Rev Environ Sci Biotechnol (2008) 7:1–26
DOI 10.1007/s11157-007-9120-9

REVIEW PAPER

Olive oil history, production and by-product management

Iosif Emmanouil Kapellakis · Konstantinos P. Tsagarakis · John C. Crowther

Received: 14 July 2006 / Accepted: 8 March 2007 / Published online: 17 April 2007

Abstract In this review paper, the history of the olive tree and the development of the main olive oil extraction systems employed in the past are presented. Furthermore, the management techniques employed from antiquity until today for the treatment of olive mill wastewater (OMW), which constitutes the most important by-product of olive oil production, are addressed and compared. Reference is also made to global olive oil production, the environmental impacts of the uncontrolled disposal of OMW, and to the legal framework concerning its management in the main olive oil producing countries. Other by-products of olive processing are also discussed.

Keywords Conventional treatment processes · Legal framework · Natural treatment · Olive mill history · Olive mill wastewater history · Olive oil production

I. E. Kapellakis (✉)
National Agricultural Research Foundation, Iraklio 713 07, Greece
e-mail: kapellakis@in.gr

K. P. Tsagarakis
Department of Environmental Engineering, Technical University of Crete, Chania 73100, Greece
e-mail: kandila@her.forthnet.gr

J. C. Crowther
School of the Built and Natural Environment, Glasgow Caledonian University, Glasgow, UK

1 History of the olive tree

The olive tree is amongst the oldest known cultivated trees in the world (Liphschitz et al. 1991). In the past, it was the symbol of friendship and peace among nations. As early as the 7th century BC, winners of the Olympic games were awarded a wreath of olive branches (Fiorino and Nizzi Griffi 1992).

Much information regarding the social and religious role of the olive tree is given both in Greek mythology and the Old Testament. One of the myths concerns the origin of the name of the city of Athens. In the conflict for the possession of the wider region (Attica) between Poseidon, God of the sea and earthquakes, and Athena, Goddess of wisdom, Athena won, as her gift of an olive tree was deemed more precious than Poseidon's gift of a salt spring. In the story of the flood in the Old Testament, Noah released a dove, which returned holding an olive branch, a sign of the retreating water and a symbol of the re-establishment of peace between God and human beings.

More than 30 species of olive trees are known (Siggelakis 1982). However, the origin of the species has been the subject of much debate (Loukas and Krimbas 1983). Although fossils dating back to the Tertiary period (1 million years ago) prove the existence of an ancestor of the olive tree in Italy (Boskou 1996), it seems certain that the olive tree, as it is known today, had its origin about 5,000 years ago in the region corresponding to ancient Persia and Mesopotamia and from there it spread to Syria and Palestine (Kiritsakis 1998; Di Giovacchino 2000). People living in these areas developed the cultivation of the olive tree and then brought it to North Africa by land or sea (Loukas and Krimbas 1983). Others believe that the tree originated in Africa, as ancient Egyptians used to practice the cultivation of olives (Balatsouras 1997a).

It was the Phoenicians who were responsible for the spread of the olive tree to western regions, as they traded with other maritime centres. Around the 28th century BC, it began to reach the Greek islands, Libya, and Carthage. The Greeks established colonies in other parts of the Mediterranean basin, such as in Spain, and hence introduced olive farming to these areas (de Graaff and Eppink 1999). The first significant improvement in olive cultivation occurred in the 8th and 7th century BC, when olives were cultivated more systematically (Fiorino and Nizzi Griffi 1992).

Later the Romans discovered olive trees through their contacts with Greek colonies in Italy. Although they were not admirers of olives and olive oil, the Romans were responsible for spreading the tree throughout their huge empire (Boskou 1996). The value of the tree lead the Roman agronomist Collumela to call the olive *the queen of trees* (Frankel et al. 1994). Romans used olive oil in their baths and as a fuel, but not for edible purposes, as they considered it a commodity of moderate quality (Fiorino and Nizzi Griffi 1992). The rise of the Roman Empire and the conquest of Greece, Asia Minor, and Egypt increased the trading channels around the Mediterranean Sea and olive oil became far more important, not only as a staple food, but also as a pharmaceutical and energy source (Chazau-Gillig 1994). In the 5th century AD, the expansion of olive growing revived again when maritime states began to flourish. In the 15th century AD, missionaries and early settlers introduced vines and olive trees to the newly discovered America. Vines spread everywhere, but olive trees were cultivated only in restricted areas in Chile, Argentina, and California, areas with similar climatic conditions to those in the Mediterranean.

2 Olive mill development and wastewater production: a parallel past

2.1 Historical development

The olive oil has been discovered by the *barefooted* and *unchitoned* primitive man, as he crushed accidentally fallen olive seeds and noticed that the segregated oil moistened and softened his tough-skinned sole (Sarakomenos 1930).

Expansion of olive tree cultivation was followed by a consequent expansion and development of olive oil extraction processes. Since ca. 5000 BC, people used to collect and squeeze olives in stone mortars (Di Giovacchino 2000). Through the outlet of an inclined stone basin, the olive paste was collected in a small pot and, after the addition of hot water, olive oil was collected from surface, as being lighter than water, (Melena 1983), and transferred to storage clay pots (Davaras 1976; Psilakis et al. 2003). Due to the fact that people produced enough oil to cover their personal needs, the olive oil production plants were initially located within houses and operated by the members of each family. This has been well evidenced during the Bronze Age in Crete (Kopaka 2005), as is indicated by the stone tools (Fig. 1), clay pots (Fig. 2), and wall paintings that were found at farmhouses and at the Palaces of Phaistos and Knossos (ca. 15th century BC). Stone tools were also found in other countries around Mediterranean Sea, such as Israel and Palestine (Dar 1996; Frankel 1999), Cyprus (Hadjisavvas 1992), and Syria (Bourbou and Bourbos 1997). In the former location, a remarkable abundance of prehistoric remains have recently been discovered in a parallel to the coast submerged belt (Galili et al. 1997). It has been reported that wooden shoes were also used for olive crushing (Psilakis et al. 1999). The flow diagram of the olive oil and waste production process during the Bronze Age is presented in Diagram 1.

During the Mycenean era (between ca. 1450 BC and ca. 1150 BC), the process used in olive oil production was similar to that of the Bronze Age. However, linguistic indications revealed that woven mats, similar to those used by the Egyptians (Alexakis 1998), were used in the olive oil production process (Psilakis et al. 2003). According to Faure (1976) and Isager and Skydsgaard (1995), crushed

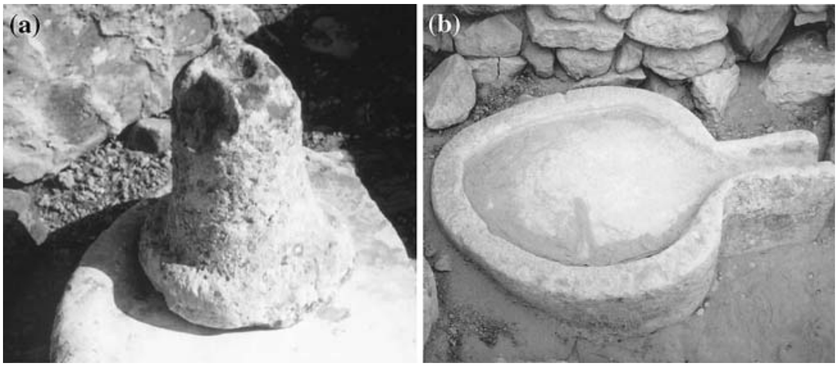

Fig. 1 Tools coming from the Minoan Palace of Phaistos and used for the extraction of olive oil from olives: (**a**) stone mortar and (**b**) oil collection pot

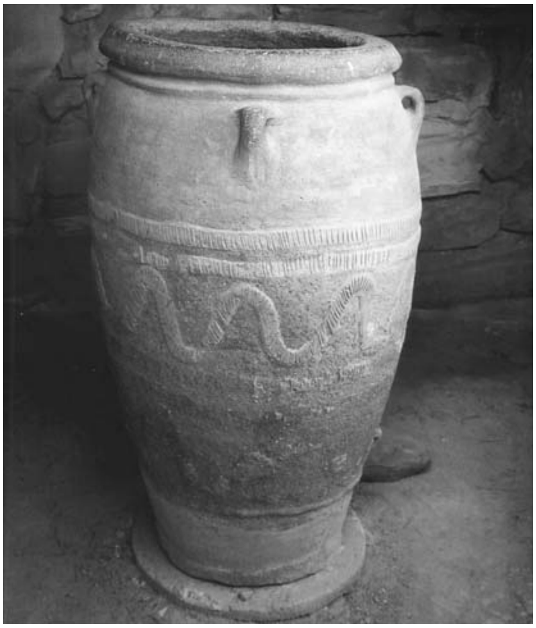

Fig. 2 A clay pot used for olive oil storage in the Minoan Palace of Phaistos

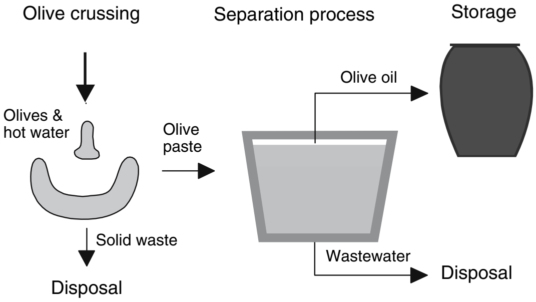

Diagram 1 Flow diagram of the olive oil and by-product production during the Bronze Age (not to scale)

olives were placed in woven mats and squeezed above the settling vats. To the authors' knowledge, mats have at least been discovered in Israel (Galili et al. 1997). The flow diagram of this process is presented in Diagram 2. During the Hellenistic period, the same olive oil production process was used.

