
Reduction strategies for polycyclic aromatic hydrocarbons in processed foods

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) are a large group of carcinogenic compounds. PAHs are ubiquitous in the environment and food, thus human beings may be exposed to PAHs through ingestion (water and food), inhalation (air and smoking) and skin contact in daily life. Dietary intake is the major source of exposure to PAHs in humans. Significant and

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harmful levels of PAHs can be generated during food processing and cooking. Although the formation of PAHs during processing is almost unavoidable, the levels can be diminished with reduction strategies. This review aims to provide comprehensive insights into the mechanisms underlying the formation of PAHs and factors influencing their formation in processed foods. The strategy for the reduction of PAHs including change in ingredients (i.e. reducing fat content), pretreatment conditions (i.e. reducing the pH), processing methods and parameters (i.e. reducing processing temperature and time), and packaging and storage conditions, are discussed. Potential novel strategies for PAH reduction are also identified and the feasibility is evaluated.

Keywords: Polycyclic aromatic hydrocarbons; contamination; food processing; food safety;

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1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a large group of ubiquitous toxic compounds consisting of two or more fused aromatic rings (Trinquier et al., 2017). The most common PAHs present in the environment and dietary sources are those containing two to seven rings (Hamidi, Hajeb, Selamat and Abdull, 2016; Martorell et al., 2010). Based on the number of fused aromatic rings, PAHs are usually classified as small or low-molecular-weight PAHs (four or less rings) and large or high-molecular-weight PAHs

(more than five rings). Small PAHs are more water-soluble than large PAHs (Ferrarese, Andreottola, and Oprea, 2008). They are less toxic than large PAHs but more volatile and therefore more readily inhaled into human airways (Gao et al., 2016; Låg, Øvrevik, Refsnes and Holme, 2020). With an increased number of aromatic rings, large PAHs are more hydrophobic and have longer half-lives (Kanaly and Harayama, 2000). Although bacteria and fungi can biodegrade both small and large PAHs, the large PAHs are more recalcitrant to microbial degradation due to their increased hydrophobicity (Potin, Veignie and Rafin, 2004).

Humans can be exposed to PAH through ingestion (water and food), inhalation (air and smoking) and skin contact (Diggs et al., 2011; Lawal and Lawal, 2017; Qin, Hu, Yang, Liu and Gao, 2021;) with the major exposure route being dietary intake (Duan et al., 2016). In 2002, the Scientific Community on Food (SCF) defined 15 genotoxic PAHs (SCF 15 PAH) out of 33 investigated as the priority compounds in the assessment of PAHs dietary intake, and proposed that benzo[a]pyrene (BaP) could be used as the biomarker of exposure to PAHs from food and environment (SCF, 2002). In 2008, the European Food Safety Authority (EFSA) defined 16 priority PAHs (EU 15+1 PAH) in the assessment of PAHs dietary intake (Figure 1), which covers the SCF 15 PAH and the additional benzo[c]fluorene (BcF) as recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (EFSA, 2008). EFSA (2008) also suggested that both PAH4 (BaP, CHR, BaA, and BbF) and PAH8 (PAH4 + BkF, BgP, DahA, and IcP) are more suitable than BaP alone as the indicator

of PAHs in food, and the levels of PAH4 provides sufficient accuracy for PAHs assessment compared with PAH8 because of the similar limit of detection. Within the PAH4, BaP is carcinogenic to human (group 1) while the other 3 PAH4 were classified as possibly carcinogenic (group 2) by International Agency for Research on Cancer (IARC, 2010). Besides, the Environmental Protection Agency (EPA) of United States also listed 16 priority PAHs (EPA 16 PAH) in foods, including the PAH8 plus another 8 other small PAHs (Purcaro, Moret and Conte, 2013). Figure 1 illustrates the abbreviations, structures and carcinogenicities of the 24 PAHs covering EU 15+1 PAH and EPA 16 PAH as determined by IARC (2010).

Up to 98% of total human PAHs exposure can arise from dietary intake (Duan et al., 2016; Singh and Agarwal, 2018; Yu et al., 2011). The PAHs in food mainly occur from food contamination or are induced during food processing. Food raw materials such as cereals, vegetables, meat and seafood can be contaminated by PAHs through air, water and soil during their growth, while food high temperature processing operations such as smoking, frying, drying and grilling are additional sources of PAHs in our daily diet (Alomirah et al., 2011; Lee, Jeong, Park and Lee, 2018).

The existence of PAHs in food causes food safety issue and threatens human health. For effective reduction of contamination of food by PAHs, it is critical to understand the origin of PAHs including key factors affecting their formation during food processing. Therefore,

many scholars have continued to review the mechanisms of PAHs formation, factors influencing PAHs formation and reduction strategies based on food types (i.e. dairy products, meat, etc.) or processing methods (i.e. baking, frying, etc.) (Onopiuk et al., 2021; Singh et al., 2016, 2020). However, food production is an end-to-end program, and each step of it, from ingredients and pretreatments, to processing formulations and parameters, and to food packaging and storage conditions, could affect the PAHs levels in the final products. A systematic analysis of the causes and reduction strategies for PAHs formation along the entire chain of food product manufacture would provide a valuable reference for food researchers and food manufacturers. Moreover, a few novel strategies and potential applications on reducing PAHs levels in foods, such as PAHs degradation by ozonation and ultraviolet (UV) irradiation, PAHs adsorption by lactic acid bacteria (LAB) cellulosic aerogel, and PAHs reduction by using fuel catalysts and edible coatings, have been recently developed (Bartkiene et al., 2017; Chen and Chen, 2005; Kim, Han and Shin, 2021; Rozentale, Ancans, Bartkiene and Viksna, 2016; Yao et al., 2020). These strategies are important to consider for their potential in various processed foods. Thus, this review focuses on the research developments of the past decade and discusses the mechanisms of PAHs formation, factors and reduction measures on the PAHs levels in processed foods from an end-to-end perspective of food manufacturing. The potential of newly developed reduction strategies and identified opportunities are also summarized to advance the control of PAHs levels in

processed foods.

2 The mechanisms of PAHs formation in food processing and storage

The harm posed by PAHs is that they are potential carcinogens. The main mechanism of carcinogenesis by PAHs is that their metabolites generated in the liver and kidneys can bind with specific DNA fragments to induce gene mutation giving rise to various types of cancers, such as lung and gastrointestinal tract cancers (Diggs et al., 2011; Okona-Mensah, Battershill, Boobis and Fielder, 2005; Qin et al., 2021).

The formation of PAHs in food is an extremely complex process. Hydrogen abstraction and acetylene addition (HACA), Diels–Alder reaction, phenyl-addition-cyclization (PAC), and methyl addition/cyclization (MAC) are considered as the possible reaction routes of PAHs formation (Kislov, Sadovnikov and Mebel, 2013; Reizer, Csizmadia, Palotás, Viskolcz and Fiser, 2019; Shukla and Koshi, 2012). Although the precise mechanism of PAHs formation is still not specified, there are three possible mechanisms regarding the pyrolysis of hydrocarbons in food, and the combustion of cooking fuel and fat droplet on open flame are widely accepted as the leading cause of PAHs formation in processed food (Alomirah et al., 2011; Singh, Agarwal and Simal-Gandara, 2020; Wang, Xie et al., 2018).

Pyrolysis promotes oxidation of organic matters in food such as proteins, fats and

carbohydrates, and small PAHs are predominantly produced at around 200 to 300 °C (Chen and Chen, 2001; McGrath, Chan and Hajajigol, 2003). Although PAH formation is favored at the temperature range of 500-900 °C (Liu et al., 2019; Singh et al., 2020), it can also be induced at lower temperature by lipid oxidation during long-time storage. For example, a significant increase of PAH8 was determined in soybean and rapeseed oils stored at 25 °C for 270 days (Zhao, Gong and Wu, 2018) and beef patties stored under refrigeration for 9 days before barbecuing (García-Lomillo, Viegas, Gonzalez-SanJose and Ferreira, 2017).

The emission of smoke generated by the incomplete combustion of fuels (such as gas and charcoal) during cooking carries the PAHs to the surface of food material. Small PAHs are usually formed below 500 °C combustion (Wang, Wang and Herath, 2017). However, as the temperature and residence time increases, small PAHs and hydrocarbon radicals formed contribute to the formation of the larger PAHs (Raj, Prada, Amer and Chung, 2012; Wang et al., 2017; Zhang, Chen and Zhang, 2021). This explains the higher concentration of large PAHs in smoked food products than in grilled foods, since the latter have a shorter cooking time for PAHs exposure (Cheng, Zhang, Ma, Zhao and Tang, 2019; Kim, Cho and Jang, 2021).

PAHs from fat droplets are also carried by smoke and attach to the surface of food. Studies showed that the PAHs were significantly decreased once the contact of fat drops and heating source are removed (Lee, Kim, Moon, Kim, Kang and Yoon, 2016; Park, Pyo, Kim

and Yoon., 2017; Rose et al., 2015). This is because the higher temperature in the flame could accelerate the pyrolysis of the fat drops (Ghorbani, Najafi Saleh, Barjasteh-Askari, Nasserri and Davoudi, 2020). Formation of small PAHs predominate with this process as they are more volatile than large PAHs (Alomirah et al., 2011; Singh et al., 2020).

Apart from these, other factors such as pH and packaging are also associated with the levels of PAHs in food (Chen and Chen, 2005; Nie et al., 2018; Wongmaneepratip and Vangnai, 2017). Thus, food processing is considered to be the most important aspect for food safety with regard to PAHs level. The following sections assess the factors for the formation of PAHs and their reduction techniques in processed food, in terms of food ingredients and pretreatments, processing parameters, processing methods, and packaging and storage.

3 Factors influencing PAHs formation and common reduction techniques in processed food

3.1 Food ingredients and pretreatments

The levels of PAHs generated during processing vary from the types of food materials. Based on the first mechanism of PAHs formation (pyrolysis and oxidation of organic matters), the content of fat, protein and carbohydrate influence PAHs levels (Alomirah et al., 2011; Singh et al., 2020; Wang, Xie et al., 2018). Furthermore, other factors, such as antioxidant level of the food formulations or foods, casing types in some foods such as

sausages, moisture content and pH are also associated with the PAHs levels in processed food (Gomes, Santos, Almeida, Elias and Roseiro, 2013; Lu, Kuhnle and Cheng, 2018; Min, Patra and Shin, 2018; Wongmaneepratip and Vangnai, 2017).

