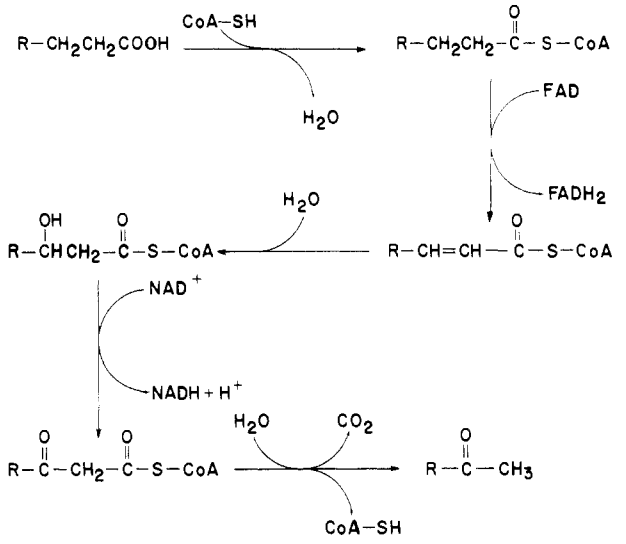
Scheme I. Proposed Enzymatic Formation Pathways of a Methyl Ketone from a Fatty Acid




to that of this compound. This unknown ketone was, therefore, identified as (8*Z*,11*Z*,14*Z*)-8,11,14-heptadecatrien-2-one.

It is known that methyl ketones are readily produced from fat and oils by lipolytic bacteria, fungi, and yeasts (Tuynenburg Muys, 1965; Cantoni et al., 1967; Scott, 1968). The proposed formation pathway of a methyl ketone from the correspoinding fatty acid is shown in Scheme I.

Crossley et al. (1962) reported that heat treating triglycerides proposed methyl ketone, which possesses one less carbon atom than the corresponding fatty acid, by β-oxidation. This reaction occurs at 190 °C in the presence of oxygen.

The above two possible pathways can be proposed for the formation of the novel ketone; however, the temperature did not exceed 45 °C throughout the experiment. It is, therefore, more reasonable to conclude that this novel ketone is formed from linolenic acid ($C_{17}H_{29}COOH$) by a metabolic breakdown.

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# Headspace Gas Chromatographic Analysis of Foods for Volatile Halocarbons

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A headspace technique for determining volatile (boiling point less than 150 °C) halocarbons (VHCs), such as chloroform, 1,1,1-trichloroethane, carbon tetrachloride, trichloroethylene, and tetrachloroethylene, in foods was developed. Food samples were placed in a septum-capped vial, 20 N $H_2SO_4$ was added to digest the sample, and water was added as a diluent. Some samples were analyzed without preliminary treatment, depending upon the matrix. The vial was equilibrated at 90 °C for 1 h. An aliquot of headspace was injected into a gas chromatograph equipped with an electron capture detector. VHCs were detected at sub-part-per-billion levels in aqueous foods, while for lipid-containing matrices, detection limits were in the 10–50-ppb range. By use of external standards or the method of standard additions, relative standard deviations of 20% or less were achieved. Fish and several processed foods, including jelly, chocolate sauce, ice cream, and mayonnaise, were analyzed by using the headspace technique.

Volatile halocarbons (VHCs), such as chloroform ($CHCl_3$), carbon tetrachloride ($CCl_4$), 1,1,1-trichloroethane (MC), trichloroethylene (TCE), and tetrachloroethylene (PCE), are used as solvents and chemical intermediates; hundreds of millions of pounds of each are produced annually (International Trade Commission, 1979). Some of these compounds are animal carcinogens (Environmental Protection Agency, 1978). They have been reported in ground and surface water (Deinzer et al., 1978; Zoeteman et al., 1980) and also in foods (McConnell et al., 1975; Page and Charbonneau, 1978). These compounds may enter the food supply through contamination of water used in food processing, as cleaning solvents for food processing equipment, through direct uptake from the environment (e.g., by fish), or through contact with packaging materials.

Because of the frequency of reports of VHCs in ground water and the potential for entering the food supply, a method was needed for the analysis of foods for the

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presence of VHCs. Determination of VHCs using solvent extraction, cleanup, and concentration by solvent evaporation was not possible because of large losses of analyte by volatilization. By taking advantage of the volatility of the VHCs, we eliminated the need for cleanup steps by analyzing the headspace vapors in equilibrium with the food sample. The quantity of analyte in an aliquot of the headspace is then related to the concentration of analyte in the sample.

