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Summarizing minimization of polycyclic aromatic hydrocarbons in thermally processed foods by different strategies

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ABSTRACT

Polycyclic Aromatic Hydrocarbons (PAHs) are environmental carcinogens which are produced in food during their processing and thermal treatment. Consumption of such foods results in diverse diseases including cancer in humans, raising a serious need to either prevent formation of these compounds in food or reduce and remove these carcinogens once they are formed. The concern is very important and reasonable as diet contributes to 88–98% exposure. Some of the processes followed to prevent PAHs formation include regulation of the cooking/processing practices/methods and revolves around deodorization and refining of oil as well as sugars, choosing right type of oil and frying process, choosing liquid smoking process over traditional practices, reducing fat content of product and preferring the right part of product (especially in meats), roasting and baking products at lower time-temperature combinations with preferences to electric oven and indirect processes, using the right heat medium (wood chips for barbequeing/charcoal grilling, using correct wraps for food products to prevent direct contact with heat and fat drippings and much more. Though these processes may be followed at household and commercial scales and have been stated in several published literature, but the absence of PAHs may not always be guaranteed in food products. This leaves the scientific community to attempt and develop strategies which can remove the already existing PAHs from food products and needs to be extensively reviewed and worked in future. This review bridges the PAHs' reduction and removal strategies [different types of ingredients (marination, spice addition), the specific kind of packaging (aluminium foils, plastic films, charred barrels, paraffin rind), the heating treatments (irradiation, microwave preheating, defatting, brewing etc.), but also the characteristics of adsorbents and filters used (active charcoal, diatomaceous earth, zeolite filters, molecular sieves), together with innovative removal apparatus] with emphasis on biological and physical-chemical factors influencing their formation/reduction or removal/degradation, mainly in heat-processed food [such as composition and surface adsorption properties, etc.]. The overall goal is to develop the understanding of the interactions amongst all factors affecting PAHs removal and draw recommendations based on conclusions of scientific evidence and propose future challenges in this area.

1. Introduction

Polycyclic Aromatic Hydrocarbons (PAHs) are carcinogenic contaminants produced in a wide range of processed food products due to their thermal processing such as roasting, grilling, smoking etc. (Onopiuk et al., 2021; Wang et al., 2022; Zhang et al., 2021). PAHs contaminated foods include diversified items such as vegetables, fruits, edible oil, smoked products, barbecue products, fried foods and dairy products. The levels of these compounds have often been reported towards higher side than the limits defined by European Union (EU), making their presence in food (especially processed) a reason of concern (Wang et al., 2022). Some of the recent articles stating the PAHs concentration in processed food are mentioned in Table 1.

Considering the toxic nature of these compounds, United States Environmental Protection Agency (USEPA) has identified 16 PAHs namely benzo[*a*]anthracene (B[*a*]A), indeno[1,2,3-*c*,*d*]pyrene (IND), acenaphthylene (ACY), dibenz[a,h]anthracene (D[ah]A), fluoranthene (FLTH), benzo[g,h,i]perylene (B[ghi]P), benzo[*a*]pyrene (B[*a*]P), benzo

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[*b*]fluoranthene (B[*b*]F), acenaphthene (ACE), pyrene (PYR), chrysene (CHRY), benzo[k]fluoranthene (B[k]F), anthracene (ANTH), fluorene (FLU), phenanthrene (PHEN) and naphthalene (NAP) as priority pollutants. The Scientific Committee of Food (SCF) in 2002 recommended monitoring of 15 PAHs, including 8 high molecular weight (HMW) PAHs that are also on the US EPA list. SCF found that 15 out of 33 PAHs evaluated, namely B[*a*]A, B[*b*]F, benzo[*j*]fluoranthene, B[k]F, B[ghi]P, B[*a*]P, cyclopenta[*cd*]pyrene, D[ah]A, dibenzo[*a,e*]pyrene, dibenzo[*a,h*]pyrene, dibenzo[*a,i*]pyrene, dibenzo[*a,l*]pyrene, IND and 5-methylcrysene exhibited clear evidence of mutagenicity/genotoxicity and, with the exception of B[ghi]P, these compounds had also shown clear carcinogenic effects in various types of biological tests in experimental animals (SCF, 2002). The use of B[*a*]P as a marker of carcinogenic PAHs in food was thus proposed based on the study of PAH profiles in food and on carcinogenicity studies on coal tar mixtures in mice (Culp et al., 1998). Of the 15 priority PAHs in the EU, 12 were identical to those identified by the International Agency for Research on Cancer (IARC) as human carcinogens. Other PAHs, which have not been defined as carcinogens, may act synergistically (IARC, 2013).

As a result, and in order to provide the basis for solving the problem of PAHs pollution, different control measures for PAHs in food have been attempted and discussed worldwide in recent years. Wang et al. (2022) had enlisted several process-driven measures which aid in reducing PAHs contamination in processed food products. Awareness about the process conditions favouring high accumulation and formation of PAHs in processed food products may help in reducing many health hazards, especially related to PAHs. However, need lies to remove PAHs already existing or present in processed food products, especially when their processing may not be much altered owing to the cost involved in the process, lack of scalable techniques and other technological advancements. This review gives an insight on the measures for food producer, processor, and consumer with respect to PAHs reduction and removal in processed food. Diversified strategies where PAHs formation has been checked in processed food, the factors controlling the reduction/removal process, and the gaps and future prospects have also been discussed (Tables 2–4).

2. PAHs carcinogenicity associated with the processed food intake

PAHs are hydrophobic and inert compounds which initiate carcinogenicity in mammalian cells through their metabolic transformation to diol epoxides. As known, these diol epoxides react with cellular macromolecules including deoxyribonucleic acid to result mutations and errors during nucleic acid replication. Also, depurination (mutation) facilitated by chemically unstable alkylation of deoxyribonucleic acid (DNA) may also occur upon one-electron oxidation based generation of other reactive intermediates (Chen and Chen, 2005). There have been three main mechanisms identified for PAHs toxicity and carcinogenicity in human cells, i.e.: 1) formation of diol-epoxides followed by DNA adducts' generation, 2) formation of free radicals and 3) formation of ortho-quinones (Ewa and Danuta, 2016). The detailed mechanism of metabolic activities favouring PAHs entrance into the body and the DNA damage are summarized respectively in Figs. 1 and 2. As observed, PAHs cause significant changes at cellular and molecular level and favour cancer of different body organs. Occupational exposure to PAHs results in vomiting, diarrhoea, nausea, eye irritation and confusion while chronic health effect involve jaundice, kidney damage, asthma-like symptoms, cataract, affected immunity, breathing problems, skin redness and inflammation. Breakdown of red blood cells (RBCs) have also been reported. Effects on embryo include pre-mature delivery, decreased body weight of offspring, heart malformation and reduced intelligent quotient as well as behavioural problems in infants. Long-term exposure as shown by animal studies led to stomach cancer, skin cancer and lung cancer depending on the route of exposure (Rengarajan et al., 2015). Since diet form 88–98% of the PAHs exposure route and hence their levels in food need to be regulated. Attempts are thus made to check the concentration of PAHs in food and regulate the risk associated with uptake of these food products for diverse populations.

3. Mechanism behind PAHs formation in processed foods

Alomirah et al. (2011) has stated three main mechanisms for PAHs formation in food. First mechanism identified is pyrolysis of organic matter as temperature reaches above 200 °C during processing while second mechanism is emergence of PAHs laden smoke during fat

Table 1
PAHs' concentration in processed food products as reported in the recent literature.

Food Products	Cooking Process	Concentration B[*a*]P (μg/kg)	Concentration total PAHs (μg/kg)	Reference
Beef Doner Kebabs	gas, charcoal, or electric	nd-7.38	PAH4 = 43.05–150.40 PAH8 = 43.05–198.10	Karslioglu & Kolsarici, 2022
Bonito, katsuobushi	Smoking	2.10	PAH4 = 6.64	Seko et al. (2022)
Chinese tea	Drying and Curing	nd-53.36	PAH4 = 37.64–106.90 16 EPA PAHs = 136.99–440.82	Guo et al. (2022)
Paprika "Pimentón de La Vera"	Smoking and Drying	22–85	PAH4 = 277 to 1208	Velazquez et al., 2022
Beef and Chicken	Charcoal-Grilling	0.271–1.341	16PAHs = 8.755–9.941	Haiba et al. (2021)
Pork, Beef, Chicken, Duck, Lamb	Grilling		16PAHs = 4.38–547.5	Pirasaheb et al., 2022
Fish	Smoking		PAH4 = 76.3–2700 12PAHs = 537-16800	Slamova et al. (2021)
Fish	Canning		16PAHs = 101-733	Iwegbue et al. (2022)
Fish, meat and plantain	Smoking	45–66	4PAHs = 445-513	Adesina (2022)
Various Ready-to-Eat Food Products		0.64	8PAHs = 1.71	Lee and Shin (2021)
Fish	Smoking	0.66–15.51	4PAHs = 2.52–121.60	Asamoah et al. (2021)
Different meats	Smoking	nd-2.61	6PAHs = 0.45–21.24	Onopiuk et al. (2022)
Fish	Smoking		16PAHs = 30-100	Racovita et al. (2021)
Cheeses	Smoking		16PAHs = 10-20	Racovita et al. (2021)
Dairy Samples	Ultra heat treatment (UHT)		20PAHs = 2.4–10.1	Yan et al. (2022)
Griddled milk bread	Baking		8PAHs = 1.25	Kim et al. (2022)
Stir-fried black sesame	Frying		8PAHs = 1.17	Kim et al. (2022)
Guarana (*Paullinia cupana*) seeds	Drying	nd-169		Junior et al. (2022)
Bread	Baking		16PAHs = 2.68–4.40	Karsi et al. (2022)
Cheeses	Smoking	17		Migdal et al. (2022)
Edible Oils and Shea Butter			16PAHs = 126-865	Iwegbue et al. (2021)
Meatballs	Barbecuing	2.33–4.30	4PAHs = 8.41–15.48	Oz (2021)

Table 2
Ingredients and food components based reduction and removal of PAHs in processed food products.

