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REVIEW

Formation and mitigation of PAHs in barbecued meat – a review

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ABSTRACT

Polycyclic Aromatic Hydrocarbons (PAHs) are chemicals, which can occur in barbecued or grilled foods, and particularly in meats. They originate from incomplete combustion of the heat source, pyrolysis of organic compounds, or fat-induced flame formation. This review therefore summarizes relevant parameters for mitigation of especially carcinogenic PAHs in barbecued meat. Consumption of PAHs increases the risk of cancer, and thus the relevance for the mitigation of PAHs formation is very high for barbecued meat products. Parameters such as heat source, barbecue geometry, and meat type as well as marinating, adding spices, and other antioxidants reduce the final benzo[*a*]pyrene and PAHs concentrations and minimize the exposure. Overall, mitigation of carcinogenic PAHs from barbecuing includes removal of visual charring, reducing fat pyrolysis by minimizing dripping from the meat onto the heat source, the use of acidic marinades or choosing leaner cuts of meat. Estimation of human exposure to barbecued meat, includes several challenges such as substantial differences in barbecuing frequencies and practices, heat sources and meat types used for grilling.

KEYWORDS
Grill; benzo[*a*]pyrene; process parameters; doneness; PAH4; polycyclic aromatic hydrocarbons

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of compounds composed of two aromatic rings and upwards. PAHs are ubiquitous in the environment and have been reported in a wide range of food matrices (e.g., Adeyeye 2020a; Guillén, Sopelana, and Partearroyo 1997; Kazerouni et al. 2001; Plaza-Bolaños, Frenich, and Vidal 2010; Singh, Varshney, and Agarwal 2016; Zelinkova and Wenzl 2015a). PAHs have mainly been found in smoked meat and fish products (Adeyeye 2020a; Duedahl-Olesen et al. 2010; Simko 2002; Stołyhwo and Sikorski 2005; Zelinkova and Wenzl 2015a), vegetable oils (Moret and Conte 2000; Zelinkova and Wenzl 2015a) and also in products like dried plant and herb material (Martena et al. 2011; Zelinkova and Wenzl 2015b) including tea and coffee, leaves and infusions (Duedahl-Olesen et al. 2015b; Orecchio, Ciotti, and Culotta 2009; Ziegenhals, Jira, and Speer 2008), cocoa- based products (Ziegenhals, Speer, and Jira 2009) and lately also dairy products (Amirdivani et al. 2019). As indicated by these food products, PAHs are often found in foods processed by improper drying processes. These includes direct drying by smoldering and roasting, similar to the ones reported for mate tea (Ziegenhals, Jira, and Speer 2008), direct drying in the sun on asphalt bitumen as reported for cocoa beans (Ziegenhals, Speer, and Jira 2009), or direct contact with combustion gases as reported for grapeseed oil (Moret, Dudine, and Conte 2000) or meat (Adeyeye 2020a).

A variety of combustion and pyrolysis processes can as mentioned result in the formation of PAHs (Singh, Varshney, and Agarwal 2016; Cheng et al. 2019). High concentrations of PAHs are therefore often found in smoked and barbecued food products (e.g., Adeyeye 2020a; Zelinkova and Wenzl 2015a) due to the combustion or pyrolysis processes, which put them in contact with smoke, meant to preserve the products and contribute to the aroma. For barbecuing or grilling of meat the smokey aroma and therefore often a direct contact between meat and smoke or heat source is an important part of the consumer sensory experience.

Unfortunately, PAHs have been found to cause various types of cancers in animal model studies (Boström et al. 2002, Phillips 1999; Schneider et al. 2002). Some of the PAHs determined to be carcinogenic are active through genotoxicity mechanisms, whereas others merely promote and progress the appearances of cancer (Baird, Hooven, and Mahadevan 2005; Bansal and Kim 2015; John et al. 2011). In 2008, the European Food Safety Authority (EFSA) published a scientific opinion, based on oral carcinogenicity, compiling information from the International Programme on Chemical Safety (IPCS), the Scientific Committee on Food (SCF) and the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (EFSA 2008). There was enough evidence for 16 PAHs of their mutagenicity and genotoxicity or carcinogenicity (EU 15 + 1 in Table 1), and in 2012 benzo[*a*]pyrene (BaP) was classified as carcinogenic to humans (group 1 compound) by the International Agency for Research on Cancer (IARC 2012). EFSA found that eight of these compounds (PAH8 in Table 1) were adequate indicators for PAH contamination in food, with only 1% information lacking by application of the sum of four PAHs (PAH4

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in Table 1). Given the severe health effects of these compounds and their ubiquity in foods in combination with the fact that the highest concentrations have been reported for the food group of meat and meat products (Zelinkova and Wenzl 2015a), we focus our study on finding means of reducing PAHs in barbecued or grilled meat to reduce the total exposure.

The final PAHs concentration of barbecued food depends on several processing parameters such as the construction of the barbecue, the fuel used, the temperature, the meat type, and the pretreatment of the meat. In the past, only specific scenarios have included such conditions. To this day, there is no source compiling all of this information into one comprehensive and cohesive publication. This review will thus include literature on different parameters affecting the formation of PAH during barbecuing and mechanisms for minimizing the concentration (mitigation). Since barbecuing and grilling contribute to the overall dietary exposure to PAH, data evaluation for scientific-based advice to the consumer with best practices for the preparation of a barbecued meal is critical. Throughout the review, the terms barbecue and grill have been used interchangeably.

Nomenclature and compounds

Several terms, including sums of a different number of PAHs compounds, are used throughout the review. These can be seen in Table 1 and covers terms such as the sum of PAH4, PAH8 presented by the EFSA (2008) and EU 15 + 1 introduced by the European Commission and finally the EPA 16 PAHs identified by the U.S. Environmental Protection Agency in 1976 (Keith 2015).

Table 1. PAH nomenclature and terms referring to sums of different PAHs compounds as described in Keith 2015, and EFSA 2008.

Compound	Abbreviation	EPA 16	EU 15 + 1	PAH8	PAH4
Naphthalene	Nap	×			
Acenaphthylene	Acy	×			
Acenaphthene	Acn	×			
Fluorene	Flu	×			
Anthracene	Ant	×			
Phenanthrene	Phe	×			
Fluoranthene	Fla	×			
Pyrene	Pyr	×			
Benz[a]anthracene	BaA	×	×	×	×
Chrysene	Chr	×	×	×	×
Cyclopenta[cd]pyrene	CcdP		×		
5-Methyl chrysene	5MChr		×		
Benzo[b]fluoranthene	BbF	×	×	×	×
Benzo[j]fluoranthene	BjF		×		
Benzo[k]fluoranthene	BkF	×	×	×	
Benzo[c]fluorene	BcF		×		
Benzo[a]pyrene	BaP	×	×	×	×
Benzo[ghi]perylene	BghiP	×	×	×	
Indeno[1,2,3-cd]pyrene	Ind	×	×	×	
Dibenz[a,h]anthracene	DBahA	×	×	×	
Dibenzo[a,e]pyrene	DBaeP		×		
Dibenzo[a,i]pyrene	DBaiP		×		
Dibenzo[a,h]pyrene	DBahP		×		
Dibenzo[a,l]pyrene	DBalP		×		

EU legislation

Some PAHs have been determined to have genotoxic and carcinogenic effects (SCF 2002; Schneider et al. 2002) and are, as previously stated, unwanted in foods. The European Scientific Committee on Food (SCF) evaluating 33 PAHs concluded in 2002 that benzo[*a*]pyrene (BaP) could be used as a marker for the total concentration of PAHs in food until data profiles of 15 other PAHs had been studied (SCF 2002). Maximum limits for BaP in certain food items such as oils and fats, infant formulae, smoked fish, and meat were then set by the European Commission in 2006 (EC Regulation No. 1881/2006). During the last 15 years, new food commodities such as cocoa fibers, banana chips, food supplements, and dried plant material have been included in the regulation, whereas raw fish and meat were removed from the list (EU Regulation No. 1881/2006 with amendments). Concomitantly, the sum PAH4 was included in the regulation in 2011 (EC regulation No. 835/2011 amending EC Regulation No.1881/2006), since BaP was found as not detectable in more than thirty percent of all studied food samples containing other PAHs (EFSA 2008). Barbecued meat was included in EU regulation section 6.1.6 in 2011 (EC Regulation No. 835/2011 amending EC Regulation No. 1881/2006) as heat-treated meat, with maximum limits of 5 µg/kg for BaP and 30 µg/kg for PAH4 in meat products sold to the consumers. These maximum limits were again confirmed in EC Regulation No. 1125/2015 amending EC Regulation 1881/2006 to the now consolidated version. A significant gap in this regulation's ability to protect consumers from incurring toxic effects due to PAH intake is that it only regulates foods produced and sold in the Food industry. Whereas for barbecuing in private homes, it is a lot harder to ensure that consumers' intake of PAHs stays within limits considered safe or as low as reasonable achievable, although guidelines for consumers have been issued by the authorities (Danish Veterinary and Food Administration 2020).

