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REVIEW

Active edible coatings and films with Mediterranean herbs to improve food shelf-life

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ABSTRACT
The main reasons for food decay are oxidative reactions and bacterial growth. The food industry has tried to solve these problems either by incorporating additives into foods or by implementing different packaging techniques. However, there is a tendency to incorporate additives into packaging materials, especially those of natural origin. Due to the multiple properties and their extensive accessibility, Mediterranean herbs have been used since ancient times for culinary and pharmacological purposes. This review includes up-to-date information concerning the materials employed and the techniques to produce Mediterranean herbal coatings and films, as well as the sensory changes during the storage within different food categories. Mediterranean herbs have polyphenols, terpenoids, and flavonoids with antimicrobial and antioxidant activity that can used to produce different active systems: sachets, multilayers, labels and coatings. Moreover, the use of essential oils or volatile essential oils from Mediterranean herbs in coatings and films can improve the properties of packaging system and extend the shelf-life of muscle products, fruit and vegetables. The impact of essential oils components in the sensory attributes of coated foods is the current main challenge for the use of these bioactive components in active packaging.

KEYWORDS
Essential oil; packing; antioxidant; antimicrobial; oxidation

Introduction

The lifestyles and daily needs have changed dramatically in recent years. For example, working parents and single-parent family have short time to prepare food. Thus, the demands for ready-to-eat meals and food with a stable and long shelf-life are continuously increasing. With the development of active and intelligent packages, the shelf-life of the food can be extended with better organoleptic quality (Realini and Marcos, 2014). In technical terms, the European Commission defined active packaging as the packaging designed to intentionally incorporate components that release or absorb substances into or onto the packaged food or its environment to prolong the shelf life or maintain or improve the quality of the packaged food (Regulation (CE), 2009).

The most common factors of food deterioration are lipid oxidation and microbial contamination (Ahmed et al., 2017). Vacuum or modified atmosphere packaging has been used to reduce oxidative processes. However, modified atmosphere packaging is limited by the type of food. Some studies have been reported the incorporation of antioxidants into foods to delay or prevent the product deterioration (Ganiari et al., 2017). Since the food industry works to limit the use of chemical additives, the most widely studied alternative is the application of natural antioxidants obtained from plant extract, herbs, spices and their essential oils (EOs) (Atarés and Chiralt, 2016).

Traditionally, Mediterranean herbs (such as thyme, sage, rosemary, oregano, and marjoram) have been used in culinary to impart unique flavors and aromas in food preparations as part of the heritage of the countries of the Mediterranean basin (basil in Genovese pesto, for instance) and in the everyday meals of the global population (Bianchi, 2015). Moreover, Mediterranean herbs have also been studied as source of bioactive compounds (especially EOs) in the protection against the development of inflammatory diseases and cancer and also has potential to improve the healthy of elderly population (Agatonovic-Kustrin et al., 2019; Elshafie and Camele, 2017).

However, some of these compounds give strong flavor, which limits their use. The possibility of using these natural antioxidants as part of the packaging has begun to be considered (Mohamed et al., 2020). Active packaging can be done in three different manners: releasing antioxidants into the food, removing oxygen or radical compounds and chelating catalysts as metal ions. In the first case, active materials are released into the food at controlled rates to

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compensate their consumption during storage. In the second and third case, scavengers or chelating agents are used (Mastromatteo et al., 2010).

Although petrochemical-based materials are cheap and easily accessible, they are not biodegradable and give rise to serious ecological problems. Hence, natural polymers, either used alone or in combination with synthetic polymers as edible films or coatings, have been created and provide ecological advantages over traditional packaging materials (Donhowe and Fennema, 1994). Due to the benefits of incorporating natural plant-based antioxidants to food packaging and an increasing amount of studies that has focused on their application, the objectives of this review are to give an overview of active edible coatings and films with Mediterranean herbs to improve food shelf-life and discuss future perspectives of food packaging.

The basis of the production of edible films and coatings

Materials used as a matrix for edible film and coating

The ideal packaging material should play as barrier properties against moisture change, gas immigration, heat exchange, microorganism growth, and well mechanical properties to form coatings and to incorporate additives. Also, it must be tasteless, odorless, colorless, flexible and easy to be heated sealing. However, there is no such natural form material available. Every year an excessive accumulation of plastic residues is generated in the world, so the packing industry is increasingly turning to natural alternatives as biopolymers that represent a more sustainable option. In this regard, natural materials used for film and coating production consist mainly of proteins, lipids and polysaccharides. Hydrocolloids have been the most researched compounds due to their excellent mechanical and structural properties, even though they have a low moisture transfer barrier capacity, whereas lipids have hydrophobic properties but low mechanical resistance that can be compensated by mixing them with hydrophilic compounds (Morillon et al., 2002). Table 1 summarizes some of the natural polymers, sorted by chemical family, that have been used in active food packaging, giving an overview of the main characteristics.

