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GAS CHROMATOGRAPHY–MASS SPECTROMETRY METHOD FOR IDENTIFYING AND DETERMINING POLYCHLORINATED BIPHENYLS

G. WILLIAM TINDALL and PERCY E. WININGER*
Tennessee Eastman Company, Kingsport, TN 37662 (U.S.A.)
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SUMMARY

The objective of this work was to develop a procedure for the detection and quantitative determination of any of the 209 possible polychlorinated biphenyls (PCBs) in a variety of commercial and environmental samples. The procedure described involves extraction of the sample with hexane, concentration of the extract by distillation, and determination of any PCBs present by computer controlled gas chromatography–mass spectrometry. This method has been successfully used for qualitative and quantitative determination of PCBs and PCB mixtures in solid, gaseous, and aqueous samples. It has also been used for screening samples to determine that no detectable PCBs are present. The method is relatively free of interferences and capable of detecting 0.01–0.2 μg/ml amounts of any PCB in sample extracts. The precision of the method is similar to that of gas chromatography methods for PCBs.

INTRODUCTION

Health and environmental concerns related to polychlorinated biphenyls (PCBs) have resulted in an increasing number of government regulations controlling their manufacture, use, and disposal. There has developed a corresponding need for laboratories to determine PCBs in environmental samples (air, water, soil), raw materials, manufactured products, and waste. Historically, only commercial mixtures of PCBs, such as the Arochlors, were of concern. PCBs and Arochlor PCB mixtures were considered the same. Analytical methods capable of identifying and determining the known commercial PCB mixtures were considered sufficient. However, it was found that some of the isomers in these mixtures are more biodegradable than others and others are more easily destroyed by incineration[1]. In addition, the recent trend in government regulations is to include any PCB or PCB mixture, as well as the Arochlor PCB mixtures. Now in many cases, the objective of a PCB analysis is to determine all of the 209 possible PCBs in a sample or to ensure their absence at significant concentrations. Accordingly, an analytical method was needed that is capable of determining significant concentrations of all possible PCBs and mixtures of PCBs.

Electron capture gas chromatography has been the recommended method for identifying and determining Arochlor PCB mixtures[1–3]. This technique can be used to

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identify and determine PCBs and PCB mixtures of known composition; for example, Arochlor PCB mixtures, as well as individual PCB compounds, provided that interfering compounds are absent. Lengthy sample cleanup procedures are used to minimize positive interferences. In addition to the problem of interferences, the electron capture gas chromatography method is of little use in analyzing samples of unknown PCB composition; for example, when a single unknown PCB is present.

Perchlorination of PCBs to decachlorobiphenyl has been suggested as a method of determining all possible PCB isomers in a sample[1,5]. The sample, or the cleaned-up sample, is perchlorinated and total PCBs determined as decachlorobiphenyl by gas chromatography. This technique is subject to serious positive interferences. Biphenyl, and other compounds, perchlorinate to yield decachlorobiphenyl or other compounds that can elute at the same retention time as decachlorobiphenyl. Biphenyl is commonly found in samples for PCB analysis. Biphenyl is used throughout industry as a heat-transfer liquid and in other applications, and biphenyl is a normal product of hydrocarbon combustion. The small amounts of biphenyl that are found in many types of PCB sample render the perchlorination method unreliable.

The method described in this paper is capable of qualitatively and quantitatively determining any of the 209 possible PCBs or PCB mixtures. This method is particularly valuable for screening a sample to ensure the absence of detectable concentrations of all possible PCBs. The method involves the simultaneous acquisition of gas chromatography–mass spectrometry (GC–MS) data for the molecular ions of each PCB group. A PCB group is defined as all the PCBs that have the same molecular weight. Mass chromatograms are successively displayed by the data system for the molecular ions of each PCB group in the appropriate retention time window for the group. The intensity ratios of the molecular ions in the appropriate retention time windows are used to determine the presence of PCBs in each PCB group. Areas of PCB peaks in the mass chromatograms are used to determine concentrations. Use of an internal standard improves precision. The method is capable of detecting individual PCBs at concentrations of from 10 to 200 μg/l, depending on the molecular weight of the PCB.

