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# DETERMINATION OF POLYCYCLIC AROMATIC HYDROCARBONS IN AUTOMOBILE EXHAUST BY MEANS OF HIGH-PERFORMANCE LIQUID CHROMATOGRAPHY WITH FLUORESCENCE DETECTION

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## SUMMARY

A chromatographic method has been developed and applied to the determination of polycyclic aromatic hydrocarbons (PAHs) in particulate matter in automobile exhaust, in petrols, and in crankcase oils. The PAHs were purified from other organic compounds by thin-layer chromatography, separated by high-performance liquid chromatography, and measured by means of on-line fluorescence detection. The identities of the PAHs were verified by comparing the emission spectra obtained by a stop-flow technique with those of standard PAHs.


## INTRODUCTION

The increase in the incidence of lung cancer in the past 50 years has focused attention on pollution by polycyclic aromatic hydrocarbons (PAHs)[1,2], because some of these have been shown to produce skin cancers and sarcomas in mice[2]. Estimates have been made of the nature of the sources and their emission of PAHs, and of the exposure of the population[3,4]. Epidemiological studies have documented that the incidence of lung cancer is higher among smokers than non-smokers[1,2], and higher among coke-oven workers than the general population[1]. In spite of the dominating effect of smoking, a number of studies have shown that the higher degree of atmospheric pollution in urban areas is apparently partly responsible for the higher rates of death from lung cancer among individuals living in cities compared with individuals from rural areas, though the precise degree of the effect of atmospheric pollution remains to be determined[1]. As the content of PAHs is higher in the urban atmosphere[5], the need for sensitive methods of determining PAHs in airborne particulate matter from different sources is evident.

Several methods for determining benzo[*a*]pyrene (B*a*P) and other PAHs have been described[6–10]. Much recent work has used gas chromatography with highly efficient glass columns or capillary columns to separate the PAHs, which are then measured by a flame ionization detector or a mass spectrometer[10–17].

The recent rapid development of high-pressure pumps and microparticulate columns for high-performance liquid chromatography (HPLC) has resulted in a

steadily growing interest in applying this technique[18]. HPLC has mainly been used in connection with on-line UV absorbance detection, *e.g.* for the analysis of PAHs in airborne particulate matter[19–22], smoke condensates[23–25], tars[10,26], and mineral oils[27,28]. Although not all compounds are well resolved into individual peaks by this method, measurements of the absorbance at two or more wavelengths have made it possible to determine the amounts of the most important PAHs[22,26].

The development of on-line fluorescence detectors seems to offer possibilities of an improved procedure[29,30]. Fluorescence detectors are generally more sensitive and therefore require smaller samples and less sampling time. Furthermore, by changing the fluorescence conditions, selectivity between the various compounds can be achieved[29,30], and hence it is easier to analyse poorly resolved components. Few publications have dealt with the analysis of PAHs using the combination of HPLC and fluorescence detection in actual samples (airborne particulate matter[31], sediments[32], petrols[33], engine oils[33–35], and water[36]). The detection procedure is normally hampered by the finding that, for example, air samples contain fluorescent compounds other than PAHs. The PAH fraction must be separated from these compounds in order to avoid a high background fluorescence. The PAHs are purified from other organic compounds by thin-layer chromatography (TLC), separated by HPLC and measured by on-line fluorescence detection, and the method has been applied to samples of particulate matter in automobile exhaust gases, to petrols, and to lubricating oils.

## EXPERIMENTAL

### *Chemicals*

Reference compounds (see Table I) were obtained from different suppliers. Each compound was checked by comparing its UV absorption spectrum with those in the literature[37]. The methanol and cyclohexane used in this study were from Ferak (p.a.; Berlin, G.F.R.); the hexane, toluene, ether, and dioxane from Merck (p.a.; Darmstadt, G.F.R.). The cyclohexane was purified by elution through a column packed with aluminium oxide (aluminia Woelm N; Super I). No interfering fluorescence was observed in the analysis of blank samples.

