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Exploring the potential of antioxidants from fruits and vegetables and strategies for their recovery

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ABSTRACT

A balanced and nutritious diet is the cornerstone of good health. The nutritional pyramid advises eating 5 servings of fruit and vegetables a day as a basic principle of a healthy diet, as they provide essential nutrients such as vitamins and minerals, as well as fiber and antioxidants. Consuming them in adequate quantities improves the immune system and helps prevent diseases such as diabetes, obesity, heart disease and even certain types of cancer. Among the most nutrient-dense nutrients in fruit and vegetables are antioxidants. This is the context for this review, in which an exhaustive analysis has been carried out of the bioactive compounds with antioxidant capacity available in the most widely produced fruit and vegetables in Spain, considering not only the studies that identify these types of compounds, but also those that analyze their effects on human health. Furthermore, it is worth highlighting the interest of the extraction of antioxidant compounds in by-product streams from fruit and vegetable processing in the context of the circular economy. Therefore, this review also includes examples of green technologies for the extraction of antioxidants from waste fractions. In this field, microwave and ultrasound-assisted extraction technologies have been found to be the most efficient, both in terms of extraction yields, which are similar to those of conventional technologies, and in terms of the operating conditions required for the process. However, enzyme cocktails or pulsed electric fields also show promising results.

1. Introduction

Free radicals are highly reactive compounds that are produced as a result of the metabolic activity of cells in biological systems (Ikonne, Ikpeazu, & Ugbogu, 2020). A certain level of these oxidative compounds exerts positive effects on the body's immune functions; however, oxidative stress associated with inappropriate dietary habits and lifestyles can trigger an imbalance between free radical production and the body's antioxidant defense mechanisms, evident through different biomarkers of molecular damage (Nimse & Pal, 2015; Dhalaria et al., 2020; Elkhatim, Elagib, & Hassan, 2018; Soares, Carvalho, Azevedo, & Fidalgo, 2019). This is the reason why free radicals can lead to undesirable health effects and consequently, there is a growing social concern to adhere to healthy consumption habits that include the intake of bioactive antioxidant compounds (Forni et al., 2021; Khan et al., 2021; Popa & Rusu, 2017; Tresserra-Rimbau, Lamuela-Raventos, & Moreno, 2018).

Worldwide dietary recommendations include the consumption of fruits and vegetables as a strategy for disease prevention, since in addition to their macro- and micronutrient and fiber content, they contain phytochemical compounds that stand out for their antioxidant properties (Ali et al., 2021; Kaur, Sandal, & Dhillon, 2017; Lorenzo, Colombo, Biella, Stockley, & Restani, 2021). Certain commercial brands have already developed and marketed products enriched in antioxidants from fruits and vegetables present in tomatoes, asparagus, mushrooms, apples and oranges, among many others. The recognition of the relevance of the role of antioxidants requires not only the work of nutrition and health professionals to identify the recommended intake levels and their effects on health, but also an assessment of the technological feasibility of those production processes that allow their recovery within the framework of the circular economy in the field of nutraceutical compounds (Belwal, Pandey, Bhatt, & Rawal, 2020; Sosa-Hernández, Escobedo-Avellaneda, Iqbal, & Welti-Chanes, 2018).

The aim of this review is to analyze the publications of the last 10 years focused on the study of the antioxidant content of the most produced fruits and vegetables in Spain, considering not only their identification and their effects on human health but also the routes of valorization and extraction of waste streams rich in this type of

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compounds. To select them, the most recent database of the Spanish Ministry of Agriculture, Fisheries and Food (year 2020) was used, in terms of productive area, yield and total production. Once selected, the Web of Science database was used to search for available publications and reports. The results of the study will allow establishing a global vision of the evaluation that integrates aspects related to nutrition, health, green chemistry, circular economy and sustainability in the nutraceutical sector.

2. Materials and methods

To carry out the literature review of available publications on antioxidants in fruits and vegetables, the time frame of the last 10 years, from 2011 to August 2021, was used as a search filter, as well as the logical operators AND, to include the fruit or vegetable together with the word "antioxidant", and OR, to cover both singular and plural forms (i.e., "tomato" OR "tomatoes").

In a previous screening, it was decided to use the following criteria: "Antioxidants OR antioxidants AND vegetables OR vegetables OR fruits", resulting in a total of 674,491 scientific publications and reports published in the last 10 years. In order to select the most relevant references, it was decided to make a preliminary classification based on those fruits and vegetables with the highest production levels in Spain. According to the data available from the Spanish Ministry of Agriculture, Fisheries and Food, Table 1 shows the fruits and vegetables with the highest production volume, associated with the highest industrial demand and household consumption. Based on these results, a second search was carried out focusing on the fruits and vegetables selected, which reduced the number of publications to a total of 341,706 manuscripts, i.e., 49% less. Subsequently, reports will be evaluated to identify the type, quantity and quality of antioxidants in the selected fruits and vegetables.

Considering the classification criterion based on the antioxidant properties of fruits and vegetables, Fig. 1 shows the number of publications on fruit, with orange standing out with a total of 63,496 reports, followed by tomato and apple with 48,763 and 40,052, respectively. In contrast, persimmon, mandarin and grapefruit are the fruits with the lowest volume of research articles. As for vegetables, it has been observed that the interest of researchers is lower compared to fruits, with the highest number of publications: 25,124, associated with mushrooms (Fig. 2). Other vegetables such as onion, lettuce and pumpkin also represent a notable number of publications, with 12,838, 12,395 and 10,800, respectively.

Regarding the most recurrent research areas, those related to "Chemistry", "Agriculture", "Plant Sciences" and "Food Science Technology" stand out (Fig. 3). For plant research topics (Fig. 4), although a similar trend is observed, the topic "Chemistry" is not as relevant as in the previous case, with a higher volume of publications on research topics related to "Environmental Science Ecology" and "Biochemistry Molecular Biology".

Another important aspect to evaluate is how the interest of experts and researchers in the study of the antioxidant capacity of fruits and vegetables has increased, decreased or remained constant. As shown in Figs. 5 and 6, for the fruits and vegetables selected, there has been an increase in the number of publications, especially for oranges and mushrooms for the two categories. There is one exception, cabbage, where a slight decrease is observed from 2016 to date.

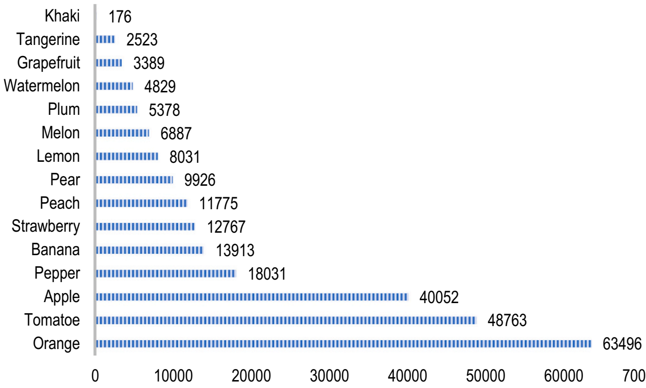

Fig. 1. Number of publications available in the WOS database from 2011 to August 2021 considering the fruit and antioxidant binomial.

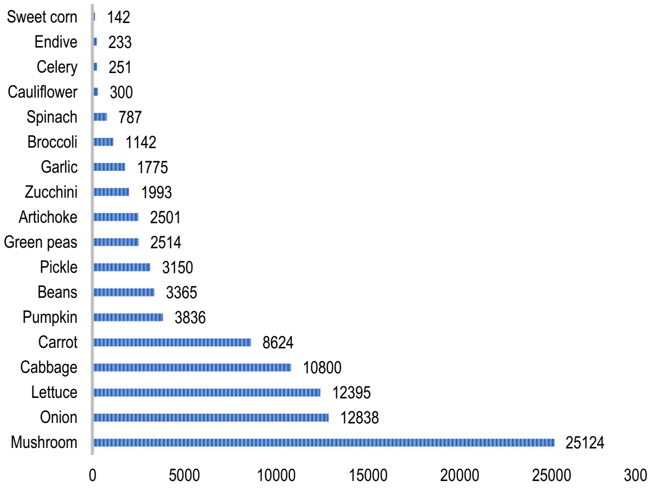

Fig. 2. Number of publications available in the WOS database from 2011 to August 2021 considering the vegetables and antioxidant binomial.

3. Natural antioxidants classification

The natural antioxidants present in fruits and vegetables fall into the category of non-enzymatic natural antioxidants. There are 4 main

Table 1
Total amount of fruits and vegetables most produced in Spain.

