

A Review of Polycyclic Aromatic Hydrocarbons in Foods

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During the last 10 years various workers have demonstrated the widespread occurrence of polycyclic aromatic hydrocarbons in the human environment. The Food and Drug Administration has been actively engaged in the development of analytical methods and their application to foodstuffs. The results of these studies indicate that various smoked products, edible vegetable oils, total diet samples, and related products of food additive significance contain trace quantities of polycyclic hydrocarbons—for example, benzo(*a*)-

pyrene and other hydrocarbons were isolated from smoked ham and smoked fish samples at part per billion levels and were also found in some vegetable oils and total diet samples. In cooperation with the U. S. Department of Agriculture, 60 foodstuffs and related products were analyzed. Levels of benzo(*a*)pyrene found in 32 products ranged up to 7.0 p.p.b. The methodology for and occurrence of such hydrocarbons in food are reviewed.

The occurrence, isolation, and identification of polycyclic aromatic hydrocarbons in the human environment have been discussed in a comprehensive survey by Gunther and Buzzetti (1965). These authors pointed out the importance of both the qualitative and quantitative aspects of their presence as residues in the total environment, since some of these compounds possess carcinogenic activity. Thus, strict surveillance is needed of their occurrence in foodstuffs and in other materials to which humans are exposed. Haenni (1968) has reviewed in detail the development of analytical control of such potential polycyclic hydrocarbon contaminants in foods and food additives. As suggested by him, it is very probable that only a small fraction of the potential compounds in this class have been recognized, identified, and toxicologically evaluated, primarily because of complex structures, the small quantities present, and the fact that analytical methods of adequate specificity and sensitivity have only recently been developed.

One of the major problems confronting the analyst has been the development of adequate separative techniques for the identification and determination of individual polycyclic hydrocarbons. As pointed out by Haenni (1968), it is the exception rather than the rule to find only one or two of these compounds occurring together. More often they occur as a minute fraction of a complete mixture of hydrocarbons, all usually closely related in physical and chemical character. Probably one of the most significant difficulties encountered in past studies is the resolution of the so-called "benzopyrene fraction" consisting of the carcinogen, benzo(*a*)pyrene, its isomer, benzo(*e*)pyrene, benzo(*k*)fluoranthene, and perylene. Such separations are of paramount importance to those engaged in regulatory work where the analyst must be able to separate and unequivocally identify the carcinogenic hydrocarbons as distinguished from the many noncarcinogenic types that have been demonstrated to be present in foodstuffs and related products of food additive significance.

Following its earlier studies on food additives the Food and Drug Administration (FDA) several years ago initiated studies on the development and application of analytical methods for polycyclic hydrocarbons in

various foodstuffs. At the 1965 annual meeting of the Association of Official Analytical Chemists (AOAC) two procedures applicable to smoked foods were described: a general method for polycyclic aromatic hydrocarbons (Howard *et al.*, 1966a) and a specific procedure for benzo(*a*)pyrene (Howard *et al.*, 1966c). Both methods were demonstrated to be sensitive to 2 p.p.b. Very briefly, they consist of an initial extraction of the hydrocarbons, followed by a partition step between dimethyl sulfoxide and an aliphatic solvent. Column chromatography on pretreated Florisil followed by paper and thin-layer separation is then used to reduce interfering background material and isolate the polycyclic compound(s). Depending on the type of product under analysis and the specific hydrocarbons to be isolated, it may be necessary to employ all of these techniques to obtain satisfactory ultraviolet and spectrophotofluorometric spectra for characterization and estimation of the compounds present. If only benzo(*a*)pyrene is to be determined, the procedure may be shortened considerably by using thin-layer chromatography on cellulose acetate directly without the lengthy paper chromatographic technique. The cellulose acetate TLC procedure is effective in the separation of the components of the aforementioned "benzopyrene fraction," whereas most other paper and thin-layer chromatographic techniques fail in this respect.

