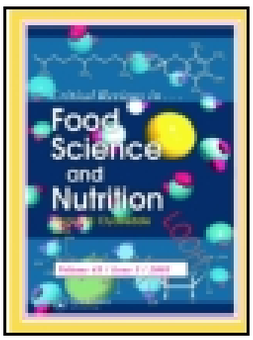

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Ozone in the Food Industry: Principles of Ozone Treatment, Mechanisms of Action, and Applications. An Overview

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Abstract

The food contamination issue requires continuous control of food at each step of the production process. High quality and safety of products are equally important factors in the food industry. They may be achieved with several, more or less technologically advanced methodologies. In this work, we review the role, contribution, importance, and impact of ozone as a decontaminating agent used to control and eliminate the presence of microorganisms in food products as well as to extend their shelf-life and remove undesirable odors. Several researchers have been focusing on the ozone's properties and applications, proving that ozone treatment technology can be applied to all types of foods, from fruits, vegetables, spices, meat and seafood products to beverages. A compilation of those works, presented in this review, can be a useful tool for establishing appropriate ozone treatment conditions, and factors affecting the improved quality and safety of food products. A critical evaluation of the advantages and disadvantages of ozone in the context of its application in the food industry is presented as well.

Keywords

ozone, food industry, microorganisms, inactivation, quality and safety

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Introduction

Safer, “healthier”, higher quality, and less processed foods are currently the main challenges faced during the development of food technologies for the manufacture of food products that will meet consumer expectations. Although there are many decontaminating methods which not only preserve food and control the growth of microorganisms in food, but also ensure the integrity of chemical composition, safety is still the key objective in the food industry. Ozone treatment is one among many existing processes that contribute to the improvement of safety and quality of food products.

Ozone treatment is a chemical method of food decontamination that involves exposing contaminated foodstuffs (fruits, vegetables, beverages, spices, herbs, meat, fish, and so on) to ozone in aqueous and/or gaseous phases. During ozonation, the inactivation of microorganisms is achieved in the gaseous phase at a constant pressure, flow rate, and specified ozone concentration depending on the level of contamination (Brodowska et al., 2014). Using ozone as a decontaminating agent, instead of traditional agents such as chlorine, is justified by its significant oxidative properties. It is about 50% stronger than chlorine, and thus is characterized by a broad spectrum of antibacterial activities. The bactericidal effects of ozone have been confirmed on a wide variety of microorganisms, including Gram-positive and Gram-negative ones, as well as bacterial spores (Guzel-Seydim et al., 2004a; Kunicka – Styczyńska and Śmigielski, 2011). Therefore, the application of ozone both improves the microbiological safety of food products, and prolongs their shelf-life without substantially changing their nutritional, chemical, and physical properties.

As a powerful antimicrobial substance, ozone has been used for years to disinfect water for various purposes, including drinking (bottled water), swimming (swimming pools), spas, marine aquaria, to prevent fouling of cooling towers, as well as to treat municipal water and sewage. Additionally, it can be used in the meat, vegetable, fruit, fish, as well as herb, spice and beverage industries (Gonçalves, 2016; Guzel-Seydim et al., 2004a; Peleg, 1976; Strittmatter et al., 1996; Tapp and Rice, 2012). However, ozone toxicity is the most important criterion for its approval in a wide variety of food branches (Pryor et al., 1995). Furthermore, it is important to monitor the dose of ozone and the people who may have been exposed to it at the workplace. This review intends to compile information that will contribute to the identification of the main achievements pointing to what still needs to be exploited, for opening further areas of research.

Principles of ozone treatment

There are different ways by which ozone may be produced. It might be generated by the exposure of air or another gas mixture which contains oxygen to a source of energy such as a high-energy electrical field (corona discharge method), ultraviolet radiation (phytochemical method), or conversion of oxygen molecules (O_2) to ozone (O_3) (chemical method). However, it has to be generated just before use because of its rapid degradation to oxygen. As it cannot be accumulated, ozone has to be produced continuously when needed. Other methods of ozone generation include electrolysis, reaction of elemental phosphorus with water, and radiochemical production. But these procedures are cost-ineffective for the food industry or are in their early stages of development. Generally, only 2 methods are used in practice: photochemical (UV) as well as corona discharge (CD), in some works referred to as a plasma technique (Gonçalves, 2009; Guzel-Seydim et al., 2004a; Pirani, 2010; Tapp and Rice, 2012). Although some

applications in the food industry include CD and UV, but separately used, the first one seems to be the most applicable. However, there are a few applications such as air fumigation, as well as modified-air packaging applications, that make use of the UV radiation (Table 1) (Tapp and Rice, 2012).

The main principle of corona discharge technology is passing dried, dust-free, oil-free air or an oxygen-containing gas mixture, or O_2 itself, through the space of a high-energy electrical field between two electrodes separated by a dielectric material, usually glass. One electrode is a grounded medium, and the other is a dielectric one. The important thing is to control the temperature of the processed gas because ozone may be decomposed by heating, namely may undergo an endothermic reaction (Eq. 1). If about 80% of the applied energy is exceeded, it is converted to heat and then, if not removed, a spontaneous degradation of ozone into oxygen ions and molecules occurs, in particular above 35 ^{o}C. Therefore, to prevent such decomposition, ozone generators have to be equipped with an electrode cooling system (Figure 1). Besides, the entropy of ozone formation is high and unfavorable (Eq. 2). Ozone cannot be produced using temperature to activate the oxygen because the standard enthalpy of formation is high (Eq. 3) (Miller et al., 2002; Patil and Bourke, 2012; Tapp and Rice, 2012).

$$3O_2 \leftrightarrow 2O_2 (\Delta H^O at 1atm, + \frac{284.5kJ}{mol}) \quad \text{(Eq. 1)}$$

$$\Delta S^O (1atm) = -\frac{69.9Jmol}{^oC} \quad \text{(Eq. 2)}$$

$$\Delta G^O (1atm) = +\frac{161.3kJ}{mol} \quad \text{(Eq. 3)}$$

When passing through the electrical field, the oxygen molecules are split into oxygen atoms (O), and thereby very active atomic oxygen radicals are formed, which in combination with the remaining oxygen molecules (O_2) are capable of generating ozone (O_3) (Eq. 5 and Eq. 6).

$$O_2 + e^- \rightarrow 2O \qquad \text{(Eq. 5)}$$

$$O_2 + O \rightarrow O_3 \qquad \text{(Eq. 6)}$$

The mixture of ozone and gas discharged from an ozone generator contains about 1 – 3% ozone if dry air is used, or 3 – 6% ozone when high-purity oxygen is applied as the feed gas. The electrodes are formed into concentric tubes or flat plates. During voltage supply (5000 V) to the corona discharge, ozone is formed between the 2 electrodes, and then oxygen molecules in the discharge gap are converted to ozone. This technology is characterized by a low electrical discharge across a gas-containing gap at a voltage gradient exceeding a certain critical value (Patil and Bourke, 2012).

The ultraviolet technology uses UV light of 140 – 190 nm wavelength to form ozone. During photodissociation of the oxygen molecules, some of them (O_2) are split into unstable oxygen radical atoms (O_1). The resulting free oxygen radical atoms attach to O_2 molecules, and react with other diatomic oxygen, thus forming O_3 molecules, or ozone (Pirani, 2010; Tapp and Rice, 2012; Voronov, 2008). In particular, low-pressure mercury lamps are used to produce the ozone as well as to treat the air. The most important advantage of high-transmission UV lamps is the emission spectrum of mercury discharge – it emits 2 high-efficiency resonance lines with wavelengths of 185 and 254 nm. The first photons (185 nm wavelength) are responsible for ozone generation. The other line, with the wavelength of 254 nm, is claimed to change the DNA of microorganisms, and thus to impair their reproductive ability. But the target wavelength of

254 nm of the UV lamps, which is mainly used to inactivate microorganisms, is useless in ozone generation, because ozone is destroyed at this wavelength (DuRon, 1982; Tapp and Rice, 2012). Therefore, a lamp wavelength enabling ozone production is 185 nm. Air passes through the high-energy UV_{185} system as a feed gas. However, this method has very limited uses, mainly due to poor yields. If ozone is produced in this way for industrial purposes, it is produced from oxygen in the air at low concentrations (about 0.03 ppm) by radiation (185 nm wavelength). The process is mostly carried out at the point of application, in closed systems (Muthukumarappan et al., 2000; Pirani, 2010).

Equipment for ozone treatment in the food processing industry

Ozone treatment may utilize ozone in aqueous and/or gaseous phases in the food processing industry. In practice, ozone treatment systems have few basic components: the gas (air or pure oxygen), an ozone generator, an electric power source, a contactor (if ozone is in the water phase), a reactor, a surplus gas elimination unit, and an ozone analyzer. Generally, in corona-discharge type generators dry air or pure oxygen are used as an oxygen source for conversion into ozone. If air is used, it is essential to dry it to a point of condensation of -65 ^{o}C to increase ozone treatment effectiveness and prevent the formation of nitrogen oxides that accelerate electrode corrosion. Usually, zeolite towers, which act as molecular screens, are used to produce pure oxygen by disabling the formation of nitrogen compounds in the air. The air should also be cooled because of rapid ozone decomposition into oxygen at temperatures over 30 ^{o}C. During ozone treatment, devices usually operate at a low frequency (50 to 60 Hz) and a high voltage (> 20,000 V), but advanced technologies require a higher frequency (1000 to 2000 Hz) and 10,000 V because only then they are more effective. Ozone-based systems for water treatment use

contactors to transfer the generated ozone to the water for disinfection. Depending on the target of ozone treatment, there are two basic contactors: with bubble diffuser chambers and containing a turbine-agitated reactor. It was studied that a multicolumn contactor with bubble diffuser is a productive transfer (Bablon et al., 1991). Besides, any contact chambers, turbine diffusers and static agitators can be helpful in accelerating the gaseous ozone to ensure mixing and maximize contact. After ozone treatment, the surplus ozone should be destroyed due to the safety considerations. It can be diluted with air, in the case of small treatment factories, or destroyed by catalytic decomposition or absorption in moist granular activated carbon in the case large treatment factories (Bablon et al., 1991).

Brodowska et al. (2014) suggested a simplified scheme of an apparatus for ozone treatment in a gaseous medium for laboratory purposes (Figure 2) involving the treatment of plant material contaminated with ozone. The apparatus allows for ozone treatment in a reactor (a cylindrical glass and steel chamber), in which a contaminated sample is treated continuously with the ozone/oxygen mixture. Additionally, the apparatus is equipped with a control system with a jolting and rotating mechanism, directly connected with the reactor, thereby intensifying movement of the plant material within the chamber. The ozone analyzers allows determining ozone concentration at the inlet and the outlet. On the other hand, the apparatus for ozone treatment in the water phase consists of similar elements (Figure 3), but the pH of the sample solution in the reactor should be continuously controlled (Brodowska et al., 2014, 2015; Naito and Sawairi, 2000).

Ozone in the aqueous phase

Aqueous ozone solutions are important in practice for water disinfection and purification. Ozone dissolved in water decays much faster than that dissolved in oxygen or air. Its solubility in water is 10 times higher than of oxygen (C_{O2} = 1.3*10^{-3} mol/dm^3; C_{O3} = 1.2*10^{-2} mol/dm^3; 293 K; 101.3 kPa) (Ullmann's Encyclopedia, 2002). It dissolves in water at pH below 7, where it does not react with water and occurs in the form of molecules. An increase in pH causes spontaneous ozone decomposition, which leads to the production of highly reactive free radicals, such as hydroxyl radicals, as well as oxygen and hydroxide ions, especially at pH above 7.5. The oxidation potential is recognized as higher for hydroxyl radicals (2.80 V) than for ozone (2.07 V). At pH 8, almost half of the introduced ozone is decomposed to various intermediate forms and to oxygen, in time no longer than 10 minutes (Manley and Niegowski, 1967; Manousaridis et al., 2005; Pekhonen, 2001; Pirani, 2010). Ozone decay leads to the production of the following radicals: hydroperoxyl ($^{\bullet}HO_2$), hydroxyl ($^{\bullet}OH$), and superoxide ($^{\bullet}O_2^-$). These free radicals are attributable to the great oxidizing power. Ozone stability is determined by water quality as well as purity – it decays within a few seconds to hours. The rate of ozone degradation to oxygen in impure solutions is higher than in pure water (Hill and Rice, 1982; Hoigne, 1998; Manousaridis et al., 2005; Miller, 2005). This fact was confirmed by Hill and Rice (1982) who had concluded that ozone was found to decompose by 50% within 20 min at 20 ^{o}C in distilled as well as tap water, and by only 10% within 85 min at 20 ^{o}C in double-distilled water (Hill and Rice, 1982). In natural waters, ozone decay is initially characterized by a rapid decline of ozone concentration, after which the phase of ozone decay appears according to the first-order reaction kinetics. A secondary half-life is introduced, which is the time needed to reduce ozone concentration from

50 to 25%. In distilled water and at ambient temperature, it is about 25 seconds at pH = 10, 17 min at pH = 7, and increases to 7 hours at pH = 4 (Hoigne, 1998; Miller, 2005). However, ozone solubility is also affected by temperature. The higher the water temperature, the lower its solubility is (Greene et al., 2012; Guzel-Seydim et al., 2004a; Miller, 2005; Pirani, 2010). For instance, ozone is more soluble in water at 0 ^{o}C (0.6401 ozone/L water) than at higher temperature - 60 ^{o}C where it is insoluble (Guzel-Seydim et al., 2004a). Apart from pH, ozone stability in the aqueous phase is also affected by alkalinity (concentration of carbonates) as well as organic matter content. The life of ozone in an aqueous solution is shorter, at higher pH, with a higher content of organic matter, and lower with the presence of carbonates. Generally, it is claimed that ozone half-life in distilled water at a concentration of a few % (by weight) at room temperature is approximately 20 – 30 min (Miller, 2005; Pryor and Rice, 2000). Liquid ozone, in contrast to the gaseous ozone which is relatively safe, may explode even at very low temperatures (Toby, 1984).

Reactions ongoing in water during ozone treatment

Ozone decays in water into free radicals having an unpaired electron – hydroxyl ($^{\bullet}OH$) radicals which are a result of the indirect reaction pathway. As it has been mentioned, *OH* radicals possess a stronger oxidizing capacity than ozone. As of most all short-lived compounds, due to their instability, radicals directly react with another molecule to reclaim the missing electron. It is claimed that reaction pathways in an ozone oxidation process can be very complex, thus leading to the formation of primary high-reactive species (Gottschalk et al., 2010; Greene et al., 2012; Gunten, 2003; Sehested et al., 1991).