Product	Production (ton)	Product	Production (ton)	Product	Production (ton)
Tomatoes	4,312,895	Apple	522,618	Orange	167,112
Pepper	1,469,969	Khaki	484,315	Plum	155,834
Peach	1,309,509	Banana	420,144	Mushroom	148,495
Onion	1,299,723	Carrot	392,774	Beans	148,016
Watermelon	1,234,850	Pear	324,049	Pumpkin	143,978
Lettuce	961,938	Strawberry	272,545	Sweet corn	132,345
Pickle	794,867	Garlic	269,094	Green peas	120,165
Lemon	650,938	Tangerine	252,741	Celery	100,978
Zucchini	631,224	Cauliflower	216,389	Endive	85,687
Cantaloupe	610,978	Cabbage	209,226	Spinach	82,880
Broccoli	527,915	Artichoke	196,965	Grapefruit	33,926

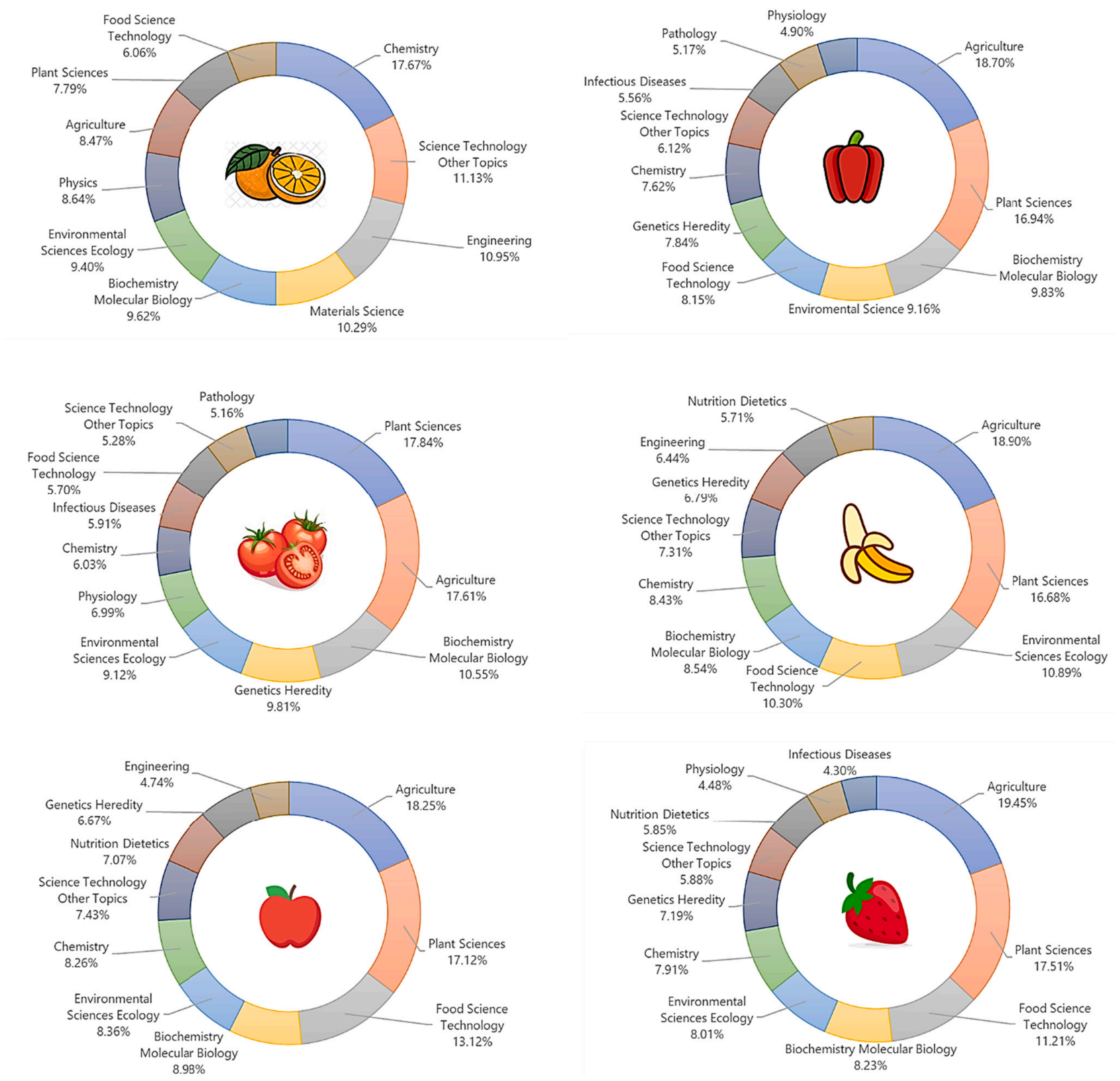

Fig. 3. Main research topics regarding fruits and antioxidants from 2011 to August 2021.

groups: vitamins, carotenoids, polyphenols and minerals. As for polyphenols, they could be classified into two main groups: flavonoids and phenolic acids.

3.1. Category I. Vitamins

Vitamins are essential elements involved in various metabolic and biological processes in the human body. Among the different vitamins required, vitamins A, C and E are the ones that stand out in fruits and vegetables. Vitamin A is considered an essential micronutrient, as it is part of several metabolic functions. Some of them are the preservation of immune defense and antioxidant functions (Noh & Mustar, 2019), which could prevent the development of diseases associated with oxidative stress and the presence of free radicals, such as certain types of cancer, cardiovascular and neurodegenerative diseases, and cell degeneration (Gelain, de Pasquali, Caregnato, Castro, & Moreira, 2012). This vitamin encompasses several nutritional organic compounds, including the alcohol form (retinol), the aldehyde-based compound (retinal), an oxidation form of retinol (retinoic acid), and the provitamin A carotenoids (most notably beta-carotene). With respect to fruits and vegetables, vitamin A can be found in the form of carotenoids, which require conversion to the retinol structure of vitamin A during the digestion process in order to be absorbed by the human body. It should be noted that, when fruits and vegetables are the source of vitamin A, such as provitamin A carotenoids, mainly half of the intake is absorbed directly into the mucosal cells (Conaway, Henning, & Lerner, 2013; Noh & Mustar, 2019). However, the most advantageous form of vitamin A in terms of antioxidant functions is in the form of retinol, as it is the most reactive structure, acting as an efficient scavenging and chain-breaking molecule in the peroxyl radical reaction (Quang Dao, Chinh Ngo, Minh Thong, & Cam Nam, 2017). On the other hand, it has been studied how efficient vitamin A could be in the most common antioxidant reaction mechanisms, which are hydrogen atom transfer (M1), electron and proton transfer (M2) and the proton loss mechanism (M3). It could also act as an antioxidant compound, in the form of retinol, by providing hydrogen atoms and/or protonated species. On the other hand the

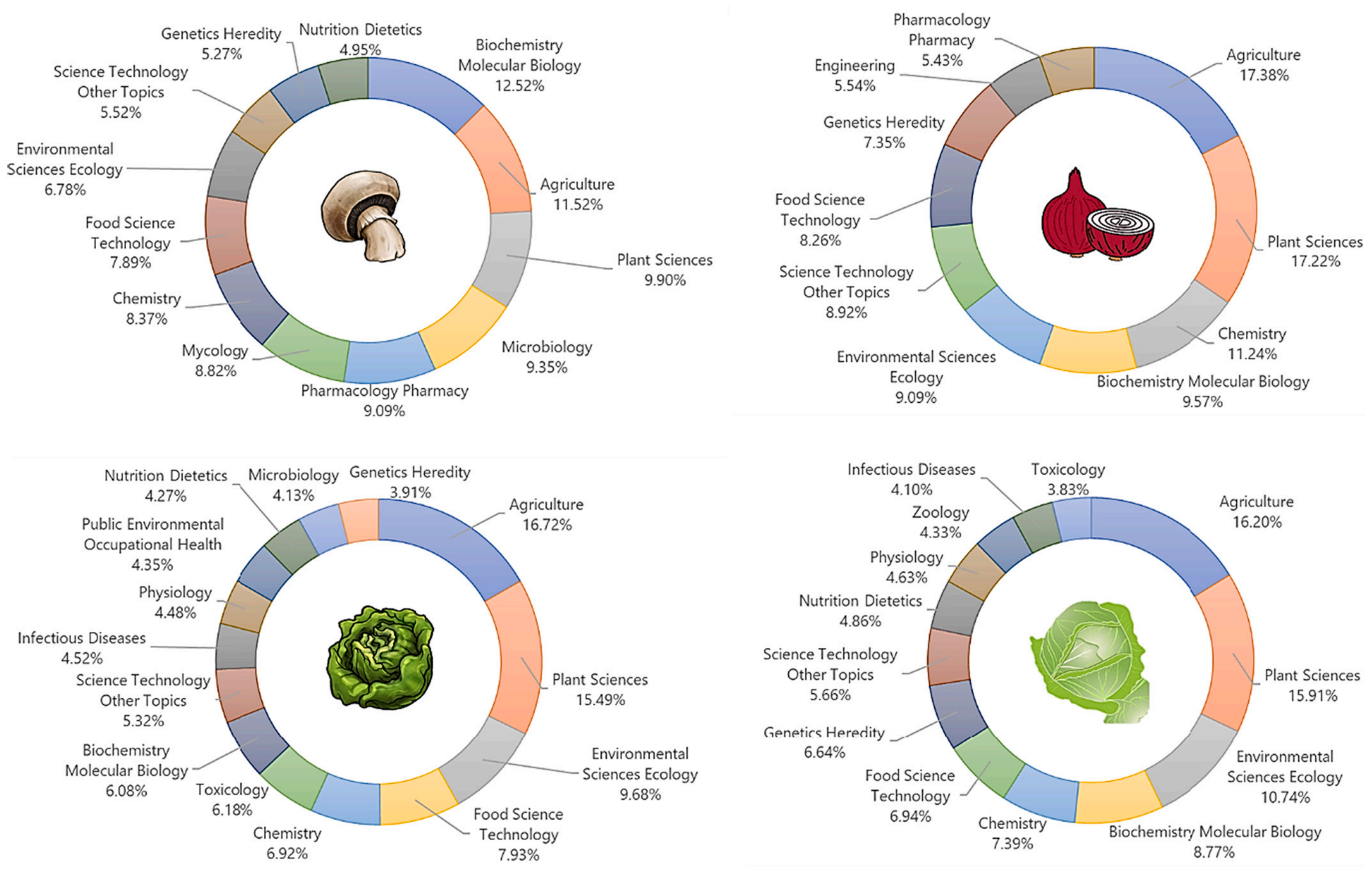

Fig. 4. Main research topics regarding vegetables and antioxidants from 2011 to August 2021.

