregation and ripening is an underlying phenomenon that has to be always taken into account. In every application that makes use of nanoparticles, it is important to ensure

the absence of aggregation and agglomeration by exploiting three major stabilisation routes (Figure 1). The steric stabilization makes use of organic ligands adsorbed onto the particles surface either during the synthesis of nanomaterials, or in a post-synthetic step. In electrostatic stabilization, charged species are adsorbed onto the surface of particles, providing sufficient electrostatic repulsion. In electro-steric stabilization, charged organic molecules comprise the steric and electrostatic effect. Usually, zeta-potential values larger than  $\pm$  30 mV are used as guidelines to define if a particle suspension is stable.

Derjaguin-Landau-Verwey and Overbeek (DLVO) theory, initially developed for planar charged surfaces, provides an excellent framework to model the interactions between charge-stabilized nanoparticles [15]. This approach, however, breaks often down at the lower nanoscale, where for instance ion specific forces between colloidal particles cannot be explained quantitatively [16].

Figure 1 summarizes the main stabilisation approaches of nanoparticles and their dispersability in media with various polarity, being the last both solid (polymers, resins, plastics) and in liquid state (solvent, oil, etc.).

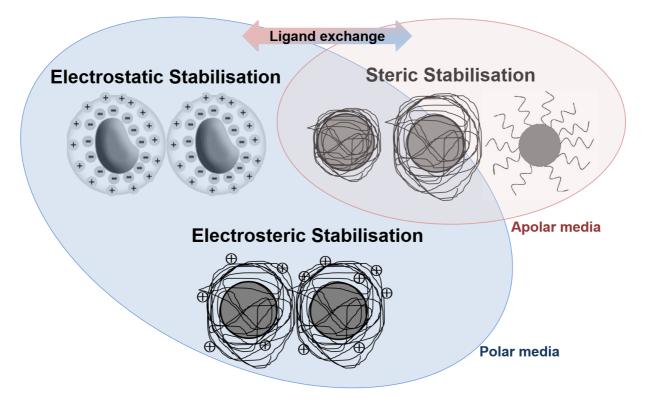


Fig. 1. Overview of the principle approaches for the stabilisation of nanoparticles in media with various polarity.

The surface functionalisation needs to be adapted to the target application. In particular, the selection of suitable ligands will define the dispersability of particles in media with various polarity. Accordingly, short and polar chain ligands and ionic species will be suitable to stabilise nanoparticles within polar media ( $\varepsilon > 50$ ), whereas the stabilisation in organic solvents with intermediate polarity ( $\varepsilon = 20 \div 50$ ) and apolar matrices ( $\varepsilon < 20$ ) will require the use of long-chain surfactants.<sup>3</sup> In some applications, it might be necessary to disperse the (nano)particles in solvents or, more general, in matrices with a dielectric constant very different from the medium where the synthesis of nanoparticles was carried out. For example, the shape and size of semiconductor nanocrystals can be precisely engineered in solvents with low dielectric constant and in the presence of long chain surfactants (i.e. oleylamine, oleic acid, tri-octyl-phosphine, and tri-octyl-phosphine oxide) that make them dispersible only in apolar solvents (i.e. chloroform, hexane, toluene). Several methods are available in the scientific literature to carry out ligand exchange processes. These methodologies enable the substitution of long chain surfactants with short chain ligands [17], or even with metal ions and complexes [18]. Also, the opposite is possible: particles synthesized in polar environment, can be re-dispersed in apolar media by

<sup>&</sup>lt;sup>3</sup>  $\varepsilon$  is the dielectric constant

realizing a steric stabilisation. In all case, the molecules adsorbed onto the particle surfaces will have to be able to create sufficient repulsive forces between particles, to keep them apart and prevent aggregation and agglomeration. Only the colloid stability will ensure the durability of the functional material.