This project coincided with that of the International Union of Pure and Applied Chemistry's (IUPAC) Subcommittee on Smoke Constituents, Trace Substances Commission, which also wished to evaluate the procedures. Because of this mutual interest, the decision was made to undertake a combined collaborative study by the AOAC and IUPAC, in the hope that it would provide the broad based support to warrant adoption of the AOAC procedures by IUPAC at least as referee methods. It was the consensus of opinion of members of the IUPAC Food Section that the benzo(*a*)pyrene method should be studied first, since it was felt that the more lengthy and complicated multicomponent procedure for specific hydrocarbons would not be practical for the initial phases of the program, and also because benzo(*a*)pyrene is the most commonly occurring carcinogen of this type. The collaborative results obtained by 11 laboratories on nine samples (smoked fish and smoked ham) fortified at levels of 4 and 10 p.p.b. of benzo(*a*)pyrene were in good agreement and

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the method was adopted as official, first action, at the 1967 AOAC meeting (Howard *et al.*, 1968b). The Trace Substances Commission recommended adoption of the method by IUPAC as a referee procedure. Since publication of the aforementioned general method, it has been found that the procedure can be considerably shortened and it is anticipated that a collaborative study will be initiated in the near future.

The above procedures have been applied to the analysis of various foods. This paper reviews the results of these studies and other relevant published data.

SOURCES AND OCCURRENCE

The possible sources of polycyclic aromatic hydrocarbon contamination of foodstuffs in the modern human environment are numerous and varied. A review of the literature reveals that curing smokes, contaminated soils, polluted air and water, modes of cooking, food additives, food processing, and endogenous sources have been considered. However, many of the investigations were conducted at random and the origin of the contaminants and extent of contamination have not been fully established or evaluated. Aside from smoke-cured foods, charcoal-broiled meats, and possibly environmental pollution, Haenni (1968) has stated that the most common sources of polycyclics as potential food contaminants are food additives of petroleum origin. Thus, analytical controls have been adopted to exclude the presence of these compounds in petroleum products of food additive significance. The complex operations involved in the collection, processing, and packaging of foods often render the determination of the contaminating source(s) extremely difficult.

SMOKED FOODS

Analyses of curing smoke and smoked foods for polycyclic aromatic hydrocarbons has received considerable attention during the last 10 years, particularly in countries outside the United States where such products constitute a major portion of the diet. These investigations were prompted by the hypothesis that a correlation might exist between a high incidence of stomach cancer in some population groups (Icelanders and Baltic fishermen) and the presence of polycyclic compounds in smoked foodstuffs. Much of the research on curing smoke has been concerned with attempts to control the polycyclic content and thus minimize the contamination. This latter area has been reviewed by Draudt (1963), Sikorski and Tilgner (1964), and Haenni (1968) and is not discussed in detail here.

Numerous studies on the benzo(*a*)pyrene content of smoked foods have been conducted by Russian and Polish investigators, but details of the procedures employed and recovery data are not readily available in the United States. As shown in Table I, varying levels of benzo(*a*)pyrene have been reported in smoked fish, sausage, and mutton. The differences in hydrocarbon content may be ascribed to the many variables involved in the smoking process, such as the type of generator, temperature of combustion, degree of smoking, etc. (Draudt, 1963). In regard to the surface deposition of the smoke constituents, various investigations have shown that the polycyclic compounds will migrate into the interior of the food, the extent of migration depend-

Table I. Levels of Benzo(*a*)pyrene Found in Smoked Foods Processed Outside United States

| Food Product | Levels, P.P.B. | References |
|------------------|----------------|--|
| Sausage and fish | 1.7-10.5 | Dobes <i>et al.</i> (1954) |
| Fish and mutton | 0.3- 2.1 | Bailey and Dungal (1958) |
| Fish | 1.7-53.0 | Tilgner and Miler (1963) Wojtelowicz <i>et al.</i> (1957) |
| Fish | 7.0 | Gorelova (1963) |
| Sausage and fish | 0.1- 1.4 | Gorelova and Dikun (1965) |
| Fish | 4.2-60.0 | Petrun and Rubenchik (1966) |

ing upon the character of the product and its storage time (Gorelova *et al.*, 1960; Tilgner, 1958).