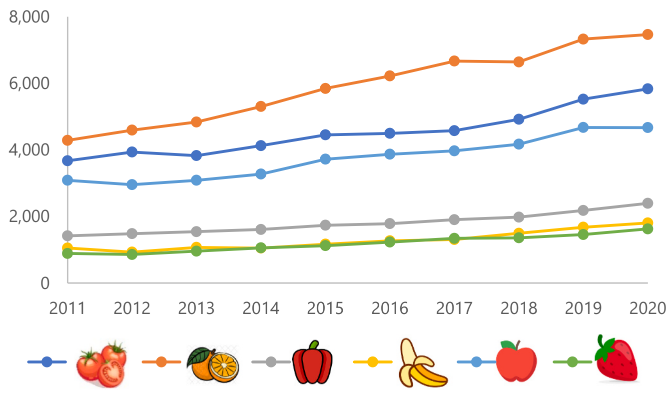

Fig. 5. Number of publications on fruits and antioxidants from 2011 to August 2021.

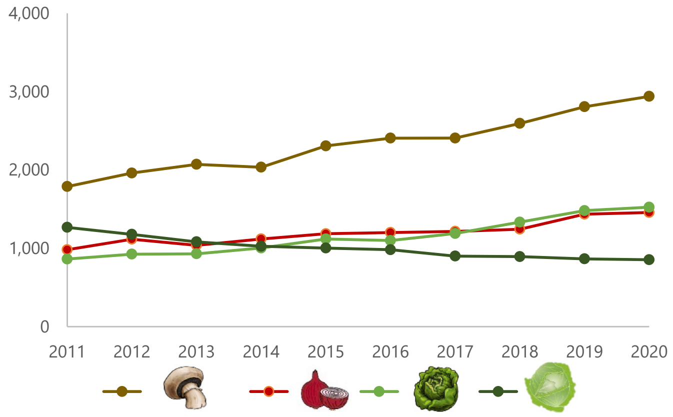

Fig. 6. Number of publications on vegetables and antioxidants from 2011 to August 2021.

potential of vitamin A to develop an addition reaction with hydroperoxyl radicals (HOO$^{\bullet}$) has also been studied (Edge & Truscott, 2018; Quang Dao et al., 2017; Rozanowska et al., 2019). Thus, the Recommended Dietary Allowance (RDA) of vitamin A amounts to 900 mg for males and 700 mg for females (Institute of Medicine, Food and Nutrition Board, 2001).

With respect to Vitamin C, also known as ascorbic acid, its antioxidant functionality is greater than that of Vitamin A. On the other hand, another important advantage of this vitamin is its ability to regenerate the antioxidant form of Vitamin E, thus allowing greater accessibility and presence of these essential compounds within the metabolic functions of the human body, leading to health benefits (Pehlivan, 2017; Pham-Huy, He, & Pham-Huy, 2008). Ascorbic acid could also act as a cofactor for different enzymes in the process of hormone and neurotransmitter biosynthesis, immune process, as well as cell regulation and regeneration. With respect to its antioxidant function, it can neutralize both ROS and nitrogen oxide species by donating a hydrogen atom, thus subsequently forming an ascorbic radical (dehydroascorbic acid), which could be reconverted to Vitamin C. Another important advantage of Vitamin C is that it can act both on the intracellular and extracellular side, as it is water soluble (Pehlivan, 2017; Popovic et al., 2015). Thus, Vitamin C is considered an antioxidant with enormous potential to prevent the development of oxidative reactions of lipids and macromolecules (Macan, Kraljević, & Raić-Malić, 2019). The RDA of Vitamin C has been defined between 65 and 90 mg/day, with 2000 mg being the upper limit that should not be exceeded to avoid side effects (Levine et al., 2001).

Similar to Vitamin A, Vitamin E entails several related compounds known as tocopherols and tocotrienols, vitamins categorized as fat-soluble. Among them, alpha-tocopherol is the one that stands out due to its ability to prevent the development of lipid oxidative reactions that lead to cell membrane damage (Lobo, Patil, Phatak, & Chandra, 2010; Popovic et al., 2015). The mechanism of action is quite easy, as it is based on the donation of a hydrogen atom from the phenolic group located in the ring of the Vitamin E structure (called the chromanol ring) (Qing, 2014). An important fact about this mechanism is that the oxidized form of alpha-tocopherol could recover its initial antioxidant active form through a reduction reaction with other antioxidants, such as vitamin C or retinol, among others (Lobo et al., 2010).

Another important benefit of Vitamin E is its participation in the immune system, through protection from bacterial infections, inhibition of the formation of mutagenic compounds and cell tissue repair, among others. This fact has led to consider Vitamin E as an essential compound to reduce the possibility of cancer cell and tumor development, given the reinforcement it provides to the immune system (Popovic et al., 2015). Regarding its RDA, its value is the lowest in comparison to vitamin A and C, with a recommendation intake value of only 15 mg/day (Traber & Manor, 2012).

Tables 2 and 3 includes the average values of Vitamins A, C, and E of the selected fruits and vegetables, respectively.

3.2. Category II. Carotenoids

Carotenoids available in fruits and vegetables can be acyclic molecules, as is the case of lycopene, or a 40-carbon chain with six-carbon rings at its ends, as is the case of α-carotene and β-carotene (Bohn et al., 2019; Perera & Yen, 2007). The biological activity and benefits on human health provided by these compounds, their interest in pharmaceutical, food and cosmetic fields is constantly growing (Kim, 2016). The ability of carotenes to reduce the presence of free radicals, to act as reactive scavengers of oxygen species and as chemical inhibitors, thus reducing oxidative stress leading to the development of processes associated with various chronic disorders, makes them powerful antioxidant substances (Fiedor & Burda, 2014; Perera & Yen, 2007). In addition to their antioxidant properties, they are considered as precursors of vitamin A, with β-carotene standing out (Chiu, Shen, Venkatakrishnan, & Wang, 2019). On the other hand, other health benefits have been identified regarding the consumption of carotene-rich foods: lower cardiovascular risk, increased immune response, reduced cancer cell proliferation, and hepatoprotective and neuroprotective functions (Chiu et al., 2019; Ikonne et al., 2020; Kopsell et al., 2010; Lakey-Beitia, Jagadeesh Kumar, Hegde, & Rao, 2019; Nagarajan, Ramanan, Raghunandan, Galanakis, & Krishnamurthy, 2017; Paiva & Russell, 2013; Sun, Tang, Chen, & Hu, 2020; Tanumihardjo, 2013). According to the recommended intake of carotenoids, it has not been standardized yet. While the German Nutrition Society reports a value of 2 mg/day as the adequate carotenoid daily intake, the value for Europeans increases to a daily value of 2.7 mg for men and 2.9 mg for women (Böhm et al., 2021).

Table 2

Vitamins content of the selected fruits.

	Vitamin A	Vitamin C	Vitamin E	*Reference*
Orange	225^{1}	53.2^{3}	0.18^{3}	(Tütem, Başkan, Ersoy, & Apak, 2020)
Tomato	614.4^{1}	36.2^{3}	0.02^{3}	(Ali et al., 2021)
Apple	5.4^{1}	4.6^{3}	0.18^{3}	(Simmonds & Howes, 2016)
Pepper	1.6^{1}	47.6^{3}	0.7^{3}	(Emmanuel-Ikpeme, Henry, & Okiri, 2014)
Banana	1.0^{2}	12.3^{3}	0.1^{3}	(USDA, 2021)
Strawberry	1.0^{1}	58.8^{3}	0.29^{3}	(Giampieri et al., 2012)
Peach	163^{1}	95^{3}	15.3^{3}	(Durst & Weaver, 2013)(
Pear	25^{1}	45^{3}	0.12^{3}	(Li, Li, Wang, & Gao, 2016)
Lemon	22^{1}	53^{3}	0.15^{3}	(El-Otmani, Ait-Oubahou, & Zacarías, 2011)
Melon	232^{2}	10.9^{3}	0.1^{3}	(USDA, 2021)
Plum	20^{2}	6.0^{3}	0.2^{3}	(BEDCA, 2021)
Watermelon	18^{2}	5.0^{3}	0.05^{3}	(BEDCA, 2021)
Grapefruit	1150^{1}	31.2^{3}	0.15^{3}	(El-Otmani et al., 2011)
Tangerine	681^{1}	26.7^{3}	0.2^{3}	(El-Otmani et al., 2011)
Kaki	163^{2}	7.0^{3}	0.73^{3}	(BEDCA, 2021)

Units: ^{1}IU ^{2}µg ^{3}mg/100 g.

Table 3

Vitamins content of the selected vegetables.