A second aspect that needs to be considered when the application of nanoparticles in large consumer products is envisaged is the potential toxicity of nanomaterials for human and the environment. While there are several studies in the literature devoted to the assessment of particle toxicity, the results are not conclusive [19]. Therefore, it is important to carry out studies on the transport of nanomaterials within the solid matrices and devices where they are incorporated in order to ensure that they are not released to the external environment. Finally, the application of nanomaterials in the field of catalysis has generated remarkable breakthrough in the last decades [20]. The enhanced activity and the size- and shape- dependent properties are ascribed to the majority of atoms residing onto the particles surface, rather than in the bulk. In heterogeneous catalysis, the use of nanoparticles has led to enhancement of catalytic activity as a trade-off between kinetic and thermodynamic aspects of adsorption on the increased number of active sites on the surface, and desorption of products from particles surface. In photocatalysis, semiconductor particles can be designed to have suitable bandgap and combined to harvest sunlight over a wide range of wavelengths. In electrocatalysis, the influence of particle size, structure and catalyst support is well-recognised [21]. Through the engineering of particles synthesis, it is possible to maximise the exposure of high index crystalline facets that notably bear the highest concentration of defects.

In general, the catalyst support has to be understood not as a neutral observer of the catalytic reactions, but as a key actor defining, together with the catalyst, the kinetics and thermodynamics of the catalytic processes [22]. The deposition of nanoparticles onto suitable catalytic supports is a key strategy to recycling and reusing the catalysts, which is usually a very expensive component.

#### Nanoparticles as additives in plastic materials

With the current scenario, the most practical approach for a short-term reduction of the amount of plastic released in the environment seems to be the mechanical recycling.

As shortly mentioned in the introduction, the mechanical recycling of plastic materials requires the addition of various amount of virgin polymer to balance the loss of mechanical strength and the degradation of the optical properties in order to give access to polymeric materials with benchmark properties over an infinite number of cycles.

Recently, it has been shown that a valuable alternative to the use of virgin polymer to enhance the mechanical and optical properties of the recycle polymer is the addition of nanofillers as additives with the purpose to push forward the reuse and the recycling of plastics [23]. This strategy has been applied to several polymeric materials, such as polyethylene terephthalate (PET), polystyrene (PS), polyethylene and high-density polyethylene (PE and HDPE), polypropylene (PP), etc.

The nano-additives that can be potentially used are classified as organic and inorganic materials. The former comprise carbon-derived components such as graphene, carbon nanotubes and nanohorns, or nanocellulose. Interestingly, several of these carbon-based materials can be nowadays derived from biomass [24, 25]. Among the inorganic nano-additives, metal oxide materials such as ZnO, TiO<sub>2</sub> and SiO<sub>2</sub> nanoparticles have found already applications in the recycling of poly(ethylene terephthalate), polypropylene and polystyrene mainly as inorganic nano-reinforcement to improve the mechanical properties [26-28].

Clays are also widely used as additives in recycled plastics, as they are particularly effective in increasing the yield stress, the modulus of tension, rigidity, hardness, and resistance to humidity [23,29,30].

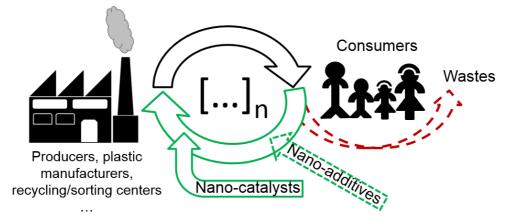


Fig. 2. General scheme of the life cycle of plastic materials from producers to consumers, and possible path alternative to the waste generation, from consumers to recycling/sorting centres and plastic manufacturers, involving the contribution of nanotechnology.

In the examples provided above the inorganic materials are usually used in small amount, in a way that they compensate the lack of mechanical strength, while maintaining a neutral colour with respect to the original material. However, nanoparticles could be used also as nano-pigments, whereby in the framework of this application they are providing beside the nano-reinforcement function also an optical response such as the colour. The colour of the original matrix can be enhanced, intensified or modified, accordingly. This approach would be particularly interesting in the cradle-to-cradle perspective, where the plastic material derived from a recycling process can find application not only in the original product but also in a new one.

Following this idea, inorganic nanoparticles can be classified based on their optical response as ultraviolet (UV), UV-visible and UV-visible-near infrared (NIR) pigments. In the first class, white solids are enclosed such as silica, TiO<sub>2</sub>, ZnO. Among the visible pigments, iron oxides can play a central role for the strength of the colour that they are able to provide and for the biocompatibility of this class of materials. Accordingly, hematite is an example of a red pigment, goethite is yellow and magnetite/maghemite are black pigments. In the NIR region, pigments can be used to exploit thermal management applications. However, the use of nanoparticles as additives in plastic to enhance the mechanical and optical properties of recycled polymers requires still extensive studies at several levels.

