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Polynuclear Aromatic Hydrocarbons in the Water Environment *

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Many polynuclear aromatic hydrocarbons (PAH) are known to be carcinogenic to animals and probably to man. This review is concerned with carcinogenic and non-carcinogenic PAH in the water environment, with emphasis on 3,4-benzpyrene (BP) because it is ubiquitous, is one of the most potent of the carcinogenic PAH and has been widely studied. Although PAH are formed in combustion and other high-temperature processes, there is also evidence for their endogenous formation in plants, which may explain their ubiquity therein. Although the solubility of these compounds in pure water is very low, they may be solubilized by such materials as detergents, or they may otherwise occur in aqueous solution associated with or adsorbed on to a variety of colloidal materials or biota, and thereby be transported through the water environment. A notable characteristic of PAH is their sensitivity to light.

PAH have been found in industrial and municipal waste effluents, and occur in soils, ground waters and surface waters, and their sediments and biota. With the exception of filtration or sorption by activated carbon, conventional water treatment processes do not efficiently remove them, and they have been found in domestic water supplies. Because of the ubiquity of PAH in the environment, it is impossible to prevent completely man's exposure to them ; nevertheless their surveillance should be continued and their concentrations in the environment should be reduced where practicable.

INTRODUCTION

Many polynuclear aromatic hydrocarbons (which will be referred to as PAH) are known to be carcinogenic to animals and probably to man (Badger, 1962; Falk et al., 1964; Hueper & Conway, 1964). The potential hazard of PAH to man in water supplies has been noted by the WHO Expert Committee on the Prevention of Cancer (1964) which states:

> Industrial effluents and atmospheric pollutants can find their way into water supplies. Water purification procedures in general use are designed to deal with bacteriological hazards and specific chemical impurities. Each chemical impurity may necessitate special measures. It is important that attention should be given to this problem in terms of carcinogenic compounds.

In surveying the literature concerning the incidence and significance of such PAH in natural and treated waters, it became apparent that the large majority of these studies were being performed in Europe. For example, Wedgwood & Cooper, in England, have studied PAH in industrial effluents and sewage (Wedgwood, 1952a, 1952b, 1953; Wedgwood & Cooper, 1953, 1954, 1955, 1956). Borneff and coworkers, in Germany, have investigated the incidence and origin of PAH in, and adjacent to, fresh water and water supplies, as well as the effectiveness of various treatment processes (Borneff, 1960, 1963a, 1963b, 1964a, 1964b, 1965, 1967, 1969; Borneff & Knerr, 1959a, 1959b, 1960; Borneff & Fischer, 1961a, 1961b, 1961c, 1962a, 1962b, 1962c, 1962d, 1963; Borneff & Kunte, 1963, 1964, 1965, 1967, 1969; Borneff, Engelhardt et al., 1968; Kunte,

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1967, 1969; Müller, 1968; Müller & Reichert, 1969; Münch, 1966; Reichert, 1968a, 1968b, 1968c). Mallet and co-workers, in France, have performed similar studies, mainly in marine waters (Mallet, 1960, 1961, 1962, 1964, 1965a, 1965b, 1966, 1967; Mallet & Héros, 1960, 1961, 1962; Mallet & Lami, 1964; Mallet & Le Theule, 1961; Mallet & Priou, 1967; Mallet & Sardou, 1964, 1965; Mallet & Schneider, 1964; Mallet & Tissier, 1965; Mallet et al., 1960, 1963a, 1963b, 1967; Binet & Mallet, 1963; Bourcart et al., 1961; Bourcart & Mallet, 1965; Depuis, 1960; Greffard & Meury, 1967; Lalou, 1963, 1965; Lalou et al., 1962; Perdriau, 1964a, 1964b), while several investigators from the USSR have investigated PAH in industrial wastes (Gortalum & Dikun, 1958; Cherkinsky et al., 1959; Grigorev, 1960; Makhinenko & Dikun, 1962; Yanysheva et al., 1962; Dikun & Makhinenko, 1963; Fedorenko, 1964; Veldre et al., 1965a, 1965b; Ershova, 1968; Ershova & Mints, 1968; Samoilovich & Redkin, 1968). Ilnitsky & Varshavskaya (1964) reviewed some of the literature up to 1962 concerning water as a possible vehicle for the transmission of carcinogens through the environment. Their review principally discussed 3,4-benzpyrene (which will be referred to as BP), which they note is increasingly polluting natural waters. A discussion on possible pollution control measures followed in a later review (Ilnitsky, 1966).

It is both timely and useful to bring the above and other recent studies to the attention of a wider group of readers, at the same time interrelating and reviewing their various aspects, especially as the recommendation has now been made that treated surface water that is used for drinking-water supplies should be examined for PAH (World Health Organization, 1970). For this purpose, a routine analysis method has been suggested for the determination of 6 PAH, of which 3 are carcinogenic (Borneff & Kunte, 1969).

Carcinogenic substances may enter natural waters, and thereby public water supplies, with the discharge of urban and domestic sewage, the release of industrial wastes, and through rain- and storm-water which contain atmospheric and surface carcinogenic contaminants. Carcinogens of natural or industrial origin adsorbed on to vegetation or incorporated into soil may leach into the ground water as well (Hueper, 1960).

The rapidly increasing urbanization and industrialization of our society is accompanied by an increased demand on water resources—rivers, lakes and underground reservoirs. Thus, the danger of exposure to carcinogenic PAH from the consumption of contaminated drinking-water will grow in the future.

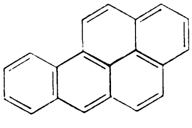
The present review is concerned primarily with PAH in the water environment, with emphasis on BP because it is ubiquitous, is one of the most potent of the carcinogenic PAH and is widely studied. Whenever BP concentrations alone are mentioned it is because no other PAH were reported. The review will also discuss other carcinogenic and non-carcinogenic PAH. Pertinent physico-chemical properties of these materials, such as solubility and stability, will be reviewed as will analytical techniques. The sources and origins of these chemicals in the aquatic environment will be considered, as well as the mode of transport to natural waters and their incidence in surface, ground, marine and drinking water. The effect of water- and waste-treatment processes in removing PAH will be reviewed. Finally, the significance of water-borne carcinogenic PAH for humans will be discussed. Contamination of air by PAH has been widely studied and reviewed elsewhere (for example, Sawicki & Cassel, 1962) and will be discussed only as a source of contamination of water.

PHYSICO-CHEMICAL PROPERTIES AND ANALYSIS OF PAH

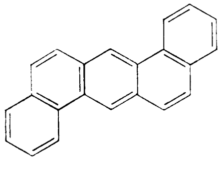
Table 1 lists the various PAH compounds referred to in this review, along with their abbreviations, empirical formulae and relative carcinogenic potency. As indicated there, BP is one of the most potent carcinogens. The chemical structures of two PAH are given in the accompanying diagram.

One of the most important characteristics of PAH relative to their incidence in water is solubility. As would be expected from their high molecular weight and lack of polar substituent groups, their solubility in pure water is extremely low. For example, after equilibrating water for two years with crystals of DBA, none of this material could be detected in

TYPICAL POLYNUCLEAR AROMATIC HYDROCARBONS



3,4-benzpyrene



1,2,5,6-dibenzanthracene

TABLE 1
LIST OF PAH COMPOUNDS

Symbol	Compound	Carcinogenic[a] potency	Empirical formula
AC	anthracene	?	$C_{14}H_{10}$
AF	alkylfluoranthene	?	
AN	acenaphthylene		$C_{12}H_8$
AP	alkylpyrene	?	
AT	anthanthrene	—	$C_{22}H_{12}$
BA	1,2-benzanthracene	+	$C_{18}H_{12}$
3,4-BF	3,4-benzfluoranthene	++	$C_{20}H_{12}$
10,11-BF	10,11-benzfluoranthene	++	$C_{20}H_{12}$
11,12-BF	11,12-benzfluoranthene	—	$C_{20}H_{12}$
BP	3,4-benzpyrene	+++	$C_{20}H_{12}$
1,2-BP	1,2-benzpyrene	±	$C_{20}H_{12}$
BPR	1,12-benzperylene	—	$C_{22}H_{12}$
CH	chrysene	+	$C_{18}H_{12}$
CR	coronene	—	$C_{24}H_{12}$
DBA	1,2,5,6-dibenzanthracene	+++	$C_{22}H_{14}$
DMBA	9,10-dimethyl-1,2-benzanthracene	(active)	$C_{20}H_{16}$
FL	fluorenthene	—	$C_{16}H_{10}$
FR	fluorene	—[b]	$C_{13}H_{10}$
IP	indeno (1,2,3-cd) pyrene	+	$C_{22}H_{12}$
MCA	3-methylcholanthrene	(active)	$C_{21}H_{16}$
PA	phenanthrene	?	$C_{14}H_{10}$
PR	perylene	—	$C_{20}H_{12}$
PY	pyrene	—	$C_{16}H_{10}$
TP	triphenylene	—	$C_{18}H_{12}$

[a] Potency notation: +++, active; ++, moderate; +, weak; —, inactive; ?, unknown. (Compounds labelled " active " are so listed by several authors). The notation is taken from Hoffman & Wynder (1962).

[b] Reference from Gräf & Nowak (1966).

distilled water by an analytical technique sensitive to 0.01 μg/litre (Borneff & Knerr, 1960). The increase in solubility of PAH by the addition of water-soluble organic compounds is a phenomenon that is important in the passage of these compounds into and through environmental waters.