Strategies	Food	Conditions	PAHs	Reduction%	References
Marination	Poultry meat	Marinating with juice for different time period	16PAHs	23.74%, 6.64%, 47.63%, 23.45% and 19.10% reduction in chicken heart, chicken drumsticks, chicken gizzard, chicken breasts and duck drumsticks respectively	Kao et al. (2012)
	Beef Satay	Honey-spices marination followed by gas-grilling	15PAHs	0.77% reduction when gas-grilled beef satay with Trigona sp. honey-spices marination, temperature of grilling 150 °C.	Hasyimah et al. (2022)
	Grilled beef meat	Seven marinade treatments containing 1) basic marinade, which include sugar, water, onion, turmeric, lemon grass, salt, garlic, coriander and cinnamon, 2) basic-oil, 3) Commercial marinade. 4) basic-oillemonjuice, 5) basic-lemon juice, 6) basicoil-tamarind and 7) commercial-tamarind at four time intervals (0, 4, 8 and 12 h)	3PAHs	47%, 48%, 52%, 57% and 70% reduction in BaP concentrations achieved when basic-oil-tamarind, commercial-tamarind, basic, basic-oil-lemon juice and basic-lemon juice marinations respectively were used.	Farhadian et al., 2011
	Charcoal grilled chicken wings	Tea marination (green tea, white tea, yellow tea, oolong tea, dark tea, and black tea)	8PAHs	38.2%, 40.6%, 45.1%, 45.5%, 45.7% and 50.7% reduction observed with black tea, dark tea, oolong tea, yellow tea, white tea and green tea.	Wang et al. (2018)
	Charcoal grilled duck meat	Andaliman (*Zanthoxylumacanthopodium*, DC) fruit juice at 0,10,20,30%	BaP	62.51% reduction mainly at 20% fruit juice treatment	Sinaga et al. (2016)
	Beef	sage and thyme extracts prepared at 0.5 to 2.0 °Brix	7PAHs	41.47% and 46.05% reduction in sage and thyme treated barbecue samples at 2° Brix. 36.63% and 33.27% reduction in sage and thyme treated pan-fried samples at 2° Brix	Buyukkurt et al., 2017
	Charcoal grilled pork	Pilsner beer, non-alcoholic Pilsner beer, and Black beer	16PAHs	13%, 25% and 53% reduction in 8PAHs achieved on marination with Pilsner beer, non-alcoholic Pilsner beer and Black beer respectively	Viegas et al. (2014)
	charcoal-grilled chicken wings	Heineken, Tsing Tao, Budweiser, Corona, Snow and Harbin extract	8PAHs	67% reduction with Heineken exhibiting excellent performance	Wang et al. (2019a, 2019b)
	Grilled chicken	Oil types and pH varies in marination (Palm oil, acid, double acid, base and double base marinade)	16PAHs	9.34–73.06% reduction when using palm oil marinade over other marinades. 49.52–58.46% reduction observed on using control marination over oil marination.	Wongmaneepratip and Vangnai, 2017
Spice application	Pork meat and gravy	Onion and garlic	6PAHs	Onion caused 60% and 90% reduction in meat and gravy while garlic caused 54% and 13.5–79% reduction in meat and gravy	Janoszka (2011)
	Deep-fried meatballs	0.5% garlic, onion, red chilli, paprika, ginger and black pepper powderadded into beef and chicken meatballs fried at 180 °C	B(*a*)A and B(*a*)P	47.02–97.9% reduction in beef meatballs, 74.8–97.2% reduction in chicken samples	Lu et al. (2018)
Antioxidant application	Meat model system	Different antioxidants (BHT, BHA, a-tocopherol, EGCG and sesamol)	8PAHs	37.46–49.43% reduction in different antioxidant treated samples than control.	Min et al. (2018)
	grilled pork	dietary antioxidants, diallyl disulphide (DADS) and quercetin, in marinade	16PAHs	100% reduction in BaP and 84% in heavy PAHs with diallyl disulphide in marinade, 23 and 55% reduction respectively with quercetin	Wongmaneepratip et al. (2019)
	fried bread youtiao	rosemary extract, tea polyphenol, and antioxidant of bamboo	16PAHs	up to 30.30%, 23.47%, 11.38% and 28.85%, respectively compared to control (without antioxidant) youtiao fried in soybean oil	Gong et al. (2018)
	Charcoal-Grilled Chicken Wings	epigallocatechin gallate (EGCG), gallocatechin (GC), catechin (C), epicatechin gallate (ECG), catechin gallate (CG), eriodictyol, naringenin, and quinic acid	16PAHs	15.1–54.5% inhibition in PAH8, quinic acid and naringenin showed excellent inhibition	Wang et al. (2019a, 2019b)
Changes in frying oil	Fried peanuts	Addition of tert-butylhydroquinone (TBHQ)	16PAHs	71.75% reduction	Zhao et al. (2017)
Seasoning	Charcoal-grilled pork	white wine vinegar, red wine vinegar, apple cider vinegar, elderberry vinegar, apple cider vinegar with raspberry juice	16PAHs	Elderberry vinegar exhibited the highest inhibition (82%), followed by white wine vinegar (79%), red wine and cider vinegars (66%), and fruit vinegar with raspberry juice (55%).	Cordeiro et al., 2020
	Roasted laver	a model system to change the mixing ratio of oil, roasting temperature, and time	PAH4	35% reduction when perilla oil was removed from the mixed oil composition and roasting continued at 350 °C for 10 s	Kang et al. (2019)
	Barbecued beef patties	Red wine pomace seasoning and high-oxygen atmospheric storage	16PAHs	44.99% reduction when the seasoned patties (2%w/w) were stored in high oxygen atmosphere for 9 days compared to control and single treatments.	Garcia-Lomillo et al., 2017

Table 3
External controls based reduction and removal of PAHs in processed food products.

Strategies	Food	Conditions	PAHs	Reduction%	References
Packaging	Smoked sprats	Different packaging material	16PAHs	75.82% reduction when smoked sprats are packed in PET than PP package at room temperature. 52.85–82.88% reduction is seen when the storage time in different package material is between 0 and 2h than other durations (2h and above) at room temperature.	Kuźmicz & Ciemniak, 2018
	liquid model and roasted duck meat	Low density polyethylene and UV treatment	16PAHs	8–73% reduction in individual PAHs after 24h storage of meat in LDPE pouches. More than 50% PAHs adsorption in all the models. 70.8–84% reduction in LDPE after 3h of UV treatment.	Chen and Chen (2005)
	Cachaca	Glass jugs and polyethylene packs	16PAHs	88.22% reduction when glass jugs preferred over polyethylene packs	Machado et al. (2014)
	Smoked sausage	o-polyamide/low density polyethylene laminated film	PAH4	81.06% reduction	Semanova et al. (2016)
	liquid smoke flavour UTP-1	bottles made of low-density polyethylene and stored for 14 days	6PAHs	98.39% reduction after 14 days.	Simko and Brunckova (1993)
Adsorbents	Sesame oil	Activated carbons (of variable mesh size) and celite	B(*a*)P	32–46% reduction with Granular Darco 12–20 (D12) and Darco 20–40 (D20)mesh charcoal-based activated carbons	Shin et al. (2015)
	Sesame oil	Activated charcoal and clay, Notit-8015	16PAHs	20.8%, 54.7% and 67.4% reduction when activated clay, Notit-8015 and WY activated charcoal was used	Shi et al. (2017)
	Fish oils	Activated carbon, wood ashes and mussel shell ashes	10PAHs	42–100% removal observed for individual PAHs with pine ashes, 10–100% with wood ashes and 80–100% with activated charcoal. No removal with mussel shell ashes	Yebra-Pimentel et al. (2014)
	Smoked cured fish	modified traditional kiln fitted with activated charcoal filters	16PAHs	21–69% reduction	Essumang et al. (2014)
	Food grade deodorized coconut oil	Activated zeolite filters	16PAHs	60% reduction	Parker et al. (2017)
	Common carp	smoked without filter (Fo), with zeolite filter (Fz), filter with granular activated carbon (Fc) and gravel filter (Fg)	16PAHs	65.9% reduction with Fz, 63.95% reduction with Fc, 51.39% reduction with Fg	Babic et al. (2018a)
	Common carp	with zeolite filter, filter with granular activated carbon and gravel filter	16PAHs	71.7% reduction through zeolite, 70.7% with activated carbon, 64.2% with gravel	Babic et al. (2018b)
	bleached soybean oil	activated carbon with bleaching earth	PAH4	100% reduction	Aliyar-Zanjani et al. (2018)
High carbonization and a preheating step for charcoal preparation for smoke development in grilling	Charcoal used for grilling food	Mangrove charcoal was prepared at carbonization temperatures of 500,750 and 1000 °C. Charcoal was preheated at different burning times at 5, 20 min and 5 h	16PAHs	96.48% reduction in PAHs when charcoal produced at 500 °C when preheated for 5h than for 5 and 20min. 95.47% and 92.96% reduction when charcoal carbonization temperatures are 750 and 1000 °C and preheated for 5min.	Chaemsai et al. (2016)
Pretreatment of charcoal	Charcoal used for grilling food	900 °C of pyrolysis min^{-1} temperature, 5 h of holding time and 40 °C of heating rate	16PAHs	99.8% reduction upon pre-treatment	Teachathanyakul et al. (2018)

dripping on heat source. The PAHs in generated smoke adhere to food product upon contact. Third mechanism stated by Alomirah et al. (2011) is incomplete combustion of fuel/charcoal and heat source on which food is kept, thereby exposing the food product to released PAHs. It has been observed that due to the complexities and diversification of cooking methods, food products, food composition and ingredients; the mechanism of food products have not been well illustrated. Attempts are being made to identify the key processes facilitating PAHs formation in food and their mechanism.

Li et al. (2021) has demonstrated positive correlation between unsaturated fatty acids and PAHs 4 content, stating major participation of these fatty acids in PAHs formation. Hasyimah et al. (2022) identified that incorporation of honey and spice marination to beef satay promoted PAHs formation. It was stated that maillard reaction favors aromatization and de-hydrocyclisation, caramelization and fat/sugars degradation; which promote PAHs formation in food products. Additionally, pyrolysis of amino-acids and proteins favour condensation of nitrogen containing organic compounds followed with catalyzation of PAHs formation reactions. Similarly, Hung et al. (2021) reported that use of sugar-say sauce in cooking, favoured PAHs formation in dried pork which might be due to the formation of lipid degradation products (possessing conjugated double bonds) in Diels-Alder reaction with dienophile compounds. During this reaction, C1–C4 compounds attach with the lipid degradation products and undergo hydrogen abstraction/acetylene addition. Further reaction involves phenyl or methyl addition/cyclization. Min et al. (2018) in their model system study reported that PAHs formation is affected greatly by existence of water and anti-oxidants demonstrating that these are critical factors which needs to be accounted. Also it was stated that temperature has more influence than time with respect to favouring PAHs formation.