3.1.1 Fats and oil

The mean exposure to PAHs for adults from fats and oil is 0.347 ng/kg bw/day, which accounts for 23.5% of PAHs in the diet (Veyrand et al., 2013). The pyrolysis of fat can form both small and large PAHs. Small PAHs, such as FL, PHE and PY, account for 52-88% of the total PAHs in fat-rich food products (Saito, Tanaka, Miyazaki and Tsuzaki, 2014). In such foods, large PAHs including PAH4 and PAH8 are readily detected (Alomirah et al., 2010; Saito et al., 2014). Due to the lipophilic nature of the PAHs, fat and oil act as their carriers and this accumulates them in both raw and processed foods (Cortazar et al., 2008; Lee, Suh and Yoon, 2019). As a result, PAHs contamination is very common in fat-rich foods such as meat, oil, fish (e.g. salmon), and in infant formula powder etc. (Ahmad Kamal, Selamat and Sanny, 2018; Cai, Wu, Zhou, Yang and Hu., 2020; Lee, Suh et al., 2019; Viegas, Novo, Pinto, Pinho and Ferreira, 2012).

Since fats and oil are the major carriers of PAHs, reducing the fat content in food materials is the most direct and frequently used method to reduce PAHs intake (Table 1). The PAHs reduction could achieve 9-95% depending on the type of processed foods, such as dry

fermented pork sausages (Gomes et al., 2013), UHT milk (Naccari et al., 2011), yogurt (Battisti, Girelli and Tarola, 2015), smoked Frankfurter-type sausages (Pöhlmann, Hitzel, Schwägele, Speer and Jira, 2013), and heated beef system (Min et al., 2018). The correlation coefficient between fat and PAHs levels in milk products ($r^2 = 0.645-0.659$) was much higher than that in meat product ($r^2 = 0.364$) (Kishikawa, Wada, Kuroda, Akiyama and Nakashima, 2003; Lu et al., 2018). Very effective reduction (95%) of PAHs levels can be achieved in yoghurt products simply by reducing the fat content from 3.9% to 0.1% (Battisti et al., 2015).

The utilization of low-fat alternatives is also an option to reduce PAHs in other processed foods, and a few examples are given in Table 1. However, this method does not always work. For example, in the study of meat and blood sausages, Santos, Gomes and Roseiro (2011) observed that the higher fat content of blood sausages resulted in higher PAHs levels compared to meat sausages. However, the *Cacholeira* blood sausages (41.8% of fat) had nearly double amount of total PAHs compared with that of *Morecla* blood sausages (46.4% of fat), and the former experienced a shorter smoking time. This finding could be due to differences in the formulation (not available in the study), as other ingredients such as proteins and carbohydrates beside fat content could affect PAHs content during smoking (Alomirah et al., 2011; Singh et al., 2020; Wang, Xie et al., 2018). Moreover, the composition of fatty acids in the fat also affects the levels of PAHs. Liu et al. (2019) reported that the number of carbons and the degree of unsaturation in fatty acids from animal source

fat enhanced the generation of PAHs during food processing.

Although using low-fat raw materials or alternatives in processed dairy and meat products significantly contributes to the lower PAHs levels, such approaches may cause unacceptable changes in the organoleptic properties such as undesirable color, flavor and taste (Chetachukwu, Thongraung and Yupanqui, 2018; Jiménez Colmenero, 2000).

In contrast to fat reduction, using vegetable oil in lieu of animal fat in pork patties to reduce PAHs formation has less impact on the sensory attributes (Lu, Kuhnle and Cheng, 2017). By replacing 40% of the pork fat with olive oil, the PAHs levels of pork patties decreased by 7.75% and 51.52% at the cooking temperature of 180 and 220 °C, respectively (Table 1). Sunflower oil and grape seed oil have also been shown to decrease PAHs, where 21.71% and 17.38% reduction at 180 °C and 220 °C were observed, respectively. The discrepancy in PAHs reduction compared to olive oil could result from olive oil having a higher smoking point (242 °C) than sunflower oil (227 °C) and grape seed oil (216 °C) (Lu et al., 2017); thus it is more stable during cooking especially at the higher temperature. More importantly, the replacement with these three types of plant oils had no significant effect ($p > 0.05$) on pH, hardness, springiness, chewiness, and color of the pork patties, suggesting retention of good sensory attributes of the patties. However, the limitation of this method could be that it is only suitable for comminuted meat products such as sausage, mince, patties etc., rather than intact meat products such as steaks and chicken wings.

3.1.2 Proteins and amino acids

Proteins and amino acids are also considered significant factors in the formation of PAHs. A strong association between protein oxidation and the formation of PAHs was found in deep-fried meat systems (Lu et al., 2018). The total PAHs levels (BaA and BaP) in sixteen types of deep-fried beef and chicken meatballs (approximate protein content of 24 % and 20% respectively) were measured. The correlation coefficient of protein carbonyl value (protein oxidation) to total PAHs was 0.598 ($p < 0.05$) followed by thiobarbituric acid-reactive substances (TBARS; lipid oxidation) value (0.364; $p < 0.05$), suggesting that both protein and lipid oxidation contributed to the PAHs levels in the tested foods.

However, the relationship between protein content and PAHs formation in processed food has not been extensively studied (Table 1). Nie et al. (2018) showed that the addition of amino acids (0.5 g/100 g) in grilled sausage increased the PAHs levels to 166% above control and the levels were dependent on the amino acid types. Although there was no apparent correlation between the PAHs content and the polarity of amino acids, basic amino acids, however, showed a greater impact ($p < 0.05$) on the formation of PAHs compared with acidic amino acids. The authors suggested that the increased pH by added basic amino acids promoted the Maillard reaction between the free amino acids and reduced sugars, which facilitated PAHs formation. Interestingly, they found that the amino acids consisting of

benzene ring, such as L-tryptophan, L-phenylalanine and L-tyrosine, led to lower PAHs formation, in which the structural stability of the benzene ring limited their cleavage into small molecules, such as aldehydes, enynes and others at a high cooking temperature.

Although protein is a main component of animal food product, most studies on high protein food concluded that it has less significance to PAHs formation than that of fat (Liu et al., 2019; Saito et al., 2014). However, due to the lack of research regarding the impact of protein types on PAHs formation, such as whey and gluten proteins, the correlation between protein and PAHs formation warrants further investigation.

3.1.3 Carbohydrates

Similar to proteins and amino acids, the effect of carbohydrates on the formation of PAHs in processed food has not been extensively studied (Table 1). In terms of individual carbohydrates, the effects of cellulose, pectin, D-glucose, and sucrose have been investigated (McGrath et al., 2001; McGrath, Chan and Hajajigol, 2003). Under the heating temperature range of 300-600 °C, most small PAHs, such as FL, PHE and AN were generated in the presence of these carbohydrates. Moreover, the total PAHs levels was found to have a positive relationship with the temperature, residence time and the particle size of the carbohydrate, which are the critical conditions that determine the formation of large PAHs (such as BaP) (McGrath et al., 2001; McGrath et al., 2003).

In a complex meat system, the aldehyde groups in reducing sugars such as D-glucose could contribute to the PAHs formation through complex reactions (Nie et al., 2018). With the addition of 0.5 g/100 g D-glucose, the concentration of PAHs in the baked pork sausages cooked in an electric oven at 240 °C for 20 min were significantly increased by 120% ($p < 0.05$). The study also indicated that the effect of the aldehyde group (in glucose) and the keto group (in fructose) are more critical parameters than molecular weight of carbohydrates on PAHs formation. Therefore, the addition of simple sugars would have more impact than addition of complex carbohydrates, such as starch, on the yield of PAHs in food processing.

The PAHs levels of various common sugars have also been reported. By investigating 57 sugar products, Tfouni and Toledo (2007) found white crystal sugar contained the highest level of PAHs, while their levels in granulated white sugar, demerara sugar (a type of raw cane sugar), brown sugar, and organic sugar (including brown, demerara and refined sugars) was 86, 93, 50, and 74% lower, respectively. They proposed that the refining procedure of sugar could reduce PAHs level, so the PAHs levels in granulated white sugar refined twice by floatation method was lower than that of crystal sugar refined once. The unexpected highest PAHs levels in crystal sugar, comparing with demerara and brown sugar, may due to the high proportion of burnt sugar cane used in processing (Tfouni and Toledo, 2007). With respect to impact of the production process on PAHs exposure, the white sugar seems to be a better choice compared to the non-refined brown sugar.

As carbohydrate-rich products, cereals are also identified as a dietary source of PAHs due to their high intake (Omodara, Amoko and Ojo, 2014; Zelinkova and Wenzl, 2015). However, apart from the impact of processing, such as baking, toasting etc., the main cause of the PAHs in cereals is the contamination of raw materials by environmental factors such as water and soil (Ciecierska and Obiedziński, 2013; Dennis et al., 1991).

Carbohydrate is obviously a factor in the levels of PAHs in processed foods, but its importance in relation to fat and oil in this aspect is unknown due to limited studies. According to SCF (2002), cereals contribute 36% daily intake of BaP, while oils account for 47%. However, since the total large PAHs formed due to presence of carbohydrates are relatively low, food products with high carbohydrate content should have minimal effects for consideration of dietary PAHs intake, despite their high levels of consumption (Ciecierska and Obiedziński, 2013).

3.1.4 Antioxidants and antioxidant-rich ingredients

The impact of antioxidants and antioxidant-rich ingredients on PAHs levels in processed foods has been well documented (Table 1). In the study of fried peanut, the existence of peanut skin (rich in antioxidants) and the addition of tert-butylhydroquinone (TBHQ, a commercial antioxidant) at the concentration of 60 mg/kg reduced PAHs levels of up to 22.63% and 71.75%, respectively (Zhao, Wu, Gong, Li and Zhuang, 2017). The addition of

30g/100g of onion (also rich in antioxidants) reduced PAHs levels of up to 67% and 91% were reported for meat and gravy, respectively, whereas addition of 15g/100g of garlic reduced them by 41-66% in meat and 14-79.4% in gravy (Janoszka, 2011).

