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Review

Sustainable Environmental Management and Valorization Options for Olive Mill Byproducts in the Middle East and North Africa (MENA) Region

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Abstract: Cultivation of olive trees and olive oil production have been considered as a legacy for the Mediterranean region. This custom represents a very important benefit for many nations in terms of wealth and health. However, huge amounts of by-products and waste are generated during olive oil production. This represents a serious environmental impact on land and water bodies if not properly handled. Olive oil extraction generates two waste streams, a solid waste called pomace and olive mill wastewater (OMWW), which has been considered as highly pollutant and phytotoxic waste. These wastes have high disposal costs and predominantly generated from small-scale enterprises that have limited financial resources to treat them properly before discharge to the environment. Besides being a serious environmental problem, OMWW has potential economic value that remains to be utilized such as: fertilizers, valuable antioxidants agents and fatty acids needed in human diet. Also, Olive pomace is a valuable renewable energy source with an energy density of 23 MJ/kg and has become an inexpensive alternative for fossil fuels. Aiming at adding value to the olive production sectors and potential valorization options for byproducts in the MENA region, international practices applied in olive mills wastes management's and treatment methods used in major oil producing countries are presented.

Keywords: olive mill wastewater; Zibar; olive Pomace; Jift; treatment; valorization; cleaner production; disposal

1. Introduction

Olives trees (*Olea europapea* L.) cultivation and olive oil production have deep roots in the history of Mediterranean region. The centuries old tradition represents a very important asset for many countries in the region, not only in terms of culture and health but also in terms of wealth. The olive oil production is increasing over time due to the increase in olive trees cultivation and customers high demand. Over the past few decades, olive oil represents an important healthy dietary trend worldwide, because it is considered as important resource of essential fatty acids and antioxidants agents in human diet [1].

According to latest estimations by Reference [2], approximately 11 Mha of olive groves were cultivated in 2015 worldwide. Roughly 50% of these groves' cultivations are in EU countries, especially Spain (24.2%), Italy (11.1%) and Greece (9.0%). Total olive fruit production in the EU accounted for 13.24 Mt, again Spain is the major producer with 7.87 Mt (60%) of total EU yield. The annual global olive oil production is around 3.32 million, with 72% produced in Europe [2].

Olive oil main producers are Spain (42%), Italy (17%) and Greece (11%), other Mediterranean countries producers are Turkey, Syria and Tunisia with (6%) each and Morocco (4%), Jordan (3%) and Lebanon (1.5%). Other smaller producers outside Mediterranean basin in Asia, Africa and America are growing with 15%, 12% and 2% of worldwide olive production, respectively [2].

Olive groves are the main agricultural plantation over the eastern Mediterranean. Olive orchards are typically small and located in hilly areas and therefore difficult to access and expensive to harvest by hand [3]. In addition to the nutritional value in its products, olive trees have significant social and economic importance in the life of the people living in Mediterranean countries. The biomass residues from olive trimming and olive mill byproducts after drying and pelleting have become an important source of renewable energy and an inexpensive alternative to traditional fossil fuels used in home heating in rural areas. In Mediterranean countries where soils are poor in organic matter and vigorous desertification processes are in play, the recycling of olive mill wastewater (OMWW) as a soil amendment should be encouraged.

The environmental impact of olive oil production is critical in all olive oil producing countries in the Mediterranean. The oil extraction processes need a significant amount of water and produces huge amounts of olive mill wastes (OMWs) in short period of time (October-January) of olive harvesting season [3–11].

Management of (OMWs) is considered a major environmental challenge in Mediterranean region. Olive oil extraction generate two types of waste streams: a liquid stream called (OMWW) or locally named “Zibar” and a solid residue known as pomace, locally referred to as “Jift.” Olive mills dispose their OMWW in settling ponds that are normally under-sized and sometimes get overloaded causing spillage to nearby valleys [3,4]. There are no proper facilities for treatment of OMWW in individual mills, so their minimization, prevention and treatment, have long been investigated to reduce their environmental impacts caused by their uncontrolled disposal [12]. The wastewater is also a major source of odor and can be harmful to plants if irrigated at high OMWW concentration.

Therefore, guidelines should be set to handle these wastes streams properly by adopting technologies that minimize their generation and increase awareness, motivation within the industry and by introducing cleaner production options for farmers to minimize their environmental impact and increase resource sustainability. Management of this wastewater has been considered in the past three decades with special attention to its safe treatment and possible valorization [13].

The effects of application of OMWW on the environment, on soil properties and on olive tree performance were studied by many local researchers [14–17]. Their results showed that application of OMWW did not have any negative effect on soil properties. It served as a beneficial soil amendment particularly in providing plant macronutrients (nitrogen, phosphorus and potassium) along with organic matter that is beneficial to soils [15–23].

To improve land fertility and olive trees performance, the recommended application of OMWW to olive orchards is 100 m^3/ha (10 L/m^2) [14–18]. It was reported that the concentrations of organic matter, phenolic compounds, potassium and microbial activities were considerably enhanced in soil amended with OMWW [14]. Furthermore, a substantial increase in plant growth, photosynthesis and fruit production were also observed without affecting oil quality parameters. On the other hand, short-term deleterious effects on soil biological activities and chemical properties were noticed after OMWW application but it can be considered insignificant after appropriate waiting time [24]. For toxicity mitigation, it was recommended applications of OMWW six months before sowing of maize [25]. Beside the right selection of suitable soils (particularly calcareous soils) and tolerant crops, like maize, this approach might be considered as an effective method to avoid problems associated with OMWW uncontrolled disposal and represents an economical opportunity by providing cheap available local fertilizer.