History

One of the first reported studies on the formation of PAHs during barbecuing included a preliminary study on charcoal broiling of 15 beef steaks followed by the analysis of 15 PAHs in the outer 1 cm of the steak (Lijinsky and Shubik 1964). Results were compared to broiled meat, and concentrations of BaP up to 8 µg/kg were reported for charcoal grilling. Their study presented the first indications of melted fat dripping onto the heat surfaces and resulting in pyrolysis with formation of PAHs at high enough temperatures, which unfortunately would be transferred by small smoke particles back to the barbecuing meat product surface. In a follow-on study in 1967, they confirmed the relation to fat dripping, fat concentration, and pyrolysis (Lijinsky and Ross 1967). They compared five different cooking methods (electric and gas broiling, charcoal, charcoal with no drip and charcoal with a limited amount of charcoal), three meat types (beef, pork, and chicken) as well as the distance to the heat source (7, 12, 25 cm). The conclusion was that parameters affecting the BaP concentration included flame contact, time, and temperature combination (indicated by heat source distance) and meat fat concentration (Lijinsky and

Ross 1967). The analysis of barbecued meat today includes the whole meat and not only the outer surface, as Lijinsky and coworkers did. Consumers typically consume the barbecued meat in its entirety, rather than just the outer surface. However, the removal of the outer surface could be included as a mitigation proposition, especially if the meat is heavily charred.

Existing studies have investigated different barbecuing techniques focusing on specific parameters characteristic for their products and the formation of PAHs, but few has included comparisons to other frying techniques, type of fuel and grill geometry used for barbecuing as well as the choice of meat type, heating time and pretreatments of the meat. A few mitigating procedures and challenges during exposure assessment of barbecued meat will be summing up the review.

Comparison to other cooking techniques

Studies of effects of various processing methods including steaming, roasting, smoking, charcoal grilling, and liquid smoke flavoring on the formation of EPA 16 PAHs in duck breast steak were reported by Chen and Lin (Chen and Lin 1997). They concluded that for duck meat, the BaP concentration and sum of the EPA 16 PAHs were highest for smoking (BaP/sum EPA PAHs = 14/154 µg/kg) followed by charcoal grilling (8.5/151 µg/kg), roasting (nd/88.5 µg/kg) and steaming (nd/4.4 µg/kg) (Chen and Lin 1997). Whereas Kazerouni and coworkers who compared oven broiled, pan-fried, roasted, stewed, baked, microwaved, and grilled meat preparations reported processing by barbecuing of well-done chicken with skin (5.9 µg/kg) to result in the highest BaP concentrations (Kazerouni et al. 2001). In general, electric heating, oven broiling or pan-frying resulted in BaP concentrations well below 0.5 µg/kg (El Badry 2010; Farhadian et al. 2010; Kazerouni et al. 2001; Larsson et al. 1983; Rose et al. 2015) indicating that these heating types are not relevant when concerns are related to the PAHs concentration. However, benzo[*a*]pyrene concentrations above 5 µg/kg was reported for marinated suya prepared by electric grilling and hot air oven heating while even higher BaP concentrations were detected for suya prepared in a traditional suya smoker (Adeyeye 2017, 2020b). The marinade might have had an undefined effect on the final product PAHs concentration.

Little or no increase in the concentration of 27 different PAHs during frying, toasting, and roasting of lamb, chicken, beef burger, beef, salmon, sausages and pork products notwithstanding cooking intensity was reported in extensive in-house cooking experiments with 256 samples (Rose et al. 2015). However, increased PAH concentrations were obtained when barbecuing by gas or charcoal with a variety of meat and heat source distances. Studies on raw meat in comparison to grilled meat revealed a significant increase in the total sum of EPA 16 PAHs for practically all grilled food products with a total risk of PAHs increasing by a factor of five (Cheng et al. 2019). The PAH profile also varies according to food type. For instance, grilled vegetables have a PAH profile dominated by lower molecular weight PAHs (two to three aromatic rings), whereas in grilled chicken wings and mutton meat, the higher molecular weight PAHs (four to six aromatic rings) are a lot more prevalent (Cheng et al. 2019; Perelló et al. 2009). Incidentally, it is these heavier PAHs that are also more likely to be associated with adverse health effects (SCF 2002).

All these results indicate that several factors influence the final PAHs formation during barbecuing and, therefore, the individual final risk evaluations. Concentrations of the most commonly studied PAH, namely benzo[*a*]pyrene (BaP), have been reported in several studies on meat barbecuing in which the meat type and heat source were the main parameters (gas or charcoal) studied. In Table 2, BaP concentrations ranging from not detected (ND) for several conditions to 63 µg/kg for barbecued pork are reported for studies including gas or charcoal barbecuing for chicken, beef, lamb, salmon, pork and sausages.

Barbecuing parameters

Burning of coal, wood, manure, and diesel oil was reported to produce much higher levels of BaP than propane, due to the complete burning of the gas with low production of smoke (Lijinsky and Shubik 1964). This thick smoke production due to the use of suboptimal fuel sources leads to the concomitant adsorption of PAHs to the surface of barbecued meat, which in turn results in elevated PAH concentrations. An excellent example of this is *rougan*, a traditional Chinese pork product, which when barbecued over an open charcoal flame (Wu et al. 1997) was found to contain levels of PAH4 up to 500 times higher as compared to other studies describing levels of PAH in barbecued pork (Aaslyng et al. 2013; Abramsson-Zetterberg, Darnerud, and Wretling 2014; Duedahl-Olesen et al. 2015a; Mottier, Parisod, and Turesky 2000; Olatunji et al. 2013; Rose et al. 2015).

Reports illustrating PAH profile differences between gas and charcoal barbecued meat has also indicated that the PAH profile depends on the heat source (Gorji et al. 2016). The type of heat source used for barbecuing has a strong influence on PAH concentrations and the final PAH profile in the barbecued meat products.

Heat source

Multiple studies have concluded that PAH concentrations in charcoal-grilled meat are higher than those found in gas-grilled meat (Alomirah et al. 2011; Anjum et al. 2019; Dost and Ideli 2012; Duedahl-Olesen et al. 2015a; Farhadian et al. 2010; Gorji et al. 2016; Kazerouni et al. 2001; Rose et al. 2015; Viegas et al. 2012; Wu et al. 1997) (Table 2). A single study on barbecued lamb has identified higher BaP concentrations for products barbecued by gas in comparison to direct charcoal-grilled products, however with much lower BaP concentrations for indirect charcoal-grilled lamb (Table 2) (Alomirah et al. 2011). By comparison of direct charcoal-grilled chicken, beef, and pork to indirect charcoal grilling, the BaP concentrations and other PAH were found to increase by a factor of five or more when direct charcoal

Table 2. Reported BaP concentrations in μg/kg for different barbecued meat products using either gas or charcoal as a heating source with references.