The study of charcoal-grilled sirloin pork (Wongmaneepratip, Jom and Vangnai, 2019) and Chinese oil fried bread (Gong, Zhao and Wu, 2018) showed that the reduction of PAHs did not have an obvious association with the concentration of the added antioxidants or antioxidative ingredients. Lu et al. (2018) indicated that *Trolox* equivalent antioxidant capacity (determined by ABTS assay) of the spices was the most important index ($p < 0.05$) on the inhibition of PAHs ($r = 0.647$) followed by total phenolic content ($r = 0.507$; $p < 0.05$) and oxygen radical antioxidant capacity ($r = 0.238$; $p > 0.05$). In their study, the effect of addition of onion, garlic, red chili, paprika, black pepper and ginger on the reduction of PAHs levels in meat products has been compared. With addition of 5g/kg of each spice, the PAHs content of deep-fried beef and chicken meatballs reduced by 65/86%, 57/86%, 65/79%, 87/74%, 47/97%, and 98/97%, respectively, in the above spice order (Lu et al., 2018).

In the study of Esfahani Mehr, Hosseini and Seyadain Ardebili (2019), nutmeg oil, ginger oil and their nanoemulsions at the concentration of 0.02 and 0.04% were added to beef patties and their effect on the formation of PAH8 in grilled beef patties during storage was investigated. The PAH8 reduction rate of oils after 90-days storage was consistent with the reduction of TBARS value and the radical scavenging antioxidant activity, and the highest

reduction was found in the sample with the 0.04% addition of nanoemulsified nutmeg oil. Since the smaller particle size of nanoemulsified oils promoted stronger radical scavenging capacity, their PAHs reduction efficiency is 6 to 12% higher than the non-nanoemulsified oils (Esfahani Mehr et al., 2019). This indicated that nano-sized antioxidants or antioxidative ingredients could have a better inhibition activity against PAHs formation, which is worthy of further investigation on other common antioxidants in nano-size, such as EGCG.

Nevertheless, two studies showed that the PAHs reduction rate was not consistent with radical scavenger activity of added antioxidative ingredients. For example, the nonalcoholic Pilsner beer had a higher radical-scavenging activity measured with DPPH assay than alcoholic Pilsner beer, but resulted in lower PAHs reduction in grilled pork when used as the marinades (Viegas, Yebra-Pimentel, Martínez-Carballo, Simal-Gandara and Ferreira, 2014). Similarly, in another study on barbecued beef patties, the addition of red wine pomace seasoning resulted in a significantly higher PAHs content (16.63 ng/g; $p < 0.05$) in the cooked sample compared to the control group (9.67 ng/g) (García-Lomillo, Viegas, Gonzalez-SanJose and Ferreira, 2017). It is because the untreated raw beef patties had a higher radical scavenger activity (2.98 $\mu\text{mol/g}$), measured by ABTS assay, than the treated group (1.99 $\mu\text{mol/g}$). They indicated that the reduced antioxidant activity by the addition of red wine pomace seasoning could be caused by the interaction between the antioxidants in the seasoning and proteins of the beef patties (García-Lomillo et al., 2017). This would suggest

that further studies should consider the antioxidant activity of whole food materials rather than the added ingredients only.

The above studies (Esfahani Mehr et al., 2019; García-Lomillo et al., 2017) showed that the addition of antioxidative ingredients could reduce the PAHs formation in meat products during storage. However, an opposite effect was observed by the addition of grape seed extract which caused a decrease in radical scavenging activity measured by both DPPH and ABTS assay and increases in the PAHs levels and TBARS value of grilled pork sausage cooked after up to 8-days storage in the sealed opaque polyethylene plastic bag (Nie et al., 2020). Although the PAHs content of the grilled sample was significantly lower ($p < 0.05$) than the control sample at day 0-4, the situation was significantly reversed on day 8 ($p < 0.05$). In comparison with the above studies (Esfahani Mehr et al., 2019; García-Lomillo et al., 2017) in which the products were cooked before storage, sausages in this research were cooked after storage. The authors explained that the added antioxidants were self-oxidized after 4 days, and this autooxidation resulted in higher PAHs formation at the end of storage (Nie et al., 2020). This would imply that products with added antioxidant ingredients need to be processed and consumed before undesirable quality changes occur. The antioxidant activities of ingredients against food PAHs formation during storage need further investigation.

3.1.5 Casing types for sausages

Casing is an essential material in the processing of meat sausages which may affect the PAHs levels (Table 1). Replacing sheep intestine casing with cellulose casing could reduce PAHs by 78% (Youssef, Abou- EL-Hawa, Hussein, and Mahmoud. 2016). Collagen casing is also recognized as a good substitute for natural animal intestine casing in reducing PAHs in sausages (García-Falcón and Simal-Gándara, 2005; Gomes et al., 2013; Mastanjević et al., 2019). The effect of cellulose and collagen casing on the reduction of PAHs is believed to be due to the absence of fats in the casings, and the smoother surface and lower porosity of these casings prevents PAHs contamination, adsorption and penetration from external source, such as smoke, into sausages (García-Falcón and Simal-Gándara, 2005; Gomes et al., 2013; Ledesma, Rendueles and Díaz, 2015).

Casings are a good barrier against PAHs penetrating from exterior into the inner layers, as up to 90% of the total PAHs have been found to deposit on the surface of sausage casings (García-Falcón and Simal-Gándara, 2005; Gomes et al., 2013; Santos et al., 2011). Therefore, Gomes et al.(2013) suggested that it is better to remove the casing before sausage consumption to reduce PAHs exposure.

3.1.6 Water content and pH

Water content is another factor affecting PAHs formation in foods. Studies showed that

the higher the water content in raw materials, the lower the PAHs levels in the processed products (Table 1). For example, up to 62% reduction of PAHs levels in ground beef heated to 80 °C was achieved by the addition of an equal weight of water into the meat mince, and the reduction rate increased up to 72% when the heating temperature raised to 200 °C (Min et al., 2018). In another study, thawing of frozen chicken in the fridge produced lower levels of PAHs than thawing by microwave or water immersion, when the thawed chickens were cooked by air-frying or deep-fried (Lee et al., 2020). These differences were related to the higher water retention in fridge thawed chicken compared to the other thawing methods. These two studies indicated that water is an important conduit for oxygen in limiting incomplete combustion of organic materials during cooking of the meat, which is a pathway for PAHs formation (Lee et al., 2020; Min et al., 2018).

pH of meat is also a controllable factor influencing PAHs formation (Table 1). Higher PAHs levels (1781 µg/kg) were detected in the grilled chicken marinated with 30 g/kg of sodium bicarbonate compared to the sample marinated with 0.21 g/kg of citric acid (535.2µg/kg) at the treatment pH of 7.51 and 3.62, respectively (Wongmaneepratip and Vangnai, 2017). This is because the increased pH enhanced the Maillard reaction which further promoted the PAHs formation (Nie et al., 2018, Wongmaneepratip and Vangnai, 2017). Although the studies on the effect of pH on the formation of PAHs in processed food products are limited, the utilization of acid marinade of raw food material appears to be a

promising method to reduce PAHs formation.

3.2 Food processing methods and processing parameters

3.2.1 Processing methods

The levels of PAHs in processed food is largely determined by the processing methods and operating conditions (Table 2). Compared to the raw material, grilling could increase PAHs levels by 3.43 times (Cheng et al., 2019). Grilling generated more PAHs than roasting in beef and pork (Chung et al., 2011), and barbecued beef had higher PAHs content than that of pan-fried (Kılıç Büyükkurt, Aykın Dinçer, Burak Çam, Candal and Erbas, 2020). Among the reported processing methods, smoking and boiling generate the most and least PAHs in food products, respectively (Manda et al., 2012; Olatunji, Fatoki, Opeolu and Ximba, 2014; Onwukeme and Okafor, 2015).

Due to the distinct nature of these processing methods, we can say that the processing method macroscopically affects the PAHs formation while the processing parameters such as processing time and temperature could be the microscopic factors. PAHs formation is also dependent on extraneous factors such as smoke from the combustion of fuel and fat droplets generated during cooking or processing.

3.2.2 Processing time

Studies have indicated that processing time directly affects the PAHs levels in processed food (Table 3). Rose et al. (2015) investigated the impact of cooking time on PAHs contamination in various fried, grilled, barbecued, toasted and roasted animal and plant food products, and the increasing levels of PAHs was observed in most products when cooking time was extended by 50%. This increase is thought to result from the extended time which allows more PAHs to be formed and accumulated in the products (Hao, Li and Yao, 2016). A similar result was also found in ground beef heated in an aluminum heating block, in which PAHs levels gradually increased with the extended heating time (Min et al., 2018). Among the studies regarding the impact of decreased processing time on PAHs levels (Table 3), the largest reduction of PAHs was found in roasted ground *Arabica* coffee (72% reduction) by reducing roasting time from 20 to 5 min (Houessou et al., 2007) and sugar-smoked seafood (100% reduction) by reducing smoking time from 6 to 3 min (Chen, Kao, Chen, Huang and Chen, 2013). Conversely, Simko, Gergely, Karovicova, Drdak and Knezo (1993) showed that extending the boiling time by 5 min (from 57 to 62 min) resulted in a 38% reduction of PAHs levels in pre-smoked sausages. They proposed that the longer boiling treatment cooked the fat out and carried the PAHs out of the sausage into the boiling water (Simko et al., 1993). It has also been suggested that meat products could be pre-boiled to partially-cooked to shorten the heating/cooking time of the subsequent high temperature cooking, such as grilling and roasting, which would result in reduced PAHs levels (Omodara et al., 2014). Moreover,

boiling was found to enhance the texture properties (reduced hardness, tenderness and shear force) and retain the sensory attributes (such as taste, flavor, color and overall acceptability) of a cooked Chinese meat dish of pork belly and chicken drumstick (Wang, Zhang, Fan, Yang and Fang, 2018). Therefore, the combination of boiling and common cooking methods could be a potential solution to improve eating quality and safety for meat products with regards to PAHs levels.

3.2.3 Processing temperature

High temperature induces the pyrolysis of organic compounds, therefore PAHs formation in foods. As documented in Table 3, lower cooking temperature reduces the PAHs levels in various processed food products, and 63-71% reduction was reported for baked bread (Ciecierska and Obiedziński, 2013), smoked salmon (Duedahl-Olesen et al., 2010) and gas-grilled beef satay (Ahmad Kamal et al., 2018).