The uncontrolled disposal of OMWs on soil results in decreases in water retention and infiltration rate, increases soil hydrophobicity and causes strong phytotoxic and antimicrobial effects, in addition to affects soil acidity, salinity, nitrogen immobilization, nutrient leakage, organic acids, lipids concentration, naturally occurred phenols and microbial activity [26].

In this study, the status of olive by-products management is presented, along with their present utilization and techniques applied in major oil producing countries. Alternative options that can be adopted for eliminating possible environmental problems associated with uncontrolled disposal of olive mill byproducts are investigated. Aiming at adding value to the olive oil industry in Middle East and North Africa (MENA) region, international best practices applied to solid waste and wastewaters treatment as well as potential valorization options are also highlighted.

2. Olive Harvesting and Processing Methods

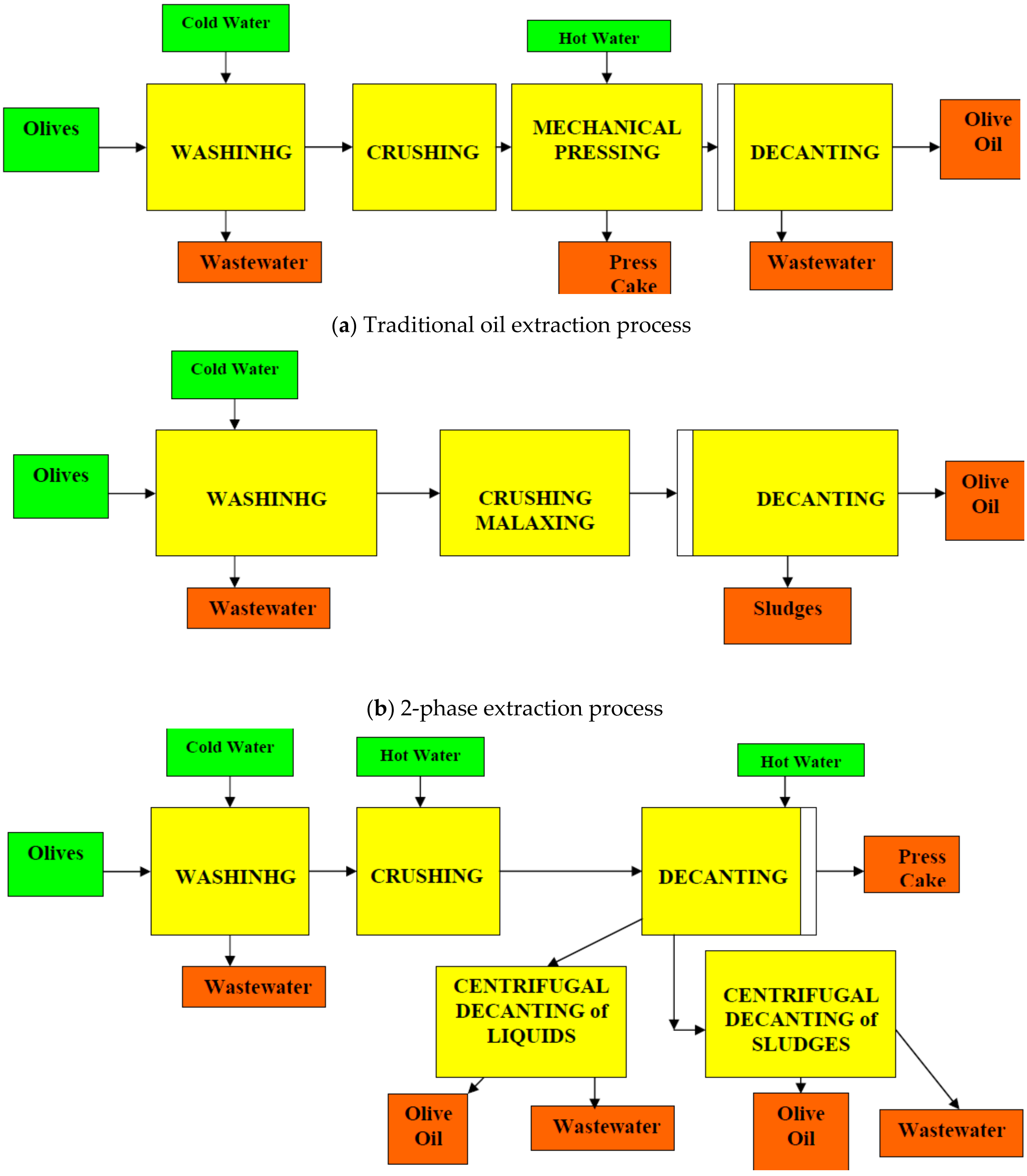
In the (MENA) region countries, olives are typically harvested by hand because of the small sizes of orchards in addition to small farm holding. Harvested fruits are gathered in sacks and transported to olive mills for further processing. The traditional press process is the oldest method for obtaining olive oil from fruits. Significant industrial development has been made in olive oil manufacturing in the past three decades aiming at reducing water consumption and related potential environmental impacts. To improve extraction efficiency, the industry developed continuous centrifugation systems to obtain better quality of olive oil produced while reducing waste generation by 75% [27]. Nowadays, most conventional olive oil mills have been replaced by continuous centrifugal systems that is, ecological decanters [27].

Olive Oil Processing Systems and By-Products Characteristics

Olive oil is extracted from olives by mechanical means. Three methods are used namely: the traditional press, the three-phase centrifugation and currently the two-phase ecological decanter systems used since 1990 [7,8]. The three-phase centrifugation system produces two by-products: solid by-product that consists mainly of olive pulps and stones called olive pomace with a humidity of about 55% and the OMWW consists of olive fruit water and water used washing and processing olives [7,28]. The three-phase method normally produces around 20% olive oil, 30% olive pomace and 50% OMWW, which indicates that wastes produced are four times more than olive oil. The two-phase systems require small quantities of water for oil extraction, thus smaller amounts of wastes are generated.

