Food colorants: challenges, opportunities and current desires of agro-industries to ensure consumer expectations and regulatory practices

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ABSTRACT

Background: Worldwide consumers seek most delightful and appealing foodstuffs, at the same time they require safer, more nutritious and healthier products. Color is one of the most important organoleptic attributes that directly affects consumers' acceptance and food selection.

Scope and approach: The present report aims to provide an extensive approach to the field of food (natural/synthetic) colorants, namely those who are currently allowed with established acceptable daily intake (ADI). It also describes the biotechnological and industrial techniques that have been used to optimize food attractiveness, shelf life and color stability, as well as the general trends and future perspectives of food science and technology in the topic of food colorants.

Key findings and conclusions: Synthetic food colorants were largely used, but have been progressively substituted by those obtained from natural origins. Numerous side effects and toxicity, at both medium and long-terms, allergic reactions, behavioral and neurocognitive effects have been related with their use. Otherwise, naturally-derived food colorants seem to provide high quality, efficiency and organoleptic properties, and also play a contributive role as health promoters. Anthocyanins, carotenoids, phenolic compounds, beet derivatives, annatto and some curcuminoids are among the most commonly used, while strict regulatory practices have been applied looking for food quality assurance.

Key words: Chemical stability; Food products; Health impact; Natural colorants; Safety; Synthetic colorants.

1. Introduction

1.1. A brief overview on food additives

Food industry has exerted a large and fascinating impact on the science evolution, but also on health and nutrition assurance together with consumers' taste and appearance satisfaction. An increasing delivery of more and more specific products with different formats, colors, tastes, smells, textures and so on, are available in supermarkets, mostly created to ensure consumers' expectations (Ayala-Zavala et al., 2011; Carocho, Barreiro, Morales, & Ferreira, 2014; M. I. Dias, Ferreira, & Barreiro, 2015). There are no doubts about the real impact that consumers' perception, opinions and desires exert on food industries. Therefore, more appealing and delightful products have been produced and offered (Carocho et al., 2014; Delgado-Vargas & Paredes-Lopez, 2003). Among the food constituents responsible for a considerable improvement of the organoleptic characteristics, additives revealed to be pivotal. In fact, they exert truly important benefits on the shelf life, microbiological quality and security of numerous foodstuffs. Not least important to highlight is their sensorial attractiveness, namely the visual perception, color and smell, and in particular food colorants revealed to be of the utmost importance in this field (Carocho et al., 2014; Ray, Raychaudhuri, & Chakraborty, 2015; Shim et al., 2011).

Color may be considered one of the most impressive and delightful attributes of foodstuffs, which directly influences preference, selection and eating desires of the consumers (Delgado-Vargas & Paredes-Lopez, 2003; Shim et al., 2011). However, despite natural food products have their own color intensity, storage conditions, manufacturing and processing practices/methods have a pronounced influence on their final coloration; thus, food additives may be considered as promising means to

mask their unpleasant characteristics. In fact, they have been highly appreciated and selected for multiple purposes, at the same time that, increasingly strict and regulatory legislation accompanied those advances, towards to ensure the good manufacturing practices and total security of consumers (Bridle & Timberlake, 1997; M. I. Dias et al., 2015; Laokuldilok, Thakeow, Kopermsub, & Utama-ang, 2016).

However, several food additives that were used over decades are no longer currently allowed, due to the real evidences of their side effects, toxicity at medium- and long-terms and high frequency of health disturbance incidents. European Food and Safety Authority (EFSA) and Food and Drug Administration (FDA) represent the most important regulatory organizations empowered to ensure the quality and security of food products, as also to protect and promote the human health (Amchova, Kotolova, & Ruda-Kucerova, 2015; Carocho et al., 2014). More important to emphasize is that not only synthetic but also commercial plant-animal derived additives have been consecutively suspended by those authorities (Amchova et al., 2015; Rodriguez-Amaya, 2016; Tumolo & Lanfer-Marquez, 2012).

1.2. The historical perspective of colorants

Despite the increasingly specific and regulatory legislation, an intense investigation on the field of food industry have reached a prestigious level (Castañeda-Ovando, Pacheco-Hernández, Páez-Hernández, Rodríguez, & Galán-Vidal, 2009; Shahid, Shahid-ul-Islam, & Mohammad, 2013). Food additives present a long history of use, and more specifically food colorants are a good example. In the broadest sense, according to FDA, a food colorant is "any dye, pigment or substance which when added or applied to a food, drug or cosmetic, or to the human body, is capable (alone *or through reactions with other substances) of imparting color*" (FDA, 2016). Apart from their direct use, i.e. application to coloring foods, they might be also used to contribute to the flavorful assurance, safety, quality and organoleptic characteristics of foodstuffs, and not least important to warrant the consumers satisfaction.

A progressive empowerment of food industry has been observed in the last decades, mainly incited by the increasing demands by consumers (Agócs & Deli, 2011; Delgado-Vargas & Paredes-Lopez, 2003; Shahid et al., 2013). More delightful, nutritive, attractive, healthy and high sensorial quality products are already available, and therefore more specific and applied methods/techniques need to be developed and then implemented to achieve the industrial goals and consumer desires. It is not new that several external conditions, such as light, air, temperature, moisture and storage conditions play a crucial role on the food color loss (Cejudo-Bastante, Hurtado, Mosquera, & Heredia, 2014; Sagdic et al., 2013; Türker & Erdogdu, 2006). Thus, food colorants are mainly applied to offset and overcome those unpleasant characteristics, as also to homogenize the color of foodstuffs, through correction of color variations and/or enhancement of the naturally occurring food color, and even making available colorless products. The final result arising from this intervention is the appearance of specifically targeted and requested products by consumers, and the commonly named "fun foods", that significantly improves their attractiveness and consequent worldwide demand (González, Gallego, & Valcárcel, 2002; Jiménez-Aguilar et al., 2011).

On the other hand, and mainly derived from the increasing demands by consumers, including their preferences, as also the most specific legislation procedures, market pressure has been facing a growing need for perfection (Lubbe & Verpoorte, 2011; Shahid et al., 2013). In fact, the use of more natural ingredients in the foodstuffs

formulation constitutes the main focus of food industries, since a pivotal interest by consumers on natural pigments for coloring foods is doubtlessly expressed. Both food manufacturing industries as also consumers have shown a growing interest for natural colorants in line with a consequent replacement of synthetic food additives (Carocho, Morales, & Ferreira, 2015; Rodriguez-Amaya, 2016; Shahid et al., 2013). Furthermore, the scientific advances go in the same direction, clearly evidencing that natural products are associated with a consequent promotion of quality of life and health improvement while synthetic dyes are critically assessed, being even some of them no longer currently available, in spite to the notable transnational disagreements (Official Journal of the European Communities Legislation, 2009; Rodriguez-Amaya, 2016).

Thus, and based on the latest advances, the present work aims to provide a general overview of the food colorants, including an extensive description of the currently available natural and synthetic food colorants, their uses and acceptable/safe doses, and also focus on the current challenges, opportunities and desires from both food industries and consumers.

2. Color food additives

2.1. From healthy to organoleptic perceptions

Food colorants are among the most interesting features at industrial and scientific level. In fact, due to increasing demands by consumers, pronounced advances and opportunities have been achieved in food industry (Carocho et al., 2015; Shahid et al., 2013). Apart from the ancient use of food additives, natural food colorants have

received a particular attention, not only for their potent ability to color foods, but also for providing some healthy benefits (Delgado-Vargas & Paredes-Lopez, 2003; Gengatharan, Dykes, & Choo, 2015; Shim et al., 2011). Associated with the increasing demand by consumers for more delightful, attractive and pleasant products, a great interest in the health effects of some food colorants has been also observed (Agócs & Deli, 2011; Sagdic et al., 2013; Shahid et al., 2013).