Although reducing the cooking temperature (or time) could reduce the PAHs levels in processed food, the processing temperature (or time) needs to be adequate to achieve safe products. Usually, when the processing temperature is decreased, the time should be raised to achieve well-cooked food, and vice versa being decreased time requires increased temperature. It is of interest to understand whether, and to what degree, the decreased temperature/time could offset the impact of increased time/temperature on the PAHs

formation in processed food. For example, by changing the heating temperature from 120 to 80 °C for ground beef, the maximum reduction in PAHs levels was 32% when cooked for 25 min, compared to 15, 20, and 30 min (Min et al., 2018). In the study of Houessou et al. (2007), PAHs content of roasted coffee was 19.81 µg/kg when roasted at 200 °C for 20 min, which is lower than that roasted at 220 °C for 5 min (20.29 µg/kg). Min et al. (2008) indicated that temperature has a greater impact on PAHs formation than time in heated ground beef, but this is not convincing as the Pearson correlation coefficients among temperature, time and PAH content was not reported in their study. As there is no research particularly focusing on the combined effect of temperature and time on PAHs formation in processed food, the correlation between temperature and time are not clear and warrant further investigation. Understanding this relationship would contribute to optimum settings for processing temperature and time to minimize the PAHs formation in processed foods.

3.2.4 The extent of exposure of food materials to a heating source

The exposure of food materials to smoke generated by the combustion of fuel and fat drops during direct heat processing significantly increases the PAHs levels. Lee et al. (2016) showed that the PAHs concentration in grilled meat product can be decreased up to 89% by collecting the fat droplets in a beaker under the meat, and up to 74% by preventing the direct contact of the meat with smoke by removal with a ventilation duct. Moreover, the reduction

rate was greater with higher fat content food as smoke favorably adheres to fatty materials (Duedahl-Olesen et al., 2010; Lee et al., 2016).

In order to reduce PAHs formation in processed foods, especially in smoking products, minimizing the exposure of food to a heating source is recommended, which includes a greater distance from the heat source and the use of indirect heating (Table 3). The levels of PAHs in processed food is inversely associated with the distance from the heating source. For example, the concentration of PAH4 in beef burgers and sausages cooked 4 cm above the wood chips and charcoal was higher than when cooked 7 and 9 cm above (Rose et al., 2015). Similarly, significant reduction of PAHs ($p < 0.05$) was achieved in smoked cheese (68%) and smoked sausages (50 and 51% with natural and collagen casing, respectively) placed at farther distance from the smoking source (Guillén et al., 2011; Mastanjević et al., 2019). Nevertheless, an opposite result was observed in another study on sausages smoked by a smoke generator located underneath. The concentration of EPA 16 PAH of a ‘superior’ sample (furthest from the smoking source; 1608.17 $\mu\text{g/kg}$) was much higher than that of an ‘inferior’ sample (closest to the smoking source; 356.66 $\mu\text{g/kg}$) (Roseiro, Gomes and Santos, 2011). Further analysis indicated that the volatile small PAHs accounted for 99.4% of the total PAHs, which are carried by the upward smoke and accumulate on the surface of the sausages (Roseiro et al., 2011).

Indirect heating refers to placing an obstacle between the heating source and food

material such as wrapping, and for smoking using a smoking source external to the smokehouse (Duedahl-Olesen et al., 2010; Farhadian, Jinap, Hanifah and Zaidul, 2011; Gomes et al., 2013). Toasting of bread wrapped in aluminum foil diminished PAHs levels by 99.5 compared to the sample directly cooked on the flame (Rey-Salgueiro, García-Falcón, Martínez-Carballo and Simal-Gándara 2008). Interestingly, wrapping food in banana leaf also reduced PAHs levels by 40 and 80% in grilled beef and chicken, respectively, which compares favorably with the 46 and 80% reduction, respectively, using standard aluminum wrapping (Farhadian et al., 2011). In addition, using non-flame methods for preheating of chicken, such as steaming and microwave, resulted in up to 63% reduction of PAHs levels compared to no preheating when the chicken was grilled (Farhadian et al., 2011). Using an external smoke generator and then filtering the smoke by removing the particles in the gas, resulted in up to 72% reduction in smoked rainbow trout (Mihalca, Tița, Tița and Mihalca, 2011). Moreover, the greater fat content in the fish resulted in higher PAHs formation inhibition by using this indirect smoking method (Duedahl-Olesen et al., 2010).

In addition, preheating charcoal to avoid exposure of foods to the initial wave of smoke is another effective method to prevent PAHs formation arising from smoking source. In the study by Chaemsai et al., (2016), the highest PAHs concentration (19.86 $\mu\text{g/g}$) was determined in the smoke from the partially glowed charcoal which was preheated for 5 min at 500 °C, while the lowest PAHs concentration (0.15 $\mu\text{g/g}$) was in the smoke of charcoal

preheated for 5 h at 1000 °C which was completely burnt. A low burning temperature for a short time resulted in the incomplete combustion of charcoal which promoted the formation of PAHs. Thus, a longer preheating time for the charcoal at a higher temperature was recommended to reduce PAHs generation from smoke (Chaemsai et al., 2016).

3.2.5 Type of heating source

Because of the generation of smoke during fuel combustion, foods processed by open-flame heating sources such as gas and charcoal are usually associated with high PAHs content (Rey-Salgueiro et al., 2008). Thus, non-smoke generation heating, such as infrared-ray and electric heating, are recommended food processing methods for PAHs reduction (Table 3). For example, the PAHs content decreased 94% by using electric heating instead of wood burning for the drying of rice (Bertinetti, Ferreira, Monks, Sanches-Filho and Elias, 2018). Additionally, in the processing of Peking duck, a Chinese delicacy, the PAHs contents in the skin and lean meat of the duck were remarkably lower when cooked by electric oven grilling (25.8 µg/kg and 1.1 µg/kg, respectively) rather than by wood fire grilling (129 µg/kg and 2.1 µg/kg, respectively) (Lin, Weigel, Tang, Schulz and Shen, 2011). Moreover, 6 large PAHs including the group 1 carcinogen BaP, which were detected in the wood fire grilled duck skin, were not detected in the electrical oven-grilled duck skin (Lin et al., 2011). Likewise, BaP was detected in charcoal grilled pork belly at a level of 8.04 µg/kg,

but not detected in infrared grilled nor electric grilled pork belly (Park et al., 2017).

In terms of the open-flame heating source, gas was widely recognized as a better fuel than charcoal and wood to reduce PAHs generation (Ghorbani et al., 2020; Omodara et al., 2014). Since the flame is generated and spread by gas nozzles, flame-free spaces are formed between each nozzle (small gas flame), which reduces the possibility of flame contact with fat droplets compared with the entitle flame arising from charcoal and wood (Ghorbani et al., 2020). As shown in Table 3, by using gas instead of charcoal in the processing, the PAHs content dropped 88% in dried rice (Bertinetti et al., 2018), 76% in grilled Turkish kebab (Terzi, Gelik and Nisbet, 2008), and 25% in grilled Iranian kebab (Gorji et al., 2016).

The effects of different fuels (e.g. wood and charcoal types) on the food PAHs are also summarized in Table 3. The PAH content in grilled meat using disposable charcoal was 1.6-fold higher than using wood as fuel (Reinik et al., 2007). In relation to charcoal types, white charcoal generated fewer PAHs in grilled beef, pork and chicken than black and extruded charcoal (Kim, Cho et al., 2021), and coconut charcoal generated fewer PAHs in grilled beef and salmon than wood charcoal (Viegas et al., 2012). These results suggested that during charcoal combustion, the less flame and smoke produced, the fewer the PAHs formed (Viegas et al., 2012).

Wood with higher resin content should be avoided as the fuel for smoking, which could cause intensive soot during combustion and then pollute the food material with PAHs

(Stumpe-Viksna, Bartkevičs, Kukare and Morozovs, 2008). Apple wood generated 90% lower PAHs than spruce wood, which was directly related to their resin content (Stumpe-Viksna et al., 2008). In another study, smoking using smoldering poplar and hickory wood reduced PAHs content by 35-55% in frankfurters and mini-salamis, respectively, compared with the commonly used beech wood (Hitzel, Pöhlmann, Schwägele, Speer and Jira, 2013).

The above research indicated that to minimize the contamination of food with PAHs from various fuels, the preference should be (most to least preferred): non-flame cooking fuel such as infrared-ray and electric heating sources, gas fuels, charcoal and low resin wood.

3.2.6 Cooking oil type

Cooking oils used for deep-frying, pan-frying and pan-grilling acts as a heat transfer medium and contribute to food texture and flavour (Choe and Min, 2007), which also significantly influences the formation of PAHs. Four recommendations have been proposed for the appropriate use of cooking oil for PAHs reduction (Table 3).

Firstly, using a non-oil cooking method, such as air-frying, could reduce 90% of PAHs content in French fries compared to deep-frying (Andrés, Arguelles, Castelló and Heredia, 2013). Without addition of external oil, the PAHs concentration significantly declined by 21% in all air-fried chicken meat compared to the deep-fried samples (Lee et al., 2020).

Secondly, refined oil was used to minimize the primary contamination of PAHs from the oil itself (Hua, Zhao, Wu and Li, 2016). In respect to the oil refining procedures, deodorization effectively reduced small PAHs, and acid neutralization and bleaching with active charcoal contributed to the reduction of large PAHs (Teixeira, Casal and Oliveira, 2007). Based on the results of previous studies (Table 3), the PAHs content in refined sunflower, soybean, olive and rapeseed oil are 71, 88, 87, and 60% less compared with their crude oils (Hua et al., 2016; Rojo Camargo, Antonioli and Vicente, 2012; Teixeira et al., 2007). The negative effect of oil refining is that the oil's antioxidants are also removed so that PAHs could increase during storage, although the total PAHs is still 23-43% less than that of the crude oil regardless of storage temperature (Zhao et al., 2018).

Thirdly, using appropriate oil could avoid extra PAHs generation during cooking. The higher the smoke point of the oil, the more suitable it is for high temperature processing as less smoke will be produced (Sarwar, Vunguturi and Aneesha, 2016). For example, since the smoke points of brown rice oil, sesame oil and perilla oil are 257, 165 and 161 °C, respectively, reducing the ratio of sesame oil and perilla oil and increasing the ratio of brown rice oil in the three oil mix resulted in up to 21 and 32% reduction, respectively, of PAHs at 380 °C in roasted laver (Kang et al., 2019). Olatunji, Fatoki, Ximba and Opeolu (2014) also indicated that the aromatization and dehydrocyclization of mono-unsaturated fatty acids could contribute to the PAHs formation in oil during high temperature cooking.