Cellulose and its derivatives, such as cellulose acetate (CAc), have been widely used to produce plastic films (Rodríguez et al., 2012). Another example is chitosan and its by-products that are biodegradable, antimicrobial and antifungal polysaccharide and nontoxic (Aider, 2010; Romanazzi et al., 2002). The deacetylated chitin (presents in the cell wall of fungi as well as the exoskeleton of crustaceans) have been used as a base for the EOs-based films and coatings (Sánchez-González et al., 2010). It also reported that the antimicrobial and antioxidant capacity of chitosan can be improved by incorporating EOs to chitosan as well as reduced the water vapor permeability in the film and the lipid oxidation of the covered food products (Kanatt et al., 2008).

Antioxidant compounds

The antioxidant constituents of the herbs could be collected and concentrated as EOs or extractions with high phenol and terpenes contents in the oleoresins (Figure 1). The extraction methods determine the antioxidant efficiency of extracting components. The available extraction methods include steam distillation, organic solvent extraction, supercritical CO_2 extraction, mechanical press, etc (Pateiro et al., 2018). EOs have been primarily associated with natural antioxidant and antimicrobial protection (e.g.: carvacrol and thymol) (Serrano et al., 2008). Phenolic compounds usually are extracted with pure organic solvents. The extraction yield depends on the selected solvents (e.g.: water, acetone, ethyl acetate, methanol, ethanol, propanol, etc), temperature, and duration of extractions (Oniszczuk et al., 2014). In addition to herbs and spices, natural extracts derived from by-products, such as olive leaves, have also been shown to have antioxidant activities (Marcos et al., 2014).

The compatibility of the antioxidant compounds with the packaging material and releasing time of the antioxidant compounds should be considered as the two factors that determine their efficiency as an active packaging (Gómez-Estaca et al., 2014). For instance, incorporating EO can reduce the water vapor permeability of hydrophilic film (Atarés and Chiralt, 2016).

Mechanism of incorporation

An edible coating is a thin layer of edible material direct or indirect covering a food (Figure 2). The direct coatings are applied by immersion of the food in a liquid solution that provides the structural matrix, while the indirect coatings are applied to the food as a previously molded solid layer to wrap of foods (Falguera et al., 2011).

There are several manners to incorporate antioxidants into packaging materials, including sachets, labels, multilayers and coating (Bolumar et al., 2011). During the manufacturing process, all the compounds are mixed, either by dissolving them in a solvent, immobilizing in the film surface, or extruding the film solution (Granda-Restrepo et al., 2009; Lacoste et al., 2005). Casting is an energy-intensive method wherein a large proportion of water is evaporated from the films (Figure 2). The evaporation step is an important procedure in casting. It is usually done with heated air and takes 2–3 days depending on the air temperature. The drying processing in manufacturing edible films could also be achieved by infrared, conduction, vacuum or low pressure overheated steam drying, and microwave (de Moraes and Laurindo, 2018; Kaya and Kaya, 2000; Mayachiew and Devahastin, 2008; Ortiz et al., 2017). Apart from that, a study indicated that a good way of improving conventional emulsification is by producing nano-emulsification, reducing also the losses of EOs due to volatilization. This approach also improves the antimicrobial capacity of the film since EO can be released more slowly (Acevedo-Fani et al., 2015). Due to above limitations (long time processing, high requirements for equipment, etc.) the production of edible films is limited to the bench scale.

Table 1. Main characteristics of natural materials used as chemical alternatives for film and coating production sorted by their biochemical families (Mohamed et al., 2020).