Meat product \ Heat source	Gas (μg/kg)	Charcoal (direct heat) (μg/kg)	Charcoal (indirect heat) (μg/kg)	Charcoal (not specified) (μg/kg)	References
Beef	–	–	–	0–24	Aaslyng et al. 2013
Beef	–	6.57–7.05	–	–	Adeyeye 2017
Beef	–	ND–0.92	–	–	Chung et al. 2011
Beef	–	–	–	ND–17.5	Duedahl-Olesen et al. 2015a
Beef	ND–0.84	–	–	7.34–12.5	Farhadian et al. 2010
Beef	–	ND–3.16	–	–	Farhadian et al. 2011
Beef	–	–	–	0.11–4.78	Farhadian et al. 2012
Beef	–	0.17–0.39	–	–	García-Lomillo et al. 2017
Beef	0.35–4.02	0.36–5.81	–	–	Gorji et al. 2016
Beef	–	0.27–0.29			Haiba et al. 2019
Beef	–	–	–	0.09–4.86	Kazerouni et al. 2001
Beef	–	2.20–5.07	0.78–1.13	–	Lee et al. 2016
Beef	–	ND–42	–	–	Maga 1986
Beef	–	0.90–1.81	–	–	Mehr, Hosseini, and Ardebili 2019
Beef	–	–	–	ND–2.67	Moazzen et al. 2013
Beef	0.01–0.02	–	–	–	Mohammadi and Valizadeh-Kakhki 2018
Beef	–	–	–	ND–0.16	Olatunji et al. 2013
Beef	–	–	–	ND–0.29	Oz and Yuzer 2016
Beef	0.07–16.6	0.23–29.1	–	–	Rose et al. 2015
Beef	–	0.41–0.5	–	–	Viegas et al. 2012
Chicken (no skin)	–	–	–	0–1.2	Aaslyng et al. 2013
Chicken (no skin)	–	–	–	ND–0.6	Duedahl-Olesen et al. 2015a
Chicken (no skin)	ND–2.83	–	–	1.61–6.46	Farhadian et al. 2010
Chicken (no skin)	0.28–5.02	0.28–5.23	–	–	Gorji et al. 2016
Chicken (no skin)	–	0.97–1.34	–	–	Haiba et al. 2019
Chicken (no skin)	0.01–0.07	–	–	–	Mohammadi and Valizadeh-Kakhki 2018
Chicken (no skin)	–	–	–	ND–0.04	Olatunji et al. 2013
Chicken (with skin)	0.96–5.30 (direct) ND–1.01 (indirect)				El Badry 2010
Chicken (with skin)		–	–	ND–3.96	El Husseini et al. 2018
Chicken (with skin)	–	ND–2.44	–	–	Farhadian et al. 2011
Chicken (with skin)		3.14–8.73	–	–	Viegas et al. 2012
Chicken (with and without skin)	0–0.93	ND–4.63	0.12–0.64	–	Alomirah et al. 2011
Chicken (with and without skin)	–	ND–4	–	–	Kao et al. 2012
Chicken (with and without skin)	–	–	–	0.39–4.57	Kazerouni et al. 2001
Chicken (not specified)	–	–	–	ND–0.25	Moazzen et al. 2013
Chicken (not specified)	–	–	–	ND–1.0	Reinik et al. 2007
Chicken (not specified)	ND–0.48	0.26–0.57	–	–	Rose et al. 2015
Chicken (not specified)	–	ND–3.68	–	–	Wang et al. 2018
Chicken (not specified)	–	0.44–1.51	–	–	Wang et al. 2019a
Chicken (not specified)	–	0.51–2.81	–	–	Wang et al. 2019b
Duck (with and without skin)	–	3.7–9.2	–	–	Chen and Lin 1997
Duck (not specified)	–	2.6–3.1	–	–	Kao et al. 2012
Goat	–	–	–	0.09–1.52	Ahmad et al. 2017
Lamb	ND–4.45	ND–4.28	ND–0.21	–	Alomirah et al. 2011
Lamb	–	–	–	ND–3.9	Duedahl-Olesen et al. 2015a
Lamb	–	ND–5.8	–	–	Kao et al. 2012
Lamb	–	–	–	ND–1.43	Moazzen et al. 2013
Lamb	–	0.32–2.81	–	–	Mottier, Parisod, and Turesky 2000
Lamb	0.18–0.59	0.54–2.39	–	–	Rose et al. 2015
Mutton	0.3–2.1 (horizontal) <0.1 (vertical)	–	–	–	Saint-Aubert et al. 1992
Salmon	–	–	–	ND–2.7	Duedahl-Olesen et al. 2015a
Salmon	–	–	–	–	Oz and Yuzer 2016
Salmon	0.22–1.94	0.38–1.01	–	–	Rose et al. 2015
Salmon	–	1.36–4.72	–	–	Viegas et al. 2012
Sardine	1–5.0 (horizontal)/ 0.15 (vertical)	–	–	–	Saint-Aubert et al. 1992
Fish (not specified)	–	–	–	ND–1.23	Farhadian et al. 2010
Pork	–	–	–	ND–4.5	Aaslyng et al. 2013
Pork	–	ND–8.49	–	–	Chung et al. 2011
Pork	–	–	–	ND–63	

(continued)

Table 2. Continued.

Meat product \ Heat source	Gas (µg/kg)	Charcoal (direct heat) (µg/kg)	Charcoal (indirect heat) (µg/kg)	Charcoal (not specified) (µg/kg)	References
					Duedahl-Olesen et al. 2015a
Pork	–	–	–	–	Kazerouni et al. 2001
Pork	–	0.68–5.99	0.56–0.66	–	Lee et al. 2016
Pork	–	–	–	0.13–0.19	Olatunji et al. 2013
Pork	–	5.51–8.04	1.28	–	Park et al. 2017
Pork	–	0.7–1.8	–	–	Reinik et al. 2007
Pork	ND–0.98	0.36–1.9	–	–	Rose et al. 2015
Pork	–	–	–	–	Tkacz, Wiek, and Kubiak 2012
Pork	–	–	–	1.07–2.71	Viegas et al. 2014
Pork	–	ND–30.2	–	–	Wongmaneepratip, Jom, and Vangnai 2019
Sausage, Pork	–	–	–	ND–1.9	Duedahl-Olesen et al. 2015a
Sausage, Pork	–	–	–	0.01–0.11	Kazerouni et al. 2001
Sausage, Pork	–	ND–1.0	–	–	Larsson et al. 1983
Sausage, Pork	–	ND–1.2	–	–	Reinik et al. 2007
Sausage, Pork	ND–1.88	0.27–3.55	–	–	Rose et al. 2015

grilling was applied (Alomirah et al. 2011; Anjum et al. 2019; Lee et al. 2016). These results illustrate similar trends as previously reported for direct and indirect smoking of fish as well (Duedahl-Olesen et al. 2010).

A thorough barbecue study with different meat types (chicken, beef, lamb, pork, salmon and sausage) including different heating techniques and fuels (gas, charcoal, charcoal with wood chips and briquettes) in the UK followed the same trends (Rose et al. 2015). For all barbecued meat products, the highest concentrations were associated with the use of charcoal with wood chips (Table 3). The use of briquettes resulted in similar or lower BaP, PAH4 and sum PAH_{27} concentrations for meats except beef burgers, when compared to gas grilling of the same products (Rose et al. 2015). For barbecuing with charcoal and flame-gas, BaP concentrations ranging from 0.76 to 7.4 µg/kg and 0.37 to 1.5 µg/kg, respectively (Table 2) were obtained with the use of a vertical flame-gas source resulting in the lowest PAH concentrations (sum of 3 PAHs) (Farhadian et al. 2010). The different PAHs concentrations for gas and charcoal is most likely due to the incomplete burning of charcoal compared to gas, and the resulting soot formation with increased PAH levels.

The repeated use of a gas barbecue for mutton chop barbecuing, resulted in similar PAH concentrations (sum of six) for the first and 20^{th} barbecuing without cleaning, whereas the 5^{th} barbecuing had PAH concentrations approximately 5 times lower (Saint-Aubert et al. 1992). In contrast, barbecuing of sardines with the same condition as described above resulted in the highest PAH concentrations for the 5^{th} barbecuing (Saint-Aubert et al. 1992). Continuous barbecuing with same charcoal resulted in an at least double concentration of BaP (from 3.1 µg/kg to 8.7 µg/kg) and the same for the sum of 8 PAH (from 25 to 61 µg/kg) due to the accumulated high molecular weight PAHs being released by reheating (Viegas et al. 2012).

Formation of PAH4 in meat barbecued during 4 consecutive heating periods of 12 minutes with considerably higher concentrations in beef ribs and pork meat from the first period indicated incomplete combustion of the charcoal with visible flames, especially in the first 12 minutes of heating (Lee et al. 2016). During the other periods, no significant differences in concentrations of PAHs were detected, indicating the importance of pre-heating charcoals before starting to barbecue. Use of three charcoal types on the formation of 16 EPA PAHs during barbecuing of beef, pork and chicken reported highest concentrations for extruded charcoal from coconut shell followed by black charcoal from oak with lowest PAH concentrations obtained when using white charcoal from broadleaf trees, no matter the meat type (Kim, Cho, and Jang 2020).

The use of wood as heat sources and their influence on the PAHs concentration was demonstrated by Larsson and coworkers, who obtained average BaP concentrations of 54.2 µg/kg in log fire flame grilled frankfurters whereas in charcoal-grilled frankfurters BaP concentrations did not exceed 1 µg/kg (Larsson et al. 1983) (Table 3). BaP concentrations between these extreme values were reported for barbecuing over smoldering spruce or pine cones and log fire embers (Table 3) also with a decreasing sum of 22 PAHs in the following order: *log fire > pine cones > log fire embers > charcoal fire > electric oven (24 µg/kg) > frying pan (12 µg/kg)* (Larsson et al. 1983). Pan-frying (n = 5) or electric broiling (n = 2) of frankfurters did similar to results reported in the previous section and did not lead to any increase in BaP levels in comparison to the raw material. Grilling in a propane flame at temperatures of 600 °C with an open and closed burner resulted in BaP levels below 1 µg/kg and 15 µg/kg, respectively (Larsson et al. 1983). So the PAH levels in barbecued foods are strongly dependent on the type of heat source used and, the extent of ventilation, rather than the temperature.