EXPERIMENTAL

Sample preparation

Normally the first step in an analysis will be the isolation of PCBs from the sample by solvent extraction using hexane. Solids are extracted in a Soxhlet extractor for 24 h. Gaseous samples are passed through a 6 × 2 cm I.D. column of Florisil absorbent[6]. The PCBs trapped by the absorbent are then extracted in a Soxhlet extractor. Aqueous samples are extracted in a separatory funnel with three 75-ml portions of hexane. Aqueous samples that tend to emulsify are extracted in a bottle instead of a separatory funnel. The water sample, hexane, and a magnetic stirring bar are added to the bottle. The contents are gently stirred so as to maintain separation of the hexane–water layers. From 30 to 60 min is long enough to achieve equilibration of a 1-l water sample with 75 ml of hexane.

Hexane extracts are concentrated by distillation. A pear-shaped flask is used for the pot and a micro-jacketed Vigreux column is used for the reflux column. The flask is heated by a water bath consisting of a 1-l beaker and a 1-l beaker heating mantle. The heating rate is controlled by adjusting the immersion of the pear-shaped flask

in the water bath. Mounting the water bath on a jack stand enables the immersion level to be conveniently controlled. Hexane extracts of up to 200 ml can be concentrated to a volume of 1 ml with this apparatus. The concentrated extracts are spiked with 2,4,6-tribromobiphenyl internal standard. An internal standard concentration of 1 mg/l is recommended.

Cleanup

Samples can be column chromatographed to remove polar interfering compounds. We prefer the procedure of Mills[7] because it does not use diethyl ether as a solvent.

Standards

Standards are prepared in heptane from pure PCB compounds or PCB mixtures. Standards are spiked with internal standard at the same concentration as samples.

Data acquisition

A Finnigan 4000 GC–MS or similar computer-controlled GC–MS is used to acquire the GC–MS data. The objectives of data acquisition are to acquire the GC–MS data that can be used to determine if any PCBs are in the sample and then to determine the concentration of any PCBs detected. The mass spectrometer is set up and tuned according to the manufacturer's instructions. Special attention must be devoted to resolution to ensure that the resolution is adequate up to mass 500. To optimize the sensitivity for high and low molecular weight PCBs, each sample is analyzed twice: once for monochloro- to hexachlorobiphenyl groups and once for the hexachloro- to decachlorobiphenyl groups. The GC–MS conditions for these analyses are given in Tables I and II.

TABLE I

GC–MS CONDITIONS FOR Cl_1–Cl_6 PCB GROUPS

Column:	6 ft. × 2 mm I. D. glass, 3% OV-101 on 80–100 mesh Chromosorb W-HP
Programme:	2 min at 130°C program 130–250°C at 8°C/min, 10 min at 250°C
Injector temperature:	250°C
Separator temperature:	270°C
Carrier gas:	He at 20 ml/min
Sample size:	4 μl
Electron energy:	70 eV
Emission current:	0.40 mA

MULTIPLE ION DETECTION PROGRAMME

PCB group	*Mass range scanned*	*Ions for identification*	*Ions for quantitation*
Cl_1	186–190	188, 190	188 (190)*
Cl_2	220–226	222, 224, 226	222 (224)
Cl_3	254–260	256, 258, 260	258
Cl_4	288–294	290, 292, 294	292 (294)
Cl_5	322–328	324, 326, 328	326 (328)
Cl_6	356–364	358, 360, 362	360 (364)
TBB (int. std.)	387–394	388, 390, 392	390

* Ion for quantitation to minimize M^+ — HCl interference (see text).