Biochemical family	Material	Characteristics
Animal Polysaccharide	Chitin and chitosan	Decreases the partial pressure of oxygen in the package. Regulates the humidity between food and environment by maintaining the temperature. Delays enzymatic browning in fruits, promotes natural flavor, controls respiration, decreases dehydration and thus improves the emulsifying effect. Good resistance to oxygen, fat and oil. Sensitive to humidity.
Plant polysaccharides	Cellulose	Odorless and tasteless. Biodegradable. Poor water vapor barrier. In particular, carboxymethyl cellulose has excellent film-forming and thermal gelation properties.
	Starch	Translucent or transparent. Colorless, tasteless and flavorless.
	Pectin	Used as a stabilizer, thickener and gelling agent. Excellent mechanical properties. Excellent barrier to oil, oxygen, and aroma. Low resistance to humidity. Fragile.
	Arabic gum	Decreases the breathing rate in fruits and the production of ethylene.
Marine polysaccharides	Alginate	Increases the water barrier, maintaining the taste. Delays the lipid oxidation. Low water vapor barriers.
	Carrageenan	Protects against surface dehydration.
Microbial polysaccharides	Pullulan	Odorless, tasteless and colorless. Heat-sealable. Not very permeable to oxygen and oil, but permeable to water.
	Gellan	Hard and brittle gel. Efficient on freshly cut vegetables.
	Xanthan gum	Effective on freshly cut fruit. Delays the ripening process.
Lipid	Oil and fats	Water resistant. Water vapor barrier.
	Essential oils	Antimicrobial activity. Antioxidant properties.
	Waxes	Reduces moisture permeability.
	Resins	Imparts hydrophobia. Good water vapor and gas barriers. Prevents senescence.
	Plasticizers	Increases strength and flexibility. Increases gas and water permeability.
	Emulsifiers	Decreases surface tension Prevents phase separation
Protein	Milk proteins	Used as a moisture barrier. Excellent gas barrier properties. Poor water vapor resistance of protein films Low mechanical strength. Abundant and cheap.
	Gelatine	Used as a moisture barrier. Impressive gas barrier properties. Poor resistance to water vapor. High film thickness. Improved mechanical properties.
	Soya protein	Used as a moisture barrier. Good gas barrier properties. Poor resistance to water vapor. Low mechanical strength. Low applications.

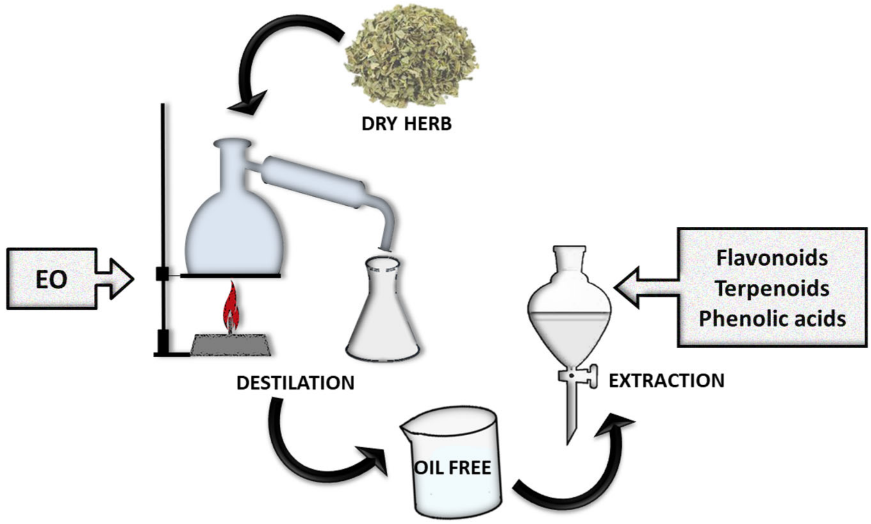

Figure 1. Overview of obtaining antioxidant compounds from herbs.

Purposes of the use of additives in food packaging

Antimicrobial

Additives with antimicrobial properties are added to food to prevent the growth of microorganisms (Appendini and Hotchkiss, 2002). However, the effectiveness of these agents could be diminished by specific compounds that may be present in the food. Novel active packaging can solve this issue by gradual releasing of selected compounds from the packaging materials into the food (Ouattar et al., 2000). Based on this theory, herbs with antimicrobial effect can be added to the films to slow food spoilage (Shankar et al., 2014). Oregano (*Origanum vulgare*) has been demonstrated its effectiveness against bacteria, yeast and fungi such as *Alternaria alternata* and *Rhizopus stolonife* (Souza et al., 2007). Pola et al. (2016) reported that the addition of oregano EO to films inhibited phytopathogens, and it could exhibit a greater inhibition at a higher concentration regardless of the type of microorganism. Although films containing 20% of the same EO was effective against *Fusarium semitectum* growth, they were not able to inhibit the proliferation of *A. alternata*, *R. stolonifer* and *Lasiodiplodia theobromaea* (Espitia et al., 2012). The effectiveness of EO from sage as an antimicrobial agent has been demonstrated. In particular, the species *S. officinalis* was shown to be active against *Staphylococcus aureus, Escherichia coli, Bacillus cereus, Salmonella enteritidis, Bacillus subtilis, Aspergillus niger* and *Candida albican* (Abdelkader et al., 2014; Miladinović and Miladinović, 2000). Gutierrez et al. (2008) demonstrated that combining EO of oregano with *Sage triloba* increased the *E. coli* lag phase, and even the combination of sage with thyme was an efficient alternative against *E. coli* and *Listeria monocytogenes*.