Gottschalk et al. (2010), as well as Sehested et al. (1991), described an indirect reaction of ozone oxidation process as a model process consisting of several steps, including initiation, radical chain reaction, and termination.

The first one – initiation – involves the acceleration of ozone decomposition initiated by an initiator, for instance, hydroxyl molecule. Firstly, hydroxide ions react with ozone (Eq. 7), resulting in the formation of an O_2^{-o} anion and an HO_2^o radical:

$$O_3 + OH^- \rightarrow O_2^{-o} + HO_2^o \qquad \text{(Eq. 7)}$$

Such a radical has an acid/base equilibrium at pKa = 4.8, but when this value is exceeded, the hydroxyl radical forms only a O_2^{-o} and that is why it no longer splits (Eq. 8):

$$HO_2^o \rightarrow O_2^{-o} + H^+ \qquad \text{(Eq. 8)}$$

The second step consists in a radical chain reaction in which OH radicals are formed (Eq. 9, Eq. 10 and Eq. 11). During this reaction, only one O_2^{-o} anion reacts with ozone resulting in an O_3^{-o} anion. Then, it decomposes quickly through hydrogen trioxide (HO_3^o) to an OH^o radical as follows:

$$O_3 + O_2^{-o} \rightarrow O_3^{-o} + O_2 \qquad \text{(Eq. 9)}$$

$$HO_3^o \leftrightarrow O_3^{-o} + H^+ \qquad \text{(Eq. 10)}$$

$$HO_3^o \rightarrow OH^o + O_2 \qquad \text{(Eq. 11)}$$

Due to the formation of an OH^o radical, the next reaction is as follows (Eq. 12 and Eq. 13):

$$OH^o + O_3 \rightarrow HO_4^o \qquad \text{(Eq. 12)}$$

$$HO_4^o \rightarrow HO_2^o + O_2 \qquad \text{(Eq. 13)}$$

The HO_2^o radicals allow initiating a higher number of reactions, which results in a chain reaction maintained by promoters. These are substances which transform hydroxyl (*OH*) radicals to superoxide (O_2^{-o}) radicals, for instance organic molecules.

The last step is termination, where radical scavengers terminate the above chain reaction. It is caused by a reaction between organic as well as inorganic substances such as CO_3^{2-} and HCO_3^-, and *OH* radicals. Secondary radicals resulting from this reaction do not produce superoxide radicals. Gottschalk et al. (2010) showed an example of the reaction of 2 radicals (Eq. 14):

$$OH^o + HO_2^o \rightarrow O_2 + H_2O \qquad \text{(Eq. 14)}$$

Due to the molecular structure of ozone, a scavenger can be an electrophilic or a nucleophilic agent. Electrophilic reactions proceed mainly upon the presence of organic contaminants in water having a high electron density, and their rate is higher in solutions containing non-aliphatic compounds. On the other hand, nucleophilic reactions will occur when there is a lack of electrons, in particular with carbon compounds containing electron-withdrawing groups, including - *COOH* and NO_2. Generally, the direct reaction of the ozone oxidation process involves a selective reaction mechanism. Noteworthy is the pH value of water which has an impact on ozone decay (Gunten, 2003; Miller, 2005).

Distribution of ozone in "pure" water at ambient temperature (21 ^oC) is as follows:

$$O_3 + OH^- \rightarrow HO_2^- + O_2 \qquad \text{(Eq. 15)}$$

$k = 70\ dm^3/mol*s$

$$O_3 + HO_2^- \rightarrow O_2^{o-} + HO^o + O_2 \qquad \text{(Eq. 16)}$$

$k = 2.8*10^6$ dm^3/mol*s

$$O_3 + O_2^{o-} \rightarrow O_3^{o-} + O_2 \qquad \text{(Eq. 17)}$$

$k = 1.6*10^9$ dm^3/mol*s

pH < ~ 8; pH > ~ 8

$$O_3^{o-} + H^+ \rightarrow HO_3^o \quad O_3^{o-} \leftrightarrow O^{o-} + O_2 \quad \text{(Eq. 18)}$$

pK = -8.2; pK = 6.2

$k_+ = 5*10^{10}$ dm^3/mol*s; $k_+ = 2.1*10^3$ dm^3/mol*s

$k = 3.3*10^2$ dm^3/mol*s; $k = 3.3*10^9$ dm^3/mol*s

$$HO_3^o \rightarrow HO^o + O_2 \qquad O^{o-} + H_2O \rightarrow HO^o + OH^- \qquad \text{(Eq. 19)}$$

$k = 1.4*10^5$ dm^3/mol*s; $k = 1*10^8$ dm^3/mol*s

$$O_3 + HO^o \rightarrow HO_2^o + O_2 \qquad \text{(Eq. 20)}$$

$k = 1*10^8$ or $2*10^9$ dm^3/mol*s

The reaction sequence (Eq. 15 – Eq. 20) has a chain character. The products formed are radicals that participate in reactions which intensify ozone distribution. In water solutions, ozone decomposition proceeds through chemical pathways. The hydroxyl radical is an important transient species and chain-propagating radical. Liquid ozone may explode if more than 20% of an ozone – oxygen mixture occurs (Greene et al., 2012; Gunten, 2003; Miller, 2005). Ozone decomposition is so rapid in the water phase of foods that its antimicrobial effect may occur mainly at the surface (Hoigne, 1998; Pirani, 2010).

Ozone in the gaseous phase

Unlike the liquid form, pure gaseous ozone is relatively safe and kinetically stable under the pressure of several kPa at room temperature. Theoretical ozone decomposition, determined from the frequency of collisions, under a pressure of 101.3 kPa at 298 K is only 0.2% per year. Moreover, noteworthy is that any contamination as well as surface reactions can lead to the chain reactions, which under high pressure makes ozone a dangerous substance (Miller, 2005; Toby, 1984). However, a mixture of gaseous ozone with oxygen or air is preferably used over pure ozone during ozone treatment. The explosion limits of the ozone-oxygen mixture depend on several factors, including ozone concentration, temperature, pressure, the presence of contaminants and catalysts, as well as size and dimensions of the reactor. Actually, due to the high risk of using pure ozone in liquid and gaseous phases, ozone is practically produced *in situ*. Safe mixtures are considered those containing more than 9 – 16 mole percent of ozone; the top value can be achieved while maintaining special precautions. Above these levels, under air pressure and at ambient temperature, a spontaneous explosion takes place. However, independently of ozone dose, rapid pressure and temperature changes, as well as mechanical shocks and an unclean apparatus should be avoided to ensure safety at work (Miller, 2005; Pryor and Rice, 2000). In addition, the risk of explosion is higher when the temperature drops, and is lower when the pressure increases. Besides, it also feasible to produce ozone containing approximately 40% of mass in oxygen at low temperatures, respectively, as well as generate it under a pressure of 1.3 MPa. One must pay attention to the fact that at temperatures close to the boiling point of oxygen (90.2 K) ozone-oxygen mixtures separate into layers rich and poor in ozone, and the rich one poses a significant danger. Therefore, in the case of work with highly

concentrated mixtures, the temperature cannot be lower than 95 K (Greene et al., 2012; Miller, 2005).

In contrast to a liquid state, gaseous ozone has a longer half-life (Rice, 1986). Bailey et al. (2002), Horvath et al. (1985), and Kirk-Othmer (1967) studied ozone decomposition in the gaseous phase at temperatures ranging between 80 and 500 ^{o}C (353 – 773 K). They noticed that the ozone decay followed the below presented reactions (Eq. 21, Eq. 22 and Eq. 23):

$$O_3 + M \rightarrow O_2 + O + M \qquad \Delta H = 103 \text{ kJ/mol} \qquad \text{(Eq. 21)}$$

$$O_2 + O + M \rightarrow O_3 + M \qquad \text{(Eq. 22)}$$

$$O_3 + O \rightarrow 2O_2 \qquad \Delta H = -389 \text{ kJ/mol} \qquad \text{(Eq. 23)}$$

where M means “the third body”, which may be O_2, O_3, N_2, He, or CO_2 (Bailey et al., 2002; Horvath et al., 1985; Kirk-Othmer, 1967). The energy emitted during explosion is estimated at 284 kJ according to the following reaction (Eq. 24) (Dietrich et al., 2000):

$$2O_3 \rightarrow 3O_2 \qquad \text{(Eq. 24)}$$

In the explosive decomposition of ozone, a significant role is probably played by atomic oxygen. It can cause further ozone decay, while a large amount of energy carried by the two oxygen molecules (Eq. 23) has an impact on an increase in the process rate. The atomic oxygen can also react with water steam, in which hydroxyl radicals are formed. Because of their high reactivity, they can cause further decay of ozone (Miller, 2005; Perry et al., 1997). The half-life of ozone decreases with an increase of ozone dose in the gaseous state and with an increase of temperature. The half-life of ozone in the gaseous solution of 5% of ozone mass at 120 ^{o}C is 11.2

min, and at 200 ^{o}C it is 0.9 s. At room temperature the ozone in an ozone-oxygen mixture decomposes within 20 to 100 hours, depending on its concentration (Kirk-Othmer, 1967).

What is more, ozone decay in the gaseous phase may be initiated by some gaseous substances, including nitrogen oxides (*NO, NO_2*), chlorine, sulfur di- and trioxide, as well as hydrogen sulfide (Dietrich et al., 2000; Miller, 2005).

Furthermore, ozone decomposition can be initiated by electrical discharges, where the atomic oxygen and oxide anion are formed (Eq. 25 and Eq. 26). Both forms of oxygen can initiate further decay of ozone (Horvath et al., 1985):

$$O_3 + e^- \rightarrow O_2 + O + e^- \qquad \text{(Eq. 25)}$$

$$O_3 + e^- \rightarrow O_2 + O^{o-} \qquad \text{(Eq. 26)}$$

Advantages and disadvantages of ozone treatment

The primary advantage of ozone treatment is most likely a non-residual feature of the process. In contrast to the chemical methods (formaldehyde, ethyl alcohol), which leave the residual compounds that may possess or tend to possess carcinogenic properties having a negative impact on human health, ozone treatment is free of the chemical residues (Brodowska et al., 2014; Patil and Bourke, 2012; Pirani, 2010; Tapp and Rice, 2012). Furthermore, ozone treatment is a promising substitute for the conventional fumigation in use (SO_2). Additionally, it can be applied to all types of foods, from fruits, vegetables, spices, meat and seafood products to beverages (Guzel-Seydim et al., 2004a; Peleg, 1976; Strittmatter et al., 1996; Tapp and Rice, 2012). An important issue is also that no matter what the product state is, it can be used for both fresh and frozen foods. Since ozone has been found to be effective in reducing the microbial contamination (pathogenic and nonpathogenic microorganisms) of a food product without having an

unfavorable effect on its visual, textural and nutritional quality and in extending its shelf life, surely it can be recommended and incorporated into the supply chain. Moreover, ozone treatment is considered as a cost-effective and eco-friendly food processing technology. Ozone use can be beneficial because of the lower costs of the purchase and maintenance of the ozone supply units compared to the cost of the purveyance of disinfectants (Glowacz et al., 2014; Greene et al., 2012).

Concerning the disadvantages of this method, first, microorganisms possess a different sensitivity to ozone which depends on several factors, including the type of product, target microorganism, the initial level of contamination, physiological state of the bacterial cells, the physical state of ozone, as well as the type of an organic material (Restaino et al., 1995; Miller et al., 2013). The fact that ozone treatment effectiveness is strongly determined by so many factors may cause some limitations in the selection of a sufficiently effective ozone dose. Additionally, some care must be taken if higher ozone concentrations are required for reducing microbial counts, because it can negatively affect food quality preservation by the emergence of toxic symptoms. Most relevant for foods is the reduction of vitamin, polyphenol, and volatile compound contents, color changes, and loss of firmness, water, and weight (Miller et al., 2013). Nonetheless, most of these changes can also be induced by the traditional food preservation methods, such as cooking, canning, pickling, freezing, and drying, etc.

Furthermore, due to the fact that ozone is unstable in water, in which some ozone-resistant compounds occur, including pesticides and chlorinated solvents, only partial oxidation may take place (Hoigné, 1998). Thus, with the cost-effective ozone generation, ozone treatment cannot be economically viable. Another disadvantage of ozone application in food is its poor acceptability

by consumers as it is believed to have toxicity properties. However, in recent years consumer's perception has changed and ozone attracted their attention due to being an alternative to chlorine which possesses a negative impact on human health and safety. Nonetheless, the effective and comprehensive information regarding the new technologies and their benefits is important for consumer acceptance (Frewer et al., 2003; Aday et al., 2014).

Mechanism of antimicrobial action of ozone

Ozone has a broad spectrum of antimicrobial activities, which results from its high reactivity, which in turn is due to the oxidizing power of free radicals. Due to its instability in both the aqueous and gaseous phases, ozone decomposes into hydroxyl, hydroperoxy, and superoxide radicals (Manousaridis et al., 2005; Pirani, 2010). However, it is claimed that molecular ozone is the main inactivator of microorganisms, researchers indicated the mentioned highly reactive by-products created during ozone decomposition as a source of potential antimicrobial activity (Hunt and Marinas, 1997; Pirani, 2010).

The inactivation of bacteria during ozone treatment is a complex process because it concerns an ozone attack on cell membrane constituents (proteins, respiratory enzymes, unsaturated fatty), cell envelopes (peptidoglycans), cytoplasm (enzymes, nucleic acids), spore coats, and virus capsids (proteins and peptidoglycan) (Greene et al., 2012; Guzel-Seydim et al., 2004a; Pirani, 2010). Some researchers suggested that there are possibly 2 primary mechanisms of microorganism inactivation by ozone. The first one includes the oxidation of sulfhydryl groups and amino acids of enzymes, peptides, and proteins to produce smaller peptides during ozone exposure, whereas the second mechanism involves the oxidation of polyunsaturated fatty acids to acid peroxides. Microorganism inactivation is claimed to occur due the damage to a cell

envelope, or its disintegration which leads to subsequent leakage of cellular contents and cell lysis (Greene et al., 2012; Victorin, 1992).