PAH concentrations in smoked meat have also previously been reported to strongly depend on the wood source with BaP concentrations ranging from 6.0 µg/kg for apple-tree, and alder smoked meat to 35 µg/kg for spruce smoked meat

Table 3. BaP, sum PAH4 and sum PAH (number of PAH included in subscript) concentrations in µg/kg of variable barbecued meat types with indications of the use of heat source.

Type of meat	Heat source	N	BaP in µg/kg	Sum PAH4 in µg/kg	Sum PAH_{total} in µg/kg	References
Sausages (frankfurters)	Log fire	17	54.2	173	$PAH_{22} = 905$	Larsson et al. 1983
	Pine cones, smoldering spruce	7	17.6	70.0	$PAH_{22} = 377$	
	Log fire embers	9	7.70	39.0	$PAH_{22} = 269$	
	Charcoal fire	13	<1.0	3.60	$PAH_{22} = 51.0$	
Sausages	Charcoal	3	3.29	15.7	$PAH_{27} = 117$	Rose et al. 2015
	Charcoal + wood chips	3	22.4	22.4	$PAH_{27} = 410$	
	Briquettes	3	0.31	3.65	$PAH_{27} = 62.3$	
	gas	3	1.50	4.48	$PAH_{27} = 43.7$	
Beef Patties (30% fat)	Mesquite wood	2	42.0	144	$PAH_{31} = 549$	Maga, 1986
	Hickory sawdust	2	1.0	6.0	$PAH_{31} = 68.0$	
Beef burger	Charcoal	3	17.3	66.6	$PAH_{27} = 563$	Rose et al. 2015
	Charcoal + wood chips	3	20.0	70.6	$PAH_{27} = 790$	
	Briquettes	3	10.4	38.7	$PAH_{27} = 449$	
	gas	3	14.8	35.2	$PAH_{27} = 547$	
Beef	Coconut shells	3	0.50	3.49	$PAH_{15} = 43.8$	Viegas et al. 2012
	Wood charcoal	3	0.41	2.33	$PAH_{15} = 48.1$	
Beef	Charcoal	3	0.27	3.86	$PAH_{27} = 37.0$	Rose et al. 2015
	Charcoal + wood chips	3	1.20	7.77	$PAH_{27} = 82.8$	
	Briquettes	3	0.18	1.74	$PAH_{27} = 30.3$	
	gas	3	0.25	0.86	$PAH_{27} = 21.1$	
Pork chop	Charcoal	1	1.90	8.07	$PAH_{27} = 84.4$	Rose et al. 2015
	Charcoal + wood chips	1	3.38	13.5	$PAH_{27} = 148$	
	Briquettes	1	0.23	1.84	$PAH_{27} = 43.0$	
	gas	1	0.98	3.03	$PAH_{27} = 39.3$	
Chicken	Charcoal	3	0.41	7.64	$PAH_{27} = 73.6$	Rose et al. 2015
	Charcoal + wood chips	3	0.73	7.07	$PAH_{27} = 102$	
	Briquettes	3	0.15	1.66	$PAH_{27} = 23.0$	
	gas	3	0.18	0.59	$PAH_{27} = 4.76$	
Salmon	Coconut shells	3	1.36	8.26	$PAH_{15} = 73.2$	Viegas et al. 2012
	Wood charcoal	3	4.72	38.0	$PAH_{15} = 213$	
Salmon	Charcoal	3	0.79	9.25	$PAH_{27} = 101$	Rose et al. 2015
	Charcoal + wood chips	3	3.24	18.2	$PAH_{27} = 171$	
	Briquettes	3	0.29	2.73	$PAH_{27} = 36.4$	
	gas	3	1.12	3.49	$PAH_{27} = 41.6$	
Lamb	Charcoal	1	2.39	11.0	$PAH_{27} = 150$	Rose et al. 2015
	Charcoal + wood chips	1	3.03	11.4	$PAH_{27} = 130$	
	Briquettes	1	0.71	3.79	$PAH_{27} = 71.3$	
	gas	1	0.59	1.88	$PAH_{27} = 22.8$	

Data from Rose et al. 2015 includes results from the shortest distance to heat source.

(Stumpe-Vīksna et al. 2008). However, studies on PAH profiles in the smoke composition of softwood (pine) and hardwood (beech) have not reported variations (Toth and Pothast 1984), whereas others have reported slight differences in the concentrations of high molecular PAHs in the final barbecued products (Larsson et al. 1983). Comparison of grilling beef patties (30% fat) with mesquite wood (64% lignin) and the hardwood hickory sawdust (54% cellulose) however, resulted in considerable different BaP, PAH4 and sum PAH_{31} concentrations (Table 3) as well as different PAH profiles with 24 PAHs present in mesquite wood grilled beef and only 16 PAHs in hickory grilled beef (Maga 1986).

A comparison of grilling with coconut shells labeled as "flameless and smokeless charcoal," to wood charcoal resulted in lower PAH concentrations with coconut shells for barbecuing of salmon compared to wood charcoal, whereas the PAH concentrations in beef were similar (Viegas et al. 2012) (Table 3). The scientists noticed that flames formed when barbecuing salmon, mainly due to fat dripping onto the heat source, which probably resulted in PAH and particle formation with higher PAH concentrations in the final wood charcoal barbecued product.

All these results indicate that conditions for smoke generation have a critical influence on the final PAH level, also indicated by the effect of the presence of oxygen for gas combustion.

Barbecue geometry

Cooking on two geometrically different butane gas barbecues resulted in higher PAH concentrations (fluoranthene and 5 PAH classified as carcinogenic) in samples barbecued at a horizontal lava-rocks barbecue, compared to a vertical barbecue, where fat dripping pyrolysis was prevented (Saint-Aubert et al. 1992). Significant differences ($p < 0.001$) in BaP concentrations were also obtained for barbecuing of pork belly using a modified charcoal grill (1.3 µg/kg), where fat dripping contact to the heat source was prevented, compared to a regular charcoal grill (8.0 µg/kg) (Park et al. 2017). Also, Lee et al. 2016 concluded that a significant reduction in the sum of PAH4 in barbecued pork and beef meats were obtained through barbecuing on devices which removed meat drippings (48–89% reduction). The total sum of 8 PAHs in meat using a stone barbecue was surprisingly similar to or even higher than sums for meat barbecued on a general wire barbecue, except for very well-done samples (Oz and Yuzer 2016). The researchers were speculating, with reason, that the use of tail fat for cleaning the stone barbecue before cooking the samples could have influenced the

Table 4. BaP concentrations in µg/kg for barbecuing of chicken, pork, and cow meats on a rack positioned 40 cm above the heat source and a disposable barbecue, 15 cm above the heat source (Data translated from Noll and de Figueiredo 1997).

Type of meat	N	BaP in µg/kg (40 cm)	BaP in µg/kg (15 cm)
Chicken leg	3	Nd	1.4
Chicken wing	3	0.97	4.3
Neck of pork	3	Nd	7.9
Chorizo, pork	3	Nd	0.4
Cow shank	6	0.56	7.1

Nd = not detected. Numbers reduced to significant digits.

surprising result (Oz and Yuzer 2016). Studies including a reduction of smoke using a ventilated barbecue illustrated a considerable decrease in PAH4 (41–74% reduction) concentrations of grilled food, due to the decrease in contact between the meat and the smoke particles (Lee et al. 2016). All the above results confirm the first findings from Lijinsky and Shubik (1964), stating that one of the most critical factors contributing to the production of PAHs in barbecuing is smoke resulting from incomplete combustion of fat dripped onto the heat source.

The distance between the barbecued meat and the heat source is also a factor in the final concentration of PAHs in the meat product. A study on BaP in charcoal-broiled beef, pork and chicken meats barbecued over a rack positioned 40 cm above the heat source, and a portable barbecue positioned 15 cm above the charcoal, reported higher concentrations no matter the meat type for the portable grill (Table 4) (Noll and de Figueiredo 1997).

