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The Potential use of Phenolic Compounds Recovered from Olive Mill Wastewater in Food Model Systems

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Authors' contributions

This work was carried out in collaboration among all authors. Authors AMM and MKA contributed equally to this work. All authors read and approved the final manuscript.

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Review Article

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ABSTRACT

In terms of economic growth, health benefits, and culture, the olive oil industry is a critical sector for many countries. However, olive mill wastewater (OMWW) is one of the most polluting by-products of the manufacture of virgin olive oil. Several studies have reported that OMWW is a valuable resource of usable compounds for recovery and valorization. Because of its high content of phenolic compounds, it may serve a significant function in food because of the phenolics' strong antioxidant value. The current paper provides a survey of OMWW's phenolic recovery methods, focusing on their application as active constituents in food products. In addition, this contribution provides an overview of key research describing the potentialities of OMWW phenolics in food model systems. The Scopus, Web of Science, and Science Direct databases were chosen as our paper references. Based on the available studies, traditional techniques like solvent extraction, membranes, and, more recently, innovative technologies that promise minimum impact on these phytochemicals' compounds are used to recover phenolics from OMWW. Various food products, such as vegetable oils, bakery products, milk beverages, and meat products, can be fortified. All of these applications are based on phenolics' antibacterial and antioxidant properties to minimize food matrix alteration and contamination.

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Keywords: Olive mill byproducts; olive mill wastewater; phenolic compounds; food matrix.

ABBREVIATIONS

AGEs : Advanced Glycation Endproducts;
APCI : Atmospheric Pressure Chemical Ionization;
BCFN : Barilla Center For Food and Nutrition;
BHA : Butylated HydroxyAnisole;
BHT : Butylated HydroxyToluene;
CML : CarboxyMethyl-Lysine;
ESI : Electrospray Ionization;
FAB : Fast Atom Bombardment;
FSI : Food Sustainability System;
GAE : Gallic Acid Equivalent;
HT : HydroxyTyrosol;
LC-MS : Liquid Chromatography-Mass Spectrometry;
MALDI : Matrix Assisted Laser Adsorption Ionization;
MF : MicroFiltration;
NF : NanoFiltration;
OMW : Olive Mill Waste,
OMWW : Olive Mill WasteWater;
PG : Propyl Gallate;
SCF : SuperCritical Fluid;
SDGs : Sustainable Development Goals;
SFS : Sustainable Food System;
SPE : Solid Phase Extraction;
RO : Reverse Osmosis;
RP-SPE : Reversed Phase-Solid Phase Extraction;
TBHQ : Tert-ButylHydroQuinone;
UF : UltraFiltration;
UHT : Ultra High-Temperature;
VOO : Virgin Olive Oil;

1. INTRODUCTION

Multiple environmental negative impacts, such as biodiversity loss, soil deterioration, and water pollution, are all exacerbated by food production. In fact, the food system is responsible for 20–30% of global greenhouse gas emissions. Humanity has a monumental task in reorganizing food systems to provide healthy diets to all people in a sustainable manner. The 2030 Agenda for Sustainable Development encapsulates 17 Sustainable Development Goals to be achieved by 2030, including responsible production and consumption. The Sustainable Development Goals (SDGs) of the United Nations place a focus on a sustainable food system. The SDGs, which were adopted in 2015, request radical changes in agriculture and food systems by 2030 in order to eliminate hunger, ensure food security, and support nutritional intake [1, 2, 3].

According to the United Nations, a sustainable food system (SFS) is one that ensures food security and nutrition for all while preserving the economic, social, and environmental foundations required for future generations to have food security and nutrition. This means that it achieves all sustainability aspects, including economic, social, and environmental sustainability. Population density, industrialization, higher revenue, changing consumption habits, as well as global warming and natural resource degradation, must all be considered [1, 2, 3].

Based on the United Nations and the Food and Agriculture Organization (FAO) reports, food system evolution has produced numerous favorable outcomes. As food industries have expanded, these outcomes have included the increase of career opportunities and the development of food choices beyond local food items, satisfying consumers' preferences in terms of organoleptic properties, and quantity. However, the quick industrialization of food supply chains has generated severe problems, with significant impacts in terms of food security and nutrition. A wide range of processed food items are commonly accessible to consumers, inducing high levels of food loss and waste and an environmental impact [1, 2, 3].

The consumption of olive oil, which is widely known for its biological activities, is increasing throughout the world due to its health benefits and great nutritional properties [4]. Unfortunately, the challenge of treating and disposing of olive mill wastewater (OMWW) is wreaking havoc on the producing countries due to its high organic content. The agri-food industry has recently been under greater pressure to address social and environmental challenges in their supply chains throughout product lifecycles. Indeed, olive oil production necessitates the use of a considerable number of resources as well as the release of pollutants that have a substantial negative impact on the environment [5].

In order to examine the simultaneous influence of food systems on human health and the environment, the Barilla Center for Food and Nutrition (BCFN) has created the Food Sustainability Index (FSI). It assesses the long-term sustainability of 78 countries' food systems based on three core pillars: food loss and waste, agriculture, and nutritional problems. The index ranges from 0 to 100, with 100 representing the highest level of sustainability [6] (Fig. 1).