### *The equipment for HPLC detection*

The equipment consisted of a Waters pump 6000 A, a Rheodyne 7120 sample injector with a 20 $\mu$l loop, a Nucleosil® 5 $C_{18}$ precolumn 6 cm × 0.46 cm, a Zorbax™ ODS column 25 cm × 0.46 cm, and a Perkin-Elmer LC 1000 fluorescence detector. The chromatogram was displayed on a Kipp-Zonen BD 41 recorder. The eluent (methanol–water, 8:1) was filtered through a mobile-phase filter before entering the pump. A Rheodyne column inlet filter was placed between the injection loop and the pre-column, and the samples were filtered through a 0.2-$\mu$m Millipore filter FG before injection, in order to prevent the deterioration of the columns as a result of particles in the injected solutions.

### *Extraction*

The particulates in automobile exhaust gases were separated and collected in two fractions. One was collected on cyclones of stainless steel and mainly contained particles with a diameter greater than 1 $\mu$m; the other was collected on glass-fibre

TABLE I

RETENTION TIME AND SENSITIVITY DATA FOR PAHs

Pre-column, Nucleosil® 5 $C_{18}$ 6 cm × 0.46 cm; main column, Zorbax™ ODS 25 cm × 0.46 cm. Solvent, methanol–water (8:1) at 1.0 ml/min; pressure, 1700 p.s.i.; temperature, 21°.

| *Compound* | *Relative retention time** | *Detection limits (ng)*** | |
|---|---|---|---|
| | | *340/425 nm* | *363/435 nm* |
| Phenanthrene | 0.93 | 5 | 1000 |
| Anthracene | 1.00 | 0.2 | 0.2 |
| Fluoranthene | 1.20 | 0.6 | 0.4 |
| 1-Methylanthracene | 1.31 | 0.3 | 0.2 |
| 9-Methylanthracene | 1.34 | 0.2 | 0.2 |
| Pyrene | 1.35 | 0.06 | 0.2 |
| 2-Methylanthracene | 1.42 | 0.4 | 0.4 |
| 2,3-Benzofluorene | 1.69 | 3 | 200 |
| Benzo[*a*]anthracene | 1.75 | 0.3 | 0.7 |
| Chrysene | 1.76 | 7 | 50 |
| Triphenylene | 1.76 | 100 | — |
| Benzo[*ghi*]fluoranthene | 1.78 | 0.3 | 0.7 |
| 9,10-Dimethylanthracene | 1.83 | 0.2 | 0.09 |
| Benzo[*j*]fluoranthene | 2.2 | >2000 | >2000 |
| Benzo[*e*]pyrene | 2.4 | 1 | 30 |
| Perylene | 2.5 | 6 | 0.3 |
| Benzo[*k*]fluoranthene | 2.6 | 0.2 | 0.2 |
| Benzo[*a*]pyrene | 2.9 | 0.1 | 0.05 |
| 1,2,3,4-Dibenzanthracene | 3.1 | 2 | 10 |
| 1,2,5,6-Dibenzanthracene | 3.6 | 0.5 | 3 |
| Benzo[*ghi*]perylene | 4.3 | 0.3 | 0.2 |
| Indeno[1,2,3-*c*,*d*]pyrene | 4.6 | 20 | — |
| Coronene | 5.7 | 40 | 50 |

* Anthracene *ca.* 11.5 min.

** Considered to be the amount injected to give peak a with a height double that of the random baseline noise level.

filters (Whatman G-FA) and consisted mainly of particles of less than 1 $\mu$m in diameter. The PAHs on the filters and the cyclones were extracted ultrasonically in cyclohexane[38]. Cyclohexane was preferred to other organic solvents because it was expected that these would dissolve undesirable polar compounds to a higher degree. The collected extracts were dried over sodium sulphate, filtered and concentrated in a rotavapor to a few ml. The rest of the cyclohexane was evaporated to 0.1 ml at 35° in a stream of nitrogen.