Nie et al. (2018) in order to identify the specific roles of these components on PAHs formation, subjected grilled pork sausages to different treatments such as R groups (aldo/keto groups in sugars), molecular weight carbohydrates, polarities and acid base amino groups. The study reported prominent role of aldehyde-based D-glucose, smaller molecular weight D-glucose, basic amino acids (L-lysine, L-arginine) and non-benzene ring amino acids in PAHs formation respectively over keto-based D-fructose, larger molecular weight

Table 4
Apparatus and treatments based reduction and removal of PAHs in processed food products.

Strategies	Food	Conditions	PAHs	Reduction%	References
Machine/apparatus	Sesame seed oil	Self-designed apparatus	BaP	70.7% reduction in BaP content at optimized condition i.e. a single washing cycle with 2-min spin-drying, 1350-rpm ventilation, and a single compression cycle. 33.33% and 25% reduction of BaP in sesame seed oil and perilla oil when self-designed apparatus was used than traditional apparatus.	Yi et al. (2017)
	plantain (Musa paradisiaca)	charcoal fired roasting equipment	16PAHs	99.7% reduction in comparison to direct roasting method	Adisa et al. (2019)
Heat pump-assisted drying	Red peppers (Capsicum annuum L.)	drying red peppers at 50–80 °C for 7–31 h followed by drying at 30 °C for 5 h	PAH4	38.5–87.6% reduction compared to air-drying	Hwang et al. (2019)
Gamma Irradiation	Ground Wheat Grains (about 200–300 μm size)	different doses of gamma rays (1, 5, 10, and 15 kGy) in a ^{60}Co package irradiator (ROBO, Russia) (dose rate 730 Gy/h).	7PAHs	35% reduction at 1 kGy and 70% reduction at doses higher than 5 kGy	Khalil et al. (2016)
	Sausages	fresh sausage was irradiated with γ-rays at doses of 0, 1, 4, and 8 kGy followed by Baking at 220 °C for 20 min		51.67% reduction when 8 kGy is used than non-irradiated form.	Li et al. (2021)
Microwave pre-heating	Olive pomace oil	Drying at 200 °C following the microwave pre-treatment	16PAHs	75% reduction	Kiralan et al. (2017)
Microbial treatment	Cold smoked pork sausages	lactic acid bacteria with antimicrobial properties	PAH4	30.79% and 12.57% reduction in before and after smoking *L.sakei* treatment than control. 14.11% reduction in after smoking treatment of *P. acidilactici*. 21.03% and 20.55% reduction with before and after *P. pentosaceus* treatment than control treatment.	Bartkiene et al. (2017)
Fat replacement	Pork patties	Replacement of fat by vegetable oils (sunflower, olive and grapeseed) and cooked at 180 °C and 220 °C	BaA and BaP	21.70% reduction when sunflower oil based replacement followed by cooking at 180 °C was done over control. 51.52% reduction over control when olive oil based replacement was done followed by cooking at 220 °C.	Lu et al. (2017)
UV treatment and temperature-time sensitivity	Rapeseed and Sunflower oil	UV application (direct and indirect)	16PAHs	56.35% and 33.87% reduction in direct and indirect radiation treatment respectively. 47.81%, reduction in sunflower oil when heated at 40 °C for 24h than 0 and 48h, 51.14% when heated at 100 °C for 2h than other time periods and 66.56% when heated for 2h at 200 °C.	Mocek and Ciemniak, 2016
Low pressure cold plasma	roasted sesame and perilla seeds	treatment for 30 min	B(*a*)P	degradation ranged between 40 and 46%	Lee et al. (2019)
Balanced extraction and purification	Propolis	Usage of a balanced mixture of ethanol and water solvents	8PAHs	96.84% and 99.32% reduction when 70:30 and 60:40 ethanol: water ratio respectively is used.	Galeotti et al. (2017)
	Fish oils	Ethanol extraction	10PAHs	60–90% removal observed for individual PAHs	Yebra-Pimentel et al. (2014)

4-(α-*d*-glucosido)-*d*-glucose and cellulose, acidic amino acids (L-glutamic acid, L-aspartate) and benzene ring possessing amino acids. Hu et al. (2021) has reported that free radical reaction favors B[*a*]P formation and can be affected by different form of sulphur compounds such as sulphur (-*S*-), thioallyl group (–S–CH2–CH═CH2); and allyl methyl trisulfide (AMTS).

As observed (Fig. 3a and b), studies focusing on mechanism determination for PAHs formation in food are very few and exhibits enormous opportunities for research.

4. Reduction and removal strategies for PAHs in thermally processed foods

4.1. Ingredients-based strategies

Ingredients form one of the important components of processed food products. Wang et al. (2022) highlighted the need to study the effect of ingredients on PAHs formation and solubility in food. Promising effect of several ingredients has been observed in reducing the PAHs contamination in processed foods.

PAHs formation in food has been considered as a consequence of food combustion followed by release of free radicals and their recombination; with major role of a) smoke interaction with food surface, b) food organic compounds' pyrolysis and c) reaction upon dripping of fat onto flames (Alomirah et al., 2011). Thus cutting down these free radicals by incorporation of some antioxidants or radical scavengers especially at pre-treatment stage is a probable solution for mitigating PAHs formation/accumulation in food products (Fig. 4). Different tea marinades prepared from white tea, green tea, yellow tea, dark tea, oolong tea and black tea demonstrated significant inhibitory effect on the formation of PAH8 in charcoal-grilled chicken wings (Wang et al., 2018). An inhibitory effect of 2% was found with oolong tea marinade while the effect was 31%, 23% and 58% with white tea, yellow tea and green tea respectively. On the contrary, an increase in PAH8 formation was found associated with dark tea (54%) and black tea (126%) marinades. It was also observed that PAH8 content significantly correlated ($R^2 = 0.915$) with total radicals in the marinades. The possible mechanism for PAH8 inhibition in the charcoal-grilled chicken wings treated with tea marinades was suggested to be the participation of radicals and phenolic compounds. As stated in the study, tea marinades cover the surface of the chicken wings. The reaction which had to occur directly between heat source and chicken wings during grilling is intercepted by the layer of tea. Since tea product possess polyphenols [mainly epigallocatechin gallate (EGCG)], the free radicals formed upon tea marination and heat source interaction during chicken wing grilling must have been quenched or scavenged by these polyphenols. Also, the oxidative sites or species present on chicken wings must have been covered by these antioxidant substances and might also have prevented Maillard reaction,

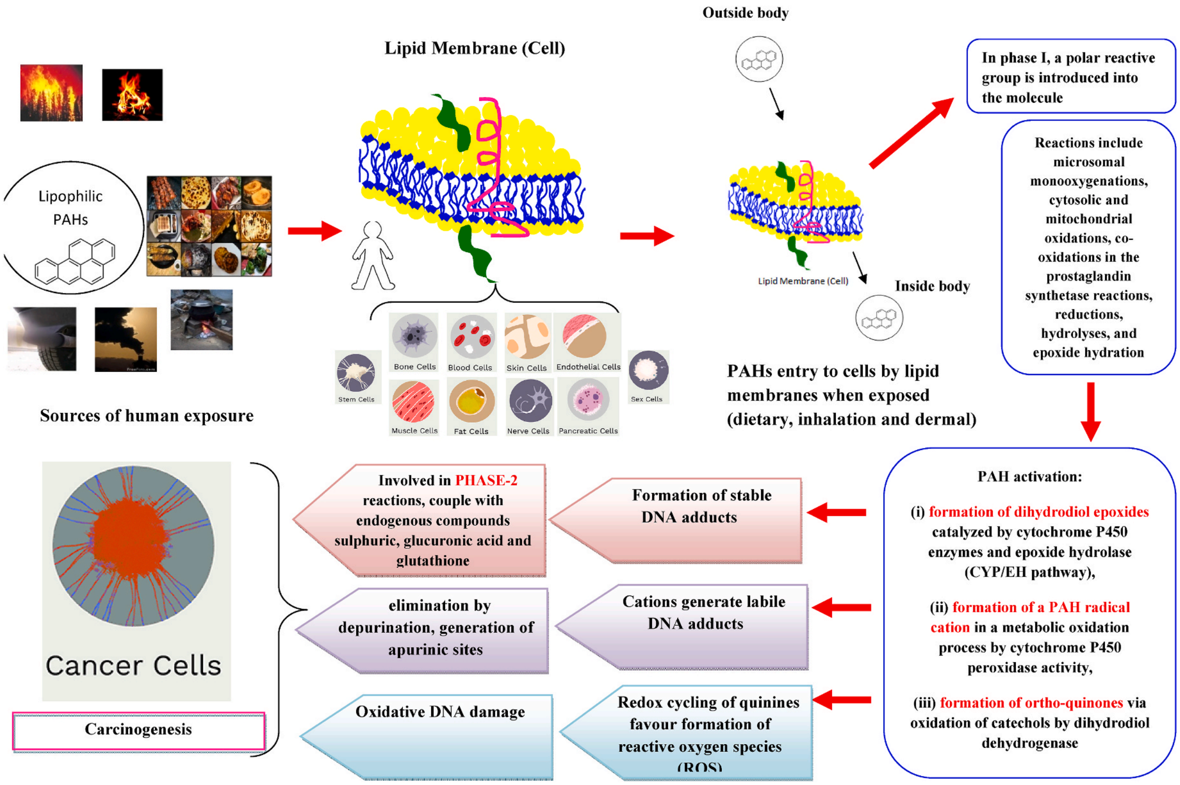

Fig. 1. Metabolic activities associated with PAHs entrance into the human body (details adopted from Ewa and Danuta, 2016).