The majority of nano-additives available in the market are synthesised on industrial scale either in gas-phase processes or in liquid phase using usually water as solvent. The pigments obtained are available as dried solids that need to be re-dispersed in order to be used in a given application. This re-dispersion in liquid or molten media requires a careful control of the solid-liquid interfaces over a wide range of temperatures by applying the strategies outlined in Fig. 1. In order to ensure a homogeneous distribution of the particles inside the recycled plastic, it is fundamental to match the polarity of the functional groups onto the particles surface with the polarity of the matrix. This approach ensures an even distribution of the nanoparticles in the organic matrix, where the presence of particles as single objects and not as aggregates is fundamental in order to ensure that the properties of particles are transferable to their ensemble.

In order to realise processes with a true environmental and economic benefit, it is necessary to ensure that the composite material obtained after the addition of the nano-additives at the first cycle, can be recycled over several cycles maintaining the same properties. This is true not only for the organic matrices, but also for the nano-additives. In other words, the stability of particles and their dispersibility has to be ascertained to ensure that the particles will not undergo aggregation and/or ripening in the successive cycles. A second important point is how to ensure that the nanoparticles remain enclosed in the plastic material and are not released in the environment. There, besides the direct potential risks for living organisms to get in contact with nanoparticulate systems of which the toxicological properties are not yet ascertained, the accumulation of non-degradable nanoparticles in the environment would indirectly threaten human health, similarly to the phenomenon of microplastics described above. If these critical points are not addressed, the applicability of nanoparticles as additives for the processing of post-consumer plastics in the framework of a circular economy seems very labile (dashed green arrow in Fig. 2).

## Nanoparticles as catalysts for the depolymerisation of plastics

Instead, the depolymerisation of plastics in the constituent monomers appears to be a more promising research field. For a comprehensive overview of the state of the art in materials and processes, the reader is invited to read the recent contribution by Sheldon and Norton [14]. In the following, it will be proposed how nanotechnology can make a breakthrough in this nascent field (Table 1).

With their high surface area, the possibility to tune the reactivity by controlling the size and shape of the nanoarchitecture and defect density, nanoparticles have been reported as active catalysts in a number of processes. The application of nanoparticles as catalysts is one of the few fields where nanotechnology has reached a market application (see for instance the catalytic converter and fuel cells). In the specific case of catalysts for plastic depolymerisation, the state of the art is still at lab-scale and can be considered at its infancy. The key challenges are here the effective immobilisation of nanoparticulate catalysts onto suitable catalytic supports in a way that the activity of the catalyst is enhanced through a cooperative effect with the substrate. Besides the enhanced activity of particles due to the large surface to volume ratio, the possibility to fabricate multifunctional materials is a clear advantage of nanotechnology. A multifunctional material comprise several domains spatially and electronically organised onto the substrates. The domains can interact with each other, or carry out subsequent steps of a complex chemical transformation. Deactivation processes such as Ostwald ripening, detachment of the particles from the catalytic support with consequent dissolution and Ostwald ripening have to be circumvent. These challenges can be tackled by strengthening the interaction of the nanocatalysts with the solid support and by exploiting encapsulation strategies in porous shell. The size of the pores has to be large enough to enable the access of the polymer to the active sites of the catalysts, and small enough with respect to the particles size to ensure that the nanoparticulate catalysts remain protected inside the porous shell.

Another important contribution of nanotechnology to the depolymerisation of plastics is the possibility to recover and reuse the catalysts over several cycles. A possible strategy to achieve the recyclability is to immobilise the catalysts onto the surface of superparamagnetic particles. These are highly crystalline materials that, in the absence of an applied field, behave as non-magnetic, but when a magnetic field is applied they are strongly attracted to the external magnet and can be separated from the solution by magnetic decantation [31]. The magnetic decantation is well-know not only at lab scale, but also in some industrial segments. Examples of superparamagnetic particles are  $Fe_3O_4$  (magnetite) and  $CoFe_2O_4$  (cobalt ferrite), for which water-based protocols for the largescale production are available.

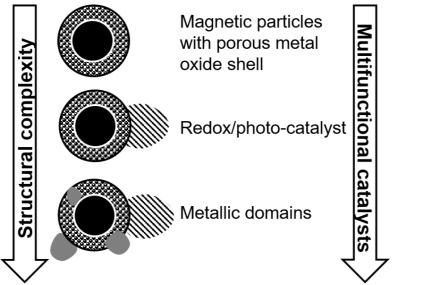


Fig. 3. Nanotechnology offers the theoretical framework for the fabrication of multifunctional materials.