Nevertheless, a destructive action of ozone on bacteria involves the successive damage of cell walls, cytoplasmic membrane, and finally the DNA structure of the bacterial cell, resulting in disability to resist an ozone attack (Oizumi et al., 1998). The first step includes the breakdown of the bacterial cell wall, under which there is a cytoplasmic membrane consisting of phospholipids. These compounds contain polyunsaturated fatty acids, which due to the ozone action undergo a peroxidation process. This results in the oxidation of unsaturated fatty acids or their residues upon the action of free radicals, and in the formation of peroxides of those compounds (Margalit et al., 2001). During the initiation phase, due to ozone activity, a hydrogen molecule is peeled off from the rest of the unsaturated fatty acid molecule. Then, a free alkyl radical with an unpaired electron in a carbon atom without a hydrogen atom is formed. The next stage involves the rearrangement of the double bonds, which results in the formation of conjugated bonds. After the initiation phase, followed by a series of chemical reactions, the lipids are completely peroxidized. The peroxidation products change the physical properties of the cell membranes, causing their depolarization and inhibition of the activities of the membrane enzymes and transport proteins. Furthermore, the reactions with strong oxidizing agents such as ozone may also lead to amino acid, protein, as well as nucleic acid oxidation when the immediate cell destruction is not sufficient. However, it is believed that the destruction of the membrane barrier is the main factor leading to the secondary DNA damage and, finally, death of the cell (Antoszewski and Madej, 1997; Antoszewski et al., 2004; Margalit et al., 2001).

Microorganism susceptibility to ozone

Numerous studies have confirmed the bactericidal effect of ozone on a variety of microorganisms, including Gram-positive and Gram-negative bacteria, as well as spore forms and vegetative cells (Brodowska et al., 2014; Pirani, 2010). The antimicrobial efficacy varies significantly depending on the experimental conditions. Therefore, it is not feasible to compare the results of ozone sensitivity to bacteria with different studies (Pirani, 2010). Selected investigations have been reported to illustrate the effect of ozone on food-related microorganisms, including Gram-positive bacteria (*Listeria monocytogenes*, *Bacillus cereus*, *Staphylococcus aureus*, *Enterococcus faecalis*) and Gram-negative ones (*Pseudomonas aeruginosa*, *Yersinia enterocolitica*), and also yeasts (*Candida albicans*, *Zygosaccharomyces bailli*) and molds (*Aspergillus niger*, *Botrytis cinerea*) (Barth et al., 1995; Greene et al., 2012; Hildebrand et al., 2008; Liew and Prange, 1994; Palou et al., 2002; Perez et al., 1999; Restaino et al., 1995; Tzortzakis et al., 2007a, b). Some of them showed that the range of *Escherichia coli* inactivation after ozone treatment was by 0.5 to 6.5-log, as affected by ozone dose (from 4.4 to 800 µg/L) and treatment duration (from 30 to 120 s) (Finch et al., 1988). Furthermore, Dave et al. (1998) report that the bacterial inactivation by ozone is strictly related to its environmental medium. They demonstrated a 6-log decrease in counts of *Salmonella enteritidis* in distilled water at low ozone concentration (1.5 ppm) (Dave et al., 1998). In another study, broiler skin was inoculated with *S. enteritidis* and exposed to an ozone-air mixture (8%, wt/wt; 15 s), resulting in a 1-log decrease after 15 s (Ramirez et al., 1994). Restaino et al. (1995) confirmed that Gram-positive bacteria such as *Listeria monocytogenes* were more sensitive to ozone exposure in the water phase than the Gram-negative ones (*Escherichia coli*, *Yersinia*

enterocolitica). Some studies, however, recognized Gram-negative bacteria as more resistant than the Gram-positive ones due to the higher content of peptidoglycan in their cell walls. Because of the fact that the primary destruction of the lipoprotein and lipopolysaccharide layers occurs in Gram-negative bacteria, an increase in cell permeability and eventual cell lysis were observed (Kim et al., 1999; Victorin, 1992).

Comparing sensitivity of bacterial spores and vegetative cells to ozone exposure, the spores are more resistant to ozone. As it has been observed, the multilayered spore coat is a major barrier protecting against ozone. Therefore, ozone treatment combined with another damaging factor is believed to enhance bacterial spore inactivation. The study conducted by Naitoh (1992a, 1992b) confirmed that ozone treatment (5 – 50 ppm; 1 – 6 h) coupled with an addition of ascorbic acid, isoascorbic acid, or metallozeolites significantly reduced the count of *B. subtilis* spores. Moreover, the same author suggested the synergistic sporicidal activity of gaseous ozone and UV irradiation, which possibly shortens the ozone contact time (Naitoh 1992a; 1992b). Furthermore, Guzel-Seydim et al. (2004b) evaluated the effect of food components (fats, proteins, carbohydrates) on a bactericidal cell's power against ozone. The efficacy of ozone to reduce counts of *Bacillus stearothermophilus* spores, vegetative cells of *E. coli*, and cells of *Staphylococcus aureus* using sterile Class C buffer, whipped cream, and 1% solutions of the locust bean gum, soluble starch, and sodium caseinate was studied as well. The results demonstrated that the use of starch and an ozone dose of 4 ppm for 10 min had no protective effect on the vegetative cells in comparison to the buffer control, while locust bean gum possessed same protection level. In turn caseinate and whipped cream ensured the greatest level of bacterial population protection against ozone (Guzel-Seydim et al., 2004b). Also, it was

suggested that *Bacillus* spores treated with ozone were inactivated as a result of outer spore component degradation. This could be explained by the spore coat layers representing almost 50% of the spore volume, and thereby preventing cortex and core exposure to the action of ozone (Greene et al., 2012; Khadre et al., 2001). Young and Setlow (2004) confirmed that not the damage of the DNA by ozone, but the loss of spores ability to germinate (caused by damage to the spore's inner membrane) was the main reason for spore inactivation.

Furthermore, ozone has a fungicidal activity (Freitas-Silva and Venancio, 2010; Miller et al., 2013; Palou et al., 2001). Freitas-Silva and Venancio (2010) suggested that the mechanism of fungal inactivation by ozone is related to membrane integrity damage. Similarly to bacteria, it has been reported that mold species are characterized by different sensitivities to ozone. Palou et al. (2001) compared the impact of ozone on *Penicillium italicum* and *Penicillium digitatum* growth. The results indicated that the first *P. italicum* was affected by ozone, while *P. digitatum* was found to be resistant (Palou et al., 2001). Moreover, the reported data showed that the gaseous form of ozone was usually used for mold growth inactivation (Miller et al., 2013). However, Zorlugenc et al. (2008) investigated, in a comparative study, the effectiveness of gaseous ozone and aqueous ozone in mold growth inactivation and removal of aflatoxin B1 from dried figs. The gaseous state of ozone was more effective than the aqueous one in toxin reduction, whereas ozonated water was recognized to be more effective in the inactivation of mold growth (Zorlugenc et al., 2008).

Some researchers have suggested that ozone may be an alternative to other fumigation methods (Sarig et al., 1996; Whangchai et al., 2006). In the study conducted by Ewell (1938) on the effects of ozone treatment plus subsequent storage condition (ozone dose from 0.6 to 1.5 ppm, at

0.6 °C and 90% relative humidity), mold reduction on hen eggs was achieved. The same storage conditions (ozone concentration from 2.5 to 3.0 ppm) enabled mold growth control in beef (Ewell, 1938).

What is more, ozone was shown to inactivate yeasts (Farooq and Akhlaque, 1983). Yeasts seem to be more sensitive to ozone than molds. Restaino et al. (1995) demonstrated that counts of *Candida albicans* and *Zygosaccharomyces bailii* were reduced by more than 4.5 log units using ozonated water, whereas *Aspergillus niger* spores were decreased by less than 1 log unit during a 5-minute treatment.

Ozone use for odor removal

Due to increasing complaints about unpleasant odors developed as a result of physical food processing mainly upon biological and chemical reactions, industries are obliged by the environmental legislation to seek effective solutions (technologies) to reduce or even eliminate the malodorous volatile organic compounds from areas closely located to the emission source (Domeno et al., 2010; Mahin, 2001; Tyndall and Colletti, 2007). Rappert and Muller (2005) suggested two ways of odor control: the first one, more preferable, included a reduction of odor production at the source of emission, and the second one – treatment of odors from gaseous streams before they are released into the environment. However, a continuous control of odor is expensive due to the large amounts of chemicals needed during repeated periods of the process which make deodorization technology more effective (Arvanitoyannis, 2012).

Though applications of ozone in the food industry concern mainly decontamination of a product surface, as well as water treatment, it is also successfully used as an agent for odor removal (Chawla, 2006). Numerous reports have claimed the substantial reductions in some

volatile organic compounds that caused an undesirable odor emission through the action of ozone. Below, we will review the main achievements reported in this respect.

One of the first reports studied the effect of ozone treatment on gaseous and liquid odor emissions from animal rendering plants (Arsovic and Burchard, 1973). Neither using chlorine nor ozone alone brought the significant reductions. A greater decrease in a malodor level was observed while combining chlorine with ozone (Arsovic and Burchard, 1973). Similarly, Mohana et al. (2009) found ozone to be suitable to remove the malodor emission from distillery spent wash due to the occurrence of skatolic, indolic, and sulfuric compounds, which remained after the distillation process.

Considering ozone treatment employment in the fish industry, the removal of off-odor compounds such as geosmin and 2-methylisoborneol (MIB) in rainbow trout was reported by Schrader et al. (2010). Water continuously treated with ozone (0.25 – 0.28 mg/L) was not sufficient to ensure significant reduction of such compounds, but this treatment improved water quality. Certainly, an increasing ozone concentration in water can decrease the level of off-odor compounds, but might as well be toxic to rainbow trout. Therefore, the low ozone dose has demonstrated that it has no benefits in a solution of off-flavor problems with geosmin and MIB (Schrader et al., 2010). These observations were confirmed in later studies of Zhang et al. (2016), which proved that ozone levels of 3.3 mg/L, 5.1 mg/L, and 7.6 mg/L were more effective in reducing geosmin content. These authors evaluated the impact of ozone treatment (ozonated water and ozone-flotation) on geosmin removal from the fish muscle of bighead carp. Washing with ozonated water or ozone-flotation was recognized to be effective in the elimination of geosmin from fish muscle. Ozone-flotation was found to have a greater impact on the scavenging

process than the ozonated water. After using ozonated water, the content of geosmin decreased by 42.09, 53.84, and 54.28% for 5, 10, and 15 min, respectively, whereas the application of ozone-flotation reduced it by 42.78, 60.58, and 69.19%. Although two methods of treatment were effective in eliminating a muddy flavor, the ozonated water ensured an increase in the SEP content, Ca^{2+}-ATPase activity, the active sulfhydryl, carbonyl content, and gel strength. Peroxide and TBA values remained virtually unchanged. Thus, the appropriate conditions of oxidation with ozonated water are required to make ozone an alternative technique of elimination of off-flavors and enhancement of the physicochemical properties of the surimi gel (Zhang et al., 2016).

Kim-Yang et al. (2005) evaluated the effect of ozone treatment on the reduction of odors produced from swine housing facility, and observed that the levels of indolic compounds greatly decreased at low gaseous ozone doses (0.01, 0.05, and 0.1 ppm). But, ozone did not significantly affect the level of volatile fatty acids and phenolic compounds in the air. The authors suggested that the higher doses of ozone would be required to achieve the greater reductions of odor level, but then it would exceed a permissible exposure limit for ozone. This conclusion was consistent with the previous data from the study by Priem (1977), which proved that a higher ozone dose of 0.2 ppm was suitable to reach a 50, and 15% reduction of ammonia level from swine barn during winter and summer ventilation conditions, respectively. Similarly, Wu et al. (1999) reported that ozone treatment was effective in removing the malodorous bacterial metabolites such as phenol, p-cresol, p-ethylphenol, and skatole. Additionally, the same ozone dose of 0.5 g/L significantly reduced the intensity of the odor from manure slurry. Besides, ozone in combination with hydrogen peroxide had no greater impact on odor removal than ozone used alone. The

temperature of the process did not affect the effectiveness of ozone treatment. It was also observed that the initial odor did not return after a 1-month storage (Wu et al., 1999). The influence of ozone treatment on the stored swine manure slurry was also the subject of Watkins et al. (1997) study. Despite obtaining a great decrease in odor, the levels of volatile fatty acids, nitrate, phosphate, as well as ammonia were not significantly reduced by ozone. Unlike these compounds, odor from indolic and phenolic metabolites was reduced to a minimal level of detection. Furthermore, ozone caused a decrease in the numbers of *E.coli* and total coliforms (Watkins et al., 1997). Riaño et al. (2014) compared the effectiveness of the oxidation processes (Fenton method, O_3, H_2O_2 and O_3/H_2O_2) in removing color and organic matter from swine manure. The results indicated that the Fenton method was more effective than ozone to reduce TCOD (Total Chemical Oxygen Demand) and color from swine manure (Riaño et al., 2014).

Alkoaik (2009) studied the effect of ozone treatment on animal manure and reported that an ozone dose of 25 mg/L of manure was suitable to remove the occurring odor - a 66% reduction was observed in the continuous operation, and a 69% reduction while using the batch operation. Additionally, they observed that in the case of an increasing ozone concentration, the odor offensiveness also increased with the presence of intermediate products, resulting from the interaction between ozone, hydrogen sulfide, and methylamine (Alkoaik, 2009). Moreover, Elenbaas-Thomas (2005) has established that the ozone dose of 0.01 ppm is a maximum safe concentration that can be used in swine facility, but does not evoke any significant impact on concentrations of sulfur compounds, including dimethyl sulfide, dimethyldisulfide, and dimethyltrisulfide, odor level, and its emission rate, as well as dust mass concentration, and bacteria reduction. It also caused an increase in ammonia level (Elenbaas-Thomas, 2005). Li et

al. (2008) evaluated effects of ozone treatment on NH_3 emissions from manure. They noticed that ozone had no impact on the NH_3 reduction, however it was found to generate high concentrations of particles, which may be very harmful for health (Li et al., 2008).

Nutritional and sensory aspects of ozone treatment

Numerous effective agents, including ozone, are used in the food processing industry to prevent microbial contamination (Whitehead, 1998). They were, however, often reported to exert negative effects on the texture, chemical content, as well as sensory attributes of food products (Tiwari and Muthukumarappan, 2012). Nutritional and sensory properties of all processed food products are affected during decontamination or sterilization techniques, and ozone treatment is no exception (Caldo et al., 2014). The impact of ozone on product quality and nutritional value has been reported by many investigators (Achen and Yousef, 2001; Barth et al., 1995; Bialka and Demirci, 2007; Fonseca and Rushing, 2006; Guzel-Seydim et al., 2004a; Perez et al., 1999; Skog and Chu, 2001; Tiwari and Muthukumarappan, 2012; Tzortzakis et al., 2007b; Zhang et al., 2005). Numerous studies showed that ozone may favorably and unfavorably affect the quality of different types of food products (Table 2).

At doses below 1 ppm (1 ppm responds to 1.96 mg/m^3), changes in the chemical composition of ozone-treated foods are considered insignificant. However, high ozone doses cause significant nutritional losses as well as deteriorate sensory properties (Akbas and Ozdemir, 2008; Guzel-Seydim et al., 2004a; Naitoh et al., 1988; Zagon et al., 1992; Zhao and Cranston, 1995). The most noticeable effect of ozone treatment on the sensory quality of fruits which has been studied is the loss of aroma. Enrichment of cold stored strawberries with ozone caused reversible losses of their flavor that were probably caused by the oxidation of volatile compounds (Nadas et al.,

2003).