Prior to 1966, little information was available on the polycyclic hydrocarbon content of foods smoked in North America. Genest and Smith (1964) developed a detection method for benzo(*a*)pyrene with a reported sensitivity of 10 to 50 p.p.b., depending on the food product analyzed; smoked fish, frankfurters, and cheese were investigated, but benzo(*a*)pyrene was not detected. Lijinsky and Shubik (1965b) reported the presence of benzo(*a*)pyrene and other polycyclic hydrocarbons in two samples of smoked haddock and salmon; levels of benzo(*a*)pyrene were 0.3 and 1.0 p.p.b., respectively. These investigators also analyzed two liquid smoke flavors, but did not find benzo(*a*)pyrene. On the basis of these results it was concluded that even the minute amount of benzo(*a*)pyrene in smoked fish would be avoided by the use of liquid smoke rather than direct smoking.

The Food and Drug Administration has also been actively engaged in the development of analytical methods and their application for the determination of polycyclic hydrocarbons in foods (Howard *et al.*, 1966a,c). As shown in Tables II and III, various foods (including both smoked and unsmoked products) were analyzed for polycyclic hydrocarbons by the aforementioned general method. Benzo(*a*)pyrene was found in smoked sturgeon, dried smoked herring, and smoked ham samples at levels of 0.8, 1.0, and 3.2 p.p.b. respectively. Its identification was confirmed by three techniques: ultraviolet and fluorescence spectrophotometry, and molecular weight (*m/e* 252) as determined by mass spectrometry. Pyrene and fluoranthene were present in all of the foods examined; however, no plausible explanation of their origin can be offered at the present time.

As a result of these preliminary findings the FDA recommended further studies to accumulate the analytical data necessary for proper evaluation of the existing situation. Accordingly, a cooperative program between the United States Department of Agriculture (U.S.D.A. Meat Inspection Division) and FDA was initiated. Sixty assorted foodstuffs and related materials were analyzed for benzo(*a*)pyrene by the two laboratories (Malanoski *et al.*, 1968). Thirty-two of these products contained the hydrocarbon. Levels reported for 21 samples were below 1 p.p.b.; higher amounts were isolated from the remaining 11 samples (Table IV). The levels of benzo(*a*)pyrene do not exceed 7.0 p.p.b., well below some of the values reported in the literature for European smoked fish. As pointed out by Draudt (1963),

Table II. Polycyclic Aromatic Hydrocarbons Found in Smoked Food Products (P.P.B.)

| Food Product | Benz(a)-anthracene | Benzo(a)-pyrene | Benzo(e)-pyrene | Benzo(g,h,i)-perylene | Fluoranthene | Pyrene | 4-Methylpyrene |
|-----------------|--------------------|-----------------|-----------------|-----------------------|--------------|--------|----------------|
| Beef, chipped | 0.4 | | | | 0.6 | 0.5 | |
| Cheese, gouda | | | | | 2.8 | 2.6 | |
| Fish | | | | | | | |
| Herring | | | | | 3.0 | 2.2 | |
| Herring (dried) | 1.7 | 1.0 | 1.2 | 1.0 | 1.8 | 1.8 | |
| Salmon | 0.5 | | 0.4 | | 3.2 | 2.0 | |
| Sturgeon | | 0.8 | | | 2.4 | 4.4 | |
| White | | | | | 4.6 | 4.0 | |
| Frankfurters | | | | | 6.4 | 3.8 | |
| Ham | 2.8 | 3.2 | 1.2 | 1.4 | 14.0 | 11.2 | 2.0 |
| Pork roll | | | | | 3.1 | 2.5 | |

Table III. Polycyclic Aromatic Hydrocarbons in Unsmoked Food Products

| Food Product | Fluoranthene, P.P.B. | Pyrene, P.P.B. |
|------------------------|----------------------|----------------|
| Cheese, cheddar | 0.8 | 0.7 |
| Fish, haddock | 1.6 | 0.8 |
| Fish, herring (salted) | 0.8 | 1.0 |
| Fish, salmon (canned) | 1.8 | 1.4 |