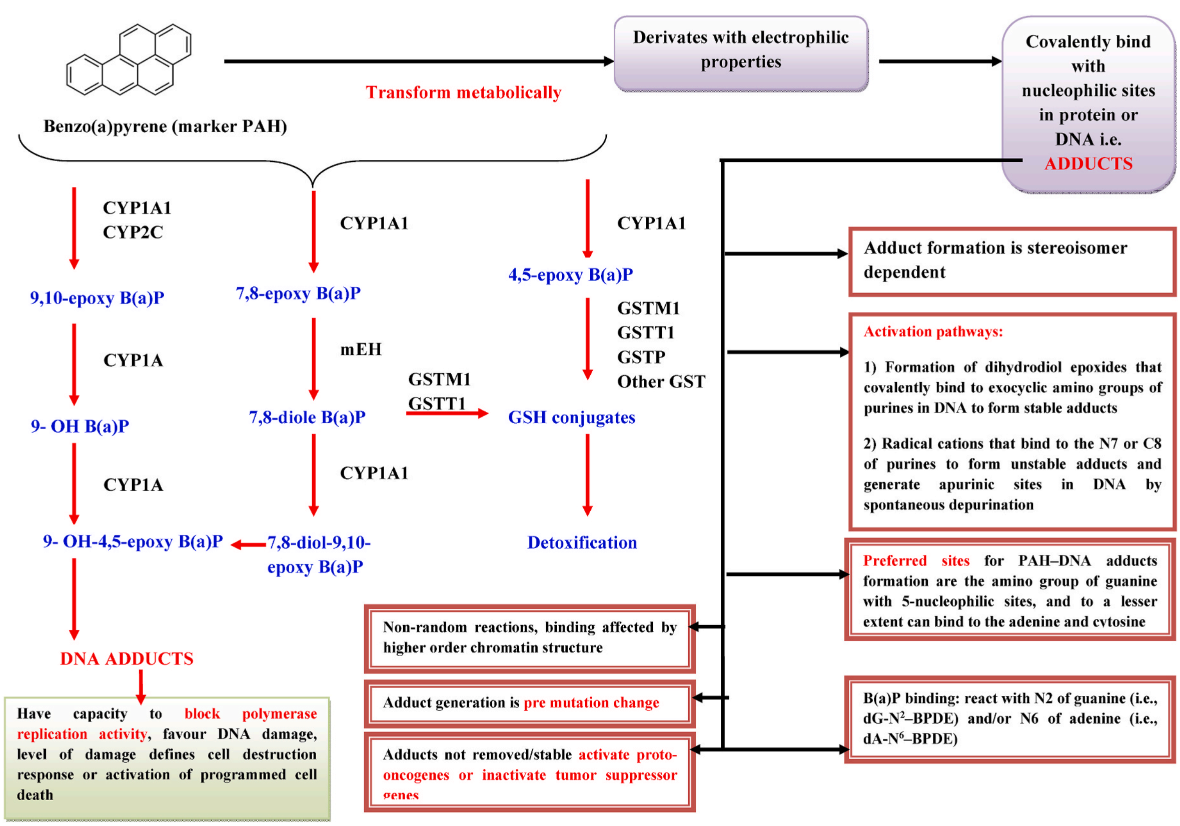

Fig. 2. Specific Metabolic activities associated with PAHs and DNA interaction (details adopted from Ewa and Danuta, 2016).

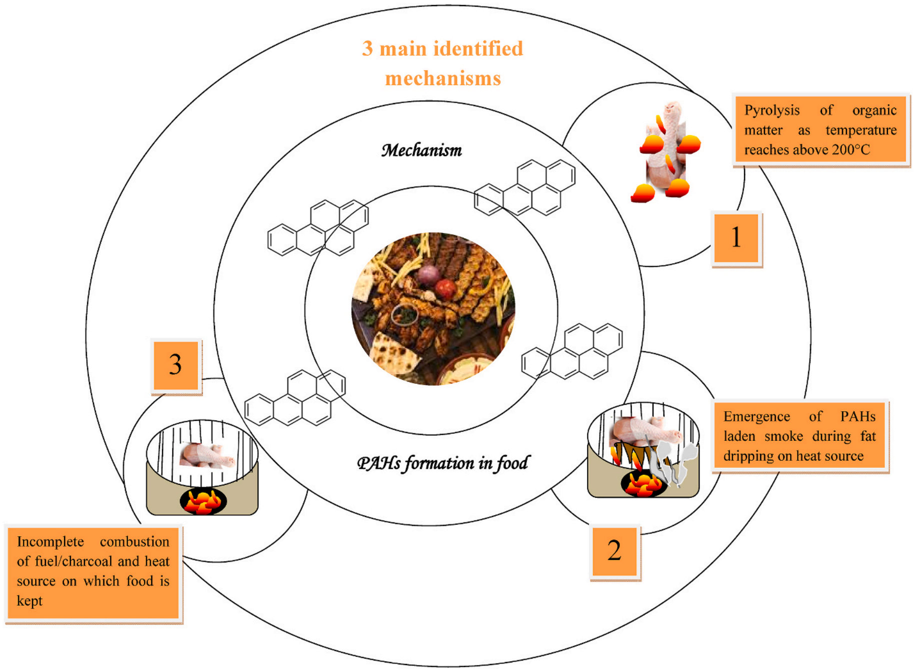

Fig. 3a. General Mechanism of PAHs formation as identified by Alomirah et al. (2011).

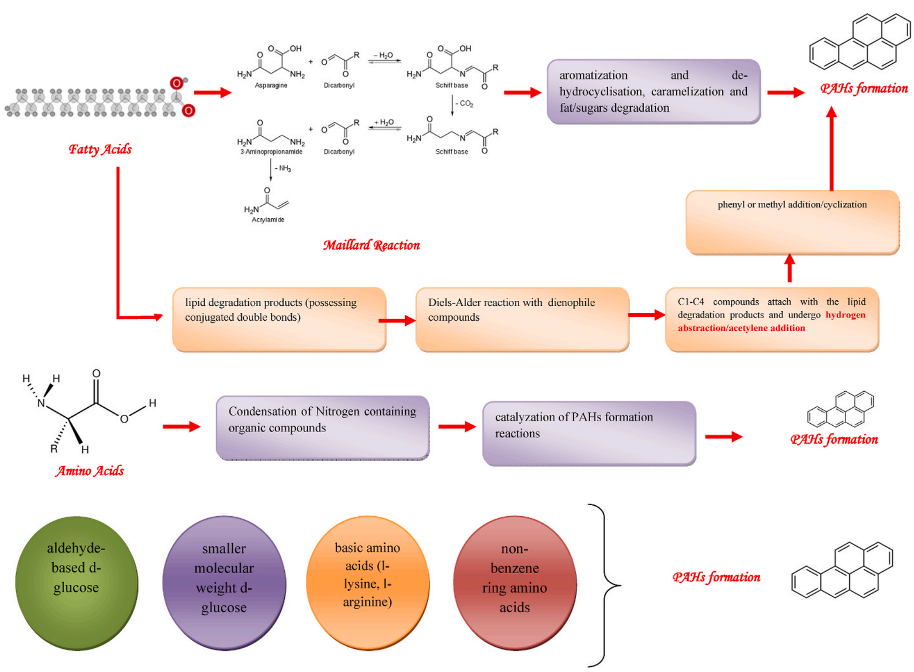

Fig. 3b. Mechanism of PAHs formation (reactions involved).

thereby reducing PAHs formation and accumulation. Since the different tea types possess variable polyphenol content (especially in their oxidized forms as they themselves undergo processing), the PAHs reduction levels achieved also differed in the study. It must however be noted that the mechanism described in this study contradicts the fact that a) phenolic compounds donate hydrogen to free radicals to become phenoxyl radical themselves, b) phenolic compounds upon pyrolysis forms PAHs and c) pectin and cellulose under pyrolysis situation act as donors for PAHs formation. It may thus be hypothesized that the probable candidate responsible for PAHs reduction may actually be different from ones investigated and thus warrants future research.

Similarly, involvement of free radicals in PAHs formation in food was reported by Viegas et al. (2014). The study utilized sundry beer marinades [Black beer (BB), non-alcoholic Pilsner beer (NPB) and Pilsner beer (PB)] in the charcoal-grilled pork and examined formation of PAH8. Lowest reduction (13%) occurred in PB marinated pork whereas NPB and BB marinade reduced PAH8 by 25% and 53% respectively. The DPPH radical scavenging activity of the marinades was found to be significantly correlated with the PAH inhibitory effect of marinades. Thus the radical scavenging and quenching activity of the beer marinades may be considered important for PAH8 inhibition. The diverse effect observed with beer types on PAHs reduction is attributed to

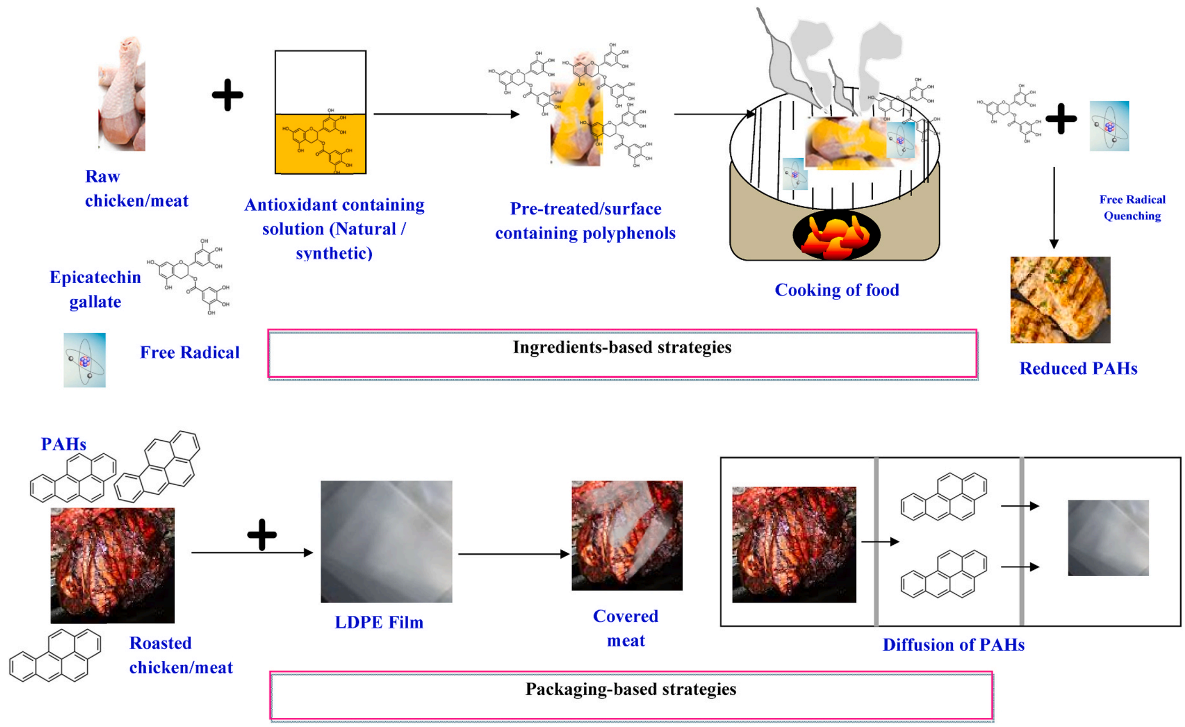

Fig. 4. Ingredients and packaging based strategies.

fermentation type, presence of sweeteners/glucose-fructose syrup, food colours/other additives; whereby food colours/additives incorporation enhanced the antioxidant activity of beers. Also, beer effect on oxidative species of meat surface favoured resistance in meat towards PAHs formation and accumulation.