Also, antioxidant capacity ascribed to the presence of vitamin C, anthocyanins, carotenoids, and polyphenols in fruits, vegetables, herbs, and spices, is significantly affected during ozone treatment due to its strong oxidizing activity. But it has been reported that plants can develop some defense mechanism when under the oxidative stress induced by ozone. Such antioxidant compounds act as scavengers of reactive oxygen species (ROS) and thereby protect cell structures from oxidative damage. The reaction between ozone and antioxidants results in the formation of ROS such as hydroxyl radical, hydrogen peroxide, and superoxide radicals inside the plant cell (Miller et al., 2013; Moldau et al., 1998). For instance, a decrease in vitamin C content was detected during ozone treatment by Alothman et al. (2010). But it was confirmed that low ozone doses ensured higher vitamin C amounts remaining in the product (Zhang et al., 2005). In contrast, degradation of phenolics in juniper berry samples was observed at ozone doses above 100 g O_3/m^3 during a 60-minute treatment, (Brodowska et al., 2015). What is more, it was revealed that ozone concentration of 50 g O_3/m^3 can destroy polyphenols after a 12-hour treatment. Additionally, the study conducted by Diao et al. (2014) confirmed that contact time with ozone influenced the phenolic content, and that it was due to the cleavage of glycosidic linkages with sugars or oxidation of polyphenols to a carbonyl group (Diao et al., 2014). Nonetheless, Dock (1999) observed no detrimental differences in the quality of apple cider treated and not treated with ozone. However, applying high ozone doses for an effective decontamination may change the sensory qualities of food products (Tiwari and Muthukumarappan, 2012). It was reported that a high ozone concentration (18 ppm) and long contact time (8 h) caused noticeable changes in natural pigments of aloe powder and bee pollen

and a significant decrease of pH in Korean red ginseng powder (Byun et al., 1997, 1998; Yook et al., 1997).

Unlike other decontamination methods (thermal processes), ozone treatment caused no damage to amino acids and did not deteriorate protein quality (Erdman, 1997). Due to the rapid reaction with unsaturated organic compounds, PUFAs may be oxidized and peroxidation increases at higher ozone doses. However, exposure to low ozone doses should not have a significant impact (Erdman, 1997).

In addition to nutritional adequacy, sensory aspects are also extremely important to the feasibility of ozone treatment of foods, particularly regarding fresh products such as fruits and vegetables, which has been reviewed by Tiwari and Muthukumarappan (2012).

Specific ozone applications in the food industry to control microorganism growth and extend the shelf life of food products

The reported data is usually characterized by significant divergence. Some studies have claimed substantial reductions in microbial contamination upon ozone treatment, whereas the opposite side postulates that ozone treatment is not effective at all. Several studies compared the effectiveness of gaseous and aqueous ozone in bacterial inactivation, reporting different results. Some investigations recognized ozonated water as significantly more effective than the gaseous state of ozone, while other researchers disagreed. Such diversity of results may be due to the different kinds of products used in each study. Therefore, ozone efficiency should be assessed concerning the product and the group of microorganisms (Miller et al., 2013).

Next to control of the growth of microorganisms, the extension of the shelf life of food products treated with ozone is also an important criterion for applying this technology in the food

industry. Equally important is the issue related to odor removal because the sensory perception, including a product's flavor, has a great impact on purchase decisions. Nevertheless, removal of pesticide residues with ozone is a significant topic in the food industry field (O'Donnell et al., 2012). It has, however, been reviewed in detail by Chan and Wu (2012), therefore we will focus on the main achievements reported in the subjects mentioned above.

Microbial spoilage elimination is one of main reasons for using ozone treatment to preserve the fresh-cut vegetables and fruits, as well as the meat, poultry, seafood, and dairy products (Abad et al., 1997; Achen and Yousef, 2001; Barth et al., 1995; Beltran et al., 2005; Bialka and Demirci, 2007; Perez et al., 1999; Tzortzakis et al., 2007b; Garcia et al., 2003; Gil et al., 2006; Fonseca and Rushing, 2006; Hildebrand et al., 2008; Jaksch et al., 2004; Zhang et al., 2005). Undeniably, in the majority of the investigated studies, ozone was effective in both bacteria and fungi reductions (Table 2).

Spices

Concerning spices (Table 2), Zhao and Cranston (1995) evaluated the impact of ozone on the volatile constituents of spices (whole black peppercorn and ground black pepper). Their findings were similar to those obtained by Perez et al. (1999), who reported a significant microbial reduction, but at higher ozone doses. Zhao and Cranston (1995) also demonstrated that the form of the spices (whole, ground) affected their aroma quality. Also treatment of whole black peppercorn or ground black pepper with ozone in the gaseous phase (6.7 mg/L) for 10 min with a flow rate of 6 L/min resulted in a decrease of microbial population by 3 - 4 log units reduction, and 3 – 6 log units reduction, respectively. In the ozone-treated ground black pepper, however, the oxidation of some volatile constituents occurred, whereas ozone had no significant effect on

the whole black peppercorn (Zhao and Cranston, 1995). It may be due to the better availability of volatile compounds in ground than in not ground plant materials, causing greater susceptibility of volatile compounds to oxidation by ozone. Spices (dried oregano) were also addressed in the study conducted by Torlak et al. (2013). Longer exposure time and higher concentrations resulted in a considerable reduction of microorganisms. The gaseous ozone (2.8 and 5.3 mg/L for 120 min) was capable to inactivate *Salmonella* by 2.8 and 3.7 log, respectively, while ozone dose of 2.8 mg/L applied for 120 min resulted in 2.7- and 1.8 log reductions in aerobic plate counts and yeast and mold counts, respectively. Besides, at the higher ozone doses (5.3 mg/L for 90 min), aerobic plate counts were reduced by more than 3.2 log units reduction. Additionally, the sensory analysis showed that ozone treatment allowed obtaining dried oregano with an acceptable taste, flavor, and appearance, as well as significantly reduced microbial counts (Torlak et al., 2013). Brodowska et al. (2015) studied the impact of ozone treatment in a dynamic bed on changes in contents of biologically-active substances of juniper berries. The spice was treated with different ozone concentrations (100.0; 130.0; and 160.0 g O_3/m^3) and contact times (30, 60, 90 min), a constant flow rate (0.1 L/min) and pressure (0.5 atm). As we have observed, during short ozone contact times (30 min), higher contents of total polyphenols were found. However, the prolonged period of treatment caused opposite results. Besides, ozone treatment was not significantly effective in bacteria and fungi reduction (Brodowska et al., 2015). Similar observations were made in another study conducted by Brodowska et al. (2014) in which cardamom seeds were treated with ozone doses of 160 to 165 g O_3/m^3 for 30 min. Ozone treatment was conducted 3 times, at 24-h intervals to ensure the least possible losses of biologically-active substances. The study revealed that after the 3rd ozone treatment the extract

from cardamom seeds had a better radical scavenging activity (IC_{50} = 24.18 ± 0.04 mg/mL) than the control sample (IC_{50} = 31.94 ± 0.05 mg/mL), as well as an improved FRAP activity (613.64 ± 49.79 mmol TE/g compared to 480.29 ± 30.91 mmol TE/g of control sample). In contrast, negative effects of ozone were noted in the case of the total polyphenol content and the total antioxidant capacity (Brodowska et al., 2014).

Noteworthy is that, next to dried vegetables, the spices are the most irradiated food products in the world (Calado et al., 2014). Comparing the effect of ozone with gamma-irradiation (2 and 4 kGy), similar reductions as in previous mentioned study (Torlak et al., 2013) of 1 and 2 log in the fungal count in hot peppers were observed (Iqbal et al., 2012).

Fruits and vegetables

Another application of ozone technology focuses on the treatment of post-harvest fruits and vegetables to extend their shelf life. Also, most of the investigated studies show that the main goal of using ozone was elimination or reduction of mold *Botrytis cinerea*, commonly known as a grey mold, from a susceptible blackberry, strawberry, grape and peach, plum, carrot, tomato, etc. (Barth et al., 1995; Hildebrand et al., 2008; Liew and Prange, 1994; Palou et al., 2002; Perez et al., 1999; Tzortzakis et al., 2007a; Tzortzakis et al., 2007b). In the study conducted by Barth et al. (1995), blackberries were exposed to ozone (0.1 and 0.3 ppm) for fungi reduction. After a 12-day storage, 20% of berries exhibited a decay of the main mold *B. cinerea*. Apart from the elimination of fungi development, the evaluation of anthocyanins, color and peroxidase (POD) activity was taken into account. The 12-day ozone storage retained anthocyanin content at the same level as the initial one. No significant differences in defects or injury of the blackberry surface were observed during ozone storage. The color of the surface was kept at a high level of

the hue angle values in berries treated with 0.1 and 0.3 ppm of ozone and stored for 5 days as well as in berries treated with 0.3 ppm of ozone during 12-day storage. The ozone-enriched blackberries had an extended shelf life and maintained their quality (Barth et al., 1995). The same aim of the research had Perez et al. (1999), who studied the effect of ozone treatment on strawberries. In contrast to the study of Barth et al. (1995), ozone was ineffective in reducing *B. cinerea*. Moreover, strawberries treated with ozone were characterized by a 3-fold decrease in vitamin C content after a 3-day storage. Besides, ozone had a negative impact on strawberry aroma, which was indicated by a 40% reduction in volatile ester content (Perez et al., 1999). Nevertheless, Kute et al. (1995) observed that strawberries stored and treated with 0.3 and 0.7 µL/L of gaseous ozone were not significantly affected – the level of ascorbic acid was unchanged. In addition, the total soluble solid content was increasing for a week during the treatment and finally has reached the higher levels than in the strawberries not treated with ozone (Kute et al., 1995).

In another investigation Liew and Prange (1994) studied the impact of ozone on carrots, and reported a 50% reduction of two postharvest pathogens - *Botrytis cinerea* and *Sclerotinia sclerotiorum* at the highest ozone dose (60 µL/L) with a flow rate of 0.5 L/min for 8 h/day. But, certain changes in carrot respiration, electrolyte leakage, as well as total color with an increasing ozone concentration were observed despite its fungal activity. Carrots not treated with ozone were characterized by more intense color – higher chroma values, and lower lightness – lower L values in CIELAB color spaces (Liew and Prange, 1994). In a later study, Sarig et al. (1996) evaluated the post-harvest decay of table grape berries after ozone treatment. Initially they observed a rapid decline in ozone concentration during the contact with organic matter, and the

final dose of ozone which reacted with the grape surface was 0.1 mg/g with a flow rate of 8 mg/min for 20 min. The study also revealed that even though the longer time of ozone exposure could be applied, the symptoms of toxicity appeared on the treated samples of grapes. However, the numbers of microbial contaminants, including bacteria, yeasts and fungi significantly decreased after a 20-min treatment with ozone. This study demonstrated also a considerable reduction of fungi *Rhizopus stolonifer* in grape berries treated with ozone, showing that ozone induced resistance to post-harvest decay expansion. Besides, the contents of phytoalexins: resveratrol and pterostilbene, produced upon ozone treatments were similar to those obtained during UV-C irradiation. Comparing the effectiveness of ozone with SO_2 treatment in relation to the control of *R. stolonifera* growth, the same level of reduction was achieved, and no observable injury or defects in the grape cluster were noticed using both methods. Due to a significant effectiveness of ozone treatment in surface sanitization of post-harvest fungal decay, as well as elicitation of stilbene phytoalexins, it can be used as a substitute for SO_2 fumigation (Sarig et al., 1996).

The effect of different sanitizers on the microbial quality of pear, baby carrots and lettuce was evaluated by Spotts and Cervantes (1992) and Singh et al. (2002). Spotts and Cervantes (1992) compared the effectiveness of pear sanitization using water treated with ozone with that treated with chlorine. The results showed similar levels of reduction in the count of *Cladosporium* spp., *Penicillium* spp. and *Altenaria* spp. after a 5-month storage using both methods. A commercial packaging test revealed, however, a lower survival rate of propagules of *Altenaria* spp. in chlorinated water than in the ozonated one (Spotts and Cervantes, 1992). Another study revealed that the inactivation of *Escherichia coli* could not be achieved in lettuce and baby carrots using

different sanitizers (water, ozone, chlorine dioxide, thyme essential oil) alone. Thus, sequential washing seems to be effective in eliminating *E. coli* (Singh et al., 2002). A gaseous ozone dose of 2.1, 5.2, and 7.6 mg/L for 5, 10, 15 min, as well as ozonated water at a dose of 5.2, 9.7, 16.5 mg/L of ozone for 1, 5, 10, 15 min were used in this study. Besides, the study included treatments with a gaseous and aqueous chlorine dioxide with doses of 0.5, 0.75, or 1.00 mg/L, and 5, 10, or 20 mg/L, respectively for 5, 10, 15 min. Additionally, sequential washing was carried out including aqueous chlorine dioxide (10 mg/L for 10 min), ozonated water (9.7 mg/L for 10 min), and thyme oil suspension (1.0 mL/L for 5 min). The 10-min washing with 9.7 or 16.5 mg/L of ozonated water, resulted in a 1.68 and 1.8 log reduction of *E. coli* population in baby carrots, respectively. Gaseous and aqueous ozone reduced *E. coli* in baby carrots by 1.11 – 2.64 logs and by 1.68 – 1.80 log units reduction, respectively, whereas ozonated water was ineffective in the case of lettuce, which may be explained by the greater ozone demands of the organic material in the medium (Restaino et al., 1995; Singh et al., 2002). In contrast to ozone, aqueous chlorine dioxide resulted in a 1.69 log reduction in baby carrots after 15 min, while a longer treatment duration was ineffective in the case of lettuce. This may result from the penetration of microorganisms through cut edges into the inaccessible sites of lettuce. Besides, it may also be caused by the fact that disinfectants in solutions cannot penetrate into the protective hydrophobic pockets, folds or minute cracks on the surface of the leafy vegetables. On the other hand, the treatment of lettuce with gaseous ClO_2 as a sanitizer caused a significant reduction of *E. coli* at the higher doses of chlorine dioxide (0.75; 1.00 mg/L) after 10 min - 1.67, and 1.91 log units reduction, respectively. At the same concentrations of chlorine dioxide, greater reductions were observed in the treated baby carrots than lettuce, which may be due to the fact that some

microorganisms present on lettuce were protected from ClO_2. Despite the effectiveness of sequential washing in inactivation *E. coli*, plant extracts, aqueous chlorine dioxide and ozonated water caused the oxidation of lettuce chlorophyll (decolorization) during an extended period of treatment with high doses of ClO_2 and O_3 (Singh et al., 2002).