TABLE II

GC–MS CONDITIONS FOR Cl_6–Cl_{10} PCB GROUPS

Column:	6 ft. × 2 mm I.D. glass, 3% OV-101 on 80–100 mesh Chromosorb W-HP
Programme:	2 min at 210°C; program 210–260°C at 8°C/min, 10 min at 260°C.
Injector temperature:	250°C
Separator temperature:	270°C
Carrier gas:	He at 20 ml/min
Sample size:	1–4 μl
Electron energy:	70 eV
Emission current:	0.40 mA

MULTIPLE ION DETECTION PROGRAMME

PCB group	*Mass range scanned*	*Ions for identification*	*Ions for quantitation*
Cl_6	356–364	358, 360, 362	360 (364)*
Cl_7	386–400 (Cl_7 + TBB)	394, 396, 398	394 (398)
Cl_8	426–434	428, 430, 432	430 (432)
Cl_9	460–468	462, 464, 466, 468	464 (468)
Cl_{10}	494–504	496, 498, 500, 502	498
TBB (int. std.)		388, 390, 392	392

* Ion for quantitation to minimize M^+ — HCl interference (see text).

Qualitative analysis

Retention time windows for each PCB group are established using the retention index data of Albro[8]. A 10-ppm standard of *n*-alkanes (C_{16} to C_{24}) is used to locate the retention times of these compounds using the gas chromatographic conditions used in the PCB analysis. The PCB retention time windows of the PCB groups on an OV-101 column packing are given relative to these hydrocarbons in Table III. The calculated retention time windows are checked by analyzing commercial PCB mixtures, for example Arochlor 1016, 1232, 1248 and 1260 PCB mixtures.

TABLE III

PCB RETENTION WINDOWS

PCB group	*Retention index windows*
Cl_1	1509–1585
Cl_2	1619–1774
Cl_3	1732–1959
Cl_4	1842–2144
Cl_5	1959–2305
Cl_6	2076–2466
Cl_7	2238–2433
Cl_8	2383–2578
Cl_9	2545–2555
Cl_{10}	2690

Mass chromatograms of two to four of the strongest molecular ions in a PCB group are displayed by the data system in the retention time window for each PCB group. A typical display is given in Fig. 1 for the tetrachlorobiphenyl group. Each peak in the chromatogram is evaluated to determine if it is a PCB peak. Peaks must

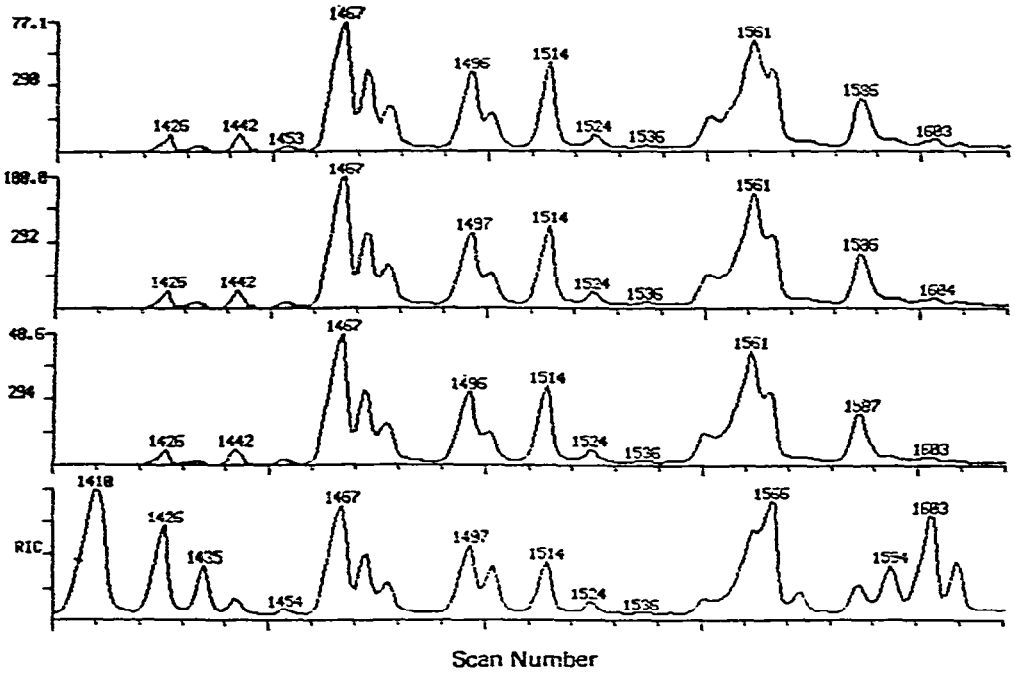

Fig. 1. Ion chromatograms for tetrachlorobiphenyls from an Arochlor PCB mixture.