Farhadian et al., (2011) explored the effect of seven marinades namely a) Commercial marinade b) basic marinade, which included onion, lemon grass, sugar, garlic, turmeric, coriander, salt, cinnamon and water c) basic-oil d) basic-lemon juice e) basic-oil-lemon juice f) basic-oil-tamarind g) commercial-tamarind and treatment duration (0, 4, 8 and 12 h) on the formation of B[*a*]P, B[*b*]F and FLTH compounds in grilled beef meat. The meat samples treated with acidic marinade (with 1.2% lemon juice) achieved a 70% reduction in PAH concentration. The antioxidant activity of spices was given as a probable reason for the observed inhibitory effect. Additionally, the role of organic sulphur compounds, present in the onion and garlic, in preventing the Maillard reaction was found to be associated with reduction in PAH's formation. In other study, pork meat dishes and gravy were analysed for PAHs in the absence and presence of onion and garlic to study their effect on the PAHs formation (Janoszka, 2011). Total concentration of 6 PAHs (B[*b*]F, B[k]F, B[*a*]A, B[*a*]P, D[ah]A, B[ghi]P) in meat samples ranged from 2.0 to 7.2 ng/g in cooked meat and from 0.05 to 0.6 ng/g in gravies. B[*a*]P concentration in meat and gravy samples was found to be 0.38–1.61 ng/g and 0.01–0.11 ng/g respectively. A reduction of 60% (pan-fried meat) and ~90% (in gravies) PAHs was observed with onion addition at the rate of 30 g/100 g. On the other hand, 15g garlic/100 g of meat was found to reduce PAHs levels in meat by 54% on an average while 13.5–79% in gravies. Destruction of fatty acid hydroperoxides by sulfhydryl and phenolic compounds and prevention of Maillard reaction was considered as a mechanism for PAHs reduction (Janoszka, 2011).

Similarly, García-Lomillo et al. (2017) analysed the effect of red wine pomace seasoning (RWPS) and high-oxygen atmosphere storage on PAHs' formation in barbecued beef patties. The levels of PAHs were low but were found to increase in control patties during 9 days of storage. This increase was found to be associated with the reduction in antioxidants concentrations. Also, the RWPS patties possessed higher PAHs levels than the control patties after cooking. RWPS is complex product, rich in flavonoids (phenolic compounds) and fibre; but it was stated that pyrolysis of these compounds favoured higher PAHs formation. It was suggested that the mechanism of phenolic compounds during the cooking of meat is different from the mechanism taking place during free radical scavenging. However, cooking of stored RWPS patties resulted in lower PAHs contamination owing to time dependent as well as non-immediate effect of the polyphenols. Moreover, RWPS accompanied with high-oxygen atmosphere may inhibit formation of cooking carcinogens in barbecued patties. This could be due to retarded pyrolysis, lipid peroxidation and loss of endogenous thiols groups under high-oxygen atmosphere. Gong et al. (2018) determined the formation of 16PAHs and 5 oxygenated PAHs in the frying oils (soybean and palm oil) after treatment with *tert*-butyl hydroquinone (TBHQ, synthesized antioxidant) and three natural antioxidants (rosemary extract, tea polyphenol and antioxidant of bamboo). The total PAHs in the youtiao fried in soybean oil reduced by 30.30% (TBHQ), 23.47% (rosemary extract), 11.38% (tea polyphenol) and 28.85% (antioxidants of bamboo) while reduction of 38.94%, 39.26%, 27.56%, and 9.45% was achieved with TBHQ, bamboo antioxidants, rosemary extract and tea polyphenol respectively in the youtiao fried in palm oil. This study further indicated the inhibitory role of natural as well as synthetic antioxidants on PAHs formation in food products.

In order to further investigate the role of oil types on PAHs reduction and the key factor responsible for same, the effect of changes in mixing ratio of oil, roasting temperature and time on PAHs levels (B[*a*]P, B[*a*]A, CHRY, B[*b*]F) in seasoned-roasted (SR) laver was determined (Kang et al., 2019). It was observed through this model system that the contribution of mixed oil (sesame oil, perilla oil and brown rice oil) and roasting process in PAHs formation in laver is up to 57% and 43% respectively while a 35% reduction in PAHs levels was observed when perilla oil was removed from mixed oil composition and roasting was performed at 350 °C for 10 s. The study demonstrated a significant role of mixed oil composition and heat treatment parameters in PAHs formation in roasted laver. It was also stated that the oils with high smoke points can degrade the products into fatty acids and facilitate the reduction process, hence a higher proportion of brown rice oil in mixed oil resulted in reduced PAHs concentration (Kang et al., 2019).

Rey-Salgueiro et al. (2009) reported that five and six ring PAHs occurred at high levels in mussels kept in aqueous natural sauce (salt, water and other spices) than on canning in pickle sauce (comprising of vinegar, salt, paprika, vegetable oil and other spices) due to their migration (about 12% of total PAHs) from mussels to canned pickle sauce. This difference could be a function of lipophilic nature of the PAHs because of which migration was observed in oil containing canned pickle sauce in comparison to aqueous natural sauce.

4.2. Packaging-based strategies

Wang et al. (2022) highlighted the use of packaging materials as a means for PAHs mitigation in processed food and also specified the need to check the viability of these methods for efficient removal. In 1993, Simko and co-workers investigated the changes in concentration of PAHs spiked (52 μg/kg) to liquid smoke flavourings (LSF) which was filled into low-density polyethylene (LDPE) packaging. It was observed that after one day of storage, the PAHs concentration dropped by more than 50% (B[*a*]P - from 5.58 to 3.71 μg/kg and total PAHs - from 45.60 to 21 μg/kg). The concentrations gradually reduced to the detection levels (B[*a*]P - 0.07 μg/kg and total PAHs - 0.73 μg/kg) at the end of 14 days' experiment. PAHs analysis in the packaging material revealed that the PAHs lost from the LSF migrated to the LDPE bottles (Fig. 4). PAHs diffused from the strongly polar medium into the non-polar packaging medium and van der Waals disperse forces were identified as the main influencing factor for the observed results. This process might have been facilitated by the non-polar nature of PAHs. In another study, LDPE was used to remove five carcinogenic PAHs (B[*a*]A, B[*a*]P, B[*b*]F, D[ah]A and IND) from three liquid models (aqueous, water-oil and water-oil + phosphatidylcholine model) and roasted meat. The LDPE film was able to remove more than 50% of the PAHs from the three liquid models with the highest removal from water-oil system and lowest from the phospholipid containing system. Reduction of PAHs could have been due to the diffusion in the solution and migration of PAHs from the polar water medium to the less polar ethylene alcohol followed by migration to the non-polar LDPE film. Similar polarity of compounds, decisive influence of van der Waals forces, viscosities of liquid media, molar mass and size of compounds and contact duration (film and media) were some of the prominent factors found to affect the sorption of different PAHs on the LDPE film. PAHs migrated to the LDPE were degraded to 70.8–84.0% by 3 h of UV irradiation, B[*a*]P being the most sensitive to UV among the 5 PAHs. This is owing to the absorption of UV light by PAHs and conversion to ketones, aromatic alcohols or ethers by oxidation of peripheral carbon atoms, thereby facilitating photolysis. On the other hand, levels of three PAHs (B[*a*]A, B[*b*]F and B[*a*]P) detected in the skin of roasted duck meat were found to reduce by 9–74% after 24 h of storage. The PAHs could have moved from the roasted meat skin to the LDPE film through contact (Chen and Chen, 2005). Simko et al. (2006) spiked B[*a*]P at 37.1 and 38.6 μg/kg concentrations in commercially available sunflower oil and the distilled sunflower oil respectively. Afterwards, the oil was stored in polyethylene terephthalate (PET) and low density polyethylene (LDPE) receptacles for at least 49 h. It was found that PAHs in the non-distilled oil stored in PET receptacle reduced to 25.9 μg/kg while distilled oil possessed 34.6 μg/kg B[*a*]P concentration. The study revealed that B(*a*)P got sorbed on the PET surface resulting in removal of PAHs from oil, but the presence of other compounds (such as fatty acids, free electron pairs etc.) affects the rate and extent of B[*a*]P removal from oil. Contrarily to the above study, LDPE in this study proved as an inappropriate material for PAHs reduction from sunflower and rapeseed oils, and maintained the B[*a*]P concentration constant throughout the experiment.

Semanova et al. (2016) packed smoked meat sausages in the o-polyamide/low density polyethylene laminated film and studied the behaviour of four PAHs (B[*a*]A, CHRY, B[*b*]F and B[*a*]P) for 180 min. In the packed sample, total concentration of four PAHs reduced by 81% and for individual PAHs the reduction ranged from 72 to 88% in the 180 min' duration. PAHs concentration remained constant in the unpacked sausage sample. The authors explained based on the diffusion of PAHs from the smoked sausage to the LDPE packaging and the process to be governed by the Fickian diffusion law. Machado et al. (2014) analysed the PAHs concentration in cachaça stored in polyethylene (PE) tank and glass bottles and found significantly higher PAH concentration in the drink stored in the PE tank (51.57 μg/L) than glass bottles (6.07 μg/L). Drink from both the storage methods was found to have 3-ring PAHs but 5- and 6- ring PAHs including B[*a*]P were identified in the drink stored in PE tank. PE tank contributed nearly 7-fold higher PAHs to the drink as compared to the glass bottles demonstrating significant impact of the storage vessel. Li et al. (2017) also reported migration of PAHs from the polystyrene food contact material to water used as food simulant. Joo et al. (2021) stated that the hydrophobic nature and surface area of plastic favours adsorption of PAHs and other contaminants on it, and is a function of heterogeneous environmental conditions inclusive of plastic characteristics (surface property, structure, binding energy and composition), release medium parameters (like pH, salinity, temperature, ionic strength) and contamination factors (charge, redox state, solubility, stability). It was also stated that in comparison to bare plastics, PVC based plastics reacted significantly with B[*a*]P and offered higher toxicity in time- and dose-dependent manner.