## Impact

Plastic materials have influenced largely the modern society with durable, light and flexible products. The nondegradable properties of plastics is of paramount importance in order to provide robustness and durability of plastic materials, but at the same time it leads to their accumulation in the environment, often under the form of microplastics that originate from photodegradation of macroscopic materials. However, the problem of plastic accumulation in the environment needs to be solved at a global level. For decades, higher income countries have exported plastics to lower income countries of the East Asia and Pacific area. However, the Chinese ban to the import of plastics from other countries poses serious challenges to the source lands that have to find an alternative to landfill and incineration [32].

Recently, waste management based on plastic recycling has gathered remarkable attention in the scientific community in both industry and academia. The recycling of plastics has important impacts on environmental, economic and social level. The plastic recycling and the development of biodegradable plastic materials is expected to have a very positive impact on the GHG abatement and will help Europe to keep the commitment to the Paris agreement. This target, however, will be achieved only if true sustainable products will be developed with environmental, social and economical benefits throughout the entire process chain, from extraction of raw materials to disposal of the final wastes.

Industry has recognized the importance of shifting the paradigm of used plastics from wastes to resources. However, it is very unlikely that the plastic recycling and the reduction of the plastic wastes released in the environment can be successful without a direct involvement of the citizens. The separation of household plastic wastes has to be pushed forward in order to ensure the homogeneous composition of the feed before recycling. This is a priority both for the mechanical recycling of plastics and becomes fundamental for their chemical depolymerisation into constituent monomers. The combination of life cycle assessment and cradle-to-cradle principles is necessary in order to quantify and design general rules for the development of materials and processes that are more sustainable for human and the environment [33]. A realistic perspective on the cradleto-cradle paradigm from an energetic point of view should not be neglected [34]. What is clear and widely accepted is that the release of plastics in the environment, either traditional or degradable, virgin or recycled, is not anymore an acceptable solution.

#### Conclusions

In summary, it has been shown that nanotechnology can contribute to the field of waste management.

Nanoparticles can be used as additives for mechanically recycled plastics to improve the mechanical and optical properties. Another possible contribution of nanotechnology is the development of nanoparticulate catalysts for the depolymerisation of plastic into the constituent monomers. For this purpose, the nanocatalysts have to be immobilised onto the surface of suitable carriers, whereby recoverable magnetic materials are an example of possible catalytic support. In both kind of applications, it is fundamental not only to control the size and shape of nanomaterials, but especially the surface properties of the nanoparticles. They will contribute in large scale to determine the dispersability of particulate additives in media with various polarity, the strength of interaction between the nanocatalysts and the catalytic support, and ultimately the catalyst performance.

#### **Conflict of interest**

There are no conflicts to declare

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# THE CRITICAL ROLE OF ADVANCED SUSTAINABILITY ASSESSMENT TOOLS IN ENHANCING THE REAL-WORLD APPLICATION OF BIOFUELS

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# Abstract

Sustainability has become of paramount importance in the biofuel industry. Accordingly, various sustainability assessment schemes such as emergy analysis, techno-economic analysis, life cycle assessment, energy accounting, and exergy analysis and its extensions (exergoeconomic, exergoenvironmental, and exergoeconoenvironmental analyses) are being employed increasingly for decision-making on biofuel production and consumption systems. In this opinion paper, after classifying and describing biofuel generations, the developed sustainability assessment tools are critically explained, and their pros and cons are discussed. Overall, among the various sustainability assessment approaches introduced so far, exergy-based methods appear to be the most promising tools for developing sustainable biofuel systems. This can be attributed to the fact that the exergy concept is deeply rooted in the well-defined principles of thermodynamics.

# Keywords

biofuels; sustainability; exergy; emergy; Life Cycle Assessment; decision-making

Fossil fuels are currently the major sources of energy globally, and this is unlikely to change anytime soon. On the other hand, pursuing the business-as-usual scenario is regarded as a serious threat to our entire species. This is mainly ascribed to the increasing atmospheric concentration of greenhouse gases (GHGs) contributed by fossil fuels. The resultant unfavorable phenomena, i.e., global warming and climate change, have already endangered public health from different perspectives and could profoundly affect the life of a child born today [1].