*Petrols and lubricating oils*

A 10.0-ml volume of the special petrol was evaporated to 0.3 ml in a stream of nitrogen, and this was used in the following clean-up procedure; 100 $\mu$l of crankcase oil was diluted with 10 ml cyclohexane, and the solution was purified by the liquid–liquid extraction method described by Grimmer *et al.*[39]. The cyclohexane extract was dried over sodium sulphate, filtered and concentrated to 0.1 ml.

*Thin-layer chromatography*

The TLC pre-fractionation procedure was a modification of that used by

Brocco *et al.*[40]. The TLC plates (20 cm × 20 cm, 0.25 mm thick, from Macherey-Nagel SIL G-25 HR) were activated at 100° for 1 h. The concentrated extract was applied as a band (6–8 mm wide), and a standard PAH mixture consisting of anthracene, 1-methylanthracene, fluoranthene, B*a*P, and benzo[*ghi*]perylene was spotted on to each TLC plate. The plates were developed by ascending elution in hexane, and air-drying in the dark for 5 min, followed by elution in toluene–cyclohexane (1:1). It is advantageous to elute the plates in hexane first in order to avoid overloading if the content of non-volatile, non-polar compounds is high. By means of the elution in the toluene–cyclohexane mixture, the PAH fraction was separated from up to six other fractions of fluorescent compounds. The compounds in the six bands were not identified, but tests with known standards indicated that they may consist of nitrogen and oxygen heterocycles and substituted PAHs (phenols and anisoles). The silica gel with the PAH fraction was scraped off the plate and eluted twice with 3 ml diethyl ether. The combined extracts were filtered, concentrated to 0.1 ml in a stream of nitrogen, and diluted with a mixture of methanol and dioxane (3:2).

*High-performance liquid chromatography*

High efficient separations of the PAHs are achieved with HPLC on an octadecylsilyl (ODS)[20–22,28,30,31,35], or an octadecyldimethylsilyl (RP-18)[36] stationary phase of microparticles (5–7 μm) together with a polar mobile phase. A decrease in the capacity of the column after prolonged use has been reported[36], but the reasons for this are not clear[41]. Therefore, a short protective pre-column was introduced: following the described procedure, 600 analyses resulted in no decrease in the capacity. The separation of the PAHs was improved by the introduction of the pre-column. Figs. 1 and 2 show the separation of eight standard compounds. Fig. 3 shows that separation