Concerning stored fruits and vegetables, ozone treatment of broccoli, cucumber, apple, pear and mushroom was evaluated by Skog and Chu (2001) who concluded that ozone doses of 0.05 – 0.4 µL/L were suitable to prolong their shelf life without a significant influence on the quality of the stored foods (Table 2). At such doses, ozone was effective in reducing ethylene concentration in the atmosphere in which apples and pears were stored. The proposed ozone concentrations were enough for antimicrobial effects (Skog and Chu, 2001). Zhang et al. (2005) also verified that an aqueous ozone at a dose of 0.18 ppm inactivated the growth of the microbial population on celery during a 9-day storage, maintaining the fresh quality provided that the senescence of tissue was constantly controlled. Nonetheless, Bradford and Suslow (2000) suggested that the ozone doses higher than 0.04 – 0.4 µL/L were needed to reach fungicidal effects. Apples treated with ozone indicated no detrimental differences in internal ethylene concentrations, which demonstrated that ethylene may have been oxidized rather than reduced by ozone. The study showed that ozone had a potential use in storage of both ethylene-producing and ethylene-sensitive fruits and vegetables in the same room (Skog and Chu, 2001). The observations made by Zhuang et al. (1996) in the study in which broccoli florets were treated with ozonated water at a dose 1 ppm for 10 and 50 min, agreed with those obtained by Skog and Chu (2001), who investigated ozone effect on surface color, ascorbic acid, and total carotenoid level, as well as microbial growth in broccoli. In those assays there were no significant changes in comparison to

samples not treated with ozone, however, a reduction of ethylene production after a 22-hour treatment was observed (Zhuang et al., 1996).

Beltran et al. (2005) assessed the impact of ozone treatment on browning process in fresh-cut potatoes (Table 2). They investigated the effectiveness of different decontaminating agents such as sodium hypochlorite, sodium sulfite, peroxyacetic acid (Tsunami®), and ozone applied alone and in combination, on potatoes that were stored under vacuum- (VP) or modified-atmosphere packaging (MAP). The obtained results showed that under MAP conditions the browning could be inhibited by the use of sodium sulfite only, but the off-flavor was noticeable. Although treatments with aqueous ozone revealed no significant reduction in microbial growth, the initial color was preserved with no browning effect in MAP or VP after a 5-day storage. Under the same conditions results for ozone – Tsunami® and Tsunami® treatments showed a moderate degree of browning. However, ozone combined with Tsunami® under VP conditions after a 14-day storage resulted in maintenance of the sensory, as well as microbial quality (Beltran et al., 2005). Also, Whangchai et al. (2006) studied the use of ozone treatment in combination with some organic acids in comparison to SO_2 to control postharvest decay and pericarp browning of longan fruit. They concluded that the fruit treated with ozone (200 µL/L for 15, 30, 60, 120 min) in combination with oxalic acid or citric acid was characterized by less browning and reduction of polyphenol oxidase (PPO). Despite an SO_2 effectiveness in microbial reduction, browning inhibition, and low PPO activity, some people may have an allergy to sulfur. Thus, combining ozone with oxalic or citric acid can be a partial alternative to sulfur dioxide fumigation to control the postharvest decay and browning (Whangchai et al., 2006).

Delicacies

With regard to dried fruits, Akbas and Ozdemir (2008) studied the effect of ozone treatment to control populations of *Escherichia coli*, *Bacillus cereus* and *Bacillus cereus* spores on dried figs. A gaseous ozone of 0.1, 0.5 and 1.0 ppm was used for 360 min to reduce *Escherichia coli* and *Bacillus cereus*, whereas the higher concentrations (1.0, 5.0, 7.0 and 9.0 ppm) were used for the same period to inactivate *B. cereus* spores. At 1 ppm of ozone after 360 min, a 3.5 log reduction in *Escherichia coli* and *Bacillus cereus* was achieved, while above this dose – a 2 log reduction of *B. cereus* spores was observed. In addition, ozone had no impact on pH, color, and moisture, as well as sweetness, rancidity, flavor, and appearance. The results indicated that ozone effectively inactivated the vegetative cells (Akbas and Ozdemir, 2008). Similar observations were made in pistachios (Akbas and Ozdemir, 2006). Ozone was found to be sufficient to inactivate *Escherichia coli* and *Bacillus cereus* with increasing doses (0.1, 0.5 and 1.0 ppm) and period of treatment (up to 360 min). The properties of pistachios such as pH, free fatty acids, color, and fatty acid composition, were slightly affected by ozone, while the peroxide value was significantly changed after ozone treatment at 1 ppm for 360 min (Akbas and Ozdemir, 2006).

Meat and poultry

Concerning meat and poultry, an aqueous ozone treatment of beef carcasses was evaluated by Castillo et al. (2003), Reagan et al. (1996), and Gorman et al. (1997) who concluded that the ozone doses of 95 mg/L (Castillo et al., 2003), 0.5% (Gorman et al., 1997), and 0.3 – 2.3 ppm (Reagan et al., 1996) were suitable to prevent the growth of bacteria (aerobic plate count, *E. coli*, *Salmonella typhimurium*). Reagan et al. (1996) compared treatment with ozonated water to hydrogen peroxide, obtaining a higher reduction in aerobic plate count for ozone - 1.30 and 1.14

log, respectively. Gorman et al. (1997) also studied the effectiveness of few washing treatments such as non-washing, washing with hydrogen peroxide, washing with 35°C water or ozonated water or trimming/washing with 35°C water, washing with a commercial sanitizer, and washing with trisodium phosphate. A 6 log reduction was achieved using ozonated water, or washing with 35°C water, or trimming/washing with 35°C water after 11-16 days of treatment. The same results were obtained after 1-3, 7-11, 16-23 and 23-29 days of treatment of meat unwashed, washed with hydrogen peroxide, washed with a commercial sanitizer, and washed with trisodium phosphate, respectively. Similarly, Castillo et al. (2003) evaluated the effect of an aqueous ozone treatment and water wash on reduction of *E. coli*, *Salmonella typhimurium*. They concluded that ozone was unsuitable for preventing the growth of pathogens. Better results were achieved with a water wash alone (Castillo et al., 2003). Also, the influence of ozone treatment on the beef brisket was reported by Gorman et al. (1995). Different decontaminating agents such as 5% hydrogen peroxide, 0.5% ozonated water, 12% trisodium phosphate, 2% acetic acid, 0.3% commercial sanitizer, and water at temperatures ranging between 16 and 74 °C, as well as spray-washing sequence (spray-washing with chemical solutions and hand-trimming followed by spray-washing with plain water/spray-washing with chemical solutions) were studied. The best results were obtained for ozonated water and hydrogen peroxide preceded by washing with plain water (Gorman et al., 1995). Cárdenas et al. (2011) studied the impact of the gaseous ozone on beef quality. An ozone dose of 72 ppm ensured a decline in the population of *E. coli* (0.6 to 1.0 log numbers) in beef samples with an unchanged color of the surface after 3 or 24 h at 0 and 4 ^{o}C. Additionally, it has been observed that the temperature is an important factor of microorganism survival – a greater reduction was observed at a lower temperature (Cárdenas et

al., 2011). Results of this work agreed with the previous study which addressed the influence of ozone on bactericidal properties. For instance, Moore et al. (2000) found that ozone concentration of 4 ppm was sufficient to prevent microbial growth. Moreover, they noticed that Gram-negative bacteria were more sensitive to ozone than the Gram-positive ones, and bacteria were more sensitive than the yeast strains (Moore et al., 2000). In a recent study conducted by Lyu et al. (2016), the effect of carbon monoxide and ozone pretreatment was assessed in relation to the quality of the vacuum packaged beef meats at different volume ratios of CO and O_3: 100% CO; 2% O_3/98% CO; 5% O_3/95% CO; 10% O_3/90% CO. Beef meats were pretreated in such a way under MAP conditions for 1.5 h, and then packaged under vacuum and evaluated after a 45-day storage at 0^oC. The sensory analysis with color evaluation indicated the higher values for the pretreated samples. However, the physical analysis, including measurement of total viable counts, metmyoglobin, thiobarbituric acid reactive substances, total volatile basic nitrogen, and pH, revealed lower values after the combined pretreatment. Nonetheless, it has been suggested that the combination of O_3 and CO can help maintaining the quality of beef, and thereby after recognition of some important issues such as interactions of protein denaturation, oxidation, and lipid oxidation, it may be a promising technology for preserving meat quality in the future (Lyu et al., 2016). Jaksch et al. (2004) reported the ozone treatment of pork meat (at doses of 100 ppm, 1000 ppm, Table 2), was sufficient to reduce microbial contamination, indicating the lower dose of ozone as more effective, as well as to extend the shelf lifetime of meat (Jaksch et al., 2004). Furthermore, Jindal et al. (1995) found ozone to be suitable during a gaseous treatment of broiler drumsticks. An ozone dose range of 0.44 - 0.54 ppm significantly reduced counts of Gram-negative and Gram-positive bacteria and extended the shelf life of

drumsticks after a 14-day storage. Similarly, Muthukumar and Muthuchamy (2012) studied the inactivation of *Listeria monocytogenes* on raw chicken samples, reporting that no bacterial growth was observed at 33 mg/min of a gaseous ozone dose for 1 to 9 min. Also, the effect of combined ozone and lyophilization on the shelf life of the chicken meat was investigated. Gaseous ozone at 0.4, 0.6 and 0.72 ppm was applied for 10, 30, 60, and 120 min. An increasing time of treatment caused a significantly lower number in total aerobic mesophilic bacteria, and after an 8-month storage bacterial counts were reduced up to 6.8 log units reduction (Cantalejo et al., 2016). Similar observations were made by Stivarius et al. (2002) who concluded that a 15-min usage of ozonated water (1%, 7.2 ^{o}C) caused a greater decrease in bacterial growth compared to the 7-min treatment. The lower values of pH were observed in combined treated samples of chicken which were reported in previous studies (Stivarius et al., 2002). Yellowness and lightness were slightly increased after the combined treatment (Cantalejo et al., 2016). These findings did not agree with those obtained by Clavijo (2005) who observed the lower values of lightness in ozonated and partially dehydrated chicken. Cantalejo et al. (2016) proved that the combined use of ozone (0.6 ppm for 10 min) and lyophilization enhanced the microbial quality of meat, at the same time preserving the high sensory values and extending the shelf life of raw chicken meat up to 8 months.

Fish

Concerning seafood products (Table 2), ozone treatment was used, for instance, to eliminate the *Vibrio* from shrimps (Blogoslawski et al., 1993). A 0.07 mg/L ozone dose applied for 5 – 7 min was sufficient to inhibit *Vibrio* bacteria growth. Ozonated seawater resulted in rising the survival rate of larval shrimps and in reducing antibiotic use (Blogoslawski et al., 1993). Also,

shrimps were the subject of investigation of Chawla et al. (2007) who reported that soaking of the peeled shrimps in ozonated water was more effective than spraying with ozonated water. Moreover, the higher ozone doses and a longer period of treatment were recognized as sufficient to reduce bacterial contamination. Ozonated water did not affect the lipid oxidation in shrimps (Chawla et al., 2007). In turn, mussels were studied by Abad et al. (1997) who showed an aqueous ozone treatment to be an effective method to preserve this kind of seafood products. The results indicated that the human pathogenic enteric viruses were eliminated by a continuous flow of ozonated marine water during 96 h of immersion (Abad et al., 1997). Additionally, chilled tilapias were verified in relation to the effect of ozone treatment. A dose of 6 ppm at 0 and 5 ^{o}C extended their shelf life by 12 days and improved their quality after a 1-month storage. The ozone treatment combined with storage at 0^{o}C seemed to be a promising method of extending the storage life of fish (Nash, 2002; Gelman et al., 2005). Nonetheless, despite these positive results no studies were conducted to evaluate the possible pro-oxidant effect of ozone on fish constituents. In contrast to chlorine, ozone does not act selectively oxidizing certain enzymatic systems, but as a general oxidizing agent. Some researchers have shown that fish products treated with ozone possess better sensory properties, are free from molds and do not putrefy. Their results demonstrated that ozone concentrations between 2.5 to 3 ppm were recognized as the most advisable at 1 – 3 ^{o}C with relative humidity of 90% to prevent fat oxidation and unwanted odor production (Gonçalves, 2009; Rice et al., 1982; Tapp and Sopher, 2002).

Dairy

Another application of ozone technology focuses on the treatment of dairy products to increase their quality and prevent microbial contamination (Table 2). Sander (1985) developed

and patented an alternative method for raw milk treatment using mild ozone doses. So far, raw milk has been treated by heating which may have a negative impact on its nutritional, as well as sensory properties. The technology proposed by Sander (1985) should eliminate these drawbacks and minimize quality deterioration. A later study conducted by Rojek et al. (1995) showed a significant decrease (99%) in the psychrotrophic count in skim milk with ozone doses of 5–35 mg/L applied for 5–25 min. Similarly, ozone ensured complete inactivation of *Listeria monocytogenes* in raw and branded milk (Sheelamary and Muthukumar, 2011). Additionally, ozone treatment of raw milk was combined with a bubbling technology (Cavalcante et al., 2013a). Ozone applied at a dose of 1.5 mg/L for 15 min together with bubbling technology resulted in bacteria and fungi reduction to about 1 log, while ozone used alone was not effective to eliminate microbial contamination (Cavalcante et al., 2013a). However, it was reported that one Swedish industry plant used ozone at the first pretreatment step of fluid milk, followed by a traditional thermal process such as pasteurization. This ensured an extended shelf life of fluid milk with maintaining lipid and protein contents – no evidence of oxidation has been noticed (Varga and Szigeti, 2016). Torlak and Sert (2013) found gaseous ozone (2.8 mg/L or 5.3 mg/L for 0.5 – 2 h) effective in inactivation *Cronobacter sakazakii* in milk powder (skim and whole milk powder) – a 3 and 1.4 log reduction was observed, respectively. However, a negative but insignificant impact on fat content in whole milk was observed after a 2-hour treatment . Concluding, the gaseous ozone treatment was found to be a promising method for reducing *Cronobacter* in skim milk powder (Torlak and Sert, 2013). In the previous study conducted by Ipsen (1989) it was confirmed that ozone treatment caused lipid oxidation. Whole and skim milk

powders were compared during ozone exposure. The first one was more sensitive to ozone due to the possible reaction between ozone and milk fat (Kurtz et al., 1969).