Of special interest is the phenomenon of solubilization (Klevens, 1950; McBain & Hutchinson, 1955), which is defined by the latter as " a process in which otherwise insoluble matter is brought into solution by colloidal matter, specifically by micelles."

The solubilization of PAH by micelles is of particular interest because the latter are formed in water by synthetic detergents. However, it should be noted that solubilization does not occur until a sufficient amount of the solubilizing agent has been added and the region of the critical micelle concentration (CMC) reached. Once the vicinity of the CMC is reached, the increase in concentration of solubilizing agent results in increased solubility of PAH, although the relationship is often not a linear one. In general, the CMC for synthetic detergents is high, one study measuring a value of 40 mg/litre of a linear alkylbenzene sulfonate in drinking-water (Böhm-Gössl & Krüger, 1965). Thus, for most natural and treated waters the detergent concentrations are lower than the minimum amount required to solubilize.

One must also consider the phenomenon of hydrotropy, that is, the increase in PAH solubility in water as a result of the introduction of other organic chemicals not associated with colloid formation; for example the increased BP water solubility by lactic acid (Ekwall & Sjöblom, 1952), purines, such as caffeine (Weil-Malherbe, 1946), acetone (Gräf & Nothhafft, 1963; Suess, 1967), and ethyl alcohol (Brock et al., 1938; Suess, 1967). There is sufficient evidence to indicate the possibility of the increased solubility of PAH as a result of the presence of a wide variety of organic compounds that may be found in these waters. Finally, it should be noted that highly polluted water containing emulsions of organic solvents would be capable of containing relatively large quantities of PAH.

The sorption of PAH on to surfaces is an important characteristic in relation to their presence in environmental waters. The ability of BP to concentrate on activated carbon, calcareous material, silica, glass and plastics by sorption has been noted (Brock et al., 1938; Borneff & Knerr, 1959a; Mallet & Schneider, 1964; Knorr & Gätzschmann, 1966; Suess, 1967). Hence, the presence of minerals and other suspended and settled particulates in environmental waters should be considered in relation to their ability to contain and carry PAH.

The stability of PAH, especially as affected by light and oxygen, is an important characteristic in relation to their presence in water or any other environment. Recently, a study was made of the degradation of BP and 12 other PAH dissolved in cyclohexane and dichloromethane (Kuratsune & Hirohata, 1962). Sunlight, fluorescent lamps, and an ultraviolet source consisting mostly of the 365 $m\mu$ and 366 $m\mu$ mercury lines, were the sources of illumination, the solutions being irradiated for up to 377 hours. The most light-sensitive compounds were naphthacene and DMBA, with BP and AC following. The other PAH generally did not decompose. Degradation was similarly affected by daylight-type fluorescent lamps and sunlight when using reasonably comparable light intensities and exposure times. Using an ultraviolet source, the decomposition of BP was compared in oxygen- and nitrogen-saturated solutions. The nitrogen-saturated solution reduced somewhat the BP degradation compared with the solution that had been exposed to air prior to sealing and irradiation. However, BP in the oxygen-saturated solution decomposed at a much higher rate. Several other investigations were performed, studying similar effects of various light sources, organic solvents in pure form or as mixed aqueous solutions, and the presence of oxygen on the degradation of BP and other PAH (Kriegel & Herforth, 1957; Woenckhaus et al., 1962; Reske & Stauff, 1963, 1964; Tanimura, 1964; Reske, 1966; Masuda & Kuratsune, 1966; Harrison & Raabe, 1967; Suess, 1967; Jäger & Kassowitzova, 1968).

A study was made of the degradation of BP sorbed on to surfaces of calcium carbonate in aqueous suspensions exposed to fluorescent light (Suess, 1967; Andelman & Suess, in press). Such a system is likely to simulate BP behaviour in natural water environments, particularly because of the demonstrated ability of BP to sorb on to mineral surfaces. The effects of light intensity, oxygen concentration, temperature, pH and ionic strength on the photodecomposition rate were determined, and the mechanism of this process was discussed. The results indicated that while higher light intensity, oxygen concentration and temperature accelerated BP degradation, pH and ionic strength in the ranges studied had no effect.

Of immediate implications for environmental waters was one study in which a comparison was made of the light sensitivity of BP in crystalline form, vegetable oil and aqueous detergent solutions, using ultraviolet irradiation and daylight (Borneff & Knerr, 1959b). The degradation was greatest in the aqueous detergent solution, next in the oil solution, and least in the crystalline form. With daylight illumination of about 8000 lux (1.2 mW/cm^2) approximately 20 hours were required to degrade 90% of the BP in the aqueous solution, and 60–90 hours in oil. The shorter ultraviolet wave lengths were particularly effective. Because these PAH have varying degrees of sensitivity, their relative concentrations in the environment need not reflect those in the contaminating sources. The presence of oxygen and other oxidizing agents will significantly increase rate of degradation of the PAH. In aqueous and mixed aqueous solutions they will degrade faster than in pure organic solvents or in crystalline form.

The analytical, concentration and separation techniques for examining PAH in environmental samples have been comprehensively reviewed (Sawicki, 1964). Sawicki emphasized that in environmental samples these compounds are generally present in very small quantities, along with many other kinds of organic materials. Thus, prior to using instrumental methods for their analysis, it is necessary to separate the PAH from other classes of compounds and from each other. In addition they must often be concentrated prior to the instrumental analysis. Solid–liquid and liquid–liquid extraction techniques have been used in the initial stages of separation and concentration, often followed by column, paper and thin-layer chromatography. Gas chromatography has also been used as a separation–analysis technique. The most frequently used instrumental methods for identification and quantitative analysis are based on the ultraviolet–visible absorption spectra, as well as the fluorescence spectra. A concise description of the simultaneous determination of 13 PAH is given by Grimmer & Hildebrandt (1965a). For a routine determination of PAH in drinking-water, however, a simplified procedure is referred to by the World Health Organization (1970), suggesting the evaluation of only 6 PAH—namely, 3,4-BF; 11,12-BF; BP; FL; BPR and IP (Borneff & Kunte, 1969) (Table 1).

It is not the intention of this review to examine in detail the techniques used in the analysis of environmental samples. Yet, an extensive series of studies should be mentioned, as they are particularly useful in describing the methodology for the examination of a wide variety of water and water-related samples. There are discussions of adsorption spectra and chromatographic techniques (Borneff & Knerr, 1959a; Kunte, 1967), extraction of BP from water and sand (Borneff & Knerr, 1959b), and fluorescence analysis (Borneff, 1960; Kunte, 1967). In Borneff's paper the greater sensitivity of fluorescence compared with ultraviolet-absorption analysis (10-fold) was noted. This is also discussed elsewhere (Sawicki, 1964), although this sensitivity ratio was there stated to be in the range of 10–1000. Extraction of PAH from activated carbon, as well as losses encountered in the subsequent steps of distillation, column and paper chromatography were discussed (Borneff & Fisher, 1961a). Descriptions were given for techniques used in the analysis of filter mud (Borneff & Fischer, 1961b), zooplankton and phytoplankton (Borneff & Fischer, 1961c), soil (Borneff & Fischer, 1962d), the centrifugate from surface water (Borneff & Fischer, 1963), and liquid–liquid extraction of river, lake and tap water (Borneff & Kunte, 1964).

Although bioassay with conventional laboratory animals has long been used in testing the carcinogenicity of specific compounds or extracts containing unknown materials, the use of bacteria for assaying carcinogenic PAH may have advantages in terms of speed and sensitivity (Won & Thomas, 1962).

A photodynamic bioassay using *Paramecium caudatum* has been developed as a presumptive index of carcinogenicity from PAH and used to analyse carbon-filter extracts of finished drinking-water (Epstein & Taylor, 1966). The correlation of this assay with detailed instrumental–chemical analyses of PAH could obviate the need for the latter and be particularly useful in the analysis of large numbers of water samples for carcinogenic PAH.

ORIGIN, SOURCE AND VEHICLES OF TRANSMISSION OF PAH

In this discussion the term " origin " will refer to the formation of PAH, while " source " will signify the object or material in which they are concentrated and from which they may be disseminated into the environment. The vehicles of transmission are industrial and domestic effluents, atmospheric fall-out, precipitation and run-off water. The ubiquity of PAH, in spite of their instability, particularly with exposure to light and oxidizing agents, and their very low solubility in water, prompt one to consider their possible origins.