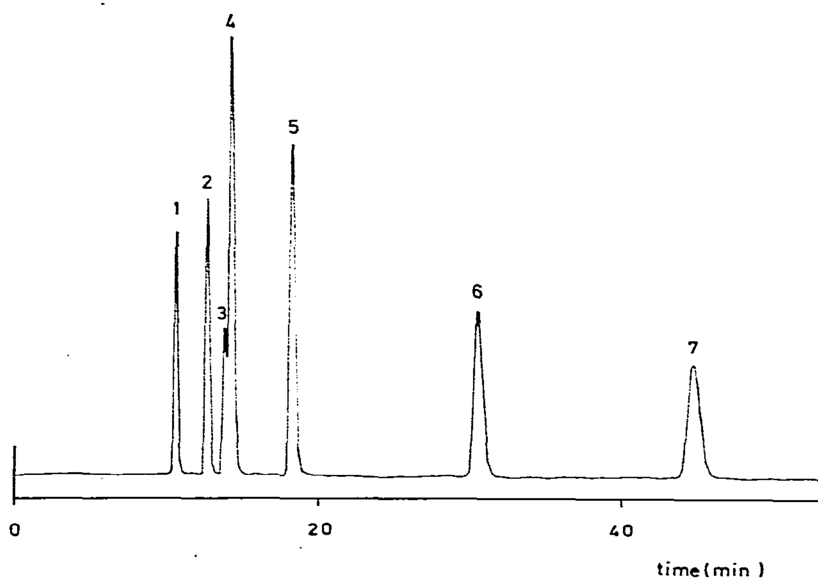


Fig. 1. Chromatogram of a standard PAH mixture at 340/425 nm. For the chromatographic conditions, see Table I. Peaks: 1 = anthracene; 2 = fluoranthene; 3 = 1-methylanthracene; 4 = pyrene; 5 = benzo[*a*]anthracene; 6 = benzo[*a*]pyrene; 7 = benzo[*ghi*]perylene.

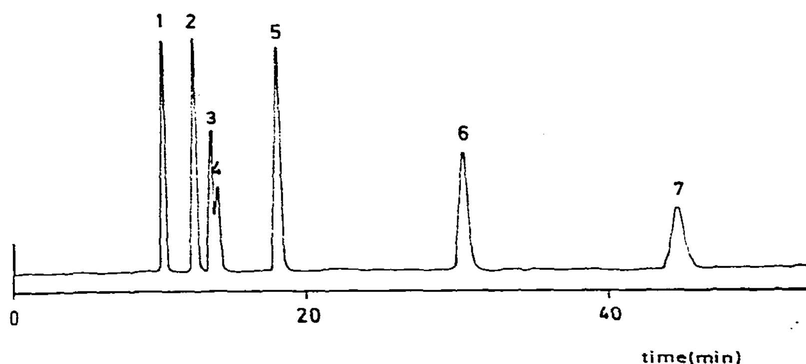


Fig. 2. As Fig. 1, except that the solvent is not deoxygenated.

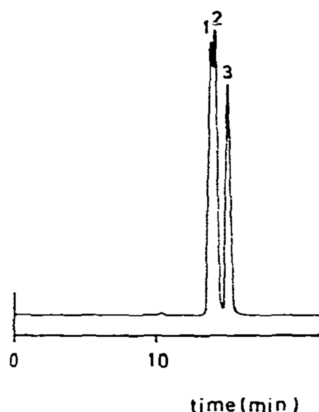


Fig. 3. Chromatogram of the three methylanthracenes at 363/435 nm. For chromatographic conditions, see Table I. Peaks: 1 = 1-methylanthracene; 2 = 9-methylanthracene; 3 = 2-methylanthracene.

of even the isomers 1-, 2-, and 9-methylanthracene can be achieved. Table I lists the relative retention times for 23 PAHs.

The characteristics of the chromatographic system were as follows:
The capacity factor of benzo[*ghi*]perylene was

$$k' = \frac{V - V_0}{V_0} = 14.2$$

where $V$ is the elution volume and $V_0$ the void volume.

The theoretical plate number for B*a*P was

$$N = 16 \left(\frac{V}{W}\right)^2 = 13{,}000$$

where $W$ is the peak volume.

The resolution between the isomers benzo[*e*]pyrene and B*a*P was

$$R = \frac{V_2 - V_1}{{}^1/_2 (W_1 + W_2)} = 4.7$$

*Fluorescence*

*Oxygen quenching.* The mobile phase was deoxygenated by bubbling argon through the eluate for at least 1 h, deaerated ultrasonically and kept under an argon atmosphere that was slowly renewed. Otherwise, the presence of small amounts of oxygen in the mobile phase would quench the fluorescence[7,31], especially of pyrene (see Figs. 1 and 2). The response of the standard mixtures thus did not change by more than a few per cent throughout a working day.