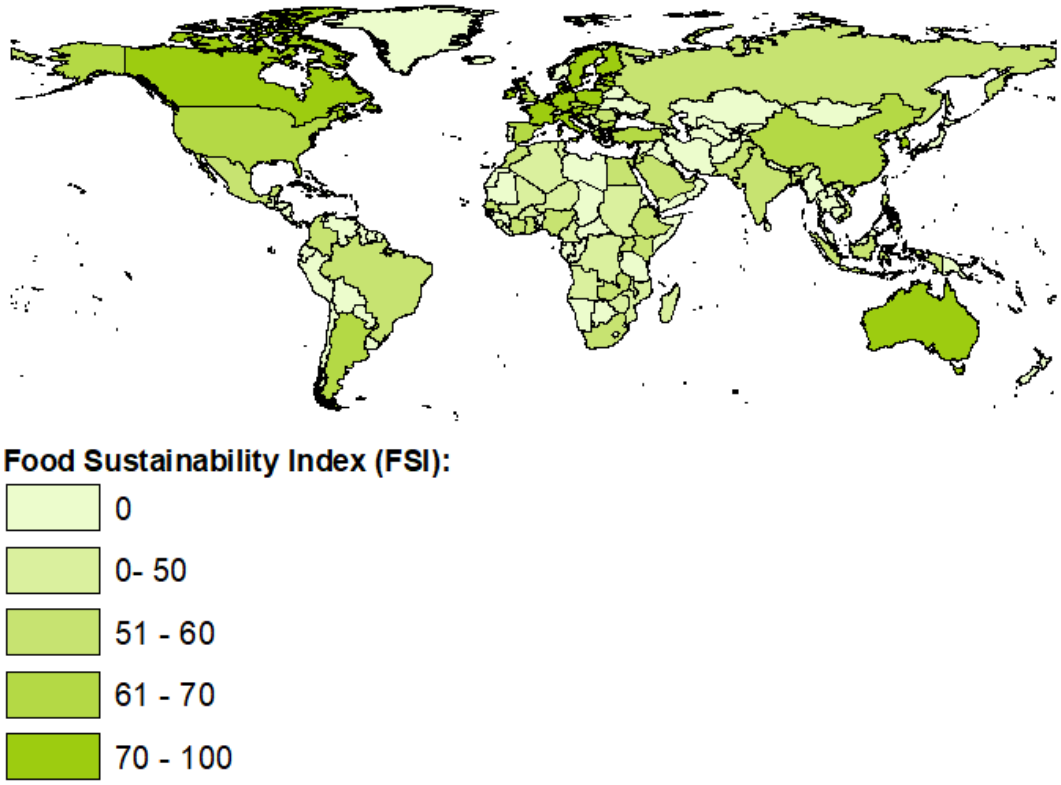

Fig. 1. Food Sustainability Index (FSI) (prepared using data of the Barilla Center for Food and Nutrition BCFN)

Olive mill wastewater (OMWW) has long been thought to be the most polluting and bothersome waste generated by olive mills. One of the most damaging effluents produced by the agro-food industry is said to be this complex medium. OMWW production is estimated to be over 30 million m^3 worldwide, with 98% of it generated in the Mediterranean basin. Thus, the management of this liquid residue has been investigated, and some extensive and detailed studies reported that olive production systems conversion (i.e., two-phase instead of three-phase), detoxification methods, and recovery of compounds from OMWW can be considered to treat olive oil processing effluents [7]. In fact, this waste could be easily turned into a valuable source of antioxidant chemicals due to the high concentration of phenolic compounds, which can be added to a variety of foods to develop a functional product with better nutritional properties. In fact, phenolic compounds have been shown to inhibit metal-induced oxidation, scavenge free radicals, serve as reductants, and even preserve food quality. In addition, the antimicrobial activities of phenolics have been demonstrated, suggesting their usage as natural additives to extend the shelf life of foods. Recovering phenolic compounds not only gives financial and nutritional benefits, but also reduces the OMWW's environmental impact [8].

Olive oil extraction entails a number of steps, including olive washing, crushing, and malaxing, as well as the extraction itself, which is the most basic stage of the entire process. The extraction process will determine the amount and physical-chemical properties of the olive oil as well as those of the waste produced. The two processes for extracting the oil are traditional pressing, which has been employed for millennia, and centrifugation, which the olive oil industry has taken over more recently [9]. This technology is referred to as three-phase because the centrifugal decanter separates three phases: olive oil, pomace, and wastewater. This process, however, necessitates the addition of warm water to dilute the olive paste, thereby generating a significant amount of OMWW [9]. Fig. 2 depicts the extraction method applied to virgin olive oil production with regard to the byproducts obtained from the three-phase system process.

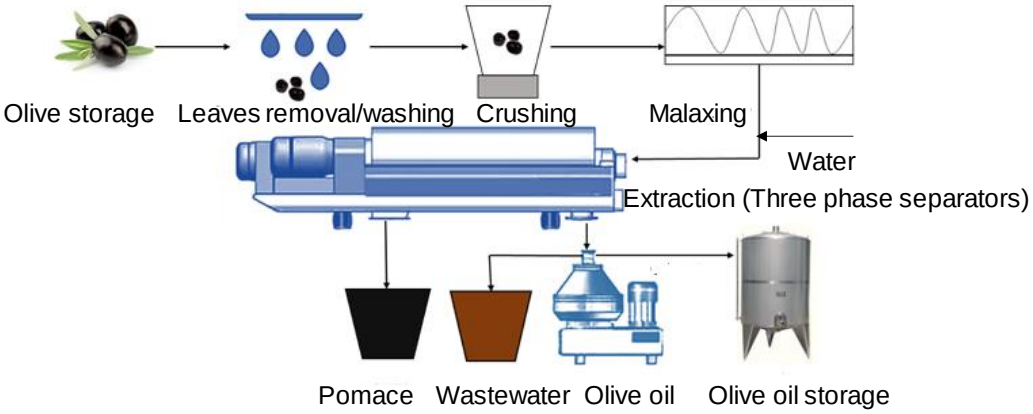

Fig. 2. Three-phase system process

Table 1. Physical and chemical properties of olive mill wastewater

	Unit	Range of values	References
pH	-	4.8−5.7	[12]
Conductivity	mS/cm	5−81	[12]
COD	g/L	16.5−156	[13]
BOD	g/L	13.4−37.5	[14]
Dry residue	g/L	11.5−90	[15]
Lipids	g/L	7	[15]
Sugar	g/L	1.3−4.3	[16]
Total nitrogen	g/L	0.06−0.9	[17]

COD: Chemical Oxygen Demand ; BOD: Biochemical Oxygen Demand

The annual production of OMWW in the world is estimated to be between 10 and 30 million m^3. The discontinuous technique (which is no longer widely employed) produces less but more concentrated waste (0.5-1 m^3 per 1000 kg) than centrifugation (1-1.5 m^3 per 1000 kg) [10]. Because OMWW contains various high-added-value compounds, the generation amount should be exploited. OMWW is mainly made up of 83–94% water, 4–16% organic compounds, and 0.4-2.5% mineral salts. In addition, OMWW contains sugars, tannins, phenolic compounds, polyalcohols, minerals, pectins, and lipids. It has a high potassium concentration and considerable amounts of nitrogen, phosphorus, calcium, magnesium, and iron when compared to other organic wastes [11]. Despite the high concentration of phenolic compounds in the olive fruit, only 2% of the initial concentration is found in the VOO, with the remainder (about 53%) present in the OMW and the pomace (approximately 45%). This is owing to the hydrophilic character of phenolics and the contact between the water phase and the oil phase during the extraction [11]. It has a black color (due to lignin polymerization with phenolic chemicals), is acidic (pH of around 5), and has high electrical conductivity (Table 1).