Reinik et al. (2007) also reported the presence of both higher BaP and PAH concentrations (12 PAHs) in home-grilled pork and chicken products, with up to a 1.6-fold higher PAH concentration with disposable (portable) barbecuing units than by traditional wood-burning barbecues (Reinik et al. 2007). However, barbecuing of sausages did not show PAH concentrations dependent on the choice of barbecue geometry and, therefore, the distance to the heat. In contrast to these results, barbecuing frankfurters to a well-done state with varying distance (1, 6, 16, and 26 cm) to the log fire heat source including 20 cm flames, showed that the PAH levels (sum of 22 compounds) in flame-grilled frankfurters were strongly dependent on the position of the samples in the flame during the barbecuing (Larsson et al. 1983). Moderate levels were found in the samples barbecued 1 cm above the fuel (BaP approximately 5 µg/kg), whereas samples in a 6–7 cm zone showed dramatically increased PAH levels (BaP ∼ 80 µg/kg). For longer distances, the levels declined with increasing distance from the fuel with BaP concentrations of approximately 25 (16 cm) and 15 (26 cm) µg/kg, respectively. A comprehensive UK heating study found that PAH levels increased when the food was barbequed closer to the heat source (4 and 7 cm compared to 9 and 11.5 cm) no matter the heat source (Rose et al. 2015). For sausage cooked over briquettes and for beef burgers, beef and salmon cooked over charcoal, the concentration of PAHs was lower when the food was closer to the heat source (Rose et al. 2015), similar to the findings of Larsson et al. 1983. The presence of PAH in the final products is probably due to the uptake of PAHs from the combustion fumes or dripping fat at different distances reflecting the PAH distribution in the smoke.

Meat type

Several barbecue studies have concluded that the final PAH concentrations depend on the meat type (Dost and Ideli 2012; Perelló et al. 2009; Saint-Aubert et al. 1992). However, no clear trends for specific meat types and PAH concentrations have been identified, mainly due to a large number of variable parameters for each of the studies reported. The final PAH concentrations do however depend on the fat content of the barbecued meat (Babaoglu, Karakaya, and Öz 2017; El Husseini et al. 2018; Kao et al. 2012; Kazerouni et al. 2001; Kafouris et al. 2020; Kim, Cho, and Jang 2020; Lee et al. 2016; Mottier, Parisod, and Turesky 2000; Oz and Yuzer 2016; Viegas et al. 2012). Furthermore, the higher the content of unsaturated fat, the more likely it is for benzene-like compounds and derivatives of PAHs to be formed (Chen and Chen 2001).

Studies comparing BaP concentrations of grilled meat have demonstrated highest BaP concentrations in barbecued pork (Chung et al. 2011; Kim, Cho, and Jang 2020) or salmon (Viegas et al. 2012) whereas other studies have reported highest concentrations in barbecued beef samples in comparison to pork, chicken or lamb (Table 2) (Aaslyng et al. 2013; Larsson et al. 1983; Moazzen et al. 2013; Mohammadi and Valizadeh-Kakhki 2018; Rose et al. 2015). Even though the fat content of barbecued meat is relevant for fat pyrolysis and PAH formation, none of the studies reported the fat concentration with BaP concentrations in beef ranging from 0.15 µg/kg to 29 µg/kg (Table 2). Four commercial Nigerian char-broiled beef suya had similar high Bap concentrations with reported values from 6.5 to 21 µg/kg (Duke and Albert 2007). West African grilled suya from cow, goat, sheep and chicken had BaP concentrations still above the maximum limit of 5 µg/kg ranging from 5.7 µg/kg for cow to 5.1 µg/kg for chicken, with PAH4 concentrations below 30 µg/kg ranging from 22.7 µg/kg for cow to 20.0 for chicken suya (Adeyeye et al. 2020a). All products had a fat content between 20.2% for cow and 18.9% for chicken.

Also, barbecuing of pork, chicken and cow showed variable meat types having the highest BaP concentration depending on the distance between the meat and the heat source (Table 4), indicating no clear meat type trends. A comparison of barbecued mutton versus sardines revealed that sardines (5.0 µg/kg) contained twice the BaP concentrations than mutton (2.1 µg/kg) (Saint-Aubert et al. 1992). In contrast, lamb steak compared to different chicken products demonstrated highest BaP concentrations in lamb (5.8 µg/kg) followed by chicken drumstick (4.0 µg/kg) (Kao et al. 2012). For both Iranian popular grilled kebab and Malaysian charcoal-grilled satay from beef and chicken, significant differences were obtained for BaP concentrations of beef (22 µg/kg and 7.4 µg/kg) and chicken (0.29 µg/kg and 2.0 µg/kg) (Farhadian et al. 2010; Mohammadi and Valizadeh-Kakhki 2018). In contrast, both Egyptian charcoal barbecued chicken and Iranian charcoal barbecued kebab had higher BaP and PAH4 concentrations than similar barbecued meat varieties (Haiba et al. 2019; Gorji et al. 2016). In both cases, the higher concentrations were expected to be due to higher fat content of the barbecued chicken products. Comparing BaP, PAH4 and the sum of 27 PAH concentrations for beef, beef burger, sausage, pork chops, salmon, lamb and chicken generally resulted in highest concentrations for beef burgers (up to twenty times) than for the lowest concentrations of chicken, no matter the heat source (Table 3) (Rose et al. 2015). Multiple studies report the lowest BaP concentrations to be found in barbecued chicken fillets, indicating that chicken tends to be associated with lower concentrations compared to other meat types (Duedahl-Olesen et al. 2015a; Larsson et al. 1983; Moazzen et al. 2013; Rose et al. 2015), possibly due to the lower fat content. However, BaP concentrations for chicken in the range of 0.28 to 5.8 µg/kg has shown highest concentrations in charcoal-grilled chicken wings with skin and lowest for gas grilled chicken meat (Gorji et al. 2016) and therefore confirmed findings of higher PAHs concentrations in chicken with skin compared to without (Chen and Lin 1997; Duedahl-Olesen et al. 2015a; El Husseini et al. 2019; Kafouris et al. 2020; Noll and de Figueiredo 1997). The skin was expected to increase the fat content of the chicken and therefore increase the possibility of fat dripping onto the heat source and forming fumes. In contradiction to these results, a study on charcoal-grilled marinated beef and skinless chicken resulted in highest EPA 16 PAH and BaP concentrations for chicken (Haiba et al. 2019). The importance of fat was illustrated with the obtained PAH4 concentrations in Turkish lamb and beef kokorec using the respective intestine in combination with lamb tallow, lamb subcutaneous or lamb tail fat. Highest values was obtained for beef combined with tail fat (23.6 µg/kg), whereas lamb products obtained highest concentrations in combination with tallow fat (8.1 µg/kg) (Babaoglu, Karakaya, and Öz 2017). Studies of the different goat organs and the formation of PAHs during barbecuing also illustrated different PAH levels, however, without the possibility to correlate data with the fat content (Ahmad et al. 2017).

A study based on traditional Lebanese barbecued products illustrated the importance of a low surface to mass ratio when barbecuing beef and chicken products. The PAH4 concentrations were more than doubled with increased surface areas with PAH4 concentrations of 2 µg/kg for steak/breast to 7.5 µg/kg for small beef pieces and 6.6 µg/kg for chicken pieces (El Husseini et al. 2019).

For barbecued frankfurters and sausages, the PAH concentrations have been reported to be lower or even much lower than other whole meat products (Abramsson-Zetterberg, Darnerud, and Wretling 2014; Larsson et al. 1983; Rose et al. 2015). While barbecuing frankfurters or sausages, only small amounts of fat will drip onto the heat source. The fat from sausages is typically bound in stable emulsions in the product. Additionally, when the sausage casing breaks during grilling, the spray from the sausages will go in multiple directions and therefore is less likely to reach the fuel source in its entirety, thus greatly diminishing or even preventing fat pyrolysis (Larsson et al. 1983; Rose et al. 2015). These speculations were confirmed by barbecuing frankfurters simultaneously with pork chops with a resulting BaP concentration rising from 0.3 µg/kg for barbecuing frankfurters alone to 2.0 µg/kg (Larsson et al. 1983).

The increased fat content of patties of 90, 80 and 70% lean beef meat confirmed the correlation to fat content and resulted in increased PAH concentrations (Fretheim 1983; Lijinsky and Shubik 1964; Maga 1986; Olatunji et al. 2013). Overall, when using mesquite wood for barbecuing patties of 90, 80, and 70% lean beef, a lower amount of PAHs were found in 90% lean patties with approximately half the total PAH concentration compared to the 70% lean products (Table 4). The 80% lean patties had levels in the middle of the two extremes, with sums of 32 PAHs of 314, 549, and 448 µg/kg (Maga 1986). In addition, barbecued beef burgers prepared from minced beef meat with BaP concentrations up to 29 µg/kg (Rose et al. 2015) indicated that the fat content and possibly a large surface to volume ratio are essential factors for the formation of PAHs and exhibiting high BaP concentrations.

Time-temperature combination

Doneness refers to how thoroughly cooked a piece of meat is, typically ranging from rare (red center and soft) to well-done (gray-brown throughout and firm). As a rule of thumb, the longer and more thorough meat is grilled, the higher the levels of PAHs tend to be (Haiba et al. 2019; Kazerouni et al. 2001; Saint-Aubert et al. 1992).

