Food Additives and Contaminants
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/tfac19

Polycyclic aromatic hydrocarbons in smoked food products and commerical liquid smoke flavourings

Enayat A. Gomaa [a], J. Ian Gray [a], Samir Rabie [a], Clemente Lopez-Bote [b] & Alden M. Booren [a]

[a] Department of Food Science and Human Nutrition, Michigan State University, East Lansing, MI, 48824, USA

[b] Facultad de Veterinaria, Universidad de Extremadura, Caceres, 10071, Spain

Version of record first published: 10 Jan 2009.

To cite this article: Enayat A. Gomaa, J. Ian Gray, Samir Rabie, Clemente Lopez-Bote & Alden M. Booren (1993): Polycyclic aromatic hydrocarbons in smoked food products and commerical liquid smoke flavourings, Food Additives and Contaminants, 10:5, 503-521

To link to this article: http://dx.doi.org/10.1080/02652039309374174

FOOD ADDITIVES AND CONTAMINANTS, 1993, VOL. 10, NO. 5, 503–521

Polycyclic aromatic hydrocarbons in smoked food products and commerical liquid smoke flavourings

ENAYAT A. OMAA,† J. IAN GRAY,†§ SAMIR RABIE,† CLEMENTE LOPEZ-BOTE‡ and ALDEN M. BOOREN†

†Department of Food Science and Human Nutrition, Michigan State University, East Lansing, MI 48824, USA; ‡Faculdad de Veterinaria, Universidad de Extremadura, 10071 Caceres, Spain

(Received 25 January 1993; accepted 11 March 1993)

Smoked foods including turkey, pork, chicken, beef and fish products were screened for the presence of carcinogenic and non-carcinogenic polycyclic aromatic hydrocarbons (PAHs). Eighteen commercial liquid smoke flavourings and seasonings were also analysed. Total PAH concentrations in smoked meat products ranged from 2·6 μg/kg in a cooked ham sample to 29·8 μg/kg in grilled pork chops, while those in fish products ranged from 9·3 μg/kg in smoked shrimp to 86·6 μg/kg in smoked salmon. Total concentrations of the carcinogenic PAHs (benzo[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*a*]pyrene, dibenzo[*a*,*h*]anthracene, and indeno[1,2,3-*c*,*d*]pyrene) ranged from non-detectable in several meat products to 7·4 μg/kg in grilled pork chops, and from 0·2 μg/kg in trout to 16·0 μg/kg in salmon. In liquid smoke flavourings and seasonings, total PAH concentrations ranged from 6·3 to 43·7 μg/kg, with the carcinogenic PAHs ranging from 0·3 to 10·2 μg/kg.

Keywords: PAHs, smoked meat, liquid smoke flavourings

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are produced from the incomplete combustion or thermal decomposition (pyrolysis) of organic material. The quantity and composition of the PAHs produced are closely related to the reaction conditions, temperature and amount of air and, therefore, may vary considerably (Tóth and Potthast 1984, Vaessen *et al.* 1988). The widespread occurrence of PAHs in the environment is well documented as is their biological and carcinogenic activity (Zedeck 1980, IARC 1983, 1987). The occurrence of PAHs in food may result from their sorption from a contaminated environment or from food preparation (Lo and Sandi 1978, Kramers and Van Der Heijden 1988). The variation in the PAH profile in food products also depends on the source of the contamination (Vaessen *et al.* 1988).

The presence of benzo[*a*]pyrene and other carcinogenic PAHs in foods has received considerable attention over the past three decades (Maga 1988). PAHs have been detected and quantified in many foods including charcoal-broiled meat, smoked/grilled foods, fats and oils, plant materials, seafood, liquid smokes and beverages (Haenni and Fischbach 1974, Fretheim 1976, Lo and Sandi 1978,

§ To whom correspondence should be addressed.

0265-203X/93 $10·00 © 1993 Taylor & Francis Ltd.

Doremire *et al.* 1979, Howard and Fazio 1980, Joe *et al.* 1984). The possible sources of PAH contamination of foodstuffs are numerous and include contaminated soils, polluted air and water, mode of cooking, food processing, type of fuel (coal, wood, manure, diesel, propane), smoke generation conditions, wood composition, temperature of pyrolyses, endogenous sources and meat lipid content (Thorsteinsson and Thordarson 1968, Tilgner 1977, Doremire *et al.* 1979, Toth and Potthast 1984, Maga 1988, Vaessen *et al.* 1988).

Historically, benzo[*a*]pyrene has been used as a reference indicator compound for carcinogenic PAHs (Rhee and Bratzler 1970). Five PAHs (benzo[*a*]anthracene, benzo[*a*]pyrene, benzo[*b*]fluoranthene, dibenzo[*a,h*]anthracene and indeno[1,2,3-*c,d*]pyrene), however, account for the bulk of all carcinogenic PAHs in food as well as in air (Lo and Sandi 1978, Lee *et al.* 1981, Fazio and Howard 1983, IARC 1983, Kramers and Van Der Heljden 1988).

The initial objective of this study was to evaluate a number of commercial smoked poultry, red meat and fish products for the presence of carcinogenic and non-carcinogenic PAHs. A second objective of this study was to develop a rapid and sensitive liquid chromatographic procedure for the separation and quantitation of PAHs in commercial liquid smokes.

Materials and methods

Samples

All commercial smoked meat products and liquid smoke seasonings were purchased from local supermarkets in the mid-Michigan area. Liquid smoke flavourings were donated by commercial companies.

Reagents

Dimethyl sulphoxide (DMSO) was obtained from J. T. Baker Chemical Co. (Phillipsburg, NJ). Cyclohexane, 1,1,2-dichloro-1,2,2,-trifluoroethane (TCTFE), hexane, methanol, ethanol, dichloromethane (DCM) and acetonitrile (HPLC grade) were purchased from E.M. Science (Gibbstown, NJ). Florisil was obtained from Fisher Scientific (Fair Lawn, NJ). Silica gel (70–230 mesh) and alumina (70–230 mesh ASTM) were purchased from Sigma Chemical Co. (St Louis, MO).

Standards

Sixteen PAHs were selected for determination in foods based on their occurrence and carcinogenicity, and available methodology (table 1). Reference PAHs were purchased from Supelco, Inc., Supelco Park (Bellefonte, PA), while the internal standard triphenylene was obtained from Sigma Chemical Co. Standard PAH solutions were stored at 4°C in glass vials wrapped in aluminium foil to avoid possible light degradation of some PAHs. Chromatographic columns were also wrapped in aluminum foil to avoid exposure to direct sunlight. Concentration of food and liquid smoke solvent extracts was performed by rotary evaporation under reduced pressure at 35°C. Further concentration of the extracts to less than 2 ml was accomplished using a slow stream of nitrogen.

Table 1. Abbreviations and carcinogenicity of polycyclic aromatic hydrocarbons (PAHs)[a].