*Detection.* The fluorescence of the PAHs was measured with an excitation at

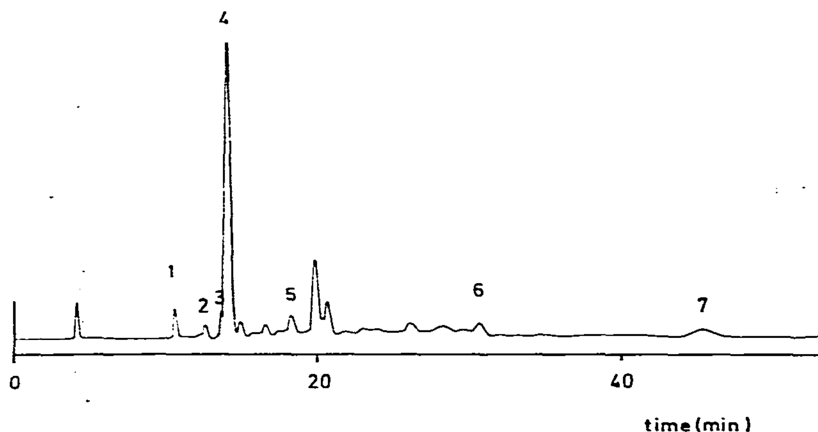


Fig. 4. Chromatogram of a filter sample at 340/425 nm. For chromatographic conditions, see Table I. Peaks: 1 = anthracene; 2 = fluoranthene; 3 = 1-methylanthracene; 4 = pyrene; 5 = benzo[*a*]-anthracene; 6 = benzo[*a*]pyrene; 7 = benzo[*ghi*]perylene.

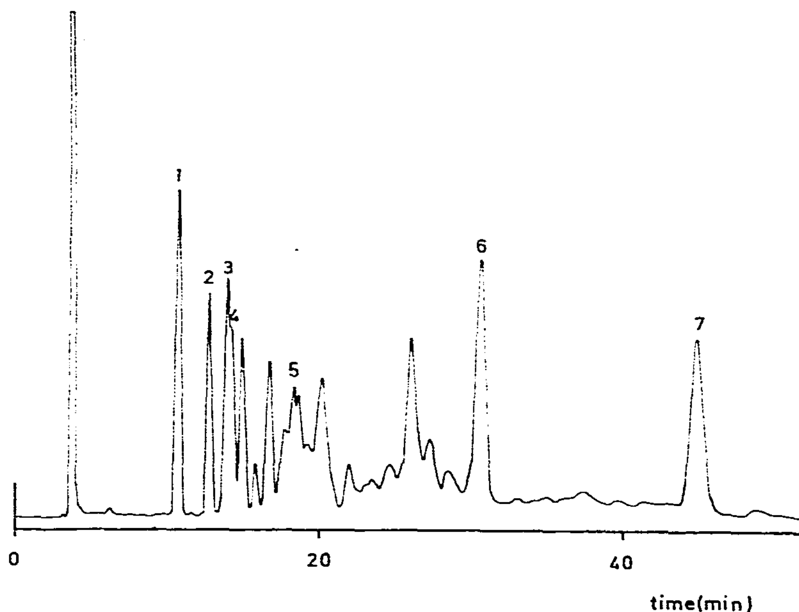


Fig. 5. Chromatogram of a filter sample at 363/435 nm. For chromatographic conditions, see Table I. Peaks: 1 = anthracene; 2 = fluoranthene; 3 = 1-methylanthracene; 4 = pyrene + 9-methylanthracene; 5 = benzo[*a*]anthracene; 6 = benzo[*a*]pyrene; 7 = benzo[*ghi*]perylene.

340 nm and an emission at 425 nm, and by the combination 363/435 nm. The choice of the excitation and emission wavelengths depends on the properties of the fluorescence detector, the separation of the PAHs and the PAH profile of the samples. Table I lists the detection limits for 23 PAHs at 340/425 nm and 363/435 nm. The selectivity of the fluorescence detector is also demonstrated by the HPLC/fluorescence trace of a sample of automobile exhaust collected on a glass-fibre filter at 340/425 nm (Fig. 4), and at 363/435 nm (Fig. 5). For instance, the peak of 1-methylanthracene is almost hidden by the peak of pyrene at 340/425 nm, whereas at 363/435 nm the peak of pyrene is suppressed. Even the selected conditions were not ideal for the determination of B*a*P; the sensitivity for this compound was at least five times better than attained with gas chromatographic methods[16].