2. METHODOLOGY

This study was conducted in order to find and document scientific papers that are relevant to the topic. The papers included in this review are strictly limited to scientific research published in peer-reviewed journals. The study was conducted in three phases: preparation, execution, and analysis. The review strategy was devised in the planning phase. The authors came to a conclusion about the criteria used to record the papers and the key questions addressed in the review. Following this stage of conceptualization, the authors proceeded on to the actual work. In this step, data mining was achieved through a review of scientific databases, the recording of all OMWW-related papers, and the final selection of those whose contexts fit the current review's purpose. The Scopus, Web of Science, and Science Direct databases were chosen as our paper references. Papers were found in these databases by searching for titles, abstracts, and keywords.

"OMWW (olive oil mill wastewater)," "OMWW's phenolics," "recovery of phenolics from OMWW," "Qualitative and quantitative determination of OMWW phenolic compounds," and "application of OMWW's phenolics in food model systems" were the main keywords employed in this procedure. The process was completed with a third phase, which was the analysis of the results. Furthermore, in order to provide a reliable and qualitative database of papers, the scientific work reported above is confined to articles in peer-reviewed scientific journals published in English. In this regard, the manuscript has made no attempt to shed light on research that has been done and published in conference proceedings, PhD or master's theses, diploma dissertations, working papers, or textbooks. As a result, limiting the study to scientific journals ensures that the analysis is thorough and unbiased. Thus, the authors examined the recorded papers and classified them into groups based on their content, as defined during the planning phase. The publications have been categorized according to the year of publication, the tested concentration of recovered phenolics, the food matrix, and the impact they investigate.

3. RESULTS AND DISCUSSION

3.1 Recovery of OMWW Phenolic Compounds

The presence of significant levels of organic chemicals in OMWW could explain their phytotoxic and bactericidal properties in part. As a result, significant research efforts have been focused on developing purification and treatment procedures that can recover the high-value phenolic chemicals from OMWW while reducing the volume and severe environmental effects of these hazardous effluents. Solvent extraction, chromatographic separations, centrifugation, membrane processes [18], and, more recently, high voltage electrical discharges, pulsed electric fields, and ultrasounds [19, 20] have been proposed to recover phenolics from OMWW [21]. However, the two most frequently utilized methods for recovering phenolic chemicals are solvent extraction and membrane separation. Despite the high cost of using significant amounts of organic solvents, solvent extraction is the most extensively utilized method for recovering phenolic compounds from OMWW [22, 23]. Several authors, including Kalogerakis et al. [24], employed ethyl acetate. Allouche et al. [25] described ethyl acetate as the most effective solvent for recovering phenols from OMWW produced using a three-phase mill.

Emmons and Guttersen's [26] method for obtaining oleuropein aglycon from OMWW entails adding citric acid to the raw material, boiling it to precipitate the solids, then extracting the oleuropein aglycon from the water immiscible component with a non-polar organic solvent mixture, preferably a 50/50 v/v hexane/acetone mixture. After that, the solvent is evaporated using a vacuum and/or heat.

De Marco et al. [28] used the SPE extraction method to recover biophenols from OMWW, stating that reversed phase-solid phase extraction (RP-SPE) yielded about 1 g of pure hydroxytyrosol/1 L of OMWW.

Galanakis et al. [29] described a method for isolating dietary fibers from OMWW, including pectins and important phenolic compounds, as well as the utilization of isolated products as food additives and antioxidants, respectively. To begin with, OMWW is centrifuged to remove the fat and then concentrated by removing the water content. Following that, it is extracted with ethanol and an organic acid. One of the following organic acids can be used in the process: citric, tartaric, malonic, maleic, malic, oxalic, adipic, or fumaric. The remaining polyphenols in the dietary fibers are then extracted and filtered with ethanol at a concentration of at least 85 percent (v/v). The polyphenol-containing liquid phase is clarified by filtration after dilution with 15-40% (v/v) ethanol.

The use of supercritical fluids (SCFs), notably supercritical CO_2, has been shown in recent studies to overcome the limitations of organic solvents such as toxicity and flammability. SCF extraction, on the other hand, comes with the drawback of requiring the use of expensive high-pressure equipment. Thus, phenolic chemicals were extracted from OMWW using a solvent and a CO_2 supercritical fluid, as described by Lafka et al. [30]. The phenolic content was measured as caffeic acid equivalents on a dry basis (% w/w) and ranged from 0.43% to 1.29% using solvent extraction against 0.76% using supercritical CO_2.

Membrane technologies are appealing for recovering phenolic compounds from olive mill wastewater because of their numerous advantages, including minimal energy consumption, no chemical requirements, and no phase shift. Despite the fact that conventional

filtration membranes are still commonly used in the treatment of OMWW, microfiltration, ultrafiltration, nanofiltration, and reverse osmosis membranes, mostly in sequential form, effectively cover the needs for the recovery, purification, and concentration of antioxidants in terms of their specific molecular weight cut-off values [20].

The large molecular weight range of OMWW chemicals makes high-purity recovery difficult. However, this can be solved via membrane technologies. Microfiltration membranes can hold microparticles as small as 0.1-10 m, whereas UF membranes can hold macromolecules as small as 1-100 nm. NF membranes can separate molecules with a diameter of 0.5-5 nm, while RO membranes retain molecules with a diameter of less than 1 nm [31].

As a result, many membrane-based approaches for recovering natural antioxidants from OMWW have been investigated. Villanova et al. [32] proposed a method for recovering tyrosol and hydroxytyrosol from OMWW. Rough filtration (RF), microfiltration, ultrafiltration, nanofiltration, and reverse osmosis units, followed by column chromatography, are recommended for recovering tyrosol and hydroxytyrosol from polyphenolic fractions or their purified components of OMWW.

Membrane filtration was used by D'Antuono et al. [33] to recover OMWW phenolics from two Italian and three Greek olive varieties. MF, UF, and NF each produced three fractions. Each fraction had a distinct level of purity as well as a varying amount of phenolic compounds.

The fractions generated by a membrane method, specifically the reverse osmosis concentrate, after utilizing NF were characterized by Zagklis et al. [34]. Adsorption/desorption resins were used to further treat these phenolic compounds. Finally, vacuum evaporation was used to concentrate the recovered phenolic compounds, yielding a final extract with a phenolic concentration of 378 g GAE/L.

Bazzarelli et al. [35] recovered purified polyphenol-concentrated retentate, using a combination of microfiltration and nanofiltration membranes, as well as osmotic membrane distillation and membrane emulsification processes.