Origin and sources of PAH in the environment

Until recently the evidence available seemed to indicate that the carcinogenic hydrocarbons in the environment are formed only at high temperatures (Badger, 1962; Badger et al. 1966). Badger (1962) notes that coal tar, which is produced by heating coal in the absence of air, consists of relatively large quantities of BP and other PAH when produced at high temperatures (e.g., 700°C), as compared with 300°C–450°C when the products are mostly paraffins, cycloparaffins, olefins, and phenols. The carcinogenic effect of petroleum asphalt, cooking oil and coal tar has been studied (Hueper & Payne, 1960; Hueper & Conway, 1964; and others). The materials associated with such high temperature pyrolysis that have been shown to contain PAH, and which may act as a source of them for the water environment, include coal tar and coal-tar pitch, shale oil (Cahnmann, 1955), and carbon black, which is used in many manufacturing processes, particularly being incorporated into automobile tyres (Badger, 1962). Thus, the wear of the latter on roads can be a source of PAH. Bitumen or asphalt used in constructing roads contain PAH, as does dust that may be collected from their surfaces and carried by run-off water (Borneff & Kunte, 1965). BP has been isolated from cracked mineral oils but carcinogenic PAH have generally not been isolated from uncracked oils (Cook et al., 1958; Badger, 1962). However, there is some evidence for the occurrence of some PAH in crude oil (Meinschein, 1959), and the presence of BP in concentrations of about 1 mg/litre has been demonstrated (Gräf & Winter, 1968). Shipping and harbour oils have been connected with contamination of water by PAH (Shimkin et al., 1951; Cahnmann & Kuratsune, 1956; Mallet, Tendron & Plessis, 1960; Mallet & Le Theule, 1961; Borneff, 1964a), as have effluents from a variety of industries using pyrolytic processes.

The question of endogenous formation of PAH in plants and micro-organisms (Mallet & Héros, 1962; Borneff, 1963b, 1964a; Gräf, 1964, 1965; Gräf & Diehl, 1966), arises because of their ubiquity in the environment, particularly in a wide variety of materials which were not likely to have been associated with pyrolytic processes. Several PAH were found in forest soils remote from human habitation and in other soils (Kern, 1947; Mallet & Héros, 1962; Binet & Mallet, 1963; Gräf, 1965; Mallet, 1965a, 1966; Zdražil & Picha, 1966). In general, the upper layer of the earth contains carcinogenic PAH in the range of 100 μg/kg–1000 μg/kg; although these soils are a source of PAH for surface waters, their contribution is small (Borneff, 1964a). The presence of BP cannot be attributed only to fall-out from polluted air; rather, it seems to be indigenous to the soil and might be the product of living organisms (Blumer, 1961). The synthesis of PAH by micro-organisms to account for their presence in filtration wash-water sludge has been suggested (Knorr, 1965), and was later verified by a laboratory study of various bacteria which accumulated BP through synthesis in amounts of 2 μg/kg–6 μg/kg of dried material (Knorr & Schenk, 1968). Laboratory culture studies of the fresh water alga *Chlorella vulgaris* have shown that it synthesizes several PAH (Borneff, 1964a; Borneff, Selenka et al., 1968a; Borneff, Selenka et al., 1968b). The extracted algae contained carcinogenic PAH in the range of 10 μg/kg–50 μg/kg (Borneff, 1964b).

The presence of PAH has been demonstrated in a wide variety of plants from diverse sources (Guddal, 1959; Borneff, 1963b; Grimmer, 1966). Of special interest is some recent research on the synthesis of PAH and their physiological functions in plants (Gräf, 1964, 1965; Gräf & Diehl, 1966; Gräf & Nowak, 1966). Wheat and rye were grown hydroponically in solutions made from reagent-grade chemicals which were free of PAH and in the presence and absence of light. Whereas the seeds contained only traces of BP the seedlings contained 10 μg–20 μg BP per kg of dried material after 8–10 days of growth. It was concluded that the BP was synthesized by the plants, both in the presence and absence of light. In another series of experiments it was shown that BP added to the soil or nutrient media accelerated the growth of kohlrabi, cauliflower, wheat, rye, and tobacco.

In the case of rye, the grain output was 3 times greater when the BP was added. It was found that BP was not stored in these experiments, but was utilized. Similar experiments with DBA, a strong carcinogen, and BA, a weaker one, suggested a direct relationship between carcinogenic potency in animals and the ability of PAH to promote growth. The investigator concluded that it is likely that there is a world-wide synthesis of PAH in plants, and that they have always been present in man's environment. Although additional research on the synthesis of PAH by plants is required to justify the conclusions that the process is a universal one, the great amount of evidence of the ubiquity of PAH supports the likelihood of such syntheses.

Vehicles of transmission of PAH

Various industrial entreprises are a potential source of PAH, and contaminate the water environment when releasing their large amounts of waste-water effluents into lakes, rivers and coastal waters (Bourcart et al., 1961; Mallet & Le Theule, 1961; Borneff, 1964a; Borneff & Kunte, 1965; Bourcart & Mallet, 1965). Industrial effluents, loaded with PAH, may be produced by refineries, industries utilizing solid and liquid fuel materials for manufacturing chemical by-products, the plastics and dyestuffs industries, high-temperature furnaces, the lime industry and others (Mallet & Le Theule, 1961). Although industrial waste-water has been generally recognized as a potential vehicle of transmission of PAH, only few quantitative investigations have been conducted. The majority of the reports were published by British and Soviet authors (Table 2). The British authors studied the PAH content in industrial effluents before entering the city sewers, while the Soviet authors put the emphasis on the study of BP concentration in the effluents of various processing stages of the solid fuel by-product industry (Gortalum

TABLE 2
PAH CONCENTRATION IN INDUSTRIAL EFFLUENTS

Industry	Source of waste-water	BP concentration (µg/litre)	Reference
Shale-oil	After treatment for dephenolization	320	Makhinenko & Dikun, 1962 Dikun & Makhinenko, 1963
		5	Gräf, 1964
		2	Veldre et al., 1965a,1965b
Coke by-products	Not indicated	present	Yanysheva et al., 1962
	After biochemical treatment	12–16	Fedorenko, 1964
	After oil separation (5 samples)	6.5, 130, 250, 290 and " big " quantity	Cherkinsky et al., 1959
	Spent gas liquor	very small quantity [a]	Wedgwood, 1952b
Coke or oil-gas works	Before discharge to sewer	not indicated [b]	Wedgwood & Cooper, 1955
	Before discharge to sewer (2 plants)	1 000 and 340 [c]	Wedgwood & Cooper, 1956
Oil-gas works	After oil separation (3 samples)	3, 6, and 30	Cherkinsky et al., 1959
Oil refinery	After oil separation (3 samples)	none detected	Cherkinsky et al., 1959
Tar paper	Not indicated	present	Grigorev, 1960
Acetylene	Not indicated	0.015–0.100 [d]	Filippov & Ruchena, 1965
Ammonium sulfate	After cooling and settling	about 10 [e]	Wedgwood & Cooper, 1955

[a] Also present were: AT, BA, CH, FL, PR, and PY.
[b] Present or suspected were: AC, AN, BPR, FR, and PA.
[c] Also present were: AC, BPR, FL, and PY.
[d] The figures were taken from *Chemical Abstracts*, but appear to be too low. It is assumed that the correct values should read 15–100 µg/litre.
[e] Also present: AP, AT, BA, 1,2-BP, CH, FL, PR, PY, and TP.

& Dikun, 1958; Dikun & Makhinenko, 1963) as well as in the finally treated waste-water (Veldre et al., 1965b). The results indicated that BP concentration increased with increased cracking temperature, but it was not detected at all in waste-water from operations under 500°C (Cherkinsky et al., 1959; Veldre et al., 1965a).

Municipal wastes (Table 3) often contain large quantities of industrial effluents (Wedgwood & Cooper, 1955, 1956). During the rainy season the sewer system may also be affected by run-off water passing over roads. In addition, human urine was found to contain BP (Mallet & Héros, 1960), while the human metabolite 8-hydroxy-3,4-BP was identified in both the urine and faeces (Iversen, 1947). It was noted that PAH were actually present in sewage sludge whether or not industrial wastes enter the system (Wedgwood, 1952b; Wedgwood & Cooper, 1956).

The presence and fate of PAH in the polluted atmosphere have been widely studied, and discussed in detail in the literature. It has been shown that some of the PAH in air find their way to the ground adsorbed on to aerosols and bacteria (Mallet & Héros, 1961), either by direct settling or through precipitation. Run-off water can then collect the PAH from areas covered with atmospheric fall-out and road-dust (Wedgwood & Cooper, 1954; Borneff, 1964a; Borneff & Kunte, 1965), as well as from freshly tarred or oiled surfaces (Hueper & Ruchhoft,

TABLE 3

PAH CONCENTRATION IN DOMESTIC EFFLUENTS

Source	Concentration (μg/litre) [a]			Reference
	BP	Carcinogenic PAH	Total PAH	
Domestic effluent from a small community after primary treatment	0.170	3.0	15.0	Borneff & Kunte, 1964
Waste effluents entering Rotach River:				Borneff & Kunte, 1965
Sample I	0.015	0.2	0.5	
Sample II	0.047	1.1	2.7	
Sample III	0.079	1.0	6.6	
Sample IV	0.100	0.5	5.1	
Sample V	0.038	0.3	0.8	
Waste effluent of Stockach	0.100	5.0	15.0	Borneff & Kunte, 1965
Waste effluent of Radolfzell	0.368	2.8	8.6	Borneff & Kunte, 1965
Waste effluents of Hegne:				Borneff & Kunte, 1965
Sample I	0.001	0.1	0.8	
Sample II	0.011	1.2	4.2	
Sample III	1.840	37.9	87.5	
Sample IV	0.450	31.6	68.0	
Vegetable cold wash-water:				Grimmer, 1966
Sample I	1.60	8.1	25.2	
Sample II	1.02	5.8	22.6	
Urine from Paris inhabitants (4 samples)	1 to 3			Mallet & Héros, 1960
Sewage of Leningrad	present			Poglazova et al., 1966
Sewage, final effluent (4 plants—8 samples)	present	0.15–1.50 PY, AN, AP and FL		Wedgwood & Cooper, 1954, 1956
Sludge from secondary treatment (humus)	present	AC, AN, AT, BA, FL, PR, and PY		Wedgwood, 1952b Wedgwood & Cooper, 1954, 1955
Humus, dried	3 000			Wedgwood & Cooper, 1954

[a] Detailed tables with figures for the various carcinogenic and non-carcinogenic PAH are presented in Borneff & Kunte, 1964, 1965; Grimmer, 1966.