Viuda-Martos et al. (2011) analyzed the antibacterial activity of EO of *Thymus vulgaris* at a concentration of 40 μL and reported inhibitory zones of 41.00, 23.50 and 20.25 mm respectively *for L. innocua, Pseudomonas fluorescens, S. marcescens* bacteria, respectively. Lower inhibition zones (10.1, 9.7, 19.4 and 13.9 and mm) were found for *B. cereus, Salmonella spp., S. aureus* and *L. innocua* due to the incorporation of *Thymus capitatus* in a lower concentration (4 mL), which indicated a concentration-dependent inhibitory effect (Bounatirou et al., 2007; Ruiz-Navajas et al., 2013).

Antimicrobial mechanisms of EOs include the disruption of enzyme systems, altering bacterial genetic sequences, destroying the phospholipid bilayer and oxidizing unsaturated fatty acids by the newly-formed hydroxyperoxidase (Arqués et al., 2008). Lucini et al. (2006) indicated that the monoterpenes in EOs could increase peroxide levels, such as alkoperoxyl, hydroxyl and alkoxyl, which can cause cellular death. Meanwhile, Sharma & Tripathi (Sharma and Tripathi, 2008) reported that EOs could cause the death of the mycelium due to the release of hyphae intracellular components and the loss of rigidity and integrity of the cell wall.

Antioxidant

The presence of oxygen inside the package, even in marginal quantities, is the main issue of food spoilage. In addition to leading to the oxidation of lipids and promoting microbial growth, oxygen also causes discoloration, textural changes, the development of abnormal odors and flavors, nutritional losses and the possible formation of toxic compounds. Antioxidants mechanisms were proposed as eliminating free radicals, either by inhibiting the initiation and propagation of oxidation reactions, or by their chelating action with catalysts (Mitsumoto et al., 2005). Among the natural antioxidants, the phenolic compounds are remarkable candidates, which can be extracted from herbs and spices and used as flavorings. The family of herbs belonging to the *Laminaceae* have been greatly studied, especially thyme, marjoram, sage, oregano, rosemary and savory (Camo et al., 2008, 2011; Ganiari et al., 2017; Nerín et al., 2006; Vital et al., 2016). The antioxidant capacity can be evaluated from the phenolic content. Phenolic compounds and the terpenoids of EOs have redox properties by their ability to donate hydrogen, which can eliminate free radicals and quench single oxygen and chelate transition metals (Liyana-Pathirana and Shahidi,

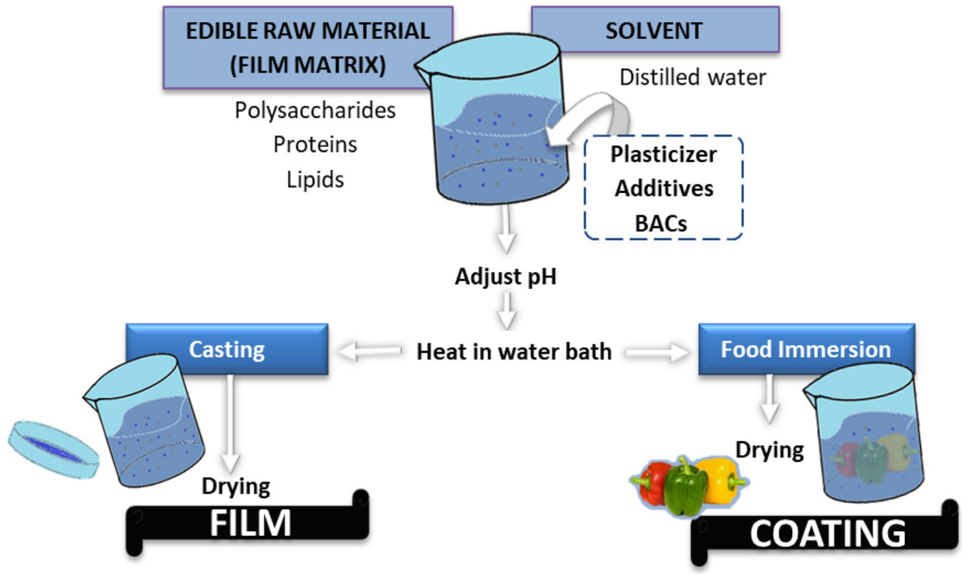

Figure 2. General process for obtaining active edible films and coating.

2006). The antioxidant activity of EOs obtained from Thymus has been studied and proven (Sarikurkcu et al., 2010). However, in a low concentration (0.5 and 1%), it did not present significant effects in cellulose films (Ruiz-Navajas et al., 2013).