To obtain a dried phenolic-rich fraction of OMWW, Sedej et al. [36] used a two-step sequential filtration approach (ultrafiltration/reverse osmosis) and various drying procedures (spray drying, freeze drying, or infrared drying). In 3-phase OMWW, spray drying led to the highest total phenolic content, antioxidant activity, and phenolic compound content.

De Almeida et al. [37] used a combination of ultrafiltration and nanofiltration to develop a polyphenol-concentrated retentate that contained oleuropein, gallic acid, syringic acid, and HT.

Ultrafiltration and nanofiltration membranes offer excellent bioactive chemical recovery techniques and are the most practical for phenolic compounds recovery from OMWW [38].

Previous scientific research found that combining various techniques such as microfiltration, nanofiltration, and reverse osmosis produced an OMWW polyphenol-concentrated retentate with a high HT content (1.52 g/L), tyrosol content (0.52 g/L), and oleuropein content (0.51 g/L), as well as antioxidant hypolipidemic and hypoglycemic characteristics *in vitro* [31]. Similarly, to extract polyphenols and carbohydrates from OMWW, a pilot membrane design combining ultrafiltration, nanofiltration, and reverse osmosis membranes was developed [39].

Other methods, like enzymatic processing, have been employed to recover phenolic compounds from OMWW. Khoufi et al. [40] reported that the OMWW submitted to the hydrolyse action of an enzyme from *Aspergilus niger* cultivated on wheat bran could be a valid source of phenolic compounds, mainly hydroxytyrosol, with interesting applications. Another study discovered that pre-treating OMWW with *Aspergillus niger* and *Trichoderma atroviride* culture broths rich in α-glucosidase increased the amount of hydroxytyrosol produced. Similar investigations with *Trametes trogii* culture media, however, revealed a substantial oxidation of phenolic chemicals due to this strain's strong laccase activity [41]. In the same context, Hamza and Sayadi [42] investigated the efficacy of enzymatic pre-treatment (with *Aspergillus niger* α-glucosidase) and membrane technology (using MF and UF). Overall, this method was successful in recovering hydroxytyrosol.

Table 2. The phenolic compounds most commonly found in olive mill wastewaters

Phenolic compound	Molecular formula	Method of extraction	Method of identification	References
3,4-dihydroxyphenylglycol (DHPG)	$C_8H_{10}O_4$	Solvent extraction	MALDI-TOF-MS	[46]
Apigenin	$C_{15}H_{10}O_5$	Ultrasound-Assisted Solid Liquid Extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Apigenin-7-*O*-glucoside	$C_{21}H_{20}O_{10}$	Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Apigenin-7-*O*-rutinoside	$C_{27}H_{30}O_{14}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
β-hydroxy-isoverbascoside	$C_{29}H_{36}O_{16}$	Membrane technology coupled to low-pressure gel filtration chromatography on a Sephadex LH-20	HPLC-DAD-MS/MS	[49]
		Membrane extraction	LC-DAD-ESI-MS^n	[33]
Caffeic acid	$C_9H_8O_4$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		Membrane extraction	LC-DAD-ESI–MS^n	[33]
Caffeoyl-6’ -secologanoside (Cafselogoside)	$C_{25}H_{28}O_{14}$	Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
		Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
Chlorogenic acid (Caffeoyl-quinic acid)	$C_{16}H_{18}O_9$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
Chrysoeriol	$C_{16}H_{12}O_6$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
Comselogoside	$C_{25}H_{27}O_{13}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS UPLC-DAD-ESI-QTOF-HRMS LC-DAD-ESI-MS^n	[48] [47]
		Ultrasound-assisted solid liquid extraction		[33]

Phenolic compound	Molecular formula	Method of extraction	Method of identification	References
		(USLE) method Membrane extraction		
Demethyloleuropein	$C_{24}H_{30}O_{13}$	Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Dihydro-oleuropein	$C_{25}H_{36}O_{13}$	Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Elenolic acid (EA)	$C_{11}H_{14}O_6$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47] [50]
		Solvent extraction	HPLC-ESI-Q-TOF-MS	
Elenolic acid (EA) derivative (decarboxylated form of hydroxyelenolic acid)	$C_{10}H_{14}O_5$	Solvent extraction	MALDI-TOF MS	[46]
		Solvent extraction	HPLC-ESI-Q-TOF-MS	[50]
Elenolic acid glucoside (Oleoside methyl ester)	$C_{17}H_{24}O_{11}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Gallic acid	$C_7H_6O_5$	Solvent extraction	HPLC-ESI-Q-TOF-MS	[50]
Hydroxytyrosol (3,4-DHPEA)	$C_8H_{10}O_3$	Adsorption resin technology	LC-ESI-MS/MS	[51]
		Solvent extraction		[48]
		Ultrasound-assisted solid liquid extraction (USLE) method	RPLC-DAD RPLC-ESI-MS UPLC-DAD-ESI-QTOF-HRMS	[47]
		Membrane extraction		[33]
		Solvent extraction		[50]

Phenolic compound	Molecular formula	Method of extraction	Method of identification	References
		Solvent extraction	LC-DAD-ESI-MS^n MALDI-TOF-MS HPLC-ESI-QTOF-MS	
Hydroxytyrosol glucoside	$C_{14}H_{20}O_8$	Solvent extraction	RPLC–DAD RPLC–ESI-MS	[48]
		Membrane extraction	LC-DAD-ESI–MS^n	[33]
		ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Hydroxytyrosil acyclodihydroelenolate	$C_{19}H_{26}O$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		Membrane extraction	LC-DAD-ESI-MS^n	[33]
Ligstroside	$C_{25}H_{32}O_{12}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Luteolin	$C_{15}H_{10}O_6$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Luteolin-4'-*O*-glucoside	$C_{21}H_{20}O_{11}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Luteolin-7-*O*-glucoside	$C_{21}H_{20}O_{11}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]