Olive oil extraction process steps include washing of olive fruit, crushing of fruit, malaxation and separation and finally the extraction of oil. Malaxation is the process of mixing milled olives for 20 to 40 min at 27 °C allowing smaller oil droplets to aggregate and be separated more easily. The effectiveness of oil extraction yield is a function of mixing time and maintaining the desired temperature. The extraction at higher temperatures implies that the oil cannot be labeled “cold pressed,” a process that is preferred by consumers. [29].

Input-output analysis of each extraction process is illustrated in Figure 1. Olive oil production starts with the washing of olives to remove any dirt and other contaminants (e.g., leaves and twigs). Water usage at the mills depends on pressing methods and operational practices. Average volume of OMWW generated ranges from 0.3 to 1.2 m^3/ton of processed olives (Table 1). In 3-phase mills, average freshwater consumptions range from 700–1000 L/ton of olives processed and the generated OMWW is 1200 L/ton, the highest of all processes. Corresponding figures for 2-phase mills are 100–120 L/ton of freshwater consumed and 200 L/ton of OMWW generated.



(**a**) Traditional oil extraction process

(**b**) 2-phase extraction process

(**c**) 3-phase extraction process

Figure 1. Process flow chart of olive oil extraction methods.

Table 1. Approximate input output analysis of water consumption and byproducts generated at olive oil mills [3,30,31].

Mill Type	Input	Quantity	Output	Quantity
Traditional	Olive Washing water	1000 kg 100–200 L	Oil Pomace Wastewater	230.4 kg 500 kg 650 Liter
3-Phase	Olive Washing water Hot water Added	1000 kg 100–120 L 700–1000 L	Oil Pomace Wastewater	256.4 kg 581.16 1200 L
2-Phase	Olive Washing water	1000 kg 100–120 Liter	Oil Pomace Wastewater	247.4 kg 735 kg 200–300

OMWW constitutes a suspension consists mainly of: 80–92% water, 3–15% organic matters such as oils and fats, carbohydrates, lipids, pectin, organic acids, sugars, mucilage, polysaccharides, phenols, tannins and lignin and 0.50–2% mineral content on a weight basis [10,32–39].

The effluent highest concentrations of biological oxygen demand (BOD) and chemical oxygen demand (COD) might reach 100 and 220 g/L, respectively [40], with high COD/BOD_5 ratio between 2–5 [4,30,41]. The chemical-physical properties of OMWW depend on olive cultivar, cultural practices, maturity (ripening stage of the olives), climate conditions, olive storage and oil production system [4,20,35,36,42–46]

The aromatic, long-chain fatty acids and polyphenolic compounds with concentration up to 80 g/L are considered the major factors related to OMWW pollution characteristics because they are toxic to soil microorganisms and plants [47–49] and considered the most polluting waste generated by the agri-food industries [50–52]. Phenolic compounds are initially produced by the olive trees and developed later during the oil extraction processes [7]. Most of these compounds are soluble in water, so they go away with olive mill by-products and just a small amount is remained in the oil [53].

The two-phase extraction system was introduced to the olive oil mill industry in 1992 [30,52] and considered as the most ecologically friendly techniques. These decanters generate smaller quantities of byproducts that are typically 75% less than what is generated while using the other two extraction methods [30], thus providing cost savings in wastewater treatment and disposal. However, the pomace produced in the two-phase system is excessively wet characterized by high humidity (62%) [54], which is considered as a major drawback of this technique [32,54].

The increase in quantity of waste generated and their disposal without proper treatment causes environmental challenges because of its phytotoxicity and high organic matter content [54,55]. To alleviate these negative impacts, environmentally friendly waste management technique is required by law. The most common management practices are the use of lagoons or storage ponds located on mill sites that both allow wastewater to evaporate naturally and for solids (along with any heavy metals) to settle to the bottom of ponds [3,4].

3. Current Management Practices of Olive Mills by Byproducts in the MENA Region

The replacement of the three-phase mills with two-phase mills has reduced consumption of water throughout oil extraction processes. This change has a significant positive impact in most Mediterranean countries as these countries are also undergoing severe water shortages [3,4]. Additionally, the use of less water translates to generation of lower quantities of process byproducts. However, environmental concerns are not eliminated totally in this process. A semi-solid waste stream with a unique physical and chemical properties is produced leading to the need for a new management practice that is different from that of handling olive husks. This requires development of a new process that is technically feasible and economically affordable for managing this waste stream.

3.1. Management Practices of OMWW

The increase in number of olive groves plantations over years has led to an increase in number of olive mills in the region. Wastewater generated at the mill is initially stored in concrete or otherwise sealed ponds and lagoons (Figure 2a–c). It is then transferred by tanks to officially designated sites. At the present time there are no OMWW treatment plants facilities operating in the MENA region. The discharge of OMWW to the municipal sewer system is totally prohibited because of its corrosive nature and the high concentration of suspended solids which may result in clogging of the wastewater network close to the olive mills.

(**a**) Concrete pool (**b**) Evaporation Lagoon

(**c**) Sealed Drying pond for OMMW (**d**) Spring polluted with OMWW

Figure 2. Wastewater storage and handling at different locations of olive mills in Irbid area.