Lastly, it is important to avoid the continual re-use of cooking oil (Olatunji, Fatoki, Ximba, et al., 2014; Singh et al., 2020). As discussed above, the PAHs content in rapeseed, soybean, peanut and olive oil was increased with the extension of frying time (Hao et al., 2016). Abundant PAHs would have been formed in the first oil frying round, which would increase the PAHs levels in the next round of frying. Therefore, discarding the used oil, regularly renewing the oil, and applying suitable frying conditions (i.e. temperature and time) and oil type to reduce the PAHs levels in the fried food products is critical.

3.3 Food packaging materials and storage temperature and time

3.3.1 Food packaging materials

Food packaging materials also influence the PAHs content in processed foods (Table 4). The utilization of low-density polyethylene (LDPE) package was found to reduce PAHs (BaA, BbFA, and BaP) by 50% in roasted duck after 24 h through the adsorption of PAHs by the packaging (Chen and Chen, 2005). The authors reported that the PAHs adsorption with plastic was more effective for the duck meat in a water-oil system containing distilled water and propylene alcohol, as the non-polar PAHs can easily migrate from polar water to the non-polar LDPE. Four other packaging plastics beside LDPE were also found to adsorb PAHs in smoked sprats, and their ability to adsorb PAHs was ranked in the order (strongest) of high-density polyethylene (HDPE), LDPE, polypropylene (PP), oxo-degradable (FO) and

polyethylene terephthalate (PET) (Kuzmicz and Ciemniak, 2018).

However, the polar adsorption of plastic packaging to PAHs may not be reliable. Firstly, when the food material (e.g. non-polar oil in fish) has a similar polarity to PAHs or a stronger adsorbing ability than the plastics, the ability of the plastic to remove the PAHs from the food could be limited. In the above study of Kuzmicz and Ciemniak (2018), it should be noted that the amount of PAHs adsorbed by plastics was less than that retained in the fish samples at the end of storage. For example, the BaP level in HDPE and fish skin was 0.1 and 2.3 µg/kg, respectively. Secondly, the plastic packaging could adsorb PAHs from external sources, such as polluted air, which may transfer into the foods. Li, Ni and Zeng et al. (2017), for example, indicated that the plastic packaging (polystyrene) could be a potential medium for migration of PAHs from a contaminated source to food materials.

Despite the PAHs adsorption by plastic materials, the efficiency of applying such materials on PAHs reduction in foods is still equivocal. Therefore, the usage of plastic packaging for PAHs reduction in different types of food (such as liquid and solid) with different PAHs contamination levels needs further investigation.

3.3.2 Storage time and temperature

Storage is an important factor for food safety and quality. Generally, a lower storage temperature is associated with reduced risk of PAHs contamination (Table 4). For example,

storing at 4 °C reduced PAHs generation by 13-22 and 26-31% due to the reduced oxidation of crude and refined oils, respectively (Zhao et al., 2018). Moreover, a lower storage temperature allowed plastic packaging materials (such as LDPE, PP, PET and FO) to absorb more PAHs from smoked fish (Kuzmicz and Ciemniak, 2018), but increased storage temperature could promote PAHs migrating from contaminated polystyrene packaging to the foods (Li et al., 2017).

Storage time has different impacts on the PAHs content, depending on the type of food. For smoked products such as smoked fish and beef sausages, the PAHs content was decreased as the length of storage time increased (Table 4). For example, after 7-days storage with exposure to oxygen and daylight, a 70% reduction in BaP concentration was observed in smoked fish (Simko, 1991). The total EPA 16 PAH in smoked beef sausages decreased to 0.0026 mg/kg from 2691 mg/kg with sheep intestine casing and 599 mg/kg with cellulose casing (Youssef et al., 2016). During storage, the PAHs can be decomposed by light and interact with other components such as antioxidants (Simko, 1991; Škaljac et al., 2014). Thus the highest content of PAHs in food is usually found immediately after the smoking process (Škaljac et al., 2014; Youssef et al., 2016). However, PAHs could penetrate from the outer layer into the inner layer of the sausages during storage and become less available to be decomposed (Simko, 2005; Škaljac et al., 2014).

In contrast, the PAHs content was significantly increased in soybean and rapeseed oils

after storage at 4 and 25 °C for 270 days (Zhao et al., 2018). The authors proposed that the increased PAHs content could be caused by a series of reactions such as polymerization, oxidation, degradation and volatilization of unsaturated fatty acids and generation of radicals during the food storage period.

The differing impacts of storage time on PAHs content in smoked products and oils could be due to different timelines of PAHs formation. Most PAHs in smoked products are induced during high temperature processing before storage, therefore storage could be a manner to reduce PAHs for such cooked products by PAHs decomposition. Comparably, since the oil is a non-heated product, the PAHs in oil are generated during storage by oxidation. Thus, to minimise the contamination of food products by PAHs, smoked products should be kept for a defined time based on the safety and quality, and oil products are safer when consumed as soon as possible.

Due to the limited research regarding PAHs formation during food storage, more details on the relationship between storage conditions such as relative humidity and light exposure with PAHs formation are required. Since the existing research suggests that the change in PAHs content during storage is associated with lipid oxidation, it would be interesting to investigate whether the fat content and fatty acid composition would have an effect on the levels of PAHs formed during food storage.

4 Novel and potential PAHs reduction strategies

Recently, some novel techniques have been developed for PAHs reduction. Unlike the methods discussed above which aim to prevent the formation of PAHs, these novel strategies focus on removing generated PAHs from processed food products and reducing the bio-accessibility of PAHs after consumption (Table 5).

4.1 Degradation of PAHs by ozonation

The utilization of ozonation to degrade contaminated PAHs in aqueous solution and soil has been widely studied. The mechanism involves ozone directly oxidizing the PAHs or generating reactive oxygen species which react with PAHs (Lee, Puligundla and Mok, 2019; Nam and Kukor, 2000). Rozentale et al. (2016) suspended smoked fish sample in a plastic box and exposed them to a continuous flow of ozone at 20 ppm for 60 min and found that the PAH4 and BaP contents decreased to 11.7 and 1.52 $\mu\text{g/kg}$ from their initial levels of 14.9 and 2.82 $\mu\text{g/kg}$, respectively. They also showed that PAHs with more fused rings were more readily oxidised by ozone (Rozentale et al., 2016). Likewise, up to 46.21% and 45.25% reduction of BaP was observed in roasted sesame and perilla seeds by oxidation induced by low-pressure cold plasma (Lee, Puligundla et al., 2019). The low-pressure releases extremely high voltages which decomposes the oxygen molecule into oxygen radicals, which binds to oxygen molecules, hence forming ozone. However, the authors also mentioned that the

oxidized derivative of BaP (diol epoxide) would still be a concern due to its similar carcinogenic mechanism to BaP in the human body. Additionally, the plasma treatment is also associated with increased lipid oxidation and acidity as well as quality deterioration in colour and texture of the foods (Thirumdas, Sarangapani and Annapure, 2015). Therefore, potential commercialization and impacts on food safety and quality using ozone requires further study.

4.2 Reduction of PAHs from fuel emission using catalysts

Due to the emission of smoke and therefore PAHs from incomplete combustion, improving the combustion of fuel could be an effective way to reduce PAHs exposure in open flame processed food. The impact of the addition of KCl, Na₂CO₃, CaO, and Fe₂O₃ on the PAHs formation in the flue gas of sewage sludge bio-oil was studied, where the total EPA 16 PAH was decreased by 72, 71, 56 and 49%, respectively, at 850 °C (Hu et al., 2019). However, they also found that these catalysts promoted PAHs formation at a lower temperature of 450 °C. In another similar study on the EPA 16 PAH, the addition of 2% Fe₃O₄ (w:w) showed different inhibition activity against the formation of the 16 PAHs at 850 °C in the flue gas of combusting coal, with the highest reduction (72%) observed for CHR (Yao et al., 2020). Similar to ozonation, the authors explained that the mechanism of this method is that the catalysts can form reactive oxygen species that oxidize the PAHs.

However, there is no reports applying this method to PAHs in processed food products,

which deserves further investigation. It is also worthy to investigate the inhibiting effect of catalysts on the PAHs formation by the combustion of other common solid cooking fuels, such as charcoal.

4.3 Photo-degradation of PAHs by UV irradiation

The content of PAHs can be reduced by UV irradiation due to its photo-degradability. For example, up to 50% reduction of 14 PAHs with more than 3 fused rings occurred in water when UV-C irradiation was applied (264 nm wavelength) for 90s (Włodarczyk-Makula, Wiśniowska, Turek and Obstój, 2016). Moreover, a maximum reduction rate of 95% of 14 PAHs (including PAH8) was reported in industrial soil by UV-C irradiation (200-280 nm wavelength) at 30 °C for 24 h with TiO₂ as a photo-degradation catalyst (Eker and Hatipoglu, 2019). The PAHs reduction efficiency by UV irradiation was positively associated with the temperature and amount of added TiO₂, but it was negatively associated with increasing UV wavelength, as longer wavelengths have less energy to degrade PAHs. By using UV-C irradiation (254 nm wavelength) at 25 °C for 24 h, Chen and Chen (2005) were able to show reduction of 11, 16, 18, 25, and 29% of IP, BbFA, DBahA, BaA and BaP in contaminated LDPE film, respectively, and longer irradiation time resulted in more reduction of PAHs content. However, it was suggested that PAHs with higher number of fused rings are more resistance to the UV-C degradation (Chen and Chen, 2005; Eker and Hatipoglu, 2019;

Włodarczyk-Makula et al., 2016).

Although UV irradiation is commonly used in food processing such as pasteurization of juice and bacterial control for meat (Koutchma, 2008), reports on the application of this technology for food-related PAHs removal is very limited. More research is needed to investigate the UV irradiation for PAHs removal in food products, including the degradation mechanisms and safety evaluation of the degraded PAHs compounds.

4.4 Adsorption of PAHs by LAB

The application of LAB to reduce PAHs in processed food products has only been tentatively studied. LAB cannot inhibit the formation of PAHs, but can bind with PAHs by their interaction with peptidoglycans of the bacterial cell wall (Shoukat, Aslam, Rehman and Zhang, 2019; Yousefi et al., 2019; Zhao et al., 2013). It was reported that 15-31% of PAH4 was reduced in cold-smoked pork sausage after the immersion of sausage in a live LAB suspension, without any apparent changes in sensory properties (Bartkiene et al., 2017). Although this method could be restricted to live LAB without heat treatment (or the bacteria will die), a few studies indicated that the dead LAB still retain efficient binding capacity to PAHs as the cell wall is not affected (Shoukat et al., 2019; Yousefi et al., 2019). Additionally, the dead bacteria would not impart any effects on quality such as creamy and viscous texture, and acid taste which were caused by live cells in food (Narvhus and Gadaga, 2003).