meet three criteria to be labelled PCB peaks for quantitation: (1) the peaks of the characteristic ions must maximize at the same retention time; (2) the peak must be in the proper retention time window; and (3) the relative peak intensities of the molecular ions must be within $\pm 15\%$ of the theoretical ratio[9]. This tolerance is arbitrary and can be made larger for very low concentrations of PCBs where statistical variations in peak intensity become large.

Quantitative analysis

Quantitation of PCBs detected in the qualitative analysis can be performed in several ways. A detailed description of one method is given below using the tetrachlorobiphenyl group as an example. GC–MS data are acquired on the sample, as well as two tetrachlorobiphenyl standards that bracket the estimated concentration of the tetrachlorobiphenyls in the sample. Data acquisition parameters given in Table I are used. Ion chromatograms for mass 292 are displayed by the data system for the sample data. The areas of the tetrachlorobiphenyl peaks are summed, ΣA_x, for the sample. The area of the mass 390 peak in the respective internal standard is measured, $A_{\text{I.s.,x}}$. The areas of the mass 292 peaks for the high and low tetrachlorobiphenyl standards are measured, A_{s1} and A_{s2}, as well as their respective internal standard areas, $A_{\text{I.s.,1}}$ and $A_{\text{I.s.,2}}$. A response factor, R, is calculated from the standard areas and their concentrations, C_1 and C_2, using eqn. 1.

$$R = \frac{C_1}{A_{s1}/A_{\text{I.s.,1}}} + \frac{C_2}{A_{s2}/A_{\text{I.s.,2}}} \; 1/2 \qquad (1)$$

The total concentration of tetrachlorobiphenyls in the sample, ΣC_{Cl_4}, is calculated using eqn. 2.

$$\Sigma C_{Cl_4} = \frac{R\Sigma A_n}{A_{i.s.,x}} \quad (2)$$

RESULTS AND DISCUSSION

Hexane is preferred for PCB analyses over the commonly-used extraction solvents. Hexane will not extract polar compounds, which results in a "cleaner" sample for analysis. The extraction efficiency of hexane for PCBs is adequate. As a test of extraction efficiency, 2 μg of a hexachlorobiphenyl in hexane was mixed with 100 g of fly ash that contained *ca.* 20% carbon. The ash was dried to deposit the PCB on the ash. It was thought that extraction of the small amount of PCB from the large, active surface of the fly ash would challenge the ability of hexane as a PCB extractant. The PCB was extracted with hexane in a Soxhlet extractor and analyzed as described by the procedure in the Experimental section. Recoveries of more than 90% were found for three experiments. The level of interfering compounds also extracted was low relative to the PCB without any sample cleanup. When methylene chloride was used in this experiment, a precipitate of other extracted compounds formed on concentration of the methylene chloride and the level of interference obscured the PCB.

We prefer the apparatus described for concentrating extraction solvent to the traditional Kuderna–Danish apparatus. The volume of solvent in the Vigreux column is less than 0.5 ml during distillation. Heat loss through the jacketed column is small, so distillation proceeds smoothly with only a small heat input to the solvent and little analyst attention. Some 200 ml of hexane can be concentrated to 1 ml without changing distillation column or pot. The distillation proceeds at high efficiency, for example, greater than 90% recovery of 1 μg of xylene was found during the distillation of 200 ml of hexane to a 1 ml final volume.

There are two types of interference that must be eliminated to achieve a successful analysis. Percent amounts of any compound in the sample that elutes during the analysis may affect the mass spectrometer source and rods. In most cases, the sensitivity of the mass spectrometer for PCBs will drop. In some cases recalibration is necessary and, in extreme cases, the source and rods will have to be cleaned before the analysis is continued. Florisil column chromatography[7] and extraction of the hexane with concentrated sulphuric acid are usually effective for removing this type of interference. The other type of interference occurs when a compound with an ion common to a PCB molecular ion elutes in a PCB retention time window. Hydrocarbons have weak ions common to most of the PCB molecular ions. High concentrations of hydrocarbons, for example petroleum products or products of combustion, may obscure low concentrations of PCBs .The above cleanup methods do not reduce the level of these hydrocarbon interferences.