However, as the time passed by, such production methods were not suitable for meeting the increased olive oil demand, due to the increased trading channels of the Roman Empire around the Mediterranean Sea. The Romans contributed to the technological development in olive processing by expediting the crushing operation with a millstone crusher, the *trapetum* (Fig. 3; named after the Greek word *trepo*, which means *revolve*), and improving the separation system with the introduction of presses (Kiourellis 2005). The wooden or iron manually-activated screw press, first used by the Greeks (50 BC) and later improved and disseminated by the Romans, represented major progress in olive processing (Di Giovacchino 2000). The flow diagram of the olive oil and by-product production process is presented in Diagram 3.

During the period followed by the fall of the Roman Empire and the Barbarian invasion, there were no innovations in olive processing, which continued to be based on the pressure exerted by a screw press (Di Giovacchino, 2000). In fact, Arabian sources claimed that in Crete the olive oil production during the Byzantine era must have been very limited (Tsougarakis 1990). During the Venetian period, olive oil production was increased, as Venetians used to encourage olive cultivation and oil production in their occupied territories (Tsougarakis 1990).

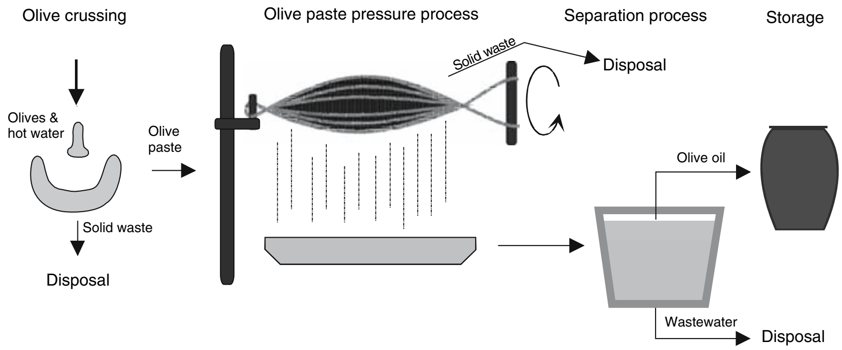

Diagram 2 Flow diagram of the olive oil and by-product production during the Mycenean era (not to scale)

Fig. 3 A trapetum, found in the broader area of Phaistos

Until the past three centuries, olive oil production was still a very difficult process and a great physical effort was required for olive crushing. However, the situation was improved by the discovery of rotating wheels (Fig. 4a) moved mostly either by horses (Coppa-Zuccari 1962) or by cows (Psilakis et al. 1999). The paste produced after the crushing of the olive seeds was still drenched with hot water, in order to achieve better separation of olive oil (Karidis 1983), and then was placed in oil diaphragms, called *boxades* (Fig. 4b). These oil diaphragms were then placed one over the other in the bottom side of the press (Fig. 4c). Then, after pressure was applied, the mixture of oil and water was extracted into a series of stone tanks, where oil was collected from the surface (Fig. 4d). The flow diagram of the olive oil and by-product production process is presented in Diagram 4.

A significant development on the olive oil extraction systems occurred in 1795 by Joseph Graham, who invented the hydraulic pressing systems (Balatsouras 1986). The expansion however, occurred during the second half of the 20th century, where a lot of technological improvements and innovations have been made.

Olive oil separators have replaced the traditional methods and the productivity has been increased with the wide-spread adoption of the hydraulic pressing

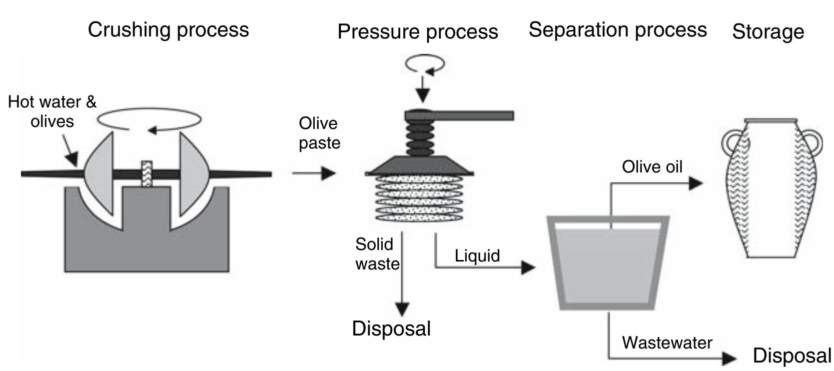

Diagram 3 Flow diagram of the olive oil and by-product production during the Roman era (not to scale)

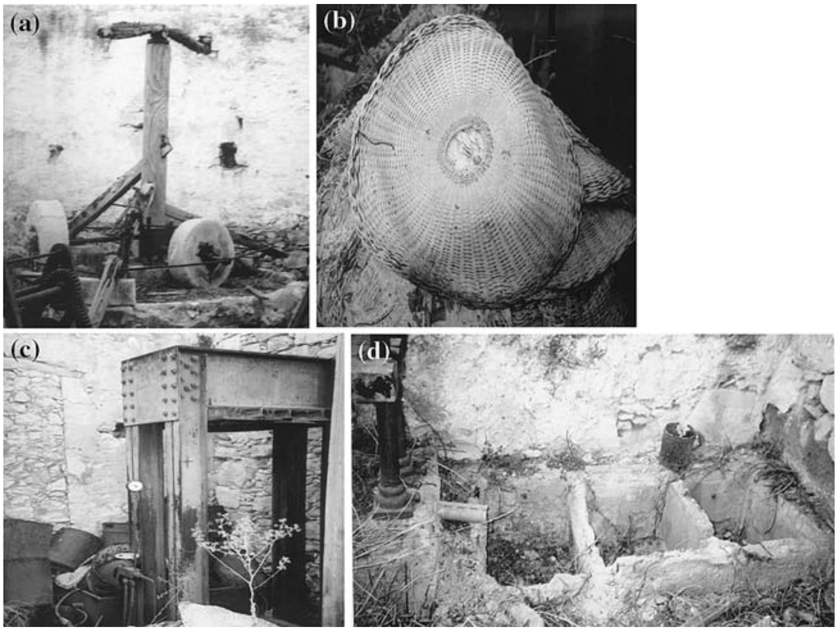

Fig. 4 Old OM: (**a**) Rotating wheels were used to crush the olives to produce the olive paste; (**b**) the oil diaphragms (*boxades*) used for putting paste from the rotating wheels were functioning as filters; (**c**) Oil diaphragms were placed one over the other in the bottom side of the press; and (**d**) the stone tanks were full of water and oil was collected from the surface as was passing through them

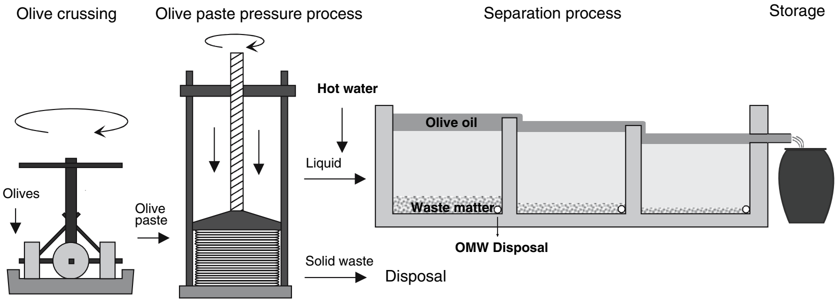

Diagram 4 Flow diagram of the olive oil and by-product production in the 20th century (not to scale)

systems, such as those in the island of Lesvos (Sifounakis 1994). However, even with this technology, productivity was not satisfactory. Today, the increased use of centrifugation rather than percolation as a separating method, results in an oil of higher quality and yield.

Nowadays, there are 600 million productive olive trees on the planet, occupying a surface area of 7 million ha (Molina Alcaide and Nefzaoui 1996). Olive oil is produced in those regions of the world where climatic conditions are as favourable as those prevailing in the Mediterranean countries. The Mediterranean region accounts for not less than 97% of the world production (de Graaff and Eppink 1999; Al-Malah et al. 2000; WWF 2001). The main olive-producing countries are Spain, Italy, Greece, Tunisia, Turkey, Morocco, Portugal, Syria and Algeria. The tree is also cultivated in France, Yugoslavia, Cyprus, Egypt, Israel, Libya, Jordan, Lebanon, Argentina, Chile, Mexico, Peru, the USA, and Australia (Table 1). The oil is no longer used for lighting except for religion purposes and in the preparation of pharmaceuticals and cosmetics.