In another study, Garcia-Falcon & Simal-Gandara (2005) investigated the effect of barrel charring process on the presence of 7PAHs in alcoholic beverages considering the type of charring (traditional or convective) and the charring intensity (light, medium or heavy) as the main parameters. It was observed that the convective charring could control the PAHs formation in wood in comparison to the traditionally charred barrels. This is because convective charring is performed using flameless procedure whereby hot air is circulated in closed/clean environment while traditional charring/toasting of barrels involve direct contact of wood-fire.

The effect of outer covering as a means of PAHs reduction was also investigated by Farhadian et al. (2011). The study included two treatments of meat samples before charcoal grilling i.e. a) steam and microwave preheating and b) aluminium and banana leaf wrapping, and found very promising results. Samples with steam and microwave preheating or aluminium wrapping treatments did not possess carcinogenic PAHs (B[*a*]P and B[*b*]F). Up to 46% and 81% reductions were observed in FLTH content in beef and chicken samples respectively. In charcoal-grilled beef samples (without wrapping) FLTH was found at 144 ng/g which reduced respectively to 83.3 and 92.2 ng/g with aluminium and banana wrapping. Banana wrapped samples possessed 0.49 ng/g and 1.19 ng/g B[*a*]P and B[*b*]F respectively. In charcoal-grilled chicken samples (without wrapping), FLTH was 165 ng/g which reduced to 45.5 and 30 ng/g in the aluminium and banana wrapped samples respectively. B[*a*]P and B[*b*]F in banana wrapped samples occurred at 1.48 and 3.29 ng/g, respectively. Significant difference ($p < 0.05$) in PAHs' reduction levels was observed between the aluminium and banana based packing of meat. It was thus concluded that aluminium packing can reduce PAHs content (B[*a*]P and B[*b*]F) in meat samples by 100%. These results were explained to be an effect of reduction in fat drippings onto the fire used for cooking meat. With banana wrapping, the burning of leaves adversely affected the reduction process as it consequently exposed meat to smoke. The burning phenomenon, however, was not found associated to aluminium wrapping. Reduction associated with steam or microwave pre-treatments is due to the less time taken by these samples to cook by charcoal grilling which led to lesser pyrolysis of food, lesser deposition and penetration of smoke particles generated from fat drippings on heat source (hot coal).

4.3. Adsorbents and filters-based strategies

Removal of PAHs from smoke has been a topic of interest for many industries (Šimko, 2005). One of the lucrative methods is treatment with adsorbents and filters. Parker et al. (2017) utilized zeolite filter for

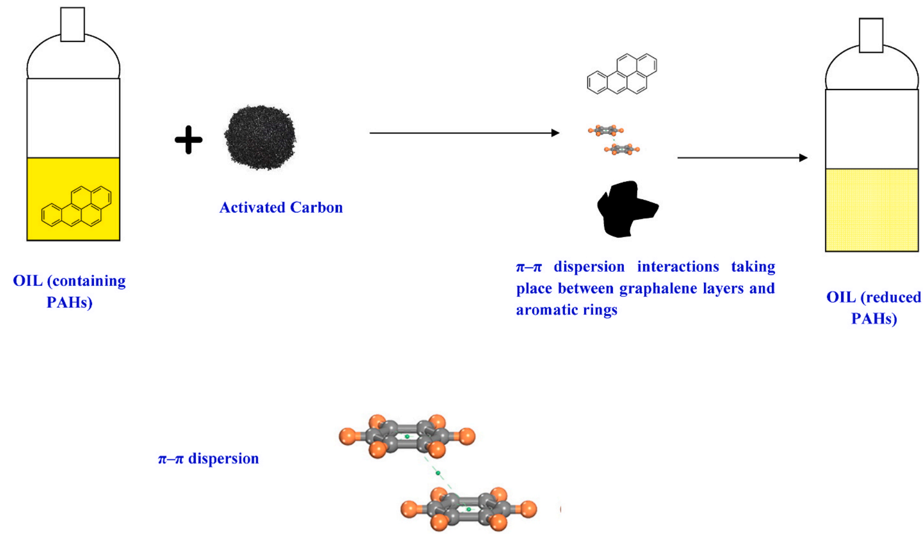

Fig. 5. Adsorbent-based strategies.

removal of PAHs from 70 types of products from food industry. It was observed that incorporation of filter reduced PAHs contamination by 90% in simple matrix or solution (Fig. 5). Zeolite also known as clinoptilolite is a naturally occurring aluminosilicate or crystalline porous materials, which has the capacity to accommodate different cations. They can interact with organic molecules possessing positive charge through superficial properties inclusive of unique modifiable surface, structure as well as chemical composition; high surface area; better adsorption and condensation properties. The three-dimensional framework is derived through tetrahedral SiO_4 and AlO_4 unit, with AlO_4 giving a negative charge to the framework making them easily exchangeable with compounds. Besides this exchange capacity, the zeolites can interact with PAHs through π-electrons in the PAH (i.e., van der Waals forces) (Fuss et al., 2021). In this study, however reduction in coconut oil was not considerable. This could be because of more lipophilic nature of the oil in comparison to simple matrices which facilitated retention of PAHs.

Shin et al. (2015) studied effect of different adsorbents (activated carbons-D12 and D40, and Celite) on B[*a*]P removal from sesame oil. It was observed that B[*a*]P contamination in oil was 20 μg/kg which reduced by 32% and 46% by D12 and D40 adsorbents respectively. Contrarily, the reduction with Celite was found to be non-significant. Activated charcoal has been considered to effectively interact with non-polar and hydrophobic organic compounds (including B[*a*]P) through their parallel stacked carbon atoms whereas Celite mostly react with planar compounds through its silanol groups (with silicon dioxide) on the surface. Conclusively, D12 was found to be most promising adsorbent. In another study, Yebra-Pimentel et al. (2014) compared the efficacy of activated carbons (AC1, AC2, AC3), and sustainable adsorbents [wood ashes from a timber industry (WA1), mussel shell ashes (SA) and pine wood ashes (WA2, WA3)] calcined at two different temperatures (500 and 800 °C, respectively) for the removal of PAHs from fish oils. The removal efficiency was found to be well correlated with the molecular weight of individual PAHs. A reduction of 80–100% occurred with activated carbons [62–100% (AC1), 83–100% (AC2) and 10–100% (AC3)] while 10–100% reduction was associated with sustainable adsorbents. Pine wood ashes however exhibited 87–100% (except for two PAHs) removal which was equivalent or at par with that of activated carbons. The observed differences in PAH removal efficiency of activated carbon and sustainable adsorbents may be attributed to the differences in adsorption rates due to the material characteristics such as pore size, BET surface area, origin, total pore volume, composition and particle size. Essumang et al. (2014) modified a traditional kiln and placed charcoal filters to remove PAHs contamination in smoke-cured fish. Total PAHs concentration was found to be 517.33–751.56 mg/kg in the smoke cured samples with no charcoal filter in the kiln, 212.56–472.98 mg/kg in the samples with “already made” activated charcoal filter and 248.64–454.77 mg/kg with “locally made” activated filter. The reduction and removal of PAHs from smoke may be attributed to its adsorption on filters facilitated by lower average temperatures of charcoal filter beds (101.5–119.3 °C and 98.5–122.3 °C for “already made” and “locally made” activated charcoal respectively], micropore volumes and lower fat dripping on fire zone (filter bed acting as a barrier).

If closely seen in the mechanism of PAHs removal through these charcoal and other adsorbates, the material properties and characteristics play an important role as discussed above. PAHs’ dimensions and molecular conformations also play important role in adsorption; whereby van der Waals forces result in activated carbon adsorption. Extreme polarities on either side (i.e. more planar or more non-polar) of adsorbate favours higher interaction, and form co-planar structures. Specifically in case of activated carbons, aromatic compounds adsorb on surface in flat position owing to the π–π dispersion interactions taking place between graphene layers and aromatic rings. Moreover, the planar PAHs are able to access and pack in the pores (slit-shape) of the activated carbons more easily and effectively than non-polar molecules and thus present higher adsorptions.

4.4. *Apparatus-based strategies*

Yi et al. (2017) developed an apparatus to reduce the B[*a*]P contents in sesame seed oil (SSO). Also, the effect of antioxidants in SSO was analysed for their role in B[*a*]P reduction. The apparatus had a provision for washing of sesame seeds followed by spin-drying process before roasting. Ventilation was maintained to prevent B[*a*]P formation and re-absorption into oil. It was observed that the B[*a*]P levels in SSO was 3.13 μg/kg when processing was done at optimized conditions without washing. On the other hand, PAHs contamination increased respectively to 2.93 μg/kg, 4.79 μg/kg, and 7.11 μg/kg upon inclusion of one, two and three washing cycles. Strong reduction (2.93 μg/kg) for B[*a*]P was achieved with 1350 rpm of ventilation. Washing facilitated more moisture absorption by seeds which further favoured dehulling process. Dehulled seeds possessed more fat and protein contents in meal and greatly contributed in B[*a*]P formation upon roasting. This is because of the fact that PAHs are lipophilic in nature, more fat in seeds favour more accumulation of PAHs. In addition to PAH reduction by ventilation, the reduction was also caused by presence of sesamol. The compound posed stronger antioxidant capacity at high temperatures by involving in diverse reactions based on free radicals, cyclization at inter-molecular level and polymerization. On the other hand, B[*a*]P content in SSO reduced from 8.98 μg/kg to 4.81 μg/kg when spin drying process was included for 1 min. Further reduction to 2.93 and 2.02 μg/kg was observed with 2 and 3 min of spin-drying. Reduction to 1.98, 1.58, 1.09, and 0.99 μg/kg was observed associated with incorporation of thermal drying for 0, 3, 5, and 10 min respectively.