With the growing and continuous search, numerous synthetic food colorants were developed to be added to improve food products quality and organoleptic characteristics, however, over time, most of them were banned due to the clearly evident side effects, signals of toxicity at short and long terms, as also health impairment abilities, including their possible carcinogenic effects (Amchova et al., 2015; Carocho et al., 2014). Thus, consumer expectations were largely affected but not changed, and requested the addition of natural pigments to foodstuffs in favor of the synthetic ones (Masone & Chanforan, 2015; Wissgott & Bortlik, 1996).

Natural food colorants revealed to be as much effective as those derived from chemical synthesis, with the subsequent benefits of: being more safe, providing health benefits besides conferring organoleptic features, exerting two or more benefits as food ingredients (in fact several food additives exerting colorant effects also act as antioxidants and even preservatives), and lastly contributing functional properties to food products (Carocho et al., 2014; Delgado-Vargas & Paredes-Lopez, 2003; Rodriguez-Amaya, 2016). The last ones, commonly called functional foods, have been increasingly demanded by consumers (Bagchi, 2006). In fact, proper regulatory practices and guidelines are still being developed and improved towards legislation and reassurance of consumers safety and life quality, and at the same time to contribute to adequate foodstuffs labeling information (Bagchi, 2006; Jauho & Niva,

2013; Viuda-Martos, Ruiz-Navajas, Fernández-López, & Pérez-Álvarez, 2010). Recent data have revealed that modern consumers with health concerns is more likely to choose functional foods for their disease-preventing properties than for their riskreducing or appearance-enhancing properties (Siró, Kápolna, Kápolna, & Lugasi, 2008). More interestingly is that not only functional foods have been increasingly sought by consumers, but also functional ingredients that improve the final quality of foodstuffs, and other agro-industrial preparations (Kammerer, Kammerer, Valet, & Carle, 2014). In this sense, natural food pigments/colorants, as well as other natural additives, are considered not only as organoleptic improvement agents, but also enhancers of nutritional status and health promoters.

2.1.1. Synthetic food colorants

Synthetic food colorants are widely used to improve the attractiveness of numerous foodstuffs. In spite a considerable amount of them have been increasingly removed and even prohibited in the food industry, blue (**Table 1**), red to orange (**Table 2**), yellow (**Table 3**) and green and white (**Table 4**) synthetic food colorants are among the most commonly used and studied in terms of security, side effects, toxicity at short, medium and long terms, as well as health impact. Their application in food products is currently allowed by FDA and EFSA, with already established acceptable daily intake (ADI) doses.

Based on the ADI values, blue (**Table 1**), followed by yellow (**Table 3**) and lastly green and white (**Table 4**) colorants seem to be the less dangerous, even at the higher ADI doses. Interestingly, titanium dioxide (E171) has not an established ADI, being thus currently used in confectionary, baked goods, cheeses, icings and toppings with

permitted maximum levels not defined. As shown in Table 2, red to orange colorants present the lower ADI doses, namely erythrosine - E127 (0.1 mg/kg b.w.), red 2G -E128 (0.1 mg/kg b.w.) and amaranth – E123 (0.8 mg/kg b.w.), which means that the occurrence of side effects and related toxicity is possible. Not least important to highlight is the occurrence of cumulative effects related with their daily intake. In fact, the majority of food products in which these colorants are applied are also daily consumed, i.e. beverages, cocktails, alcoholic drinks, fish and meat products, and candied cherries (widely consumed by children and teenagers). Apart from allergic reactions, several reports have inclusively reported that artificial food colorants highly affect children's behavior (Gostner, Becker, Ueberall, & Fuchs, 2015; Jiménez-Aguilar et al., 2011; Masone & Chanforan, 2015). Attention deficit hyperactivity disorder (ADHD) is the most common, with six synthetic food colorants being currently indicated as having negative effects on the concentration activity (Council Regulation (EC) 1333/2008), namely tartrazine (E102), quinolone yellow (E104), sunset yellow FCF (E110), carmoisine/ azorubine (E122), Ponceau 4R (E124), Allura Red AC (E129).

Thus, it is feasible to infer that the risk of organic saturation and consequently the occurrence of side effects and toxicity will be markedly improved. Moreover, most of the food colorants are applied in sugar products and beverages (alcoholic and non-alcoholic), which incite the consumption of these products.

2.1.2. Natural food colorants

With the increasing demand by consumers for naturally-derived and safer food ingredients, absent of toxic side effects and even health promoters, numerous

experiments have been carried out to provide more effective and selective food colorants. As an example, brown to black food colorants still continue to be highly explored, both derived from synthesis as also from natural sources; their food applications, chemical stability, side effects and related toxicity are among the main parameters exploited, towards determining the most effective and safer ADI. Thus, for these currently approved food colorants, E codes were approved and ADI were established, namely to caramel - E150 (160-200 mg/kg b.w.), authorized to be used in sauces, biscuits, crisps, pickles and several alcoholic and non-alcoholic beverages (EFSA, 2011a); brilliant black – E151 (1 mg/kg b.w.), used in several cheeses, wine, sauces, and drinks (EFSA, 2010b); vegetable carbon – E153 (not established), used in jam and jelly crystals (EFSA, 2012b); brown FK - E154 (0.15 mg/kg b.w.), authorized in smoked and cured fish, meat and crisps (EFSA, 2010d); and brown HT - E155 (1.5 mg/kg b.w.), used in several biscuits, chocolate and cakes (EFSA, 2010e). Another interesting area under research is the use of yeast-derived natural pigments; one of the latest studied is monascin, a secondary yellow natural pigment produced by the genus Monascus. Appart from their interesting food coloring attributes, several biological activities, such as anti-cancer, anti-inflammatory, anti-diabetic, and anticholesterolemic effects has been also reported (Patakova, 2013; C. Wang et al., 2015). But, like does not exist a general consensus about their safe use, no E code was stablished, whereas their use considered illegal in Germany and in Asian countries is largely applied in food products (Wild, 2000). Thus, and considering these aspects, further investigation on this field should be carry out.

However, other naturally-occurring food colorants have been also studied, namely anthocyanins (**Table 5**), beet colorants (**Table 6**), carotenoids (**Table 7**) and phenolic compounds (**Table 8**). Annato, carminic acid and some curcuminoids, particularly

curcumin, have been also limited investigated, while many others still to be examined and their use is not yet authorized with an E code (**Table 9**).

Anthocyanins (Table 5) are the most widely studied natural food colorants, being obtained from flowers, fruits, leaves and even whole plants. Commercial anthocyanins, namely cyanidin 3-glucoside, pelargonidin 3-glucoside and peonidin 3glucoside have been also used, and their effectiveness has been increasingly assessed. It is really important to highlight that external interferences highly affect the anthocyanin pigment colors, namely pH, temperature, humidity, salinity, stress conditions and even storage conditions. Thus, the anthocyanins color may vary from red to purple and blue color (Cabrita, Fossen, & Andersen, 2000; Jiménez-Aguilar et al., 2011; Nontasan, Moongngarm, & Deeseenthum, 2012; Türker & Erdogdu, 2006). As a particular example, Cabrita et al. (2000) evaluated the effects of pH, and temperature during storage on the anthocyanins stability and color, and described that in strong acidic medium reddish color is the most prominent, while at relative neutral conditions bluish color dominates. Furthermore, for the anthocyanins 3-glucosides a maximum level of stability was obtained at pH values 8-9, while for other ones a pronounced stability was obtained at pH values ranging from 5 to 7 (Cabrita et al., 2000). Regarding red-purple colorants derived from beets (Table 6), betacyanins and betalains are the most commonly studied and were already approved (E162) to be safely used. Interestingly, not only *Beta vulgaris* L. root is a source of these natural colorants, but also fruit of Hylocereus polyrhizus (Weber) Britton & Rose (Stintzing, Schieber, & Carle, 2002), Opuntia ficus-indica [L.] Miller (Cassano, Conidi, & Drioli, 2010; Otálora et al., 2015), Opuntia stricta (Haw.) Haw. (Obón, Castellar, Alacid, & Fernández-López, 2009) and Rivina humilis L. (Khan & Giridhar, 2014) are also rich

in these ingredients, widely used in burgers, desserts, ice creams, jams, jellies, soups, sauces, sweets, drinks, dairy products and yogurts.