2.2 Processing steps of the modern olive oil mills

Olive oil is present in the form of small drops in the vacuoles of mesocarpal cells in the olive fruit. It is also scattered to a lesser extent in the colloidal system of the cell's cytoplasm and to a lesser degree, in the epicarp and endosperm (Balatsouras 1999). To release most of the oil present in the olive during processing, it is necessary to use water in various steps. Water is used in washing, mixing (when olives

Table 1 Geographic distribution of world olive oil production (Di Giovacchino 2000)

Country/Region	Area[a]		Production[b]	
	Hectares	Percentage	Average[c]	Percentage
European Union	4,507	51.73	1,425	74.4
Spain	2,227	25.56	602.2	31.4
Italy	1,141	13.10	451.3	23.5
Greece	718.50	8.24	332.0	17.3
Portugal	400.00	4.59	37.4	2.0
France	20.00	0.23	2.3	0.1
Other EU countries	75.16	0.86	7.0	0.4
Tunisia	1,624	18.64	172.9	9.0
Turkey	881.00	10.11	92.0	4.8
Morocco	450.00	5.17	46.3	2.4
Syria	421.50	4.84	81.0	4.2
Algeria	206.28	2.37	29.0	1.5
Argentina	40.00	0.46	9.1	0.5
United States	15.80	0.18	1.4	0.1
Others	490.52	5.63	52.1	2.7
Mediterranean countries	8,606	98.80	1,874	97.65
Total	8,711	100.00	1,916	100.00

[a] 1,000 ha

[b] 1,000 tonnes

[c] Average for the period 1990–1997

are entirely dry) and diluting the paste, and in the final separation of the olive oil. Water used in these stages corresponds to 10%, 40%, and 20% out of the initial olive weight, respectively. To show the mass yield, 1 kg of olive oil is produced after the processing of approximately 5 kg of olives. The main processing steps needed to obtain olive oil include: feeding, leaf removal and washing, crushing, mixing, separating the olive oil and centrifuging the oil. There are three different systems, depending on the extraction method: (a) pressure, (b) centrifugation, and (c) selective filtration process (percolation). A typical layout of the extraction process, based on centrifugation, together with the main processes is given in Fig. 5.

2.2.1 *Feeding, leaf removal and washing*

After their collection from the orchards, olives are put into a large feeding hopper attached to a moving belt. Removal of the leaves and washing are necessary in order to remove from the olives all foreign material that could be harmful to the machinery or contaminate the product, e.g. the presence of leaves imparts a bitter taste to the oil.

2.2.2 *Crushing*

Crushing is the first main step in olive fruit processing. The purpose of crushing is to tear the flesh cells to facilitate the release of the oil from the vacuoles. It consists of a large bowl in which two or three heavy wheels rotate at high speeds, crushing the olives.

2.2.3 *Mixing*

After the olive fruit has been crushed, the resulting paste is mixed. Mixing or malaxation entails stirring the olive mash slowly and constantly for about 30 min. The purpose of this operation is to increase the percentage of available oil. It also aids in the coalescence of small oil drops into larger ones, thereby facilitating separation of the oil and water phases. It aids in breaking up the oil/water emulsion drops as

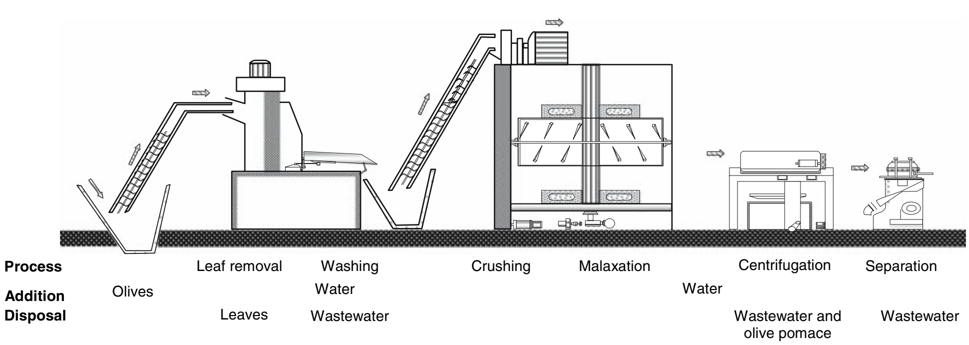

Fig. 5 A modern OM based on centrifugation process

well. Malaxators differ in shape, size and layout among olive mills (OM). To avoid oxidation, they are made of stainless steel. Depending on the inside available area, malaxators are divided in vertical and horizontal, with the former being less used due to additional access requirements. The basic apparatus units consist of cylindrical vats with rotating blades and double walls. A rotating helix with several wings mixes the paste, usually in low speeds (19–20 rpm) depending on the paste. For olives that have reached maturity, 20–30 min of malaxation is sufficient.

For greater efficiency, malaxators have double walls for circulation of the heating water. A temperature increase results in lower viscosity of the oil and greater olive oil yield (Kiritsakis 1998). However, the temperature of the water should be no more than 30°C to prevent destruction of volatile constituents, change of oil colour to reddish and an increase in acidity. To avoid the above negative effects, modern malaxators are equipped with automatic thermostats.

2.2.4 Olive oil extraction processes

The main constituents of olive paste before this step are: olive oil, small pieces of kernel (pit), water and cellular debris of the crushed olives. Three different extraction processes may be applied for the separation of oil from the other constituents. These are pressure, centrifugation and selective filtration process (Sinolea).

Pressure process As mentioned previously, pressure process is the oldest and most widespread method for processing olive fruit to obtain olive oil. A photo taken during the 1960s from an old OM is shown in Fig. 6. With major modifications, especially during the second half of the previous century, it is still used today. The invention of the hydraulic press was a revolution for the operation of old mills and hydraulic presses are still used in improved traditional mills. Machinery used for this process has been improved and is more powerful and reliable. The olive paste of 2–3 cm thickness obtained from the previous stages is placed uniformly in oil diaphragms, which are then placed in moving units (trolleys) with central shaft. A metal tray and a cloth without paste are placed after every 3–4 diaphragms to obtain uniform application and a more stable load. Then the moving unit along with its load is placed under a hydraulic pressure unit. When applying pressure, the liquid phases (oil and water) run through the olive cake.

Centrifugation process Centrifugation is a relatively new process for separation of oil from olive paste. It is based on the differences in density of the olive paste constituents (olive oil, water and insoluble solids). Separation is accomplished through a horizontal centrifuge (decanter). After crushing and mixing, the olive oil is either completely free or in the form of small droplets inside microgels, or emulsified in the aqueous phase. Free olive oil is separated by the centrifuge, while the oil locked in the microgels is released by adding water.

Decanters consist of a cylindrical conical bowl. Inside there is a hollow, similarly-shaped component with helical blades. A slight difference between the

Fig. 6 Pressure process OM in Crete (Courtesy of Mr. J. Daskalakis, Sivas 1967)

speed at which the bowl rotates and that at which the inner screw gyrates results in the movement of the pomace to one end of the centrifuge, while the two other constituents of the olive paste (oil and olive mill wastewater-OMW) are pushed to the other end. The oily must (oil with a small amount of water and water containing a small amount of oil) passes through vertical centrifuges which revolve at 6,000–7,000 rpm for the final separation of the oil. Each decanter has a maximum hourly capacity, depending on its manufacturing characteristics. In general, this capacity ranges from 1.5 to 6.5 tonnes per hour. When this capacity is exceeded, decanter performance is not efficient which results in poor separation of oil from OMW.

A factor that affects the oil yield is the amount of water added to the paste. The optimum amount of water needed to dilute the paste is determined by the characteristics of the olive paste and the centrifuge. It is estimated empirically by observing the oil, which should be clean, and the water, which should not be very viscous, as they flow out of the decanter. The main advantages of centrifugal processing systems are (Kiritsakis 1998):

- Limited labour is needed, since the process is continuous and automated;
- Stainless steel materials are always used and thus the oil is well protected from contamination;
- Since no diaphragms are used, the risk of contaminating the oil is eliminated; and
- Better yield performance, as most of the oil is collected.

The main disadvantages of centrifugal processing systems are (Kiritsakis 1998):

- Water and energy demanding, while a significant amount of phenols (natural antioxidants) are lost during the centrifuge process in the OMW;
- The olive pomace contains a high percentage of moisture; and
- Increased production of OMW, which is approximately 50% more than the pressure process.

Decanters, with minor modifications can operate either as three-phase or two-phase systems. The former produce olive pomace, olive oil, and OMW in a portion of 40%, 20% and 100% of the weight of the olives, respectively, while the latter produce a better quality of olive oil as it does not consume water for the oil separation and the resulting two phases comprised olive oil and a slurry olive pomace containing a small part of wastewater, which is 10% of the olive weight, or alternatively 10% of the total volume of OMW generated in the three-phase. The two-phase systems have been called ‘‘ecological’’ decanters. However, this characterisation is not correct, as the resulting slurry pomace cannot be managed easily, while the polluting load is more concentrated. Another disadvantage of the two-phase systems is due to the increased moisture content when olive pomace has to be further processed to produce olive pomace oil. Olive pomace obtained in two-phase systems has to be heated to higher temperatures than that obtained in the three-phase systems, and risks the generation of carcinogenic substances (benzo-a-pyrene) within this oil.