Table V. Levels of Benzo(a)pyrene in Vegetable Oils

| Vegetable Oil | No. of Oils Examined | No. of Oils Found | Range, P.P.B. | Av. P.P.B. |
|---------------|----------------------|-------------------|---------------|------------|
| Soybean | 9 | 2 | 1.2-1.5 | 1.4 |
| Cottonseed | 6 | 2 | 0.4-0.4 | 0.4 |
| Corn | 8 | 7 | 0.4-1.0 | 0.7 |
| Olive | 5 | 3 | 0.4-0.5 | 0.5 |
| Peanut | 1 | 1 | — | 0.6 |

lightly smoked products are common in the United States in contrast to some countries in which foods are heavily smoked for preservative purposes. For example, Dungal (1959) in his study of Icelandic smoked food-stuffs warned against prolonged smoking exposure. He concluded that foods smoked only briefly are probably not hazardous, but products exposed for weeks or months, as is frequently the case with lamb and mutton in Iceland, may contain dangerous quantities of carcinogenic hydrocarbons.

VEGETABLE OILS

Various European workers have reported the presence of polycyclic contaminants in edible vegetable oils. Jung and Morand (1962, 1963, 1964) reported the detection of pyrene, benzo(e)pyrene, and benzo(a)pyrene in different oils. Ciusa *et al.* (1965) found a number of polycyclic hydrocarbons including phenanthrene, pyrene, fluoranthene, benz(a)anthracene, chrysene, and perylene in pressed and refined olive oils, with higher quantities of the hydrocarbons being isolated from refined products. No benzo(a)pyrene, however, was detected. Concurrently, studies were initiated to

develop a quantitative method for application to refined oils processed in the United States. The results of the investigation were reported at the 1966 AOAC meeting (Howard *et al.*, 1966b). With the exception of safflower oil (only three samples), trace quantities of polycyclic hydrocarbons, including benzo(a)pyrene, were found in at least one of each type of oils analyzed: soybean, cottonseed, corn, olive, and peanut. As shown in Table V, the highest content (1.5 p.p.b.) of benzo(a)pyrene was found in soybean oil; however, only two of the nine samples analyzed contained it. In the corn oil products the compound was isolated from seven of the eight samples investigated. Other hydrocarbons found included benzo(g,h,i)perylene, benz(a)anthracene, benzo(e)pyrene, pyrene, fluoranthene, and benzo(k)fluoranthene. In subsequent studies attempts were made to establish the source of contamination. Fifteen commercial hexanes used in the solvent extraction of edible oils were analyzed on the thesis that they contribute polycyclic residues (Howard *et al.*, 1968a). Although nine samples were found to contain polycyclics, no carcinogenic types were detected. However, analysis of several crude oil samples which had not been subjected to solvent extraction revealed the presence of benzo(a)pyrene. These studies would appear to indicate that such contamination occurs in the initial processing operation (expeller, etc.) or was present in the starting material, as suggested by several reports in the literature. Haenni (1968) has suggested that environmental contamination of agricultural products or endogenous existence should be considered possible contributing factors. The latter possibility has also been postulated by Graf and Diehl (1966), whereas Guddal (1959) claims that some polycyclic compounds are actually resorbed by plants grown in contaminated soils.

The results of the above studies with respect to the presence of polycyclic compounds in refined and crude oils have been confirmed by German investigators (Biernoth and Rost, 1967; Grimmer and Hildebrandt, 1967). Grimmer and Hildebrandt (1967) also con-

Table IV. Food Products Found to Contain Highest Levels of Benzo(a)pyrene in FDA-USDA Cooperative Program

| Food Product | Av. P.P.B. |
|-----------------------|------------|
| Smoked hot sausage | 0.8 |
| Smoked ham | 1.0 |
| | 1.2 |
| Smoked turkey fat | 2.1 |
| Barbecued pork | 4.5 |
| Barbecued beef | 2.5 |
| Smoked whitefish | 4.3 |
| Smoked whiting | 6.9 |
| Smoked chubs | 1.3 |
| Smoked cod (kippered) | 4.5 |
| Cooking oil (re-used) | 1.4 |

cluded that hexane solvents were not the source of contamination.