Other purposes

Antioxidants could be used in packaging as oxygen scavengers. As a result of their action, lipid oxidation and the resulting organoleptic and nutrition loss can be avoided. The proliferation of aerobic microorganisms is also prevented. An economic alternative option is to use vacuum and modified atmosphere packaging, which could decrease the amount of preservatives and can be labeled as natural (Ahmed et al., 2017). As a complement to these oxygen scavengers, CO_2 emitters can be used as the relative level of O_2 is reduced by increasing amount of CO_2. As a consequence, aerobic bacteria and fungi activities are inhibited (Suppakul et al., 2003). During the food storage, fungal and bacterial growth is greatly increased in foods, as the water activity is elevated (e.g.: meat, fish, and poultry). In these cases, the use of absorbent pads, sheets and blankets as well as the incorporation of desiccants can assist in the control of exudates and purge inside the packaging.

Food applications

Due to their numerous properties, especially as antioxidants and antimicrobial activity, the extracts of herbs and spices are a promising alternative for active food packaging, which has been reflected in a great number of recent researches (Ganiari et al., 2017). Mediterranean herbs have been employed in food system since antient times for both medicinal and food purposes. Among the Mediterranean aromatic plants, rosemary, garlic, lavender, leek, olive leaf, onion, oregano, pepper, mint, sage and satureja display potential to be explored as source of active compounds (Alirezalu et al., 2020). However, not all of them have received the same attention, the research about their effectiveness is still scarce (Table 2).

Meat and meat products

It has been proved that clove, thyme, oregano, rosemary and garlic are powerful antimicrobial compounds to extend shelf-life of muscle products (Emiroğ lu et al., 2010; Matiacevich et al., 2015; Soni et al., 2016; Zinoviadou et al., 2009). Regarding antibacterial properties, the EO of rosemary has been used in the vacuum packaging of beef to reduce the populations of microorganisms, such as *Psychrotrophs*, *Brochothrix thermosphacta*, *Pseudomonas* spp. and *Enterobacteriaceae* (Sirocchi et al., 2017). Bolumar et al. (2011) investigated the incorporation of rosemary extract into packaging under high pressure treatment, and reported a lower oxidation level of chicken meat during storage.

In a further study, the effect of rosemary extract was compared with the effect of a commercial oxygen scavenger at different pressure treatments (600, 700 and 800 MPa) (Bolumar et al., 2016). The results obtained with the rosemary-based packaging showed fluctuations in lipid oxidation up to day 17, but thereafter the levels decreased to the same range as with the oxygen scavenger packaged samples on day 25. Therefore, rosemary-based packaging can be considered an excellent system. In another experiment, polyphenols extracted from rosemary were incorporated into tapioca starch films in order to verify its antioxidant property, which were not only demonstrated but also observed to be better barriers against ultraviolet light (Piñeros-Hernandez et al., 2017).

Studies have shown that oregano oil incorporated in polypropylene packaging has a greater anti-oxidation effect on lamb fillets samples than rosemary (Camo et al., 2008), which also displayed a positive relationship between the amount of added extract and the antioxidant capacity (Camo et al., 2011). In contrast, Oussalah et al. (2004)

Table 2. Main results of selected studies carried out until now on Mediterranean herbal-based films and coatings on meat and meat products.

Food application	Mediterranean herb	Matrix	Results	Reference
Beef	Cumin	Shahri Balangu mucilage	Lower values of Total viable counts, *Psychotropics*, *Staphylococci*, *coliforms* and fungi. Lower water losses and minor lipid oxidation Improved hardness	(Behbahani et al., 2020)
Beef	Olive by-products	Pectin-Gelatine	59-68 % improved lipid stabilization 100 % reduction of lipid oxidation for 7 days using hydroxytyrosol and beeswax in the film	(Cañas et al., 2017)
Beef muscle	Oregano	Milk proteins	Significant decrease in the level of *E. coli* O157:H7 and *Pseudomonas* spp. Significant increase in TBARS content on day 4, but stable until day 7 without significant difference with the control.	(Oussalah et al., 2004)
Beef muscle	Oregano	Milk proteins	No difference in water content and water vapor permeability. Increased elongation properties at concentrations of 1.0%. Significant reduction in Viable Total Count values and *Pseudomonas* spp.	(Zinoviadou et al., 2009)
Beef	Oregano	Polypropylene	Significant increase in the shelf life of beef from 14 to 23 days by adding at least 1% extract. Concentrations greater than 4% unacceptable development of oregano odor	(Camo et al., 2011)
Beef patties	Oregano, Thyme	Soya protein	Increased inhibition of *S. aureus* and strong antimicrobial activity against *E. coli* and *E.coli* O157:H7 with concentrations up to 3% EO. No inhibitory effect against *S. aureus*, *E. coli* or *E.coli* O157:H7. Significant reduction of *Pseudomonas* pp. and coliforms during cold storage.	(Emiroğlu et al., 2010)
Beef meat	Rosemary	Polyethylene	Lower counts of the *Psychophilic* bacteria, *Brochothrix thermosphacta*, *Enterobacteriaceae* and *Pseudomonas* spp. Extended shelf life up to day 15 depending on storage conditions. Positive influence on color parameters, in particular redness	(Sirocchi et al., 2017)
Lamb steak	Oregano Rosemary	Polypropylene	Increased oxidative stability in combination with modified atmosphere packaging, especially with oregano. Extension of the smell and fresh color for 5 days.	(Camo et al., 2008)
Lamb	Garlic	Alginate	Reduced lightness, redness, lipid oxidation, and water loss but increased yellowness no effect in pH	(Guerrero et al., 2020)
Chicken	Clove	Polyethylene	Improved UV barrier of the film and influenced yellow color of film Inhibition of *Salmonella* Typhimurium and *Listeria monocytogenes*	(Mulla et al., 2017)
Chicken patties	Oregano Thyme	Carrageenan	Higher antimicrobial effect in the mixture of both EO (0.02% (w/v) and 0.03% (w/v) for oregano and thyme respectively).	(Soni et al., 2016)
Chicken breast fillets	Thyme	Alginate	Extended shelf-life for 1.75 days at 4 °C of storage	(Matiacevich et al., 2015)
Chicken breast and thigh	Rosemary	Polyethylene	Lower degree of lipid oxidation on day 25 of storage when combined with high pressure treatment. More effective than oxygen scavengers.	(Bolumar et al., 2011, 2016)