Moreover, the bound LAB-PAH complex was stable in an alkaline phosphate buffer saline which might reduce the bioaccessibility of PAHs for uptake in the intestine (Yousefi et al., 2019). Because LAB are food compatible and known to confer health benefits to humans (Kechagia et al., 2013), the application of LAB for PAHs reduction has significant potential in the food industry.

4.5 Adsorption of PAHs by cellulosic aerogel

As mentioned previously, active carbons are used to adsorb the PAHs during oil refining (Rojo Camargo et al., 2012; Teixeira et al., 2007). Cellulose also possesses an excellent adsorption ability to metal ions and organic substances such as PAHs. Kim, Han, et al. (2021) generated cellulosic aerogel with a porous structure for better adsorption capacity, which they applied over a smoke regulator in a smoking chamber to adsorb PAHs. The PAHs content in smoked pork sausages was significantly reduced ($p < 0.05$) by 9-24%, depending on the solution, smoking time and temperature. Using cellulosic aerogel as the filter reduced the PAHs in extracted oil from roasted sesame seeds. Kim, Kim and Shin (2021) showed that the mean levels of PAH4 was reduced to 0.8-1 $\mu\text{g/kg}$ from 0.69-2.97 $\mu\text{g/kg}$ in the oil from the roasted sesame seeds. The researchers also indicated that the PAHs adsorption rate (from 31.56 to 47.32%) was increased with the increased proportion of cellulosic aerogel (from 0.3 to 5%; w/w), and no significant changes in acid value, peroxide value and color parameters

were detected in the treated oil.

The PAHs removal by cellulosic aerogels is based on their absorption onto functional groups of cellulose, and the porous structure enhances this process with greater cellulose surface exposure (Kim, Han, et al., 2021; Kim, Kim, et al., 2021). A significant advantage of this physical removal technique is to avoid generating toxic compounds during the process. There is potential to explore reduction of PAHs in food using cellulose derivatives that could present greater absorptivity, such as methylcellulose, hydroxypropyl cellulose, and carboxymethyl cellulose, which deserve further research.

4.6 Application of edible coating

Edible coatings have been widely used to improve food quality and safety, such as salt reduction, for antimicrobial purposes, and to improve texture and appearances (Galus, Kibar, Gniewosz, and Kraśniewska, 2020; Xiong, Deng, Warner and Fang, 2020). To date, there is no study showing the impact of edible coating on PAHs content of any food products, although it may have potential to reduce PAHs through two mechanisms as follows.

Firstly, the edible coating may reduce the PAHs content by reducing oil uptake. The edible coating acts as a barrier to inhibit the oil particles penetrating into the food. Kurek, Ščetar and Galić (2017) comprehensively reviewed the application of different edible coatings on the reduction of oil uptake in diverse processed foods, and the highest reduction

rate was 91% in fried pastry mix. As discussed above, oil is not only a significant source producing PAHs, but also an effective PAH carrier in food. Thus, reducing oil uptake by edible coating could be an efficient method for PAHs reduction.

Secondly, the edible coating could also adsorb PAHs and then reduce its content in food. The ability of cellulosic aerogels to adsorb PAHs has been discussed above. The cellulose derivatives, methylcellulose, hydroxypropyl methylcellulose, hydroxypropyl cellulose, carboxymethylcellulose, and microcrystalline cellulose, are commonly used edible coating materials (Kurek et al., 2017), and potentially have similar functions in PAHs removal to cellulosic aerogel. Moreover, chitin, chitosan and their derivatives are also widely used edible coating materials, which have been used to reduce PAH contamination in water and to adsorb small PAHs such as NA, AC, AN, PY, etc.. (Björklund and Li, 2018; Crisafulli et al., 2008; Naing, Yau Li and Lee, 2016).

These studies suggested that edible coatings could be developed as a new method to reduce the PAHs occurrence in food, in addition to their already widely recognised functions in improving food quality and safety such as antioxidant and antimicrobial properties (Al-Tayyar, Youssef and Al-Hindi, 2020). More importantly, since the release of PAHs from the bound complex in the human digestive tract is a critical safety concern, the binding stability of the coating materials to PAHs in simulated digestive fluids should be investigated.

5 Conclusion

In this review, the main causes of exposure to PAHs and the mechanisms of PAHs formation in processed food have been critically evaluated. Extensive studies on the factors, such as fat content and processing methods, affecting PAHs formation have been the main area of research on PAHs in the past decade. These factors are present at each step of food processing from formulation of the food including composition and pretreatments, to processing methods and parameters, and to food packaging and storage conditions. Common strategies used for the reduction of PAHs, such as reducing fat content in food formulations, application of no- or low-smoke cooking methods, selection of suitable cooking fuels, avoiding continual reuse of frying oil, and selection of suitable packaging materials etc., have been proposed. These methods are mainly focused on inhibiting the generation of PAHs and have various reduction efficiencies. Furthermore, some novel and potential strategies to reduce PAHs including their application and potential directions for research have been identified, which mainly target the removal of PAHs by oxidation and adsorption. These strategies could be incorporated with traditional methods to enhance the PAHs reduction in processed food. However, the potential food safety issues of some applications such as ozonation and UV irradiation require further research. Comparably, the application of edible coating to processed foods shows significant potential for PAHs reduction. A wider and more in-depth understanding of the mechanisms and factors on PAHs formation in food will

facilitate new reduction strategies to enhance food safety and quality.

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Figure captions

Figure 1 Structures, abbreviations and carcinogenicities of polycyclic aromatic hydrocarbons (PAHs) in foods, in which 16 PAHs are defined by Environmental Protection Agency (EPA) of United States (EPA 16 PAH) and 15 +1 PAHs are defined by European Food Safety Authority (EU 15+1 PAH)

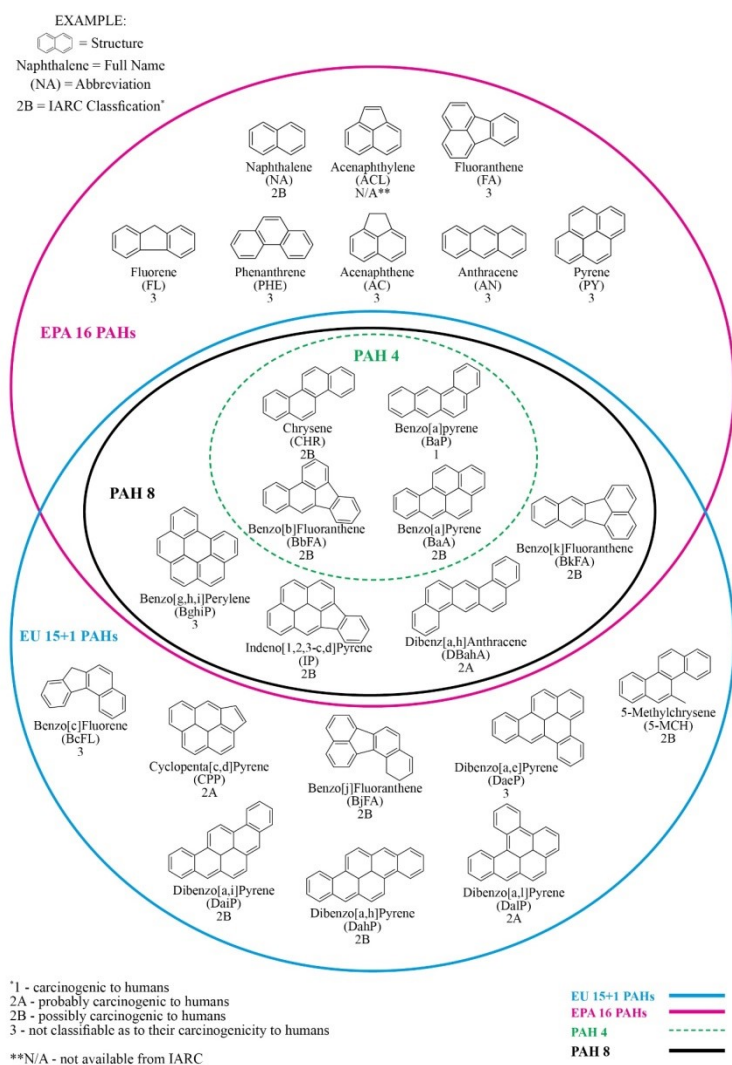


Figure 1 Table 1 Common PAHs

Methods	Food materials	Conditions	PAHs	PAHs reduction (%)	References
Reducing fat content	Dry fermented pork	20% fat reduced	EPA 16 PAH	19% reduction	(Gomes et al., 2013)

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**in food
matrix**

sausages

UHT milk	2.3% fat reduced	PAH4 + PHE, PY+AN and BghiP	30% reduction	(Naccari et al., 2011)
Yogurt	3.8% fat reduced	Most EPA 16 PAH (excludin g IP)	95% reduced	(Battisti et al., 2015)
Smoked Frankfurter-t ype sausages	10, 20 and 29% fat reduced	EU 15+1 PAH	37, 61 and 69% reduction with 10, 20 and 29% fat reduced, respectivel y	(Pöhlmann et al., 2013)
Heated beef system	5, 9, 13, 17 and 20% methyl linolenate (fatty acid methyl ester) reduced	PAH 8	9, 19, 26, 34, and 49% reduced with 5, 9, 13, 17 and 20% methyl linolenate	(Min et al., 2018)

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				reduced, respectivel y	
Using low-fat alternativ es	Portuguese smoked blood sausages	6.6% difference in fat (fat content of <i>Chouriço mouro</i> and <i>Morecla</i> are 53 and 46.4%)	EPA 16 PAH	145% reduced	(Santos et al., 2011)
	Grilled beef	7.3% difference in fat (Fat content of loin and ribs are 31.7% and 24.4%)	PAH 4	22% reduced	(Lee et al., 2016)
	Grilled pork	16.95 difference in fat (fat content of neck lean and belly are 9.5% and 26.5)		47% reduced	
Changing fat type	Oven cooked pork patties	40% back fat replaced by olive oil	BaP and BaA	7.75 and 51.52 % reduction at 180°C and 220°C, respectivel y	(Lu et al., 2017)
		40% back fat replaced by sunflower oil		21.71% reduction (at 180 °C)	
		40% back fat replaced by grape seed oil		17.38% reduction	