Bomfeh et al. (2019) also used an improved kiln FAO-Thiaroye Technique (FTT, introduced by the Food and Agriculture Organization of the United Nations) and determined the PAHs contamination in smoked fishes. It was observed that B[*a*]P and PAH4 was 91- and 63-fold lower in FTT products than metal drum (a type of traditional kiln) products, and 215- and 183-fold lower in comparison to Chorkor smoker (a type of traditional kiln) products. The highest accumulation of PAHs in Chorkor smoker products is due to the concentration of smoke around fish since many trays are incorporated at a time in such system. Operational system in the traditional kilns involve use of wood as fuel and also involves fat drippings, which results in PAHs accumulation in fishes. Contrarily, FTT design works with fully-lit charcoal than fuelwood, prevent dripping of fats onto heat source and prevent direct contact between product and heat source. Moreover, the design facilitates filtering of smoke, thereby resulting in indirect smoke flavouring. Heat retention stones work well with charcoals and smoke generated from sugarcane baggase gets filtered through the moistened luffa sponge; thereby an advantage is added to FTT to prevent PAHs accumulation in food product. It was also stated that usage of luffa sponge (a readily available renewable resource) in FTT makes it more economical and sustainable approach than activated charcoal option.

4.5. Treatments-based strategies

The removal of PAHs from wheat kernels using gamma irradiation (0, 5, 10, and 15 kGy doses) was investigated by Khalil and Al-Bachir (2015). It was observed that highest irradiation dose was most effective against all PAH compounds decreasing their concentration from 154 μg/kg to 21 μg/kg while B[*a*]P reduced by more than 50%. Contrarily, 5 kGy gamma-irradiation (γ-ray) adversely affected ACY and PHEN compounds by increasing their content by 260% and 340% respectively whereas 84%, 40%, 42%, and 35% reduction occurred in NAPH, ACE, FLU and ANTH compounds at same dose of treatment. On the other hand, some HMW PAHs (FLTH, PYR, D[ah]A, B[ghi]P, IND) disappeared completely at this dose. More effective removal of HMWs occurred at 10 kGy of dose. However, the response for individual PAHs differed. B[*b*]F and B[*a*]P concentrations reduced by 88% and 30%, but CHRY and B[k]F compounds were least affected by this dose of radiation. In another study, PAHs reduction of about 35% occurred in wheat grains when subjected to 1 kGy gamma-irradiation dose (Khalil et al., 2016). This reduction may be attributed to the ozonolysis-radiolysis effect whereby these compounds are made to decompose to form aldehydes or carboxylic acids. The mechanism is initiated by formation of free radicals. For example, fluoranthene (FLTH) decomposition begins with multiple site reaction of FLTH molecule with OH-radicals to form OH adducts which dimerize and react with other bulky radicals of matrix. These then convert to $HO_2\cdot/O_2^{\cdot-}$ species in presence of air (H) and eq^- to form peroxyl radicals. These peroxyl radicals then split or open to react with other species to form low molecules (aldehydes, carboxylic acids etc.) (Popov and Getoff, 2005).

Kiralan et al. (2017) investigated the effect of microwave heating [low – 60W, medium-low – 150W, and medium – 270W each for 270, 120 and 90 s] and vacuum drying pre-treatment of olive pomace (before conventional drying process) as a mitigation strategy. Total PAHs were found to reduce by 75% with significant effect on four of the PAHs. Conventionally dried oil pomace possessed 3471 ± 993 μg/kg of total 15PAHs which reduced to 2292 ± 438 μg/kg, 1999 ± 59 μg/kg, and 875 ± 138 μg/kg with microwave pre-treatment at 270W for 90 s, 150W for 120 s and 60W for 270 s respectively prior to drying application. Low power condition (150W for 270 s) was identified to give the best results. Moreover, vacuum drying resulted in 815 ± 246 mg/kg of PAHs content. The effect of microwave and steam pre-heating on the PAHs (B[*a*]P, B[*b*]F and FLTH] contamination in charcoal grilled beef and chicken samples has also been reported by Farhadian et al. (2011). The concentration of FLTH in charcoal grilled beef samples was found to be 143 ng/g which reduced to 77.6 ng/g and 86.3 ng/g with steam and microwave pre-treatments respectively. Similarly, charcoal grilled chicken possessed 165 ng/g FLTH which reduced to 79.1 and 65.0 ng/g with steam and microwave pre-treatment. On the other hand, 100% reduction was observed for the other two PAHs (B[*a*]A and B[*b*]F) in both chicken and beef samples. The duration of charcoal grilling with pre-treatment was lower (2–4 min) than charcoal grilling process (8 min) without pre-treatment which could have resulted in reduced pyrolysis duration, lower penetration of smoke generated from fat drippings and lower depositions consequently reduced PAHs. It has been stated that microwave energy is absorbed by contaminant and the food material which convert to heat and results in selective heating (generated when radiation passes material having lower dielectric loss factor and reaches molecules possessing high dielectric loss factor). PAHs have higher dielectric properties than matrix and thus convert absorbed MW energy to heat; thereby leading to higher localized temperature of contaminants. The difference brought in boiling point of contaminant and matrix then favour removal of contaminant by thermal decomposition. However, the parameters such as physical and chemical properties of matrix, interaction/adsorption-desorption of contaminants on matrix molecules, surface area etc. are critical factors affecting the extent PAHs removal (Falciglia et al., 2016). Li et al. (2021) has however stated that the use of γ-ray or high-energy electron is however prohibited in most of the countries. Harell et al. (2018) stated that irradiated foods raise serious concern with respect to their genomic instability and oncogenic potential. This exhibits the need to investigate effects of such consumables through long term and prospective clinical trials. All the more, such irradiations may degrade PAHs to other toxic forms and extensive studies are required to assure safety of these procedures.

Probiotics are microbes which balance the intestinal flora of humans/animals, leading to several health benefits. Bartkiene et al. (2017) determined the effect of lactic acid bacteria (LAB) on the formation of PAHs in smoked sausages. Significant effect of fermentation was observed on the content of B[*a*]A, B[*b*]F, B[*a*]P and CHRY compounds. The efficiency of these microbes in detoxification, xenobiotics' transformations and conversion to less toxic compounds/metabolites were considered as probable mechanisms of PAHs reduction. Yousefi et al. (2019) stated that probiotics can biotransform xenobiotics to less toxic forms as well as can physically interact with toxins and absorb them on their surfaces. This binding at cell wall is regulated by several other parameters such temperature, matrix/media, pH, strain type, population, toxin type and concentration etc. This binding capacity of microbes is in general facilitated by the anionic functional groups (teichoic acids, peptidoglycan, and teichuronic acids) and other anionic cellular characteristics. Presence of enveloped proteins, lipoteichoic acids and other carbohydrate contents give the microbial cell capacity to bind to hydrophobic compounds.

On the other hand, Mahugija and Njale (2018) observed changes in PAHs content of smoked fishes upon washing with warm water (60 °C). A reduction of ~35.8%–100%, 0.6%–100% and 2.9%–100% was found for individual PAH content in smoked *S. victoria*, *L. niloticus* and *Haplochromis* spp samples respectively. Reduction in PAHs content with washing process was found to vary with fish species Washing process in general removes the PAHs adsorbed on the surface of the fish through desorption and may either reduce or eliminate the existing PAHs.

One of the main factors contributing to PAHs contamination during cooking is use of coal. Type of charcoal, volatile content and the process of its formation play a major role in PAHs release upon burning. Chaemsai et al. (2016) had investigated the effect of carbonization temperature of coal on PAHs releasing in smoke. Coal carbonized at 500

°C released 19.86 μg/g, 1.23 μg/g and 0.69 μg/g PAHs with a burning duration of 5 min, 20 min and 5 h respectively. The amount of PAHs changed to 0.95, 1.25 and 0.28 μg/g for the coal carbonized at 750 °C and 1.38, 0.82 and 0.15 μg/g for the coal carbonized at 1000 °C with a burning duration of 5 min, 20 min and 5 h respectively. It was concluded that food cooked using the coal carbonized at higher temperature may possess lower PAHs content. It has been stated that the coal carbonized at higher temperature possess more of carbon, facilitating lower formation of methane, limited release of volatiles and PAHs upon combustion. Another possible treatment is the usage of glow smoke for hot smoked sausages (Jira et al., 2013). Glow smoke may be produced by controlling smoke generation temperature below 600 °C and thus the smoke produced and deposited on food product in contact may be less.

Nowadays, replacement of saturated fats with unsaturated fatty acids has gained immense popularity among researchers and meat processors. For example, introduction of olive oil enhances mono-unsaturated fatty acids (MUFAs) content in final products while sunflower oil and grapes seed oil enhances polyunsaturated fatty acids (PUFAs). Lu et al. (2017) analysed the effect of replacing pork back fat with different fats (olive oil, sunflower oil, and grapes seed oil) on PAHs concentration. Also, the effect of cooking temperature on PAHs formation in pork patties was observed. The total PAHs content was found to range from 1.59 ± 0.26 ng/g to 3.84 ± 0.21 ng/g with a dominance of B[*a*]P in all the samples (1.44–3.53 ng/g). The formation of B[*a*]P was unaltered by cooking temperature, however fat type had a significant effect on PAHs formation. Olive oil and grapes seed oil inhibited B[*a*]P formation in the patties cooked at 220 °C, while no effect was observed at 180 °C. In contrast, sunflower oil inhibited B[*a*]P formation at 180 °C but enhanced its formation at 220 °C. The authors concluded that high temperature conditions promoted PAHs occurrence in food owing to pyrolysis of fat, and the difference in smoking point of the tested oils explained the variability in PAHs content in the food product. Lower smoke point (227 °C and 216 °C) is associated with sunflower oil and grapes seed oil due to the presence of PUFAs which make them susceptible to decomposition at a lower temperature than olive oil (242 °C). The free radicals generated through decomposition of oil accelerate PAHs formation and thus was responsible for observed phenomenon.

5. Factors affecting PAHs removal

Concerns over their adverse effects in food and environment, and therefore human health, have resulted in extensive studies on various types of PAHs removal methods. Thus, awareness about the process conditions favouring high accumulation and formation of PAHs in processed food products may help in reducing many health hazards related to PAHs.

5.1. Composition and surface adsorption properties

Simko and Brunckova (1993) stated that the use of packaging film or material for PAHs reduction in smoke flavours is highly dependent on the van der Waals disperse forces, laws of mass transfer and diffusion. van der Waals forces specifically affect the sorption of non-polar PAHs on the packaging material. The other factor which was found to affect the contamination reduction from packaging is the thickness of the film. Simko et al. (1995) stated that the PAHs are likely to re-diffuse from film to the food material if the thickness of film is low. Diffusion coefficient or migration properties of individual PAHs also play a role during the process. The other influential factors include: surface affinity based on polarity, viscosity of liquid media, molar mass and size of the compounds, exposure of packaging material to radiation such as UV facilitate conversion of PAHs to aromatic ethers, alcohols, and ketones by oxidation of peripheral carbon atoms, nature of storage vessel and existing contamination in vessels, presence of auxochromes and chromophores etc. (Chen and Chen, 2005; Li et al., 2017; Machado et al., 2014).