Carotenoids are another group of naturally-derived food colorants with a renowned impact and demand, mainly due to their prominent coloring attributes and bioactive properties, among other health benefits. Their antioxidative potential is widely known and scientifically recognized, being used in large scale by the food industry also as natural preservatives (M. G. Dias, Camões, & Oliveira, 2009; Rodriguez-Amaya, 2016). These pigments have been also increasingly used for cosmetic, pharmaceutical and nutraceutical purposes, with available supplements containing both individual and mixtures of these ingredients (Martín, Mattea, Gutiérrez, Miguel, & Cocero, 2007; Rodriguez-Amaya, 2016). However, their food colorant attributes are also highly appreciated. As shown in Table 7, carotenoids are particularly selected by food industries for inclusion in foodstuffs with high fatty acid content. In fact, butter and margarines, cakes, milk products and soft drinks possess, respectively, a high, moderate and low percentage of lipids, and therefore possess different susceptibilities to oxidation process. Not only vegetable sources, such as plant roots, flowers, leaves and even the whole matrix are used as raw material to extract carotenoid pigments, but also algae/microalgae, fungus/yeasts and aquatic animals (Danesi, Rangel-Yagui, Carvalho, & Sato, 2002; Grewe, Menge, & Griehl, 2007; Hong, Suo, Han, & Li, 2009; K Nabae et al., 2005). For example, astaxanthin (E161j) is isolated from animal sources, while β -carotene (E160a) may be both extracted from the roots of *Daucus* carota L. and even from fungus (Blakeslea trispora). The most common color attributes of carotenoids are yellow to orange and even red color. As previously highlighted, lutein (E161b) and astaxanthin (E161j) are the carotenoids most commonly used for pharmaceutical and nutraceutical purposes, being used not only to

confer bioactive and functional properties but also colorant attributes (Carocho et al., 2015; Devasagayam et al., 2004).

Another promising class of natural food colorants is phenolic compounds (**Table 8**). Flavanones (naringin), flavones (4',5,7-trihydroxyflavones and apigenin) and flavonols (fisetin, myricetin, myricitin, quercetin and rutin) have been the most widely investigated, but so far commercial products are the most commonly used. Only myricetin and myricitrin were from plant origin, namely isolated from *Myrica cerifera* L. roots. The colorant attributes of many other phenolic compounds have been also studied but their safety, stability and spectrum of activity still remains unclear (Carocho & Ferreira, 2013; Grotewold, 2006; Robbins, 2003). In fact, phenolic compounds do not possess approved E code and ADI, and despite their wide recognition as prominent antioxidants and health-promoters and functional ingredients, the use of this large group of bioactive molecules as food additives continues to be poorly investigated. Lastly, it is of the utmost importance to refer that apart from their biological attributes, phenolic compounds are among the main responsible agents for the wide variety of naturally-occurred food colors (Jiménez-Aguilar et al., 2011; Shahid et al., 2013).

Furthermore, curcumin (E100), one of the most important and widely known curcuminoids derived from *Curcuma longa* L. rhizomes is also widely used as food colorant for multiple purposes (**Table 9**). Annato (E160b) is other natural food colorant, with a long history of use. Bixin and norbixin are the main components of this yellow-red pigment, extracted from *Bixa orellana* L. seeds. The yellow to red-orange natural food colorant, carminic acid (E120) already exists from synthetic origin, with an established ADI of 5 mg/kg b.w., being largely used for several purposes, such as in cakes, cookies, beverages, jam, jelly, ice creams, sausages, pies,

dried fish, yogurt, gelatins, cider, tomato, dairy products, cherries, non-carbonated drinks, chewing gums, pills and cough drops (Bibi, Galvis, Grasselli, & Fernández-Lahore, 2012; Huang, Chiu, Sue, & Cheng, 2003; Huang et al., 2002; Masone & Chanforan, 2015). Despite synthetic food colorants have presented a large utilization and lower-associated costs than those derived from natural origin, they have been progressively substituted by naturally-derived food colorants, which are safer, specific, absent of side effects and related toxicity, and are also able to confer healthimproving effects and functional benefits (Carocho et al., 2014; M. I. Dias et al., 2015). There are other food colorants under investigation, such as c-phycocyanin, a blue pigment isolated from Arthrospira platensis (cyanobacteria) (Martelli, Folli, Visai, Daglia, & Ferrari, 2014); c-phycoerythrin, a red-orange pigment from bluegreen algae (Mishra, Shrivastav, Pancha, Jain, & Mishra, 2010); hot-air and freeze dried aerial parts of Crithmum maritimum L., which provides, respectively, a very interesting grey and green color when added to pasta, sauces, rice, fish and meat (Renna & Gonnella, 2012). Genipin, a blue pigment derived both from fruits of Gardenia jasminoides Ellis and Genipa americana L. also revealed prominent attractive potentialities when added to beverages, juices, nectars, desserts and gels (Gao, Zhang, Cui, & Yan, 2014; Hou, Tsai, Lai, Chen, & Chao, 2008; Ramos-De-La-Peña et al., 2014). The red pigment madder color, isolated from Rubia tinctorum L. roots also revealed to improve the general acceptability of hams, sausages, boiled fish, paste, beverages and even some confectionaries products, as well as the violet pigment violacein isolated from the bacteria Chromobacterium violaceum UTM 5 when added to yogurt and jelly (Venil et al., 2015).

Other naturally-occurring pigments, but from commercial origin, have been also studied, such as geniposide, monascorubrin and purple corn color (Kyoko Nabae et al., 2008; Ozaki et al., 2002; Wada et al., 2007). In fact, for the majority of naturallyoccurring food pigments, studies using natural pigments and also those from commercial sources, have been carried out. These procedures are considered very important mainly for two reasons: the first one is to compare both the efficiency and efficacy between pigments according to their origin, and the second one to determine if the safety and probability of side effects, and related toxicity are similar. Overall, and despite the current worldwide advances in the food industry, particularly on the field of food colorants, it is still necessary to further the knowledge on this expansive area.

2.2. Food industries: challenges, opportunities and unexpected demands

Considering the increasingly specific and directional consumer claims/requirements, a high level of market pressure has been put on food industries (Agócs & Deli, 2011; Carocho et al., 2014). The satisfaction of consumer needs and expectations are among the most important goals of agro-industrial and biotechnological industries, but not least important to highlight is the real contribution of the scientific research to food science and technology (Ayala-Zavala et al., 2011; Carocho et al., 2014).

A direct and bidirectional collaboration between food industries and scientific research institutions has been observed; the first one applies the achieved scientific knowledge to produce highly valuable and nutritionally-enriched food products, while the second one provides the food manufacturing industries with latest advances on the field of food science, aiming to the elaboration and subsequent commercialization of more natural, healthier, safer and functional products/ingredients. Moreover, scientific research centers also carry out high end investigations to assess the real effects of new

molecules/ingredients/foods on health and wellbeing, at short and long terms, and then, based on the obtained results, several optimization procedures are applied by food industries to properly ensure the consumers safety. Not least important to highlight is the progressive and increasingly strict regulatory legislation on food additives, and particularly on food colorants, clearly distinguishing permitted from prohibited and safe from harmful food colorants (Carocho et al., 2014; Shahid et al., 2013; T. Zou, He, Yasen, & Li, 2013). Lists of permitted and prohibited food colorants have been progressively updated (Council Regulation (EC) 1129/2011; Council Regulation (EC) 1333/2008), but prominent discrepancies are observed among different countries of European Union and even other countries (Chemical Engineering, 2002; Europe Environment, 2005; Official Journal of the European Communities Legislation, 2009). Most of them refer to synthetic food colorants, as soon as measurable health impairment effects are observed and scientifically proved (Carocho et al., 2015; Delgado-Vargas & Paredes-Lopez, 2003; Shim et al., 2011), in spite of already published contradictory results (Amchova et al., 2015; Kapadia et al., 1998). In line with this, a progressive substitution of synthetic with natural food colorants has been intensified, up to a point where numerous used natural pigments still continue to be poorly studied and their real functions are unknown. Although some natural pigments have already been used as substitutes of their related synthetic ones, the wide benefits and related use/application of many others has not been tested yet. Therefore, gaining knowledge on this field could be considered a promissory advance in order to develop more specific and functional foods/products.