(at 220 °C)					
Reducing amino acids	Grilled pork sausages with	Reduced (0.5/100g) L-proline, L-tryptophan, L – phenylalanine, (L -tyrosine, L – threonine, L –serine, L -lysine, L –arginine, L -glutamic acid, and L -aspartate acid	PAH8 + NA, AC, FL and FA	Up to 64% reduction	(Nie et al., 2018)
Reducing and using different sugars	Grilled pork sausages	Reduced (0.5/100g) D-glucose, D-fructose, 4-(α -D-Glucosido)-D- glucose, and cellulose	PAH8 + NA, AC, FL and FA	Up to 55% reduction	(Nie et al., 2018)
	Sugar	Crystal sugar, granulated sugar, Demerara sugar, brown sugar, and organic sugar	BaA, BbFA, BkFA, BaP, and DBahA	86, 93, 50, and 74% reduction replacing crystal sugar by granulated sugar, Demerara sugar, brown sugar, and organic sugar, respectivel y	(Tfouni and Toledo, 2007)

Addition of Antioxidants	Fry with peanut skin		8.24-15.06 % reduction in TBHQ-free oil;	
	Fried peanuts	EPA 16 PAH	20.06-22.6 3% reduction in TBHQ-added oil	(Zhao et al., 2017)
		TBHQ addition (60 mg/kg)	41.42-71.7 5% reduced	
Pan fried pork	Addition of onion (30g/100g)	Most PAH8 (excluding CHR and IP)	54-67% and 91% reduction in meat and gravy	(Janoszka, 2011)
	Addition of garlic (15g/100g)		41 -66% and 14-79.4% reduction in meat and gravy	
Deep-fried beef meatballs	Addition of garlic and onion, red chili, paprika, black paper	BaA and BaP	65, 57, 65, 87, 47, and 98 %	(Lu et al., 2018)

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	and ginger (5g/kg)	reduction, respectivel y	
Deep-fried chicken meatballs		86%, 86%, 79%, 74%, 97%, and 97% reduction, respectivel y	
Charcoal-gri lled sirloin pork	Addition of diallyl disulfide (100 and 500mg/kg)	33 and 67 % reduction of BaP and total PAHs by 100 mg/kg	
		100 and 84 % reduction of BaP and total PAHs by 500 mg/kg	Wongmaneepr atip et al., 2019)
	Addition of quercetin (100 and 500mg/kg)	16 and 61% reduction of BaP and total PAHs by 100	

			mg/kg	
			21 and 55% reduction of BaP and total PAHs by 500 mg/kg	
Charcoal-Grilled loin Pork steak	Addition of Pilsner beer, nonalcoholic Pilsner beer and Black beer (1 ml/g)	PAH8	13, 25 and 53% reduction of PAH8, respectively	(Viegas et al., 2014)
Chinese fried bread, youtiao	Addition of TBHQ, rosemary extract, tea polyphenol, and antioxidants of bamboo into soybean/palm oil as the frying oil (60, 120, and 180 mg/kg)	EPA 16 PAH	Up to 30, 26, 13 and 28.85 % reduction in soybean oil, respectively; 36, 29, 10 and 39% reduction in palm oil, respectively	(Gong et al., 2018)
barbecued	Addition of 2% red	anthracene	45%	(García-Lomil

	beef patties	wine pomace seasoning (w/w, seasoning/patty)	ne (A), fluoranthene (FA), pyrene (PYR) and PAH8	reduction at day 9	lo et al., 2017)
	grilled beef patties	Addition of nutmeg and ginger oil and their nanoemulsions (0.02 and 0.04%)	PAH8	22.21 to 38.48% reduction at Day 90; nanoemulsified oil had a 1.34 to 2.84% higher reduction rate	(Esfahani Mehr et al., 2019)
Using different sausage	Smoked beef sausage	Sheep casing replaced by cellulose casing	EPA 16 PAH	78% reduction	(Youssef et al., 2016)

	Smoked pork sausage (<i>Slavonska kobasica</i>)	Natural casing replaced by collagen casing	EPA 16 PAH	34-36% reduction	(Mastanjević et al., 2019)
	Dry fermented pork sausages	Hog casing replaced by collagen casing	EPA 16 PAH	58-63% reduction (with 20% fat content) 44-82% reduction (with 40% fat content)	(Gomes et al., 2013)
	Smoked pork sausage (Chorizo)	Tripe casing replaced by collagen casing	FA and Most PAH8 (excluding CHR)	25% reduction	(García-Falcón and Simal-Gándara, 2005)
Increasing water content	Heated beef system	Addition of 1:1 water (beef: water by weight)	PAH8	45-72% reduction	(Min et al., 2018)

	Fried Chicken	Thawed by fridge, microwave, and water immersion	PAH4	Up to 8 and 10% reduction in air-fried and deep-fried chicken using fridge-thawed, respectively	(Lee et al., 2020)
Reduction of pH	Grilled chicken	Addition of acid and base marinade	EPA 16 PAH	70% reduction in double acid marinade compared to double based marinade of sample	(Wongmaneepratip and Vangnai, 2017)

Table 2. Effect of different processing methods on the formation of food PAHs

Food Material	Processing Methods	PAHs	Impact on PAHs	References
Sausage and	Grilling and frying	EPA 16	Frying generates	(Samiee et al.,

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burger		PAH	more PAHs than grilling	2020)
Meats and fishes	Smoking, frying, grilling	PAH8	Smoking generated the most PAHs	(Manda et al., 2012)
Chicken meat	Boiling, frying, barbecue, and roasting	EPA 16 PAH	Frying generates the more PAHs followed by roasting, barbecue, and boiling	(Onwukeme and Okafor, 2015)
Beef, goat meat and pork	Direct-heat charcoal roasting, shallow-pan frying and electric-oven grilling	EPA 16 PAH	Electric-oven grilling was a better method than the other two	(Onyango et al., 2012)
Beef and Pork	Charcoal Roasting and grilling	Most PAH8 (excluded BaA)	Grilling generates more PAHs than roasting	(Chung et al., 2011)
Beef	Barbecue and Pan-frying	PAH4 + PHE, FA, and PY	Barbecue generates more PAHs than Pan-frying	(Kılıç Büyükkurt et al., 2020)
Beef stripe, pork and chicken fillet	Smoking, grilling and boiling	BkFA, BaP, IP, and BghiP	Smoking generates the more PAHs followed by grilling and boiling	(Olatunji, Fatoki, Opeolu, et al., 2014)

Table 3 Common PAHs reduction strategies by changes of food processing parameters

Food	Conditions	PAHs	PAHs reduction (%)
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materials			
Deep-fried potatoes	30 min reduced (45 min to 15 min) in rapeseed oil, soybean oil, peanut oil and olive oil	EPA 16 PAH	Up to 60% of reduction in rapeseed oil
Deep-fried chicken nuggets			Up to 55% of reduction in rapeseed oil
Roasted ground <i>Arabica</i> coffee	15 min reduced (20 min to 5min)	Most of EPA 16 PAH (excluding NA, ACL, AC, FL and IP)	Up to 72% reduction (at 250 °C for lot b) Up to 70% reduction (at 260 °C for lot a)
Heated beef system	15min reduced (30 min to 15 min)	PAH8	Up to 40% in wet condition; 56% in dry condition
Sugar-smoked poultry meat	3 min reduced (6 min to 3 min)	EPA 16 PAH	12, 53, 45, 34 and 68% reduction in chicken heart, chicken drumstick, chicken gizzard, chicken breast and duck drumstick, respectively
Sugar-smoked red meat			21, 41, 54, 73, and 42% reduction in beef steak, pork knuckle, ham, lamb steak and pork chop, respectively
Sugar-smoked seafood			67, 62, 71 and 100% reduction in octopus, salmon, shrimp and squid, respectively
Boiled smoked sausages	57 min extended (5 min to 62 min)	BaP	38%
Baltonowski bread	10 and 20 °C decreased	SCF 15 PAH + PHE, NA and FA	With 10 °C decreased, 22% reduction in crumb and loaf of bread, 32% reduction in crust;

			With 20 °C decreased, 47, 46 and 48% reduction in crumb, loaf of bread, and crust, respectively
Rye bread			With 10 °C decreased, 22% reduction in crumb and crust, and 21% reduction in loaf of bread; With 20 °C decreased, 36, 34 and 40% reduction in crumb, loaf of bread, and crust, respectively
Whole-meal rye bread			With 10 °C decreased, 26, 28 and 23% reduction in crumb, loaf of bread, and crust, respectively With 20 °C decreased, 63, 47 and 35% reduction in crumb, loaf of bread, and crust, respectively
Gas-grilled beef satay	50, 100, 150 and 200 °C decreased	EPA 16 PAH	In control group, 21, 37, 49, and 71% reduction with 50, 100, 150 and 200°C decreased, respectively; In marinade group, 27, 43, 58 and 70% reduction with 50, 100, 150 and 200°C decreased, respectively;
Smoking salmon	Cold-smoking (15-30°C) and hot-smoking (65-80°C)	25 PAHs (including EPA 16 PAH and EU 15+1 PAH)	70% reduction in cold-smoked salmon
Roasted ground <i>Arabica</i>	10 °C decreased	Most of EPA 16 PAH (excluding NA, ACL, AC,	Up to 64% reduction for lot a; Up to 73% reduction for lot b

coffee		FL and IP)	
Heated beef system	40, 80, and 120 °C decreased	PAH 8	<p>In wet condition, up to 36, 46 and 62% reduction, when 40, 80, and 120 °C decreased, respectively</p> <p>In dry condition, up to 45, 61 and 74% reduction, when 40, 80, and 120 °C decreased, respectively</p>
Grilled meats	Fat droplets were collected under the grilling grid	PAH4	85, 76, 48 and 89% reduction in beef loin, beef rib, pork neck lean and pork belly, respectively
Grilled meats	A ventilation duct was placed between flame and meat		73, 71, 41 and 74% reduction in beef loin, beef rib, pork neck lean and pork belly, respectively
Smoked cheese	A lower and higher position placed in smokehouse	60 PAHs (including EPA 16 PAH and EU 15+1 PAH)	68% reduction by placing at higher shelf of smokehouse
Smoked sausage (<i>Slavonska kobasica</i>)	Hanging 2 and 3 m from the fire	EPA 16 PAH	50 and 51% reduction by placing at 3m in natural and collagen casing, respectively
Smoked fish	Using external smoke source	25 PAHs (including EPA 16 PAH and EU 15+1 PAH)	54, 64 and 23% reduction in Herring (20% fat), Mackerel (30% fat) and Trout fish (8%), respectively
Grilled beef and chicken	<p>Wrapped by aluminum and banana leaf;</p> <p>Steam and microwave</p>	FA, BbFA, and BaP	<p>46 and 40% reduction in beef sample wrapped by aluminum and banana, respectively;</p> <p>74 and 80% reduction in chicken sample wrapped by aluminum and banana,</p>