TOTAL DIET SAMPLES

In addition to the studies on smoked foods and vegetable oils, the FDA has continued its methods development program in applications to other dietary products—"total diet samples." These samples, described in detail previously (Cummings, 1966; Duggan *et al.*, 1966) have been used successfully by FDA since 1961 to monitor radioactive contaminants (Laug *et al.*, 1963), pesticide residues (Cummings, 1965, 1966; Duggan *et al.*, 1966; Duggan and Johnson, 1967; Mills, 1963), and nutrient content (Deutsch *et al.*, 1963) in our food supply. Very briefly, such surveillance is carried out with established methods for the analysis of sample categories comprised of combinations of similar food items. The great variations in the types and number of foods, their patterns of use, environmental factors, and permissible levels make it impractical to provide adequate control through a product-by-product analysis (Haenni, 1968).

Multidetector procedures have been developed for the determination of polycyclic hydrocarbons in root vegetables, dairy products, oils, fats, and shortenings, meat, fish, and poultry, and beverages (Howard *et al.*, 1968c), but have not as yet been validated by collaborative study. The following compounds were found in shortening composites at levels below 0.5 p.p.b.: benzo(*a*)pyrene, benzo(*k*)fluoranthene, benzo(*b*)fluoranthene, benzo(*e*)pyrene, benzo(*g,h,i*)perylene, benzo(*a*)anthracene, pyrene, fluoranthene, and phenanthrene. Pyrene and fluoranthene were the only hydrocarbons found in the other composites listed above.

FRUITS AND PLANTS

At present there is little information on the extent of contamination of our food supply via air pollutants. Gunther *et al.* (1967) reported the presence of about 25 p.p.m. of anthracene and six unidentified polycyclics in the rinds of oranges grown in atmospheric-polluted areas, but not in fruit harvested in uncontaminated locations. On the basis of these findings they emphasized the need to investigate the polycyclic content of food plants, particularly those grown in areas of heavy air pollution, industrialization, and extensive petroleum oil spraying. Bolling (1964) indicated that wheat, corn, oats, and barley grown in industrial surroundings contained higher levels of contaminants than crops from more remote areas. Graf and Diehl (1966) isolated eight polycyclic hydrocarbons from various plant leaves, including benzo(*a*)pyrene at levels of 8 to 40 p.p.b. Lower levels were found in fruit and grains. These authors concluded that the hydrocarbons were endogenous.

The possible migration of polycyclic hydrocarbons from contaminated soils into plants has also received some attention but remains incompletely resolved. Benzo(*a*)pyrene (levels ranging from 40 to 1300 p.p.b.) and numerous other hydrocarbons have been found in soil (Blumer, 1961) and postulated as derived from the pyrolysis of wood or alternatively as possible products of organisms which contribute their organic matter to the soils. The latter concept, including subsequent translocation to plants, has been proposed by Mallet

(1964), who found 10 to 14 p.p.b. of benzo(*a*)pyrene in tree leaves and 43 p.p.b. in decaying organic matter under the trees. Further evidence for the occurrence of this mechanism was reported by Doerr (1965), who showed that barley roots in soil or water cultures absorbed benzo(*a*)pyrene with subsequent transference to the shoots. Guddal (1959) also claims that various hydrocarbons are resorbed by plants grown in contaminated soils.