There are small gaps related to policy, legal and institutional framework for OMWW management in the region, which has sometimes resulted in environmental mismanagement, pollution and potential threat to scarce surface and groundwater resources as shown in Figure 2d. Due to its characteristics, OMWW is considered as an industrial wastewater and is regulated as such. It was reported that one cubic meter of OMWW has the equivalent environmental impact of 100–200 m^3 of domestic sewage [10], while their discharge to lakes, rivers or sea have a severe effect on aquatic ecosystems because of eutrophication potential [55].

Olive mills in the MENA region are typically small to medium size enterprises, geographically scattered around countries sides, lacking financial resources and technical capabilities needed to develop an appropriate on sites OMWW treatment plant. Treatment methods have to be simple and inexpensive in order to help the olive mill owners to adopt it and raise their awareness to best management practices, especially in upgrading olive mills in order to enhance their global competitiveness and increase exports of processed oil. Many olive mills have implemented good manufacturing practices and have been able to get the Hazard Analysis and Critical Control Point (HACCP) certificate, representing expertise in a system of food safety protocols that are internationally recognized.

3.2. Management Practices of Olive Pomace (Jift)

The three-phase olive mill produce huge amounts of OMWW, in addition to huge amounts of solid wastes. Olive pomace (Jift) is the solid by-product, obtained by olives processing composed of fibrous part (fruit pulp and olive skin), pit, together containing approximately 5–8% of residual olive oil and water content between 25–70%, depending on extraction systems [20,56–58].

The pomace generated per ton of processed olives is roughly 870 and 620 kg in the "two-phase" and "three-phase" systems, respectively [57] and the wet soft texture of the pomace restricts its transportation [20]. Pomace has an acidic pH (≈5.2), oil and fats (8–14%) on dry matter basis and by elevated levels of phenolic substances (1–3%, on dry matter basis) [8,59].

The pomace is sometimes used as animal feed [60] or composted to produce a stable organic soil amendment with fertilization value [60]. Pomace (Jift) is also used as a substrate for the production of activated carbon [61,62], as a source of bio-pesticides [63], in co-firing with coal at power stations [64] and as a source of residual oil for the soap industry [65]. Unused pomace if disposed improperly might cause serious environmental consequences [3]. Because of its organic content, this material can be used as a soil amendment after it has been treated, thus improving soil properties [13] leading to increased soil productivities [66].

The olive pomace calorific value largely exceeds that of other wood or agricultural biomass and releases considerable energy during its utilization. The olive pomace with heat energy of 23 MJ/kg [67–69] is considered a valuable resource of renewable energy and has become an alternative to natural gas and liquid fossil fuels normally used in house heating [70]. Many olive mills pressed the dried pomace to form blocks or briquettes and sell them to local communities as a fuel for home heating. Therefore, olive pomace has become a source of income for mills owners. Prior to 2000, small quantities of pomace (Jift) have been used for space heating at rural mills during winter. Currently, a majority of olive pomace produced in the region is upgraded by means of agglomeration, including briquetting and pelletizing [71,72].

Pelletizing is the process of compressing or molding crude or exhausted olive pomace into the shape of a pellet obtained by mechanical compression. The process starts with drying, grinding, conditioning, and pelletizing to be used later as upgraded solid fuel for space heating. Several alternatives for managing solid waste and wastewater streams generated by the olive oil industry were analyzed by Reference [73]. Pellets manufacturing for domestic water heating rank first for environmental benefits, while briquette manufacturing ranked second for use in domestic stoves for space heating [73].

Olive leaves and olive pomace can be composted to produce a high-quality soil fertilizer as demonstrated by many experiments conducted in pilot study and full-scale plants by Chowdhury et al., 2014 [74]. The produced compost has also low thermal conductivity less than 0.60 W/m^2 K, therefore, can be used as cheap natural light weight fertilizer and good insulator to save energy in green roof buildings [75]. Important parameters that affects the composting process such as total pile volume, moisture content, particle size, porosity, nutritional balance, temperature, aeration, and pH have been extensively studied [76].

3.3. Other Solid Byproducts

A small amount of twigs and leaves are also generated at first step where the olives are washed and cleaned which account for 5% of the weight of the olives [77,78]. However, these byproducts can be used as low-cost fuel resource for heating and cooking in countryside in addition to animal feed. Olive leaves are also recognized as antioxidants resource of which can be extracted and sold to be used in herbal tea with anti-hypertensive, antioxidants, and diuretic effect [14]. Olive leaves antioxidants have also been assessed to determine their capacity for preventing oxidation of animal meat [79,80].

4. Olive Mill Waste Management Practices

There are no exclusive solutions exist yet, but several options have been recommended for valorization of olive mill waste streams. Several factors are to be considered when selecting the best management practice. These include the quantity of waste to process, required investment in infrastructure, available land for application, agronomic benefits that follow and the local regulations. Many countries in the Mediterranean faces' desertification, therefore, the application of organic matter to agricultural soils is very popular and can help recover topsoil fertility and slow down soil erosion processes in hilly areas. In organic farming, the use of these natural byproducts represents a significant plant fertilizer value and contributes to close the residual resources cycle. Consequently, the use of olive mill by-product as fertilizers and soil amendments on olive orchards will preserve the olive trees ecosystem and contribute to sustainable agriculture.

4.1. Characteristics, Treatment, Valorization and Utilization of OMWW

The olive oil production in the Mediterranean region is around 2 million tons annually, resulting in approximately of 30 million m^3 of OMWW and 20 million tons of olive pomace [76]. Oil mill wastewater (OMWW) has reddish-black color because of the existence of high concentration of phenolic compounds (10,650 mg/L), a strong unpleasant odor, a high concentration of fats, oil and grease (FOG) of (10,650 mg/L) and an extremely high organic load (COD and BOD_5) [13]. This organic load is approximately 400 times higher than that of typical domestic wastewaters [13]. Additionally, OMWW is acidic (pH 4 to 5), has an electrical conductivity (EC) ranging between 5.5 and 12.0 dS/m and a high concentration of polyphenols [9,10].