It is probably possible to eliminate the hydrocarbon interference by using a mass spectrometer with greater resolution than that of the Finnigan 4000. Elimination of the interference of hydrocarbons was attempted by using the Finnigan 4000 in the negative ion chemical ionization mode with methane reagent gas. The signal for the negative parent ions for most PCBs was found to be much weaker than for the positive molecular ions. While the interference of hydrocarbons on the PCB parent ions is

diminished using the negative ion mode, the overall detection limit for PCBs was not adequate for our work. Apparently, negative PCB ions readily lose Cl^- so that most of the negative ion signal consists of Cl^-. By monitoring the negative mass 35 or 37, extremely small amounts of PCBs could be detected (less than 1 μg/l) in the presence of hydrocarbons. Unfortunately, many other chlorinated compounds also gave strong mass 35 and 37 negative ions. Because the mass 35 or 37 ion gives no clue to the parent molecule, the negative ion approach is not useful for qualitative work and the evaluation of it was stopped.

The accuracy-limiting part of the procedure is the selection of standards. In the Experimental section, we suggest using a standard for each PCB group. The underlying assumption is that all PCBs of the same molecular weight have the same response factor. In fact, GC–MS response factors within a PCB group may vary as much as a factor of two[10]. Frequently it is not known which isomer is being quantitated, and, even if it is known, standards are available in reasonable amounts and cost for only a few PCBs. Another approach is to use a commercial PCB mixture of known composition as a standard. The response factors of the several isomers in each PCB group may average to yield a more accurate response factor for each PCB group. Unfortunately, commercial PCB mixtures of known compositions are not generally available to most laboratories[3]. For our standard, we select one PCB from each PCB group, based on cost and availability, and accept the possible bias in the quantitative results.

The choice of internal standard is critical. To be of value, its chemistry in the source must match that of the PCBs. We have evaluated internal standards that have resulted in poorer precision than if no internal standard were used. The best internal standard for a PCB analysis is a PCB. However, for a general purpose procedure, a PCB cannot be used. We found tribromobiphenyl to be a reasonable substitute. It responds to source changes much like a PCB and its molecular weight is great enough that interferences with its ions are rarely encountered.

The mass spectrometer is tuned according to manufacturer's recommendations. Proper resolution is critical. If the mass peaks are not adequately resolved, then another type of interference is encountered. If a compound co-elutes with a PCB that has relatively stronger ions one mass more or less than a PCB, the data system will have difficulty apportioning peak area between the adjacent masses. If the peaks are underresolved, some or all of the weaker PCB mass peaks may be merged with the stronger adjacent mass peak. Symptoms of this problem will be evident in displays of the ion chromatograms for the PCB ions. Instead of the usual Gaussian curves for the eluting PCB ions, these curves will have discontinuities at certain scan numbers. The pieces of area missing from the PCB ion chromatogram will appear in the ion chromatogram one mass more or less than the PCB ion. This problem is not uncommon in actual samples when the PCB concentration is very low. Adjusting the mass spectrometer for greater resolution will solve the problem. Overresolving mass peaks will result in a considerable reduction of sensitivity.

For quantitation, the strongest molecular ion is used. If there are interferences for that ion, some other ion may be used (see below). If interferences on all ions are known to be absent, another technique can be used with a considerable improvement in detection limit. Data are acquired with less-than-unit mass resolution which improves sensitivity. For quantitation, the sum of the ions over the mass range of the

molecular ions are displayed by the data system instead of each individual ion. The resulting peak is then measured for quantitation.