*Identification*

The components of the samples were identified by comparing the retention times and the emission spectra obtained by a stop-flow technique with those of standards PAHs[30]. Thus, anthracene, fluoranthene, 1-methylanthracene, pyrene, benzo[*a*]-anthracene, B*a*P and benzo[*ghi*]perylene were identified at 340/425 nm, and anthracene, fluoranthene, 1-methylanthracene, benzo[*a*]anthracene, B*a*P, and benzo[*ghi*]-perylene were identified at 363/435 nm as components of automobile exhaust gases.

*Quantitation*

The quantitation of the PAHs in the samples was performed by comparing the measured peak heights with those of a standard mixture. In all cases examined, the response of the fluorescence detector was linear over three decades with the amount of PAHs injected. An important effect interfering in trace analysis is that arising from absorption of the fluorescent light by another non-fluorescent component[7]. A column with a low capacity (the number of theoretical plates was *ca.* 2000) was used to investigate whether any component absorbed the light emitted from another component in the cases where the two components were poorly resolved and the fluorescence of the first was suppressed. However, no evidence of this phenomenon was observed in the different combinations with relevant concentrations. It must, therefore, be assumed that the concentrations of the components in the measuring cell are sufficiently small to avoid inner-filter or concentration effects[7]. This assumption was further verified by the fact that the emission spectra of the components identified in the samples were not distorted, and also by the fact that the results obtained by analysing the samples at two different combinations of excitation and emission wavelength were consistent (Table II). The reason why there is a larger amount of B*a*P at 340/425 nm is that B*a*P is poorly resolved from an unidentified compound (see Fig. 4). At 363/435 nm the fluorescence of this compound is suppressed (see Fig. 5) and hence the analysis of B*a*P is not distorted.

*Control experiments*

The recoveries obtained by this method were tested by processing known quantities of the standards through the clean-up procedure. Table III shows the percentage recoveries for three determination. As a further assessment of the reproducibility of the method, a set of two equivalent filter and three equivalent cyclone

TABLE II

THE AMOUNTS OF PAHs ON A FILTER SAMPLE ANALYSED AT TWO DIFFERENT COMBINATIONS OF EXCITATION AND EMISSION WAVELENGTHS

| *Compound* | *Amount (μg)* | |
|---|---|---|
| | *340/425 nm* | *363/435 nm* |
| Anthracene | 17.9 | 17.7 |
| Fluoranthene | 14.9 | 15.1 |
| 1-Methylanthracene | 12 | 12.0 |
| Pyrene | 20.5 | – |
| Benzo[*a*]anthracene | 11.3 | 11.7 |
| Benzo[*a*]pyrene | 2.7 | 2.3 |
| Benzo[*ghi*]perylene | 1.6 | 1.5 |

TABLE III

RECOVERY TEST OF THREE STANDARD PAH MIXTURES

| *Compound* | *Amount (μg)* | *Recovery (%)* | | | | *S.D. (%)* |
|---|---|---|---|---|---|---|
| | | *I* | *II* | *III* | *Mean* | |
| Anthracene | 6.59 | 83 | 93 | 81 | 86 | 7 |
| Fluoranthene | 5.94 | 98 | 98 | 96 | 97 | 1 |
| 1-Methylanthracene | 8.82 | 90 | 97 | 87 | 91 | 6 |
| Pyrene | 2.18 | 98 | 102 | 98 | 99 | 2 |
| Benzo[*a*]anthracene | 15.38 | 100 | 99 | 98 | 99 | 1 |
| Benzo[*a*]pyrene | 1.15 | 102 | 97 | 96 | 98 | 4 |
| Benzo[*ghi*]perylene | 6.21 | 100 | 101 | 99 | 100 | 1 |