	Vitamin A	Vitamin C	Vitamin E	*Reference*
Mushroom	N/A	4.0^{3}	0.12^{3}	(BEDCA, 2021)
Onion	N/A	6.9^{3}	0.45^{3}	(BEDCA, 2021)
Lettuce	436^{2}	4.6^{3}	0.14^{3}	(USDA, 2021)
Cabbage	4.0^{2}	49^{3}	0.2^{3}	(BEDCA, 2021)
Carrot	1346^{2}	7.0^{3}	0.50^{3}	(BEDCA, 2021)
Pumpkin	34^{2}	12^{3}	0.1^{3}	(BEDCA, 2021)
Beans	53^{2}	18^{3}	0.1^{3}	(BEDCA, 2021)
Pickle	4.0^{2}	2.1^{3}	0.12^{3}	(USDA, 2021)
Green peas	53^{2}	18^{3}	0.1^{3}	(BEDCA, 2021)
Artichoke	4.0^{2}	6^{3}	traces	(BEDCA, 2021)
Zucchini	4.0^{2}	20^{3}	traces	(BEDCA, 2021)
Garlic	traces	14^{3}	0.1^{3}	(BEDCA, 2021)
Broccoli	8.0^{2}	91.3^{3}	0.15^{3}	(USDA, 2021)
Spinach	306^{2}	30.3^{3}	1.72^{3}	(USDA, 2021)
Cauliflower	traces	47^{3}	0.12^{3}	(BEDCA, 2021)
Celery	95^{2}	8^{3}	0.2^{3}	(BEDCA, 2021)
Endive	17^{2}	10^{3}	1^{3}	(BEDCA, 2021)
Sweet corn	25^{2}	6^{3}	0.4^{3}	(BEDCA, 2021)

Units: ^{1}IU ^{2}µg ^{3}mg/100 g.

Among the different carotenoids available in fruits and vegetables, the interest of β-cryptoxanthin has increased in recent years (Burri, 2015; Burri, Chang, & Neidlinger, 2011; Frihart, 2016). The main reason for its growing demand is due to its bioaccessibility and bioavailability, which is higher compared to other carotenoids (Jiao, Reuss, & Wang, 2019). Bioaccessibility is based on the release of phenolic compounds present in the food by the action of digestive enzymes, which subsequently makes these compounds available for subsequent absorption in the gastrointestinal tract. On the other hand, bioavailability refers to the fraction of these compounds that have been stored and are therefore available to be used in the different physical and metabolic functions of the human body (Blancas-Benítez, Montalvo-González, González-Aguilar, & Sáyago-Ayerdi, 2019; Cory, Passarelli, Szeto, Tamez, & Mattei, 2018; del Perales-Vázquez et al., 2020).

The bioaccessibility and bioavailability properties of phenolic compounds differ significantly among different types of fruits and vegetables. Their values will depend on the molecular form in which they are found within the food matrix. When they are completely bound to the macromolecules present in the molecular structure of the food, the accessibility to this matrix is much more complex and reduced (Cory et al., 2018). Therefore, the microstructure of fruits and vegetables considerably affects the nutritional value and the benefits that can be obtained from their intake. Those fruits and vegetables in which macromolecules have a greater tendency to be found in free forms, rather than as part of conjugates, and therefore a greater benefit for antioxidant action on human metabolism.

Although that β-cryptoxanthin has a number of beneficial functions for human health, such as reducing the risk of metabolic syndrome (Haidari, Hojhabrimanesh, Helli, Seyedian, & Ahmadi-Angali, 2018; Llopis et al., 2019), osteoporosis (Hirata et al., 2019; Regu et al., 2017), inflammatory disorders (Liua et al., 2016) and certain types of cancer (Iskandar et al., 2016; Millán et al., 2015), undoubtedly the most significant and important is its ability to be a precursor of vitamin A (Burri, 2015; Burri et al., 2011; Llopis et al., 2019). But why is that β-cryptoxanthin is considered as the best source of provitamin A? It is based on the fact that the amount of this carotenoid in fruits is higher than in vegetables, in which carotenoid compounds (i.e., α-carotene and β-carotene) are prominent, and the carotenoid-vitamin A conversion of fruit foods is more efficient than in the case of vegetables. In fruits, carotenoids fuse in the chromoplast in the form of oil droplets, which are easily dissolved and absorbed, whereas in vegetables they bind to fused chloroplasts in leaves, a much less accessible molecular structure (Burri, 2015).

Among carotenoids, lycopene provides the highest potential to scavenge oxygen free radicals, due to its high reactivity derived from its molecular structure: a linear carbon chain with thirteen double bonds leading to the ability for easy release of electrons capable of being donated to attack free radical species (Adetunji et al., 2021; Başaran, Bacanli, & Ahmet Başaran, 2017; Caseiro et al., 2020). Compared to other strong antioxidant carotenoids, its antioxidant capacity is the highest, being more than 10 times higher compared to tocopherol and twice compared to beta-carotene (Böhm, 2012; Grabowska et al., 2019; Singh & Goyal, 2008). This high reactivity also provides lycopene with important health benefits as it is able to reduce oxidative stress, ROS generation and oxidation of lipids, proteins and DNA, metabolic mechanisms that are directly related to the occurrence of cardiovascular, degenerative, carcinogenic and immunological diseases, among others (Bacanli, Basaran, & Basaran, 2017; Chen, Huang, & Chen, 2019; Przybylska, 2020).

Another beneficial feature when talking about lycopene is based on the fact that its antioxidant capacity is not adversely affected by the sterilization and cooking steps performed during food processing; on the contrary, its stability at high temperatures gives it an even greater reactivity, leading to a higher antioxidant potential (Singh & Goyal, 2008). On the other hand, another important characteristic of lycopene is that it is able to regenerate non-enzymatic antioxidants, such as vitamins. In addition, recent studies have shown that it could also protect vitamin E from inactivation (Caseiro et al., 2020). Regarding its mechanism of antioxidant action, it could develop following three different pathways: radical addition, electron transfer or the formation of an allylic hydrogen (Caseiro et al., 2020; Przybylska, 2020).

Tables 4 and 5 includes the average values of total carotenoids of the selected fruits and vegetables, respectively.

3.3. *Category III. Phenolic acids*

Phenolic acids naturally present in fruits and vegetables exhibit high antioxidant potential (Huang, Xiao, Burton-Freeman, & Edirisinghe, 2016; Süntar & Yakıncı, 2020; Thakur, Singh, & Khedkar, 2020). They are secondary metabolites that are formed by an aromatic ring, namely benzene, in which the hydrogen atoms are replaced by carboxylic acids and/or hydroxyl groups (Chandrasekara, 2019; Chen et al., 2020). One of the main advantages of phenolic acids, compared to flavonoids, is that they are in free form, which favors not only their bioavailability, but also their solubility, which translates into greater ease of absorption in the digestive tract (Chen et al., 2020). Within phenolic acids, a second classification can be made: benzoic or hydroxybenzoic acids, compounds formed by a benzene group with a carboxylic group as a substitute for a hydrogen molecule, and cinnamic acids, conjugated acid compounds derived from cinnamate, formed by an acrylic acid carrying a benzene molecule (Llopis et al., 2019).

Table 4
Total carotenoids (TC) content of selected fruits.

	TC[1]	*Reference*		TC[1]	*Reference*
Orange	0.25	(Leong et al., 2022)	**Lemon**	0.01	(Leong et al., 2022)
Tomato	1.27	(Leong et al., 2022)	**Melon**	0.10	(Leong et al., 2022)
Apple	3.50	(Ampomah-Dwamena et al., 2012)	**Plum**	0.17	(Leong et al., 2022)
Pepper	1.98	(Leong et al., 2022)	**Watermelon**	4.25	(Leong et al., 2022)
Banana	0.02	(Leong et al., 2022)	**Grapefruit**	0.09	(Leong et al., 2022)
Strawberry	31	(Žlabur et al., 2020)	**Tangerine**	0.44	(Leong et al., 2022)
Peach	15	(Brown et al., 2014)	**Kaki**	0.88	(Zhou et al., 2011)

[1] mg/100 g.

Table 5
Total carotenoids (TC) content of selected vegetables.

	TC[1]	*Reference*		TC[1]	*Reference*
Mushroom	4.12	(Wong et al., 2014)	**Artichoke**	0.50	(Kostić et al., 2021)
Onion	0.02	(Müller, 1997)	**Zucchini**	0.36	(Leong, Chen, Varjani, & Chang, 2022)
Lettuce	8.48	(Müller, 1997)	**Garlic**	0.75	(Leong et al., 2022)
Cabbage	0.43	(Müller, 1997)	**Broccoli**	0.21	(Kostić et al., 2021)
Carrot	10.29	(Müller, 1997)	**Spinach**	17.31	(Müller, 1997)
Pumpkin	0.69	(Leong et al., 2022)	**Cauliflower**	0.04	(Müller, 1997)
Beans	1.46	(Müller, 1997)(	**Celery**	0.93	(Leong et al., 2022)
Pickle	0.21	(Leong et al., 2022)	**Endive**	3.60	(Müller, 1997)
Green peas	0.62	(Leong et al., 2022)	**Sweet corn**	3.97	(Wei et al., 2020)

[1] mg/100 g.

All the beneficial properties for human health and well-being associated with phenolic acids are the result of what is known as the "biochemical scavenger effect", i.e., they favor mechanisms to reduce free radicals through the formation of stable chemical complexes (Bento-Silva et al., 2019; Cory et al., 2018). In general, they have a high efficiency and potential to reduce free hydroxyl ($OH^{\bullet}$), superoxides ($O_2^{-\bullet}$) and peroxyl ($ROO^{\bullet}$) radicals, and also act against other non-radical compounds that are also involved in oxidative stress and cell damage, such as hydrogen peroxide (H_2O_2) and hypochlorous acid ($HClO$) (Badhani, Sharma, & Kakkar, 2015). Moreover, they also have the advantage of reducing oxidative stress, phenolic acids contribute positively to the regulation of the immune response of the human body (Cory et al., 2018; Tresserra-Rimbau et al., 2018).