For the qualitative and quantitative procedures to be universally applicable to any PCB determination, the GC–MS data for each PCB group must be unique. The GC–MS data for any PCB group must not be influenced by PCBs in other PCB groups. The mass spectrum of a typical PCB shows that the molecular ion initially fragments by loss of two chlorines and to a lesser extent by loss of HCl and Cl[11]. The loss of two chlorine atoms from a molecular ion will produce an ion cluster that has several masses common to the molecular ions of the PCB group with two fewer chlorines. As an example, the spectra for hexachlorobiphenyl and tetrachlorobiphenyl are shown in Fig. 2.

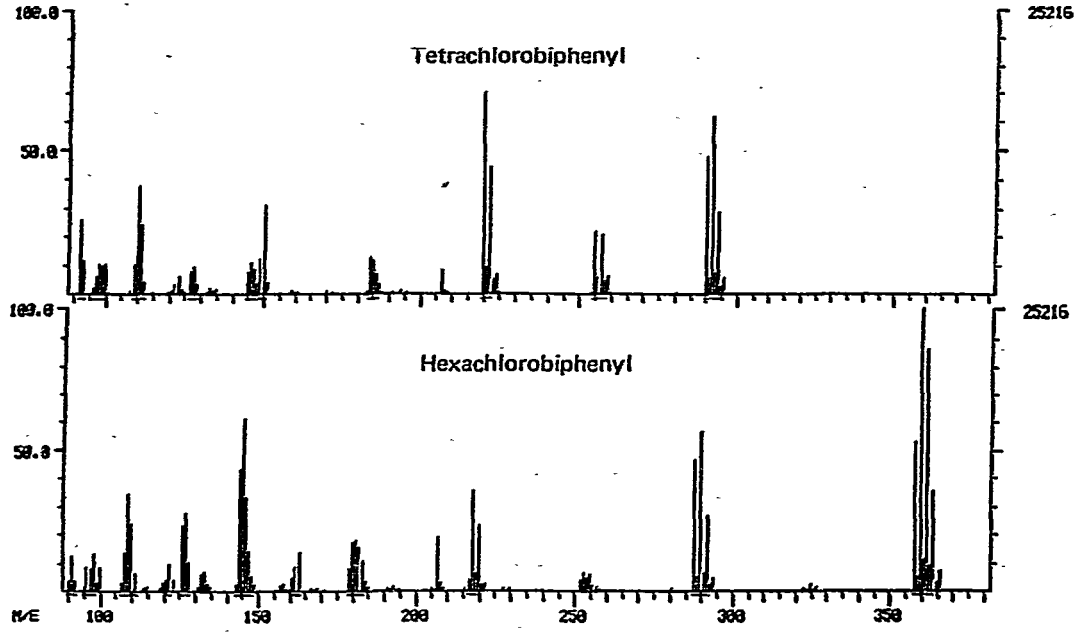

Fig. 2. Comparison of spectra of tetrachlorobiphenyl and hexachlorobiphenyl.

The use of gas chromatography retention windows minimizes the interference of PCB groups differing by two chlorines. The calculated data in Table III show that there is potential overlap of gas chromatography retention time windows of hexachlorobiphenyls on tetrachlorobiphenyls, heptachlorobiphenyls on pentachlorobiphenyls and octachlorobiphenyls on hexachlorobiphenyls. The calculated retention times for all but four of the hexachlorobiphenyls and heptachlorobiphenyls are uncertain, so these isomers may in fact not overlap the tetrachloro- and pentachlorobiphenyl groups[8]. The other overlapping hexachloro- and heptachloroisomers have a 2,4,6-substitution which is not normally found in PCB samples. Hence, there is potential interference between hexachloro- and tetrachloro-, and heptachloro- and pentachloro-groups; but, in practice, this interference probably would never be encountered.

Of the twelve possible octachlorobiphenyl isomers, nine overlap the retention time window of hexachlorobiphenyl. If any of these nine isomers are encountered, their $M - Cl_2$ ions that will be detected in the hexachlorobiphenyl window should not be counted as a hexachlorobiphenyl.