	preheating		respectively; 49 and 44% reduction in beef sample preheated by steam and microwave, respectively; 55 and 63% reduction in chicken sample preheated by steam and microwave, respectively;
Toasted bread	Wrapped by aluminum foil and toasted by a frying pan	PAH8 + 5-MCH, BjFA, and DalP	99.5% and 97.5% reduction by using aluminum foil and frying pan
Smoked fish	Using external smoke source	SCF 15 PAH	72% reduction in rainbow trout processed by Alder logs; 48% reduction in brook trout processed by beech chips
Smoke of charcoal	Charcoal was preheated for 5 min, 20 min and 5 h, and at 500, 750 and 1000 °C	EPA 16 PAH	99% reduction in PAH4 preheated for 5 h at 1000 °C by compared with it preheated for 5min at 500 °C
Roasted Peking duck	Using wood and electric oven	Most EPA 16 PAH and EU 15+1 PAH (excluding BcFL, BjFA, NA, ACL, and AC)	80 and 48% reduction in skin and lean meat, respectively by using electrical oven
Grilled pork belly	Charcoal, infrared-ray and electric grilling	BaP	100% (8.04 µg/kg in charcoal gilled sample, and BaP was not detected in infrared-ray nor electric grilled sample)
Dried Rice	Mature wood, rice husk, gas,	EPA 16 PAH	65, 88 and 94% reduction in brown rice using rich husk, gas and electric heating,

	and electric heating		respectively, comparing to using mature wood; 26, 61 and 94% reduction in parboiled brown rice using rich husk, gas and electric heating, respectively, comparing to using mature wood; 48, 54 and 77% reduction in polished rice using rich husk, gas and electric heating, respectively, comparing to using mature wood
Grilled Turkish kebab	Gas and charcoal	BaP	76% reduction using gas
Grilled Iranian kebab	Gas and charcoal	Most EPA 16 PAH (excluding NA)	25, 29, 18, and 24% reduction by using gas in Juje Kabab, Juje Kabab Bal, Kabab Barg and Kabab Koobideh, respectively
Grilled pork, sausage, and chicken	Wood and disposable charcoal	PAH8 + 5-MCH, BjFA, DaIP and DaeP	35, 9 and 34% reduction by using wood in pork, sausage, and chicken, respectively
Grilled beef, pork, and chicken	White, black and extruded charcoal	EPA 16 PAH	60, 87 and 75% reduction by using white charcoal rather to extruded charcoal in beef, pork and chicken, respectively; 49, 83 and 62% reduction by using black charcoal rather to extruded charcoal in beef, pork and chicken, respectively
Grilled beef and salmon	Wood and coconut charcoal	EPA 16 PAH	9 and 64% reduction by using coconut charcoal rather to wood charcoal in beef and salmon, respectively
Smouldering-smoked	oak, poplar, hickory, spruce, fir, alder, beech, and beech	EU 15+1 PAH	35-55% reduction using poplar and hickory led

frankfurters and mini-salamis.	with an apple-smoking spice mix, cherry-smoking spice mix, and a mix of juniper berries and bay leaves		compared to beach wood
Smoked pork	Apple, alder, alder + juniper, spruce, maple, hazel, plum, aspen, bird-cherry, rowantree and charcoal	SCF 15 PAH	90% reduction by using apple wood instead of spruce which generated the highest level of PAHs; 40% reduction by using apple wood instead of charcoal
Fried chicken	Using air frying instead of deep-fat frying	PAH 4	24, 26 and 15% reduction in microwave-thawed thighs, wings, and breasts, respectively; 25, 25 and 12% reduction in fridge-thawed thighs, wings, and breasts, respectively; 26, 18 and 21% reduction in water-immersion-thawed thighs, wings, and breasts, respectively;
Sunflower, soybean and olive oil	Refining by neutralization, bleaching, and deodorization	EPA 16 PAH (ACL was not detected)	32, 53 and 71% reduction in refined sunflower oil by neutralization, bleaching, and deodorization, respectively; 32, 32 and 87% reduction in refined soybean oil by neutralization, bleaching, and deodorization, respectively; 25, 17 and 82% reduction in refined olive oil by neutralization, bleaching, and deodorization, respectively;

Soybean and rapeseed oil	Refining by neutralization, bleaching, and deodorization	EPA 16 PAH	31, 52 and 60% reduction in refined soybean oil by neutralization, bleaching, and deodorization, respectively; 30, 48 and 55% reduction in refined rapeseed oil by neutralization, bleaching, and deodorization, respectively;
Soybean oil	Refining by neutralization, bleaching, and deodorization	Most SCF 15 PAH (excluding CPP and BghiP)	Up to 88% reduction through refining mainly contributed by neutralization and deodorization
Soybean and rapeseed oil	Refining	EPA 16 PAH	50, 35 and 43% reduction in refined soybean oil before storage, stored at 25 and 4 °C, respectively; 46, 23 and 34% reduction in refined soybean oil before storage, stored at 25 and 4 °C, respectively;
Roasted laver with brown rice and chill oil based oil	Different ratio of brown rice oil and perilla oil, and brown rice oil and sesame oil	PAH4	15 and 21% reduction when the ratio of brown rice oil: sesame oil changed from 84.7%:1.2% to 85.3%:0.6% and 85.9%:0%, respectively; 8 and 32% reduction when the ratio of brown rice oil: perilla oil changed from 90.2%:6.8% to 93.6%:3.4% and 97%:0%, respectively

Table 4 Common PAHs reduction strategies by changes of food packaging and storage temperature and time

Methods	Food	Conditions	PAHs	PAHs
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materials				
Roasted duck	Packed in LDPE pouches for 24 h	BaA, BbFA, BaP, DBahA and IP (later two were not detected)	8, 54 and 73% reduction in BaA, BbFA and BaP respectively	
Soybean and rapeseed oil	Stored at 25 and 4 °C for 270 days	EPA 16 PAH	22 and 31% reduction in crude and refined soybean oil, respectively, stored at 4°C; 13 and 26% reduction in crude and refined rapeseed oil, respectively, stored at 4°C	
Smoked fish	Stored at 18 °C without control of oxygen and daylight for 7days	BaP	79% reduction	
Smoked beef sausage	Stored at 4 °C in fridge for 90 days	EPA 16 PAH	Almost 100% reduction in both sheep casing and cellulose casing sausage	

Table 5 Novel and potential PAHs reduction strategies for processed food

Method Type	Materials	Conditions	PAHs	PAHs reduction	References
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Author Manuscript					
Ozonation					
Smoke fish					
60 min ozonation					
PAH4					
22% reduction in PAHs and 46% reduction in BaP					
(Rozentale et al., 2016)					
Roasted sesame and perilla seeds					
30 min degradation by low-pressure cold plasma					
BaP					
46.21, 40.56, 45.20 and 45.25% reduction in regular roasted sesame seeds, over-roasted sesame seeds, regular roasted perilla seeds, and over-roasted perilla seeds, respectively					
(Lee, Puligundla et al., 2019)					
Addition of fuel catalysts					
Flue gas from sewage					
Addition of CaO, KCl, Na ₂ CO ₃ and					
EPA 16 PAH					
72, 71, 56 and 49% reduction					
(Hu et al., 2019)					

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	sludge bio-oil	Fe ₂ O ₃		with the addition of addition of KCl, Na ₂ CO ₃ , CaO and Fe ₂ O ₃ , respectively , at 850 °C	
	Coal	Addition of Fe ₃ O ₄	EPA 16 PAH	All the 16 PAHs were reduced 850 °C; CHR had the highest reduction, about 72%	(Yao et al., 2020)
Photo-degradation by UV irradiation	Coking wastewater	Under UV irradiation (UV-C; wavelength 264 nm) for 30, 60 and 90 s	Most EPA 16 PAHs (excluding ACL)	10-12.3% reduction of total 15 PAHs; 5.0-9.1% reduction of 2-ring PAHs; 38-50% reduction of 3-ring PAHs;	(Włodarczyk-Mak uła et al., 2016)

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				26-45% reduction of 4-ring PAHs;
				34-43% reduction of 5-ring PAHs;
				36.6-40% reduction of 6-ring of PAHs;
				No obvious linear relationship between irradiation time and PAHs concentratio n
Industrial soil	With/withou t addition of TiO ₂ ; Irradiation temperature at 18 or 30°C; irradiation	PAH 8 + PHE, AN, FA and PY	Maximum 95% reduction of total 12 PAHs with 10% and 20% addition of TiO ₂ at 30	(Eker and Hatipoglu, 2019)

		wavelength of 200-280 nm (UV-C) or 315-400 nm (UV-A) for 24 h		°C with the wavelength of 200-280 nm (UV-C); Reduction efficiency increased with increased temperature and decreased wavelength	
Contaminat ed LDPE film	Irradiation for 1, 2 and 3 h	BaP, BaA, BbFA, IP and DBahA	11, 16, 18, 25, and 29% reduction of IP, BbFA, DBahA, BaA and BaP, respectively ;	(Chen and Chen, 2005)	Reduction efficiency increased with decreased number of fused rings

Adsorption by LAB	Cold smoked pork sausage	Immersed in live LAB suspension	PAH4	15-31% reduction	(Bartkiene et al., 2017)
Adsorption by cellulosic aerogel	Smoked pork sausage	Functionalized by NaOH/urea, LiBr and LiOH/urea	PAH 4	6-18% reduction by NaOH/urea functionalized cellulosic aerogel; 7.9-24% reduction by LiBr functionalized cellulosic aerogel; 7.9-19% reduction by LiOH/urea functionalized cellulosic aerogel	(Kim, Han, et al., 2021)
	Extraction of oil from roasted	NaOH/urea functionalized cellulosic	PAH 4	34-47% reduction depending	(Kim, Kim, et al., 2021)

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