Steaks, hamburgers and chicken with skin barbecued for more extended periods (well-done) tend to contain higher levels of BaP (4.9, 1.5, and 4.6 µg/kg respectively) than if the barbecuing time is shorter (medium-done steak: 4.2 µg/kg, medium done hamburger: 0.1 µg/kg) (Kazerouni et al. 2001). Barbecuing frankfurters to well-done state no matter the distance to the heat source (1, 6, 16, 26 cm) resulted in highest heating time for the longest distance (4.5 min) and shortest time at 6 cm (1.2 min) with simultaneously final highest BaP concentration (~ 83 µg/kg) (Larsson et al. 1983). Barbecuing frankfurters in 20 cm flames over a log fire for 3.5 min at various distances (1, 7, 13, 20 cm) from the flame resulted in a burnt product (1 cm distance; BaP around 12 µg/kg), well-done (7 cm distance; BaP ~ 27 µg/kg), medium (13 cm distance; BaP ~ 22 µg/kg) and light (BaP ~ 6 µg/kg) barbecued frankfurter (Larsson et al. 1983). This trend in BaP levels indicates a correlation between doneness and PAH concentration.

Formation of six PAHs including BaP in horizontal barbecued sardines fried 2 (rare), 4 (medium), and 8 (well done) minutes on each side increased for all treatments compared to raw material. After 4 minutes of barbecuing, the maximum levels of 30 µg/kg and 185 µg/kg for BaP and the sum of six PAHs were reached, with similar concentrations after 8 minutes barbecuing (Saint-Aubert et al. 1992). Total EPA 16 PAHs concentrations changed from 151 µg/kg to 200 µg/kg and finally 300 µg/kg for grilling duck breast without skin for 0.5, 1, and 1.5 h, respectively (Chen and

Lin 2001). The BaP concentration, however, reached a maximum of 5 μg/kg after 1 h. In addition, duck breast with skin reached a final BaP concentration though at a higher level of 8.4 μg/kg after barbecuing for 1 h and total EPA 16 PAH concentrations of 182; 276 and 319 μg/kg for 0.5; 1 and 1.5 h (Chen and Lin 2001).

In Rose et al. 2015, indications of an increase of PAH (27) concentrations with increased barbecuing times were illustrated (Rose et al. 2015). However, the study concluded that a combination of distance and cooking time may result in a moderate increase in PAHs in some foods (Rose et al. 2015). An interesting finding of this study was that doubling the cooking time for beef burgers, no matter the heat source, and for chicken using charcoal, surprisingly led to a decrease in the final PAH concentrations (Rose et al. 2015). It is well known that chicken meat needs less time than beef to be barbecued, and results on kebab indicated that total PAHs concentrations for chicken samples were considerably lower than beef samples (Gorji et al. 2016). A study including two barbecuing times (doubling the time) for each barbecued product (chicken, pork, seafood, and lamb) typically resulted in increased total EPA 16 PAH concentrations with increased heating time, except for squid and pork chop, which were unchanged (Kao et al. 2012).

Studies on 15 PAHs in marinated grilled beef satay at temperatures in increased steps of 50 degrees Celsius from 150 °C to 350 °C showed that the concentration of PAHs increased markedly from 300 °C to 350 °C. PAH4 concentrations at the two temperatures exceeded the maximum limit of 30 μg/kg with concentrations of 51.1 μg/kg and 67.7 μg/kg, respectively. Without marinade only suya grilled at 350 °C had PAH4 concentrations above the limit, namely 43.7 μg/kg (Kamal, Selamat, and Sanny 2018). The formation of PAHs being more affected by changes in temperature than time was confirmed in a meat model study on 8 PAHs at temperatures of 80 °C to 200 °C in steps of 40 °C and times from 15 to 30 minutes in 5 minutes intervals (Min, Patra, and Shin 2018).

In a study by Haiba et al. 2019, beef and chicken meat samples were grilled on a satay charcoal grill, at a distance of 3–5 cm, after being marinated in a sauce of lemon juice, chopped onion, garlic, pepper, and salt, at two different levels of doneness. The well-done samples had an average of 14% higher concentrations of PAHs than medium-done samples. The levels for the sum of 16 EPA PAHs were lower than other studies, ranging from 3.7 to 6.3 μg/kg (Haiba et al. 2019). The low distance, as indicated by Rose and coworkers and the use of marinade, were likely contributing factors in this. Overall, the level of doneness indeed indicated an increase in PAH concentrations in charcoal-grilled meat samples, which translates to increased health risk to consumers (Haiba et al. 2019).

When investigating barbecue type alongside doneness, Oz and Yuzer 2016 found none of the PAH 8's in beef samples barbecued to rare and medium on the wire barbecue, whereas when the cooking time was increased to produce well-done and very well-done cooked samples, the total sum of PAH8 increased with cooking time to 0.8 and 0.9 μg/kg, respectively. On the stone barbecue, PAH 8's were not detected in the rare cooked samples, the highest levels were detected in the medium-grilled samples (2.6 μg/kg), and increasing cooking time and doneness, similar to Larsson and coworkers, decreased the levels of the PAH8 in the samples to 0.9 and 0.8 μg/kg, respectively (Oz and Yuzer 2016). In comparison, also Adeyeye commented that especially well-done meat contained PAHs (Adeyeye 2020a). Epidemiologic studies recommending low consumption of carcinogens and especially avoidance of well and very well-done meat (Anderson et al. 2002) support these observations.

Authors recommend that the barbecuing temperature should be reduced with concomitant longer cooking time (Anjum et al. 2019; Oz and Yuzer 2016) or with less convincing documentation the barbecuing time could be reduced in general to avoid charring and overcooking (Farhadian et al. 2010; Kazerouni et al. 2001; Mottier, Parisod, and Turesky 2000; Perelló et al. 2009). Given the contradictory results in the literature and the variability in experimental parameters, it is challenging to draw overarching conclusions based on time and temperature alone.

Pretreatment of meat

Pretreatment procedures such as steaming, microwave preheating, and wrapping in aluminum foil or banana leaves before charcoal barbecuing have been found to reduce the BaP concentrations (Farhadian et al. 2011). In samples of beef (charcoal grilling, BaP = 3.2 μg/kg) and chicken (charcoal grilling, BaP = 2.4 μg/kg) reductions, up to 100% was achieved (BaP not detected), except for banana wrapping, which only reduced the BaP concentrations 81 and 65% for beef and chicken, respectively. However, Fla was detected in all samples, no matter treatment, with a reduction between 32 and 81%, both for banana wrapped beef and chicken, respectively (Farhadian et al. 2011). The use of aluminum trays significantly reduced (67 to 86%) the concentration of 7 carcinogenic PAH as well as Fla and Pyr in barbecued pork steak dependent on the pretreatment (Tkacz, Wiek, and Kubiak 2012). As expected, wrapped meat or meat in trays prevent fat from dripping onto the heat source and thereby reduce the possibilities of generating PAH.

Barbecuing chicken without pretreatment increased the concentrations of the EPA 16 PAHs in meat from 0.2 μg/kg in raw chicken to 44–72 μg/kg dependent on the grilling method, compared to 3.2–29 μg/kg with a pretreatment including spices or marinades (El Badry 2010). Simultaneously, the concentration of carcinogenic PAHs increased up to 200 times without pretreatment and 10 to 120 times with pretreatment.

Experiments on marinades have included several different approaches and have not shown an explicit effect on the sum of PAHs levels. Several types of marinades have been studied, and the ones containing higher amounts of antioxidants were associated with inhibition of PAH generation. The key antioxidant-rich ingredients were onions and garlic (Janoszka 2011), spices (Gong, Zhao, and Wu 2018; Rey-Salgueiro et al. 2009), beer (Viegas et al. 2014; Wang et al.