Selective filtration process This process is also called Sinolea, after the company which first introduced this method of phase separation. Besides pressure and centrifugation, selective filtration combined with centrifugation is used for the separation of olive oil from the olive paste. It is based on the different interfacial tension of oil and water coming in contact with a steel plate. When the steel plate is plunged into olive paste, it will be coated with oil due to the lower interfacial tension of the oil than that of the water. The most common selective filtration unit is the Sinolea unit. It is made up of one or more units that can handle about 350–370 kg of olive paste, each of which has a stainless steel grating unit at the bottom. Five to seven thousand moving sheets are slotted through the slits in the grating unit and slowly penetrate the paste in reciprocating motion. When the steel plate is moved into the paste, it will preferably be coated with oil which then drips off, creating a flow of oily must consisting almost solely of oil. When this process is repeated many times, most of the oil from the paste is recovered. The oil obtained by this process is called Sinolea oil and can be considered as prime oil. To extract the remaining oil, the olive paste is further moistened, undergoes new mixing and is subjected to centrifugation, with the process described in previous paragraph. The oil obtained by this process is called Decanter oil and the combination of the two processes results in a high yield. However, combined selective filtration and centrifugation is not used broadly due to high operational and maintenance costs, high energy demand, and the frequent need to clean the moving sheets.

2.2.5 *Final centrifugation of olive oil*

Irrespective of the process applied for oil separation from the other constituents of the olive fruit, a final centrifugation of the oil is needed. Separation takes place in a vertical centrifugation unit that rotates at lower speeds than the previous one described. This unit consists of a stationary part and a mobile part which turns very quickly. A large number of cone-shaped disks are attached to the centrifugal unit. The liquid phase is distributed on the total surface in thin layers and the centrifugation in the vertical centrifuge results in a final separation of the oil from the water and other substances.

2.3 Olive oil by-products

During olive processing three main residual products are generated. These are the olive pomace, the twigs and leaves, and the OMW. The flow diagram of the by-products of olive processing is presented in Fig. 7.

Olive pomace is the remained pulpy material after removing most of the oil from the olive paste. The commercial value depends on its oil and water content: The three-phase pomace, with low moisture content, has a better commercial value than the one obtained in a two-phase process system. Regarding the improvement occurred during the last decades, it

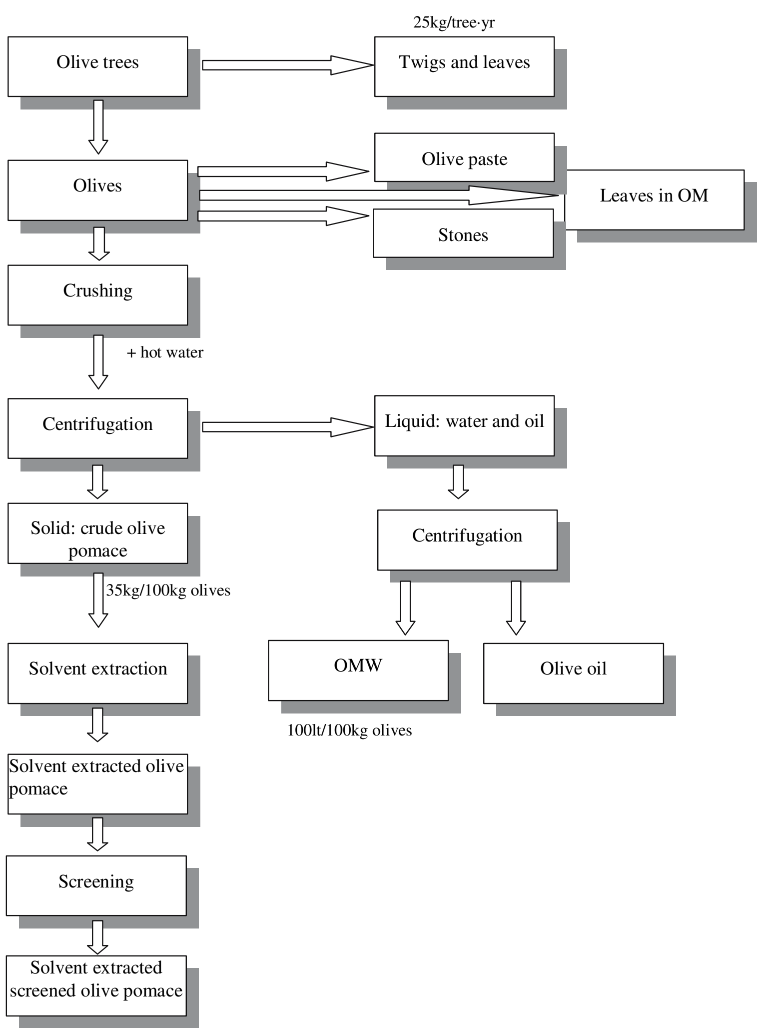

Fig. 7 By-products of the olive tree culture and olive oil industry (Molina Alcaide and Nefzaoui 1996)

Table 2 Composition of OMW (Michelakis and Koutsaftakis 1989; Balatsouras 1997b; Tamburino et al. 1999; Antoniou 2001)

Constituents	Processing system	
	Pressure	Centrifugation
pH	4.5–5	4.7–5.2
Polluting load		
COD (g/l)	120–130	60–180
BOD_5 (g/l)	90–100	20–55
SS (%)	0.1	0.9
TS (%)	12.0	6
Organic constituents (%)		
Sugars	2.0–8.0	0.5–2.6
Nitrogen compounds	0.5–2.0	0.1–0.3
Organic acids	0.1–1.5	0.2–0.4
Polyalcohols	1.0–1.5	0.3–0.5
Pentoses, tannins	1.0–1.5	0.2–0.5
Polyphenols	2.0–2.4	0.3–0.8
Lipids	0.03–1.0	0.5–2.3
Inorganic constituents (%)		
P	0.11	0.03
K	0.72	0.27
Ca	0.07	0.02
Mg	0.04	0.01
Na	0.09	0.03
Cl	0.03	0.01

is worth mentioning that a traditional OM dating back to 1975 produced pomace with an oil content of more than 15%, compared to 3.5% of modern OM (Koutsaftakis and Stefanoudaki 1994). Leaves and twigs are approximately 25 kg/tree·yr. They do not have any commercial value and they are either used in animal nutrition or are landfilled. OMW is described in the following pages.

3 Olive mill wastewater

3.1 General characteristics

OMW (*aqua reflue* in Italy; *alpechin* in Spain; *katsigaros* in Greece; *zebar* in Arab countries) is a dark red to black-coloured, mildly acidic liquid of high conductivity, obtained from mechanical olive processing during olive oil production. The negative environmental effects for olive oil producing countries are more noticeable today, mainly due to: (a) the industrialisation of agriculture which has brought about a noticeable increase in olive oil production worldwide during the last thirty years, (b) the conversion of pressure-type OM into centrifuge-type OM, (c) the dispersed location of a great number of small sized OM, and (d) the increased sensitivity of the public to environmental problems. It is very difficult to calculate the volume of OMW produced worldwide, as this depends on many parameters, such as the olive variety, olive seed maturity, cultivation techniques, and geological-climatic conditions.

However, it is strongly dependent on the processing system: In pressure systems the volume produced varies from 40 to 60 l per 100 kg of olives, while in two-phase systems it is approximately 10 l per 100 kg of olives and in three-phase centrifugation systems it varies from 80 l to 120 l (average 100 l) per 100 kg of olives.

OMW consists of water (83–92% by weight), sugars, other organic substances, including readily fermentable proteins, organic acids (acetic, fumaric, glyceric, and oxalic acid), small amounts of emulsified olive oil, phenols, waxy and resinous substances, vitamins, and traces of pesticides (Table 2).

Phenolic compounds include many organic substances that all have the common characteristic of possessing an aromatic ring with one or more substituent hydroxyl groups and a functional side chain. These phenolic compounds are responsible for the toxicity of OMW towards bacteria, plants and animals. Phenols are not found in their free forms in olives, but mostly as glucosides, tannins, antocianins, and lignin, and, in comparison to other constituents, they are present in small quantities at concentrations which range from 50 mg/l to 200 mg/l. They play an important role in the protection of the plant against oxidation, UV-radiation, and micro-organisms. The 50 phenols that have already been identified in OMW belong to three important categories of phenolic compounds (Miranda et al. 2001): (a) cinnamic acid derivatives, (b) benzoic acid derivatives, and (c) compounds related to tyrosol (Table 3).