3. Multidimensional applications of food colorants

3.1. A general overview of food attractiveness optimization

Over the years, most accurate and reliable methods, and even increasingly specific analytical procedures have been developed and applied for a wide variety of purposes by food industries. Detection of undeclared substances, illegal ingredients and the abundance of many other additives, both allowed and prohibited, have deserved a particular interest (Bonan, Fedrizzi, Menotta, & Elisabetta, 2013; Xing et al., 2012). Most of the label information is incomplete and even ambiguous, up to a point that consumers require adequate information on food packaging, including the addition of warning and/or safety messages (Masone & Chanforan, 2015; Sanjay et al., 2007; Shim et al., 2011). Furthermore, another important aspect, which has also received special attention, is the real stability of food colorants and respective stored foodstuffs.

Color appreciation comprises one of the earliest aesthetic parameters considered by consumers during foodstuffs selection, being a direct predictive parameter that ensures good quality (Sagdic et al., 2013; Todaro et al., 2009). Visual cues present a doubtless influence on food preference, acceptability and lastly food choice. Therefore, it is clearly evident that food industries aim to provide increasingly uniform, attractive and pleasant colored foodstuffs, to fully satisfy consumers' expectations and current needs (Cai, Sun, & Corke, 2005; Junqueira-Goncalves et al., 2011; Otálora, Carriazo, Iturriaga, Nazareno, & Osorio, 2015; Sagdic et al., 2013). Several reports have confirmed the real interference that some external factors exert on the food color stability; temperature, light, air/oxygen, pH, chemical structure, solvents, packaging materials and storage conditions are among the most important factors that can reduce food attractiveness (Jiménez-Aguilar et al., 2011; Lemos, Aliyu, & Hungerford, 2012; Zhu et al., 2015). Anthocyanin pigments are a good

example of natural food colorants highly affected by those external agents; in fact, they are very unstable compounds and highly susceptible to degradation (Assous, Abdel-Hady, & Medany, 2014; Jiménez-Aguilar et al., 2011; Sagdic et al., 2013; Zhu et al., 2015). Therefore, their final coloration will vary significantly, at the same time that various degrees of susceptibility to external factors may be also observed among different species belonging to the same plant family (Sagdic et al., 2013). For example, Sagdic et al. (2013) observed that red tulip anthocyanins are less sensitive to temperature than violet tulip anthocyanins that exhibited a lower chemical stability. Jiménez-Aguilar et al. (2011) evaluated the color stability of spray-dried blueberry, a very important source of anthocyanins, and observed a progressive increase of air outlet temperature together with a loss of total phenolics and anthocyanins and a decrease in the antioxidant activity. On the other hand, Tan et al. (2014) aiming to assess both the effects of temperature (under refrigerated and room temperature conditions) and pH (from 1.0 to 11.5) on a purple-red anthocyanin-derived extract of Rhoeo spathacea (Swartz) Stearn leaves, concluded that it exhibited a phenomenal color stability under a range of pH values and temperatures. Over a period of 60 days, the authors observed a remarkable stability of anthocyanin-derived extract in acidic pH, and a complete stability when solid food (jelly) and liquid food (barley water) were used (J. B. L. Tan, Lim, & Lee, 2014). Furthermore, Munawar & Jamil. (2014), aiming to access the color changes, anthocyanin stability and antioxidant activity of several spray dried plant pigments, observed that the most prominent antioxidant potential was obtained to those possess higher anthocyanin content, being again emphasized the negative impact of storage temperature on the final chemical stability and acceptability of products. But, even so, the authors highlighted the safety and healthy abilities of the studied natural colorant, may even be upcoming used as functional ingredient in food products (Munawar & Jamil, 2014).

Many other examples related with natural pigments stability might be highlighted but the most interesting feature that indeed deserves a particular attention is that highly specific and reliable procedures, and industrial techniques have been effectively used to overcome the problem of poor stability of numerous food pigments, as also to identify and quantify their relative abundance in numerous foodstuffs.

3.2. Agro-industrial strategies to ensure consumer satisfaction

The wide variety of food colorants currently applied in food industry has been highly scrutinized, both from regulatory agencies and subsequent updated legislation as also scientific researchers, that exert a meticulous and increasingly detailed study of food colorants, including a wide variety of evaluation criteria (Konczak et al., 2005; Shahid et al., 2013; Venil, Zakaria, & Ahmad, 2013). World consumers also perform a significant interference in these aspects, because they are increasingly informed and more exigent in relation to the quality, safety, credibility and scientific assurance of commercialized foodstuffs and their chemical additives, even requesting "clean labels" (Carocho et al., 2014, 2015; Gengatharan et al., 2015). Nowadays, the consumer satisfaction is not only related with the taste, appearance, smell and attractiveness of foodstuffs, but also with their health impact, improvement of life quality and longevity (Giusti & Wrolstad, 2003; Kammerer et al., 2014; Xi et al., 2007). Thus, food industries must simultaneously satisfy not only the organoleptic requirements but also health claims of consumers, and therefore, it becomes of the utmost importance to establish a direct collaboration with research institutions, and

listen to the worlds consumers requirements (mainly through satisfaction questionnaires).

More efficient techniques have been applied by food industries to optimize the extraction of naturally-derived pigments, to ensure their stability, to avoid color loses of those pigments and the appearance of unpleasant characteristics (Cerón, Higuita, & Cardona, 2012; M. I. Dias et al., 2015; Ray et al., 2015; Santos, Albuquerque, & Meireles, 2011; Sivakumar, Vijaeeswarri, & Anna, 2011; Yang & Zhai, 2010b). In addition, modern food processing and packaging techniques have been also developed, not only to improve the shelf life of natural products and to reduce the subsequent addition of synthetic additives, but also to contribute for the production of safer products, without affecting their attractiveness by consumers and at the same time to improve their subsequent demands (Buchweitz, Brauch, Carle, & Kammerer, 2013; Chung, Rojanasasithara, Mutilangi, & McClements, 2015; Fernandez, Torres-Giner, & Lagaron, 2009; Komolprasert, Diel, & Sadler, 2006).

3.3. Industrial and biotechnological procedures to offset colorants instability

As previously highlighted, several analytical techniques have been developed and ameliorated both to easily identify and quantify food colorants present in a wide variety of foodstuffs. In fact, some food colorants which are no longer permitted in EU, still continue to be identified, even at lower doses, in several food products (Official Journal of the European Communities Legislation, 2009). The observed side effects, related toxicity, at medium and long terms, as well as the cumulative effects of their ingestion were more than evident factors that led to their suspension. However, and since regulatory legislation varies from different countries, an increasingly strict evaluation has become increasingly necessary in order to ensure consumers safety and security (Bonan et al., 2013; Kammerer et al., 2014; Shahid et al., 2013; T. Zou et al., 2013).

Among the most commonly used analytical techniques to identify and/or quantify food colorants, are spectrometry, thin-layer chromatography, adsorptive voltammetry and differential pulse chromatography, which have been progressively substituted by capillary electrophoresis and ion chromatography, once revealed to be less timeconsuming to pretreat samples and applicable to complex colorant mixtures (Chen, Mou, Hou, Riviello, & Ni, 1998; Huang, Shih, & Chen, 2002; Karanikolopoulos, Gerakis, Papadopoulou, & Mastrantoni, 2015). But, with the progressive use of those techniques sensitivity problems are still frequent and the robustness of these methods is still significantly affected and limited. Thus, more robust and high resolute, selective and sensitive analytical techniques are increasingly preferred, namely highchromatography performance reversed-phase liquid and ion-pair liquid chromatography coupled with UV or diode-array detectors (Karanikolopoulos et al., 2015; Reinholds, Bartkevics, Silvis, Ruth, & Esslinger, 2015).

