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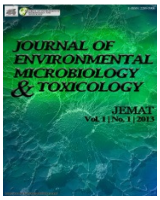

Effects of Cooking on Toxins Levels and Influence of Food Additives on Toxins Level in Food Products

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Abstract

Cooking is a process that could reduce the contamination level of toxic chemicals such polyaromatic hydrocarbons (PAHs) in several foodstuffs. However, this effect depends largely on the cooking process (grilling or frying), cooking material selection (charcoal, gas or wood), type of toxicant and the specific food item. This review discussed in detail pertaining to the effect of cooking and addition of food additives on the toxicant levels in foodstuff.

INTRODUCTION

Concentrations of various environmental pollutants such as Cd, total Hg, polychlorinated dibenzo-p-dioxins and furans (PCDD/PCDFs), dioxin-like polychlorinated biphenyls (DL-PCBs), polybrominated diphenyl ethers (PBDEs), polychlorinated naphthalenes (PCDNs) and hexachlorobenzene (HCB) in food samples had been determined in Catalonia (Northeast Spain) during 2000-2002. The dietary intakes of chemical contaminants included in the survey were again assessed in 2006 among the population of Catalonia[1]. Similar studies had been carried out in Canada where Canadian total Diet Survey (TDS) is performed annually to estimate the dietary exposure of toxic contaminants. In addition to that, an expansive PCDD/Fs/DL-PCBs monitoring program for foods had been established in Taiwan in 2000 [2]. These studies appeared to agree that food consumption was considered more important than ambient exposure to toxic contaminants though it brought impacts on health as well.

Polycyclic aromatic hydrocarbons (PAHs) is a class of environmental pollutants produced by pyrolysis or incomplete combustion of organic substances like petroleum fuels, wood, garbage, tobacco and charbroiled meat [3]. PAHs can be found abundantly in the environment. Contaminated soil, polluted water and air can be the possible sources of PAHs [4,5]. PAHs can also be found in food when undergoes heat processing such as smoke curing, broiling, roasting and grilling over open fires or charcoal [6]. Risks of 33 PAHs have been assessed and 15 PAHs, namely benzo[a]anthracene (BaA), cyclopenta[c,d]pyrene (CPP), chrysene (CHR), 5-methylchrysene (5MC), benzo[b]fluoranthene (BbF), benzo[j]fluoranthene (BiF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-c,d)pyrene (IcP), dibenzo[a,h]anthracene (DhA), benzo[g,h,i]perylene (BgP), dibenzo[a,e]pyrene (DeP), dibenzo[a,l]pyrene (Dlp), dibenzo[a,e)pyrene (DeP), dibenzo[a,i]pyrene (DiP), dibenzo[a,h]pyrene (Dhp) were reported to be the most carcinogenic, cytotoxic and mutagenic. In addition to that, the European Food Safety Authority (EFSA) recommends that benzo[c]fluorene (BsL) should be assessed as supported by the Joint FAO/WHO Experts Committee on Food Additives (JECFA) in 2005. Hence, these compounds are identified as 16 EU priority PAHs and always been examined in food samples. Polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs) and polychlorinated diphenyl ethers (PCDEs) are groups of chlorinated compounds found commonly in environment. These compounds are harmful to humans due to its potential negative effects of immunotoxicity and carcinogenicity. Recently formation of polychlorinated naphthalenes (PCNs) in food has become one of the interests of researchers as it shares similar structure with and pose similar toxicity PCDD/Fs [7]. Human exposed to these toxic contaminants mainly through dietary intake such as consumption of meat, fish and dairy products.

It is known that cooking processes widely modify the physiochemical and nutritional qualities of a number of foods. Thus, it is unsurprised that the toxins levels in food may undergo some changes. The focus has now been shifted to the effects of cooking on the levels of toxins in food stuffs. Several studies had been performed to evaluate the effects of cooking on the levels of PCDDs, PCDFs, DL-PCBS, PCDEs and PAHs in foodstuffs. The levels of toxins presented in food items including livestock, poultry, fish, seafood, milk, dairy products, oil, eggs, etc had been tested in these studies. Though the mechanism on how cooking affect the level of toxins in food is still unknown, the researchers believed that heat treatment did bring impacts on it. In fact, some studies also investigate the suitability of certain PAH compound which used as a marker for other carcinogenic PAHs. Despite of that, researcher found that presence of chlorine-containing additives and sucralose during cooking may affect the level of toxins in food samples. These clearly demonstrate the interest of researchers nowadays.

Studies Related to Effects of Cooking on levels of PCDDs, PCDFs, DL- PCBS, PCDEs and PAHs

In order to further investigate the effect of heat treatment on concentrations of toxins present (PCDDs, PCDFs, DL-PCBs PCDEs and PAHs) in cooked food, various investigations had been conducted. Meat products, fish, seafood and miscellaneous food samples had been included in the studies. Table 1 depicts the recent studies that had been done in the related field.