Phenolic compound	Molecular formula	Method of extraction	Method of identification	References
Luteolin-7-*O*-rutinoside	$C_{27}H_{30}O_{15}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
Luteolin-*O*-rutinoside	$C_{27}H_{30}O_{15}$	Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Oleacin (3,4-DHPEA-EDA)	$C_{17}H_{20}O_6$	Solvent extraction	MALDI-TOF MS	[46]
		Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
			UPLC-DAD-ESI-QTOF-HRMS	[47]
		Ultrasound-assisted solid liquid extraction (USLE) method		
Olenoside A and B	$C_{11}H_{14}O_5$	Solvent extraction	HR-ESI-MALDI-TOF-MS	[52]
Oleocanthal (*p*-HPEA-EDA)	$C_{17}H_{20}O_5$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
			UPLC-DAD-ESI-QTOF-HRMS	[47]
		Ultrasound-assisted solid liquid extraction (USLE) method		
Oleoside	$C_{16}H_{22}O_{11}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
			UPLC-DAD-ESI-QTOF-HRMS	[47]
		Ultrasound-assisted solid liquid extraction (USLE) method	LC-DAD-ESI-MS^n	[33]
		Membrane extraction		
Oleuropein	$C_{25}H_{32}O_{13}$	Solvent extraction	RPLC-DAD RPLC–ESI-MS	[48]
			UPLC-DAD-ESI-QTOF-HRMS	[47]
		Ultrasound-assisted solid liquid extraction (USLE) method	LC-DAD-ESI-MS^n	[33]
		Membrane extraction		
Oleuropein aglycone	$C_{19}H_{22}O_8$	Solvent extraction	MALDI-TOF-MS	[46]

Phenolic compound	Molecular formula	Method of extraction	Method of identification	References
(3,4-DHPEA-EA)				
			RPLC-DAD	[48]
		Solvent extraction	RPLC-ESI-MS	
			UPLC-DAD-ESI-QTOF-HRMS	[47]
		Ultrasound-assisted solid liquid extraction (USLE) method		
Oleuropein aglycone derivative	$C_{16}H_{26}O_{10}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		Membrane extraction	LC-DAD-ESI-MS^n	[33]
Oleuropein glucoside isomers	$C_{31}H_{42}O_{18}$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]
		Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
Oleuroside	$C_{25}H_{32}O_{13}$	Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
p-coumaric acid	$C_9H_8O_3$	Ultrasound-assisted solid liquid extraction (USLE) method	UPLC-DAD-ESI-QTOF-HRMS	[47]
		Membrane extraction	LCDAD-ESI-MS^n	[33]
p-Hydroxybenzoic acid (4-hydroxybenzoic acid)	$C_7H_6O_3$	Solvent extraction	HPLC-ESI-Q-TOF-MS	[50]
Protocatechuic acid	$C_7H_6O_4$	Solvent extraction	HPLC-ESI-Q-TOF-MS	[50]
Quercetin	$C_{15}H_{10}O_7$	Solvent extraction	RPLC–DAD RPLC–ESI-MS	[48]
Quercetin-3-*O*-rhamnoside	$C_{21}H_{20}O_{11}$	Solvent extraction	RPLC–DAD RPLC–ESI-MS	[48]
Quinic acid	$C_7H_{12}O_6$	Membrane extraction	LC-DAD-ESI–MS^n	[33]
Rutin	$C_{27}H_{30}O_1$	Solvent extraction	RPLC-DAD RPLC-ESI-MS	[48]

Phenolic compound	Molecular formula	Method of extraction	Method of identification	References
			UPLC-DAD-ESI-QTOF-HRMS	[47]
		Ultrasound-assisted solid liquid extraction (USLE) method	LC-DAD-ESI-MS^n	[33]
		Solvent extraction		
Tyrosol	$C_8H_{10}O_2$	Adsorption resin technology	LC-ESI-MS^2	[51]
				[48]
			RPLC-DAD	
		Solvent extraction	RPLC-ESI-MS	[33]
			LC-DAD-ESI-MS^n	[50]
		Solvent extraction		
			HPLC-ESI-Q-TOF-MS	
		Solvent extraction		
Tyrosol glucoside	$C_{14}H_{20}O_7$	Solvent extraction	RPLC-DAD	[48]
			RPLC-ESI-MS	
			UPLC-DAD-ESI-QTOF-HRMS	[47]
		Ultrasound-assisted solid liquid extraction (USLE) method		
Verbascoside	$C_{29}H_{36}O_{15}$	Solvent extraction	LC-DAD-ESI-MS^n	[33]
			UPLC-DAD-ESI-QTOF-HRMS	[47]
		Ultrasound-assisted solid liquid extraction (USLE) method		

MALDI-TOF-MS: matrix-assisted laser desorption/ionization (MALDI) time-of-flight mass spectrometry, UPLC-DAD-ESI-QTOF-HRMS: ultra-high pressure liquid chromatography system with diode array and electrospray ionization quadrupole time-of-flight high resolution mass spectrometry, RPLC–DAD: reversed phase HPLC–photodiode array detection, RPLC–ESI-MS: RPLC–electrospray ionisation mass spectrometry, LC-DAD-ESI–MS^n: liquid chromatography/diode array detection/electrospray ion trap tandem mass spectrometry, LC-ESI-MS/MS : liquid chromatography–electrospray ionization tandem mass spectrometry

Table 3. Range of contents of phenolics quantified in OMWW

Phenolic compound	Contents range	Method of quantification	Reference
Apigenin	2.5–6.5 [a]	RP-HPLC-UV-MS	[53]
Caffeic acid	0.014–0.017[c]	RP-HPLC-UV-MS	[53]
Elenolic acid (EA)	4.9–11.7[a]	RP-HPLC-UV-MS	[53]
Gallic acid	3.86–6.71[a]	HPLC-ESI-QTOF-MS	[50]
	22.2–61.0[a]	RP-HPLC-UV-MS	[53]
Hydroxytyrosol (3,4-DHPEA)	483.0–1733.2[a]	HPLC-ESI-QTOF-MS	[50]
	157.2–245.1[a]	RP-HPLC-UV-MS	[53]
	544–1560[a]	HPLC-DAD	[54]
	1230–1290[a]	RPLC-DAD	[48]
	0.25–18.2[b]	RPLC-ESI-MS LC-ESI-MS/MS	[51]
Hydroxytyrosol glucoside	1300 -1700[a]	RPLC-DAD RPLC-ESI-MS	[48]
Ligstroside	0.0087–0.0092[c]	-	[4]
Luteolin	2.5–36.2[a]	RP-HPLC-UV-MS RPLC-DAD	[53]
	270–510[a]	RPLC-ESI-MS	[48]
Luteolin-7-*O*-glucoside	0–0.0214[c]	-	[4]
Luteolin-hexoside	3.2–24.2[a]	RP-HPLC-UV-MS	[53]
Oleacin (3,4-DHPEA-EDA)	11300–45951[a]	HPLC-DAD	[54]
Oleuropein derivative	5400–7600[a]	RP-HPLC-UV-MS RP-HPLC-UV-MS RPLC-DAD	[53] [48]
Oleuroside	200–400[a]	RP-HPLC-UV-MS RPLC-DAD	[48]
p-coumaric acid	15.9–21.8[a]	RP-HPLC-UV-MS	[53]
p-hydroxybenzoic acid (4-hydroxybenzoic acid	1.75–6.15[a]	HPLC-ESI-QTOF-MS	[50]
Protocatechuic acid	2.77–5.29[a]	HPLC-ESI-QTOF-MS	[50]
	25.3–136.7[a]	RP-HPLC-UV-MS	[53]
Rutin	440–640[a]	RPLC-DAD RPLC-ESI-MS	[48]