IUPAC name	Abbreviation	Carcinogenicity[b]
Naphthalene	N	?
Acenaphthylene	APL	?
Acenaphthene	AP	?
Fluorene	Fl.	?
Phenanthrene	Phen	?
Anthracene	A	?
Fluoranthene	F	
Pyrene	Py	−
Benzo[*a*]anthracene	BaA	+
Chrysene	Ch	±
Benzo[*b*]fluoranthene	BbF	+
Benzo[*k*]fluoranthene	BkF	−(±)
Benzo[*a*]pyrene	BaP	+ + +
Dibenzo[*a,h*]anthracene	DBahA	+ + +
Benzo[*g,h,i*]perylene	BghiP	−(+ +)
Indeno[1,2,3 *c,d*]pyrene	IcdPy	+

[a] Adapted from Toth and Potthast (1984) and IARC (1983, 1987).
[b] + + +, very carcinogenic; + +, carcinogenic; +, somewhat carcinogenic; −, not carcinogenic; ± uncertain; ?, inadequate information.

Analytical methods

Extraction and purification of PAHs in smoked foods. Extraction of PAHs from foods and subsequent clean-up of the extracts were carried out as described by Joe *et al.* (1984) and Fazio (1987).

Extraction of PAHs in liquid smokes. The extraction and clean-up procedure was adapted from that of Black *et al.* (1979). Twenty five grams of liquid smoke were saponified for 3 h with 9 g KOH and 100 ml methanol in a round bottom boiling flask equipped with a Friedreich condenser. When the flask was cooled, the contents were filtered into a separatory funnel through an ethanol-rinsed glass wool pad. The flask was rinsed with 250 ml cyclohexane and 100 ml 80% methanol in water (*v/v*) and added to the separator funnel through the glass wool pad. The contents were shaken vigorously for 2 min. After separation of the layers, the aqueous layer was extracted for 2 min with a fresh aliquot of cyclohexane (250 ml) and 80% methanol (100 ml). Again, the aqueous layer was separated and extracted with fresh cyclohexane (100 ml) and 80% methanol (50 ml). The combined cyclohexane extracts were washed with three (100 ml) aliquots of warm water (45°C), dried over anhydrous sodium sulphate, and then concentrated to approximately 15 ml in a rotary evaporator.

Sample clean-up. The concentrated extract was purified by Florisil column chromatography. A glass column (14·5 × 250 mm) was plugged with glass wool and packed with 10 g deactivated Florisil (2% water), 2·5 g anhydrous Na_2SO_4 and

1 cm sand. The column was prewashed with 75 ml of hexane. The concentrated cyclohexane extract was applied to the top of the Florisil column and eluted using a sequential solvent system. The first fraction was eluted with 50 ml of hexane, the second with 100 ml of 30% DCM in hexane (v/v), the third fraction with 100 ml of 50% DCM in hexane, and the fourth with 100 ml of 70% DCM in hexane. Fractions 2, 3 and 4 were combined, and rotary evaporated to near dryness under reduced pressure at 35°C. The residue was dissolved in acetonitrile, filtered through a filter (pore size 0·45 μm, Millipore Corporation, Bedford, MA) and then concentrated using a slow stream of nitrogen for HPLC analysis. During the elution process, the column was monitored occasionally with an ultraviolet light (254, 365 μm) to locate the fluorescent PAH fraction. Complete elution of the PAHs from the column was checked by sonicating the Florisil packing material with 50 ml of 70% DCM in hexane, filtering, concentrating, and analysing by high performance liquid chromatography (HPLC) The first fraction was also concentrated and analysed by HPLC to check the absence of PAHs of interest.

Quantitation of PAHs in extracts. PAHs in the extracts from the smoked foods and liquid smokes were analysed by HPLC. The HPLC system consisted of a model U6K universal injector, solvent delivery system (model 501), fluorescence detector (model 420-AC) with a 254 nm excitation filter and 375 nm emission filter, and a variable wavelength detector set at 254 nm (Waters Associates, Inc. Milford, MA). The HPLC system, solvent programme and peak integration parameters were controlled by a baseline 810 chromatograph workstation (Dynamic Solutions, Millipore Corp.). Samples were analysed using a Supelcosil LC-PAH column (25 cm × 4·6 mm o.d., Supelco, Inc.).

PAHs were separated at ambient temperature using a gradient elution programme with a flow rate of 1 ml/min. The initial mobile phase was 60% acetonitrile in distilled water (5 min), which was then gradiently changed to 100% acetonitrile in 15 min, held at 100% for 15 min, then decreased to 60% acetonitrile over 10 min, and held at 60% for 15 min. Twenty μl of the sample extracts or the standard solution were injected. The standard solution was injected after every fourth sample extract.

Recoveries of PAHs

To evaluate the accuracy and reliability of the clean-up procedure developed for liquid smokes, cyclohexane was spiked with a mixture of standard PAHs to give individual PAH concentrations ranging from 3 to 5 μg PAH/l cyclohexane and was applied to the Florisil column.

Liquid smoke procedure. Recoveries of PAHs in the liquid smokes were determined in triplicate by spiking the samples with a mixture of standard PAHs, sufficient to produce levels equivalent to 5 μg PAH/kg hickory liquid smoke seasoning.

Smoked food procedure. Recoveries of PAHs in the smoked foods were determined in triplicate by spiking smoked salmon or turkey breast with a mixture of PAHs sufficient to produce concentrations equivalent to 3·2 μg PAH/kg.

The identification of the PAHs was determined by comparing the retention times of UV and fluorescence peaks in the sample chromatograms with those of known PAH standards.

Results and discussion

The fluorescent detection limits for each PAH in the standard solution ranged from 0·01 μg/kg for A to 0·4 μg/kg for DB*ah*A (table 2). Values for ultraviolet detection ranged from 0·02 μg/kg for B*k*F to 0·56 μg/kg for A. The detection limits depend on the compound and on the detectors used (NIOSH 1985). Chromatograms for the PAH standard mixture (0·4 ng of each PAH) and the internal standard (triphenylene) obtained by fluorescent and ultraviolet detection are shown in figure 1. Four PAH compounds, N, APL, AP and FL, did not appear in the fluorescence chromatogram because they do not fluoresce at an excitation wavelength near 375 nm (Facio 1987). Thus, only the UV chromatograms were used for calculating the concentrations of these four PAHs in the food products and liquid smokes. These PAHs, however, are not included in the total PAH concentrations because they are not regarded as carcinogenic (table 1). The concentrations of the 12 other PAHs were calculated from both the ultraviolet and fluorescence responses.

Percent recoveries of PAHs for the two extraction procedures are presented in tables 2 and 3. Each recovery value represents the average of triplicate determinations. For the smoked meat analytical procedure, the average recoveries ranged from 80% ± 6·7 to 112% ± 8·2 with a mean coefficient of variation (CV) of 5·1% for ultraviolet detection, and from 75% ± 3·5 to 98 ± 8·2, with a mean CV of 5·8% for fluorescence detection (table 2). For the liquid smoke analytical

Table 2. Percent recovery of polycyclic aromatic hydrocarbons added to smoked meat products.