Shin et al. (2015) reported that the surface properties, pore size distribution, porosity, presence of functional groups differ in activated charcoals and thus guide the efficiency of PAHs removal. Microspore volume is an important parameter which also determines PAHs adsorption by activated carbon. Results also differ with the type of oil, the increased mesh size of charcoal (favours removal) and dimensions of B[*a*]P (similar pore size favour adsorption). Yebra Pimentel et al. (2014) also highlighted origin, composition and particle size of the adsorbent as influential factors. Molecular conformations and dimensions of PAHs were identified as other factors. Planar and non-polar PAH's been found to interact more with adsorbent (activated carbon). It was also reported that calcination temperature enhances number of micropores and surface activity thereby incrementing adsorption potential of the material. Essumang et al. (2014) reported a major effect of using filter in kiln on PAHs levels especially for fat-rich (sardines, mackerel etc.) fishes as the filter acts as barrier for fat drippings. It was highlighted that activated charcoal has smaller grain sizes; good packing and large surface area and thus enhances PAHs adsorption when used as filter in kiln. Lamichhane et al. (2016) had reported role of PAHs' solubility, salinity (aqueous solutions), pH of solution, and temperature of adsorption reaction as the other parameters governing adsorption/absorption capacity of the adsorbents.

5.2. Factors influencing their formation reduction or removal increase

Farhadian et al. (2011) reported that PAHs accumulation in meat is also dependent upon the rate of pyrolysis, deposition of the smoke particles, fat dripping and penetration of smoke; and play major role in reducing PAHs when microwave and steam treatments are followed. Similarly, reduction of PAHs using probiotics was found to depend on the type of strain used, food product type, conditions of food processing and for growth of microbes, viability of cells etc. (Bartkiene et al., 2017). Dietary fibre and wheat bran was found to have strong affinity for PAHs (Zhang et al., 2016). Extraction of fibre removed hydrophilic residues (starch) from bran and enhanced the roughness of fibre surface. This led to higher fibre affinity for PAHs than bran. This suggested that fibre can efficiently remove toxins and may reduce the risk associated with dietary intake of these toxic compounds.

Similarly, the reduction achieved through washing of smoked fish was found to be a function of type or species targeted, severity of washing, temperature of water, duration of washing process and the stage at which washing is conducted (before or after cooking) etc. (Mahugija and Njale, 2018). Chaemsai et al. (2016) highlighted the role of temperature at which charcoal is formed in the observed PAHs reduction. Also, presence of volatile matter in the charcoal was found to play a significant role. Lu et al. (2017) stated that reduction is also a factor of the type of fat present in a food product. Yi et al. (2017) stated that dehulling of sesame seeds may result in abundance of glutamic acid and thus contributes towards PAHs formation. Bomfeh et al. (2019) stated that kiln design and processing fuel largely determine PAHs levels in smoked fish.

6. Future prospects/challenges

Evidence of a reduction of PAHs in meat and fish products has been documented with different strategies. To name a few, by heating or wrapping meat products or using a casing before grilling, PAH contamination is reduced. The presence of filters in the smoking equipment reduces the amount of PAH in the smoked fish. Using marinades on meat/fish before grilling or smoking can lead to a reduction in PAHs in these processed foods. Packing smoked fish or meat in low-density polyethylene bags can also reduce PAH levels. Future research studies are necessary in this regard to completely understand the PAH's formation processes for developing efficient mitigation measures.

6.1. Packaging material

It has been found that introduction of recycled plastics as packaging material may act as a source of PAHs. Possibility lies that the PAHs may migrate from packaging materials into the non-polar food products such as oils, bacons, milk fats etc. Therefore, it is necessary to control contamination level in recycled plastics and guarantee their toxicological safety. Attempts are also needed to prevent direct contact between food and packaging materials (Simko et al., 1994). Some studies have shown that recycling itself may not affect PAHs in the recycled packaging material since the boiling points of PAHs are higher than the temperature achieved during recycling process (Simko et al., 1995). Thus future research studies must be performed to determine the relationship between recycled packaging and PAHs contamination in stored food products. Also, information on different packaging materials must be explored as a way for elevating food safety. The role of auxochromes and chromophores on PAHs formation in packaging material must also be investigated.

It has been observed that calculated diffusion coefficients may help determine transfer of PAHs from packaging material in food products. Constants may be utilized for other food geometries and making predictions for them (Semanova et al., 2016). However, this needs to be confirmed through future research studies. Li et al. (2017) stated that PAHs migration in real foods (consumed daily) than simulated model may be affected by water used as food simulant. Also, the time-temperature combination may affect the migration value. Besides this, knowledge of measurable by-products and processing aids (which are used in plastic synthesis and may be present in end product) must be developed for safety assessment of plastics as food contact materials. Any impurities attached may thus migrate depending on plastic structure and food components and may also influence migration of PAHs.

6.2. Adsorbents

There is a lack of cognizance about PAH adsorption capacity of activated carbons of different commercial grades (Yebra-Pimentel et al., 2014). The capacity of zeolite needs to be determined for PAHs removal from smoked foods such as smoked sauces, smoked spices, and other key food ingredients (Parker et al., 2017). However, any addition must not be done at the cost of consumers' safety and food quality. Thus studies on these aspects are warranted.

6.3. Apparatus

There is a need of macroscale modelling of roasting to study emission rate of roasting gases and corresponding thermal distribution of the roasting machine in order to facilitate B[*a*]P reduction through the developed apparatus (Yi et al., 2017). Also chemical changes in sesame seed oil (SSO) obtained at different conditions needs to be explored to clarify reduction mechanism and understanding about developed technology.

6.4. Treatments

Khalil et al. (2016) demonstrated that PAHs reduction through gamma irradiation exhibits linear and exponential trends. Future studies focusing on wider irradiation dose ranges for different kinds of foods must be explored to further determine possibility of PAHs mitigation.

Alternative strategies must be worked out for reduction of PAHs in olive pomace. Kiralan et al. (2017) suggested innovation at industrial level to incorporate microwave preheating and microwave system (tunnel form) for PAHs reduction in such products. Scaling-up inclusive of mathematical modelling studies are needed for such developments. Knowledge of dielectric properties (dielectric constant and relative dielectric loss factor) need to be enhanced to effectively develop mathematical models and control temperature change during microwave pre-drying process of olive pomace. Maintaining temperature and surface moisture uniformity is another problem to be dealt while using microwaves. Research must be conducted to substitute rotation based microwave system with air impingement, and infrared heating to maintain uniformity and constant temperature.

There is a need to determine mechanisms by which microbes and probiotics prevent colon cancer (Nair et al., 2021). Studies focusing on effect of washing of raw food on PAHs concentration in processed food are scarce (Mahugija and Njale, 2018). There is also a need to develop cognizance about the composition of charcoals used for cooking as in most of the cases the information is not available due to their production at different place from the place of use (Chaemsai et al., 2016). The study stated that high carbonization temperature reduces PAHs contamination in the smoke generated from charcoal. However, the process involves cost and low yield. Thus, attempts must be made towards developing cost effective measures.

Future research is needed to understand the role of antioxidants in oils on PAHs reduction (Lu et al., 2017). The study stated lack of knowledge about impact of tocopherols and phenolic compounds on the PAHs in meat.

7. Conclusions and recommendations

Since severe health consequences are attached with the chemical contamination of food, it becomes necessary to develop regulations and actions to limit their occurrence and accumulation throughout the supply chain. The limits defined must not be too close to the detection limits (LODs), as these add the analytical challenges and affects the enforcement of safety during food supply. Also, the concept of risk and the levels of contaminants in context with Acceptable Daily Intake (ADI) must be effectively communicated to the public. Polycyclic Aromatic Hydrocarbons (PAHs) may be categorized in the same group as per their severity and application of similar analytical methods used for detection and process control mechanisms. This may aid in designing the mitigation efforts effectively. Priority for addressing chemical contamination differs among different regulatory bodies. As a result, formulating the comprehensive program for controlling the contamination at global scale becomes complex. Thus need occurs in considering several factors such as contamination source, ingredient's origin, food adulteration, contaminants development during manufacturing, etc. in an effort to control the level of contamination. Many PAHs formation reduction mechanisms remain unknown. Although some studies observed a removal/degradation increase, there is not clear information about degradation derivatives, if any. Scientific principles may be established to develop a risk based program for managing contamination in food ingredients and will aid in ensuring products safety and global regulatory compliance.

Some recommendations which must be taken into account while suggesting a process/method as PAHs removal/reduction are:

1) Use of packaging material: Packaging material has shown promising results in reducing PAHs in wrapped foods. But, it must also be noted that the packaging material itself does not add any other contaminant to food.
2) It must be noted that when food comes in contact with external factors/additions (direct addition of activation charcoal to oil etc.); these factors do not harm or change food properties adversely. These products must not act as hazards. Proper removal/extraction of these factors after PAHs removal must be ensured.
3) Combination of marinates/spices etc. used as food PAHs regulators must be formulated considering their additives effect on human body, gastro-intestinal digestion, pH and basic element of food. Any alteration and random mixing without proper knowledge of the basic nature of these ingredients may promote PAHs formation or related toxic products and may affect human health.

4) The probiotics may degrade the PAHs in food and may enter human body. It must be ensured that the form of these probiotics does not change when they come in contact with PAHs. As well it is needed to know how these probiotics will get affected when the food is processed. The information on these aspects is not well illustrated and studied, and requires research.
5) It is further recommended to follow a holistic approach to determine the effect of a strategy for reducing PAHs in food as some applications instead of reducing PAHs has shown opposite effect. Also cumulative and interactive effect of two contaminants (one being PAHs and other present in any ingredient) needs to be studied. Recommendations drafted by scientific community must also consider the limitations of the studies stating PAHs' reduction and removal strategies.
6) It has been observed that most of the studies focusing on PAHs' reduction/removal as well as machine designing have been performed for meat products. A huge gap lies to identify these strategies for other processed products.

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Declaration of interests

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

Data availability

Data will be made available on request.

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