Phenolic compound	Contents range	Method of quantification	Reference
Tyrosol	218.4–581.0[a]	HPLC-ESI-QTOF-MS	[50]
	1180–1560[a]	RP-HPLC-UV-MS RPLC-DAD	[48]
	0.19–4.32[b]	RPLC-ESI-MS	[51]
Vanillic acid	1.68–62.7[a]	HPLC-ESI-QTOF-MS	[50]
	0.0174–0.0198[b]	-	[4]
Verbascoside	14496–24100[a]	HPLC-DAD	[54]
	1620–1760[a]	RP-HPLC-UV-MS	[48]

Contents are given as [a]: mg/Kg , [b]: mg/mL , and [c]: weight %

3.2 Qualitative and Quantitative Determination of OMWW Phenolic Compounds

The qualitative and quantitative determination of OMWW phenolic compounds is difficult due to the matrix's complexity and the heterogeneity of the fraction of interest. However, technological advancements in analytical instruments over the previous few decades are assisting in overcoming the aforementioned barrier. In this regard, the employment of high-resolution separation techniques that aid in the subsequent detection of individual components substantially assists in the improvement of the proposed methods' selectivity and sensitivity. On the other hand, the lack of commercially available pure standards for a large number of OMWW phenolics, as well as difficulties in completely resolving complex chromatographic profiles, has made the use of MS detection almost mandatory due to its ability to confirm identity and quantify overlapped peaks. Many scientific reports have demonstrated the importance of the LC-MS for the identification of phenolic compounds in olive processing byproducts [43, 44]. In mass spectrometry, there are a variety of ionization processes that are entirely compatible with liquid chromatography (LC), such as atmospheric pressure chemical Ionization (APCI), electrospray ionization (ESI), Fast Atom Bombardment (FAB) and matrix assisted laser desorption ionization (MALDI).

OMWW is a rich source of polyphenols, with secoiridoid derivatives such as hydroxytyrosol, the dialdehydic form of decarboxymethyl oleuropein aglycone, tyrosol, and verbascoside being particularly important [45]. During the extraction process, olive phenolic compounds are divided between the water and oil phases. However, because they are water-soluble substrates with strong polarity, the bulk percentage is missing in the oil phase. The extraction and identification of phenolics from OMWW seems promising because they're powerful natural antioxidants that have sparked a lot of interest in the cosmetic, food, and pharmaceutical industries. OMWW has been shown to contain almost forty distinct phenolic chemicals [4].

The ability to characterize OMWWs in terms of phenolic content is critical for developing effective re-evaluation approaches. As a result of the complexity of OMWWs, analytical procedures capable of providing a complete qualitative and quantitative screening of their composition have been developed. Several analytical approaches have been published in the literature for the identification and quantification of specific phenolic compounds in OMWW. Molecular formulas and the method of extraction and identification of the phenolic compounds in OMWW are listed in Table 2.

The cultivar, pedoclimatic conditions, maturity of the fruit, processing conditions, and degree of hydrolysis of OMWW (related to its storage conditions) are all factors that influence the concentrations of the phenolic compounds in the OMWW [55]. Due to these various factors, including the phenolic recovery approach and the analytical technique used, both qualitative and quantitative profiles are dependent. 4-hydroxybenzoic acid, protocatechuic acid, and vanillic acid were all discovered in OMWW [56, 57, 58]. Furthermore, the presence of polymeric phenols has been attributed to the effluent's characteristic brownish-black color [59]. The inherent unpredictability of the wide variety of analytical parameters and methodologies used to extract and evaluate the phenolic compounds, as described above, could possibly explain the diversity of phenolics in OMWW. Nonetheless, the phenolic concentration of OMWW from different origins varies greatly, complicating the identification and quantification procedures.

In addition to its phenolic content, OMWW includes numerous valuable nutrients, including sugar, proteins, and phosphate. OMWW has also been shown to contain soluble dietary fibres, particularly pectin substances with excellent gelling properties [29].

This article presents a literature review of major research outlining OMWW's phenolic recovery techniques and the possible applications of OMWW’s phenolics for the development of functional food products. The purpose of this research is to present a critical overview of pertinent scientific studies centered on OMWW's phenolics and their possible application in food model systems. It summarizes current knowledge in the valorization of a byproducts of a vital agro-food product, such as virgin olive oil.

3.3 Using Recovered Phenolics in Food Model Systems

Recently, OMWW has been proposed for the formulation of functional foods with a wide range of effects. Indeed, phenolics identified in olive

mill by-products have been extensively reported in the literature for their bioactive potential. Their valorization not only aids in environmental conservation but also provides natural bio-ingredients for re-use in food and non-food applications. Natural phenolic compounds, particularly those recovered from food industry by-products, are promising antioxidants for use in various foods as substitutes for synthetic antioxidants. From both an environmental and food technology standpoint, the use of OMWW as a valuable source of phenolic compounds capable of adding functional value to food items is of great interest.

In fact, Visioli et al. [60] investigated phenolic extracts obtained from OMWW that demonstrated high antioxidant potential.

According to Kachouri and Hamdi [61], the incorporation of OMWW (fermented by *Lactobacillus plantarum*) into olive oil facilitated the decrease of phenolics in wastewater residuals and the rise of phenolics in olive oil. This is due to *Lactobacillus plantarum*'s ability to depolymerize high-molecular-weight phenolics in OMWW, which allows them to potentially transfer from wastewater to oil. These researchers discovered that combining OMWW with fermented *L. plantarum* with plain or non-fermented OMWW resulted in a considerable increase in polyphenol concentration in the oil, with 703 and 112 mg/L of oil, respectively. Individual phenolic compounds, particularly oleuropein content, showed a similar pattern in this investigation, with 401.8 and 140.4 mg/L in oil samples with and without fermented *L. plantarum*, respectively.

The antioxidant potential of OMWW extract was compared to that of BHA and BHT. Peroxide values were lower when OMWW extract was added at 500 ppm compared to BHA [62].