Therefore, in order to improve the general stability and the attractiveness of numerous foodstuffs, some techniques have been applied. More interestingly is that some of them are also used to neutralize and/or to remove several unpleasant characteristics of foodstuffs and even food colorants. Silva et al. (2005) investigating the influence of different hydrodistillation procedures on the turmeric deodorization observed that distillation under high vacuum and using a rotatory evaporator were not efficient. On the other hand, distillation of medium size grated turmeric using a Clevenger apparatus led to a pronounced decrease of turmeric flavor. Then, the authors assessed the general acceptability of both turmeric-derived colorants in gelatin, and the gelatin

prepared with deodorized turmeric was preferred (Silva, Nelson, Drummond, Dufossé, & Glória, 2005). In a similar way, Laokuldilok et al. (2016) evaluated the odor masking ability and encapsulation efficiency of turmeric extract prepared by a binary blend of wall materials. They concluded that the optimal formulation was the microcapsules consisting of 5% of core loading in addition with 20 g/L of β cyclodextrin, once that a high amount of curcuminoids was encapsulated and corresponded to the formulation with low volatile release (Laokuldilok et al., 2016). Furthermore, Martins et al. (2013) evaluating the spray drying effects on curcuminoid and curcumin content as well as the related solubility of microparticles containing curcuma extract, concluded that the solubility of curcuminoids was markedly improved 100-fold by using microparticles. In fact, encapsulation by drying technology and microencapsulation techniques have gained progressively a particular reliability up to a point that are used both in food industries, but also for pharmaceutical and nutraceutical purposes (M. I. Dias et al., 2015; Fernandez et al., 2009; Otálora et al., 2015; Ray et al., 2015). As an example, Venil et al. (2015) aiming to assess the spray drying effect on the stability of an encapsulated violet pigment produced from Chromobacterium violaceum UTM 5, described that the high stability of this pigment was achieved at pH 7, temperature 25-60 °C and under dark conditions, during entire storage period (Venil et al., 2015). Khalil et al. (2012) also evaluated the lutein stability and bioavailability under food processing conditions and application of spray drying, and concluded that the emulsification of medium-chain triacylglycerols oil markedly improved the stability and bioavailability of lutein esters (Khalil et al., 2012).

On the other hand, thermal treatments have been also revealed to significantly influence color and chemical stability of some food colorants and their related enriched foodstuffs. Khan and Giridhar (2014) aiming to evaluate the stability of betalains derived from *Rivina humilis* L. berry juice observed that at 90 °C for 36 min and at 25 °C for 48 days, up to 95% and 96% of betacyanins were degradated, respectively, while at 5 °C for 90 days only 15% of them were destroyed (Khan & Giridhar, 2014). Jiménez-Aguilar et al. (2011) carried out a deepen evaluation of the color variation and concentration of chemical compounds (responsible for the final coloration) on spray-dried powders derived from blueberry extracts, with added mequite gum. The authors observed that the lowest losses of bioactive colorant chemical compounds and minor color variations were found in samples dried at 140 °C and 9.1 mL/min, followed by microencapsulates stored at 4 °C for 4 weeks in the absence of light.

Lastly, gamma irradiation techniques have been also increasingly investigated, as well as upcoming procedures that improve the shelf life of numerous foodstuffs, at the same time ensure the relative stability of numerous food additives (among them food colorants). For example, Komolprasert et al. (2006) assessing the effects of 10- and 20-kGy gamma irradiation in a polystyrene polymer containing either yellow or blue colorant, concluded that both yellow and blue colorants are relatively stable under these gamma irradiation doses (Komolprasert et al., 2006).

Overall, and despite the prominent advances, it still continues to be imperative to deepen knowledge on this field, towards providing more feasible, secure and highly specific procedures to contribute to the consumers' satisfaction, security and long term health security, at the same time that food industry achieves its strategic goals and produce increasingly valuable foodstuffs.

4. Current desires and futures perspectives

Consumer demands and their preferences for naturally-derived colorants have increased exponentially, widely associated with the image of healthy, safe and good quality products, which constitutes a great challenge to food industries and related food science research institutions (Carocho et al., 2014; M. I. Dias et al., 2015; Shahid et al., 2013). There are no doubts that nature is highly rich in color pigments and that the majority of plants has not yet been exploited for their coloring properties/abilities. Otherwise, and despite these drawbacks, the chemical stability of numerous naturallyderived food pigments is markedly affected by several external factors, such as pH, temperature, light, oxygen, solvents, presence of enzymes, proteins and metallic ions, as well as their structure and concentration used (Lemos et al., 2012; Ravichandran et al., 2013). To overcome this constraint, increasingly deepen experiments have been carried out to provide new and highly specific sources and procedures to improve the extraction efficiency and related stability of those food pigments (Jiménez-Aguilar et al., 2011; Martins, Pereira, Siqueira, Salomão, & Freitas, 2013; Obón et al., 2009; Otálora et al., 2015). Thus, new naturally-derived food pigments are prepared to satisfy consumer expectations. In parallel with this, increasingly effective techniques are needed to retain the stability of natural food pigments and to ensure the final attractiveness of enriched-foodstuffs, during the manufacturing and processing practices, as well as storage conditions.

Overall, and despite the current advances in the field of food science, many other natural sources of food pigments need to be assessed for their coloring properties, whereas for the current ones, sufficient quantities should become available mainly by industrial extraction and subsequent use.

5. Conclusions

Natural food colorants have been demanded and become increasingly popular among worldwide consumers; their safety, functional and biological potential, health effects, as well as the whole impact at short and long terms, still continues to be exploited and markedly benefits of their consumption have been stated. In comparison to synthetic colorants with great perceptions of undesirable and harmful effects, natural ones tend to be perceived by consumers as safer. However, allergenic and intolerance reactions still continue to be reported comparing to the use of synthetic colorants. Thus, despite natural pigments offer a strong advantage when compared to the synthetic ones, its safety and whole effects needs to be assessed, in order to conclusively demonstrate health improving effects.

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Table 1. Blue synthetic f	food colorants.
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Colorant	E code	ADI	Uses	References
Brilliant blue FCF	E133	10 mg/kg b.w.	Dairy powders, color	(Bonan et al., 2013; Chen et al., 1998; EFSA, 2010c; Huang et
			beverages, jellies, candies,	al., 2003, 2002; Kapadia et al., 1998; Kong et al., 2015; FJ.
			condiments, icings, syrups,	Liu, Liu, Li, & Tang, 2015; K. Ma, Yang, Jiang, Zhao, & Cai,
			extracts	2012; Medeiros, Lourencao, Rocha-Filho, & Fatibello-Filho,
				2012b, 2012a; Minioti, Sakellariou, & Thomaidis, 2007; Ni,
				Wang, & Kokot, 2009; Pan, Ushio, & Ohshima, 2005; Qi et
				al., 2011; Vidotti, Costa, & Oliveira, 2006)
Indigo carmine	E132	5 mg/kg b.w.	Ice-cream, sweets, baked	(Berzas, Flores, Llerena, & Fariñas, 1999; Bonan et al., 2013;
(syn.: indigotine)			goods, confectionery, and	Chen et al., 1998; EFSA, 2014; Huang et al., 2003, 2002;
			biscuits	Kapadia et al., 1998; Kong et al., 2015; Minioti et al., 2007;
				Pan et al., 2005; Qi et al., 2011; Yuzhen Wang et al., 2009)
Patent blue V	E131	15 mg/kg b.w.	Scotch eggs and jelly sweets	(Berzas et al., 1999; Bonan et al., 2013; EFSA, 2013; Minioti
				et al., 2007)