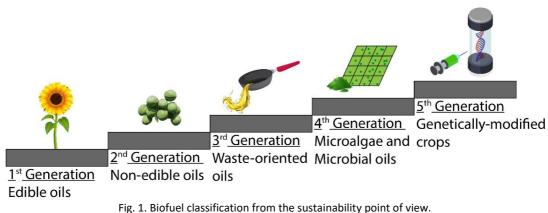
Various types of renewable energies have been at the center of attention for quite some time now, intending to mitigate the tragedy by reducing the global share of carbon-intensive energy carriers. However, their penetration into the transportation sector has been trivial. This sector requires  $5.6 \times 10^{20}$  J/yr (560 EJ/yr), accounting for 30% of the world's fossil fuel annual consumption [2]. Substituting fossil fuels with their renewable counterparts at such a huge scale is a major challenge. Moreover, most renewable energy carriers such as solar energy and wind energy can hardly be considered as short-term or even mid-term solutions, given their current intrinsic technological limitations. Hence, among the existing options, biofuels such as bioethanol and biodiesel seem to be the most promising alternatives to at least partially replace their carbon-intensive fossil-oriented counterparts, i.e., gasoline and diesel, respectively.

Biofuels are less carbon-intensive vs. fossil fuels. For instance, biodiesel combustion lowers unburned hydrocarbon, carbon monoxide, and smoke emissions by 20%, 30%, and 50%, respectively, compared with mineral diesel [3]. From the global warming perspective, it should also be highlighted that the carbon emitted through the combustion of biofuels is of biogenic origin and hence, does not contribute to the long carbon cycle also known as the geogenic carbon cycle. Combustion of each liter of mineral diesel, for instance, leads to the

emission of 2.67 kg of  $CO_2$  of geogenic origin, which could be entirely prevented by using biodiesel [4]. Biofuels are also advantageous from the application point of view for several reasons; 1) they share similar physicochemical properties with fossil fuels, 2) they can be used in in-use engines with little or no modifications, 3) they are indigenously available, among others.

Despite the above-mentioned promising features, biofuels, and in particular, their first-generation, have also been criticized for their sustainability features. First-generation biofuels amounting to over 95% of the global biofuel production are produced from edible resources, e.g., corn bioethanol and soybean biodiesel [5]. Despite their economic viability in geographical locations with abundant fertile lands and water resources, their sustainability is heavily questioned, citing their land-use change (LUC) and the resultant contribution to the net flux of carbon to the atmosphere [6]. In addition to that, by diverting edible resources with food and feed applications to the biofuel industry, global food security has been more than ever threatened. Biofuels production has been widely criticized for its unfavorable impact on crop prices and contribution to hunger [7]. These shortcomings place first-generation biofuels in conflict with the United Nations' Sustainable Development Goals (SDGs) and particularly SDG 2. These downsides have been the main motivation for the shift from first-generation biofuels to the next generations.

From the sustainability point of view, the most comprehensive classification of biofuels was presented by Hajjari et al. [8], based on which a distinction was made between non-edible resources or biomass (Figure 1). More specifically, non-edible energy crops were designated as feedstocks resulting in the production of secondgeneration biofuels, while waste-oriented resources such as waste cooking oil (WCO) and bagasse were identified as feedstocks associated with the third-generation biofuels [3]. It should be noted that although the non-edible energy crops pose no apparent threat to global food security, they are no different from their first-generation counterparts from the LUC and water security perspectives unless cultivated on marginal lands and through non-agricultural water resources. However, even under the latter circumstances and to achieve the maximal yields, non-edible energy crops are still associated with environmental burdens arising from carrying out the routine agricultural practices such as fertilizer application, harvest, and transfer. On the contrary, thirdgeneration feedstocks are simply the different sources of waste generated through completely different processes and are therefore attributed to no upstream GHG emissions nor carbon footprints. While such a comparison is largely accurate but it overlooks an important feature of non-edible energy crops; i.e., soil and root carbon storage contribution. In a recent investigation, Yang and Tilman [9] proved that soil and root carbon storage contributed by these crops (especially high plant diversity mixtures) is the main source of GHG savings rather than fossil fuel displacement. They argued that the former removes CO<sub>2</sub> from the atmosphere directly, whereas the latter benefit stems from the avoidance of future fossil fuel GHG emissions, a pathway that is dependent on a wide array of assumptions and uncertainties, especially the rebound effect of the fuel market [9].