Table 1: Recent research which studied effects of different cooking methods on levels of toxins in different food items

Research	Cooking method	Food items	Toxins
Chen et al. [2]	Stir-fried Boiling Roasting Frying	Livestock Poultry Fish Seafood Milk Dairy products Oil Eggs	PCDDs PCDFs PCBs
Djinovic et al. [8]	Smoking	Beef ham Pork ham Bacon without skin Bacon with skin Cajna sausage Sremska sausage	PAHs
Perelló et al. [9]	Frying Grilling Boiling Roasting	Fish (sardine, hake, tuna) Meat (veal steak, loin of pork, breast, thigh of chicken, steak and rib of lamb) String bean Potato Rice Olive oil	PBDEs HCB PAHs
Perelló et al. [9]	Frying Grilling Boiling Roasting	Fish (sardine, hake, tuna) Meat (veal steak, loin of pork, breast, thigh of chicken, steak and rib of lamb) String bean Potato Rice Olive oil	PCDDs PCDFs PCBs PCDEs
Jira et. al, [10]	Smoking	Raw sausage Raw ham Cooked ham Frankfurter-type sausage Liver sausage	PAHs
Chung et. al, [6]	Charcoal grilling Charcoal roasting Gas roasting	Ham Bacon Sausage Chicken Beef Pork	PAHs
Rawn et al. [11]	Baking Boiling Frying	Finfish Non-finfish	PCBs PCDDs PCDFs

MATERIALS AND METHOD

All the food samples in these studies were purchased randomly either from local market or supermarkets. Cooking processes like frying, grilling and boiling were carried out using kitchen gas while roasting was done using an electric convention oven [9,10] whereas in the study of Chen et.al., [2] and Rawn et al. [11], the cooked food samples were prepared according to the recipes typically used in Taiwan and Canada respectively. The food samples were first homogenized and then extracted. The extractions of samples were varied according to the food samples tested. For PCDD/PCDFs, US EPA method 8290 was performed; for PCBs, US EPA method 1668 was utilized and for PCDEs, US EPA method 1625 and CARB method 429 was employed [10]. All the homogenized samples in these studies were added with internal standard mixture containing 15 $^{13}C_2$ – labeled PCDD/F and 12 $^{13}C_2$ – labeled PCB standards in order to control the whole sample preparation process. After completed the extraction process, multi-step clean-up procedure was carried out by using different solutions dependent on the types of food samples.

Analytical procedures to evaluate levels of PCBs and PCDD/Fs performed in these studies are similar where isotope dilution high-resolution gas chromatography and high-resolution mass spectrometry (HRGC-HRMS) method was employed. The cleaned extracts were injected and analyzed separately using Agilent 6890 Capillary Gas Chromatograph equipped with DB5-MS capillary column and coupled to Waters Autospec Ultima High Resolution Mass Spectrometer [10,11]. Fisons Gas Chromatogaph 8000 linked to Micromass Autospec Ultima EBE tri-sector mass spectrometer can also be used to determine the level of PCBs and PCDD/Fs as defined in US EPA Method 1613B [2].

Sample preparation for PAHs analysis involves three stages, namely accelerated solvent extraction, gel permeation chromatography and solid phase extraction [8,12]. Fast gas chromatography and high resolution mass spectrometry (GC-HRMA) was performed to analyze the content of PAHs in food samples in these studies while high pressure liquid chromatograph coupled to fluorescence detector (HPLC-fluorescence) was utilized in study of Chung et al. [6].

RESULTS AND DISCUSSION

A summary of findings of the studies had been presented in Table 2. Based on the table, it was found that the results of the studies were in accordance where influence of cooking on the levels of toxic contaminants depends not only on the particular cooking process, but even more the specific food item [6,8-12]. For instance, Perelló et al. [9] reported that though grilling and frying can reduce the level of PBDEs and HCB in sardine, these cooking processes brought adverse effects in hake where the contaminants contents were enhanced. Another typical example would be potato sample where the concentration of toxins only increased when it was fried but showed no changes when it was boiled. These studies also revealed that sardine sample has high content of PBDEs, PCDD/Fs and PCBs. Fortunately these contaminants can be reduced by frying and grilling. On the other hand, no notably changes were shown in string bean, rice and olive oil.