In comparison to vitamin E and C, hydroxytyrosol, recovered from OMWW, has shown higher antiradical effects and has thus been utilized to prevent lipid oxidation in fish products [63].

Servili et al. [54] increased the concentration of phenolic compounds in VOO and EVOO by adding phenolics recovered from OMWW. The crude phenolic extract was obtained by membrane filtration, and it was then extracted with ethyl acetate and ethanol. The crude phenolic extract was also added during the extraction process, specifically before the malaxation stage. In comparison to negative control, adding 5 or 10% of this phenolic concentrate resulted in a considerably increased content of total phenolic compounds, particularly 3,4-DHPEA and 3,4-DHPEA-EDA.

The effect of OMWW phenolic compounds added to milk beverages on beneficial bacteria in yogurt and similar products was investigated. The addition of phenolic extract had minimal effect on the concentration of *Lactobacillus* and *Streptococcus* bacteria during fermentation [65].

The use of OMWW for the creation of a functional beverage was proposed by Zbakh et al. [66]. Commercial products contain a variety of chemicals, including antioxidants like ascorbic acid, chelators like ethylenediaminetetraacetic acid (EDTA), and acidifiers like citric acid. When OMWW extract was used in beverages, no additional antioxidants were required.

Troise et al. [67] estimated the antioxidant potential of OMWW phenolic extract in UHT milk samples and investigated its potential for the inhibition of the Maillard reaction by adding phenolic extract at 0.1 and 0.05% w/v. The authors reported an inhibition of reactive carbonyl species formation in samples prior to heat treatment. They also revealed greater stability of tested samples without any sensorial negative attributes.

To extend the shelf-life at 4°C, a phenolic concentrate from OMWW was employed to treat the surface of fresh chicken breasts [68]. *Enterobacteriaceae* and *Pseudomonas* spp. showed a delay in growth in the dipping samples. In addition, the bactericidal activity of OMWW phenolic extract was assessed on a variety of spoilers, starters, and food-borne bacteria (*Staphylococcus* spp., *Listeria* spp., *Escherichia* spp., *Salmonella* spp., *Pseudomonas* spp., *Lactobacillus* spp., and *Pediococcus* spp.) in order to propose them as natural additives for extending food shelf life. Resistance to phenolics was lowest in *Staphylococcus aureus* and *Listeria monocytogenes*. Fasolato et al. [68] found that gram-negative bacteria (*S. Typhimurium* and *Pseudomonas* spp.) were unaffected by the tested doses, but starter cultures (*Staphylococcus xylosus*) proliferated at a much slower rate. Fasolato et al. [68] discovered that the phenolic extract (whose tested concentration was equivalent to 38.6 g/L) was efficient in extending the shelf life of fresh

chicken breast. When compared to the control, the results revealed an inhibition of growth of both *Enterobacteriaceae* and *Pseudomonas* spp., as well as a 2 day improvement in shelf life.

Using a surface treatment, Chavez-Lopez et al. [69] did a similar study in which OMWW phenolics were applied to fermented sausages to suppress mold populations. The researchers discovered that soaking the product in a 2.5% phenolic solution inhibited some fungi [69].

Previous research has demonstrated that a polyphenol-rich extract from OMWW can preserve the α-tocopherol content while frying refined olive oil and prevent the generation of undesirable volatile compounds [70]. Esposto et al. [70] investigated the impact of purified phenolic concentrate recovered from OMWW (100, 200, 400, and 1200 mg/kg) on improving oil stability during a frying process, compared to those of a refined olive oil containing BHT and an EVOO with a high phenolic content. When added at a concentration of at least 400 mg polyphenols/Kg, this OMWW extract, which mostly comprises tyrosol, hydroxytyrosol, 3,4-DHPEA-EDA, and verbascoside, was found to have a stronger ability to retain α-tocopherol content than BHT, decreasing oxidation.

The effectiveness of commercially available powder containing phenolics recovered from OMWW in comparison to other antioxidants in inhibiting microbial growth of bread and rusks during storage has been studied [71]. According to the findings, the commercially available powder was able to extend the shelf life of bread and rusk samples due to its antibacterial activity.

Phenolic compounds from OMWW act as antioxidants, radical scavengers, and texture improvers in food emulsions as well as antimicrobial molecules in meat products [72].

Chavez-López et al. [73] used OMWW-extracted phenolics to improve quality parameters and increase the shelf life of fermented sausages. In both *in vitro* and *in vivo* experiments, the extracts reduced fungal growth and spore germination in fermented sausages in a dose-dependent manner. Tested fungi were all significantly inhibited in situ after being treated with 2.5 % OMWW phenolic extract.

De Leonardis et al. [74] recommended combining lard with phenolics as a "novel food," demonstrating that OMWW's natural antioxidants were particularly effective in preventing lard from oxidizing. The phenolic extract considerably enhanced the oxidative stability of fat and, when evaluated on mouse cell lines, the applied levels (100-200 ppm) were not cytotoxic (embryonic fibroblasts).

Other researchers incorporated pure OMWW phenolic compounds into white meat hamburgers to elucidate how they impacted product shelf life after 11 days at 4 °C. The inhibition of total mesophilic count was effective at all concentrations tested, notably at the highest concentration [75].

The effect of OMWW-extracted polyphenols in white meat burgers packaged in PVC on increasing sensory and sanitary qualities was studied by Veneziani et al. [75]. The incorporation of the phenolic extract at different concentrations (0.75 and 1.50 g/kg) inhibited the growth of mesophilic aerobic bacteria in comparison to the negative control, inducing a 24 hour extension of shelf life.

Galanakis et al. [76] investigated the antioxidant effect of OMWW phenolic extract in combination with other antioxidants, finding that it reduced oxidative damage during bread and rusk baking and had an antimicrobial effect against *S. aureus*, *B. subtilis*, *E. coli*, and *P. aeruginosa* (at 200 mg/kg of flour).

Cedola et al. [77] enriched bakery products by adding OMWW that had previously been submitted to ultrafiltration and evaluated the chemical composition and sensory quality attributes of the resulting products. Both the bread dough and the spaghetti formulation at a final concentration of 30% w/w were made with ultrafiltered OMWW. The findings revealed that adding OMWW to bread and pasta improved their chemical composition considerably without compromising their sensory characteristics.
Roila et al. [78] used phenolic extract (250 g/mL and 500 g/mL) to prevent the growth of *Pseudomonas fluorescens* and *Enterobacteriaceae* in mozzarella cheese.