Quantities of byproducts generated, and their characteristic are a function of the extraction method used [4,5,29]. Characteristics of OMWW and pomace from a 3-phase extraction process are presented in Table 2. The chemical characteristics of OMWW prevents their direct discharge into domestic wastewater treatment plants [15,81,82]. If applied to soils or disposed in Wadis (water streams) without proper treatment, OMWW can cause serious environmental problems. Land application of OMWW has been widely used because of its valuable plant nutrients (nitrogen, phosphate, potassium, iron and magnesium) and high organic content [14,83,84]; even though some chemical components in OMWW might result in soil and water pollution, in addition to the risk of phytotoxicity [85–88].

The addition of OMWW and pomace to agriculture land might increase land yield by increasing soil organic matter and by providing valuable nutrients such as (nitrogen, phosphate and potassium) as demonstrated by several studies [10,37–39,56,58,89–92]. This is very important for most soils in the Mediterranean agricultural lands, which have deficiencies in soil nutrients and organic matter [37,57,93]. The organic content of OMWs can improve the structure of soil aggregates and, consequently increase soil porosity and water retention capacity [94,95]. However, an incorrect application of OMWs might result in reduction of crop yield due to temporary immobilization of mineral nitrogen [57].

Table 2. Chemical Composition of oil mill wastewater (OMWW), Pomace and Composts [8].

Parameters	OMMW	Wet Olive Pomace	Composts
pH	4–6	5–7	50–10
Dry matter (%)	6–7	50–71	
Water (%)	83	70	20
BOD (g/L)	35–110		
COD (g/L)	40–220		
EC (dS/m)	5–12	1–5	2–7.3
Organic matter (g/kg)	46–62	840–980	260–900
TOC (g/kg)	34–40	490–540	110–580
TN (g/kg)	0.60–2.10	7–19	11–54
C/N	52–54	28–73	9–36
p (g/kg)	0.15–0.30	0.7–2.2	1–30
K (g/kg)	2–9	7–30	6–44
Na (g/kg)	0.1–0.4	0.5–1.6	2–41
Ca (g/kg)	0.20–0.6	1.5–9	7–72
Mg (g/kg)	0.04–0.22	0.7–4	1–57
Fe (mgkg)	18–120	80–1470	100–410
Cu (mg/kg)	1.5–6	12–29	1.5–80
Mn (mg/kg)	1–12	5–39	13–130
Zn (mg/kg)	2.4–12	10–37	38–138
Phenols (%)	1–11	0.5–2.4	0.1–4

OMWs spreading should be applied cautiously because the unstabilized organic matters might inhibit or reduce crops growth. This might be attributed to phenols, fatty acids, tannins and high C/N ratio leading to a nitrogen competition between crop roots and soil microorganisms and roots anoxia because of oxygen consumption by microorganisms' [11,19,20,95,96]. The phytotoxic and antimicrobial impacts is primarily due to phenolic compounds ability to mix with other organic materials such as proteins, therefore changing the cell membrane permeability and intercellular transfer mechanisms [59]. At the same time, substantial amounts of organic compounds, potassium and phosphorous are added to the cultivated soils as organic fertilization [58].

Several studies have reported many advantages of rational use of OMWs as soil amendment without taking in their account potential dangers for crops and environment, on the other hand other studies showed negative effects on soil and surrounding environment mainly surface and ground water. These differences between these studies might be because of dissimilar experimental circumstances such as: soil type, cultural practices, methods of spreading, dosages used, crop phenological stage, climatic conditions and ground water depth. To avoid any problems a rational use of OMWs require proper application method and respecting regulations (if exist) [25,58].

4.2. Potential Valorization Options for Olive Mill Wastewaters (OMWWs)

Research is oriented toward flexible and efficient treatment of OMWW to degrade phenolic compounds and organic matter, to reduce its phytotoxicity and chemical oxygen demand [63]. OMWW management and treatment are facing many difficulties such as: high territorial scattering, seasonal operation, its high organic content, phenols and long chain fatty acids which are not easy biodegradable [97].

Treatment that could ensure solutions for the depollution of the waste, should compensate high capital investment and operation costs with the opportunity of recycling and recovering some valuable components. Many depolluting treatment technologies are existing; however, they require a high investment costs and a high level of technological know-how.

Further integrated approaches should be established combining minimization, recycling, treatment, valorization and energy generation measures to recover highly valued materials and reaching the safe limits of purified effluent to be reused later in irrigation, construction and industrial applications.

One of these methods is the biorefinery approach [98], which makes olive oil production process environmentally friendly and the treatment cost effective.

Biorefineries include a wide range of technologies to separate main biomass components (lignin, carbohydrates, protein, etc.) and convert them into biofuels and chemicals. The approach has been applied to several biomasses and a broad spectrum of different large scale biorefineries using a single feedstock. Benchmark and sustainability study of OMWW treatment methods was performed by Reference [99], indicating that the most useful methods for reduction of organic compounds are photo-Fenton, electrolysis, membrane filtration and supercritical water oxidation. Reduced environmental impact was obtained by coagulation, anaerobic digestion and lime treatments. The lowest cost-effective processes are membrane filtration and composting because of the added value of antioxidant phenolic compounds recovered and composts material, respectively [100].

Numerous studies have shown that phenolic compounds have many biological properties such as: antioxidant, anti-inflammatory, anticarcinogenic, antimicrobial activities and free radical scavenging [100,101]. Olive oil by-products bioactive characteristics were assessed by Reference [102], the study reported a large variety of biological activities including the antiallergic activity of OMWW, antibacterial activity of leaves and flowers, in addition to the collagen production, therefore have the potential for more development to be used in pharmacies applications and skin care industry [102].