Formation of PAHs largely depends on the cooking processes as PAHs will only formed during pyrolysis or incomplete combustion. Generally PAHs content in food samples increased the most during roasting [6,9]. This may be due to the direct contact of food sample with the flame during open air combustion, pyrolysis of fats in meat generates PAH that become deposited on the meat [13]. Christy et al. [14] also reported that thermal degradation of steroids such as cholesterol is responsible for formation of carcinogenic and mutagenic PAHs. With this regard, the difference in method of roasting on levels of toxic contaminants present in food sample was also investigated [6]. The study revealed that charcoal-barbequed chicken sample had higher PAHs and BaP content as compared to that using gas and wood.

The suitability of BaP as a marker for the sum of 16 EU priority PAHs and 12 IARC possible or probably carcinogenic PAHs was another focus in these studies. Dijnovic et al. [8] had investigated the appropriateness of BaP as marker while Jira, [12] had compared both the BaP and “PAH4”. According to the Scientific Committee on Food (SCF), BaP can be used as an indicator for the occurrence and impact of carcinogenic PAHs in food. However, the more recent report of European Food Safety Authority (EFSA) in 2008, it concluded that BaP is not suitable to be used as marker, perhaps the sum of the four PAH compounds, BaP, CHR, BaA and BbF (“PAH4”) should be employed. Based on the study of Dijnovic et al. [8], BaP is a good marker as very strong correlation coefficient with 16 EU priority PAHs and 12 IARC possible or probably carcinogenic PAHs (R= 0.97; R= 0.99) had been generated.

However, Jira, [12] demonstrated a better correlation coefficient (R=0.99) for PAH4 to the sum content of 16 EU Priority PAH as compared to that of BaP (R= 0.90). Nevertheless, he pointed few disadvantages of using “PAH4” instead of BaP as indicator for PAH in food surveillance. Firstly, chromatography separation of CHR and TP is time consuming which is not feasible for routine measurements. On top of that, PAH compounds with very different carcinogenic potential are summed up to a total content without weighting. In other words, the toxicological potential of BbF, BaA and CHR were relatively lower than BaP, the sum content of “PAH4” might dominated by PAH compounds with lower toxicology potential. The carcinogenic potential of food might be overestimated.

Effects of addition of food additives on levels of PCB, PCDD/Fs and PCNs

Polychlorinated naphthalenes (PCNs) had gained concerns recently due to its similar structure and toxicity to polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs). The formation of PCNs occurs through the chlorination of naphthalene or a product of de novo synthesis from PAHs [15]. In conjunction with that, formation of polychlorinated biphenyls (PCBs) may also happen when PAHs react with chlorine at presence of catalysts during the cooking process under certain thermal conditions [15]. Complex foodstuff matrices may provide carbon sources whereas metallic elements like copper, zinc and iron which commonly found in food and utensils may act as catalysts [16]. With the combination of such substances during cooking process which conducted at high temperature, this in turn produces PCNs. Furthermore, chlorine-containing compounds are easily found in foodstuffs, in food additives and flavourings. Hence, these substances are speculated to contribute to the formation of PCNs during cooking.

Sucralose which acts as artificial sweetener and sugar substitute in food products like products baked at high temperature contains chlorine. Sucralose was found not to be stable under high temperatures. Rahn and Yaylayan, [17] concluded that sucralose manage to chlorinate glycerol to generate toxic chloropropanols at 250°C. Thus, a number of relevant studies had been conducted to determine the influence of chlorine-containing compounds on the formation of toxins especially PCNs. A summary of the findings of these studies is presented in Table 3.

Based on the two studies conducted by Dong et al. [16,18], presence of sucralose or chloropropanol (1,3-DCP) compounds could act as chlorine sources and promote the formation of PCNs, PCDD/Fs and di-PCBs in oil fumes. Before and after the samples (beef or oil) heated, the concentrations of toxins produced in samples and in oil fumes were calculated. It was found that with addition of sucralose and 1.3-DCP compounds, levels of dl-PCBs, dl-PCNs and PCDD/Fs in oil fume showed significant increment, however no notably changes demonstrated on the samples. This confirmed the assumption of removal or transference of PCBs, PCNs and PCDD/Fs may occur by thermal decomposition and volatilization. The toxins present in the sample (beef or oil) volatized during heating and thus the toxins are transformed from the solid to the gas phase. Toxins transformed from raw materials to oil fumes during the cooking process and rendered relatively lower level of toxins present in the food sample.