1954). A part of the run-off water gets into the city sewers or the natural surface water, while another part of it may infiltrate through the soil into the ground-water.

Only a few samples of precipitation and run-off water have been analysed for their PAH content. Nevertheless, PAH were recovered from rain, and freshly fallen snow (Cooper & Lindsey, 1953; Gilbert & Lindsey, 1955).

PAH IN ENVIRONMENTAL WATERS

Borneff & Kunte (1964) divided fresh waters into 4 categories with respect to their carcinogenic PAH concentration (Table 4), these being correlated in general with their degree of pollution. However, in this review environmental waters are divided into 4 different categories. The first two, marine and surface water, include flora, fauna and sediments,

TABLE 4
CONCENTRATION OF CARCINOGENIC PAH IN FRESH WATERS[a]

Water type	Concentration range (μg/litre)
Ground-water	0.001–0.010
Treated river and lake water	0.010–0.025
Surface water	0.025–0.100
Surface water, strongly contaminated	>0.100

[a] Taken from Borneff & Kunte, 1964.

in addition to the water itself. The other two categories are ground- and drinking-water.

The majority of the investigations in the marine environment were performed by Mallet & co-workers (Suess, 1970). Systematic studies were made of the Atlantic, Channel and Mediterranean coasts of France (Mallet et al., 1960; Mallet, 1960, 1961; Bourcart et al., 1961; Mallet & Le Theule, 1961; Lalou et al., 1962; Mallet & Sardou, 1964; Mallet & Schneider, 1964; Mallet & Lami, 1964; Perdriau, 1964a; Mallet, 1965; Mallet & Sardou, 1965; Lalou, 1965; Greffard & Meury, 1967; Mallet, 1967) the Bay of Naples, Italy (Bourcart & Mallet, 1965), and the western coast of Greenland (Mallet, Perdriau & Perdriau, 1963a, 1963b; Perdriau, 1964b) (Tables 5–8). The latter, a practically unpopulated coast with little shipping and water vegetation was, nevertheless, found to contain on the average the same order of BP contamination as the French coasts, thus indicating the ubiquity of BP in the oceans. In a number of locations along the French coast the fish and molluscs contained notable amounts of BP, while the alluvial deposits at the same location showed only traces, possibly indicating that certain marine organisms are able to concentrate and fix hydrocarbons.

In the past it has been suggested that hydrocarbons may possibly be transported in the oceans by sea streams from industrialized and heavily polluted areas to remote and unpopulated regions (Bourcart et al., 1961; Mallet & Sardou, 1964). However, recent reports on endogenous synthesis of PAH in flora (see previous section) indicate that they may be formed in these areas. This can be supported by studies on the origin of petroleum which showed the presence of aromatic (and other) hydrocarbons in phytoplankton and which postulated that the amount of aliphatic and aromatic hydrocarbon compounds produced by phytoplankton per square kilometre of ocean may be as high as 3 tons (2.7 metric tonnes) per year (Smith, 1954). In addition, plankton may be able to fix them from exogenous sources (Mallet

TABLE 5
CONCENTRATION OF BP IN MARINE PLANKTON

Source	BP concentration (μg/kg of dry sample)	Reference
Greenland[a]	5.5	Mallet, Perdriau & Perdriau, 1963a
Italy[b]	6.1–21.2	Bourcart & Mallet, 1965
French Channel coast	400	Mallet & Sardou, 1964
French Mediterranean coast[c]	not detected to 5	Mallet & Sardou, 1964, 1965
Estuary, French Channel coast[d]	100	Mallet & Lami, 1964
Ditto[e]	350	Mallet & Lami, 1964
Diatoms serving as filters in food industry	5.5	Mallet & Schneider, 1964

[a] One sample from depth of 30 m.
[b] Six samples collected on water surface and from a depth of 2 m.
[c] Fifteen samples.
[d] Plankton debris in foam immediately downstream of dam.
[e] Plankton debris west of outlet, downstream from dam.

TABLE 6

CONCENTRATION OF BP IN MARINE ALGAE

Source	BP concentration (μg/kg of dry sample)	Reference
Greenland, west coast [a]	60	Mallet, Perdriau & Perdriau, 1963a
Greenland, west coast [b]	60	Mallet, Perdriau & Perdriau, 1963a
Italy	2.2	Bourcart & Mallet, 1965
French Channel coast	not detected	Mallet, 1961

[a] Sample from depth of 40 m.
[b] Samples from bottom of the sea and shore.

TABLE 7

CONCENTRATION OF BP IN MARINE FAUNA

Source	Sample	BP concentration (μg/kg of dry sample)	Reference
Greenland, west coast [a]	Cod fish	15	Mallet, Perdriau & Perdriau, 1963a
	Mollusc	60	
	Holothurian	not detected	
	Mussel: shell	18	
	body	55	
Italy, Bay of Naples [b]	Mussel: shell	11	Bourcart & Mallet, 1965
	body	130 and 540	
Italy, Bay of Naples [c]	Mollusc	2.4	
Italy, Bay of Naples [d]	Sardine	65	
Various French coasts [e]	Shrimp, oyster, mussel, mollusc, crab, etc.	not detected to traces to 1.5–90	Mallet, 1961
French Atlantic coast	Oyster: shell	3.5 μg/dozen	Mallet, Tendron & Plessis, 1960
	body	0.4 μg/dozen	
French Channel coast	Oyster: lower shell	70	Mallet & Schneider, 1964
	upper shell	112	
France, Toulon harbour	Mussel	16–22 [f]	Greffard & Meury, 1967
Alabama	Oyster: shell	24	Mallet & Schneider, 1964
Virginia	Oyster	2–6 [g]	Cahnmann & Kuratsune, 1957
California	Goose barnacle	present	Koe & Zechmeister, 1952
California	Thatched barnacle	present	Shimkin et al., 1951; Koe & Zechmeister, 1952

[a] Sample from depth of 40 m.
[b] Two samples.
[c] Sample from depth of 35 m.
[d] Sample from water surface.
[e] Mallet (1961) presents a detailed list of 25 samples covering 13 species.
[f] Total PAH was about 1100–3400 μg/kg.
[g] Total carcinogenic PAH was about 300 μg/kg, and total PAH about 1200 μg/kg.

TABLE 8
CONCENTRATION OF BP IN MARINE SEDIMENTS

Source	Sample	Depth (m)	BP concentration (µg/kg of dry sample)	Reference
Greenland, west coast	Sand	0.20	5	Mallet, Perdriau & Perdriau, 1963*a*
Italy, Bay of Naples (6 locations)	Mud, sand, shell [a]	15–45	1 000–3 000	Bourcart & Mallet, 1965
	Sand, shell	13	7.5	
	Mud, sand	2–65	10–530	
	Mud, sand, shell [b]	55	260–960	
	Muddy sand	120	1.4	
	Mud, sand [c]		100–560	
French Mediterranean coast		0–0.03	1 800	Lalou et al., 1962
		0.03–0.08	3 600	
		0.08–0.13	5 000	
		0.13–0.18	2 500	
		0.23–0.28	2 200	
		0.33–0.38	730	
		0.48–0.53	420	
		1.00	26	
		2.00	16	
French Mediterranean coast	Sand	14	400	Bourcart et al., 1961
	Black mud	16	1 500	
	Sand	48	75	
	Sand	58	traces	
	Beige mud	82	400	
Estuary, French Mediterranean	Sand		34	Bourcart et al., 1961
	Sand	1	20	
	Sand	4	15	
	Sand	5	25	
	Mud	102	not detected	
French Mediterranean coast			50	Bourcart et al., 1961
French Mediterranean coast			20	Bourcart et al., 1961
French Channel coast	Mud		15 000	Mallet, Tendron & Plessis, 1960
French Channel and Atlantic coasts (11 locations)	Mud, sand		not detected to 1 700	Mallet & Le Theule, 1961

[a] Five samples were collected 300 m from shore. Area is highly industrialized.
[b] Four samples were collected in the vicinity of volcanic pollution.
[c] Four samples. This island is affected by pollution.