SEAFOODS

The uptake of polycyclic hydrocarbons by marine life in polluted waters has been investigated. Cahnmann and Kuratsune (1957) isolated estimated levels of 2 to 6 p.p.b. of benzo(*a*)pyrene and other polycyclic compounds (total level about 200 p.p.b.) from oysters collected from an area moderately contaminated with petroleum oils. These authors assumed that the quantities of hydrocarbons in oysters vary with their habitat. This assumption was based on previous findings of other investigators (Koe, 1951; Koe and Zechmeister, 1952; Shimkin *et al.*, 1951; Zechmeister and Koe, 1952) who showed variations in the levels of compounds as well as composition of the hydrocarbon mixture, depending upon the environment. In more recent studies Mallet (1964) and Mallet and Priou (1967) reported benzo(*a*)pyrene in marine sediments approximately 500 years old as well as progressive contamination of mollusks, worms, algae, etc., with benzo(*a*)pyrene derivatives of unknown origin. Saccini-Cicatelli (1965, 1966) observed that worms placed in benzo(*a*)pyrene-contaminated water took up as much as 88.2 p.p.m. of the hydrocarbon and retained up to 350 p.p.b. when placed in pure water.

COOKING OF FOODS

The mode of cooking as a possible source of contamination of foodstuffs is also worthy of comment here. For example, benzo(*a*)pyrene has been found in charred crusts from biscuits (Kuratsune, 1956) and in some samples of commercial roasted coffees (Kuratsune and Hueper, 1962). Since most foods contain polysaccharides, Davies and Wilmshurst (1960) studied the pyrolytic behavior of starch in the absence of air and at atmospheric pressure, with the following results: At 370° to 390° C. benzo(*a*)pyrene was present at a level of 0.7 p.p.b. in the distillation residue; at 650° C. 17 p.p.b.

Somewhat similar studies were carried out by Masuda *et al.* (1967) on carbohydrates, amino acids, and fatty acids. No polycyclic compounds were found at 300° C.; however, 19 hydrocarbons including benzo(*a*)pyrene were isolated from the above materials at 500° and 700° C. At 500° C., more polycyclics were formed from the carbohydrates, whereas at 700° C. greater yields were derived from the fatty acids. Many cooking processes utilize the 370° to 390° C. range, whereas the surface temperatures of baking bread may approach 400° C. and boiling cooking fats, 400° to 600° C.

The formation of polycyclic hydrocarbons in meat as a result of cooking on a grill was reported by Seppilli and Scassellati-Sforzolini (1963) in 1963 and by Lijinsky and Shubik (1965a) in 1965. In the latter study benzo(*a*)pyrene was found at levels of 5 to 8 p.p.b. in charcoal-broiled steaks and 10.5 p.p.b. in barbecued ribs. Various other polycyclic compounds

were also identified. The authors concluded that the fat or other carbon-hydrogen-oxygen-containing compounds in the meat were the probable pyrolysis sources of the hydrocarbons.

The effect of variations in methods of cooking on the content of benzo(a)pyrene and other polycyclic hydrocarbons in meat was discussed by Lijinsky and Ross (1967). In this study, the authors confirmed that the production of polycyclic hydrocarbons in charcoal broiling is dependent on the fat content of the food. The closeness to the source of heat is also an important factor. For example, levels of benzo(a)pyrene as high as 50 p.p.b. were found in thick T-bone steaks which were cooked close to the coals for long periods, whereas concentrations were markedly reduced in samples prepared (to the same end point) at a greater distance from the heat source. The authors concluded that if the production of carcinogens, such as benzo(a)pyrene, in cooking is to be minimized, the method of preparation should avoid contact of the food with cooking flames, cook for a longer period at lower temperatures, and use meat with a minimum of fat.

CONCLUSIONS

The results of the studies cited in this review demonstrate the occurrence of trace quantities of benzo(a)pyrene and other polycyclic hydrocarbons in a wide variety of foodstuffs and related products. Many of the investigations have been conducted at random and in some cases the analytical procedures have not been validated. Obviously, there is need for further research to develop data on the extent of polycyclic hydrocarbon contamination in foods and more firmly establish the sources of such pollution. During the last few years data have been accumulated with respect to smoked foods; however, this is in contrast to other sources, such as the effects of contaminated soil, water, and air. As pointed out (Gunther and Buzzetti, 1965; Lijinsky and Shubik, 1965a), it is of major importance to develop accurate data with respect to the many facets of our total environment, so that the over-all public health hazard may be effectively evaluated and controlled.

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