Table 2: Findings of studies

Research		**Finding**
Perelló et al. [9]	PBDEs	Sardine contained highest level of PBDEs but reduced significantly in grilled and fried samples All cooking processes enhanced the levels of PBDEs in hake especially after roasting All cooking processes enhanced the levels of PBDEs in pork All cooking processes reduced levels of PBDEs in veal, lamb, string bean, rice and olive oil Levels of PBDEs in chicken and potato were variable depends on particular cooking process (roasted chicken and fried potato have higher concentrations of PBDEs)
	HCB	Sardine contained highest level of HCB but reduced in cooked samples All cooking processes enhanced the levels of HCB in hake All cooking processes enhanced the levels of HCB in meat products especially roasted chicken No difference was observed in string bean, rice and olive oil Significant increase was observed in fried potato
	PAHs	Frying enhanced the levels of PAHs in all food samples Highest total PAH levels correspond to roasted sample Total PAHs in boiling rice is twice more than the raw sample Highest sum of total carcinogenic PAHs was found in olive oil sample, followed by roasted hake and fried sardine.
Perelló et al. [10]	PCDD/Fs and PCBs	All cooking processes reduced the levels of PCDD/Fs in sardine, veal steak and loin pork All cooking processes enhanced the levels of PCDD/Fs in hake and tuna No difference was observed in chicken, string bean, rice, olive oil Total PCDD/Fs in fried potato is three times higher than in raw sample
	PCDEs	Raw sardine sample contained highest level of PCDEs, followed by oil sample All cooking processes enhanced total PCDEs level in fish and meat products except fried sardine and roasted chicken where their PCDEs levels were lower than the raw sample Cooking reduced the level of PCDEs in string bean, rice and olive oil
Rawn et al. [11]	PCB	Frying enhanced the level of PCB in six finfish samples include catfish, croaker, grouper, monkfish, pomfret and whiting Boiling enhanced the level of PCDD/Fs in non-finfish samples include cuttlefish, sea quirt, skate and squid Though difference between the raw and cooked sample was observed, the differences were generally small
Dijnovic et al. [8]	PAHs	Raw ham and frankfurter-type sausage had the highest level of BaP whereas cooked ham has the lowest BaP level. Frankfurter-type sausage had the highest level of PAH4 while cooked ham had the lowest PAH4 content PAH4 has better correlation coefficient for the sum content of 15+1 EU priority PAH as compared to BaP

Hence, Dong et al. [16] suggested that appropriate use of chlorine-containing additives and flavourings during cooking could help decrease the risk of human exposure to dl-PCBs. However, his more recent research [18] concluded that these unintentionally polychlorinated aromatic compounds in the oil fumes especially during deep frying should be avoided as these may pose risk to human health.

CONCLUSIONS

Though previous studies had suggested that cooking might able to reduce the levels of chemical contaminants in some foodstuff, the recent studies do not corroborate with that as polychlorinated aromatic hydrocarbons tend to be produced during cooking processes such as grilling and frying. On top of that, the studies shown that instead of the types of cooking processes, the influence of cooking on toxins levels depends largely on the specific food items. The materials used in grilling such as charcoal, gas and wood make difference on the levels of toxins produced where charcoal-barbecued meat showed the highest concentration of PAHs. Though PAH4 had been suggested to be a better marker for toxicology potential in food products as compared to BaP, it comes with some limitations and there is a need for more studies to determine its suitability of use as an indicator. Levels of toxins

Table 3; Findings of this study

Research	Sample	Toxins	Findings
Dong et al. [16]	Beef with oil Beef w/o oil Beef with sucralose Beef with chloropropanol 1,3-DCP	dl-PCBs	Cooking reduced the levels of dl-PCBs in cooked beef sample Presence of sucralose (45g) increase dl-PCBs in cooked beef but not significant Concentration of dl-PCBs in cooked beef sample was the highest in the presence of 1,3-DCP (40.2g) Presence of sucralose and 1,3-DCP in beef sample increase the levels of di-PCBs in oil fumes Appropriate use of chlorine-containing additives and flavourings during cooking could reduce risk of human exposure to dl-PCBs
Dong et al. [18]	Peanut oil with sodium chloride	dl-PCNs	PCNs couldn't be detected in the peanut oil and olive oil which added with sodium chloride either in the cooking oil or oil fumes at 200°C and 245°C
	Peanut oil with sucralose	dl-PCBs	PCNs couldn't be detected in the olive oil which added with sucralose either in the cooking oil or oil fumes at 200°C
	Olive oil with sodium chloride	PCDD/Fs	When peanut and olive oil heated with sucralose at 245°C, relatively high levels of PCNs, dl-PCBs and PCDD/Fs were detected in oil fumes but not in cooking oil
	Olive oil with sucralose		

present in oil fume increase significantly when food samples added with chlorine-containing compound. This proved the assumption that toxins in sample will volatized during cooking and be collected in the oil fumes. Nevertheless, the appropriateness of using chlorine-containing compound to reduce the risk of human exposure to dl-PCBs is in doubt. The dietary consumption of the toxins might be reduced, however the unintentionally produced toxins are found abundantly in the oil fumes which in turn increase the potential for human exposure. Further investigations need to be conducted to assure the suitability of using chlorine-containing compound as tools to reduce the risk of human exposure to dl-PCBs.

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