4.3. OMWWs Treatment Methods

The difficulties in installing a waste treatment facility to treat olive mill by-products are related to its high organic loads which is hard to biodegrade, seasonal operation of olive mills (October-January), regional scattering and the high treatments cost particularly for small-middle size enterprises. OMWW must be treated preceding its discharge to the environment. All known treatment methods used for industrial and domestic wastes treatment have been tested on OMWW but unfortunately none of them seemed completely appropriate to be implemented in OMWW treatment.

Many treatment approaches were tested for OMWW treatment, such as physical methods (filtration, ultrafiltration/reverse osmosis and evaporating ponds) [81]; chemical methods (electrolytic oxidation, Fenton oxidation, neutralization with lime and combustion); physiochemical methods (floatation, adsorption, sedimentation, settling, sand, membrane infiltration and coagulation–flocculation, ion exchange and electrocoagulation) and biological approaches (anaerobic or aerobic degradation, biofiltration, activated sludge, composting and lagooning or direct irrigation on soils) [81].

Physical and physicochemical treatment methods lack sustainability, while the biological methods are effective and viable but they are not appropriate due to slow degradation rates of OMWW that is, requires a longer lag phase. Some of the earlier proposed treatment methods are applied in real olive mills after laboratory investigations, unfortunately none of them was cost effective and technically feasible [103]. However, the use of a combination of these technologies has indicated a satisfactory result.

The application of biological methods (anaerobic/aerobic digestion and composting) could be a possible solution for treatment of OMWW, especially if the waste high toxicity is reduced first. This might be achieved with advanced oxidation processes (AOPs) [104]. AOPs are chemical treatment processes relies on highly reactive agents in the reactions, like hydroxyl radicals causing the destruction of the targeted contaminant [105]. AOPs technologies have been effectively used for strong industrial pollutants, such as pesticides, phenolic compounds, surfactants, coloring materials and in pharmaceuticals industries [106]. Normally, they are used as pre-treatment steps in biological treatment methods to reduce toxic organic compounds concentrations which slow down the biological activities [8].

Constructed wetlands are currently used to treat different wastewaters such as municipal, industrial and food-industrial [107–111] and recently has also been used for treatment of OMWW [112]. High organic contents of OMWW required employment of several pre-treatment stages such as coagulation [113], electrochemical oxidation [114] and biological trickling filters [110] which makes the constructed wetlands just a polishing treatment process. Some studies have reported high removal efficiency of organic matter (86–95%) using free water surface wetland systems, however the organic surface loads were (5–57 g/m^2/day) [110,115]. Constructed wetlands has other benefits such as low construction and operation costs mainly for systems which will operate for long times in addition to environmental and landscaping benefits [108–110,116]. However, constructed wetlands cannot be used everywhere because it is land-based process and need adequate space.

Unfortunately, complete reduction of OMWW pollutants cannot be obtained by applying single method. Many management alternatives have been developed for handling, treatment and valorization of OMWWs while reducing their environmental impacts. These methods are categorized as physical, chemical or biological treatment methods. None of them has been applied commercially since they have lacking sustainability in long-term basis [116]. Targeting the reduction of OMWW phytotoxicity, current methods used for treatment of OMWW are summarized below.

4.3.1. Physical Methods

Physical methods are normally used as a pre-treatment step to remove the enclosed solids particles.

Evaporation

The simplest and most economical wastewater management technique is the evaporation of water from storage ponds or lagoons. It is widely used method in the Mediterranean region where favorable warm climate conditions prevail, such as in Northern Africa, southern Europe and eastern Mediterranean countries. Lagoons are very simple and inexpensive, but risks of surrounding environment contamination exist (mainly soil and water) [57]. The process is very slow and requires large land areas; it can sometimes produce unpleasant odors as a result of the decomposition of organic materials and fermentation processes, attract insects and rodents and help their breeding, and groundwater contamination might occur if the lagoon is not properly sealed [11,59]. After solids settling, evaporation of OMWW produces a sludge rich in residual oil and organic matter, which can be used as a renewable energy resource instead of fuel for heating. Several studies recommended OMWW composting as a solution for this waste [11]. Lagooning is rapidly disappearing, so the transport of OMWW and olive pomace must be handled with suitable methods of transportation to avoid any leaks or sanitary incidents.

Direct Application to Soil

Several scholars have tested direct application of OMWW to soils as an organic fertilizer and analyzed its positive and negative impacts on soils. Advantages are linked to its elevated nutrients content, mainly potassium; while, negative impacts are attributed to its salt content (measured as electrical conductivity), low pH and inherent phytotoxicity caused by polyphenols [116]. Some organic compounds mobility in soils can be reduced by OMWW application on agriculture land. Its use was recommended to reduce the leaching of herbicides (clopyralid and metamitron) in cultivated lands as reported by Reference [117]. Also, its high anti-microbial capability may be beneficial in sanitizing the soil against some plant pathogens such as (Rhizoctonia Solani) as confirmed by Reference [117].