TABLE IV

THE REPRODUCIBILITY OF THE ANALYSIS OF THE PAHs IN TWO EQUIVALENT CYCLOHEXANE EXTRACTS OF A FILTERS

| *Compound* | *Amount (μg)* | | | *S.D. (%)* |
|---|---|---|---|---|
| | *I* | *II* | *Mean* | |
| Anthrancene | 2.71 | 2.65 | 2.68 | 2.0 |
| Fluoranthene | 2.76 | 2.77 | 2.77 | 0.3 |
| 1-Methylanthracene | 1.53 | 1.42 | 1.48 | 6.6 |
| Pyrene | 4.47 | 4.43 | 4.45 | 0.8 |
| Benzo[*a*]anthracene | 2.27 | 2.30 | 2.29 | 1.2 |
| Benzo[*a*]pyrene | 0.34 | 0.35 | 0.35 | 2.6 |
| Benzo[*ghi*]perylene | 0.83 | 0.79 | 0.81 | 4.4 |
| | | | Mean | 3% |

extracts was analysed (Tables IV and V). Table VI gives the variation coefficients for the determinations of the PAHs in a filter sample. Figs. 4 and 5 show typical chromatograms of PAHs in the filter samples.

TABLE V

THE REPRODUCIBILITY OF THE ANALYSIS OF THE PAHs IN THREE EQUIVALENT CYCLOHEXANE EXTRACTS OF A CYCLONE

| *Compound* | *Amount (μg)* | | | | *S.D. (%)* |
|---|---|---|---|---|---|
| | *I* | *II* | *III* | *Mean* | |
| Anthracene | 0.35 | 0.30 | 0.33 | 0.33 | 9 |
| Fluoranthene | 0.45 | 0.44 | 0.44 | 0.44 | 1 |
| 1-Methylanthracene | 0.22 | 0.19 | 0.21 | 0.21 | 9 |
| Pyrene | 0.61 | 0.61 | 0.63 | 0.62 | 2 |
| Benzo[*a*]anthracene | 0.38 | 0.35 | 0.35 | 0.36 | 5 |
| Benzo[*a*]pyrene | 0.026 | 0.025 | 0.024 | 0.025 | 5 |
| Benzo[*ghi*]perylene | 0.098 | 0.084 | 0.093 | 0.092 | 9 |
| | | | | Mean | 6% |

TABLE VI

THE VARIATION COEFFICIENTS FOR A TRIPLICATE DETERMINATION OF THE PAHs IN A FILTER EXTRACT

The determinations were performed on three days.

| *Compound* | *Amount (μg)* | | | | *S.D. (%)* |
|---|---|---|---|---|---|
| | *I* | *II* | *III* | *Mean* | |
| Anthracene | 12.8 | 13.0 | 12.0 | 12.6 | 5 |
| Fluoranthene | 30.3 | 30.4 | 30.9 | 30.5 | 1 |
| Pyrene | 95.5 | 96.3 | 98.7 | 96.8 | 2 |
| Benzo[*a*]anthracene | 9.8 | 9.4 | 8.8 | 9.3 | 6 |
| Benzo[*a*]pyrene | 4.20 | 4.21 | 4.30 | 4.2 | 1 |
| Benzo[*ghi*]perylene | 12.3 | 12.2 | 12.4 | 12.3 | 1 |
| | | | Mean of S.D. | | 3% |

## CONCLUSIONS

The procedure outlined offers a number of advantages over other methods for the routine analysis of PAHs. The use of TLC for purifying the PAH fraction improves the quality of the chromatograms, as other fluorescent compounds are eliminated. By appropriate choice of the excitation and emission wavelengths, it is possible to analyse poorly resolved components by means of on-line fluorescence detection. In addition to the selectivity, fluorescence is remarkably sensitive for some of the PAHs, especially the important B*a*P.

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