observed that the antioxidant capacity of oregano oil was not effective in films based on milk protein although the antimicrobial capacity was remarkable. One drawback of active packaging is the potential of cross odor contamination, which could lowered the desired sensory quality. In this sense, Camo et al. (2011) reported that the level of oregano extract should be below 2% (g/g).

There are many types of lesser investigated herbs that could be ideal candidates as active packaging additives. For instance, the addition of clove oil in film caused a strong inhibition in growth of *Salmonella* Typhimurium and *Listeria monocytogenes* inoculated in minced chicken (Mulla et al., 2017). In the case of garlic essential oil, its use in alginate film delayed lipid oxidation but affected the color of thawed lamb meat during refrigerated storage (Guerrero et al., 2020). Cumin has been investigated for its antimicrobial effects and it can reduce lipid oxidation (Behbahani et al., 2020). Olive extracts can be applied in food processing as an antioxidant additives (DeJong and Lanari, 2009). Hydroxytyrosol and 3,4-dihydroxyphenylglycol are two phenols present in olives and their antioxidant effectiveness as an edible film in beef has been recently examined (Bermúdez-Oria et al., 2019). The films based on these two compounds displayed a good oxygen barrier and a good

Table 3. Main results of selected studies carried out until now on Mediterranean herbal-based films and coatings on seafood.

Food application	Mediterranean herb	Matrix	Results	Reference
Carp	Oregano	Gelatine	Antimicrobial activity with up to 1% OE. Good inhibition activity for *Escherichia coli*. Thickness increments, solubility declination and tensile strength reductions (33.2% less) of film after the use of 4% EO.	(Wu et al., 2014)
Smoked sardine	Rosemary, oregano	Gelatine	Significant higher values of phenols in oregano samples, so presents higher antioxidant stability Lower values of TBARS and peroxides in both enriched batches after 21 days of storage Lower microbial counts in oregano high pressure treated samples until the end of storage but not so much without high pressure treatment.	(Gómez-Estaca et al., 2007)
Smoked Rainbow Trout	Rosemary	Polyethylene	Depending on the sensory quality delay of the deterioration from day 28 (control) to the day 56 or 63, depending on the concentration of the extract. Significantly lower values of totox (indicator of total oxidative stability) The higher the extract concentration in the container, the lower the TBA values, so the protective effect on secondary oxidation will be greater.	(Çoban and Can, 2013)
Smoked eel fillets	Rosemary	Carboxy-methyl-cellulose	Significant lower growth rate for Total Aerobic Bacteria and Lactic Acid Bacteria at the end of storage but low effect on *Pseudomonads* spp. in 800 ppm of polyphenols ethanol extract treatment. Conjugated dienes values stable during the first 20 days in every treatments. After, the higher extract concentration, the lower CD formation at the end of storage.	(Choulitoudi et al., 2017)
Large yellow croaker	Rosemary	Chitosan	Lower Total Viable Counts in raw fish during storage of 20 days. Effective in retarding peroxide values and lipid oxidation. According to a sensorial panel, self-life increments from 12 days to 19 days.	(Li et al., 2012)
gilthead seabream fillets	Satureja	Carboxy-methyl-cellulose	Extract obtained by ethyl acetate showed only antimicrobial properties, extract obtained by ethanol presented antioxidant activity, but essential oil provided both characteristics. Ethyl acetate extract combined with essential oil made shelf-life a 35% longer due to the better antimicrobial effect.	(Choulitoudi et al., 2016)

mechanical property(Bermúdez-Oria et al., 2017). However, there is few studies about the application in packaging.