3.2 Environmental impacts

As OMW is produced from the diluted juice of crushed olives, it can be safely assumed that it is completely biodegradable. However, phenols and

Table 3 Categories and main phenolic compounds

Categories	Cinnamic acid derivatives	Benzoic acid derivatives	Compounds related to tyrosol
Compounds and families	Cinnamic *p*-Cumaric Caffeic Ferulic	Benzoic *p*-Hydroxybenzoic Protocatechuic Vanillic Veratric Gallic Syringic	Tyrosol Hydroxytyrosol *p*-Hydroxyphenylacetic

lipids decompose at reaction rates much slower than other constituents, such as sugars or short chain volatile acids. This resistance to biodegradation together with the high organic content of OMW results in it being responsible for several environmental impacts. The most important environmental impacts of OMW are the following:

(a) Threat to aquatic life. OMW has a hundred times higher organic load concentration than municipal wastewater and when it is discharged into fresh water, the oxygen availability is reduced, causing an imbalance of the whole ecosystem. In addition, the large nutrient concentration in OMW promotes the development of algae and consequent eutrophication.
(b) Odours. Fermentation phenomena take place when OMW is stored in open tanks and/or discharged on the land or into natural waters. As a result, methane and other pungent gases (hydrogen sulphide, etc.) are emitted. This leads to considerable pollution by odours even at great distances, especially during the oil production period (Niaounakis and Halvadakis 2004).
(c) Impenetrable film. The lipids in OMW form an impenetrable film on the surface of water, which blocks out sunlight and oxygen to microorganisms in the receiving water, leading to a reduction of plant growth and increasing erosion.
(d) Discolouring of natural waters. The change in colour of natural waters can be attributed to the oxidation and subsequent polymerisation of tannins producing darkly coloured polyphenols, which are difficult to remove from the effluent.
(e) Toxicity. As OMW is characterised by the presence of several phytotoxic volatile acids and phenolic compounds, it is very toxic. Since phenols, together with alcohols, aldehydes, and organic acids, result in a very low pH, OMW must be treated in order to remove the phenolic fraction (Adhoum and Monser 2004).

3.3 Legal framework

The existing legal framework with regards to OMW highlights two significant problems. The first is the lack of a common and specific policy among the EU members. As OMW is produced only in the Mediterranean region, EU policy has not brought into force any common guidelines for OMW management. Therefore, the lack of a common policy has resulted in different legal frameworks dealing with agro-industrial effluents being implemented throughout the Mediterranean region. For example, guidelines for land application of OMW in Italy (Tamburino et al. 1999) and Spain (Cabrera et al. 1996) have long been adopted, but in Greece apart from being prohibited, each of the 52 individual prefectures has its own policy. In some cases, the policy between other prefectures is even contradictory (Kapellakis et al. 2003).

The second problem has to do with the lack of a framework for separating soil from water receivers. The presence of organic matter as well as many inorganic compounds (nitrogen, phosphorus, potassium) causes severe pollution when OMW is disposed of into water bodies, but in soil it prevents erosion and can be beneficial to soil fertility. The legal frameworks of the main olive oil producing countries, according to the data presented in Table 1, together with the EU legal framework, are presented.

3.3.1 Spain

By the end of the 1970s, OMW was produced in a three-phase process and the disposal of large quantities of OMW had become the main pollution problem in many river basins, such as in the Guadalquivir Basin, the main area of olive oil production in Spain. For this reason, the Spanish Government in 1981 established a legal framework to prohibit the discharge of untreated OMW into rivers and subsidised the construction of ponds for its storage during the milling period, aiming at the evaporation of its water during the warm Andalusian summer. About 1,000 evaporation ponds have been constructed in the Guadalquivir Basin, resulting in the improvement of water quality in the nearby rivers and streams (Cabrera et al. 1996). During the 1991–1992 harvesting season a shift from the three-phase to two-phase system was implemented. As its introduction coincided with a dry season, the elimination of the addition of water for the oil separation was very well received at that time by the Spanish olive oil sector (Alvarado 1998). Nowadays, more than 90% of the OM operate with the two-phase system. Similar management schemes have been employed by other Mediterranean countries (Herold et al. 2000; Ben Rouina et al. 1999; Ammar and Ben Rouina 1999; Garcia-Ortiz et al. 1999).

3.3.2 Italy

Unlike Spain, penetration rates of two-phase systems in Italy are negligible (<5%). Over 10,000 OM operate with the most common extraction technology still based on simple pressure. Until recently, the legal framework was based on Law No. 319/1976 (Borsani and Ferrando 1996), which established general wastewater effluent limits, irrespective of the characteristics and the use of the receiving water or lands. This was later amended by Law No. 119/1987, which allowed the application of partially treated (50% COD reduction) OMW onto the land. However, as treatment requirements were very difficult to achieve, the introduction of Law No. 574/1996 changed the management practices as it allowed the direct application of 50 m^3 OMW/ha yr from OM with pressure process and 80 m^3 OMW/ha yr from centrifuge systems (Rinaldi et al. 2003). Recently, special permission in Portugal also allows the spreading of OMW on land (Casa et al. 2003). Nowadays, although land application is promoted, Italian legislation has been and probably will continue to be a limit to the storage and land application of OMW, because of the small OMW quantity allowed for agronomic application.

3.3.3 Greece

The main principles for OMW management are based on Law 1650/86 ''*For the Protection of the Environment*'' (Government Journal 160/A/86). According to this Law, OM involve activities with high potential to cause environmental degradation, and the OM owners are obliged to develop an environmental impact assessment. The Law No. E1b/221/65 ''*Health Arrangement Action*'' (Ministry of Public Health 1965) focuses on the issue of the disposal of the treated effluent. Furthermore, the 271/91 EU Directive (EU 1991) was fully adopted by Greek legislation in the 5673/400/97 Ministerial Decision concerning limitations and conditions required for the treatment of municipal wastewaters (Ministry of Public Health 1997). Under the jurisdiction of the ''*The Transfer of the Competence for Approval of Environmental Conditions for Certain Projects to Local Prefecture Authorities*'' Ministerial Decision, Greek legislation gave additional authorities to the Prefectures. Each Prefecture was then responsible for adopting the various OMW management practices. For example, OMW management in the Prefecture of' Messinia is based on the modification of a three-phase decanter system into a two-phase, in the Prefecture of Lesvos OMW was discharged until recently untreated onto aquatic ecosystems (Halvadakis 2000), while in the Prefecture of Iraklio both the two-phase systems and disposal in the aquatic environment are forbidden.

In Crete, until 1987 OMW was discharged uncontrolled into the environment. It has been estimated that by that time 80–90% of the total OMW produced, was disposed of in ephemeral rivers (Voreadou 1994). Since then, local public authorities prohibited uncontrolled disposal in water bodies and obliged the owners of the mills to construct treatment units (obligatory since 1987) and evaporation lagoons (obligatory since 1993) (Kapellakis et al. 2002).

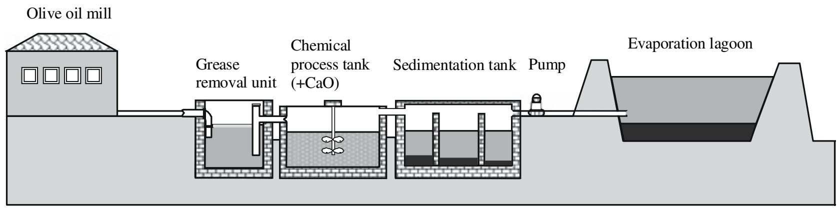

Fig. 8 Typical flow diagram of the OMW management scheme (not to scale)

OMW immediately after production has to pass through the treatment unit (grease removal, mixing with $Ca(OH)_2$ and sedimentation) and then has to be discharged into evaporation lagoons (Fig. 8). Both proposed units involve low construction and operation and maintenance (O&M) costs that do not require employment of specialised personnel. In cases when the law has been breached, the responsible authorities are the Port Police, in which accusations against the OM are anonymous and the Health Department, which belongs to the local authorities.

3.3.4 European policy

According to the European wastewater policy, the main reference to wastewater (WW) is given in Directive 271/91 on ''*Urban Wastewater Treatment*'' (EU 1991) (this was later amended by Directive 15/98). The objective of this directive was to protect the environment from the adverse effects of the discharge of urban wastewater and of wastewater from industrial sectors of the agro-food industry.

Amongst other things, it states that discharges of biodegradable industrial wastewater from certain industrial sectors that do not enter urban wastewater treatment plants before being discharged into receiving waters, should be subject to the appropriate treatment. Also, the recycling of sludge arising from wastewater treatment should be encouraged; though it is necessary to monitor receiving waters and the disposal of sludge to ensure that the environment is well protected from any adverse effects.

Thus, OMW treated effluent should be released into the receiving waters only when it meets the relevant quality standards and the provisions required by this Directive. The directive also states that treated wastewater should be reused whenever appropriate. Disposal routes should minimise the adverse effects on the environment.

4 Olive mill wastewater management

4.1 Evidence from the past

In the past, most of OMW production occurred in farmhouses located close to the olive orchards, rather than within towns. This might be due to the preference to carry olive oil rather than olive seeds from the olive orchards in the countryside to towns. Indeed, in present time olive oil production is 375 l/ha; for a yield of 15% of olive oil, the corresponding amount in olive seeds is 2,500 kg/ha.

During the Bronze Age, the content of the settling vat was discharged after olive oil collection. The resulted wastewater consisted of leaves and dirt (Sarakomenos 1930) as well as of a mixture of squeezed olive seeds with water. Since the olives were beaten off the tree and collected by bare hands (Davaras 1976), the quantity of leaves in the by-product stream was limited. The wastewater was characterised by high toxicity, distinctive smell, and dark red to black colour, and was discharged directly onto the soil. In fact, Balatsouras (1997b) insisted that the easiest and cost-effective method to manage OMW in antiquity was its disposal in fallow or arable land, or into ephemeral rivers. Similar findings have been reported by Hadjisavvas (1992), who reported that below the outlet of an olive press in Cyprus many olive pits have been discovered within a layer of blackened soil.