Morandi et al. (2009) evaluated the effect of gaseous ozone on inactivation of *L. monocytogenes* in three types of Italian cheese such as Ricotta Salata di Pecora, Taleggio PDO and Gorgonzola PDO, at different stages of ripening. Ozone applied at a dose of 4 ppm for 8 min was sufficient to reduce *L. monocytogenes* counts below 10 CFU/g in Ricotta, whereas it was ineffective in Taleggio and Gorgonzola during a 3- or 6-day ripening. As it has been observed, ozone inhibits the ripening process (Morandi et al., 2009). Furthermore, ozonated water (2 mg/L for 1 – 2 min) was studied to improve the microbial quality of cheese. The results showed that although ozone reduced counts of total aerobic mesophiles, lactic acid bacteria, yeast and mold, it did not influence the survival rates of these microorganisms after a 30-day storage. Nevertheless, ozone allowed preserving the initial physicochemical properties of cheese (Cavalcante et al., 2013b). Another study recognized ozonated water as an effective disinfectant of mozzarella cheese surface. The water pretreatment with 2 mg/L of ozone resulted in an improvement of the microbiological quality and extension of the shelf life of the final product (Segat et al., 2014).

Beverages

Another application of ozone technology focuses on the treatment of juices to control their quality. Usually, ozone is applied in the gaseous form into the juice in a stirred-tank reactor or bubble columns (Silva and Brandão, 2013). Numerous microbial studies have demonstrated that the spoilage and potentially pathogenic species occurring in the fruit and vegetable juices may be reduced by 5 log numbers upon ozone treatment (Tiwari and Muthukumarappan, 2012). The

effect of ozone on the microbial and nutritional quality of several fruit juices is presented in Table 2.

Safety and legislative aspects

The principal objectives that have to be provided to ensure the food of an acceptable level of its safety as well as quality is the achievement of adequate consumer protection and trade facilitation. The priority for everyone who is involved with the food industry is keeping food supply safe in order to prevent human illness and to prevent losses during decontaminating operations (Whitehead, 1998).

Ozone treatment has been outspread in recent years for seeking 'greener' food additives. Due to that fact, many consumers express their concerns about ingredients of the consumed food, as well as each operation which is responsible for bringing food 'from farm to fork'. The consumer survey reveals an increase of consumer awareness of the food supply chain, which further leads to their perceptions of appearing food processes. This situation, in which consumers demand minimally processed food to maintain nutritional and taste properties and require a minimum of preparation, may be some kind of a paradox. Therefore, to meet such demands related to novel food processes, the developers of innovative food products are forced to seek new 'better' technologies. However, numerous research studies suggest that any new technology is accompanied by public perceptions associated with possible risks as well as benefits. Thus, effective and comprehensive information regarding the new technologies and their benefits are significant for consumer acceptance (Frewer et al., 2003; O'Donnell et al., 2012).

In accordance with the above, ozone treatment has become more and more popular, but it is still not as widespread as other commonly used technologies due to the high cost of ozone

generation. It must be produced just before use because it decomposes rapidly, and because of its toxicity, which gives consumers a negative perception of the safety which ozone can deliver. An increasing interest in ozone potential for food applications resulted in elaborating a rule by the US Food and Drug Administration (FDA) on the ozone applications (Frewer et al., 2003). In 1893 (in Oudshoorn, the Netherlands) ozone had the first large-scale application of water treatment (Langlais et al., 1991). A few years later in 1906 (in Nice, France), ozone was also used for the treatment of drinking water. Since that time, ozone has found applications in many fields, including meat preservation (1909, in Germany), shellfish purification (1936, in France), prevention of the growth of yeasts and molds on fruit (1939), eggs and cheese in storage (1942, in the USA), oxidation of iron and manganese in drinking water (1957, in Germany), oxidation of micropollutants such as phenolic compounds and several pesticides (1965, in Switzerland), as well as control of the color of water surfaces (1965, UK, Ireland) and the presence of algae (1970, in France) (Tiwari and Rice, 2012). However, prior to mid-1997, the ozone applications in the food processing or treatment were just a few, or not used commercially in the United States, and even the use of ozone was considered illegal. It was mainly caused by the regulations of the U.S. Food and Drug Administration (FDA), which has not approved ozone for use in direct contact with foods. According to the FDA, any material which comes into contact with food is defined as a food additive. Thus, any food additive has to be approved by an appropriate and specific food additive regulation. Besides, a significant problem in the approval process of a Food Additive Petition (FAP) was the lack of a specified minimum amount of ozone exposure below which it was not effective enough for its intended purpose, and the maximum amount above which it caused damage to the food (FDA, 1997; O'Donnell et al., 2012).

Such affirmations were precisely defined around 1980, when the International Bottled Water Association (IBWA) petitioned the FDA to affirm that the application of ozone to disinfect bottled water under specified conditions was Generally Recognized As Safe (GRAS) (Graham et al., 1997; Rice and Graham, 2012). The conditions included the maximum dosage of ozone which has been established as 0.4 mg/L over 4 min contact time. Besides, the water treated with ozone must meet the potable water requirements of the U.S. Environmental Protecting Agency. The IBWA's petition was approved, and in the early 1980s, a FDA regulation affirming GRAS Status for ozone use formally appeared (FDA, 1982). However, the GRAS regulation for ozone disinfection of bottled water in 1982 contained the statement [21 C.F.R. 184.1(b)(2)] that required "All other food additive applications for ozone must be the subject of appropriate Food Additive Petitions". This was not enough to gain FDA approval for other uses of ozone in direct contact with food (FDA, 1982). Finally, in 1997 an Expert Panel of Food Scientists gathered by the Electric Power Research Institute (EPRI) concluded with the following statement: "The available information supports the safety of ozone when used as a food disinfectant or sanitizer, and further, that the available information supports a GRAS classification of ozone as a disinfectant or sanitizer for foods when used at levels and by methods of application consistent with good manufacturing practices" (EPRI, 1997). This document declared ozone to possess GRAS affirmation, which has stimulated considerable interest in ozone applications for many areas in the food industry.

A final ruling, which was released in June 2001 by the FDA, approved ozone applications in the gaseous, or aqueous forms, as an antimicrobial agent in the food processing industry (FDA, 2001).

Whatever form of ozone is applied in food, its health and safety aspects also apply to the workers. Among all disinfectants, ozone is unhealthy for humans who are expected to experience exposure to it in sufficient concentrations for sufficient periods of exposure. The toxic properties of ozone may cause specific symptoms, such as drying of the throat, headache, irritation to the nose, possibly severe illness, and even death (Muthukumarappan et al., 2000; Pirani, 2010). The typical symptoms observed in humans after ozone exposure, as well as ozone doses and the periods of exposure recommended by US Occupational Safety and Health Administration (OSHA), are shown in Table 3. In the United States, the levels of ozone exposure in the workplace environment have been adopted by OSHA (Muthukumarappan et al., 2000; Pryor and Rice, 2000).

According to the above, ozone exposure must not be higher than 0.1 ppm by volume (0.2 mg/m^3 NTP) under normal working conditions for 8 h daily, or 40 h a week without adverse effects, as well as 0.3 ppm by volume (0.6 mg/m^3 NTP) for 15 minutes, and not more than 4 times per day with intervals of at least 1 h between short-time exposures (Pryor and Rice, 2000; Rakness, 2005). Therefore, all processes involving ozone should be planned with appropriate precautions to avoid ozone exposure during work (Rice, 2012).

Furthermore, the term ‘safety’ also refers to the equipment and instrumentation which is used during food production process. Ozone may interact with the equipment, as well as surfaces, therefore it is essential to take into consideration only materials which are compatible with ozone. Most materials are resistant to ozone at concentrations of 1 – 3 ppm. As it has been observed, at high concentrations ozone may cause corrosion of equipment (Singh and Singh, 1999). Plastics such as PTFE (Teflon), PVDF (Kynar), PVC (rigid and flexible), and ECTFE

(Halar) are the most frequently applied materials in the food industry and they exhibit good behavior in the presence of ozone and resistance to corrosion during ozone exposure. Accordingly, it is a very important issue to take into account all the materials that could come into contact with ozone during food processing with respect to their potential ozone resistance (Green et al., 1999; Pirani, 2010; Singh and Singh, 1999).

Last, but not least, during ozone generation from oxygen as the feed gas, the workers must pay attention that the flammability of many organic materials can increase dramatically. Besides, it was noticed that the exposure of some organic materials of construction to oxygen can cause their decomposition. Thus, if oxygen is used to produce ozone, which is usually applied in most food processing plants, some relevant precautions should be taken into consideration to avoid unwanted fires resulting from stray sparks or flames caused by oxygen leaks (Rice, 2012).

Conclusions

The food industry is still seeking for more effective applications to ensure the safer food products to consumers. All data reported in this review showed that ozone treatment can be a suitable choice for food preservation. Although there are some negative reports regarding the impact of ozone in different types of food, ozone treatment may undeniably be used as a sterilizing agent, especially for stored food products. The advantages of using ozone in the food industry such as preserving the quality of an initial product and extending the shelf life proved that statement. Moreover, the prevention of an undesirable odor emission through the effect of ozone is an additional benefit of this technology. However, one must take into consideration that even though ozone does not leave any residues due to a quick decomposition of its structure, some restrictions should be applied in the case of human exposure to it.

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Table. 1. Comparison of the main methods of ozone generation in the food processing industry (based on Gonçalves, 2009; Miller, 2005; Tapp and Rice, 2012).

Ozone generation			
Corona discharge (CD) technology		**Ultra-violet (UV) technology**	
Advantages	**Disadvantages**	**Advantages**	**Disadvantages**
High ozone doses Effective in water applications Fast removal of organic odor compounds Long use of equipment without the need for maintenance Most applicable	Higher cost than UV Relatively high capital costs	Fewer by-products than CD Output hardly affected by humidity Lower cost than CD Simple equipment construction Relatively low capital costs	Limited applications Poor yields Difficulty in ozone production by UV lamps

Table 2. Overview of the effect of ozone treatment on microbial and nutritional quality of selected food products.

Product type	Food product	Form of ozone	Target of ozone treatment	Ozone treatment conditions	Observed changes	References
Fruit	Apple	aqueous	Bacteria decay (*E. coli* O157:H7; *E. coli* O157:H7)	An aqueous ozone concentration: 24 to 25 mg/L, agitated for 1, 3, or 5 min using a magnetic stirrer at 2, 22, 45 ^{o}C	Ozone accessibility to the attached cells on all regions of the apple. More effective results after treatments were obtained when ozone was bubbled for 3 min in the wash water, and apples were dipped in the pre-ozonated water. A decline in counts of *E. coli* (3.7 and 2.6 log numbers on the apple surface) after 3 min was observed.	Achen and Yousef (2001)
		gaseous	Extend the shelf	A 107-day storage	Ozone treatment resulted in a	Skog and

			life	at 0 ^{o}C, 95 – 98% RH*, ozone concentration: 0.05 – 0.4 μL/L	significant reduction of ethylene level. No differences in the quality deterioration, firmness, total soluble solids, or titratable acidity were observed. No symptoms of an ozone injury were noticed.	Chu (2001)
	Blackberry	gaseous	Fungal decay (*Botrytis cinerea*)	A 12-day storage at 2 ^{o}C, ozone dose: 0.0, 0.1 and 0.3 ppm	During storage, none of the visual changes were observed. The color of the blackberry surface retained better after a 5-day exposure at 0.1 and 0.3 ppm of ozone, and a 12-day exposure at the same ozone dose. On the last day of treatments, the anthocyanin	Barth et al. (1995)

					content of juice was the same as initial levels. POD* was greater in control samples and those treated with 0.1 ppm of ozone, and was the lowest at a dose of 0.3 ppm after 12 days.	
	Pear	gaseous	Extend the shelf life	A 107-day storage at 0 ^{o}C, 95 – 98% RH, ozone concentration: 0.05 – 0.4 µL/L	Ozone treatment resulted in a significant reduction of ethylene level. No differences in the quality deterioration, firmness and total soluble solids were observed. No symptoms of an ozone injury were noticed.	Skog and Chu (2001)
	Strawberry	gaseous	Fungal decay (*Botrytis*	A 3-day storage at 2 ^{o}C in	Ozone was ineffective in reducing fungal decay. There	Perez et al. (1999)

			cinerea)	atmosphere containing 0.35 ppm of ozone, then fruits were stored at 20 °C to mimic retail conditions	were observed significant differences in sugar and ascorbic acid content in strawberries treated with ozone. Also, a negative impact of ozone treatment on the strawberry aroma was noticed – a 40% reduction of volatile esters.	
	Grape and peach	gaseous	Fungal decay (*Monilinia fructicola, Botrytis cinerea, Mucor piriformis* or *Penicillium*	A 4-week storage at 5 °C and 90% RH with continuous ozone exposure of 0.3 ppm	Not significant reduction of the decay of fungi was observed. After ozone treatment, an increase of the water loss during 5 weeks of storage was indicated.	Palou et al. (2002)

			expansum)			
	Plum	gaseous	Fungal decay (*Botrytis cinerea* (gray mold))	Storage at 13 °C, 'clean air' or low-level ozone enrichment: 0.1 µmol/mol	An essential decrease in the spore production and a visible lesion development was observed.	Tzortzakis et al. (2007b)
	Watermelon	aqueous	Bacteria decay (APC*)	Ozonated water: 0.4 µL/L, chlorine: 40 µL/L, and UV-C light: 1.4–13.7 kJ/m^2 at 254 nm	Chlorine and ozone were not effective in reducing bacteria growth. A negative effect of ozone and chlorine treatment on the quality (color) of watermelon was indicated.	Fonseca and Rushing (2006)
	Blueberry	aqueous, gaseous	Bacteria decay (*E. coli* O157:H7, *Salmonella*	4 different gaseous ozone treatments: continuous ozone	A gaseous ozone was effective in inactivation of *E. coli* and *S. enterica*, a 2.2, and 3.0 log reduction was observed,	Bialka and Demirci (2007)

			enterica)	exposure, pressurized ozone exposure, and 2 combined treatments after a 64-min exposure. Aqueous ozone treatments were conducted at 20 ^{o}C and 4 ^{o}C after a 64-min treatment	respectively. Aqueous ozone treatments resulted in 2.9, and 4.3 log reductions in *E. coli* and *S. enterica*, respectively. The sensory and color analysis confirmed no differences between treated and untreated blueberries.	
Vegetables	Carrot	aqueous	Fungal decay (*Botrytis cinerea* and *Sclerotinia*	A 28-day exposure with ozone - air mixture: 0	At high ozone concentration a fungistatic activity (a 50% reduction) was observed. An increase in the carrot	Liew and Prange (1994)

			sclerotiorum)	(control), 7.5, 15, 30 or 60 µl/L, a flow rate: 0.5 L/min, 8 h per day, at 2, 8 and 16 ^{o}C	respiration rate, electrolyte leakage, color differences was revealed. Carrots enriched with ozone were lighter (higher L* values) and had lower chroma values in color than control carrots.	
		gaseous	Fungal decay (*Sclerotinia sclerotiorum* and *Botrytis cinerea*)	A 6-month storage at 0.5 ℃ and >95% relative humidity with ozone exposure: 50 ±10 nl/L	A decline in a lesion size and aerial mycelium was observed. Ozone treatment caused an injury, such as blotches of brownish discolored periderm. However, it had no impact on the fresh weight loss, sprouting of carrot crowns or	Hildebrand et al. (2008)

					concentrations of glucose, fructose, sucrose or galactose.	
	Baby carrot	aqueous, gaseous	Bacteria decay (*E. coli* O157:H7)	Gaseous ozone treatment: 2.1; 5.2; and 7.6 mg/L of ozone for 5, 10, 15 min, 80% RH, 22 ^{o}C. Ozonated water: 5.2; 9.7; 16.5 mg/L of ozone for 1, 5, 10, 15 min. Sequential washing: aqueous chlorine dioxide:	After washing treatments with 9.7 or 16.5mg/L of ozonated water, *E. coli* populations, only after a 10-min treatment, were significantly reduced by 1.68 and 1.8 log numbers, respectively. Gaseous ozone reduced *E. coli* by 1.11–2.64 log numbers.	Singh et al. (2002)