TABLE 9
CONCENTRATION OF PAH IN SURFACE WATERS

Source	Concentration (µg/litre)[a]			Reference
	BP	Carcinogenic PAH	Total PAH	
Bodensee		0.003		Borneff & Fischer, 1962b
		0.0004[c]		Borneff, 1964a
	0.0013	0.030	0.065	Borneff & Kunte, 1964
Alprhine		0.005[c]		Borneff, 1964a
River Rhine			present	Holluta & Talsky, 1955
River Rhine		0.050 to 0.500[d]		Borneff & Fischer, 1962a
River Rhine at Mainz		0.080[c]		Borneff, 1964a
River Rhine at Mainz (Mar. 1964)	0.049	0.240	0.73	Borneff & Kunte, 1964
(Mar. 1964)	0.114	0.730	1.50	
River Main, at Seligenstadt	0.0024	0.155	0.48	Borneff & Kunte, 1964
River Danube, at Ulm				Borneff & Kunte, 1964
(Apr. 1964)	0.0006	0.055	0.28	
(May 1964)		0.078	0.20	
River Gersprenz, at Münster				Borneff & Kunte, 1964
(Jan. 1964)	0.0096	0.055	0.16	
(Apr. 1964)		0.038	0.12	
River Aach, at Stockach[c]		0.50		Borneff, 1964a
River Aach, at Stockach[c]				Borneff & Kunte, 1965
Sample I	0.043	1.30	3.0	
Sample II	0.016	0.90	2.5	
Sample III	0.004	0.50	1.4	
Sample IV	0.005	1.10	3.1	
River Schussen (Bodensee)		0.50[c]		Borneff, 1964a
River Schussen	0.01	0.20	1.0	Borneff & Kunte, 1965
River Argen (Bodensee)		0.07[c]		Borneff, 1964a
River Seine	Considerable amount			Mallet, 1965, 1966
River Plyussa:				Dikun & Makhinenko, 1963
at discharge site of shale-oil effluent	12			
3500 m downstream	1			
at the water intake of Narvy	0.1			
A river:				
15 m below discharge of coke by-product effluent	8–12			Fedorenko, 1964
500 m downstream	2–3			
Peat (turf) water	0.05			Gräf, 1965

[a] Detailed tables with figures for the various carcinogenic and non-carcinogenic PAH are presented in Borneff & Kunte, 1964, 1965.

[b] Extrapolated from rapid sand filtration lake mud analyses.

[c] Extrapolated from centrifugate fractions.

[d] Extrapolated from activated-carbon adsorption analyses.

& Sardou, 1964), and marine fauna may be contaminated with BP, regardless of whether in polluted or unpolluted locations (Table 7). PAH are not considered normal metabolic products of marine fauna, but may be ingested in the course of filter-feeding (Shimkin et al., 1951; Koe & Zechmeister, 1952; Zechmeister & Koe, 1952; Mallet, Tendron & Plessis, 1960; Mallet, 1960). This may also explain the high BP concentration found in the calcareous shell of some organisms (Mallet & Schneider, 1964) (Table 7).

The sources of PAH in the marine sediments include: surface effluents, ships, volcanic debris (Bourcart & Mallet, 1965), and activity of organisms, including bacteria. The latter may explain the very high BP concentration found at some points under the bottom of the Bay of Villefranche (Lalou et al., 1962) (Table 8).

Carcinogenic PAH have been detected in many surface waters (Table 9). Mountain streams, such as the Alprhine, and relatively non-polluted lakes, such as the Bodensee (Lake Constance), have significantly lower concentrations of PAH compared with polluted rivers, such as the lower Rhine. Industrial and municipal effluents are an important source of PAH in surface water. Phytoplankton,

TABLE 10

CONCENTRATION OF PAH IN SURFACE WATER ENVIRONMENT

Source	Sample	Concentration (µg/kg of dry sample)[a]			Reference
		BP	Carcinogenic PAH	Total PAH	
Laboratory	Filamentous algae		10–50		Borneff, 1964a
Bodensee	Phytoplankton[b]	2	170	700	Borneff & Fischer, 1962c
Bodensee	Suspended solids[c]	50	1 250	1 500	Borneff & Fischer, 1962b
Bodensee	Suspended solids[d]	200	3 200	6 000	Borneff & Fischer, 1963; Borneff, 1964a
Bodensee	Suspended solids[e]		3 000		Borneff, 1964a
Alprhine, at Bregenz	Suspended solids[e]: Aug. 1962		150	620	Borneff, 1964a; Borneff & Kunte, 1965
	Sept. 1962		15	63	
River Rhine	Suspended solids[e]	300	2 600	8 000	Borneff & Fischer, 1963
River Rhine, at Mainz	Suspended solids[e]		3 000		Borneff, 1964a
River Main, at Seligenstadt			5 000		Borneff, 1964a
River Aach, at Stockach	Suspended solids[e]: Aug. 1962	600	6 700	17 000	Borneff, 1964a; Borneff & Kunte, 1965
	Aug. 1962	2 000	6 400	50 000	
	June 1963	500	19 000	56 000	
River Schussen (Bodensee)	Suspended solids[e]	400	18 000	55 000	Borneff, 1964a; Borneff & Kunte, 1965
River Argen (Bodensee)	Suspended solids[e]	100	3 700	9 600	Borneff, 1964a; Borneff & Kunte, 1965
Fresh-water pool, Italy	Tubifex worms	50			Scaccini-Cicatelli, 1965, 1966

[a] Detailed tables with figures for the carcinogenic and non-carcinogenic PAH are presented in Borneff & Fischer, 1962b, 1962c, 1963; Borneff & Kunte, 1965.

[b] Up to 95% were diatoms, *Asterionella formosa*.

[c] Coarse particles only, from rapid, sand-filter backwash water settled in a sedimentation basin.

[d] Rapid, sand-filter mud centrifugate collected from backwash water after complete suspension (down to 1 µ).

[e] Centrifugate of water sample (down to 1 µ).

one of the sources of PAH in the Bodensee, is, however, only a minor contributor when compared with industrial pollution and run-off water from roads (Borneff & Fischer, 1963; Borneff, 1964a; Borneff & Kunte, 1965).

Suspended solids have been studied as vehicles of transmission of PAH in the water environment. The suspended solid fractions have been obtained either by centrifuging the water sample or by collecting rapid-sand-filtration mud (Table 10). Bottom sediments demonstrated high concentrations of BP as well (Table 11). The values are comparable in range to those for marine sediments (Table 8).

An investigation of PAH in ground-water was performed with the conclusion that, in general, uncontaminated ground-water has the lowest concentration of carcinogenic PAH of all natural waters, ranging from 0.001 µg/litre–0.010 µg/litre (Borneff, 1964a; Borneff & Kunte, 1964) (Table 12).

Potable water samples from the following sources were examined: untreated ground-water, mixed ground- and bank-filtered-water (Table 12), and treated river and lake water (Tables 13 and 14), all samples being taken at the tap. It was concluded that, in general, the carcinogenic PAH concentration in most drinking-water does not exceed 0.025 µg/litre (Borneff & Kunte, 1964). On the other hand, BP could not be detected in the chloroform and alcohol extracts of carbon filters from finished water at different locations in the United States, although there was some evidence of unidentified polynuclear hydrocarbons (Epstein & Taylor, 1966). However, there is evidence that recovery from activated carbon may be incomplete. Because of this it may be preferable in analysing water for its PAH content to concentrate by liquid–liquid extraction, as is

TABLE 11

CONCENTRATION OF BP IN RIVER BOTTOM SEDIMENTS

Source	Depth (m)	BP concentration (µg/kg of dry sample)	Reference
River Seine, about 40 km downstream from Paris:			Mallet, 1965a, 1966
Core sample I	5.9	3 000	
	7.1	29	
	9	4	
	10	4	
Core sample II	0.8	8 500	
	3	6 500	
	10	50	
Core sample III	2	10 000	
Core sample IV	5	390	
Core sample V	9	100	
River Seine mud		15 000	Depuis, 1960
Bottom deposit of a river, below a coke by-product plant		8 200–17 000	Fedorenko, 1964

TABLE 12

PAH CONCENTRATION IN GROUND-WATER

Source	Concentration (µg/litre)[a]			Date of sample collection	Reference
	BP	Carcinogenic PAH	Total PAH		
Ground-water at Finthen		0.002			Borneff, 1964a
Ground-water at Mainz		0.005			Borneff, 1964a
Ground-water[b]:					Borneff & Kunte, 1964
Sample I	0.0001	0.001	0.045	May 1963	
Sample II	0.0006	0.004	0.034	Aug. 1963	
Mixed ground- and bank-filtered-water[b]	0.0003	0.009	0.140	May 1963	Borneff & Kunte, 1964

[a] Detailed tables with figures for the various carcinogenic and non-carcinogenic PAH are presented in Borneff & Kunte, 1964.

[b] Sampling locations not known.

TABLE 13

CONCENTRATION OF CARCINOGENIC PAH IN PURE WATER SAMPLES[a]

Source	Concentration (μg/litre)
Tap water at Mainz mixed from different sources	0.007
Bodensee, centrifuged	0.010
Well water, south of Frankfurt	0.025
River Main, centrifuged	0.800

[a] Taken from Borneff, 1964a.

currently recommended by the World Health Organization (1970).

After studies of the carcinogenic characteristics of pipe coating materials it was suggested that the latter were possible sources of PAH in drinking-water (Druckrey et al., 1960; Zdražil & Picha, 1962; Hettche, 1963; Căzăceanu & Trandafirescu, 1966). In laboratory studies PAH have been detected in water which was in contact with a protective varnish containing a coal-tar base (Rondia, 1966), as well as with bitumen used for painting the inside of pipes (Borneff & Kunte, 1965) (Table 15). The results of the latter experiment suggest the possibility of an increase in the initial PAH concentration in treated water while flowing through long bituminous water lines. However, after sufficient hardening and leaching, these materials probably become an insignificant source of PAH (Borneff & Kunte, 1964, 1965).