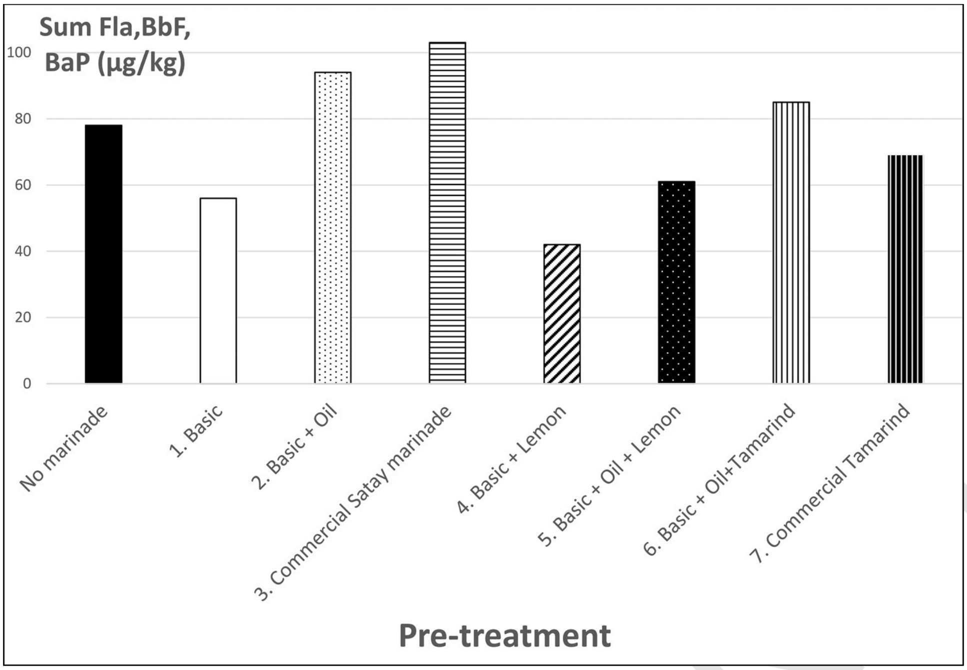

Figure 1. Sum of Fla, BbF, and BaP in µg/kg for pretreatment of skewed beef (t = 8 h) before charcoal barbecuing for 8 minutes on a satay-type grill, N = 2 (Data from Farhadian et al. 2012).

2019b), tea (Park et al. 2017; Wang et al. 2018), and lemon (Farhadian et al. 2012).

Seven marinade treatments were applied in four time intervals to the beef before charcoal barbecuing in Farhadian et al. 2012. The marinades contained the following ingredients: 1) "basic" marinade (sugar, water, onion, turmeric, lemongrass, salt, garlic, coriander, and cinnamon); 2) "basic" with oil; 3) commercial marinade; 4) "basic" with oil and lemon juice; 5) "basic" with lemon juice; 6) "basic" with oil and tamarind; 7) commercial tamarind, and the four time intervals were of 0, 4, 8, and 12 h. The duration of marinating did not show a significant factor in BaP or PAH reduction (Farhadian et al. 2012). However, a considerable reduction (70%) was shown for beef treated with acidic marinade (5) with lemon juice pH 5.2 compared to "basic" marinade (1) pH of 5.7, followed by the "basic" marinade (1) (Figure 1). The addition of oil to the marinade resulted in the lowest reduction in the BaP concentration with a concomitant increase in the total sum of three PAHs (Farhadian et al. 2012), indicating the formation of non-carcinogenic PAHs by oil addition. The authors suggested that the reduction in levels of PAHs generated could be attributed to the antioxidant activities of sulfhydryl compounds in garlic and onion. Such compounds are known to act as scavengers of free radicals, capturing electrophilic compounds and, through this mechanism, inhibit the formation of PAHs (Farhadian et al. 2012). Other authors also believed in the antioxidative effect, which was used to explain the reduction of PAH 4 in charcoal grilled pork sprayed with marinades based on vinegar (Cordeiro et al. 2020). The PAH4 concentration was reduced between 50 and 82%, with white wine and elderberry vinegar being most effective, whereas vinegar with raspberry juice was the least effective. No matter which vinegar was used all PAH4 and BaP concentrations were reduced to levels well below the maximum limits (Cordeiro et al. 2020). An even larger reduction of 11 PAHs was obtained for the application of spices to Egyptian charcoal grilled kebab and kofta (Eldaly et al. 2016).

Beer marinade also reduced PAH8 concentrations in grilled pork from 13 to 53% and showed a positive correlation with the simultaneously studied free radical scavenging ability. (Viegas et al. 2014). Others reported that beer marinades demonstrate different effects on the formation of PAH8 for charcoal barbecuing of chicken wings (Wang et al. 2019b). Heineken (4.3 µg/kg) and Tsing Tao (8.9 µg/kg) reduced the PAH8 generation compared to the control (13 µg/kg), while Budweiser (13 µg/kg) and Corona (13 µg/kg) resulted in no change, The Harbin (18 µg/kg) and Snow (18 µg/kg) showed a negative and therefore increasing effect on the PAH8 generation (Wang et al. 2019b). Heineken beer, with the most effective reduction in the PAH formation (67%), also had the highest phenol content and showed excellent abilities to scavenge free radicals (27%) (Wang et al. 2019b). The presence of phenolic compounds in beer, acting as inhibitors in free radical reaction pathways led to a reduction in the formation of PAHs in barbecued pork marinated with black beer, nonalcoholic Pilsner beer, and Pilsner beer (Viegas et al. 2014). A PAH8 reduction in comparison to the charcoal barbecued control of 53% was obtained with black beer marinade > nonalcoholic Pilsner beer (25%) > Pilsner beer (13%) (Viegas et al. 2014). The phenolic compounds in beer are principally flavonoids and phenolic acids, which may provide hydrogen to radical groups while undergoing oxidation to phenoxyl radicals (Wang et al. 2019b). Also, the addition of eight phenolic extracts from green tea (natural antioxidants) was found to decrease the BaP concentration in charcoal barbecued chicken wings (Wang et al. 2019a). However, wine marinades increased both BaP concentrations and the sum of PAH for pork steak from the neck (Tkacz, Wiek, and Kubiak 2012), indicating that the use of wine marinade resulted in the addition of extra material for pyrolysis and PAH formation, with a more potent effect than the radical-scavenging effect of the phenolic compounds from the wine. In general, marinade of red wine pomace seasonings decreased the PAH concentration in barbecued beef patties after storage in a high oxygen atmosphere but had the opposite effect immediately after barbecuing (García-Lomillo et al. 2017). Confirmation of the results by Farhadian et al. 2012 that the addition of oils to marinades would increase PAH formation was reported for 16 EPA PAH in a newer study (Wongmaneepratip and Vangnai 2017), who also found the effect to be lower for sunflower oil than palm oil. An extension of experiments with marinades of different pH included a 70% increased 16 EPA PAHs concentration for alkali marinade ($pH = 7.5$) as well as a darker color of the charcoal barbecued chicken. Treatment with other marinades, e.g., acidic marinade (pH 4.4), reduced the 16 EPA PAH concentrations, however not significantly (Wongmaneepratip and Vangnai 2017). Effects of marinades prepared from green tea, white tea, yellow tea, oolong tea, dark tea, and black tea on the formation of PAH8s in charcoal barbecued chicken wings were also studied (Wang et al. 2018). Results revealed that green tea showed the highest inhibitory effect on PAH8 formation (57%) followed by white tea (31%) > yellow tea (23%) > Oolong tea (2%) while increased PAH8 concentrations were detected with dark tea (54%) and black tea (126%) (Wang et al. 2018). Tea marinade also prevented the formation of BaP in charcoal barbecued pork belly significantly, with yerba mate tea exhibiting significantly higher radical-scavenging activity than green tea marinade (Park et al. 2017). In combination with radical scavenging activity, also inhibition of lipid oxidation in proportion to the concentration of tea infusion was considered as the overall effect. In contrast to Farhadian et al. 2012, they found that the highest BaP reduction was obtained after 8 h marinating (Park et al. 2017).

The effect of the addition of three natural antioxidants (rosemary extracts, tea polyphenol, and antioxidant bamboo) in soybean and palm oil on the levels of 16 PAH in typical Chinese fried food, youtiao were investigated. A synthesized antioxidant tert-butylhydroquinone (TBHQ) was added to the frying oil for comparison and was found to result in up to 39% reduction of PAHs when added to palm oil (Gong, Zhao, and Wu 2018). Addition of diallyl disulfide (DADS) and quercetin to a basic marinade for sirloin pork before charcoal barbecuing showed reductions in the final EPA 16 PAHs concentrations for both, however, only significant for the addition of 500 mg DADS (Wongmaneepratip, Jom, and Vangnai 2019). DADS is the predominant oil-soluble organosulfide in essential garlic oil, which has demonstrated an inhibitory effect on carcinogenic compounds in meat model systems (Wongmaneepratip, Jom, and Vangnai 2019).

Anjum et al. (2019) studied the content 6 PAHs (Ant, Fla, Nap, Chr, DbahA, and BaA) in grilled chicken, fish, and beef meat samples after they were marinated using a mixture of red chili, coriander, salt, cumin, and garlic paste, and additional mutton meat samples with only salt sprinkled before grilling. They confirmed the previous results with lower PAH concentrations in marinated samples however without the detection of BaA in any of the samples in the study (Anjum et al. 2019).