Table 2. Red to orange synthetic food colorants.	•
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Colorant	E code	ADI	Uses	Reference
Allura Red AC	E129	7 mg/kg b.w.	Soft drinks and processed meats	(Bonan et al., 2013; Chen et al., 1998; EFSA, 2009a; El-Sheikh & Al-Degs, 2013; Huang et al., 2003, 2002; Kapadia et al., 1998; Karanikolopoulos et al., 2015; Kong et al., 2015; K. Ma et al., 2012; Masone & Chanforan, 2015; Minioti et al., 2007; Obón, Castellar, Cascales, & Fernández-López, 2005; Pan et al., 2005; Qi et al., 2011; Xie et al., 2012; T. Zou et al., 2013)
Amaranth	E123	0.8 mg/kg b.w.	Beverages, alcoholic drinks and fish roe	 al., 2011, Ale et al., 2012, 1. 200 et al., 2013) (Basu & Kumar, 2015b; Bonan et al., 2013; Chen et al., 1998; Daoud, Mesmoudi, & Ghalem, 2013; EFSA, 2010a; Karanikolopoulos et al., 2015; Kong et al., 2015; K. Ma et al., 2012; M. Ma, Luo, Chen, Su, & Yao, 2006; Minioti et al., 2007; Mpountoukas et al., 2010; Ni et al., 2009; Obón et al., 2005; Pan et al., 2005; Qi et al., 2011; Ryvolová, Táborský, Vrábel, Krásenský, & Preisler, 2007; Yuzhen Wang et al., 2009; Xie et al., 2012; Xing et al., 2012)
β-carotene	E160a	5 mg/kg b.w.	Sauces, milk, spice blends, marinades, beverages, coatings, fruit juices, margarines	(Campardelli, Adami, & Reverchon, 2012;
Carminic acid	E120	5 mg/kg b.w.	Cakes, cookies, beverages, jam, jelly, ice cream, sausages, pies, dried fish, yogurt, gelatins, cider, tomato, dairy products, cherries, non-carbonated drinks, chewing gum, pills and cough drops	(EFSA, 2015b; Kapadia et al., 1998; FJ. Liu et al., 2015)

Carmoisine (syn.: azorubine)	E122	4 mg/kg b.w.	Blancmange, Swiss rolls, jams, jellies, yoghurts, bread- crumbs, mouthwash and cheesecakes	(Basu & Kumar, 2014, 2015a; Bonan et al., 2013; Datta, Mahapatra, & Halder, 2013; EFSA, 2009b; Karanikolopoulos et al., 2015; Masone & Chanforan, 2015; Minioti et al., 2007; Obón et al., 2005; Ryvolová et al., 2007)
Erythrosine	E127	0.1 mg/kg b.w.	Cocktails and candied cherries	 (Chequer, Venâncio, Bianchi, & Antunes, 2012; EFSA, 2011b; Ganesan, Margolles-Clark, Song, & Buchwald, 2011; Kapadia et al., 1998; Karanikolopoulos et al., 2015; Kong et al., 2015; K. Ma et al., 2012; Minioti et al., 2007; Mpountoukas et al., 2010; Obón et al., 2005; Pan et al., 2005; Qi et al., 2011; Ryvolová et al., 2007; Xie et al., 2012; T. Zou et al., 2013)
Lithol Rubin BK	E180	1.5 mg/kg b.w.	Cheese rind	(EFSA, 2010g; Kapadia et al., 1998)
Ponceau 4R	E124	4 mg/kg b.w.	Non-alcoholic drinks, sweets, jellies	(Bonan et al., 2013; Capitán-Vallvey, Fernández, Orbe, & Avidad, 1998; Chen et al., 1998; Daoud et al., 2013; EFSA, 2009c; Huang et al., 2003, 2002; Karanikolopoulos et al., 2015; Kong et al., 2015; K. Ma et al., 2012; M. Ma et al., 2006; Minioti et al., 2007; Ni et al., 2009; Obón et al., 2005; Pan et al., 2005; Qi et al., 2011; Ryvolová et al., 2007; Xie et al., 2012; Xing et al., 2012; T. Zou et al., 2013)
Red 2G (syn.: azophloxine)	E128	0.1 mg/kg b.w.	Confectionery and meat products	(Bonan et al., 2013; EFSA, 2007; Karanikolopoulos et al., 2015; Minioti et al., 2007; Obón et al., 2005; Ryvolová et al., 2007; T. Zou et al., 2013)

Colorant	E code	ADI	Uses		Reference
Quinoline yellow	E104	10 mg/kg b.w.	Fruit and vegetable	s juice	(Bonan et al., 2013; EFSA, 2009d; Kapadia et al., 1998 Masone & Chanforan, 2015; Minioti et al., 2007 Shahabadi & Maghsudi, 2013; Xing et al., 2012)
Sunset yellow FCF	E110	2.5 mg/kg b.w.	Non-alcoholic sweets, jellies	drinks,	
Tartrazine	E102	7.5 mg/kg b.w.	Non-alcoholic sweets, jellies	drinks,	

 Table 3. Yellow synthetic food colorants.

 Table 4. Other synthetic food colorants.

Colorant	E code	ADI	Uses	Reference
Green				
Copper chlorophyllin-complexes	E141ii	7.5 mg/kg b.w.	Green table fresh olives	(EFSA, 2015a; Gandul-Rojas, Roca, & Gallardo-Guerrero, 2012; Mortensen & Geppel, 2007)
Green S	E142	5 mg/kg b.w.	Canned peas, cake mixes, mint jelly and sauce	(EFSA, 2010f; Minioti et al., 2007)
White				
Titanium dioxide	E171	Not specified	Confectionary, baked goods, cheeses, icings, toppings	(EFSA, 2006; Gu et al., 2015)

Table 5. Natural food colorants belonging to the anthocyanins class (E163).

Source	Uses	Reference
3-Deoxyanthocyanidins		
Sorghum bicolor (L.) Moench seeds	Breads, cakes, cookies, and tortillas	(Dykes, Rooney, & Rooney, 2013)
Anthocyanin-derived extracts		
Acacia decurrens Willd. bark	Beverages, candies, dry mixed	(Sivakumar et al., 2011)
Ajuga reptans L. flowers	concentrates, chewing gums, yoghurts,	(Terahara et al., 2001)
Brassica oleracea L. leaves	sauces, soft drinks, sweet jelly, confectionary and dairy products	(Chandrasekhar, Madhusudhan, & Raghavarao, 2012)
Brassica oleracea L. ssp. capitata f. rubra		(Buchweitz et al., 2013)
Canna indica L. flowers		(Srivastava & Vankar, 2010)
Celosia cristata L. flowers		(Sivakumar et al., 2011)
<i>Coffea arabica</i> L. husks		(Prata & Oliveira, 2007)
Daucus carota L. roots		(Assous et al., 2014)
Euterpe oleracea Mart. fruits		(Coïsson, Travaglia, Piana, Capasso, & Arlorio, 2005)
Hibiscus rosa-sinensis L. flowers		(Mak, Chuah, Ahmad, & Bhat, 2013)
Hippeastrum reticulatum var. striatifolium bulb		(Nitteranon et al., 2014)
<i>Ipomoea batatas</i> (L.) Lam. tubers		(Cipriano, Ekici, Barnes, Gomes, & Talcott, 2015)
Mirabilis jalapa L. flowers		(Sivakumar et al., 2011)
Musa paradisiaca Linnaeus bracts		(Pazmio-Durán, Giusti, Wrolstad, & Glória, 2001a)
Oryza sativa L. grains		(Loypimai, Moongngarm, Chottanom, & Moontree, 2015)
Oxalis triangularis ssp. papilionaceae leaves		(Pazmio-Durán, Giusti, Wrolstad, & Glória, 2001b)