There is considerable overlap of retention time windows of adjacent PCB groups. Loss of Cl from a ^{13}C-containing PCB molecular ion and loss of HCl from the molecular ion will result in ions that could interfere with the molecular ions of the next lower molecular weight group. The relative abundance of $M - Cl$ ions is rarely over 5%[11] so the loss of Cl from a ^{13}C-PCB will never generate an ion of over 1% relative abundance, and this potential interference can be ignored. The relative abundance of the $M - HCl$ ion is usually less than 5%[11]. If the sample contained a high concentration of a PCB, then the weak $M - HCl$ ions of this PCB could be detected in the retention window of the next lower molecular weight PCB group. The fact that these ions came from an $M - HCl$ ion would be evident from the coincident retention time and the ion masses in the $M - HCl$ cluster. The $M - HCl$ ion cluster would contain an ion of mass 2 less than the M^+ ion cluster of the next lower PCB group.

In a quantitative analysis, the interference of the $M - HCl$ ions can be made less than 5% by proper selection of the ions to integrate for quantitation. The ions to use for quantitation if $M - HCl$ interference is a problem are shown in Tables I and II.

In summary, the abundant $M - Cl_2$ ions of octachlorobiphenyls may appear in the hexachlorobiphenyl retention time window, and $M - HCl$ ions from a high concentration of a PCB may appear as weak ions in the retention time window of the next lower molecular weight PCB group. Neither of these potential interferences is likely seriously to limit the application of this procedure to the qualitative and quantitative analysis of PCBs.

The detection limits for one specific PCB from each PCB group were measured using standards prepared in heptane. These data are given in Table IV. These detection limits represent the concentrations of each PCB group that can be routinely detected and quantitated with certainty in a sample free of interferences. Because of the way the raw GC–MS data are treated by the data system, particularly the setting of thresholds, we felt our judgement of detection limits was more useful than a value arrived at by statistical analysis. These detection limits are easily achieved on a routine basis if the

TABLE IV

PCB GROUP DETECTION LIMITS

PCB group	*Detection limit (ng/ml)*
Cl_1	10
Cl_2	10
Cl_3	10
Cl_4	20
Cl_5	20
Cl_6	20
Cl_7	100
Cl_8	100
Cl_9	200
Cl_{10}	200

mass spectrometer source is clean. As the mass spectrometer source and analyzer become dirty, the detection limit gets larger. We suggest that the standards be analyzed regularly to assess the sensitivity of the GC–MS system.

The precision of this analysis was estimated by computing the coefficient of variation of the response factor (concentration of standard/area of PCB peak) for a 0.3 μg/ml hexachlorobiphenyl standard analyzed every third analysis during a 12-h period of PCB analyses. The coefficient of variation was 15%. The extracts analyzed during this time period contained up to 100 μg/ml concentrations of other unidentified organic compounds. These extracts are considered relatively clean. The computed response factor showed obvious trends during this 12-h period. The use of the internal standard greatly improved this measure of precision. When the response factor was computed using the internal standard (concentration standard × area internal standard peak/area of PCB peak), the coefficient of variation of this response factor measured over this 12-h period was 2%. Obviously, the internal standard was effective in compensating for the drift in mass spectrometer sensitivity during this 12-h interval.

As a measure of overall method precision, spiked samples were analyzed in triplicate several days apart over a several-week period. These extracts contained much higher concentrations of other compounds. The internal standard was used. The concentration of the PCB spike was *ca.* 1–10 μg/ml. The coefficient of variation calculated for the results was 20%. The principal cause of the poorer precision in this experiment is the persistent effect of the other eluting compounds on the sensitivity of the mass spectrometer source. These effects are not entirely compensated for by the internal standard. This precision is similar to that reported for PCB analyses by gas chromatography[12]. More frequent recalibration probably would have improved this precision.

In summary, the accuracy and the precision of the analyses will depend on how often the instrument is calibrated and how much other compounds in the sample affect the sensitivity of the source during the course of analyses. At worst, the coefficient of variation of the method should be ±20%.

CONCLUSION

The procedure described can be used to detect and quantitatively determine any PCB or PCB mixture in a variety of samples. The method is relatively free of interferences and is sensitive. The method is capable of precision as good as gas chromatography procedures.

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