4.4. Physical-Chemical Treatment

Physicochemical methods are relatively cheap but they cannot eliminate the pollution load of OMWW completely. In these processes, chemicals are added to create electro-coagulation, precipitation and destruction of dissolved organic compounds. Calcium hydroxide, aluminum sulphate and lime pre-treatment are used to decrease pollution from OMWW because lime is available at lower cost compared to other chemicals [118]. Another approach applied by several mills is the Electro Osmosis Dewatering (EOD) method, which involves placing a colloidal material between two electrodes to extract water from OMWW. Sodium chloride was found to be a very effective electrolyte for use in the EOD technique [119]. Applied electro-coagulation using coupled iron aluminum electrodes accompanied with ozonation for treatment of OMWW treatment resulted in removal COD and TSS at 47.6% and 82.6%, respectively [120]. Additionally, they observed significant elimination of the high levels of organic compounds after 70 min of treatment. Srikanth et al., 2019 [121] used integrated purification of OMWW through pre-treatments using chitosan and alum coagulants and biological methods. They reported removal efficiencies of 57.2% and 62.9% for COD and phenols, respectively using Alum as coagulants which appears to be the best from technical and economical point of view with treatment cost of 5.86 €.m^{-3}.

5. Biotechnological Treatments of OMMW

Biological treatments, such as aerobic and anaerobic methods used microorganisms to degrade the complex organic material of OMWW and considered the most environmentally friendly approach. Aerobic methods have higher removal efficiency than anaerobic; but aerobic method required continues mechanical aeration which make the process costly [122,123]. However, these methods are sensitive to toxic substances that hardly biodegradable like polyphenols [123] which reduce the efficiency of these methods. However, in terms of economics and feasibility, these treatment methods have not yet been commercially available for small scale enterprises.

5.1. Aerobic Treatments

Aerobic treatment in the presence of natural occurring microorganisms (such as fungi, protozoa, bacteria and other microbes) is an effective method for treatment of OMWW to reduce its pollution. In aerobic processes, oxygen is provided either by agitating the reactor or by supplying air by compressor. It is based on targeting the degradation of phenolic compounds, the major contributor to phytotoxicity. These options are scientifically appealing; however, they are not widely used commercially possibly because of cost considerations [10]. The aerobic method cannot detoxify OMWW inhibitory compounds such as polyphenols and lipids [124]. The capital investments in aerobic methods are lower than anaerobic methods but the running costs are significantly higher due to the need of continuous air supply.

5.2. Composting and Cocomposting of OMWs to Produce Soil Amendments

Composting is nature's way of recycling, it biodegrades organic wastes, such as crop residues, leaves, grass trimmings, wood, food waste, cartons, feathers and turning them to a valuable organic fertilizer. Composting is natural biological process where various microorganisms break organic matter down into simpler substances under controlled aerobic conditions. Composting of organic materials is a recovery system to produce high-quality organic amendments to be used in agricultural land to replace animal manure or chemical fertilizers application to intensive crop cultivations and the peat moss in nursery [11].

Composting is the primary choice for management of OMWs to convert it to fertilizer, thus returning nutrients to agricultural soils. Composting also reduces harmful effects of nutrient overloading or phytotoxicity noticed when spreading untreated OMWW directly into agricultural land. Being a liquid, the OMWW must absorbed by a firm substrate such as lignocellulosic matrix before proceeding to the composting process. Literature review showed that compost obtained from OMWs is of exceptional quality [76,125,126].

Composting has gained momentum over the past few decades as soil amendments because agricultural soils continue to lose their fertility and the use of soil amendments is needed to replenish organic material and nutrient content. Compost fulfils these needs at affordable prices, too. Additionally, compost can improve soil water capacity, cation exchange, increase microbial activity and reduce pesticide numbers [127].

5.3. Anaerobic Digestion

Anaerobic treatment of OMWW results in the conversion of organic matter to biogas (a mixture of methane and carbon dioxide, with other trace gases) that has significant energy value [125]. Residual solids and treated OMWW has value as a soil amendment, the potential use of OMWW in agriculture is one of the main goals of anaerobic digestion. The primary bottleneck in this process is the methanogenic bacteria inhibition by the presence of organic acids and phenolic compounds in the OMWW [7]. A pretreatment stage to remove undesirable compounds was recommended to be used prior to initiating the up flow anaerobic sludge blanket (UASB) treatment system [30]. Also, Filidei et al. (2003) suggested pretreatment by sedimentation and filtration preceding anaerobic digestion as a practical approach for OMWW treatment [127]. Ammary (2005) used anaerobic sequencing batch reactors (ASBR) for OMWW treatment and reported an 83% reduction of COD [128]. Furthermore, another researcher used an UASB reactor for OMWW treatment and reported COD removal in the range 75–85%, at 5 days of Hydraulic Retention Time (HRT) and influent COD concentration of 40 g/L [129]. Khatib et al., (2009) used an UASB reactor and achieved COD removal between 46–84% and organic load reduction from 27 g/L to less than 5 g/L [130] which permits the treated OMMW to be discharged to municipal wastewater treatment plants.

5.4. Biophysical and Biochemical Treatments Options

A combined aerobic biodegradation followed by ultrasonic irradiation treatment was used for the reduction of toxic phenolic compounds in OMWW [131]. They reported 81% degradation of total phenols and 80% reduction in influent COD when OMWW was exposed to an ultrasonic field for 90 min. Advanced oxidation with ozone (O_3), aerobic biodegradation and photo-degradation by UV radiation for COD elimination were reported by Reference [122]. They observed removal of 91% of influent COD in biological and UV/O_3 treatment on pretreated OMWW.

6. Valorization Options for Two-Phase Olive Mill Waste

The two-phase olive mills reduce water consumption and produce only wet pomace with high moisture content (65%) called "alperujo" in Spain. The material is stench and has fluffy texture that makes its handling and transportation not easy [20,48]. Its management requires special measures such as: storage tanks equipped with drainage valves, pump and septic container [11].