Source: Adapted from Hajjari et al. [8].

It is also important to note that to take full advantage of the carbon storage benefits of non-edible energy crops, they should be cultivated on marginal lands and irrigated by non-conventional water resources. This could be simply justified by taking into account the adverse impacts of LUC on the carbon intensity of the whole process. It has been documented that LUC caused by the cultivation of non-edible energy crops could contribute to the emission of 17 to 420 times more  $CO_2$  than the yearly GHG mitigations expected *via* fossil fuel displacement [10–12].

As mentioned earlier, third-generation feedstocks are not associated with any upstream environmental burdens; however, it should not be overlooked that they generally require further processing instead to be quality-wise on par with their first- and second-generation counterparts. Such further processing is attributed to some material and energy inputs and, therefore, imposes some environmental burdens. For instance, WCO should undergo physical filtration to eliminate food debris, followed by de-watering and esterification to remove water and free fatty acid contents, respectively, before it can be introduced into a feasible transesterification process [13]. On the contrary, soybean oil (first-generation feedstock) and non-edible safflower oil (second-generation feedstock) do not require such pretreatments. Therefore, the use of waste-based resources for biofuel production should also be thoroughly scrutinized from the sustainability perspectives using advanced sustainability assessment tools.

The fourth- and fifth-generation biofuels are generated from microalgae and genetically-engineered crops and algae, respectively. Despite the promising features of these generations from the sustainability perspective, on most occasions, the costs of the biofuels produced are not competitive yet [14]. In light of that, recent efforts have been largely dedicated to developing multi-products biorefinery systems to mitigate the challenges faced and enhance the economic feasibility of these generations of biofuels [15]. For instance, in a recent work, Kim et al. [16] presented an innovative closed-loop biorefinery process by integrating the use of low-recalcitrant engineered biomass with its pretreatment using lignin-derived deep eutectic solvents. The authors reported that the integrated process led to near-theoretical sugar yields.

Bearing in mind the above arguments, any decision-making and subsequent expansion of the biofuel industry should be subject to a thorough sustainability assessment ensuring that the various aspects of a production cycle will be taken into account. Advanced sustainability assessment tools such as emergy analysis, techno-economic analysis, life cycle assessment (LCA), energy, and exergy analysis and its subsidiaries (exergoeconomic, exergoenvironmental, and exergoeconoenvironmental analyses) are promising approaches to serve that purpose [17,18].

The emergy concept is based on two main pillars, i.e., systems ecology rooted in general systems theory and biophysical principles governing the systems [19]. Emergy is developed to quantify the available energy previously used in direct and indirect forms in creating a good or service [20]. This long-run sustainability assessment tool can adequately evaluate the environmental burdens caused by a good or service [17]. Using transformities (i.e., unit solar-equivalent Joule), all kinds of support to a system under investigation, including energetic/non-energetic and material/non-material fluxes, are converted into a homogeneous unit, i.e., solar emjoule (sej) [21]. The total environmental production cost of a good or service is obtained by summing the emergy values of all inflows, which are obtained by multiplying their quantities by the corresponding transformities [21]. The most critical step of an emergy analysis is to choose the proper transformity coefficients being used to express all influxes in solar energy equivalents [17]. Like other sustainability indicators, the emergy concept suffers from its conceptual and methodological limitations [22]. The majority of the issues associated with this method, including inaccuracy, irreproducibility, inconsistency, and incompleteness, can be effectively addressed by mimicking the inventory modeling fundamentals of the LCA methodology.

Techno-economic analysis is one of the best ways to assess and compare the economic viability and profitability of energy projects through identifying their short- and long-term outcomes [18]. This method includes hints to address technical and economic bottlenecks of newly emerged energy technologies that could pave the way for their commercialization. However, this analysis does not consider the thermodynamic and environmental aspects of energy systems, rendering it susceptible to offering misleading recommendations and conclusions [18].

LCA is one of the most popularized methodologies to assess the environmental impacts associated with making and using a good or service over its entire life cycle [23]. This approach can track the cradle-to-grave environmental consequences of a good or service, i.e., from raw material extraction to product manufacturing, distribution, use, maintenance, recycling, and disposal [24]. An LCA analysis is started by gathering the relevant inflows and outflows of a system under investigation, proceeded by quantifying their related environmental burdens, and completed by translating and interpreting the outcomes of the inventory analysis and impact assessment stages [25]. LCA methodology can offer a broad spectrum of insights on natural resource consumption, climate change, human health, and ecological degradation associated with a good or service [26]. Nevertheless, subjectivity, inaccuracy, uncertainty, and variability are the main inherent problems of this approach [17].