	Detection method[a,b]			
	Ultraviolet		Fluorescence	
PAH compounds[c]	Mean ± SD	CV%	Mean ± SD	CV%
N	96 ± 4·9	5·1	nd	
APL	94 ± 2·4	2·6	nd	
AP	80 ± 6·7	8·4	nd	
FL	99 ± 7·3	7·3	nd	
Phen	100 ± 5·7	5·7	95 ± 5·7	6·0
A	112 ± 8·2	7·3	98 ± 8·2	8·3
F	94 ± 3·3	3·5	75 ± 3·5	4·6
Py	90 ± 2·5	2·8	85 ± 4·5	5·3
BaA	95 ± 4·9	5·1	79 ± 4·1	5·2
Ch	92 ± 1·6	1·7	80 ± 2·5	3·1
BbF	103 ± 2·5	2·4	95 ± 3·3	3·4
BkF	107 ± 2·5	2·3	88 ± 7·3	8·3
BaP	95 ± 9·8	8·8	80 ± 8·2	10·3
DB*ah*A	100 ± 6·5	6·5	81 ± 6·5	8·0
B*gh*iP	98 ± 8·0	8·2	95 ± 4·1	4·3
Ind*c*Py	100 ± 2·5	2·5	80 ± 2·5	3·1

[a] LC determination with fluorescence and UV detection.
[b] Values represent average of three determinations.
[c] Abbreviation description, see table 1.
SD: Standard deviation, CV: Coefficient of variation, nd: not detected.

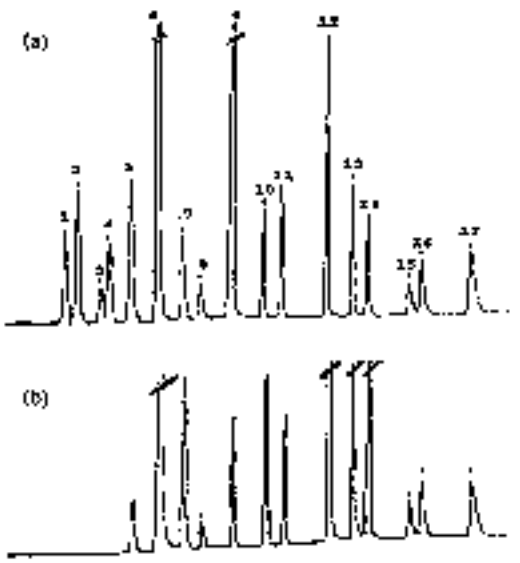

Figure 1. Liquid chromatograms for PAH standards injected on LC-column. (A), UV, 254 nm; (B), fluorescence, 305 nm. 1, N; 2, APL; 3, AP; 4, FL; 5, Phen; 6, A; 7, F; 8, Py; 9, internal standard; 10, BaA; 11, Ch; 12, BbF; 13, BkF; 14, BaP; 15, DBahA; 16, BghiP; 17, IcdPY.

Table 3. Percent recovery of polycyclic aromatic hydrocarbons added to liquid smoke flavourings.

	Detection method[a,b]			
	Ultraviolet		Fluorescence	
PAH compounds[c]	Mean ± SD	CV%	Mean ± SD	CV%
N	95 ± 9·9	11·6	nd	
APL	95 ± 6·9	8·1	nd	
AP	79 ± 8·1	10·2	nd	
FL	100 ± 2·0	2·0	nd	
Phen	100 ± 8·2	8·2	82 ± 8·0	10·7
A	98 ± 5·1	5·2	71 ± 8·9	9·1
F	100 ± 7·0	7·0	100 ± 7·4	7·4
Py	106 ± 7·2	6·7	110 ± 0·9	1·2
BaA	95 ± 3·2	3·4	90 ± 4·5	5·0
Ch	96 ± 4·0	4·2	80 ± 7·0	8·7
BbF	90 ± 9·8	10·9	83 ± 3·7	4·4
BkF	101 ± 5·0	4·8	99 ± 2·5	2·5
BaP	76 ± 9·0	11·8	66 ± 9·8	11·6
DBahA	66 ± 7·0	10·5	63 ± 16·1	25·9
BghiP	98 ± 5·0	5·1	93 ± 1·4	1·5
IcdPy	90 ± 9·0	10·0	86 ± 5·7	6·6

[a] LC determination with fluorescence and UV detection.
[b] Values represent averages of three determinations.
[c] Abbreviation description, see table 1.
SD, Standard deviation; CV, coefficient of variation; nd, not detected

procedure, the average recoveries ranged from 66% ± 7·0 to 106% ± 7·2 with a mean CV of 7·5% for ultraviolet detection, and from 63% ± 16·3 to 110% ± 0·9, with a mean CV 8·0% for fluorescence (table 3). The low coefficients of variation of the mean value for the two extraction procedures indicate that the repeatability of the procedures is very good. In general, the average recovery for the liquid smoke method was lower than that for the smoked food analytical method. For the liquid smoke procedure, a recovery study involving cyclohexane spiked with standard PAHs indicated that most of the loss occurred during the column clean-up. The losses of certain PAHs can be accounted for by the irreversible adsorption of some PAHs onto the Florisil column or by photoreaction of B*a*P as cited by Lawrence and Dorcas (1984), and observed earlier by Insco (1964) and Sawicki, *et al.* (1965). However, the recoveries in this study were greater than those of Lawrence and Dorcas (1984), due possibly to the modified clean-up procedure applied to the Florisil column.

PAH content of smoked foods

The smoked food products analysed in this survey were purchased from local supermarkets in the mid-Michigan area. A total of 62 samples were analysed and included poultry (turkey and chicken), red meat (pork and beef) and fish products.

PAH content of smoked poultry products. Fourteen turkey samples and eight chicken samples produced by eleven commercial processors were analysed (table 4a). The values shown are the average of duplicate analyses per product. Some of products were processed with liquid smoke flavourings and others with natural smoke (wood smoke).

For the smoked turkey products, the concentrations of the individual PAHs ranged from non-detectable in most of the samples to 3·5 μg/kg for Phen in wood-smoked sausage. The total PAH concentration ranged from 2·8 μg/kg in breast processed with liquid smoke to 9·6 μg/kg for wood-smoked sausage. In all the samples, Phen and Py were the major compounds present. The concentrations of PAHs with known carcinogenicity, i.e. B*a*A, B*b*F, B*a*P, DB*ah*A, and I*cd*Py, ranged from non-detectable in liquid smoke-processed breast samples to 1·9 μg/kg in wood-smoked breast samples. The percentage contributions of the carcinogenic compounds to the total PAH concentrations in the samples processed with liquid smoke were 0, 5·3, and 12·5% for breast, sausage and bacon, respectively. The corresponding values for the wood-smoked products were 32·2, 19·0, 11·5 and 17·2% for breast, sausage, sausage (different processor) and bacon, respectively. With respect to B*a*P, 57% of the samples had no trace of the compound, while the remaining samples had concentrations of <0·4 μg/kg. These results indicate that products processed with natural wood smoke (breast, sausage and bacon) had higher total PAH and carcinogenic PAH contents than those processed with liquid smoke flavourings (table 4a).

The concentrations of individual PAHs in the smoked chicken products ranged from non-detectable in sliced breast (liquid smoke flavouring added) to 6·1 μg/kg for Phen in barbecued wings. Total PAH concentrations ranged from 4·5 μg/kg in sliced breast processed with liquid smoke to 22·4 μg/kg in barbecued wings. The concentration of the carcinogenic PAHs (B*a*A, B*b*F, B*a*P, DB*ah*A, and I*cd*Py) ranged from non-detectable for sliced breast (liquid smoke flavouring) to 5·5 μg/kg in barbecued wings. B*a*P was not detected in the sliced breast (liquid smoke added)

Table 4a. PAH concentrations in selected commercial smoked poultry (turkey and chicken) products.