As for the type of phenolic acids that can be found in foods of vegetable origin, the most notable are gallic acid and syringic acid within the category of benzoic acid derivatives, and couramic acid, ferulic acid and chlorogenic acid within the group of cinnamic acid derivatives. Thus, the total phenolic content of the fruits and vegetables selected are included in Tables 6 and 7.

3.4. *Category IV. Flavonoids*

Flavonoids are secondary metabolites and phytochemicals based on a linear carbon chain with two phenolic rings synthesized in fruit and vegetables as a natural microbial infection response (Forni et al., 2021). This category of antioxidant compounds present in fruit and vegetables encompasses a subcategorization in six subgroups: flavones, flavonols, flavanones, catechins, anthocyanidins and isoflavones. This classification is based on the different degree of the phenolic ring saturation with respect to the heterocyclic ring (Dias, Pinto, & Silva, 2021).

Beyond the health benefits associated with the intake of flavonoid compounds, such as reducing cardiovascular disease and cancer, and strengthening the immune system (Adetunji et al., 2021; Dias et al., 2021; Farooqui & Farooqui, 2018; Samtiya, Aluko, Dhewa, & Moreno-Rojas, 2021), flavonoids can provide a positive influence on metabolic processes involving enzymes (Alara, Abdurahman, & Ukaegbu, 2021).

Table 6
Total phenolic content (TPC) of selected fruits.

	TPC[1]	*Reference*		TPC[1]	*Reference*
Orange	35.6	(Elkhatim et al., 2018)	**Lemon**	49.8	(Elkhatim et al., 2018)
Tomato	10.25	(Deng et al., 2013)	**Melon**	1.83	(Ganji, Singh, & Friedman, 2019)
Apple	31.9	(Krawitzky et al., 2014)	**Plum**	0.76	(Gil, Tomás-Barberán, Hess-Pierce, & Kader, 2002)
Pepper	6.43	(Deng et al., 2013)	**Watermelon**	0.17	(Neglo et al., 2021)ne
Banana	0.38	(Awele Okolie et al., 2016)	**Grapefruit**	77.3	(Elkhatim et al., 2018)
Strawberry	2.72	(W. Huang et al., 2012)	**Tangerine**	0.40	(Zhang et al., 2018)
Peach	0.41	(Gil et al., 2002)	**Kaki**	36.4	(Ercisli, Akbulut, Ozdemir, Sengul, & Orhan, 2008)

[1] mg GAE equivalent/g.

Table 7
Total phenolic content (TPC) of selected vegetables.

	TPC[1]	*Reference*		TPC[1]	*Reference*
Mushroom	4.12	(Wong et al., 2014)	**Artichoke**	30.16	(Chen, Long, Liu, Shao, & Liu, 2014)
Onion	6.20	(Deng et al., 2013)	**Zucchini**	0.10	(Bayili et al., 2011)
Lettuce	7.87	(Deng et al., 2013)	**Garlic**	0.74	(Bayili et al., 2011)
Cabbage	6.24	(Deng et al., 2013)	**Spinach**	0.18	(Bayili et al., 2011)
Carrot	0.58	(Arkoub-Djermoune et al., 2020)	**Cauliflower**	0.57	(Li et al., 2018)
Pumpkin	0.22	(Hussain et al., 2021)	**Celery**	6.80	(Deng et al., 2013)
Beans	0.92	(Madrera et al., 2021)	**Endive**	0.78	(Khalaf, El-Saadani, El-Desouky, Abdeldaiem, & Elmehy, 2018)
Pickle	0.56	(Yunusa et al., 2018)	**Sweet corn**	0.18	(Bajčan et al., 2013)
Green peas	0.85	(Hegedusová et al., 2015)			

[1] mg of GAE equivalent/g.

In a recent report that aim to correlate flavonoid intake and its contribution to reducing the risk of cardiovascular and/or cancer diseases, it was shown that consumption of at least 500 mg/day of flavonoids is particularly beneficial for alcohol and tobacco consumers (Bondonno et al., 2019).

Three different pathways have been identified in the mechanism of antioxidant action of flavonoids: (1) using their high reactivity for the removal of reactive oxidative species before they can lead to negative effects on other molecules, such as proteins, (2) inciting endogenous mechanisms using gene expression, thus achieving benefits in the response to reactive oxidative species exposure, or (3) inhibiting the formation of reactive oxygen or nitrogen species (ROS and RNS, respectively), by chelating trace elements and/or oxidative enzymatic activities (Dhalaria et al., 2020; Dias et al., 2021; Khan et al., 2021; Lobo et al., 2010). Furthermore, it should be considered that the bioavailability of flavonoids to provide full health benefits to the human body is limited, due to low absorption and early transformation into conjugated derivatives that could not have metabolic and biological activity analogous to the primary ones (Farooqui & Farooqui, 2018).

In the case of flavonols, these compounds are characterized by the presence of a ketone group on their molecular structure, with quercetin being the one that stands out (Panche, Diwan, & Chandra, 2016). Their antioxidant benefit is the result of the presence of free hydroxyl groups on their molecular structure (Dhalaria et al., 2020). It should be noted that quercetin cannot be metabolized by the human body, so dietary intakes such as fruits and vegetables can supply enough quercetin for the human body (David, Arulmoli, & Parasuraman, 2016).

Flavones are found in fruits and vegetables in the form of aglycones or glycosides. They are characterized by the presence of a saturated bond between positions C2-C3 and, the main difference with flavanols is based on the fact that a hydroxyl group is lacking at position 3 (Prasain, Barnes, & Wyss, 2018). In addition to reducing the presence of ROS and RNS, their ability to act as biomarkers of cardiovascular diseases, such as cardiac pathologies and stroke, has been reported (Hostetler, Ralston, & Schwartz, 2017).

The main difference between flavones and flavanones, another category of flavonoids, is based on the absence of the double bond at the C2 position, which results in a chiral carbon that provides flavones with important bioactive properties (Duodu & Awika, 2019). This fact leads to a higher interaction with biological receptors and a higher bioavailability compared to flavanols, as they have a better absorption capacity and a lower degradation by microbiota (Nazzaro et al., 2020; Tomás-Barberán, Gil-Izquierdo, & Moreno, 2009). Regarding their main source, it has been reported that citrus fruits are where flavanones are found in higher amounts (Awika, 2017).

As for catechins, the main source is green tea, where they are found in high concentrations, but a significant amount is also available in apples, grapes and blackberries (Arts, van de Putte, & Hollman, 2000; Evatt & Griffiths, 2013). The main problem associated with catechins is due to the fact that they are unstable and can be easily degraded, thus reducing their health benefits (Albuquerque et al., 2017; Mbaveng, Zhao, & Kuete, 2014).

Anthocyanins are phytochemicals and natural pigments found in fruits and vegetables, being responsible for purple, red and blue colors. The highest content of this category of flavonoids is present in berries, grapes and tropical fruits (Khoo, Azlan, Tang, & Lim, 2017), and they are a flavonol-derived compound. One of the functional characteristics for which they stand out is their antioxidant capacity, which can be developed through two mechanisms: donation of a hydrogen atom or an electron, both reducing reactive oxidized species into stable compounds (Tena, Martín, & Asuero, 2020). The main drawback that could be identified is based on its low bioavailability and rapid absorption, which prevents taking advantage of its beneficial properties for human health (Martín, Kuskoski, Navas, & Asuero, 2017).

Finally, isoflavones could be considered as the category of flavonoids with the lowest content in fruits and vegetables. Their main source is legumes, especially soybeans (Das, Goud, & Das, 2019). In plants, they are commonly found as inactive glycosides, which will be activated by the intestinal mucosa and bacteria through a hydrolyzing mechanism, transforming them into aglycones, which can be easily absorbed into systemic circulation directly or after subsequent metabolism in the bowel by intestinal bacteria (Popa & Rusu, 2017). Regarding their properties, isoflavones are considered as strong antioxidant species, as they are able to neutralize reactive radicals, prevent peroxidation mechanisms of lipid molecules and reduce the development of chain reactions.

Tables 8 and 9 includes the average values of total flavonoid content of the selected fruits and vegetables, respectively.

Table 8
Total flavonoid content (TFC) of selected fruits.

	TFC[1]	*Reference*		TFC[1]	*Reference*
Orange	335	(Olyad et al., 2020)(	**Lemon**	888	(Olyad, Atomsa, Chimdessa, & Gonfa, 2020)
Tomato	107.8	(Hernández-Fuentes et al., 2017)	**Melon**	15.1	(Saeed et al., 2019)
Apple	37.8	(Pandey et al., 2020)	**Plum**	37.6	(Lin & Tang, 2007)
Pepper	10.4	(Lin & Tang, 2007)	**Watermelon**	73.1	(Saeed et al., 2019)
Banana	46.3	(Saeed et al., 2019)	**Grapefruit**	10.6	(Liu et al., 2018)
Strawberry	14.6	(Lin & Tang, 2007)	**Tangerine**	420	(Olyad et al., 2020)
Peach	17.6	(Mihaylova et al., 2021)	**Kaki**	1.90	(Denev & Yordanov, 2013)

[1] mg QE equivalent/100 g.

Table 9
Total flavonoid content (TFC) of selected vegetables.