According to Troise et al. [79], spray-dried OMWW decreases the Maillard reaction in a cookie model system, demonstrating that OMWW is a multi-functional additive capable of interfering at multiple stages of the Maillard reaction. Multiple mechanisms promoted the control of lysine and asparagine changes in the cookie model system because of the chemical

Table 4. Using recovered phenolics in food model systems

Food matrix	Additive	Tested Concentration	Result	Mechanisms	References
Fermented Sauges	Polyphenols	The samples are dipped in 2.5–5% W/V.	Antifungal activity	At the concentrations employed, OMWW has antifungal action that is species dependent.	[69]
White meat burgers	Purified phenolic extract	0.75-1.50 g/kg	Antibacterial activity	Retarding the growth of aerobic mesophilic bacteria	[75]
Fresh breast of chicken	Crude phenolic concentrate	38.6 g/L	Antioxidant and antibacterial activities	TBAR levels that are much lower Growth of *Enterobacteriaceae* and *Pseudomonas* spp. is delayed by 2 days.	[75]
Lard	Crude phenolic extract	100-200 ppm	Antioxidant activity	Prevention against oxidation of lard Extension of shelf life	[74]
Milk	Olive oil mill wastewater polyphenol powders	0.1 and 0.05 % W/V	Functional milk	Increasing product stability	[67]
Milk beverage	crude phenolic concentrate	100-200 mg/mL	Fortified beverage	Formulation of fortified beverage	[54]
Butter	crude phenolic extract	2, 4, 6, and 8 mg/100g	Antioxidant activity	Confering resistance against oxidation	[80]
Cheese	crude phenolic concentrate	250 and 500 µg/mL	Antimicrobial activity	Increasing shelf life	[78]
Cooked and cold stored meat ball	Crude phenolic extract	50-100 mg/L	Antioxidant and activity	Inhibition of lipid peroxidation	[29]
Bread and pasta	Purified phenolic extract	900 ml of OMWW for bread and 30% w/w for pasta	Antioxidant activity and food fortification	Enhancing chemical composition without compromising the sensory properties	[77]
Cooked beef and pork	Crude phenolic extract	50-100 mg/Kg of meat	Antioxidant activity	Improving lipid stability during cooking	

Food matrix	Additive	Tested Concentration	Result	Mechanisms	References
Raw and cooked fresh pork sausages	Crude phenolic extract	750, 1500 mg/kg	Antioxidant activity	Inhibition of lipid oxidation and inhibition of oxidative degradation of cholesterol	
Fermented salami	Purified phenolic extract	0.15%	Antibacterial activity	*Listeria monocytogenes* growth inhibition after 45 days of fermentation	[75]

nature of secoiridoids. Indeed, OMWW inhibited the production of protein-bound Amadori compounds, CML dicarbonyls, acrylamide, and 5-hydroxymethylfurfural in cookies, demonstrating the efficacy of secoiridoids-based functional ingredients to prevent the formation of AGEs.

In a butter formulation, extracts of both OMWW were added at various concentrations, revealing that the concentration equivalent to 8mg/100 mg of butter provides protection against oxidation during storage at 25°C for 3 months, inhibiting the growth of *S. aureus*, total coliforms, yeast, and molds [80].
The incorporation of OMWW phenolic extract into foodstuffs will contribute to resolving the issue of OMWW's high pollution charge and maximize the extract's utility. Using recovered phenolics in food model systems should be carefully evaluated in order to achieve the intended effect while avoiding unfavorable effects in terms of product stability and sensory impact. Despite the fact that the bitter and pungent sensory note of virgin olive oil is desired—even within a particular range set by consumer acceptance—there is little study on the negative effects of adding an excessive amount of OMWW food products.

Because of their complex matrix and the composition of their bioactive chemicals, the design of functional foodstuffs necessitates research into the stability and interactions of phenolic compounds with other dietary ingredients. More research is also needed on the sensory impact of OMWW. Table 4 lists some examples of the potential use of phenolics recovered from OMWW in food model systems.

4. CONCLUSIONS AND FUTURE DIRECTIVES

Environmental concerns related to olive processing wastes have been extensively reported. Given the seriousness of the environmental impact of olive processing wastes, many options for valuing OMWW have been proposed. However, many elements should be considered when choosing the appropriate valuing method, including the overall amount of effluent, investment costs, the industrial or agricultural environment, and most importantly, the legislation. OMWW phenols are far too precious to be depleted or released into the environment. As a result, recovering phenols and repurposing them in various products and markets should be prioritized and more investigated.

Detoxification, production system change, and recovery of important components have been the most popular treatments to date. Traditional techniques such as solvent extractions, membranes, and, more recently, innovative technologies are used to recover phenols from olive mill waste and other food processing by-products. The potential of using components from olive mill waste dates back several years. All of these efforts have resulted in the industrial recovery of phenols from OMWW, as well as their use as natural preservatives and bioactive substances. Various foods, such as vegetable oils, table olives, lard, bakery products, milk, drinks, and meat products, have been investigated. The antioxidant potential of phenolics, as well as their antimicrobial activity, have been demonstrated in all of these applications.

In the food industry, phenolic compounds have the great promise to be used as food preservatives. In fact, phenolic compounds have been thoroughly researched for use in food products to extend the shelf life of foodstuffs. Natural food ingredients have been increasingly popular in recent years. The use of phenolic compounds instead of synthetic antioxidants, such as butylated hydroxyanisole (BHA, E-320), butylated hydroxytoluene (BHT, E-321), tert-butylhydroquinone (TBHQ, E-319), and propyl gallate (PG, E-310), are an interesting alternative to replacing chemical additives in the food matrix. Due to their possible harmful impact on human health, the chemicals are subjected to a maximum concentration limit in foods. The value of natural food preservatives with antioxidant and antibacterial characteristics for food production and consumer health has been largely investigated. Because of the detrimental effects of synthetic chemicals, natural antioxidants and antimicrobials have gained acceptance as replacements. Natural antioxidants and antimicrobials, on the other hand, necessitate more research so that the optimal doses can be properly applied to foods without affecting sensory properties.

While considerable progress has been made in terms of OMWW valorization approaches, there is still much more study to be done. In fact, the stability of phenolic compounds in OMWW-based foodstuffs during processing and storage should be thoroughly investigated. More efforts should be made in an attempt to develop various alternatives to avoid possible inactivation by direct addition. Spraying, coating, and dipping

treatments prior to packaging can be considered as viable solutions. A different approach would be to utilize one or more chemicals that could have synergistic effects at lower doses without affecting the food's sensory qualities.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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