	Concentration of PAHs (μg/kg)[a,b]													
Smoked products	Phen	A	F	Py	BaA[d]	Ch	BbF[e]	BkF	BaP[e]	DBahA[d]	BghiP	Ind[1]Py[e]	Total PAHs	Total[f] PAHs (car.)[g]
Turkey														
Breast[c]	0·4	1·0	0·1	0·4	nd	0·2	nd	0·2	nd	nd	0·3	nd	2·8	nd
Breast[d]	0·7	nd	0·8	0·8	1·0	1·4	0·5	0·2	0·1	0·3	0·1	nd	5·9	1·9
Sausage[d]	2·9	0·3	1·5	1·1	0·6	0·9	0·8	0·1	0·1	0·1	nd	nd	8·4	1·6
Sausage[d]	3·5	0·5	1·7	2·2	0·5	0·3	0·5	nd	nd	nd	0·3	0·1	9·6	1·1
Sausage[c]	0·9	0·7	0·5	0·3	nd	0·4	0·1	0·8	nd	0·1	nd	nd	3·8	0·2
Bacon[c]	1·0	0·6	nd	0·9	0·3	0·2	0·1	0·1	nd	nd	nd	nd	3·2	0·4
Bacon[d]	1·6	1·6	0·8	1·6	0·7	0·4	nd	0·9	0·4	0·3	0·6	0·2	9·3	1·6
Chicken														
Sausage[d]	1·8	0·5	1·0	1·4	0·9	1·3	0·1	0·4	0·1	0·1	0·4	0·3	8·1	1·7
Sliced breast[c]	1·5	1·0	0·9	0·8	nd	0·2	nd	0·1	nd	nd	nd	nd	4·5	nd
Barbecued wings	6·1	1·4	2·5	3·2	1·1	1·9	1·1	0·8	0·8	2·0	1·1	0·5	22·4	5·5
Barbecued chicken	5·2	1·9	2·7	0·9	0·5	0·7	0·8	0·2	0·7	1·0	1·0	0·4	17·3	3·2

[a] Average of duplicate analyses of two samples per commercial processor (four determinations).
[b] Abbreviation description, see table 1.
[c] Liquid smoke flavouring added.
[d] Wood-smoked.
[e] Carcinogenic.
nd: not detected.

but was present in those samples that were wood-smoked or barbecued. The percent contributions of the carcinogenic compounds to the total PAH concentrations were 20·9, 24·5, and 18·5% for sausage, barbecued wings and barbecued chicken, respectively. These data indicate that wood-smoked and barbecued products have the highest concentrations not only of total PAHs but also of the carcinogenic compounds. While no details of the barbecuing process appeared on the packages, it is well established that barbecuing of meats will result in the formation of significant quantities of PAHs. For example, Lijinsky and Ross (1967) and Doremire *et al.* (1979) reported that charcoal broiling of meats presents an unique situation, in that the amount of fat originally present in the meat, the closeness of the meat to the heat source, and the exposure time of the meat to the heat can all contribute to PAH formation. These investigators also concluded that to minimize the production of carcinogens such as BaP, the method of preparation should avoid contact of the meat with the cooking flames, cook for a longer time at lower temperatures, and use meat with a minimum of fat. The data obtained in this study for turkey and chicken breast products agree with those reported by Fazio (1987) for the same products. In all the samples, Phen and Py were the major compounds. These data can be explained by the results of Maga (1986) who reported that Phen and Py were the principal PAHs in condensed mesquite and hickory wood smokes.

Smoked turkey and chicken products were also screened for the presence of the four non-carcinogenic PAHs (N, APL, AP and FL) which were detected only by UV absorption (table 4b). Naphthalene was not present in any of the turkey and chicken products. The concentrations of APL, AP and FL varied from non-detectable in some turkey and chicken products to as high as 368 μg/kg (turkey sausage) and 220 μg/kg (barbecued chicken) for APL. The non-carcinogenic

Table 4b. Concentrations of UV-detected polycyclic aromatic hydrocarbons in commercial smoked poultry (turkey and chicken) products.

Smoked products	Concentrations of non-carcinogenic PAHs (μg/kg)[a,b]				
	N	APL	AP	FL	Total PAHs
Turkey					
Breast[c]	nd	nd	6	4	10
Breast[d]	nd	nd	256	40	296
Sausage[c]	nd	nd	nd	175	178
Sausage[d]	nd	368	29	40	437
Sausage[e]	nd	nd	nd	nd	nd
Bacon[e]	nd	nd	nd	1	1
Bacon[d]	nd	2	1	1	4
Chicken					
Sausage[d]	nd	5	3	nd	8
Sliced breast[c]	nd	nd	nd	nd	nd
Barbecued wings	nd	37	43	2	57
Barbecued chicken	nd	220	45	3	268

[a] Abbreviated description, see table 1.
[b] Average of duplicate analyses of two samples per commercial processor (four determinations).
[c] Liquid smoke flavouring added.
[d] Wood-smoked.
nd: not detected

compounds were not detectable in 18% of the turkey and chicken samples. However, 36% of the samples had concentrations of < 10 μg/kg, while 45% of the samples had concentrations ranging from 57 to 437 μg/kg. These data also reveal that samples processed with liquid smoke have smaller concentrations of these compounds than samples processed with wood smoke (table 4b). Limited information is available about the presence of these four non-carcinogenic PAHs in smoked foods. Maga (1988) reported that the presence of N and APL may be due to contamination of the products from air. Kushwaha *et al.* (1985) evaluated the PAH composition of various commercial charcoal briquets (coal, wood) and detected different levels of N, APL and F, as well as other PAH compounds. The smoke source can dramatically influence both the PAH level and the type of compounds present in the smoke and subsequently their deposition on the surface of smoked foods (Maga 1988).

PAH content of smoked red meat products. Sixteen smoked pork and six smoked beef samples obtained from eleven commercial processors were analysed (table 5a). The values presented are the average of duplicate analyses of two samples per product.

The concentrations of individual PAHs in the pork samples ranged from non-detectable in several samples to 8·7 μg/kg for Phen in the sausage samples. Total PAH concentrations ranged from 2·6 μg/kg in cooked ham (liquid smoke flavouring added) to 29·8 μg/kg and 29·7 μg/kg in grilled pork chops and sausage (wood-smoked), respectively. The combined concentrations of the five carcinogenic PAHs ranged from non-detectable for cooked ham samples to 7·4 μg/kg for pork chops and 6·8 μg/kg for sausage. The concentrations in the bacon, sliced and whole ham samples were greater than those reported by Fazio (1987). The greater concentration of PAHs detected in the grilled pork chops again can be explained by the findings of Lijinsky and Ross (1967) who observed a correlation between the fat content and the accumulation of BaP in grilled meat. They theorized that the fat drippings fall on the hot coals and are pyrolysed, producing BaP and other PAHS which are subsequently deposited onto the surface of the meat. The percentage contributions of the five carcinogenic compounds to the total PAH concentrations in the samples processed with liquid smoke were 0% for cooked ham and 4·4% for bacon. Corresponding values for the products processed with natural wood smoke were 7·6% for bacon, 17·9% for the whole smoked hams, 22·9% for sausage, 23·1% for sliced smoked ham, 24·8% for grilled pork chops and 31·8% for another sausage sample. BaP was not detected in the cooked ham processed with liquid smoke. The concentrations of BaP detected in this survey were smaller than those reported by Doremire *et al.* (1979) for pork grilled over charcoal (25·8 to 31·6 ug/kg), while the BaP concentrations in the bacon samples were similar to those reported by Lintas *et al.* (1979). Again, the results obtained for the pork samples indicated that those samples produced by natural wood smoke had greater concentrations of total and carcinogenic PAHs than samples processed with liquid smokes.