Fish

Typical deteriorating reactions in fish products include lipid oxidation, enzyme reactions, protein degradation, metabolic activities and microorganism activity. The cross-contaminated microorganism can quickly proliferate and limit the products shelf life. In addition, due to the high content of polyunsaturated lipids in fish, they are more likely to be oxidized (Çoban and Can, 2013).

The application of high pressure and herbs to fish could prevent microbial growth and increase products microbiological stability during storage (Table 3). Gómez-Estaca et al.

Table 4. Main results of selected studies carried out until now on Mediterranean herbal-based films and coatings on fruits and vegetables.

Food application	Mediterranean herb	Matrix	Results	Reference
Butternut squash	Olive Rosemary Garlic Oregano	Chitin and Chitosan	Improves weight loss, percentage of decay, total soluble solids content, ascorbic acid content and acidity. Higher concentration of total phenols and anthocyanins.	(Ponce et al., 2008)
Fresh-cut apricots	Oregano	Basil-seed gum coatings	Reductions of total plate count yeasts and molds populations. Total soluble phenolic and antioxidant activity was enhanced significantly at the end of cold storage.	(Hashemi et al., 2017)
Avocadoes	Thyme	Chitosan	Significant reduction of the microbial incidence and severity of anthracnose after 5 days at 20 °C.	(Bill et al., 2014)
Strawberries	Garlic EO	Cellulose	Significant delay effect on the weight loss.	(Dong and Wang, 2017)
Strawberries	Lavender Thyme	Chitosan	Lower weight loss. Mould-free storage life extension from 2 days (control) to 8 days with the acceptable overall sensory scores.	(Sangsuwan et al., 2016)
Apples Strawberries	olive waste extracts	Chitosan	Beneficial impact on the quality maintenance. Reduced cell wall deterioration activity Antifungal activity against *P. expansum* and *R. stolonifer*	(Khalifa et al., 2016)
Apples	olive waste extracts	Chitosan	Improvements in the nutritional quality. Lower weight loss. Delays increased release of anthocyanins, total phenolics, flavonoids, carotenoids, chlorophyll. Effective antioxidant properties.	(Khalifa et al., 2017)

(2007) reported the effects of oregano-based films with and without high pressure application in the smoked sardine samples. The results indicated that the incorporation of oregano provided a higher amount of phenols in the muscle deriving from the film. Similar results were found in carp muscles (Wu et al., 2014).

Not many studies are available the application of satureja as part of the packaging components, even though is one of the most common herbs belonging to the *Laminaceae* family and its antimicrobial properties are well known. Choulitoudi et al. (2016) reported antimicrobial and antioxidant properties from the EO and the extracts obtained from ethyl acetate and ethanol extraction and their combinations on filleted sea bream. The study indicated that the extract from ethyl acetate did not display any protective effect against oxidative reactions due to the low amount of phenolic acids and the presence of carvacrol which can antagonize with rosmarinic acid and flavonoids. However, the EO and the combination of the extracts were able to reduce substantially the final population of Pseudomonas. The Gram-negative bacteria are resistant to EO due the presence of a thick layer of phospholipids, lipoproteins and lipopolysaccharides impermeable to phenolic compounds (Yi et al., 2010). It has been reported that application of rosemary alcoholic extract at 800 mg/L as a coating additive has a noticeably effect on those bacteria (Choulitoudi et al., 2017). A sensory study also showed that 0.2% (w/v) rosemary coatings in packaging materials could extend the self-life of refrigerated fresh fish during one week by delaying oxidative reactions and minimizing rancid and putrid tastes and odors (Li et al., 2012).

Fruits and vegetables

The demand of ready-to-eat fruits and vegetables is increasing, but they can be easily contaminated during slicing or packaging operations (Qadri et al., 2016). Also, sugar fermentation can occur when yeasts contaminate the damaged tissues. Application of films and coatings provides the possibility of reducing microorganism growth and extending shelf life (Ciolacu et al., 2013).

Bioactive compounds can be extracted from agricultural by-products. These wastes can cause economic losses and environmental problems if they are not processed appropriately. These compounds can be used as antioxidants, antifungals and antibacterial agents to provide additional values

Table 5. Main results of selected studies carried out until now on Mediterranean herbal-based films and coatings on other foods.