EFFECT OF WATER AND WASTE-WATER TREATMENT ON PAH

The effect of some water treatment processes on the removal of PAH has been studied. Table 14 shows the concentrations of BP, carcinogenic and total PAH found in untreated and treated river and lake water. As indicated there, the combination of rapid sand-filtration and chlorination or ozonation

TABLE 14

EFFECT OF WATER TREATMENT ON PAH CONCENTRATION[a]

Water source	Treatment	Concentration (μg/litre)			Date of sampling (1964)
		BP	Carcinogenic PAH	Total PAH	
River Rhine[b]					
Sample I	Untreated	0.114	0.73	1.49	Mar. 3
Sample II	Untreated	0.049	0.24	0.73	Mar. 3
	Bank-filtered and activated carbon filter	0.0005	0.015	0.13	May 11
Bodensee[c]	Untreated	0.0013	0.030	0.065	May 20
Sample I[d]	Rapid sand filtration and chlorination or ozonation	0.0115	0.054	0.234	Mar. 20
Sample II		0.0002	0.007	0.025	May 20
Sample III		0.0005	0.010	0.039	Apr. 7
Sample IV		0.0014	0.017	0.062	May 21
Sample V		0.0024	0.025	0.072	May 21
Sample VI		0.0040	0.028	0.066	May 19

[a] Taken from Borneff & Kunte, 1964.

[b] The 2 untreated samples were taken at Mainz on the same day with an interval of 6 hours. The treated sample was taken at an unknown location 2 months later. Therefore, the treatment effect cannot be directly evaluated.

[c] The locations from which the various samples were collected are unknown, and the sampling dates varied. Therefore, the treatment effect cannot be directly evaluated. However, it is known that the untreated sample and the treated second sample were collected on the same day.

[d] No satisfactory explanation can be given to the high values.

TABLE 15

CONCENTRATION OF PAH IN BITUMEN AND CONTACT-WATER[a]

Sample	Concentration (μg/kg, and μg/litre respectively)[b]		
	BP	Carcinogenic PAH	Total PAH
Bitumen	100	3 400	3 500
Water: Sample I[c]	0.0175	0.090	0.200
Sample II[d]	0.0035	0.043	0.110

[a] Taken from Borneff & Kunte, 1965.

[b] Detailed tables with figures for the various carcinogenic and non-carcinogenic PAH are presented in Borneff & Kunte, 1965.

[c] The sample was taken from distilled water after 10 days in contact with a coating of bitumen.

[d] After Sample I was taken, the bitumenous coating was continuously washed for 4 weeks with water before a fresh distilled batch of water was put in for 10 days.

were not particularly effective in reducing the concentrations of these materials, while the activated carbon was primarily responsible for their reduction in the Rhine.

River water, after seeping through the river-bank soil, still contains PAH, probably because the filtration does not efficiently remove colloidal materials (Mallet, 1965, 1966). PAH have been recovered from activated carbon used to filter such river-bank-filtered water (Borneff & Fischer, 1962a; Borneff, 1964a). Similarly, the efficiency of rapid sand-filtration is not high; yet the examination of filter mud from backwash water of Bodensee rapid sand-filters indicates a high PAH concentration, thus showing some removal (Table 16). Although it is generally true that the better the sedimentation of the suspended solids, the better the water quality, it has to be kept in mind that no significant reduction of PAH in water should be expected because these compounds are mostly associated with particles too small to settle in a simple clarification process (Borneff & Fischer, 1963; Borneff & Kunte, 1965). On the other hand, flocculation of raw river water was found more efficient in removal of PAH (Lawerenz, 1967).

A number of laboratory studies were conducted on the effectiveness of oxidizing agents, such as chlorine and ozone, on the reduction of BP and other carcinogenic PAH in water. The studies using chlorine (Gräf & Nothhaft, 1963; Trakhtam & Manita, 1966) (Table 17), indicated relatively slow removal compared with the typical disinfection time of ½ hour by chlorine. Thus, while some reduction of BP is achieved, the efficiency of this treatment is questionable. Chlorination does not significantly increase the removal of PAH in lake-water treatment (Borneff, 1964a; Borneff & Kunte, 1964) (Table 14). On the other hand, experiments with gaseous chlordioxide in doses and contact periods commonly practised in water treatment indicated a very high percentage of BP removal, and yielded non-carcinogenic products (Reichert, 1968a, 1968b, 1968c, Borneff, 1969).

Similarly, the treatment of a BP aqueous solution with ozone-enriched air achieved a reduction of 99% within 30 minutes. However, interference in the process efficiency by organic trace contaminants has been observed (Borneff, 1969). The removal of PAH

TABLE 16

PAH RETAINED ON ACTIVATED-CARBON AND RAPID-SAND-FILTER MUD

Source of PAH	Concentration (μg/kg of dry sample)[a]			Reference
	BP	Carcinogenic PAH	Total PAH	
Activated-carbon	1	150	8 700	Borneff & Fischer, 1962a
The coarse settled particles from the backwash water of a rapid sand filter, collected from a sedimentation basin	50	1 250	15 000	Borneff & Fischer, 1962b
The centrifuged particles (down to 1μ) from the backwash water of a rapid sand filter	200	3 200	6 200	Borneff & Fischer, 1963

[a] Detailed tables with figures for the various carcinogenic PAH are presented in Borneff & Fischer, 1962a, 1962b, 1963.

TABLE 17

EFFECT OF CHLORINATION ON BP CONCENTRATION

Initial chlorine dose (mg/litre)	Initial BP concentration (μg/litre)	Time (h) needed to reduce BP concentration to:					Reference
		1/2 value	1/5 value	1/8 value	1/15 value	Zero	
0.3	5	3	—	—	—	—	Gräf & Nothhafft, 1963
0.5	5	2	20	—	—	—	
0.5	2	2	—	—	—	13	
0.3	1	—	1/2	—	2	—	Trakhtman & Manita, 1966
0.5	1	—	—	1/2	1	—	

from drinking-water using ozone was further discussed by Reichert (1969). When comparing the degradation achieved for a number of examined carcinogenic PAH, BP was found to be the most resistant (Ilnitsky, 1968). The resistance of BP to degradation depended on its state, and was greater when it was adsorbed on soil particles than when in solution.

Filtration through activated carbon is the most effective conventional process for the removal of PAH. Furthermore, the examination of fresh activated carbon did not show the presence of PAH (Borneff & Fischer, 1961a), indicating that in this respect it is suitable for use in water treatment (Borneff, 1964a). Studies on the use of activated carbon for the treatment of river-bank-filtered water indicate that the carbon removed approximately 99% of the PAH (Borneff & Fischer, 1962a).

It has been previously noted that BP and other PAH are sensitive to light of various wavelengths. It is thus likely that ultraviolet irradiation as a water treatment process will reduce the concentrations of these materials.

As a significant part of the carcinogenic PAH may be present in raw water while adsorbed on particles or in suspension, mechanical treatment such as sedimentation and filtration as well as flocculation may have some effect on their removal. Adsorption on activated carbon has been found most effective. The best over-all results may be achieved from combined processes including both mechanical and chemical ones. By such methods removal of up to 99% has been achieved. However, no known method has yet been found to reduce the concentration of carcinogenic PAH below about 0.01 μg/litre, their common concentration in non-polluted ground-water (Reichert, 1968; Borneff, 1969).

TABLE 18

REDUCTION OF PYRENE BY A SEWAGE TREATMENT[a]

PY concentration (μg/litre)			% reduction
In raw sewage	In feed water to sand filter	In final effluent	
1.00		0.36[b]	64
0.43		0.15–0.23[c]	46–65
	0.51	0.45	12

[a] Taken from Wedgwood & Cooper, 1956.

[b] From an efficient trickling filtration sewage-treatment plant using recirculation.

[c] From an overloaded trickling-filtration sewage-treatment plant.

PAH in significant concentrations are found in effluents from municipal waste-water treatment plants, partially due to the slow rate of oxidation in biological processes (Wedgwood & Cooper, 1956), indicating the limited efficiency of the treatment processes. Table 18 presents data concerning the reduction of pyrene in sewage by trickling-filter treatment plants. The removal is not high. The activated-sludge process when studied in the laboratory did not show any significant ability to oxidize carcinogenic PAH even after 144 hours (Lutin et al., 1965).

The effect of dephenolization of shale-oil waste-water on the BP concentration was investigated (Gortalum & Dikun, 1958; Dikun & Makhinenko, 1963; Veldre et al., 1965a) (Table 19). It can be concluded that in many cases complete removal of BP is not achieved and additional treatment is necessary. However, filtration through a coke bed effects a high degree of removal.

TABLE 19

REDUCTION OF BP BY INDUSTRIAL EFFLUENT TREATMENT PROCESSES

Type of treatment	BP concentration (μg/litre)		% removal	Reference
	Before treatment	After treatment		
Dephenolization	10 900	310 [a]	97	Dikun & Makhinenko, 1963
Dephenolization	200	5	97.5	Gortalum & Dikun, 1958
Dephenolization	5–20	2	60–90	Veldre et al., 1965a
Dephenolization	44–64	12–16 [b]	75	Fedorenko, 1964
Filtration through coke bed [c]	1 000	20	98	Wedgwood & Cooper, 1956

[a] A butyl acetate process was used for dephenolization.

[b] A biological process was used for dephenolization.

[c] Similar removal efficiencies were achieved for AC and PY.