The concentrations of individual PAHs in smoked beef products ranged from non-detectable for some PAHs to 2·8 μg/kg for Py in breakfast beef. Total PAH concentrations ranged from 5·6 μg/kg for sausage processed with a liquid smoke flavouring to 14·4 μg/kg for naturally-smoked sausage. The combined concentrations of the five carcinogenic compounds ranged from 0·6 μg/kg in liquid

Table 5a. PAH concentrations in selected commercial smoked red meat (pork and beef) products.

	Concentration of PAHs (μg/kg)[a,b]													
Smoked products	Phen	A	F	Py	BaA[c]	Ch	B*b*F[c]	B*k*F	BaP[c]	DBaRA[c]	BghiP	IcdPy[c]	Total PAHs	Total[c] PAHs (carc.)
Pork														
Bacon[c]	0·4	0·2	1·2	1·2	nd	0·5	nd	0·1	0·2	nd	0·3	nd	4·1	0·2
Bacon[d]	1·5	0·9	3·6	2·0	0·2	0·5	nd	nd	0·5	nd	nd	nd	9·2	0·7
Sausage[d]	2·9	0·3	0·8	0·8	0·2	0·3	0·4	0·6	1·8	0·2	0·3	0·2	8·8	2·8
Sausage[d]	8·7	6·4	1·9	4·0	4·0	1·5	0·2	nd	2·3	nd	0·5	0·2	29·7	6·8
Cooked ham[c]	0·5	1·0	0·5	0·3	nd	0·2	nd	0·1	nd	nd	nd	nd	2·6	nd
Grilled pork chops	5·7	1·5	4·6	3·8	1·6	1·5	0·4	5·0	2·5	0·8	0·3	0·1	29·8	7·4
Whole ham[d]	2·1	0·5	4·1	0·9	0·1	nd	nd	nd	1·1	0·5	0·2	nd	9·5	1·7
Sliced ham[d]	0·8	nd	1·6	1·1	0·9	0·3	nd	0·1	0·1	0·5	0·7	nd	6·5	1·5
Beef														
Sausage[c]	0·4	0·3	0·4	1·6	nd	2·0	0·3	0·1	0·2	0·1	nd	nd	5·6	0·6
Sausage[d]	2·0	1·3	1·2	1·9	2·0	1·3	0·1	3·5	1·1	0·1	nd	0·9	14·4	4·2
Breakfast beef[d]	1·2	0·4	0·8	2·8	0·9	nd	1·1	0·1	0·4	nd	0·4	nd	9·7	2·4

[a] Average of duplicate analyses of two samples per commerical processor (four determinations).
[b] Abbreviation description, see table 1.
[c] Liquid smoke flavouring added.
[d] Wood-smoked.
[e] Carcinogenic.
nd: not detected.

Table 5b. Concentrations of UV-detected polycyclic aromatic hydrocarbons in commercial smoked red meat products (pork and beef).

Smoked products	Concentrations of non-carcinogenic PAHs(μg/kg)[a,b]				
	N	APL	AP	FL	Total PAHs
Pork					
Bacon[c]	nd	nd	nd	3	3
Bacon[d]	nd	18	3	5	26
Sausage[d]	nd	35	120	60	195
Sausage	nd	2056	63	1	2120
Cooked ham[c]	nd	nd	9	nd	9
Pork chops	nd	2835	12	7	2854
Whole ham[d]	nd	21	70	1	92
Sliced ham[d]	nd	19	2	1	22
Beef					
Sausage[c]	nd	nd	8	2	10
Sausage[d]	nd	1245	329	282	1656
Breakfast beef[d]	nd	11	15	nd	26

[a] Abbreviation description, see table 1.
[b] Average of duplicate analyses of two samples per commercial processor (four determinations).
[c] Liquid smoke flavouring added.
[d] Wood-smoked.
nd: not detected.

smoked-flavoured sausage to 4·2 μg/kg in wood-smoked sausage. The percent contributions of the carcinogenic compounds to the total PAH concentration were 10·7%, 29·2% and 24·7% for the liquid smoked-flavoured sausage, naturally smoked sausage and breakfast beef, respectively. The results for breakfast beef agree with those of Joe *et al.* (1984) who reported that the carcinogens, B*a*A, B*b*F, B*a*P, and I*c*dPy, were present in more than 50% of the breakfast beef products analysed. Our results also show that sausage processed with liquid smoke had smaller B*a*P concentrations than those in sausage and breakfast beef processed with wood-smoke.

Smoked red meat products were also screened for the presence of N, APL, AP and FL (table 5b). N was not detected in many of the products. However, the concentrations of APL, AP and FL varied from non-detectable in pork and beef samples to a large APL concentration in pork chops (2835 μg/kg). The noncarcinogenic compounds were detected in 55% of the samples with concentrations ranging from 3 to 26 μg/kg, while 44% of the samples had concentrations ranging from 92 to 2854 μg/kg. Again, the results indicate that samples processed with liquid smoke (bacon, ham and sausage) have smaller total non-carcinogenic PAH concentrations than those samples processed with wood smoke (table 5b).

PAH content of smoked fish products. The concentration of PAHs in eighteen fish products including trout, shrimp, herring, salmon, oysters, whitefish and siscos chubs obtained from seven different commercial processing are presented in table 6a. The values are the average of duplicate analyses of two samples per product, except for salmon and oysters, where four samples were analysed in

Table 6b. PAH concentrations in selected commercial smoked fish products.

Compounds[a]	Concentrations of PAHs (μg/kg)						
	Trout[b]	Shrimp[b]	Herring[b]	Salmon[c]	Oysters[c]	Whitefish[b]	Siscos chubs[b]
Phen	2·1	4·0	18·1	23·3	20·8	9·1	3·4
A	0·3	nd	1·7	2·5	4·4	1·0	0·2
F	1·4	1·2	15·8	11·1	15·6	2·4	5·0
Py	5·2	3·6	4·7	24·1	11·0	9·8	6·3
BaA[e]	nd	0·0	3·5	5·3	9·3	8·3	1·3
Ch	0·5	0·0	5·8	3·0	1·8	0·5	3·4
BbF[d]	0·1	nd	0·3	1·8	1·3	0·1	nd
BkF	nd	nd	0·1	3·2	0·1	0·9	5·0
BaP[e]	nd	nd	0·9	3·9	1·0	0·8	nd
DBahA[d]	0·1	0·1	1·7	1·9	0·5	1·1	0·2
BghiP	0·1	0·1	1·7	3·5	1·3	0·4	1·0
IcdPy[d]	nd	0·1	0·9	1·2	0·8	0·8	nd
Total PAHs	11·8	9·3	55·2	86·6	69·9	28·2	25·8
Total carcinogenic PAHs	0·2	0·2	7·3	16·1	14·9	4·1	1·5

[a] Abbreviation description, see table 1.
[b] Average of duplicate analyses of two samples per commercial processor (four determinations).
[c] Average of duplicate analyses of four samples per commercial processor (eight determinations).
[d] Carcinogenic.
nd: not detected.

duplicate. PAHs were found in all the samples, the concentrations ranging from non-detectable for some individual PAHs in trout, shrimp, and siscos chubs, to 24·1 μg/kg for Py in smoked ; salmon. The greatest total PAH concentrations were found in salmon and oysters (86·6 μg/kg and 69·9 μg/kg, respectively), while the smallest concentration was detected in shrimp (9·3 μg/kg). The wide variation in PAH concentrations can be directly related to the smoking conditions (Malanoski *et al.* 1968).