Punica granatum L. rind		(Sivakumar et al., 2011)
Rhoeo spathacea (Swartz) Stearn leaves		(J. B. L. Tan et al., 2014)
Rubus glaucus Benth fruits		(Cerón et al., 2012)
Sambucus nigra L.		(Buchweitz et al., 2012)
<i>Tulipa gesneriana</i> L. 'Ben Van Zanten' variety, flowers		(Sagdic et al., 2013)
<i>Tulipa gesneriana</i> L. 'Leo Visser' variety, flowers		(Sagdic et al., 2013) (Sagdic et al., 2013)
<i>Tulipa gesneriana</i> L. 'Negrita' variety, flowers		(Sagdic et al., 2013)
<i>Tulipa gesneriana</i> L. 'Red Ring Hood' variety, flowers		(Sagdic et al., 2013) (Sagdic et al., 2013)
<i>Tulipa gesneriana</i> L. 'Yokohama' variety, flowers		(Sagdic et al., 2013) (Sagdic et al., 2013)
Tagetes erecta L. flowers		(Sivakumar et al., 2011)
Vaccinium ashei var. Rabbiteye fruits		(Jiménez-Aguilar et al., 2011)
Vaccinium pahalae Skottsb. shots		(Smith, Madhavi, Fang, &
vacemam panalae Skouso. shots		Tomczak, 1997)
<i>Vitis vinifera</i> L. marc		(Vatai et al., 2008)
Zea mays L. cv Zihei cob		(Yang & Zhai, 2010a)
Zea mays L. cv Heizhenzhu cob		(Yang & Zhai, 2010b)
		(Yang & Zhai, 2010a)
Zea mays L., cv Zihei grains		(Tailg & Zilai, 2010a)
Cyanidin 3-glucoside	Alashalia and usus alashalia harrana a	(Lange Counting & Varanalla
Pistacia lentiscus L. fruits	Alcoholic and non-alcoholic beverages,	(Longo, Scardino, & Vasapollo,
	frozen dairy desserts, candy, baked	2007)
Santalum album L. fruits	foods, and gelatin	(Harsha, Khan, Prabhakar, &
		Giridhar, 2013)
Commercial		(Masone & Chanforan, 2015)
Cyanidin 3-rutinoside		
Phillyrea latifolia L. fruits		(Longo et al., 2007)
<i>Rubia peregrina</i> L. fruits		(Longo et al., 2007)
Delphinidin 3-rutinoside		
Abies koreana E. H. Wilson		(Cabrita et al., 2000)
Solanum melongeana L. fruits		(Todaro et al., 2009)

Malvidin 3-glucoside	
Vaccinium spp.	(Cabrita et al., 2000)
Methyl pyrano-anthocyanidins	
<i>Vitis vinifera</i> L. skin	(Zhu et al., 2015)
Pelargonidin 3-glucoside	
Fragaria ananassa L. maximus	(Cabrita et al., 2000)
Commercial	(Kapadia, Balasubramanian,
	Tokuda, Iwashima, & Nishino,
	1997)
Peonidin 3-glucoside	
Commercial	(Masone & Chanforan, 2015)
<i>Oriza sativa</i> L.	(Cabrita et al., 2000)
Petunidin 3-glucoside	
Abies koreana E. H. Wilson	(Cabrita et al., 2000)

Table 6. Natural red-purple food colorants belonging to the beets class (E162).

Colorant	Source	Uses	Reference
Betacyanins	Hylocereus polyrhizus (Weber) Britton & Rose fruits	Burgers, desserts, ice cream,	(Stintzing et al., 2002)
Betalains	<i>Beta vulgaris</i> L. roots	jams, jellies, soups, sauces,	(Ravichandran et al., 2013)
	Opuntia ficus-indica [L.] Miller fruits	sweets, drinks, dairy	(Cassano et al., 2010)
		products, yogurts	(Otálora et al., 2015)
	Opuntia stricta (Haw.) Haw. fruits		(Obón et al., 2009)
	Rivina humilis L. fruits		(Khan & Giridhar, 2014)

 Table 7. Natural food colorants belonging to the carotenoids class, approved and under investigation.

Colorant	E code	Pigment color	Source	Uses	Reference
α-Carotene	E160a	Red-orange	Daucus carota L. roots	Butter and margarines, cakes,	(Sun & Temelli, 2006)
β-Carotene	E160a	Red-orange	Blakeslea trispora (fungus)	milk products and soft drinks	(K Nabae et al., 2005)
			Commercial		(Kohno et al., 2014;
					Martín et al., 2007)
			Daucus carota L. roots		(Sun & Temelli, 2006)
<i>trans</i> -β-Carotene	E160a	Red-orange	Commercial	Butter and margarines, cakes, milk products and soft drinks	(Kapadia et al., 1997)
Lutein (or xanthophylls)	E161b	Yellow-orange	Commercial	Dairy products, soft drinks, sugar confectionary, salads	(Martín et al., 2007; Sobral et al., 2016)
			Daucus carota L. roots	C 27	(Sun & Temelli, 2006)
			Tagetes erecta L. flowers		(Mejía, Loarca-Piña, &
					Ramos-Gómez, 1997)
			Tagetes spp. flowers		(Khalil et al., 2012)
Astaxanthin	E161j	Red-orange	Scenedesmus sp. (microalgae)	Food, nutraceutical and	(Grewe et al., 2007)
			Xanthophyllomyces	pharmaceutical industries	(Grewe et al., 2007)
			dendrorhous (yeast)		
			<i>Haematococcus pluvialis</i> (aquatic animal)		(Hong et al., 2009)
Under investigation			(
Crocetin	-	Yellow	Commercial	Jams and jellies, non- alcoholic drinks	(Ozaki et al., 2002; Wada et al., 2007)
			Crocus sativus L.		(Xi et al., 2007)
Crocin	-		Commercial		(Ozaki et al., 2002) (Wada et al., 2007)

Table 8. Natural food colorants under investigation that belong to phenolic compounds class.

Colorant	Source	Reference
Flavanones		
Naringin	Commercial	(Kapadia et al., 1997)
Flavones		· · ·
4',5,7-trihydroxyflavone	Commercial	(Kapadia et al., 1997)
Apigenin	Commercial	(Kapadia et al., 1997)
Flavonols		
Fisetin	Commercial	(Kapadia et al., 1997)
Myricetin	<i>Myrica cerifera</i> L. roots	(Kapadia et al., 1997)
Myricitrin	Myrica cerifera L.	(Kapadia et al., 1997)
Quercetin	Commercial	(Kapadia et al., 1997)
Rutin	Commercial	(Kapadia et al., 1997)
Other phenolic constituents		
Carthamin	Carthamus tinctorius L. flowers	(HX. Li et al., 2009)
	Commercial	(Kapadia et al., 1997)
Flavonoids and total phenolics	Senna bicapsularis L. flowers	(Mak et al., 2013)
Phenolics	Euterpe oleracea Mart. fruits	(Coïsson et al., 2005)
Phloridzin	Malus domestica Borkh. fruits	(Guyot, Serrand, Quéré,
		Sanoner, & Renard, 2007)
Pigment [*]	Cinnamomum burmannii (Nees & T. Nees) Blume fruits	(M. Tan et al., 2011)
Polyphenols	Mastisia cordata Bonpl. fruits	(Cerón, Ng, El-Halwagi, &
	-	Cardona, 2014)
Safflomin A	Commercial	(Kapadia et al., 1997)
Safflomin B		· -

* maybe a melanin or phenolic pigment; studied phenolic compounds presented a color spectrum varying from yellow-orange to red or even dark

purple

Colorant	E code	Pigment color	Source	Uses	Reference
Approved Annato (bixin E160b and/or norbixin)	E160b	Yellow- red	<i>Bixa orellana</i> L. seeds	Cakes, biscuits, rice, flour, soft drinks, smoked fish, sausages, meat products, dairy products, snack food, ice creams	(A. R. Agner, Barbisan, Scolastici, & Salvadori, 2004; Aniele R. Agner, Bazo, Ribeiro, & Salvadori, 2005; Anantharaman et al., 2015; Bautista, Moreira, Batista,
			Commercial		Miranda, & Gomes, 2004) (Kohno et al., 2014; Lima, Azevedo, Ribeiro, & Salvadori, 2003; Sobral et al., 2016)
Carminic acid	E120	Yellow to red-orange	Commercial	Cakes, cookies, beverages, jam, jelly, ice cream, sausages, pies, dried fish, yogurt, gelatins, cider, tomato, dairy products, cherries, non-carbonated drinks, chewing gum, pills and cough drops.	(Bibi et al., 2012; Huang et al., 2003, 2002; Masone & Chanforan, 2015)
Chlorophyll	E140	Green	<i>Spinacea oleracea</i> L. leaves	Beverages, fruit juices, pasta, dairy products, soups, sweeter	(Fernandes, Gomes, & Lanfer- Marquez, 2007)
Curcumin	E100	Yellow- orange	Spirulina pratensis (algae) Curcuma longa L. rhizomes	preparations Fish and baked products, dairy products, ice cream, yoghurts, yellow cakes, biscuits, sweets,	(Danesi et al., 2002) (Gómez-Estaca, Gavara, & Hernández-Muñoz, 2015; Silva et al., 2005)
			Commercial	cereals, sauces, gelatines	(Han & Yang, 2005; Kapadia et al., 1997; Maier et al., 2010;

Table 9. Other natural food colorants from plant origin, already approved and under investigation.