Regarding the Minoan Crete, in the cases where olive oil was produced within the Palaces, OMW was probably discharged into the sewerage system, similar to the system used in accidental leakages of olive oil in the storehouses (Alexiou 1964). This aspect is favoured by OMW's distinctive smell, which, due to inadequate ventilation, could make the atmosphere within the storehouse intolerable, as well as by its oily content that would have been responsible for a slippery floor. In any case, OMW was discharged onto the land or rivers outside the Palaces and the employees of the olive presses have witnessed the negative effects of the cumulative OMW disposal activity.

During the Mycenean and the Roman era, similar management practises to those of the previous period were employed. The difference lies on the woven mats, as they were permeable for the oil and water pressed, but able to retain the exhausted solid material paste (Isager and Skysdgaard 1995). Probably, when these mats were cleaned, the remained by-product was washed away and disposed of with the washing water in the soil. It is worth mentioning that there are many authors (i.e., Pliny the Elder; Varro Marcus Terrentius; Marcus Porcius Cato; etc.) who lived during the Roman era and left in their writings detailed information on the potential uses and environmental effects of OMW (Niaounakis and Halvadakis 2006a). On the contrary, the period after the Romans and until the recent years can be characterised by a limited interest on olive oil and OMW management practices. Therefore, OMW was probably managed according to the previous and/or to next periods.

During the 20th century, more information on OMW management is available. After the pressure process, the exhausted paste was gathered from the edge of the woven mats and transferred from the woven mats to a cavity. This paste was used as fuel matter, as animal forage, in cooking, or sold to olive pomace factories. Regarding the collected OMW from the settling vats, as it still contained a part of olive oil, it was stored either into specific pots or into excavated holes (Fig. 9). After long retention, olive oil was collected from the surface, while the sediment has been used in the livestock to sheep's protection from ticks. Shepards used to boil the water from the murga and then spread it with a flock of fleece into the sheeps to eliminate ticks (Geronymakis 1998). Fragkaki (1969) referred that fetsa was collected by pedlars, who in turn sold it to soap factories. In the majority of the cases, the remained OMW after storage was discharged untreated outside the OM territories (Markakis 1998). Through its route, OMW stagnated in many places within the village and ended up either onto nearby fields (Michelakis 1991) or into ephemeral rivers (Voreadou 1994).

Fig. 9 An old olive mill located in the Monastery of Odigitria, south Crete, Greece: The olive press (1), the settling vat (2), and the excavated hole in the ground, used for OMW disposal and remaining olive oil recovery (3)

Different opinions have been recorded about the effect of OMW disposal in the environment. For instance, in the village of Magarikari (pref. of Iraklio, Messara basin), OMW was disposed of into a field with fruit-bearing trees and people have witnessed a high crop production. However, in the village of Sivas, located close to the above mentioned village, people have witnessed that the continuous flow of OMW has resulted in toxic effects on similar trees. In any case, it can be concluded that controlled OMW application can be beneficial to soil fertility, while excessive application might destroy the plantations (Kapellakis et al. 2006).

4.2 State of the art on conventional treatment methods

Worldwide more than 1,000 studies have been published on OMW treatment methods (Niaounakis and Halvadakis 2006b). It is worth mentioning that the first reference to OMW management was published in 1953 by Fiestas Ros de Ursinos in the *Grasas y Aceites* Journal in Spain.

Owing to the fact that a great variety of components are found in the OMW, to date various technical methods have been used in its treatment, based on biological, physical, and physico-chemical methods. The former method involves anaerobic and aerobic processes to degrade the pollutants present in the waste stream. The physical and the physico-chemical methods may considered together and they mainly include thermal, flocculation/clarification, membrane technology, Fenton reaction, electrochemical oxidation, adsorption, and ion exchange processes. The biological and physico-chemical treatment methods are discussed in the next section.

4.2.1 Biological treatment

Aerobic processes The treatment based on aerobic process relies on those bacteria that thrive in conditions where there is adequacy of oxygen and pollutants to degrade. The organic matter can be found in OMW in the form of particulate organic carbon. This form of carbon is hydrolyzed and produces lower molecular weight organic compounds. Under aerobic conditions, these compounds are oxidized by microbes to CO_2, water, and oxidized forms of N and S.

The bioreactors, in which the aerobic process occurs, are glass columns or plastic pots filled with sieved soil and, after the aerobic reactions have taken place, most of the pollutants in the remaining fraction are used to produce new bacteria cells, which then have to be removed from the water. It is worth mentioning that bioreactors have to maintain their degrading potential in the physiological condition of immobilised cells. In fact, bioreactors with immobilised cells, such as *Aspergillus* strains (Vassilev et al. 1997) are generally the most effective in the biological treatment of organic pollutant contaminated waters (Bertin et al. 2001; Kotsou et al. 2004). In the presence of fungus, their enzymatic system is activated by the presence of organic compounds contained in OMW, the biodegradation of which results to OMW detoxification (Fountoulakis et al. 2002). Other fungal species used solely or combined to other processes to reduce the phenolic content of OMW include *Geotrichum candidum* (Assas et al. 2002; Fadil et al. 2003), *Lactobacillus plantarum* (Ayed and Hamdi 2003), *Phanerochaete chrysisporium* (Kissi et al. 2001; Dhouib et al. 2006), *Panus tigrinus* (D'Annibale et al. 2004a; 2006, and *Lentinus edodes* (D'Annibale et al. 1998).

The best known aerobic processes are the activated sludge systems, sequencing batch reactors, rotating (disk) biological contactor, and trickling filters. They can only operate efficiently if the concentration of the feed is relatively low, in the order of 1 g/l COD (Baourakis et al. 2000). However, it has been proved efficient even at higher loading rates. Eusebio et al. (2005) achieved about a 70% COD reduction for a loading rate of 40 g/l at a hydraulic retention time of 3 days. In fact, higher concentrations can only be tolerated if the plant operates at a long hydraulic retention time and/or with high recycle ratios (i.e., high flow rates of the treated effluent are recycled back to the inlet of the plant). Therefore, the high initial concentration of OMW and the slow removal kinetics of the refractory fraction of OMW pollutants combined with the fact that the refractory fraction also accounts for the colour of these effluents, makes OMW unsuitable for direct treatment by aerobic biological treatments (Niaounakis and Halvadakis 2004). Some physico-chemical pretreatments have therefore been investigated: Low-frequency ultrasound irradiation (Atanassova et al. 2005) and ozonation (Andreozzi et al. 1998; Beltran-Heredia et al. 1999, 2000) both reduce the antioxidant activity of OMW by removing up to 50% of the phenolic compounds, hence facilitating the latter biological treatments. An alternative method for improving the performance of aerobic treatment of OMW is to mix it with municipal wastewaters. In this case it is possible to use a sewage treatment plant (and associated facilities) to treat the additional wastes at a reduced cost.

Anaerobic processes In the 1970s and 80s, anaerobic digestion proved to be a promising alternative to the established aerobic treatment (Schink 2002). Solely or combined with other methods (Konstas et al. 1994; Andreozzi et al. 1998; Sabbah et al. 2004; Bertin et al. 2004) it is increasingly being used to treat wastewaters, because it allows the recovery of an appreciable fraction of the chemical energy in the waste as methane and it produces much less waste sludge than aerobic processes (Dalis et al. 1996; Erguder et al. 2000). Due to its high organic content, OMW is suitable for anaerobic treatment, even if high purification efficiencies cannot be achieved (Dalis 1989; Dalis et al. 1994; Beccari et al. 1996).

Decomposition of the organic compounds is carried out in air tight vessels by anaerobic bacteria (Rozzi and Malpei 1996) in a three-stage process

(Martin et al. 1991; Benitez et al. 1997; Zemman et al. 1997; Sabbah et al. 2004). In the first stage, the bacteria degrade complex organic materials into simpler compounds; namely, polysaccharides and polyphenols are converted to their monomers (monosaccharides and phenols, respectively). During the second stage, acetogenic bacteria convert the phenols and the monosaccharide into organic acids, such as acetic, lactic and formic acids, and alcohol. Finally, in the third stage, methanogenic bacteria, which are characterised by their sensitivity to pH, convert the organic acids into biogas (a mixture of 60–80% methane and other gases, mainly carbon dioxide).

Several anaerobic processes have been tested for OMW treatment. The processes, which differ in the way micro-organisms are retained in the fermentor, are as follows (Hamdi 1996): anaerobic contact, upflow anaerobic sludge blanket (UASB), and anaerobic filters.

Anaerobic contact: The anaerobic contact process has been tested by several researchers (Georgakakis 1989; Tsonis 1991). All these studies refer to the digestion of OMW ranging from 15 to 70 kg COD/m^3 (Rozzi and Malpei 1996). Results indicate that this process can be used as a roughing treatment for OMW with COD removal efficiencies of up to 70–80% at volumetric loading rates in the range of 2–4 kg COD/m^3 d (Beccari et al. 1996). The hydraulic retention time ranges from 20 days to 25 days (Rozzi and Malpei 1996).

Upflow anaerobic sludge blanket: Compared to the anaerobic contact process, it is quicker (Borja et al. 1996), more effective at COD removal and with higher bioenergy recovery. However, the process is unstable, especially at high COD concentrations, due to the inhibitory effect of polyphenols in the waste, lack of ammonia and the low alkalinity of OMW (Angelidaki et al. 2002). Therefore, the use of UASB process to treat OMW necessitates either adding urea as a nitrogen supplement and diluting the OMW with considerable amounts of water, resulting in large effluent amounts, or co-digesting it with other types of waste. It has been proved that co-digestion with manure could be a cost effective method as nitrogen is not added in the process (Angelidaki and Ahring 1997).