HUMAN EXPOSURE TO PAH

Before considering the possible effects of PAH on the health of man, it is useful to consider the quantities to which he is exposed. It has been postulated that as a result of the likely world-wide synthesis of PAH by plants, as well as their other origins, there exist environmental concentrations of BP of the order of magnitude of 10 μg/kg–20 μg/kg of dry organic substance (Gräf & Diehl, 1966). BP, although a most potent carcinogen, represents only a relatively small portion of the total carcinogenic PAH found in environmental samples. The majority of investigators have dealt only with BP or another single compound, and only a few studies touched on a variety of carcinogenic PAH in the same environmental samples (Borneff & Fischer, 1962a, 1962b, 1962c, 1963; Borneff, 1964a; Borneff & Kunte, 1964, 1965, 1967). The latter studies led to the conclusion that there is no correlation between the concentration of BP and that of the total carcinogenic PAH in the environmental samples. However, BP does generally constitute between 1% and 20% of the total carcinogenic PAH (Tables 3, 9, 10, 12, 14, 15 and 16).

It has been previously noted that in one series of studies the general range of concentration of carcinogenic PAH in ground-water and treated surface water is between 0.001 μg/litre and 0.025 μg/litre (Table 4). This may be taken as a likely range of concentration in drinking-water supplies, although higher values have been reported (Table 13).

There have been numerous reports on the pollution of air by BP and other carcinogenic PAH (for example, Hoffmann & Wynder, 1962). Such substances may occur in air, sorbed on to a variety of particulate materials, and be inhaled by man. Carcinogenic PAH have also been identified in tobacco smoke (Bentley & Burgan, 1958; Wynder & Hoffmann, 1959). It has also been found that carcinogenic PAH occur in fresh and air-cured tobacco leaves indicating that combustion is not required for their production (Cooper & Campbell, 1955; Campbell & Cooper, 1955; Campbell & Lindsey, 1956, 1957; Bentley & Burgan, 1958, 1960; Shanta & Krishnamurthi, 1963; Schmähl, 1964). Man may also be exposed to carcinogenic PAH from the use of unsmoked tobacco as snuff. Examples of levels of BP in such materials are 54 μg/kg in a mixture of pipe tobacco and 270 μg/kg in Zulu snuff.

River and marine sediments, suspended solids, zoo- and phytoplankton and worms, have all been shown to contain various amounts of carcinogenic PAH (Tables 5, 6, 8, 10 and 11). Aqueous biota serve as a food and source of PAH for edible fish and shellfish (Table 7), from which the PAH may reach man. BP has been found in fish from the river Seine (Depuis, 1960) codfish (Mallet, Perdrian & Perdriau, 1963a), bluefish (Smith, 1954), and sardines (Bourcart & Mallet, 1965). PAH have also been detected in various oysters and edible molluscs (Cahnmann & Kuratsune, 1957; Mallet et al., 1960; Mallet, 1961; Mallet et al., 1963b; Mallet & Schneider, 1964; Perdriau, 1964b; Bourcart & Mallet, 1965).

Carcinogenic PAH have been extracted from a large variety of fresh plants, and were detected in considerable amounts in vegetables, salads, grains, fruits and edible mushrooms (Gräf, 1964, 1965; Grimmer & Hildebrandt, 1965b, 1965c, 1966; Grimmer, 1966; Gräf & Diehl, 1966), as well as in vegetable oils (Jung & Morand, 1962, 1963; Ciusa et al., 1965; Ramel et al., 1965; Borneff & Fábián, 1966; Craft & Norman, 1966; Howard et al., 1966b; Le Clerc et al., 1966; Biernoth & Rost, 1967, 1968; Grimmer & Hildebrandt, 1967a, 1968; Fábián, 1968a); from these reports it appears that the BP content of active plant tissue is in the range of 10 μg/kg–20 μg/kg. In coconut oil its content may rise to about 50 μg/kg while in most other examined vegetable oils BP content was found to amount to only a few μg/kg.

Numerous reports have been published on the existence and concentration of carcinogenic PAH in various fried, grilled, roasted and smoked fish and meat products as well as in coffee, with particular attention being given to BP (Bailey & Dungal, 1958; Gorelova & Dikun, 1958a, 1958b, 1961, 1965; Kuratsune & Hueper, 1958, 1960; Dansi & Zanini, 1959; Dungal, 1959, 1961a, 1961b; Gorelova et al., 1959, 1960, 1963a, 1963b; Nugmanov et al., 1961; Shabad, 1961; Lijinsky & Shubik, 1964, 1965a, 1965b; Dikun, 1965a, 1965b; Ramel et al., 1965; Grimmer, 1966; Grimmer & Hildebrandt, 1966, 1967a, 1967b; Howard et al., 1966a, 1966c; Le Clerc et al., 1966; Kuratsune et al., 1966; Masuda et al., 1966; Sigurjonsson, 1966a, 1966b; Lijinsky, 1967; Fábián, 1968b, 1969; Thorsteinsson & Thordarson, 1968; Kraybill, 1969; Thorsteinsson, 1969). Levels of BP in such processed foods are, for example, 37 μg/kg in smoked fish and 200 μg/kg in coffee soot.

A recent review (Haenni, 1968) discusses the scientific rationale and the analytical principles underlying control in the United States of America of the contamination of the food supply by potentially carcinogenic PAH arising from the use of food additives. The review supplements and brings up to date an earlier paper by Gunther & Buzzetti (1965) which is even broader in scope.

HEALTH CONSIDERATIONS

In order to consider the possible effect of PAH on man, it is pertinent to discuss their relative carcinogenicities. For the most part conclusions as to the potency of these carcinogens are based on laboratory animal experiments only, although one indirect and two direct applications of BP to the human skin have been reported (Klar, 1938; Cottini & Mazzone, 1939; Rhoads et al., 1954). The potency of these substances varies with the species. It is believed that some PAH are indeed carcinogenic to man, the most ubiquitous and potent one being possibly BP (Falk et al., 1964). On the other hand, the importance of weak carcinogens has been emphasized, as these may be the significant agents in human carcinogenesis (Kennaway, 1955).

A number of attempts have been made to compare the potency of the various carcinogenic PAH. Iball (1939) has introduced the " carcinogenic index ", defined by the percentage of tumours produced over the average latent period in days. Berenblum (1945) has suggested a series of 12 " carcinogenic grades ". For simplicity Badger's system involved 4 grades of carcinogenicity (Badger, 1948). More recently a " relative activity index " has been developed using 3 grades only (Wynder & Hoffmann, 1959; Hoffmann & Wynder, 1962). Table 1 lists a variety of PAH graded by their method. In all the previously reported methods the grading has been based on the relative potency of the carcinogens to the skin of mice, BP being among the most active.

BP, although a potent carcinogen, constitutes only a relatively small portion of the total carcinogenic PAH found in environmental samples. Most investigators have determined the levels of a single, or at most a few, carcinogenic PAH in environmental samples. The work of Borneff indicates that BP generally constitutes between 1% and 20% of the total carcinogenic PAH (Tables 3, 9, 10, 12, 14, 15 and 16).

Although being carcinogenic to animal tissue, PAH do not produce carcinomas or cell changes in plants (Manil & Straszewska, 1950; Blochinger, 1961; and others). Further, it has previously been noted that PAH may promote the growth of plants and that this characteristic may be related to their carcinogenicity in animals (Gräf, 1964, 1965).

As some derivatives of BP and other carcinogenic PAH are formed during disinfection of water with oxidizing agents and ultraviolet radiation, and through various biochemical reactions, it is of interest to briefly examine the carcinogenicity of such derivatives. Except with the addition of methyl groups and homologous alkyl derivatives, most BP derivatives at most have weak carcinogenic activity (Butenandt & Dannenberg, 1956). However, 10-chloro compounds do cause tumours. 5-Chlorobenzpyrene, formed during chlorination (Reichert, 1968a, 1968b), was first suspected to be carcinogenic

(Gräf & Nothhafft, 1963) but proved in later experiments not to be so (Müller & Reichert, 1969). The quinones, on the other hand, some of which are also formed during chlorination (Reichert, 1968a, 1968b), do not produce tumours (Butenandt & Dannenberg, 1956), and may inhibit other carcinogens (Buu-Hoï, 1959).

There are very few studies concerning the statistical correlation between cancer and drinking-water. Furthermore, no epidemiological evidence is available as to the excessive liability to cancer of a population obtaining its drinking-water from bodies of polluted raw-water (Hueper & Conway, 1964). It was, nevertheless, noted that 4 London boroughs, supplied largely by well-water, had lower cancer mortalities than most of the other boroughs supplied with river water (Stocks, 1947). Another study concluded that the highest cancer death rates are for communities supplied by river water, followed by well water, and heath water (Diehl & Tromp, 1954; Tromp, 1955). None of these studies attempted to correlate cancer morbidity with concentrations of PAH. Finally it should be noted that one epidemiological study of the incidence of gastric cancer concluded that social factors and the kinds of soils present reduced the correlations otherwise obtained with the type of domestic water supply (Wynne-Griffith & Davies, 1954; Davies & Wynne-Griffith, 1954).