Under investigationC-Phycocyanin-BlueArthrospira platensis (cyanobacterium)Food, nutraceutical and pharmaceutical industries chewing gum, jellies(Martelli et al., 2014)C-Phycoerythrin-Red- orange (cyanobacteria)Blue-green algae (cyanobacteria)Food, nutraceutical industries chewing gum, jellies(Mishra et al., 2010)Fennel (freeze- dried)-Green Grey dried)Critimuum maritimum L. aerial partsPasta, sauces, rice, fish, meat and other preparations(Renna & Gonnella, 2012)Genipin-BlueCommercialBeverages, juices, nectars, deserts and gels(CC. Li et al., 2012; Wada al., 2007)Geniposide-YellowCommercial Rubia tinctorum L. rootsNoodles and confectioneries Confectionaries, hams, sausages, boiled fish, paste and beverages(Ozaki et al., 2002) (Ozaki et al., 2002)Madder color-RedRubia tinctorum L. rootsConfectionaries, hams, sausages, boiled fish, paste and beverages(Inoue, Shibutani, Masutom Toyoda, Takagi, Uneyama, 2008)						Martins et al., 2013; Masone & Chanforan, 2015; Yu Wang, Lu, Wu, & Lv, 2009)
C-Phycoerythrin-Red- orange (cyanobacterium) Blue-green algae (cyanobacteria)pharmaceutical industries chewing gum, jellies(Mishra et al., 2010)Fennel (freeze- dried)-Green 	Under investigation	on				
C-Phycoerythrin - Red- orange (cyanobacteria) Blue-green algae (cyanobacteria) chewing gum, jellies (Mishra et al., 2010) Fennel (freeze- dried) - Green Crithmum maritimum L. aerial parts Pasta, sauces, rice, fish, meat and other preparations (Renna & Gonnella, 2012) Fennel (hot-air dried) - Grey Blue Commercial Beverages, juices, nectars, desserts and gels (CC. Li et al., 2012; Wada al., 2007) Genipin - Blue Commercial Beverages, juices, nectars, fruits (Gao et al., 2014; Hou et al. 2008) Geniposide - Yellow Commercial Noodles and confectioneries (Ozaki et al., 2002) Madder color - Red Rubia tinctorum L. roots Confectionaries, hams, sausages, boiled fish, paste and beverages (Inoue, Shibutani, Masutom Toyoda, Takagi, Takahashi, al., 2008) Melanin - Black Auricularia auricular (mushroom) Not specified (Y. Zou, Xie, Fan, Gu, & H 2010)	C-Phycocyanin	-	Blue		·	(Martelli et al., 2014)
Fennel (freeze- dried)GreenCrithmum maritimum L. aerial partsPasta, sauces, rice, fish, meat and other preparations(Renna & Gonnella, 2012)Fennel (hot-air dried)-GreyGreyaerial partsand other preparations(CC. Li et al., 2012; Wada desserts and gelsGenipin-BlueCommercialBeverages, juices, nectars, desserts and gels(CC. Li et al., 2012; Wada al., 2007)Gardenia jasminoides Ellis fruits Genipa americana L. fruitsGao et al., 2014; Hou et al. 2008)(Gao et al., 2014; Hou et al. 2008)Geniposide-YellowCommercialNoodles and confectioneries Noodles and confectioneries(Ozaki et al., 2002)Madder color-RedRubia tinctorum L. rootsConfectionaries, hams, sausages, boiled fish, paste and beverages(Inoue, Shibutani, Masutorr Toyoda, Takagi, Takahashi, al., 2008)Melanin-BlackAuricularia auricular (mushroom)Not specified (Y. Zou, Xie, Fan, Gu, & H 2010)	C-Phycoerythrin	-		Blue-green algae	-	(Mishra et al., 2010)
dried) Genipin-BlueCommercialBeverages, juices, nectars, desserts and gels(CC. Li et al., 2012; Wada al., 2007)Gardenia jasminoides Ellis fruits Genipa americana L. fruitsGardenia jasminoides Ellis fruits Genipa americana L. fruits(Gao et al., 2014; Hou et al. 2008)Geniposide-YellowCommercialNoodles and confectioneries Noodles and confectioneries(Ozaki et al., 2002) (Ozaki et al., 2002)Madder color-RedRubia tinctorum L. rootsConfectionaries, hams, sausages, boiled fish, paste and beverages(Inoue, Shibutani, Masutorn Toyoda, Takagi, Uneyama, 2008)Melanin-BlackAuricularia auricular (mushroom)Not specified (Y. Zou, Xie, Fan, Gu, & H 2010)	· · · · · · · · · · · · · · · · · · ·	-	-	Crithmum maritimum L.		(Renna & Gonnella, 2012)
Image: Constraint of the sector of the se	· · · · · · · · · · · · · · · · · · ·	-	Grey			
fruits Genipa americana L. fruits2008) (Ramos-De-La-Peña et al., 2Geniposide Madder color-Yellow YellowCommercial CommercialNoodles and confectioneries Confectionaries, hams, sausages, boiled fish, paste and beverages(Ozaki et al., 2002) (Ozaki et al., 2002)Madder color-RedRubia tinctorum L. rootsConfectionaries, hams, sausages, boiled fish, paste and beverages(Inoue, Shibutani, Masutom Toyoda, Takagi, Takahashi, al., 2008)Melanin-BlackAuricularia auricular (mushroom)Not specified (Y. Zou, Xie, Fan, Gu, & H 2010)	/	-	Blue	Commercial		(CC. Li et al., 2012; Wada et al., 2007)
Melanin - Black <i>Auricularia auricular</i> Not specified (Y. Zou, Xie, Fan, Gu, & H (mushroom) (Inoue, Shibutani, Masutom Toyoda, Takagi, Uneyama, 2008) (Y. Zou, Xie, Fan, Gu, & H 2010)	1	-	Yellow	fruits <i>Genipa americana</i> L. fruits Commercial Commercial	Noodles and confectioneries Confectionaries, hams, sausages,	(Ramos-De-La-Peña et al., 2014) (Ozaki et al., 2002) (Ozaki et al., 2002) (Inoue, Shibutani, Masutomi, Toyoda, Takagi, Takahashi, et
	Melanin	_	Black		Not specified	 (Inoue, Shibutani, Masutomi, Toyoda, Takagi, Uneyama, et al., 2008) (Y. Zou, Xie, Fan, Gu, & Han,
	Monascorubrin	-	Red	· · · · · · · · · · · · · · · · · · ·	Meat, fish and other marine	,

				products, jam, ice-cream, sauces	
Purple corn color	-	Purple	Commercial	Beverages, jellies, and candies	(Kyoko Nabae et al., 200
Violacein	-	Violet	Chromobacterium	Yogurt and jelly	(Venil et al., 2015)
			<i>violaceum</i> UTM 5 (bacteria)		