The combined concentrations of the five carcinogenic PAHs ranged from 0·2 μg/kg for trout and shrimp, to 16·1 and 14·9 μg/kg for salmon and oysters, respectively. The smoked oyster data agree with those reported by Fazio (1987), while the PAH concentrations for salmon were somewhat greater. The percentage contributions of the five carcinogenic compounds to the total PAH concentrations in the smoked fish samples were 1·7, 2·2, 13·2, 18·6, 21·3, 14·5 and 5·8% for trout, shrimp, herring, salmon, oyster, whitefish and siscos, respectively.

The results in this survey are generally smaller than those reported by Larsson (1982) who quantitated the PAH contents in Swedish commercially smoked fish as influenced by wood source. Fish smoked with alder had a greater total PAH concentration (439 μg/kg) than samples smoked with spruce/juniper (271 μg/kg). The levels of BaP in the trout and salmon samples are quite similar to those reported by Dunn and Fee (1979). However, they are considerably smaller than those reported for shrimp and oysters. Our results for BaP were also smaller than values cited by Malanoski *et al.* (1968), i.e. 4·3 μg/kg for smoked whitefish and 6·9 μg/kg for smoked whiting. The variation in reported values of PAHs in smoked fish may well be related to the method of smoking. For example, Sreinig (1976) reported that direct smoking of fish produced BaP concentrations greater than

Table 6b. Concentrations of UV-detected polycyclic aromatic hydrocarbons in commercial smoked fish products.

Smoked products	Concentrations of non-carcinogenic PHAs (μg/kg)[a,b]				
	N	APL	AP	FL	Total PAHs
Trout	nd	883	24	nd	137
Shrimp	nd	40	37	11	88
Herring	nd	973	855	210	2038
Salmon	nd	376	677	260	1513
Oysters	nd	10	67	38	115
Whitefish	nd	1280	480	110	1790
Siscos chubs	nd	1800	640	350	2790

[a] Abbreviation description, see table 1.
[b] Average of duplicate analyses of two samples per commercial processor (four determinations).
[c] Liquid smoke flavouring added.
[d] Wood smoked.
nd: not detected.

1 μg/kg. In comparison to indirect smoking where levels were consistently smaller than 1 μg/kg.

Fish products were also screened for the presence of the four non-carcinogenic PAHs, N, APL, AP and FL (table 6b). Naphthalene was not present in any of the samples. However, APL, AP and FL were detected in all the fish samples (with one exception, FL in trout), with APL concentrations as low as 10 μg/kg in oyster, and as high as 1800 μg/kg in siscos chubs. The total non-carcinogenic compounds ranged from 88 μg/kg for shrimp samples to 2790 μg/kg for siscos chubs.

In general, fish products had higher concentrations of non-carcinogenic compounds than the smoked poultry and red meat products.

PAH content of liquid smokes

Eleven liquid smoke flavourings from three different commercial companies and four liquid smoke seasonings purchased at supermarkets, were surveyed for the presence of PAHs (table 7a). Individual PAH concentrations ranged from non-detectable in several samples to as high as 17·9 μg/kg (Py in sample 3 from Company C). With respect to total PAHs, Company A (sample 1) had the smallest quantity (6·3 μg/kg), while Company B (sample 4) had the greatest concentration (43·7 μg/kg). The total concentrations of the five carcinogenic PAHs ranged from 0·3 μg/kg (Company A, sample 1) to 10·2 μg/kg (Company C, sample 2). Liquid smoke seasonings (mesquite and hickory) had not only smaller total PAH concentrations, but also had smaller concentrations of the carcinogenic PAHs compared to the liquid smoke flavourings. There was some variation in total PAH and carcinogenic PAH concentrations in liquid smoke types produced by the same company, e.g. Company A varied from 6·3 to 35·3 μg/kg for total PAHs and from 0·3 to 7·2 μg/kg for total carcinogenic compounds. Smokes from Companies B and C also had a wide range of concentrations.

Liquid smokes vary in PAH profile and content as a consequence of the type of wood used, the procedure used to produce the liquid smokes, and the temperature used (Draudt 1963). Liquid smokes produced by Company A had

Table 7a. Polycyclic aromatic hydrocarbons in selected commercial liquid smoke flavourings and seasonings.

	Concentrations of PAHs (μg/kg)												
	Company A				Company B				Company C				
Compound[a]	1	2	3	4	1	2	3	4	1	2	3	Mesquite[b] seasoning	Hickory[b] seasoning
Phen	1·0	2·9	3·2	4·2	6·9	1·7	2·7	7·6	3·7	4·2	2·7	5·0	2·4
A	1·0	0·9	0·2	2·9	0·7	1·9	0·1	6·8	0·1	0·2	0·1	0·6	0·2
F	0·8	5·8	2·0	4·1	2·1	4·8	1·5	6·4	2·6	2·0	1·1	3·5	1·4
Py	1·3	2·1	4·5	16·7	3·6	5·4	2·1	8·9	5·1	6·7	17·9	3·0	2·8
BaA[c]	nd	1·5	1·5	0·8	1·2	nd	0·8	3·7	0·8	0·2	1·2	0·7	0·3
Ch	0·4	2·2	4·9	0·2	3·0	0·5	4·1	4·0	3·2	3·1	0·2	0·4	0·6
BbF[c]	nd	0·4	1·2	0·1	1·4	1·5	1·1	1·9	1·5	1·3	0·1	0·5	0·2
BkF	0·6	0·8	0·3	1·6	0·5	1·6	0·7	0·5	0·7	0·2	1·1	0·4	0·2
BaP[c]	nd	0·1	1·8	2·1	3·2	7·8	2·9	2·0	0·7	1·4	1·5	0·1	0·1
DBahA[c]	0·3	1·7	2·5	1·3	0·2	1·1	1·1	0·8	1·0	2·0	1·4	nd	nd
BgHP	0·9	1·6	0·6	0·8	0·9	0·4	0·2	0·6	nd	0·4	0·2	0·5	0·1
IcdPy[c]	nd	nd	0·2	0·5	0·3	0·3	0·2	0·5	nd	1·1	1·1	0·5	0·1
Total PAHs	6·2	20·0	22·9	35·3	24·0	21·1	17·7	43·7	19·2	27·0	16·9	14·8	6·4
Total carcinogenic PAHs	0·3	3·7	7·2	4·8	6·3	6·9	6·3	6·9	3·8	10·2	7·3	1·6	0·7

[a] Abbreviation description, see table 1.
[b] Data for liquid smoke seasoning are the average of duplicate analyses of two samples per brand name (four determinations).
[c] Carcinogenic.
Data for Companies A, B and C are average of duplicate analyses of each sample.
nd: not detected.