				10 mg/L for 10 min; ozonated water: 9.7 mg/L for 10 min; and thyme oil suspension: 1.0 mL/L for 5min.		
	Lettuce	aqueous	Bacteria decay (*Shigella sonnei*)	Ozonated water: 1.6 and 2.2 ppm for 1 min; 2 ppm for 5 min with UV-C activation, and 5 ppm for 5 min	A 3.7 and 5.6 log reduction of *Shigella sonnei* was achieved after a 1-min treatment. A nutrient broth was affected by ozone. Treatments with ozone (5.4 ppm) caused an extension of lag-phases for injured cells recovered at 10 ^{o}C. Besides, the cells, recovered in the	Gil et al. (2006)

					nutrient broth at 10 ^{o}C, were unable to grow after washing with 16.5 ppm of ozonated water. At 2 ppm of ozone dose, *Shigella sonnei* population decreased by 0.9 and 1.4 log units with or without UV-C activation, respectively. Moreover, a 1.8 log reduction in *Shigella sonnei* counts in lettuce samples treated with 5 ppm for 5 min was observed.	
		aqueous, gaseous	Bacteria decay (*E. coli* O157:H7)	Gaseous ozone treatment: 2.1; 5.2; and 7.6 mg/L	Ozone was effective in reduction of *E. coli*. Gaseous treatments resulted in the	Singh et al. (2002)

				of ozone for 5, 10, 15 min, 80% RH, 22 ^{o}C, respectively. Ozonated water: 5.2; 9.7; 16.5 mg/L of ozone for 1, 5, 10, 15 min. Sequential washing: aqueous chlorine dioxide: 10 mg/L for 10 min; ozonated water: 9.7 mg/L for 10 min; and thyme oil	higher log reductions in comparison to aqueous washing. After washing treatments with 9.7 or 16.5mg/L of ozonated water, *E. coli* populations, only after a 10-min treatment, were significantly reduced by 1.41, and 1.42 log numbers, respectively. Gaseous ozone reduced *E. coli* by 0.79–1.79 log numbers. Decolorization of lettuce leaves during a long exposure time was observed.	

				suspension: 1.0 mL/L for 5min		
		aqueous, gaseous	Bacteria decay (mesophilic and psychrotrophic bacteria count)	Ozone treatment: 2 mM or 4.93% v/v of ozone, a flow rate 0.5 L/min for 5 min	Treatment with ozone resulted in a 4 log reduction of mesophilic bacteria count after 5 min. A decline in psychrotrophic bacteria count (4.6 log numbers after 5 min) was observed.	Kim et al. (1999)
	Iceberg lettuce	aqueous	Bacteria decay (APC)	Aqueous ozone: 2.5; 5.0; and 7.5 mg/L of ozone, chlorine: 200 mg/L, and chlorine-ozone solutions –	Treatments with ozone resulted in a 0.6 and 0.8 log reduction in the APC. It was indicated that lettuce treated with ozone had a significantly lower APC reduction than chlorine (200 mg/L). The	Garcia et al. (2003)

				different combinations, for instance, 150 mg/L of chlorine and 7.5 mg/L of ozone	highest reduction was achieved using chlorine-ozone combination (150 mg/L of chlorine and 7.5 mg/L of ozone) – a 1.4 log reduction was observed. None of the ozone-chlorine treatments were significantly better than using chlorine alone at the same concentration as the combination. Iceberg lettuce treated with chlorine, ozone, or a combination of them resulted in a prolongation of its shelf life up to 16, 20, or 25 days, respectively. No visible	

					turbidity changes in water after ozone-chlorine, or ozone treatment were noticed.	
	Celery	aqueous	Bacteria decay (Total bacteria), extend the shelf life	Ozonated water: 0.03, 0.08, and 0.18 ppm for 5 min and a 3-, 6-, 9-day storage at 4 ^{o}C	A decline in the count of total bacteria after treatment with the highest ozone dose was observed - a 1.69 log reduction. On the nineth day of the storage, a spoilage of tissue was noticed. The PPO* activity of the fresh-cut celery was affected by ozonated water and an increase in the efficacy of inhibition of PPO activity with the increasing ozone concentration in water	Zhang et al. (2005)

					was observed. A 9-day storage showed that the fresh quality can be maintained while the senescence of tissue was continuously controlled. Total sugar content was slightly affected by ozone. Vitamin C content decreased with the increasing ozone dose.	
	Fresh-cut potato strips	aqueous	Bacteria decay (LAB*, coliforms, and anaerobic bacteria)	A 14-day storage at 4 ^{o}C, a flow rate: 150 NL/h, ozone was dissolved in deionized water	Ozone was not effective in reducing total microbial populations. No browning before and after ozone treatments was observed.	Beltran et al. (2005)

	Tomato	gaseous	Extend the shelf life	Storage at 13 °C, 95% RH, ozone exposure: 0.005 – 1.0 μmol/mol	No significant effect on the fruit weight was observed. The ozone treatment did not have a negative impact on the firmness of tomato. No significant changes in fruit soluble sugar content were noticed. Enrichment with ozone resulted in an increase in ascorbic acid content. After ozone treatment a 2-3-fold increase in β-carotene, lutein and lycopene content was observed. Not statistically significant differences in phenolic content were	Tzortzakis et al. (2007a)

					observed. Sensory analysis showed a preference for fruits after low-level of ozone treatment.	
	Broccoli	gaseous	Extend the shelf life	A 7-day storage at 10 ^{o}C and a 21-day storage at 3 ^{o}C, 95 – 98% RH, ozone concentration: 0.04µL/L	The slightly changes in color were observed. Floret opening and yellowing were at a low level after ozone treatment. An increased base browning was observed, but, the brown area may be removed by trimming of the stalks. An extending of the storage of life was demonstrated.	Skog and Chu (2001)
	Cucumbers	gaseous	Extend the shelf	A 12-day storage	A reduction of microbial	Skog and

			life	at 10 ^{o}C and a 17-day storage at 3 ^{o}C, 95 – 98% RH, ozone concentration: 0.04µL/L	counts was achieved. No significant differences were visually observed. No differences in firmness were noticed. A desiccated appearance was revealed.	Chu (2001)
Mushrooms	Button mushrooms	gaseous	Extend the shelf life	A 14-day storage at 4 ^{o}C, 95 – 98% RH, ozone concentration: 0.04µL/L	The browning effect was stronger observed at higher doses of ozone. Chroma was similar to the control samples after low-ozone treatment.	Skog and Chu (2001)
Meat, poultry	Beef (carcasses)	aqueous	Bacteria decay (*E. coli* 0157:H7 and *Salmonella typhimurium*)	Ozonated water: 95 mg/L of ozone	Similar pathogen reduction after ozone treatment was achieved.	Castillo et al. (2003)
		aqueous	Bacteria decay	Ozonated water:	Ozone treatment resulted in a	Reagan et al.

			(APC)	0.3 – 2.3 ppm of ozone	reduction of contamination from carcass surface by 1.30 CFU/cm^2.	(1996)
		aqueous	Bacteria decay	Ozonated water: 0.5% of ozone	A 6 log reduction using ozonated water after 11 – 16 days of treatment was achieved.	Gorman et al. (1997)
	Beef (brisket)	aqueous	Bacteria decay (*E. coli*)	Ozonated water: 0.5% of ozone	Ozone treatment was effective in reduction of *E. coli*.	Gorman et al. (1995)
	Pork	gaseous	Bacteria decay Extend the shelf life	Ozone treatment: 100 ppm and 1000 ppm of ozone for 10 min, 42 h	Ozone treatment was effective in reduction of microbial growth, and prolongation of the shelf lifetime.	Jaksch et al. (2004)
	Poultry (chicken	aqueous	Bacteria decay	Aqueous ozone treatment: 6 – 8	Ozone was effective in reduction of microbial counts.	EPRI (1999)

	carcasses)			mg/L		
	Poultry (drumsticks)	gaseous	Bacteria decay	Ozone treatment: 0.44, or 0.54 ppm of ozone	Ozone was effective in the reduction of the levels of bacteria population. Ozone treatment extended the shelf life of drumsticks to 2 days.	Jindal et al. (1995)
Seafood	Shrimp	aqueous	Bacteria decay (*Vibrio* bacteria)	Ozonized seawater: 0.07 mg/L of ozone, for 5 – 7 min	Ozone was effective in elimination of *Vibriosis*. Through one year of ozone usage, the survival rates of larval shrimp were increased, and the antibiotic use was reduced.	Blogoslawski et al. (1993)
	Mussels	aqueous	Human pathogenic enteric viruses	Ozonated marine water with a continuous flow,	Ozone treatment may prevent the occurrence of enteric viruses before public	Abad et al. (1997)

				96 h of immersion	consumption.	
	Rainbow trout	aqueous	Bacteria decay (BGD* caused by *Flavobacterium branchiophilum*)	Ozonated water: 0.025, 0.036 – 0.039 kg ozone/kg feed fed	Ozone treatment reduced BGD mortality. However, it was ineffective in heterotrophic bacteria reduction due to a short period of ozone treatment – 35 s.	Bullock et al. (1997)
	Tilapia	aqueous	Extend the shelf life	Ozonated water: 6 ppm at 0 and 5 ^{o}C during a 30-day storage	Ozone was found to prolong the shelf life of tilapias by 12 days, and improve their quality at 0 ^{o}C.	Gelman et al. (2005)
Eggs and dairy	Eggs	gaseous	Bacteria decay	Ozone concentration: 1 – 4 ppm for 20 min	Ozone reduced a number of bacteria with increasing hatchability.	Rauch (1996)
	Skim milk	gaseous	Reduction of microbial	Ozone dose: 5–35 mg/L for 5–25	Ozone was effective in reduction of psychrotrophic	Rojek et al. (1995)

			population	min	counts by 2.4 log numbers.	
	Raw milk	gaseous	Microbial count reduction	Mild ozone treatment	Ozone treatment was recognized to be suitable to minimize the product quality deterioration.	Sander (1985)
	Raw and branded milk	gaseous	Bacteria decay (*Listeria monocytogenes*)	Ozone generation rate: 0.2 g/h for 15 min	Ozone completely eliminated *Listeria monocytogenes* after a 15-min treatment.	Sheelamary and Muthukumar (2011)
	Skim and whole milk powder	gaseous	Bacteria decay (*Cronobacter sakazakii* ATCC 51329)	Ozone treatment: 2.8 mg/L or 5.3 mg/L of ozone for 0.5–2 h	Ozone resulted in a 3 log reduction in skim milk and a 1.4 log reduction in whole milk powder after 2 h. A negative impact, but not significant, on the fat content after a 2-hour treatment in	Torlak and Sert (2013)

					whole milk was observed. Gaseous ozone treatment was found to be a promising method for the reduction of *Cronobacter* in skim milk powder.	
	Italian cheeses (Ricotta Salata di Pecora, Taleggio PDO and Gorgonzola PDO)	gaseous	Bacteria decay (*Listeria monocytogenes*)	Ozone dose: 4 ppm for 8 min at different stages of ripening	Ozone completely eliminated *L. monocytogenes* from Ricotta, whereas was unsuitable for Taleggio and Gorgonzola during a 3- or 6-day ripening. Also, ozone inhibited the ripening process.	Morandi et al. (2009)
	Cheese	aqueous	Microbial count	Ozone dose: 2	Ozone was effective in	Cavalcante et

			reduction	mg/L for 1-2 min, a 30-day storage	reduction of total aerobic mesophilic, lactic acid bacteria, yeast and mold count, but it did not influence the survival rates of these microorganisms after a 30-day storage. Ozone allowed retaining the initial physicochemical properties of cheese.	al. (2013b)
	Mozzarella cheese	aqueous	Bacteria decay (Total plate count, *Pseudomonas* spp.) Extend the shelf	Ozonated water: 2 mg/L of ozone at 15 ^{o}C, a 21-day storage	The 3.58 and 6.08 log reductions in total plate count, and *Pseudomonas* counts were noticed, respectively.	Segat et al. (2014)

			life			
	Parmesan cheese	gaseous	Fungal decay	Ozone treatment: 0.24 ppm for 40 days of storage	The 0.74, 0.93 and 2.07 log reductions in Parmesan cheese surface, shelf surface, and air of cheese ripening room were observed, respectively.	Pinto et al. (2007)
Fruit juices	Apple	gaseous	Yeast decay (*Saccharomyces cerevisiae* ATCC 9763) Extend the shelf life	Ozone exposure: 33-40 ppm for 8 min at 15 – 18 ^{o}C, and a 30-day storage at 4, 8, 12, and 16 ^{o}C	Ozone inhibited the growth of *S. cerevisiae* in all samples, which may be caused by the oxidizing action of ozone, exerting additional stress before its growth. Ozone significantly extended apple shelf life.	Patil et al. (2011)
		gaseous	Bacteria decay (*E. coli* ATCC	Ozone treatment: 0.4 ppm up to 10	Ozone treatment resulted in a 5 log reduction in *E. coli* for 5	Patil et al. (2010)