Food application	Mediterranean herb	Matrix	Results	Reference
Oilseed kernels	Thyme	Carboxy-methyl-cellulose	Lower values of peroxides and conjugated dienes Lower roasting sunflower, oxidizer and cardboard odor intensity rating Detectable essential oil flavor by sensory panel	(Riveros et al., 2016)
Sliced bread	Oregano	Resins	Reduction of the growth rate of yeasts and molds, being a function of the EO concentration No Salmonella spp. was detected during the 15-day storage at room temperature.	(Passarinho et al., 2014)
Sliced bread	Garlic	Polyethylene Ethylene-vinyl alcohol copolymer Zein	The 0.50% EO samples totally inhibited fungal growth in vitro for all coating polymers tested during the entire storage. Total inhibition of *P. expansum* in zein films. Slight but tolerable garlic flavor in samples packaged in films containing 0.5% EO and 1% bread crust aroma.	(Heras-Mozos et al., 2019)
Shredded low-fat Mozzarella	Rosemary Thyme	Starch	1% mixed Eos showed reductions in *L. monocytogenes* Lactic Acid and Total Aerobic bacteria were significantly reduced by both the individual EO and the mixture	(Han et al., 2014)
Kashar cheese	Thyme	Milk proteins (Whey protein and Casein)	Lower weight loss Lower counts of *E.coli*, *L. monocitogenes* and *S. aureus* during 60 days.	(Kavas et al., 2015)

(Moudache et al., 2016). In this sense, Khalifa et al. (2016) investigated the application of olive waste (leaves and pomace) as a film based to extend the shelf life of apples and strawberries during the storage. Olive leaf wastes were found to be more effective than olive pomace against fungal growth, especially against *Penicillium expansum* and *Rhizopus stolonifer*. It is relevant to remember that strawberries have a very short shelf life (approx. 5 days at 0–4 °C) as they are susceptible to mechanical damages, water loss, physiological deterioration, texture softening and microbiological decomposition, especially by *Botrytis cinerea* growth (Gianfranco Romanazzi and Feliziani, 2014; Vu et al., 2011). Table 4 lists Mediterranean herbs as active edible coatings and films components and their application in fruits and vegetables. Garlic can inhibit a wide range of microorganisms due to its high allicin content. Its usage in films (at concentrations ranging from 1% to 3%) has shown significantly ability to delay the quality decay during storage as well as to maintain the soluble solids content in fruit and vegetable as the respiration and metabolic activities were minimized (Dong and Wang, 2017; Perdones et al., 2012) Conversely, the application of lavender and red thyme in chitosan-based films showed higher rates of respiration in strawberries (Daferera et al., 2003; Sangsuwan et al., 2016). Moreover, oregano and thyme have been studied as natural antimicrobial active compounds on avocados, apricots, and pumpkin (Bill et al., 2014; Hashemi et al., 2017; Ponce et al., 2008).

Others

The use of Mediterranean herbal-based films and coatings to preserve cheese, bread and sunflower grains is indicated in Table 5. Cheese is the most diverse category of dairy products whose shelf life depends on surface microorganisms (Galus and Kadzińska, 2015). It has been reported that it is feasible to reduce the levels of *Escherichia coli*, *Listeria monocytogenes* and *Staphylococcus aureus* with films containing thyme EOs at 1.5 (v/v) concentration in kashar cheese (Kavas et al., 2015). However, it is relevant to mention that conventional antimicrobial packaging methods are not very effective in foods with high porosity. In another study, the addition of volatile EOs into sachets was tested on mozzarella cheese (Han et al., 2014). The results showed that rosemary and thyme could reduce lactic acid formation and decrease the total aerobic bacteria counts, and their mixture significantly reduced *Listeria monocytogenes* growth during storage.

In another study, sachets with volatile EOs (e.g.: oregano) controlled the growth of fungi in bread loaves without any sensory changes (Passarinho et al., 2014). In addition, a flavor corrector (e.g.: bread crust flavor) was developed to cover the off-odor from EO and reduce its sensory impacts (Heras-Mozos et al., 2019). Riveros et al. (2016) investigated thyme EO as a coating material in roasted sunflower grains. The results indicated a reduction in oxidation rates during the storage as the formation of by-products in oxidative reaction was minimized, but the addition of thyme EO can be detected by sensory panel.

Conclusions and future trends

Conventional packaging methods are not suitable to preserve highly perishable foods. Vacuum or MAP packaging are not completely effective as the residual oxygen in the container can trigger oxidation and provide oxygen for bacterial growth. Edible films and coatings with EOs or Mediterranean herbal extracts have been proven to meet these needs. Numerous studies have been done on meat and fish, and to a lesser extent on fruits and vegetables. The results have shown that microbial growth, such as *Listeria monocytogenes* or Pseudomonas, can be inhibited and reduced. The antimicrobial effect varies depending the herb types, the form of EOs or Mediterranean herbal extracts incorporation, their concentration, the matrix and the targeted preserved food. In addition, the detectable off-odor from the EO components also cause a lower consumer acceptance. In this sense, future studies should explore the use a camouflage strategy, such as encapsulation, to overcome this challenge.

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