Table 7b. Concentrations of UV-detected polycyclic aromatic hydrocarbons in selected commerical liquid smoke flavourings and seasonings.

	Concentrations of PAHs (μg/kg)												
	Company A				Company B				Company C				
Compound[a]	1	2	3	4	1	2	3	4	1	2	3	Mesquite[b] seasoning	Hickory[b] seasoning
N	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
APL	17	290	1900	602	1384	1029	168	3214	2100	2043	271	150	73
AP	7	142	140	922	404	50	50	2018	126	177	2921	330	115
TL	nd	62	8	67	4	5	3	138	2	8	8	5	2
Total PAHs	24	599	2048	5380	1792	1384	221	6526	2228	2678	3279	337	190

[a]Abbreviation description, see Table 1.
[b]Data for liquid smoke seasonings are the average of duplicate analyses of two samples per brand name (four determinations).
Data for Companies A, B and C are the average of duplicate analyses of each sample.
nd: not detected.

average total and carcinogenic PAH concentrations of 21·1 and 4·0 μg/kg, respectively. Corresponding values for liquid smokes produced by Company B were 28·2 and 7·4 μg/kg, respectively, while those from Company C had total and carcinogenic PAH concentrations of 27·7 and 7·1 μg/kg, respectively. Mesquite seasoning had approximately double the total and carcinogenic PAH concentrations of hickory seasoning. B*a*P contents were much smaller than those reported by Potthast (1979) for 12 commercial liquid smoke products, the latter concentrations ranging from less than 0·05 μg/l to 2600 μg/l. Heuning (1976) detected up to 20 μg/kg of B*a*P in smoke-flavoured seasonings, while White *et al.* (1971) reported trance amounts of some three and four-ring PAH compounds in three of seven commercial liquid smokes. Heuning (1976) also reported that the maximum dose of liquid smoke seasoning used for smoking food products was 5 g/kg meat product and for that reason, the PAH content of food products processed with liquid smoke flavourings or seasonings was below the established German standard of 1 μg/kg.

The concentrations of the four non-carcinogenic PAHs (N, APL, AP and FL) in the liquid smokes are presented in table 7b. APL, AP and FL were present in almost all the samples with concentrations as low as 2 μg/kg for FL (Company C, sample 1) and as high as 3224 μg/kg for APL (Company B, sample 4). The total noncarcinogenic concentrations ranged from 24 μg/kg (Company A, sample 1) to 6526 μg/kg (Company B, sample 4). Potthast (1979) identified 25 PAH compounds in smoke condensates and the major PAH in smoke was Phen and closely related compounds. Maga (1986) compared the PAH composition of mesquite and hickory wood smokes and reported that the former had 31 compounds (1250 mg/kg) while hickory smoke had 22 compounds (688 mg/kg).

Conclusions

Results of this limited survey revealed the presence of PAHs in all of the smoked foods analysed. Smoked meat products (turkey, chicken, pork and beef) processed with liquid smoke flavourings had smaller concentrations of total PAHs and total carcinogenic PAHs compared to those processed with natural wood smoke. Carcinogenic PAHs were not detected in 10% of the samples, 24% had less than 1 μg/kg, while the remainder of the samples had concentrations greater than 1 μg/kg. With respect to B*a*P, 31% of the samples had no detectable quantities, while 42% of the samples contained less than 1 μg/kg. On the other hand, the remaining samples, oyster, salmon, pork chops, sausage, pork sausage and barbecued wings, had concentrations of 3·9, 2·9, 2·5, 2·3 and 1·8 μg/kg, respectively. These results concur with those of Malanoski *et al.* (1968) who reported B*a*P concentrations of less than 1 μg/kg in 21 samples out of 60 assorted foodstuffs analysed. Eleven out of 60 samples had B*a*P concentrations up to 7 μg/kg, including barbecued pork (4·5 μg/kg), barbecued beef (2·5 μ/kg), smoked whitefish (4·3 μg/kg), and smoked whiting (6·9 μg/kg). Toth and Blaas (1972a) also reported that the total quantity of potentially harmful PAHs in smoked meat was five to ten times the amount of B*a*P detected. With some exceptions, our values for total carcinogen PAHs smoked meat were 1·4 to 14·5 times the amount of B*a*P detected. However, the total carcinogenic PAH concentrations were based on the sum of the concentrations of only five carcinogenic PAHs. The PAH data of Howard *et al.* (1966), Toth and Blaas (1972b) and Joe *et al.* (1984) revealed the

presence of F and Py in at least 95% of the samples analysed, while the carcinogens, BaA, BbF, BaP, DBahA and IcdPy, were found in 30–87% of the samples tested.

It is apparent from this limited survey that major differences in PAH concentrations exist between smoked products and these may be ascribed to the many variables involved in the smoking process, including the type of generator, temperature of combustion, degree of smoking, time of preparation, and fat content of the products (Draudt 1963). Furthermore, it should be pointed out that many PAHs are not classified as carcinogenic, simply because of lack of adequate toxicological data.

In conclusion, results of this survey generally indicate similar values to those published by Fazio (1987). However, because of the carcinogenic nature of PAHs, liquid smokes should be further screened for the presence of these compounds as should be meat products processed with these smokes.

Acknowledgements

This study was supported in part by a grant from the United State Department of Agriculture-Agricultural Research Service and by the Michigan Agricultural Experiment Station.

References

Black, J. M., Denenski, P. P., and Zaessle, W. F., 1979, Routine liquid chromatographic method for assessing polynuclear aromatic hydrocarbon pollution in fresh water environments. *Bulletin of Environmental Contamination and Toxicology*, 22, 278–284.

Doerjbe, M. F., Harmon, G. E., and Pratt, D. E., 1979, 3,4-Benzopyrene in charcoal grilled meats. *Journal of Food Science*, 44, 622–623.

Draudt, H. N., 1963, The meat smoking process: a review. *Food Technology*, 17, 85–90.

Duns, B. P., and Fee, J., 1979, Polycyclic aromatic hydrocarbon carcinogens in commercial seafoods. *Canadian Journal of Fishery and Aquatic Science*, 36, 1469.

Fazio, T., 1987, *Food Additives Analytical Manual* edited by J. Sherma (Arlington, VA: Association of Official Analytical Chemists), Vol. II, pp. 222–235.

Fazio, T., and Howard, J. W., 1983, Polycyclic aromatic hydrocarbons in foods. *Handbook of Polycyclic Aromatic Hydrocarbons*, edited by A. Bjorseth (New York: M. Dekker), pp. 461–505.

Frenheim, K., 1986, Carcinogenic polycyclic aromatic hydrocarbons in Norwegian smoked meat sausages. *Journal of Agricultural and Food Chemistry*, 34, 916–919.