	TFC[1]	*Reference*		TFC[1]	*Reference*
Mushroom	7.8	(Azieana et al., 2017)	**Artichoke**		
Onion	30.6	(Lin & Tang, 2007)	**Zucchini**		
Lettuce	12.1	(Gan et al., 2016)	**Garlic**	12.8	(Saeed et al., 2019)
Cabbage	51.3	(Liang et al., 2019)	**Spinach**	133	(Lin & Tang, 2007)
Carrot	15.7	(Arkoub-Djermoune et al., 2020)	**Cauliflower**	6.3	(Saeed et al., 2019)
Pumpkin	77.1	(Hussain et al., 2021)	**Celery**	77	(Jung et al., 2011)
Beans	12.6	(Saeed et al., 2019)	**Endive**	N/A	–
Pickle	6.3	(Saeed et al., 2019)	**Sweet corn**	46	(Nawaz, Muzaffar, Aslam, & Ahmad, 2018)
Green peas	45.8	(Vanessa et al., 2017)			

[1] mg QE equivalent/100 g.

3.5. *Category V. Minerals*

The beneficial effect of minerals in fruits and vegetables is attributed to their potential to preserve water balance in cell membranes (through maintenance of electroneutrality), acting as cofactors for enzymes (Fellows, 2017). The minerals found in the highest proportions are selenium, magnesium, selenium, zinc and copper.

Selenium is present in antioxidant enzymes such as glutathione peroxidase, responsible for catalyzing the reduction reactions of hydrogen peroxide and peroxide radicals (Fanucchi, 2014), iodothyronines, which are able to catalyze the elimination of iodide and thyroxine (Germain, Galton, & Hernandez, 2009) and thioredoxin reductase, enzymes able to maintain thioredoxins in reduced form (Turanov, Hatfield, & Gladyshev, 2010). On the other hand, selenium is also capable of forming part of selenoprotein compounds with antioxidant properties (Tinggi, 2008). Several health benefits of selenium has been reported, including cellular and molecular protection, hormone biosynthesis, and prevention of diseases such as atherosclerosis, certain types of cancer, and cardiovascular and coronary risks (Barciela, Herrero, García-Martín, & Peña, 2008; Prashanth, Kattapagari, Chitturi, Baddam, & Prasad, 2015; VA & EN, 2004).

With respect to magnesium, its relevance has been described based on its participation in a multitude of enzymatic reactions (Szenthmihályi, Szilágyi, Balla, Ujhelyi, & Blázovics, 2014). In fact, manganese superoxide dismutase is the main enzyme with antioxidant functions, essential to prevent oxidative stress in mitochondria, where ATP is synthesized, producing at the same time a superoxide radical (Erikson & Aschner, 2019). On the other hand, On the other hand, it also participates as a cofactor in the ATP metabolic process (Castellanos-Gutiérrez, Sánchez-Pimienta, Carriquiry, da Costa, & Ariza, 2018).

Zinc is considered as an essential trace element and a cofactor for more than 2000 transcriptional factors and for more than 300 enzymes (Marreiro et al., 2017). It participates in different antioxidant processes, protecting biomolecules from oxidation, fostering the activation of enzymes and reducing oxidation reaction activities of nitric acid synthase, lipid peroxidation products and NADPH oxidase (Prasad, 2014). On the other hand, Zn is also able to inhibit reactive nitrogen and oxygen species, such as hydrogen peroxide, hydroxyl radicals, superoxide anions and peroxynitrites, and could also act directly as an antioxidant, when used in thiol groups (Olechnowicz, Tinkov, Skalny, & Suliburska, 2017). In addition, it should also be noted that both excess and deficiency of Zn could lead to oxidative stress, so maintaining an adequate level of Zn is essential to ensure human health (Jarosz, Olbert, Wyszogrodzka, Młyniec, & Librowski, 2017; Lee, 2018).

Finally, copper is also considered as an essential trace element, as its deficiency has been shown to foster cellular oxidative damage. Its presence is necessary because it can act as a cofactor for different oxidation processes that convert ROS into water (Nimse & Pal, 2015). Cu is also required for several biological and metabolic processes related not only to antioxidant defense, but also to immune activities, molecule synthesis, enzyme activation, and iron metabolism (Bost et al., 2016).

Regarding the Recommended Dietary Allowance (RDA) for the aforementioned minerals, their values depend on the countries' scientific agencies and authorities. But, according to the World Health Organization (WHO) the dairy intake values amounts to 25–34 μg for selenium (Vinceti et al., 2018), around 400 mg for magnesium (Agostoni et al., 2015), 9.9 mg for zinc (Caulfield & Black, 2004) and 11 μg/kg of body weight for the recommended daily intake of copper (Bresson et al.,

Table 10
Mineral content of selected fruits.

	Copper	Manganese	Selenium	Zinc	*Reference*
Orange	0.11[1]	0.08[1]	1.80[1]	0.21[1]	(Czech et al., 2020)
Tomato	0.03[1]	0.09[1]	2.50[2]	0.08[1]	(USDA, 2021)
Apple	0.03[1]	0.04[1]	N/D	0.04[1]	(Florkowski, Banks, Shewfelt, & Prussia, 2021)
Pepper	0.83[1]	2.74[1]	N/D	3.01[1]	(Bernardo, Martinez, Alvarez, Fernandez, & Lopez, 2008)
Banana	0.10[1]	0.26[1]	2.50[2]	0.16[1]	(USDA, 2021)
Strawberry	0.05[1]	0.39[1]	Traces	0.14[1]	(Giampieri et al., 2012)
Peach	0.08[1]	0.03[1]	2.10[2]	0.23[1]	(USDA, 2021)
Pear	0.07[1]	0.03[1]	0.20[2]	0.07[1]	(USDA, 2021)
Lemon	0.04[1]	0.05[1]	2.77[1]	0.22[1]	(Czech et al., 2020)
Melon	0.08[1]	0.05[1]	1.70[2]	0.44[1]	(USDA, 2021)
Plum	0.06[1]	0.05[1]	0.50[2]	0.10[1]	(Florkowski et al., 2021)
Watermelon	0.04[1]	0.03[1]	0.40[1]	0.1[1]	(Florkowski et al., 2021)
Grapefruit	0.06[1]	0.06[1]	1.48[1]	0.25[1]	(Czech et al., 2020)
Tangerine	0.04[1]	0.07[1]	2.58[1]	0.26[1]	(Czech et al., 2020)
Kaki	N/D	N/D	0.60[2]	traces	(BEDCA, 2021)

Units: [1]mg [2]μg/100 g.

2015).

Tables 10 and 11 includes the average values of minerals of the selected fruits and vegetables, respectively.

4. Strategies for the valorization of food waste as resources for the recovery of antioxidants

Agricultural residues encompass all residues generated as a result of agricultural exploitation, transformation and commercialization processes. A number of stages can be distinguished in which these residues are generated: pre-harvest, harvest, sorting, storage, transport, traders, processing, packaging, distribution, wholesale, retail, consumption and export. According to FAO, approximately 10–20% of waste is generated at the agricultural and post-harvest stages, while this value rises to 15–20% during the fruit and vegetable processing stages (FAO, 2019). Its production values amount to 0.5 billion tons per year, which represents practically 50% of all organic waste generated (García & Raghavan, 2021). This extremely high amount of waste generated not only poses a problem in its management and it is estimated that, approximately, the waste associated with agro-industrial processes amounts to 3.3 gigatons of CO_2 equivalent, which corresponds to a value of 7% of global GHG emissions (FAO, 2019).

On the other hand, it is also important to take into account the type of fruit or vegetable being processed and the fraction of recoverable waste. For example, citrus fruits are mostly used for the production of juices, while the pulp, seeds and peels are managed as waste, representing at least 50% by weight of the fruit (Bampidis & Robinson, 2006). This is why the valorization of these streams offers a more efficient use scenario, since they are a good source of sugars, minerals, vitamins, oils, polyphenols, and also antioxidant compounds (Boukroufa, Boutekedjiret, Petigny, Rakotomanomana, & Chemat, 2015; Soares et al., 2019).

Another example of a by-product of the food industry is apple pomace, a waste of the apple juice production process that represents 20–30% by weight of the weight of this fruit (Yates, Erdman, Shao, Dolan, & Griffiths, 2017). Its most common use is animal feed or compost, although an efficient utilization of this residue could turn it into a high-value resource, given its high composition in polyphenols, phenolic acids, flavonoids, among others (Esparza, Jiménez-Moreno, Bimbela, Ancín-Azpilicueta, & Gandía, 2020; Yates et al., 2017). In the case of tomatoes, the percentage of residue is not as high compared to apples or citrus, amounting to approximately 4%, including peels and seeds (Del Valle, Cámara, & Torija, 2006). However, since it is not used as animal feed, the option of its valorization to obtain high value-added products is of interest, given its interesting composition in antioxidant compounds such as phenolic compounds and flavonoids.

Table 11
Mineral content of selected vegetables.

	Copper	Manganese	Selenium	Zinc	*Reference*
Mushroom	0.11^1	0.09^1	traces	0.68^1	(USDA, 2021)
Onion	0.05^1	0.10^1	traces	0.12^1	(USDA, 2021)
Lettuce	0.05^1	0.13^1	traces	0.25^1	(USDA, 2021)
Cabbage	N/D	0.21^1	N/D	0.26^1	(Anunciação, Leao, de Jesus, & Ferreira, 2011)
Carrot	0.08^1	0.14^1	N/D	0.28^1	(USDA, 2021)
Pumpkin	0.05^1	0.05^1	N/D	0.26^1	(Khatib & Muhieddine, 2019)
Beans	0.04^1	0.18^1	N/D	0.19^1	(USDA, 2021)
Pickle	0.03^1	0.06^1	N/D	0.11^1	(USDA, 2021)
Green peas			1.90^2	0.80^1	(BEDCA, 2021)
Artichoke		0.12^1	0.20^2	0.05^1	(El Sohaimy, 2014)
Zucchini	0.05^1	0.18^1	0.20^2	0.30^1	(USDA, 2021)
Garlic			9.80^2		(USDA, 2021)
Broccoli	0.06^1	0.20^1	1.60^2	0.42^1	(USDA, 2021)
Spinach	0.08^1	0.43^1	2.50^2	0.42^1	(USDA, 2021)
Cauliflower	–	–	0.40^2	0.60^1	(BEDCA, 2021)
Celery	–	–	0.40^2	0.10^1	(BEDCA, 2021)
Endive	–	–	2.80^2	0.30^1	(BEDCA, 2021)
Sweet corn	–	–	0.80^2	0.30^1	(BEDCA, 2021)

Units: ^{1}mg ^{2}μg/100 g.