			25922 and NCTC 12900)	min at 20 ^{o}C	min. Its color and total phenolic count were significantly changed.	
		gaseous	Patulin decay	Ozone treatment: 12% w/w	The results showed that patulin can be destroyed by ozone with maintaining an initial level of sugars.	Cataldo (2008)
		gaseous	Extend the shelf life	Ozone dose: 860 ppm for 28 min at 20 ^{o}C, and a 21-day storage at 4 – 6 ^{o}C	A decrease in soluble solid and sucrose content was observed. Juices treated with ozone had a greater sedimentation.	Choi and Nielsen (2005)
		gaseous	Bacteria decay (*E. coli* O157:H7, *Salmonella*)	Ozone treatment: 9 g/h of ozone for 60 and 90 min during a 24-h	A higher reduction in *Salmonella* counts than in *E. coli* were observed.	Williams et al. (2005)

				storage at 4 ^{o}C		
	Blackberry	gaseous	Extend the shelf life	Ozone dose: 0 – 7.8% during 10 min at 20 ^{o}C	Ozone dose, as well as contact time, were recognized as important factors possessing the impact on anthocyanin content and color degradation.	Tiwari et al. (2009)
	Grape	gaseous	Extend the shelf life	Ozone dose: 0 – 7.8% during 10 min at 20 ^{o}C	Ozone caused a significant degradation of color, and a decrease in anthocyanin content. However, no changes in pH, Brix, titratable acidity were observed.	Tiwari et al. (2009)
	Orange	gaseous	Bacteria decay (*E. coli* ATCC 25922 and NCTC 12900)	Ozone treatment: 75-78 ppm up to 18 min	Ozone was effective in a 5 log reduction of *E. coli*.	Patil et al. (2009)

		gaseous	Extend the shelf life	Ozone dose: 0.6 – 10% during 10 min at 20 ^{o}C	The effect of ozone on pH, Brix, titratable acidity, cloud value, color, non-enzymatic browning, as well as ascorbic acid content was taken into consideration. No significant changes in this context were observed, but the ascorbic acid content and color were significantly affected.	Tiwari et al. (2008)
		gaseous	Yeast decay (*S. cerevisiae*) Extend the shelf life	Ozone dose: 0.9 g/h for 90 min, a 5-day storage at 10 ^{o}C	There was no significant difference in *S. cerevisiae* counts in juice before and after ozone treatment. Also, a certain decrease in ascorbic acid level was observed.	Angelino et al. (2003)

		gaseous	Bacteria decay (*E. coli* O157:H7, *Salmonella*)	Ozone dose: 9 g/h for 60 and 90 min during a 24-hour storage at 4 ^{o}C	Ozone treatment was found to be more effective in *Salmonella* than *E. coli* reduction.	Williams et al. (2005)
	Strawberry	gaseous	Extend the shelf life	Ozone dose: 1.6 – 7.8% during 10 min at 20 ^{o}C	Anthocyanin, and ascorbic acid level, as well as color changes after ozone treatment, were evaluated. Ozone significantly reduced anthocyanins (98.2 %) and ascorbic acid (85.8 %) at its highest dose.	Tiwari et al. (2009)
Vegetable juices	Tomato	gaseous	Extend the shelf life	Ozone dose: 1.6 – 7.8% during 10 min at 20 ^{o}C	The effect of ozone on pH, Brix, titratable acidity, cloud value, color, non-enzymatic browning, as well as ascorbic	Tiwari et al. (2009)

					acid content was taken into consideration. Ozone did not change pH, Brix, titratable acidity, cloud value and non-enzymatic browning. Significant changes in color and ascorbic acid content (96%) were noticed.	
Spices	Cardamom seeds	gaseous	Microbial count reduction	Ozone dose: 160 to 165 g O_3/m^3 during 30 min, a constant flow: 0.1 L/min, and pressure: 0.5 atm. Ozone treatment was conducted 3	The study revealed that extract from cardamom seeds after the 3rd treatment possessed a better radical scavenging activity as well as an improved FRAP activity than the initial sample. However, total polyphenol content and	Brodowska et al. (2014)

				times, at 24-h intervals	total antioxidant capacity were negatively affected by ozone. Ozone was not sufficient to reduce microbial counts.	
	Juniper berries	gaseous	Microbial count reduction	Ozone concentrations: 100.0; 130.0; and 160.0 g O_3/m^3, and times: 30, 60, 90 min, a constant flow: 0.1 L/min, and pressure: 0.5 atm	During short ozone contact times (30 min), the higher amounts of total polyphenol content were observed. However, a prolonged period of treatment caused reverse results. Besides, ozone treatment was not significantly effective in the reduction of bacteria and fungi.	Brodowska et al. (2015)
	Whole	gaseous	Microbial count	Ozone exposure:	Both a whole black	Zhao and

	black peppercorn and ground black pepper		reduction	6.7 mg/L for 10 min, a flow rate: 6 L/min	peppercorn and a ground black pepper were significantly affected by ozone – 3 – 4 log and 3- 6 log reductions were observed, respectively. Additionally, some volatile constituents occurring in the ground black pepper were oxidized by ozone, whereas did not affect the whole black peppercorn.	Cranston (1995)
	Oregano	gaseous	Bacteria decay (*Salmonella*, APC) Yeast and mold decay	Ozone dose: 2.8 and 5.3 mg/L during 30, 90, and 120 min	Ozone (2.8 and 5.3 mg/L for 120 min) was effective in inactivation of *Salmonella* by 2.8 and 3.7 log numbers, respectively, whereas ozone	Torlak et al. (2013)

					dose of 2.8 mg/L for 120 min resulted in 2.7 and 1.8 log reductions in aerobic plate counts, and yeast and mold counts, respectively. At higher ozone doses (5.3 mg/L for 90 min) aerobic plate counts were reduced by more than 3.2 log numbers. No changes in taste and flavor were observed.	
Delicacies	Figs	gaseous	Bacteria decay (*Bacillus cereus, Bacillus cereus* spores, *Escherichia coli*)	Ozone doses: 0.1, 0.5 and 1.0 ppm up to 360 min, and 1.0, 5.0, 7.0 and 9.0 ppm during 360 min, at	A dose of 1 ppm for 360 min was effective to inactivate *E. coli* and *B. cereus* – a 3 log reduction was observed. The higher concentrations resulted in 2 log reductions in the	Akbas and Ozdemir (2008)

				20 ^{o}C, RH 70%	number of *B. cereus* spores. In addition, ozone did not affect color, pH and moisture values. Sweetness, rancidity, flavor and appearance after ozone treatment were retained.	
	Pistachios	gaseous	Bacteria decay (*Bacillus cereus, Escherichia coli*)	Ozone treatment: 0.1, 0.5 and 1.0 ppm up to 360 min, at 20 ^{o}C, RH 70%	Ozone was found to be sufficient to reduce *E. coli* and *B. cereus* with an increasing dose and period of treatment. The properties such as pH, free fatty acids, color and fatty acid composition were slightly affected by ozone.	Akbas and Ozdemir (2006)

*POD – peroxidases; *RH – relative humidity; *L – lightness, *PPO – polyphenol oxidase, *APC - aerobic plate count, *LAB - lactic acid bacteria, *BGD - bacterial gill disease

Table 3. Ozone toxicity levels and typical symptoms depending on the ozone concentration and the length of exposure (based on Muthukumarappan et al., 2000; Pirani, 2010; Pryor and Rice, 2000; Rakness, 2005).

Length of exposure	**Ozone concentration, ppm**	**Toxicity symptoms**
detectable odor, a moment	0.01 – 0.05	nd
exceeded OSHA, 8h limit	0.1	sharp irritation to the nose, sore throat
few minutes	> 0.1	minor eye, nose and throat irritation; headache, shortness of breath
exceeded OSHA, 15 min limit	0.3	loss of vision
after a 3-6-hour exposure	0.1 – 0.5	increased loss of vision
few minutes	1.0 – 2.0	distinct irritation on the upper part of the throat, headache, chest pain, cough, dry throat
few minutes	5.0 -10.0	increased pulse, edema of lungs
few minutes	> 50.0	potentially fatal

few minutes	>1700	lethal in few minutes

nd – not detected

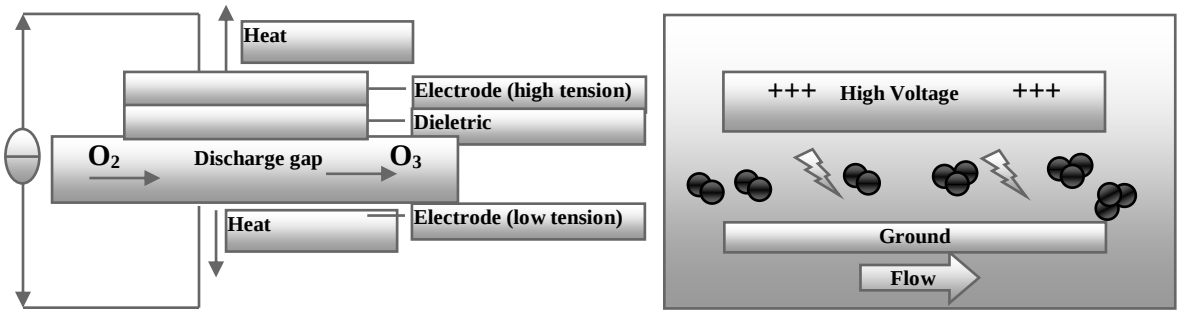

Fig. 1. Scheme of corona discharge technology (adapted from Gonçalves 2009).

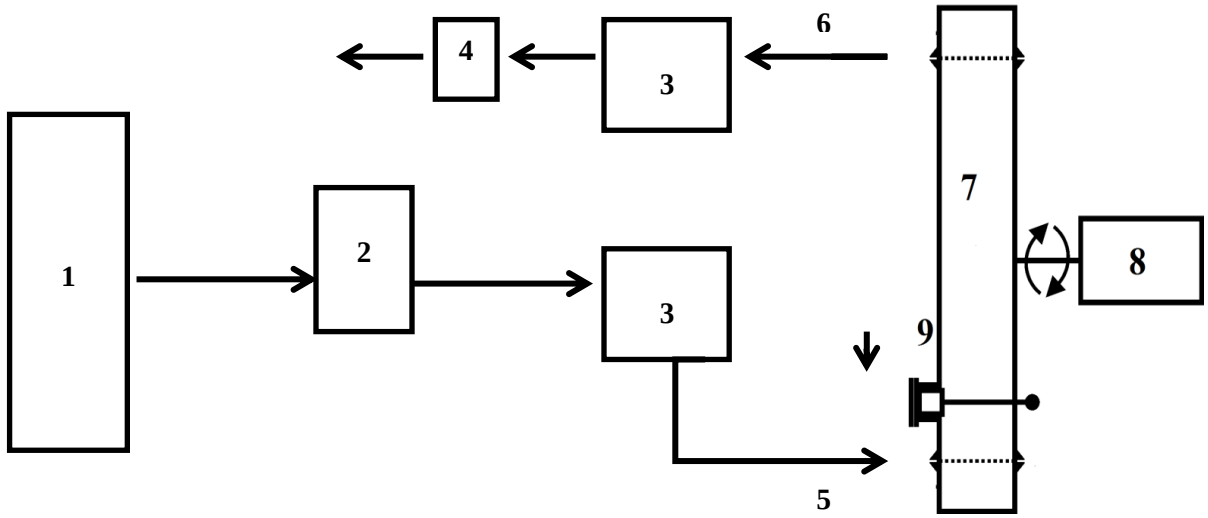

Fig. 2. Ozone treatment system for gaseous phase used for laboratory purposes (1-oxygen bottle, 2-ozone generator, 3-ozone analyzer, 4-surplus gas elimination unit, 5-inlet of ozone, 6-outlet of ozone, 7-reactor, 8-control system with jolting and rotating mechanism, 9-supply and disposal of plant material treated with ozone) (adapted from Brodowska et al., 2015).

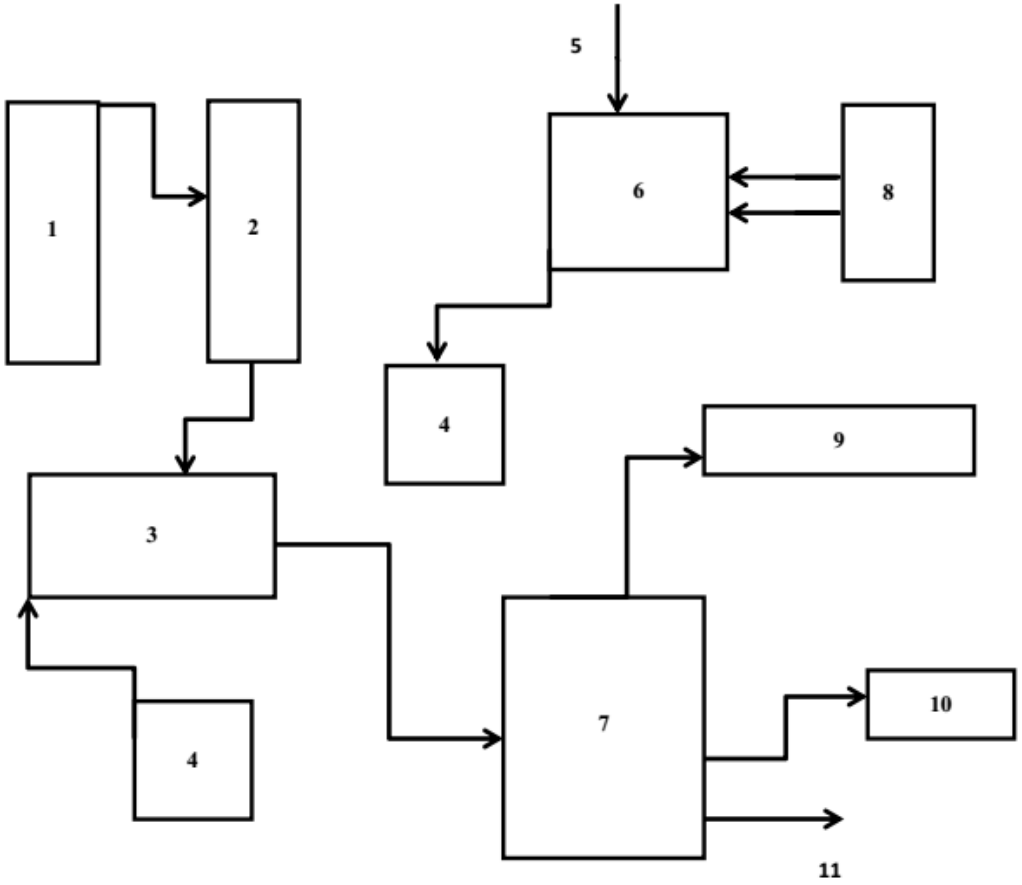

Fig. 3. Ozone treatment system for aqueous phase (1-oxygen cylinder, 2-ozone generator, 3-ozone dissolutor, 4-pump, 5-water, 6-water vessel, 7-dissolution vessel, 8-chiller, 9-ozone decomposition catalyst, 10-ozone monitor, 11-ozone-containing water) (based on Naito and Sawari, 2000).