Although many experiments to determine the carcinogenic activity of BP have been conducted on various laboratory test animals, it is not within the scope of this review to summarize, compare or evaluate the experimental results. Yet it should be noted that the possible distribution in the body of ingested BP has been demonstrated (Poel, 1963); when fed to mice the BP was absorbed from the alimentary tract and distributed to various body organs. Fore-stomach tumours have developed in some mice within a year when administered with as little as 0.0625 mg of BP per dose twice weekly in 0.1 ml Tween-60 by gastric intubation. However, differences in the carcinogenic susceptibility to BP among various species of test animals have been observed. For example, BP is highly potent for the skin of mice, but only weakly active for that of rabbits (Hueper & Conway, 1964). Therefore, it is not possible to deduce the effect of BP on humans from the results of experiments on animals.

Experimental work with mice, studying the relative tumour yield obtained with subcutaneous injections of various amounts of BP (Payne & Hueper, 1960), has further confirmed "... the general impression that repeated exposure to carcinogenic chemicals are more effective than an equivalent single exposure. ... Repeated doses of a carcinogen are, moreover, bound to overcome any modifying or inhibiting influences which any constitutional and genetically dependent factor of the host organism may exert on the development of the cancerous response" (Hueper & Conway, 1964). Therefore, the experiments described (Payne & Hueper, 1960; Poel, 1963; etc.) indicate the potential danger of continuous exposure to carcinogenic PAH introduced into the gastro-intestinal tract by water and food, even at low concentrations. However, it can be assumed that a high percentage of the carcinogenic PAH or their metabolites will be eliminated quickly through the urine and faeces (Chang, 1943; Weigert & Mottram, 1946a, 1946b; Gerarde, 1960).

It should be kept in mind that the usual relationship of effective doses of toxic compounds in proportion to body weight may not necessarily hold for carcinogens because a tumour can develop at the point of contact with the carcinogenic material. Therefore, the probability of cancer being produced in the human should not be directly related to animal size (Gerarde, 1960).

Until recently, threshold limits for carcinogenic PAH have not been determined (Borneff & Fischer, 1962b), and many investigators believe that even the slightest exposure will cause irreversible effects (Ilnitsky & Varshavskaya, 1964). It is thus observed that carcinogens differ from some other toxic agents in that no safe level exists (Druckrey, 1954; Boyland, 1958). Therefore, it has been suggested that the threshold limit be set practically at zero (Gerarde, 1960). However, this recommendation is negated by the presence of carcinogenic PAH in so many plant tissues as to render the setting of such a limit impracticable.

Man is exposed to carcinogenic PAH from many sources. A comparative study of their relative importance has, however, not yet been done. It has been suggested that the carcinogenic material ingested by man from drinking-water should not surpass a tenth of this material taken-up from normal urban air. This would limit the maximum permissible concentration (MPC) in water to 0.017 μg/litre, or with a daily consumption of 2.5 litres of drinking water, to about 15 μg yearly, and about 1 mg in a lifetime (Borneff & Fischer, 1962b; Kruse, 1965). The first recommendation has recently been made, concerning PAH in drinking-water supplies, proposing the normal concentration of 6 PAH, selected for easy determination, to be 0.1 μg/litre, and limiting the maximum recommended concentration (MRC) to 0.2 μg/litre (Borneff & Kunte, 1969; World Health Organization, 1970). Of the 6 selected PAH only 3 are carcinogenic (3,4-BF, BP and IP) with an MRC of about 0.03 μg/litre, of which BP comprises about one fourth. Calculations based on 4 drinking-water samples (Table 13) indicate an annual human consumption of carcinogenic PAH of about 6 μg, 9 μg, 22 μg, and 70 μg, respectively, for the population served.

It may be estimated that bank-filtered water from the Rhine would result in a yearly total ingestion of 50 to 500 μg of carcinogenic PAH (Borneff & Fischer, 1962a). Similarly, a river water source containing 0.1 μg/litre of BP would result in 90 μg per year (Dikun & Makhinenko, 1963). However, any current limits set on the amounts of these materials should be taken with reservation because they are not based on knowledge of their toxic effects.

Although currently the consumption of PAH-containing water is probably not dangerous, the combination of sources cannot be excluded as a potential hazard. When one considers that animal experiments have shown that repeated exposure to carcinogens is more effective than an equivalent single dose, one should not neglect the possibility of cancer from the repeated life-long exposure to carcinogens in air, food and water. Kotin (1958) said that repeated observations on the cumulative effect of carcinogens strongly suggest that given enough time and a sufficient exposure in the presence of other pathogenetic factors, the inevitability of cancer formation is one of the few certainties of life.

Therefore, the presence of PAH carcinogens in water, food and air is undesirable. Moreover, because of the technically unavoidable contamination from background, consideration should be given to avoiding increasing the level of carcinogenic PAH in food, such as from additives or processing, as well as limiting where practicable their concentrations in water and air.

CONCLUSIONS

The report of the WHO Expert Committee on the Prevention of Cancer (1964) touches on the environmental aspects and states:

> Effective measures are needed to prevent the introduction of carcinogenic industrial wastes into the atmosphere and into public waters serving as sources of drinking-water supply. Such precautions are particularly necessary when these wastes are stable and cumulative. Although at present no clear evidence exists that such carcinogenic industrial contaminants of air and water have become an actual environmental cancer hazard to the general population, they should be viewed with serious concern, so as to forestall such complications in the future.

The conclusions that can be drawn from reviewing the literature are as follows:

(1) The health significance of traces of carcinogenic PAH in drinking-water is not yet known.

(2) With the present trend to reduce the pollution of surface water, attention should be given to reducing the amounts of carcinogenic PAH.

(3) Good ground-water may contain on the average 0.001 μg/litre–0.01 μg/litre of carcinogenic PAH, probably leached from the soil by infiltrating surface water, thus making such concentrations technically unavoidable. However, waters with higher concentrations, such as in lakes and rivers, should be examined for their carcinogenic content when considering their use as raw supplies.

(4) Detergents and mineral oils in surface water may affect solubility of PAH. Generally, however, the present concentration of detergents is probably too low to have a significant effect.

(5) River-bank filtration generally does not remove enough carcinogens, nor does sedimentation, rapid sand-filtration, or chlorination. On the other hand, activated-carbon filtration at a sufficiently low filtration rate eliminates these carcinogenic substances to a great extent. Treatment with chlordioxide or ozone also appears to be promising. Because most current treatment processes do not free the water of carcinogenic PAH, these processes, as well as new methods, should be examined and developed with a view to improving their performance.

(6) The difference in concentration of PAH carcinogens naturally present in water (from soil, plants and plankton) on the one hand, and in contaminated effluents (of municipal and industrial origin, and road run-off) on the other, suggests that PAH are indicators of water contamination.

(7) Because of the technically unavoidable natural background of carcinogenic PAH to which man is exposed from water, food and air, and due to the probably cumulative effect of those substances, the introduction of carcinogenic additives in food, such as dyes and preservatives, should be avoided, or at least limited as much as possible.

RÉSUMÉ

LES HYDROCARBURES AROMATIQUES POLYCYCLIQUES DANS LE MILIEU, ET EN PARTICULIER DANS L'EAU

On sait que beaucoup d'hydrocarbures aromatiques polycycliques sont cancérogènes pour l'animal, et probablement aussi pour l'homme. L'étude actuelle passe en revue les divers aspects du problème que pose la présence dans l'eau de ces composés, parmi lesquels le benzo-3,4 pyrène retient particulièrement l'attention en raison de son ubiquité, de son pouvoir cancérogène élevé et des nombreuses recherches qui lui ont été consacrées.

Les hydrocarbures aromatiques polycycliques sont généralement le produit de la combustion et de processus exigeant de très hautes températures, mais leur omniprésence dans le milieu amène à considérer comme vraisemblable leur synthèse par les plantes et les micro-organismes. On les trouve dans des conditions excluant toute contamination résultant d'activités humaines, dans les forêts vierges, les boues, les sédiments et dans la flore et la faune de beaucoup d'eaux naturelles. Bien que spontanément très peu solubles dans l'eau pure, ils peuvent être solubilisés par certaines substances comme les détergents et les huiles minérales. Ils peuvent être véhiculés à distance après avoir été adsorbés sur des éléments de la faune et de la flore aquatiques. Une de leurs particularités est d'être sensibles à la lumière.

De multiples activités entraînent la production d'hydrocarbures aromatiques polycycliques et les effluents industriels en déversent de grandes quantités dans le milieu. Les eaux d'égout en contiennent, de même que l'urine humaine. L'examen d'eaux de diverses origines destinées ultérieurement aux usages domestiques montre que la concentration des hydrocarbures cancérogènes est minimale dans les eaux souterraines et qu'elle s'accroît dans les eaux traitées et surtout dans les eaux de surface. La plupart des techniques classiques de traitement des eaux (filtration rapide sur sable, chloration, traitement par l'ozone) n'ont qu'une efficacité limitée en ce qui regarde l'élimination de ces composés. La filtration sur charbon activé donne les meilleurs résultats.

Il est impossible de mettre l'homme complètement à l'abri des effets des hydrocarbures aromatiques polycycliques qui sont partout présents dans le milieu. Ils représentent donc une menace potentielle, bien que les concentrations décelées jusqu'à présent ne paraissent pas dangereuses.

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[1] If not otherwise marked, the reference is published in English. For reasons of simplicity, PAH substances in the reference titles are referred to as follows: PAH—polycyclic/nuclear aromatic hydrocarbons; PH—polycyclic/nuclear hydrocarbons; PA—polycyclic/nuclear aromates; AH—aromatic hydrocarbons; BP—3,4-benzpyrene or benzo(a)pyrene.

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