Other important parameters

Results show that there were significant differences in the levels of PAHs in covered and uncovered grill procedures, where covering the meat samples minimized the rate of 6 PAHs deposition in grilled meat (Anjum et al. 2019). Previously, elimination of PAH or at least 50% reduction of the concentrations was found for smoked products by the use of plastic packaging based on low-density polyethylene laminated film (Guillén, Sopelana, and Partearroyo 2000; Semanová et al. 2016). Also, the choice of sausage casing was previously illustrated to result in lower final PAH concentrations during smoking for cellulose casings compared to hog casing (Gomes et al. 2013) and when compared to sheep and collagen casings (Pöhlmann et al. 2013). It has however not been studied further whether this will affect the PAH concentration during barbecuing, expecting some casings to be more sticky to PAH than others.

Difficulties in assessing PAH exposure from barbecuing

A small to no increase in cancer risks have been associated with higher consumption of grilled or barbecued meat, especially red meat (John et al. 2011). Risks have mainly been based upon the presence of BaP in barbecued meat, whereas EFSA in 2008 concluded that BaP was only detected in approximately 30% of all foods containing carcinogenic PAHs (EFSA 2008).

EFSA has based its risk evaluation on the estimation of the margin of exposure (MOE) on the guidance that values below 10,000 can reasonably be considered to be associated with carcinogenic risks (EFSA 2005). Studies have resulted in MOE values above 10,000 for the intake of barbecued meat only, indicating that the intake of barbecued meat is of low health concern (Duedahl-Olesen et al. 2015a). However, it is essential to note that other food products contribute to the daily exposure to PAHs, such as smoked products, cereals, vegetable oils, etc., confirmed by EFSA findings of MOE values below 10,000 for European consumers' total diet (EFSA 2008). Therefore, it is vital to keep each source of PAHs as low as reasonably achievable.

The challenge to include consumption of barbecued meat in intake estimates is that national food consumption surveys commonly do not include food preparation methods. A survey in 2013 included a questionnaire for people participating in a barbecue experiment, but results were biased by only including participants already known to barbecue (Aaslyng et al. 2013). A worst-case evaluation in Norway in 2007 included barbecuing 30 times per year based on

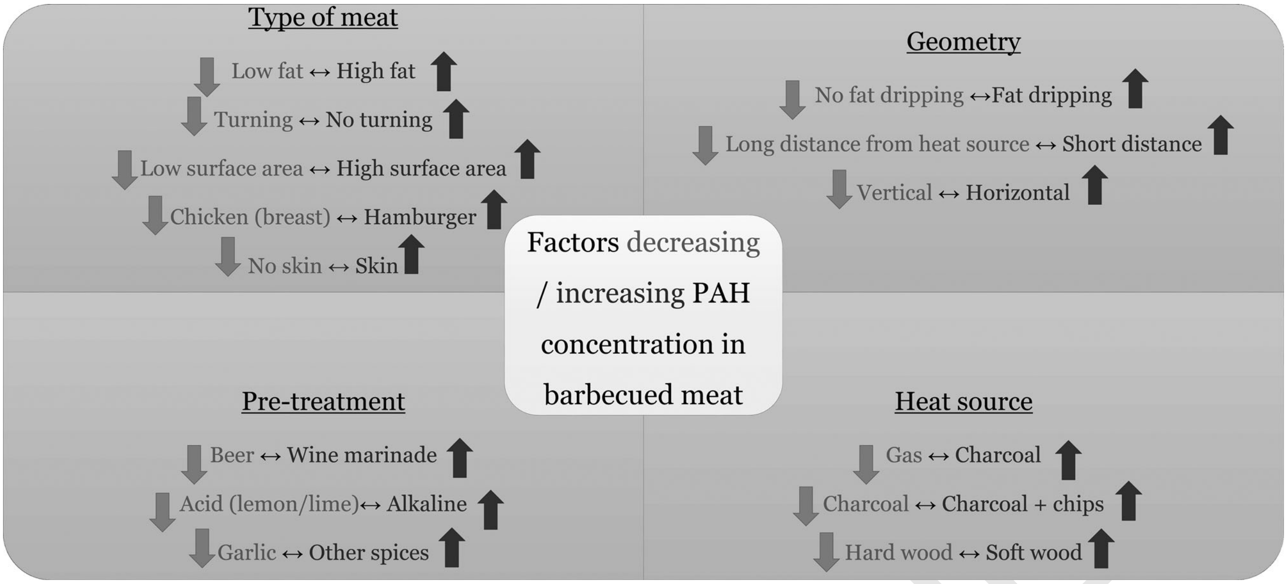

Figure 2. Summary of recommendations for minimizing PAH intake from home-barbecued meat.

maximum BaP concentrations in the barbecued meat, resulted in a MOE of 13,600 for barbecuing alone and by addition of 50% more BaP/day the MOE was below 10,000 indicating an increased cancer risk (Knutsen et al. 2007). For Danish consumers who frequently barbecue an estimate of barbecuing 30 times per year only corresponds to every day during a summer holiday and does not take into account the all-year popularity of barbecuing. A probabilistic approach assessing the cancer risk due to BaP in barbecued meat concluded that for the Danish population barbecuing would result in one to approximately 4100 new cancer cases over a lifetime (Jakobsen et al. 2018). The breadth of the range reflects the broad uncertainty of the estimate.

The exposure to barbecue fumes and the emission of PAH increase during barbecuing (Dyremark et al. 1995; Wu et al. 2015). Dyremark et al. 1995 estimated the amount of emitted PAH due to barbecuing to be a minor source with significant local peak exposures. Whereas, Wu et al. 2015 evaluated the exposure for consumers spending 1 h per day near a charcoal barbecue (including an unforeseen dermal contact) to be significant and with a total cancer incident of one to 100 for a lifetime of one million American adults (Wu et al. 2015). An evaluation of barbecue grill workers' exposure to PAHs revealed that an 8 h exposure to a charcoal barbecue would be well below the occupational threshold value of 200 µg per m^3 for PAH (Oliveira et al. 2019). Preventive measures to reduce the exposure was recommended to include adequate ventilation of the barbecue, regular washing of the skin, and use of clean working clothes (Oliveira et al. 2019).

Concluding remarks on best practice

Studies of processed foods have generally identified sources of PAHs with varying conclusions on which processing method results in the highest PAH concentrations. These conclusions are summarized in Figure 2.

A consistent reduction of the PAH levels has been obtained by broiling with electric and gas heat or broiling over charcoal in a no-drip pan (Chen and Lin 1997; El Badry, 2010; Farhadian et al. 2010; Kazerouni et al. 2001; Larsson et al. 1983; Lijinsky and Ross 1967; Rose et al. 2015).

However, it is difficult to draw definite conclusions on the choice of meat-type for barbecuing, even though it influences the final PAH concentrations. If not overcooked/burnt, the concentrations of PAHs in sausages and chicken breast tend to be on the lower end of the range.

Prevent meat charring by keeping an eye on your meat and remove black soot areas before consumption, which will reduce the PAHs concentration considerably.

It is crucial to minimize the pyrolysis of fat-containing "juices" since the resulting PAH-containing smoke will adhere to the product's surface and increase contamination. Minimized fat dripping can be obtained by keeping the fat content of the meat as low as possible, through the use of a pan or tray for the meat, wrapping the meat, or using a grill constructed in a way that fat cannot drip down (e.g., indirect heating), or by moving the heat source laterally, such as with vertical barbecues.

Mitigation of PAH can also be done by using marinades as pretreatment before barbecuing. Marinades of garlic and onions, tea marinades, beer, and especially acidic marinades without oil, reduce PAH concentrations considerably.

Pre-burn charcoals to avoid flames, and in general, for other heating materials, the consumer should make sure that the combustion is complete to reduce the PAH concentrations of the barbecued products. If possible, avoid reheating of charcoal. Keep an appropriate distance to the charcoal during barbecuing (>25 cm). Therefore, avoid the use of disposable barbecues.

Gas barbecues will result in lower PAH concentrations if adequately cleaned, and avoiding the use of contaminated oils (e.g., lubricating oil) or grease for cleaning.

For commercial barbecuing, PAH levels can be reduced by the use of continuous rotating skewer barbecues as well as efficient kitchen ventilation, which will remove steam and smoke formed.

This review has only been dealing with parameters affecting the PAH concentration of barbecued meat, but other health risks exist as well when barbecuing, such as the risk for microbial contamination.

Future studies on controlled grilling parameters and meat types are relevant for clarifying the combined effects of such parameters on the resulting PAH concentrations in the meat. It is also of interest to study whether a correct pairing of ingredients of a meal with e.g., anti-carcinogenic vegetables, can counterbalance the adverse health effects of PAHs.

Declaration of interest statement

Authors declare no conflict of interest.

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