Regarding vegetables, their leaves and roots, considered as non-edible wastes, can be considered as high value inputs for the extraction of polyphenols, such is the case of broccoli, carrots, cauliflowers, cabbages and celery, which present a phenolic compound content of between 32.71 and 267 mg GAE/100 g of fresh weight (Sepúlveda, Contreras, Cerro, & Quintulén, 2021).

Therefore, the extraction of bioactive compounds requires the use of different extraction techniques, which will depend on the process yields, the type of input and the properties required for the final compounds. At first, a distinction can be made between conventional and innovative extraction techniques, the latter being called green technologies, since they entail reduced consumption of organic solvents, process times, chemicals and energy (García & Raghavan, 2021). The effectiveness of these new extraction techniques will depend on a number of operational parameters, solute-solvent ratio, pressure, time and temperature, and properties of the substrates, such as their chemical and molecular structure (Saini, Panesar, & Bera, 2019).

4.1. *Valorization of tomato waste*

Among different extraction alternatives for phenolics present in tomato by-products, the efficiency and feasibility of conventional organic solvent extraction and microwave-assisted extraction (MAE) have been compared (Li et al., 2012), and it has been concluded that MAE is a more efficient technology, as it implies a 6–9% increase in antioxidant potential values. Another procedure that could be used is ultrasound-assisted extraction (UAE), which attains higher purity and yield of the extracted compound, and a reduced need for post-treatment of waste streams. In fact, a recent study has used several antioxidant assays to demonstrate the efficiency of UAE, an increase of 4%, 2%, 8% and 13% (compared to conventional solvent solid extraction) has been achieved for TPC, TFC, ABTS and ORAC values, respectively.

The enzymatic cocktail produced by *Aspergilllus* strains have been applied for lycopene recovery (Lavecchia & Zuorro, 2008) and compared to the use of a hexane:acetone:ethanol mixture (50:25:25 *v*/v). This procedure leads to the extraction of 440 mg of lycopene/100 g of dried tomato, which represents a considerable rise in recovery percentages, from 3 to 30% for the conventional system to 77–98% for the process that considers the use of enzymes.

High Hydrostatic Pressure-Assisted Extraction (HHPE) is also considered an efficient method for the extraction of antioxidant compounds. A recent study has demonstrated the efficiency of using HHPE in the recovery of polyphenols from tomato peel waste, resulting in 100 mg phenolics/kg of tomato waste.

Finally, Pulse Electric Field (PEF) methodology has been shown to be an effective pre-treatment method to increase the yield in the extraction of carotenoid compounds. In fact, the use of a moderate intensity and an energy input of 5 kJ/kg is sufficient to achieve promising results. Right after the PEF pretreatment, a solvent extraction step is developed, using acetone or ethyl lactate, leading to obtain a lycopene extract with 18% antioxidant potential.

4.2. *Recovery of antioxidants from orange peels*

In the case of orange peel waste valorization for the recovery of antioxidant compounds, different methods have been compared: Conventional Solid Extraction (CSE), UAE, Super Critical Extraction (SC-CO_2) and MAE (Boudhrioua, 2016). The results reported in terms of extraction yields and antioxidant recovery showed that both MAE and UAE lead to higher antioxidant extraction yields, 41% and 31% higher compared to those of CSE, respectively. In contrast, however, the use of SC-CO_2 does not imply superior extraction yields, with a decrease of

16%. When ranking the different options, the selection of UAE technology is preferable due to its lower cost and higher simplicity compared to the MAE technology (Montenegro-Landívar et al., 2021).

In the case of D-limonene extraction from citrus residues, two alternatives have been considered: solvent-free microware extraction technology (SFME) or steam explosion (SD), reaching similar extraction yields although the times required for each process are significantly different: 240 min for SD and only 22 min for SFME (Boukroufa, Boutekedjiret, & Chemat, 2017). Once extracted, D-limonene was used as solvent for the extractive step of antioxidant compounds, considering two options, UAE or CE (Conventional Extraction). When evaluating D-limonene as an extraction solvent, promising results were obtained, as analogous results were appreciable when compared to conventional n-hexane. Moreover, the UAE methodology leads to a 40% higher value in carotenoid recovery. Furthermore, it is also noteworthy that a recent study has shown that the valorization of citrus waste for D-limonene extraction in an interesting biorefinery context represents lower environmental burdens in the overall process (Santiago, Moreira, Feijoo, & González-García, 2020).

Other products associated with the valorization of orange processing residues are polyphenols, pectin and essential oils (Boukroufa et al., 2015). The procedure is based on the development of three main stages: a microwave hydro diffusion and gravity (MHG), where the essential oils are recovered, followed by the UAE and MAE stages for the recovery of polyphenols and pectin. Compared to conventional extraction, the use of UAE results in an increase of 3.46 units in the amount of polyphenols and an increase of 1.17 units of pectin. Thus, this study demonstrates that the use of green technologies offers several advantage for the extraction of antioxidant compounds, in terms of extraction yields and material and energy resources.

4.3. *Valorization of apple pomace*

The study developed by Vilas-Boas et al. (2021) is based on the comparative evaluation of two “green” extraction methodologies, UAE and MAE, with conventional extraction at low (25 °C) and high temperature (80 °C), using 50% ethanol as extraction solvent (Vilas-Boas et al., 2021). When process time is evaluated, MAE is the most recommended option, requiring only 90 s, while 30 min, 1 h and 20 min are needed for UAE, 25CE and 80CE respectively. The results obtained showed that the “green” MAE technology results in the highest TPC value compared to the UAE and CE technologies at low temperature. It is worth mentioning that, with respect to the UAE technology, the values are also promising, with antioxidant recovery values comparable with CE process and the additional advantages associated with UAE in terms of reduced solvent use, reduced process time and high efficiency make it an option with high potential.

On the other hand, UAE can also be combined with the use of pectinases, the so-called ultrasound-assisted enzymatic extraction (UAEE). This procedure has yielded promising antioxidant extraction results, with 4.62-fold and 1.7-fold recovery compared to MAE and UAE, respectively (Zhang, Poojary, Choudhary, Rai, & Tiwari, 2018). Although this methodology is efficient in terms of extraction efficiency, other factors should be considered such as the economic viability, which will be affected by the use of enzymes on a large scale. In addition, from an environmental point of view, some research reports have also identified a significant environmental contribution, especially in the categories of eutrophication and ecotoxicity (Arias, Feijoo, & Moreira, 2021).

4.4. *Valorization of onion by-products*

González-de-Peredo et al. (2021) have considered the use of UAE as the green extraction method for onion antioxidants (González-de-Peredo et al., 2021). Although the optimum percentage of methanol for flavonol recovery has been 79%, it decreases to 62.5% when antioxidant potentiality is studied. Thus, 76.9% of methanol purity has been selected as the target value. On the other hand, as for the time cycle, the optimum value is closer to the requirements for antioxidant value, which is 0.94 s. Finally, for the solid-liquid ratio, an average value between flavonols and antioxidant activity value has been selected, amounting to 0.2:12.8 (g:mL) (González-de-Peredo et al., 2021).

One of the most studied valorization processes regarding the use of onion residues as a resource is the extraction of quercetin, given its excellent properties and applications as a bioactive compound. Although the most developed extraction technique is the conventional one, that is, an extraction using ethanol/methanol (Santiago et al., 2020), in the search for more sustainable process alternatives, different green extraction methodologies have been applied to obtain this flavonoid. Table 12 includes the quercetin recovery values, and their comparison with other extraction technologies, using the following green extraction techniques: microware-ultrasound assisted extraction (MUAE, (Velisdeh, Najafpour, Mohammadi, & Poureini, 2021)), supercritical water extraction (SWE, (Ko, Cheigh, Cho, & Chung, 2011)), on-line extraction enzymatic hydrolysis (On-line EEH, (Lindahl, Liu, Khan, Nordberg Karlsson, & Turner, 2013)) and microware assisted extraction (MAE, (Jin et al., 2011)).

4.5. *Inedible mushrooms by-products valorization*

Dong-Ping et al. studied the extraction antioxidants from mushrooms using UAE technology, using ethanol as solvent (Xu et al., 2016), leading to a TPC value of 91.51 mg GAE/g and an antioxidant activity power of 347 μmol Trolox/g. The results obtained are promising, as when compared to conventional extraction techniques, such as maceration (70.06 mg GAE/g, 204.34 μmol Trolox/g) and Soxhlet (72.68 mg GAE/g, 276.76 μmol Trolox/g). Tian et al. have also studied the use of UAE technology for the recovery of antioxidant compounds from mushrooms (Tian et al., 2012), reaching analogous conclusions: UAE as an effective method for antioxidants recovery due to the increase by a 155% on extraction yields when compared to conventional hot water extraction and by 28% in comparison with MAE.