Haenni, E. O., and Tschbach, H., 1974, Trace polynuclear aromatic hydrocarbons analysis. *The Contribution of Chemistry to Food Supplies*. IUPAC–IUFOST Symposium (London: Butterworths), p. 219–225.

Henning, W., 1976, How dangerous are smoke-flavored seasonings? *Ernaehrungswirtschaft*, 3, 108–111.

Howard, J. W., and Fazio, T., 1980, Review of polycyclic aromatic hydrocarbons in foods. Analytical methodology and reported findings of polycyclic aromatic hydrocarbons in foods. *Journal of the Association of Official Analytical Chemists*, 63, 1077–1104.

Howard, J. W., White, R. H., Fry, B. E., and Tlrichi, E. W., 1966, Extraction and estimation of polycyclic aromatic hydrocarbons in smoked foods. IV. Benz[a]pyrene. *Journal of the Association of Official Analytical Chemists*, 49, 611–617.

IARC, 1983, *Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans. Polynuclear Aromatic Compounds. Part 1. Chemicals, Environmental and Experimental Data, Vol. 32* (Lyon: IARC).

IARC, 1987, *Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans. Overall Evaluation of Carcinogenicity: An Updating of IARC Monographs Volumes 1 to 42, Supplement 7* (Lyon: IARC).

Insco, N. N., 1964, Photochemical changes in thin layer chromatograms of polycyclic aromatic hydrocarbons. *Analytical Chemistry*, 36, 2505–2506.

Jhr, F. L. Jr., Salemme, J., and Fazio, T., 1984, Liquid chromatographic determination of trace residues of polynuclear aromatic hydrocarbons in smoked foods. *Journal of the Association of Official Analytical Chemists*, 67, 1076–1082.

KRAMERS, P. G. N., and VAN DER HELJDEN, C. A., 1988, Polycyclic aromatic hydrocarbons (PAH): carcinogenicity data and risk extrapolations. *Toxicological and Environmental Chemistry*, **16**, 341–351.

KESHWARA, S. C., CLARKSON, S. G., and MEISKARI, K. A., 1985, Polycyclic aromatic hydrocarbons in barbecue briquets. *Journal of Food Safety*, **7**, 177–180.

LARSSON, B. K., 1982, Polycyclic aromatic hydrocarbons in smoked fish. *Zeitschrift für Lebensmittel-Untersuchung und-Forschung*, **174**, 101–104.

LAWRENCE, J. F., and DOMAS, F. W., 1984, Determination of polycyclic aromatic hydrocarbons in some Canadian commercial fish, shellfish, and meat products by liquid chromatography with confirmation by capillary gas chromatography–mass spectrometry. *Journal of Agricultural and Food Chemistry*, **32**, 789–794.

LEE, M. L., NOVOTNY, M. V., and BARTLE, K. D., 1981, *Analytical Chemistry of Polycyclic Aromatic Compounds* (New York: Academic Press), pp. 443–449.

LIJINSKY, W., and ROSS, A. E., 1967, Production of carcinogenic polynuclear hydrocarbons in the cooking of food. *Food and Cosmetics Toxicology*, **5**, 343–347.

LINTAS, C., DE MATTHAEIS, M. C., and MERLI, F., 1979, Determination of benzo[*a*]pyrene in smoked, cooked and roasted food products. *Food and Cosmetics Toxicology*, **17**, 125–128.

LO, M.-T., and SANDI, E., 1978, Polycyclic aromatic hydrocarbons (polynuclears) in foods. *Residue Reviews*, **69**, 35–86.

MAGA, J. A., 1986, Polycyclic aromatic hydrocarbon (PAH) composition of mesquite (*Prosopis juliflora*) smoke and grilled beef. *Journal of Agricultural and Food Chemistry*, **34**, 249–251.

MAGA, J. A., 1988, *Smoke in Food Processing. Potential Health Concerns Associated with Smoke*, (Boca Raton, Florida: CRC Press, Inc.).

MALANOSKI, A. J., GIFFIELD, F. L., BARNES, C. J., WORTHINGTON, J. W., and JOE, F. L., 1968, Survey of polycyclic aromatic hydrocarbons in smoked foods. *Journal of the Association of Official Analytical Chemists*, **51**, 174–181.

NIOSH, 1985, *Manual of Analytical Methods U.S. Dep. HHS*, NIOSH, Cincinnati, USA, Suppl., 3rd edn, Methods Nos. 5506 and 5515; Polynuclear Aromatic Hydrocarbons. DHHS (NIOSH) Publ. No. 84–100.

POTTHAST, K., 1979, The influence of smoking technology on the composition of polycyclic hydrocarbons in smoked meat products, smoke condensates and in waste gases from smoking plants. *Fleischwirtschaft*, **59**, 1515–1521.

SCHMÄHL, D., HOWARD, P., and BASU, D., 1981, Health and ecological assessment of polynuclear aromatic hydrocarbons. *Journal of Environmental Pathology and Toxicology*, **5**, 1–350.

SAMAKI, E., MEISKER, J. L., and MORGAN, M. J., 1965, The quantitative composition of air pollution source effluents in terms of azaheterocyclic compounds and polynuclear aromatic hydrocarbons. *International Journal of Air and Water Pollution*, **9**, 291–298.

STEINKE, J., 1976, Benzo[*a*]pyrene contents of smoked fish in relation to smoking procedure. *Zeitschrift für Lebensmittel-Untersuchung und-Forschung*, **162**, 235–239.

THORSTEINSSON, T., and THORDARSON, G., 1968, Polycyclic hydrocarbons in singed food in Iceland. *Cancer*, **21**, 190–192.

TILGNER, D. J., 1977, The phenomena of quality in the smoke curing process. *Advances in Smoking of Foods* IUPAC–IUFOST Symposia, pp. 1629–1638.

TÓTH, L., and BLAAS, W., 1972a, The effect of smoking technology on the content of carcinogenic hydrocarbons in smoked meat products. I. Effect of various smoking methods. *Fleischwirtschaft*, **52**, 1121–1125.

TÓTH, L., and BLAAS, W., 1972b, The effect of smoking technology on the content of carcinogenic hydrocarbons in smoked meat products. II. Effect of the temperature at which the wood smolders and cooling, washing and filtration of the smoke. *Fleischwirtschaft*, **52**, 1419–1423.

TÓTH, L., and POTTHAST, K., 1984, Chemical aspects of the smoking of meat and meat products. *Advances in Food Research*, **29**, 87–158.

RHEE, K. S., and BRATZLER, L. J., 1970, Benzo[*a*]pyrene in smoked meat products. *Journal of Food Science*, **35**, 146–149.

VAESSEN, H. A. M. G., JEKEL, A. A., and WILPERS, A. A. M. M., 1988, Dietary intake of polycyclic aromatic hydrocarbons. *Toxicological and Environmental Chemistry*, **16**, 281–294.

WHITE, R. H., HOWARD, L. W., and BARNES, C. J., 1971, Determination of polycyclic aromatic hydrocarbons in liquid smoke flavors. *Journal of Agricultural and Food Chemistry*, **19**, 143–146.

ZEDECK, M. S., 1980, Polycyclic aromatic hydrocarbons—a review. *Journal of Environmental Pathology and Toxicology*, **3**, 537–567.