Anaerobic filters: They can operate either as upflow or downflow reactors, with the former being used mostly. The reason is that fixed bed reactors operating in an upflow direction offer higher COD loading and removal and a much faster start-up than those which operate in a downflow direction (Mechichi and Sayadi 2005). Anaerobic filters are favoured since they require very little process control, due to the shorter start-up time, and they can withstand high temporary overloads. Short start-up times are suitable for industries like OM, which operate seasonally. These advantages would suggest that this process is the most suitable for OMW treatment (Rozzi et al. 1989). However, in terms of process stability, anaerobic digestion still lags behind aerobic biological treatment and physico-chemical processes (Mechichi and Sayadi 2005).

4.2.2 Physico-chemical treatment

Thermal processes *Distillation and evaporation processes*: In these processes a concentrated solution (concentrated paste) and a volatile stream consisting of water vapour and volatile substances are produced, which result in large reductions of up to 90% in COD and 70–75% of the volume (Di Giacomo et al. 1991). Consequently, the more concentrated the OMW is, the more economical the distillation treatment per unit mass of concentrated COD is (Rozzi and Malpei 1996; Niaounakis and Halvadakis 2006b).

Combustion or incineration: This process involves the rapid chemical reaction of feed and oxygen to form CO_2, water, and heat. Since it is widely used for the disposal of waste material, it can be suitable for strong wastewaters, such as OMW, with the ability to sustain combustion without pre-drying. Due to the seasonal production of OMW, the incinerator should be either run for this period or require also other types of fuel for the rest of the year.

Flocculation-clarification This process involves the use of different inorganic coagulants, such as ferric chloride, aluminium sulphate, ferrous chloride, calcium chloride, and ferric sulphate (Massignan et al. 1988), as this process is not very efficient when used alone, because most organics found in OMW are difficult to precipitate (e.g. sugars and volatile acids). It is therefore used with anaerobic biological processes, either as a pre-treatment (Zouari 1998) or as a post-treatment stage (Fiestas Ros de Ursinos and Borja-Padilla 1991).

Membrane processes In membrane processes, wastewater is pressurised by a pump and then filtered through a special membrane. The aim of this process is to separate from a suspension/emulsion substances of a size greater than a particular value, technically named ''membrane cut-off'' (Borsani and Ferrando 1996). These substances are either organic compounds (Canepa et al. 1987) or olive oil droplets (Economou et al. 1994).

The main methods in membrane technology for OMW treatment are ultrafiltration, microfiltration, and reverse osmosis. In microfiltration, pores have a diameter of 2 μm, therefore complete colloidal constituents removal is achieved. Ultrafiltration has a cut-off of 0.1 μm and as well as removing the colloidal constituents, most of the suspended pollutants (i.e. phenolic substances) are also removed (Halet et al. 1997). However, this method is not efficient in the removal of dissolved substances, such as those determined by COD. Due to the small pore size, severe fouling of the membrane occurs, which affects performance. To achieve a better performance, the ultrafiltration process is combined with centrifugation where COD removal efficiency up to 90% has been reported (Turano et al. 2002), or with advanced oxidation process (Gernjak et al. 2004; Drouiche et al. 2004) or with reverse osmosis where it is possible to obtain a COD reduction of about 99% (Canepa et al. 1988).

Fenton reaction It is a method of chemical oxidation and coagulation of the organic compounds present in OMW, when hydrogen peroxide and ferrous sulphate (Fenton reagent) are added. Research has revealed that the Fenton reagent could moderately reduce COD and to a greater extent the phenolic compounds (Beltran-Heredia et al. 2001; Rivas et al. 2001).

Electrochemical oxidation process Although using electricity to treat water was first proposed in UK in 1889 (Chen 2004), electrochemical oxidation is a relatively new promising process in the field of OMW. In this process, the pollutants are destroyed by either direct or indirect oxidation process. In the former, when the pollutants are added to the reactor, they are first adsorbed on the anode surface of the electrode and then destroyed by the anodic electron transfer reaction. In the latter, strong oxidants such as hypochlorite/chlorine, are electrochemically generated. The pollutants are then destroyed in the bulk of the solution by the electrogeneration of active chlorine (Rajkumar and Palanivelu 2004).

Due to the high conductivity of OMW and the existence of chloride ions, electrochemical treatment of OMW has been reported to be very effective. Panizza and Gerisola (2006) have treated OMW with electrochemical oxidation and found that this process can reduce the COD from 26.5 g/l to less than 1g/l, and can completely remove the aromatic content and the colour of OMW. Gotsi et al. (2005) insisted that treatment efficiency is affected by the operating parameters, such as voltage, salinity, recirculation rate, and initial concentration of OMW.

Adsorption In this process, adsorbates, which are the dissolved compounds of OMW, are attached to the surface of the adsorbent, such as activated carbon or bentonite. The adsorbates are colouring substances hardly or non biodegradable pollutants (Curi et al. 1980). It is mainly used in combination with other treatment methods.

Ion exchange This process involves the substitution of ions in solution using chemical reagents. Research on ion exchange is in its infantry, however, it may be used in the elimination of phenols and polyphenols.

4.3 Natural treatment

Land application is the oldest practice for the disposal of OMW (Cabrera et al. 1996). It has long been practised. In fact, Marcus Porcius Cato (234–149 BC) advised applying OMW to improve soil fertility (Tomati and Galli 1992). Many studies have been published on the effects of spreading OMW on soils with various crops, the results of which indicate that this practice generally has positive effects for both the plants and the hosting soils (Di Giovacchino et al. 1997). However, negative effects do appear when some of the parameters governing land application have been underestimated or excluded from planning, such as the nature of soil, the water table, type of crop, and soil moisture content.

Land application serves three main objectives: (a) to limit wastewater management costs, (b) to reconnect the natural cycles of organic matter and nutrients, and (c) to exploit the fertilising properties of OMW. Several research projects have been carried out on the effectiveness and the potential problems of OMW land application and all concluded that land treatment efficiency is affected by plant selection, soil and land characteristics, and climate conditions. During the past 20 years, the application of OMW to agricultural land has been the subject of much research, particularly in the Mediterranean region. A summary of the results on soil, plant and effluent characteristics follows.

4.3.1 Soil properties

Several studies have been conducted on the direct application of OMW to soil as a fertiliser, either as a fresh liquid or sludge, sometimes with contradictory results (Morisot and Tournier 1986; Saviozzi et al. 1993; Riffaldi et al. 1993). Controlled land application of OMW improves the fertility status and productivity of soil by controlling the nutritional and biological equilibrium in the soil-plant system (Cabrera et al. 1996; Cox et al. 1997; Rinaldi et al. 2003). After applying OMW doses of less than 1000 m^3/ha, Garcia-Ortiz et al. (1999) reported that soils with high levels of carbonates and alkaline pH are the best for OMW land application, but acid soils and sands, which are poor in bases, change their structure and become nutritionally unbalanced when OMW is applied.

Regarding chemical and physico-chemical parameters, Marsilio et al. (1990) argued that the spreading of OMW on cultivated soil does not cause particular problems. Moreover, results obtained by Levi-Minzi et al. (1992) and Ben Rouina et al. (1999) showed that the enriching effect of OMW on organic matter, nitrogen, phosphorus, and exchangeable K, made the application of OMW an attractive option. Immediately after irrigation with OMW, the soil showed intense microbial activity, with a relative abundance of nitrogen fixers. The application of 6,100 m^3/ha OMW over a period of three years has been shown to cause quantitative increase in the lipidic fraction of the soil upper layer (Gonzalez-Vila et al. 1995). Lopez et al. (1996) found that OMW promoted only minor changes below 50 cm depth. Furthermore, the controlled application of 300 l/m^2 yr of OMW led to an increase in the organic carbon content of the soils and a reduction in soil porosity (Cox et al. 1997). Tamburino et al. (1999) referred to a test carried out by Morisot (1979), which showed that the application of 100 m^3/ha of OMW is equivalent to a medium nitrogen and phosphate fertilisation (50–60 kg of nitrogen under organic form and 70–200 kg phosphates as P_2O_5).

It is worth mentioning the characterisation of soils that were subject to uncontrolled OMW application. Paredes et al. (1987) collected samples from an alkaline soil polluted by OMW and noticed higher levels of soil salinity due to potassium and sodium replacement of soil cations. The pH was practically unchanged and soil C/N ratio was increased. Similar results were also observed by Sierra et al. (2001).

4.3.2 Plant characteristics

Positive effects are observed when OMW is applied to a range of different crops (Ben Rouina et al. 1999; Paredes et al. 1999; Rinaldi et al. 2003; Cereti et al. 2004). However, special consideration should be given to the temporary phytotoxic effect of OMW (Wang et al. 1967), which is directly correlated with the volume of OMW applied and the type of crop (D'Annibale et al. 2004b). Therefore, crops can be sown from three weeks to five months after the last application of OMW (Tamburino et al. 1999).