Regarding MAE technology, even though it values are not as higher as using UAE, it has also been considered for the recovery of mushroom bioactive compounds (Özyürek, Bener, Güçlü, & Apak, 2014; Xiaokang et al., 2020). The report developed by Muhammad et al. obtained a TPC value of 467.69 mg GAE/100 g and an antioxidant activity of 2930 μM TE/g (Ameer, Shahbaz, & Kwon, 2017). Also Xiaokang et al. demonstrated the effectiveness of this technology as the amount of antioxidants

Table 12

Quercetin recovery values using different green extraction techniques and it comparison with other extraction technologies.

	Operation conditions	Quercetin recovery	Comparison to other extraction technologies
MUAE	60 s microware irradiation, 15 min sonication, 70 °C, 70% ethanol and liquid to solid ratio of 30 mL/g	10.32%	UAE: 8.34% MAE: 5.03%
SWE	165 °C, 15 min, 90–131 bar.	92.40% (16.29 mg/g onion skin)	CE (with methanol, 60 °C, 2 h): 3 mg/g CE (with ethanol, 60 °C, 2 h): 2 mg/g CE (water, 100 °C, 3 h): 4 mg/g
On-line EEH	β-glucosidase, 84 °C, pH 5.5, 5% ethanol, 72 h.	9.1 μmol/g fw onion	CE: 8.1 μmol/g fw PHE: 8.5 μmol/g fw
MAE	700 W, irradiation at 10s intervals, 69.7% ethanol, 117 s.	4.75 mg/g	CE (16.5 min, 59.2 °C, 59.3% ethanol): 3.70 mg/g UAE (21.7 min, 606.4 W, 43.85 ethanol): 3.76 mg/g

extracted were significantly higher than those using organic-solvent extraction, 3.68 mg GAE/g and 4.16 mg TE/g, respectively (Xiaokang et al., 2020).

Parniakov et al., 2014 have combined two technologies (pressurized extraction (PE) + pulse electric field (PEF)) for the extraction of polyphenols, leading to a clearer and more colloidal stable extract in comparison with the conventional pressure extraction technology. Besides these aforementioned advantages, the yield and amount of biocompounds extracted was also higher, which provides an effective extraction method, with comparable results of hot water extraction technology (Parniakov, Lebovka, Van Hecke, & Vorobiev, 2013).

4.6. Cabbage wastes for valorization

Cabbage stands out due to it high content in phenolics, especially anthocyanins, which exhibits high antioxidant activity (Leja, Kamińska, & Kołton, 2010). The procedures based on EAE-M (Enzyme Assisted Extraction with maceration,(Tecucianu, Draghici, & Oancea, 2020)), MAE (Sookjitsumran, Devahastin, Mujumdar, & Chiewchan, 2016) and UAE (Ravanfar, Tamadon, & Niakousari, 2015) are some of the extraction technologies that have been selected. Table 13 includes the description of the extraction process, yield values and antioxidant recoveries for the above mentioned methodologies.

5. Could antioxidants act as pro-oxidants?

The antioxidant benefit derived from the consumption of fruits and vegetables is well known. Their concentration and balanced composition of vitamins and minerals make them healthy and beneficial foods in the defense against the appearance of free radicals that cause damage to biomolecules in our body (Miranda-Díaz, García-Sánchez, Cardona-Muñoz, & Mendonça Junior, 2020). On the other hand, some studies have concluded that certain biological compounds with antioxidant capacity can act as pro-oxidants. While environmental factors have been pointed out as being responsible for the antioxidant or pro-oxidant activities of bioactive compounds, further research is needed to evaluate and state with certainty how antioxidants could also act as pro-oxidants (Rahal et al., 2014).

Recognizing this, recent research articles have reported that at least three aspects could affect the conversion of an antioxidant compound into a pro-oxidant: the concentration of the antioxidant, its redox potential and the interaction with metal ions (D'Angelo et al., 2017; Sharifi-Rad et al., 2020; Sotler et al., 2019). It is important to bear in mind this dual capacity, as acting as pro-oxidants leads to important health consequences, as the number of free radicals and reactive oxygen species (ROS) would be higher, causing damage to biomolecules such as DNA, lipids and proteins (Guardado, Molina, Joo, & Uriarte, 2012; Tan, Norhaizan, & Liew, 2018).

Vitamin C is one of the most explored antioxidants in this topic. While a potent antioxidant benefit can be achieved when consumed at low doses (up to 100 mg/kg body weight), when the upper limit of 1000 mg/kg body weight is reached, a pro-oxidant effect can be observed. This undesirable pro-oxidant capacity of vitamin C could also be developed in the presence of metal ions, in particular Fe3+ and Cu2+, which are reduced and lead to the production of hydroxyl radicals developing Fenton reactions (Smirnoff, 2018; Sotler et al., 2019). This same mechanism of action can be observed with phenolic compounds and flavonoids, both their high concentration and the presence of transition metals, leading to a pro-oxidant action. This activity is the result of the interaction between metal cations and the phenolic/flavonoid compound itself (Sotler et al., 2019). Copper and iron transition metals can modify the redox potential of phenols, which increases their ability to donate electrons and thus to develop redox reactions leading to the formation of free radicals (Kalinowska, Gryko, Wróblewska, Jabłońska-Trypuć, & Karpowicz, 2020).

Table 13
Anthocyanins recovery values using different green extraction techniques and it comparison with other extraction technologies.

	Operation conditions	Anthocyanins	Comparison to other extraction technologies
EAE-m	50% ethanol, 15/1 solvent/solid ratio, cellulase and pectinase, 180 min, 47 °C, pH 4.7	1079 mg/100 g DW	UAE: 664.3 mg/100 g EAE: 708 mg/100 g DW
MAE	0.37 W/g, 9 min, ethanol, 10/1 solvent/solid ratio	Recovery: 54.6% TPC and 63.7% FRAP	CE (7 h): 43% TPC, 52% FRAP PSE (9 min): 55.8% TPC, 53.5% FRAP
uae	100 W, 30 min, 15 °C	Yield: 21 mg/L	–

6. Discussion and conclusions

In the search for functional foods that promote the good health of consumers and the protection of their human organism against the onset of diseases, bioactive compounds, characterized by their powerful antioxidant activity, have been highlighted (Abreu-Naranjo et al., 2020; da Silva et al., 2021; Kiokias & Oreopoulou, 2021). Although different foods can provide antioxidant properties, it is fruits and vegetables that stand out for their richness (Dhalaria et al., 2020; Saini et al., 2019; Samtiya et al., 2021). In addition, consumers are increasingly demanding the presence in the market of foods with less chemical processing, with a low content of synthetic additives. Therefore, in order to provide healthy foods, with a content of antioxidant compounds and, at the same time, of natural origin, it is possible to opt for their extraction from natural-based resources. Therefore, in recent years, much effort has been devoted to the development of efficient technologies for the extraction of antioxidants from fruits and vegetables (Sharma & Bhat, 2021; Zhang et al., 2018). Although Soxhlet extraction is the most widely used, its high production costs and the amount of the solvent required make it an uncompetitive technology when compared to the production of synthetic-based antioxidants. It is in this context where the so-called "green technologies" have emerged, whose extraction yields are better than other technologies, requiring less consumption of materials, process time and production costs (Ameer et al., 2017; Chemat et al., 2020; Rombaut, Tixier, Bily, & Chemat, 2014; Wong-Paz, Aguilar-Zárate, Veana, & Muñiz-Márquez, 2020).

On the other hand, taking into account the importance of the concepts of circular economy and sustainability in the daily life of consumers, it has also been identified that important efforts have been made in the use of non-edible parts of fruits and vegetables, as well as in the waste streams of food industries, for the extraction of bioactive antioxidant compounds, given the high content of these in the aforementioned by-products, thus reducing production costs and, in turn, promoting the concept of waste as resource (Fierascu, Fierascu, Avramescu, & Sieniawska, 2019; Makris & Şahin, 2019; Montenegro-Landívar et al., 2021; Plazzotta, Ibarz, Manzocco, & Martín-Belloso, 2020; Saini et al., 2019).

In this context, the present review has been developed with the aim of compiling the data available in high impact research articles detailing the composition in antioxidant compounds of the most commonly produced fruits and vegetables in Spain. In this way, it is possible to identify which fruits and vegetables are richer in antioxidants and, therefore, their intake and their inclusion in the diet of the population contributes to good health. In addition, a critical analysis of some of the extraction methodologies, both conventional and green, used for the extraction of valuable bioactive compounds has been carried out. Through the analysis of research reports based on the use of different green technologies for the extraction of antioxidant compounds, it has been concluded that the most efficient are the MAE and UAE technologies, both from the point of view of extraction yields, analogous to those of conventional technologies, and of the operating conditions required for the process. However, other technologies based on the use of enzymatic cocktails or pulsed electric fields are also showing promising results.

Finally, it is worth mentioning that the potential of using inedible parts of fruits and vegetables, as well as waste streams from food processing industries, has also been reflected in numerous research articles, which is a very favorable aspect to address and achieve the purposes of changing the pattern of linear production to that of circular economy and sustainable development.

CRediT authorship contribution statement

Ana Arias: Methodology, Formal analysis, Investigation, Writing – original draft. **Gumersindo Feijoo:** Writing – review & editing. **Maria Teresa Moreira:** Conceptualization, Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interest or personal relationships that could have appeared to influence the work reported in this paper.

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