6.1. Physical Chemical Treatments—Second Extraction of Oil after Drying

It is possible to obtain olive oil residue from alperujo by further extraction with solvents after drying process but the high moisture content of the alperujo requires high energy input and causes some technical problems. The high cost of the drying process because of greater energy requirement in addition to lower demand for second extraction oil has led researchers to consider unconventional techniques for its extraction [8].

6.2. Potential Thermal Energy

Thermal or electric energy by combustion of exhausted olive cake after the second extraction is generally used as fuel in husk mills [132]. Presently dried husk combustion is applied in majority of the mills because it contains reasonable high calorific value (23 MJ/kg) [7]. Two different potential energy recovery alternatives of the solid and liquid residues delivered by the olive oil industry. The first one is the upgrading of olive oil industry solid waste through pelletizing, briquetting and torrefaction; and the thermochemical conversion of olive oil industry residues, through pyrolysis and gasification (thermochemical routes) [132].

6.3. Direct Application to Soil

Alperujo has been applied directly to near agricultural lands as soil amendment because of its elevated potassium content and therefore no transportation is required. Even its toxicity is less than OMWW; it might cause some imbalances in soil nutrition as shown by several studies, because of its high C/N ratio which modifies the nitrogen cycle especially for acidic soils [133,134].

6.4. Biotechnological Treatments

6.4.1. Anaerobic Treatment

Biogas production and partially stabilized organic substance are obtained during anaerobic process [10]. Biogas can be used for energy generation of and the organic substance can be used as soil amendment. It is estimated that methane production from olive husk biogas reached 75–80% [135].

6.4.2. Solid Fermentation

Alperujo has been also used in livestock nutrition but because of its low proteins content, it is advice to add protein supplement to the livestock feed [136]. Solid fermentation also improved nutritional properties as reported by Reference [61]. They found and increase in protein percentage in olive husk between 6–40% by the solid fermentation treatment. Started by microorganisms in the solid medium, the process has been applied successfully for animal feeds, enzymes and fuel production.

6.4.3. Composting

Due to its semisolid consistency, several researchers recommended the co-composting of a mixture of alperujo with other agricultural wastes such as straw, sawdust and bark chips as bulking agents before composting. The ending product contained substantial amounts of mineral nutrients, no phototoxic effects and good humification. The composting can be considered as affordable approach to recycle of olive oil byproducts, which represent a good alternative to combustion. It is recommended to compost it with available manure in the neighboring areas around the olive mills [137].

6.5. Valuable Products Extraction

Cardoso et al., (2003) reported positive results in studying the economic feasibility of pectin's extraction from alperujo, which might be considered as cheap substrate for the extraction of important compound such as pectin's [138]. These compounds are usually used as stabilizers, gelling factors and emulsifiers in food industries [11], therefore a new strong source of pectin, using agricultural wastes as raw materials. It is reported also that alperujo is excellent source of phenols with different biological behaviors. Obied et al. (2005) reported that total phenols reached 98% in olive mill waste [139]. Tyrosol, hydroxytyrosol, caffeic acid, oleocanthal and oleuropein are the main phenolic compounds [140] found in OMWW that can be used in the pharmaceutical industry. Many scientific researches confirmed the antioxidant, cardio protective, anti-microbial, anti-hypertensive and anti-carcinogenic behavior of these components, which might be entered in cosmetic, pharmaceutical and food productions. New technologies have been emerged to improve extraction of these compounds [141].

7. Additional Valorization Options

Many very interesting but not widespread ways of valorization the olive mill waste have been developed. For example, Pagnanelli et al. [141] recommended its usage as a heavy metal sorbent substance in aqueous solutions treatment. It is applied to agricultural lands to strengthen and extend sorption of insecticides (imidacloprid) and herbicides (simazine), thus reducing their biodegradation, slow down their leaching which reduces groundwater pollution risk [140,141].

A new material for the manufacturing of containers by recycling and mixing alperujo with thermoplastic polymers was studied by Reference [141]. A comprehensive approach for recycling olive pressing byproducts was proposed by Reference [115]. Many Marketable compounds can be extracted which are valuable in pharmaceutical and cosmetic industries. Purification of wastewater and solid residues composting are also included in this integrated approach which can be used in olive orchard fertigation.

8. Conclusions

In this study the current management's methods for olive mill wastes and treatment methods and techniques applied for the management of olive mill wastewater and solid waste in major oil producing countries were investigated. Due to favoring warm weather conditions and large open areas in the MENA region, liquid waste disposed in evaporations lagoons equipped with suitable mitigation measures such as rubber lining to prevent any leakage to ground water and alleviate any environmental impacts is recommended for OMMW pretreatment followed by constructed wetland treatment which are financially affordable by mill owners.

Centralized treatment plants are considered more suitable to treat OMWW, but it is accompanied with extra high transportation costs. Also, the olive mills are geographically scattered around country sides, which put some difficulties on building centralized large-scale treatment plants or management systems. Byproducts streams have potential economic value that remains to be utilized such as fertilizers after pretreatment and composting. Appropriate legislation is required to control application rate of untreated OMWW to the agricultural lands. Also, olive pomace is a valuable renewable energy source with an energy density of 23 MJ/kg and has become an inexpensive alternative for fossil fuels which used for home heating in country sides.

The solution to the OMWW disposal problems needs full cooperation of all partners involved in olive oil processing industry and consolidate their efforts. Two phase system should be adopted to minimize OMWW generation from the three phase systems. National planning is needed to implement a master plan for OMWW management on the ground. This will be accomplished by connecting all stakeholders through planning, research, regulatory, institutional, financial, and technical means. The key factors for successful of OMWW management require a suitable legislations, proper inspection, enforcement, and proper disposal and treatment facilities. An integration of these alternatives, with law enforcement support, will help solve issues with olive mill waste (OMWW) management in the region.

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