The first law-based energy analysis is one of the most extensively applied approaches for measuring and comparing the performance of energy conversion systems. Despite the useful information provided by this analysis, it fails to account for the quality loss of energy caused by internal irreversibilities in thermodynamic processes [27]. Most of the problems, if not all, associated with energy analysis, can be adequately addressed using the exergy concept in which both the quality and quantity of resources are taken into consideration [28]. The thermodynamic property exergy can reliably quantify the upper limit of useful work that can be obtained from an energy system. When the maximum amount of work is produced, the system under consideration reaches a state through reversible processes in which it is in equilibrium with the reference environment. In general, exergy analysis is used to quantify, locate, and interpret the thermodynamic losses of energy conversion systems [29].

Besides, the enhanced versions of exergy analysis, i.e., exergoeconomic and exergoenvironmental approaches, can offer more detailed information about technical, economic, and environmental aspects of energy conversion systems. These approaches can provide a deep understanding of economic costs and environmental impacts associated with equipment and thermodynamic losses, as illustrated in Figure 2 [18]. The exergoeconoenvironmental approach elaborated by Aghbashlo and Rosen [30] can yield further complementary information concerning the thermodynamic, economic, and environmental aspects of energy systems. Advanced analyses developed by Tsatsaronis and Morosuk [31] can also boost the quality of the results obtained from exergy-based approaches by providing extra information about the thermodynamic, economic, and environmental interactions among the units of energy systems. These analyses can also quantify the avoidable portion of the thermodynamic losses, economic costs, and environmental impacts.

Similar to the other available sustainability ranking approaches, the reliability of the results of exergy analysis is substantially influenced by the cut-off criteria specified for the boundaries. Furthermore, the conclusions derived from exergy-based analyses are affected to some extent by the assumptions made regarding the reference conditions. Overall, among the various sustainability rating tools explained above, exergy-based approaches appear to be the most promising tools for developing sustainable biofuel systems. This can be attributed to the fact that the exergy concept is deeply rooted in the well-defined principles of thermodynamics.

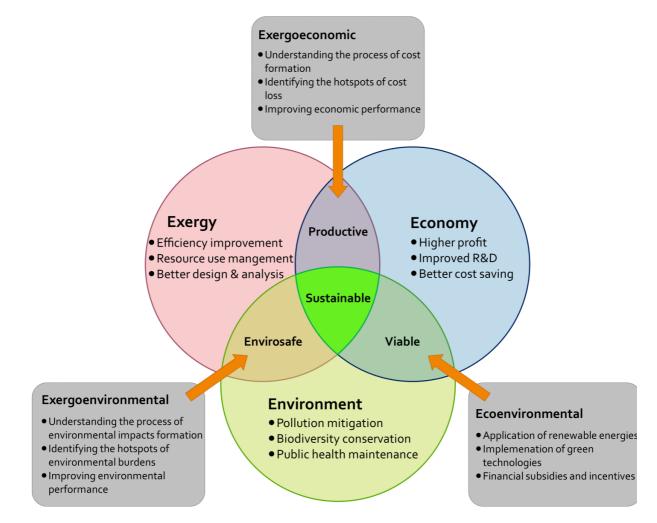


Fig. 2. The interactions among exergy, economy, and environment [18]. With permission from Elsevier. Copyright© 2020 Elsevier. License Number: 4918831100244.

# Impact

Climate change and its adverse impact on public health are believed to have been driving the world toward an irreversible point, at least as much as our species is concerned. Hence, there has been a growing interest in various strategies aimed at mitigating greenhouse gas emissions, including fossil fuel replacement with alternative energy carriers such as biofuels. To date, different generations of biofuels have been developed, each associated with pros and cons. Accordingly, any decision-making and subsequent expansion of the biofuel industry should be subject to a thorough sustainability assessment ensuring that the various aspects of a production cycle will be taken into account. Advanced sustainability assessment tools such as emergy analysis, techno-economic analysis, life cycle assessment (LCA), energy, and exergy analysis and its subsidiaries (exergoeconomic, exergoenvironmental, and exergoeconoenvironmental analyses) are promising approaches to serve that purpose and are critically discussed in this work.

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