Briccoli-Bati and Lombardo (1990) studied the effects of various doses of OMW on the growth of olive plants in pots. Positive effects on growth were observed with the low doses but a subtoxic effect was observed after the third application with a dose of 0.4 l and a lethal effect with a 0.8 l dose. Marsilio et al. (1990) applied increased and repeated OMW quantities (from 2 l up to 16 l per tree) to a young olive grove and presented evidence that, even at high doses, the young plants' growth was not reduced and that groundwater pollution was unlikely. In contrast, Ben Rouina et al. (1999) found that, when OMW is applied at high doses (6 and 8 l/pot filled with 16 kg of soil) to growing olive trees, it was phytotoxic and plants died. However, when OMW was applied before planting, 100% of the trees survived and regarding to plant growth, the total shoot elongation was significantly dependent on the OMW dose. Komilis et al. (2005) applied pre-treated OMW to tomato and chicory seeds and concluded that water dilution is the most statistically significant pre-treatment technique affecting OMW phytotoxicity, compared to phase separation through a settling basin, aeration to promote biological degradation, and pH neutralisation. Andrich et al. (1992) irrigated four land plots in an olive grove with different amounts of OMW (0, 80, 160, 320 m^3/ha) and found that OMW did not cause toxic effects on olive trees and their products. Similarly, Bonari et al. (1993) did not observe any harmful effects when OMW doses of up to 80 m^3/ha were applied to cultivated and weed species, but Garcia-Ortiz et al. (1999) and Rinaldi et al. (2003) found a delay, due to OMW application, in the emergence of plants. They propose allowing a minimum period of 45 days before planting the seedlings in order to prevent phytotoxic effects and that not more than 100 l/m^2 OMW should be applied. Di Giovacchino et al. (1997) found that grapevines treated with 50 l/m^2 of OMW yielded on average 48% more fruit and had an average weight of 35% greater than that of the control. Kapellakis et al. (2004) applied OMW to constructed wetlands planted with *Phragmites australis* and reported relatively high dry weight biomass production, ranging from 66.67 tons/ha to 74.79 tons/ha.

4.3.3 Effluent characteristics

The effluent characteristics after treatment in natural systems has been widely investigated. In a study, 1,500 kgCOD/ha·d OMW were applied to two constructed wetland units (in the second effluent recirculation was tested). 74% removal for COD and 86% for TKN was reported for the unit without effluent recirculation, but these increased to 79% and 93%, respectively, in the presence of effluent recirculation (Kapellakis et al. 2004). Oukili et al. (2001) eliminated the phenolic compounds, which are responsible for the black-brownish colour, in clayey soils in the presence of hydrogen peroxide (H_2O_2). They defined optimal physico-chemical parameters for OMW bleaching and concluded that high bleaching could be reached after 24 h exposure to OMW at 7% weight/volume (W/V) clay material [with a clay content of 50% weight/weight (W/W)] in the presence of 0.5% volume/volume (V/V) H_2O_2, in which an 87% decrease of polyphenols and a 66% decrease of the COD occurred. Furthermore, Cabrera et al. (1996) showed that a 2 m layer of calcareous clayey soil almost completely removed the organic and inorganic components of OMW when it was applied in doses of 5,000–10,000 m^3/ha yr. In the first infiltration with the raw OMW applied, BOD_5 and COD removal efficiency within 1 m of soil was 82.6% and 84.8%, respectively. They also noticed that removal efficiency of the soil for BOD_5, COD, N, and P was maintained throughout the 3 years duration of the experiment.

4.4 Natural vs. conventional treatment

It is well known that OMW can be used as a valuable source for irrigation water, fertiliser, antioxidants, fuel, fodder, and compost. However, OMW belongs to the farmers (since is produced during processing of their olives) but OM owners are responsible for its management. Farmers could spread it on their land without causing any negative effects, as on average one tree produces 10 l OMW. However, since farmers are not aware of the beneficial agronomic value of OMW, research is focused on finding solutions for OMW treatment.

Selection of suitable OMW treatment technology will always be controversial, because numerous parameters have to be considered and scientists insist on the advantages of their own particular treatment technologies. Both natural and artificial treatment orientations have advantages and disadvantages and the aim of this section is to address the most important of these. In general, however, the difference is that processes in land treatment occur at natural rates, while in artificial treatment the rates are faster and depend on the component to be eliminated.

4.4.1 Advantages of natural treatment

Due to the variety of constituents found in OMW, more than one conventional treatment technology has to be employed and thus the investment of considerable amounts of money is required. Therefore, a cost-effective solution based on artificial treatment technologies should always consider the co-treatment of OMW produced in more than one OM (centralised or co-operative management).

The adoption of such a plan presents even more difficulties, since such solutions are energy-demanding and require high construction and O&M costs, and specialised personnel. Of all the pilot studies described in literature that have been, or are currently introducing pioneering ideas to deal with OMW, none has yet been established as economically appropriate. In addition, it should be noted that, due to the high acidity and the concentration of suspended solids in OMW, maintenance has to be carried out regularly, not only in the transfer pipe network but also in the installations. Therefore, construction and *O&M* costs for central plants that would treat exclusively OMW effluents would be very high, with uncertain possibility of payback. A solution to the problem is even more difficult to obtain in view of the fact that OM are seasonally operated enterprises, mainly small-sized and widely dispersed. Possible mixing of OMW with municipal wastewaters to achieve dilution cannot take place in existing plants, which are designed for a specific population equivalent, but has to be planned from the beginning.

Due to larger area requirements of land treatment systems, OMW can be treated under decentralised conditions, i.e. the development of natural systems to treat effluents from only a few OMs. Adoption of decentralised management is possible because natural treatment systems represent easy-to-use technologies, completely adopted to the environment, with abundant construction materials, locally available, and with little need for adjustment in operation. In addition, they are effective in reducing most of the pollutants-especially after recycling of the effluent (Kapellakis et al. 2004) and they are characterised by low construction, operation and maintenance costs, minimum energy demands, and low land requirements of not more than 0.5 ha/OM; area that is already required for the evaporation lagoons. The change in use of the evaporation lagoons should be investigated as a feasible option, as construction of such systems in the areas previously occupied by the lagoons can minimise land requirements. Furthermore, the climatic conditions in olive cultivation countries, with high average temperatures, favour such systems, as they are characterised by high reaction rates.

4.4.2 Advantages of conventional treatment

OMW storage lagoons are currently sources of pollution. Hence, the establishment of centralised treatment plants would lead to the cessation in operation of these lagoons and a considerable reduction in pollution. Moreover, treatment in centralised plants would be more effective than in on-site conventional treatment plants, due to economies of scale. Effective OMW treatment would be too expensive for OM owners to have individual onsite systems.

Although there are benefits of OMW application to soil, many problems are created and various negative aspects have to be addressed. In general, this practice is not very popular, as there is no need for irrigation during winter (the season for OMW production). In addition, possible storage of OMW to irrigate during periods with high water demand results in wastewater with high salinity, which can lead to increased concentration of salts in the ground. Another important drawback of land application is that it cannot be extended beyond two years on the same ground. This is because an oily almost impermeable film is formed, which prevents the oxygenation of the soil. Other disadvantages are the dispersion in the environment of foul smelling and possibly pathogenic substances, as well as the phytotoxic effects on plant roots due to polyphenols. Furthermore, modelling the performance of a land treatment system is not as easy as in conventional methods, since various parameters have to be taken into account (e.g. nature of soil, climatic conditions, topography). It should also be noted that there is a lack of similar pilot studies and there is poor social acceptance as the public is afraid of the effects of the use of OMW for the irrigation of crops and finally the existing legal framework does not promote land application of effluents in general.

5 Beneficial uses of compounds derived after olive mill wastewater treatment

Research on the beneficial use of the compounds derived after OMW treatment is limited. However, due to the phenolic content, OMW is an exploitable source of natural bioactive compounds, which could be used in energy recovery (Caputo et al. 2003; Eroglu et al. 2006), and construction industry (Hytiris et al. 2004), agriculture, food, cosmetic and pharmaceutical industry, as antioxidants, preservatives or even prophylactic agents against certain human diseases (Bazoti et al. 2006).

6 Conclusion

The olive tree has been cultivated for thousands years and, due to its significance as a crop, has spread worldwide and nowadays its products represent some of the most important agricultural goods. Many olive cultivation countries around the world base a considerable part of their economy on olive oil production. However, this activity leads to severe environmental degradation, as OMW, which is produced when processing olives, cannot be managed properly. The situation has worsened during the past few decades, mainly due to the industrialisation of agriculture, which has brought a noticeable increase in olive oil production worldwide, the conversion of the classic-type OM into centrifuge-type OM, and the dispersed location of a great number of small sized OMs, which has resulted in an increase of pollution due to the lack of reliable mitigation measures.

Given the lack of solutions based on the olive oil process line, efforts focused on end-of-pipe solutions; based either on the new conventional methods or on the ancient natural treatment techniques. Optimisation of conventional methods over the last years has led to a considerable reduction of various constituents. On the other hand, given the low operation, maintenance, and energy requirements, land treatment systems could be useful for achieving sustainable wastewater management.

Acknowledgements Figures are courtesy of the authors, unless indicated. The second author was sponsored by the Greek National Scholarships Institute, when this paper was under preparation.

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