Headspace analysis using similar techniques has been used to analyze both hydrophilic (Drozd and Novak, 1977) and hydrophobic (Drozd and Novak, 1978; Piet et al., 1978; Dietz and Singley, 1979; Lukacovic et al., 1981) compounds in water and in some foods (Gilbert et al., 1978). A method for the analysis of fish was developed and later adapted to determine VHC residues in a wide variety of foods. The method developed involves the equilibration of a food sample with the headspace in a sealed vial and analysis of the headspace by gas chromatography (GC).

This procedure has been used to determine VHCs at part-per-billion (nanograms per gram) levels in a variety of foods including ice cream, jelly, chocolate sauce, mayonnaise, and corn oil and environmental samples such as fish and water.

## EXPERIMENTAL SECTION

**Apparatus and Reagents.** An automated headspace analyzer, a Perkin-Elmer F-42 gas chromatograph equipped with an electron capture detector (ECD), was primarily used. Manual injection of headspace using a gas-tight syringe into a Varian 3700 gas chromatograph equipped with an ECD was also used. A Precision Sampling Model A-2 Pressure-Lok gas-tight syringe (5 or 10 mL) gave the best results for manual injections. The circulating water bath of the automated headspace analyzer was used to heat the sample vials to 90 °C before analysis. For manual injection, a circulating water bath (90 °C) was used, and the gas-tight syringe was heated in a 90 °C oven immediately before use. Gas chromatography–mass spectrometry (GC–MS) was performed with a Finnigan 4000 quadrupole mass spectrometer in the electron impact mode.

Glass screw-cap vials (40 mL) with Teflon-lined septa (Supelco) were used to store standards. These vials permitted syringe withdrawal of solution through the septa. Crimp-top vials (24 mL) with Teflon-lined rubber septa and metal seals (Perkin-Elmer) were used to equilibrate the samples for headspace analysis.

Reagent-grade 2-propanol was used as the solvent for standards. Reagent-grade concentrated sulfuric acid was used to prepare the digestion medium, 20 N $H_2SO_4$. Distilled water, used for diluting samples and preparing the digestion medium, was purged with nitrogen or helium to remove traces of VHCs. Glassware was cleaned by rinsing with 2-propanol, followed by heating 20 min at 90 °C. Because VHCs are widely used as solvents, reagent and glassware blanks were analyzed to ensure that no contaminants were present. Periodic analysis of laboratory blanks consisting of a food-simulating matrix (corn oil) which had been exposed to the ambient atmosphere in Petri dishes during the experiments revealed no detectable contamination at part-per-billion levels.

**Preparation of Standards.** Chemicals (usually of 95% or greater purity) were obtained from chemical supply houses and used without further purification. Stock solutions, of concentrations from 10 to 300 mg/mL, were prepared by diluting weighed standards in volumetric flasks. They were stored in screw-cap vials filled so that no headspace existed above the solution and were placed in a freezer. Working standards, ranging from 0.5 to 500 μg/mL, were prepared by serial dilution using microliter syringes and were stored in the same fashion as the stock solutions. These solutions can be kept indefinitely at part-per-million and higher concentrations if they are tightly sealed with an intact Teflon-lined septum and kept in a freezer. Working standards were prepared every 2–4 weeks when used daily. Septa which had been pierced by syringe needles were replaced at the end of each day of use.

**Preparation of Samples.** Solids such as meat and fish (30–50 g) were ground while partially frozen. These and other solids and viscous liquids were stored frozen in glass jars or vials with Teflon-lined screw caps until analysis. Aqueous liquids were refrigerated in completely filled glass bottles sealed with Teflon-lined screw caps. Loss of VHCs through adsorption or migration through organic polymers such as polyolefins occurs rapidly; these materials were therefore not used.

**Procedure.** Food samples were treated differently depending on the physical nature of the sample. Liquids were analyzed undiluted. Semisolids (e.g., butter) and viscous liquids, if free flowing at 90 °C, were also analyzed undiluted. Water-miscible foods (e.g., jellies) were diluted with distilled water or, if water-immiscible (e.g., meat), digested in 20 N $H_2SO_4$.

The general procedure used was as follows: 1–2 g of the sample was weighed into a tared crimp-top vial; 1–5 mL of distilled water or 15 mL of digestion medium (20 N $H_2SO_4$) was added to the vial if dissolution or digestion was needed; an internal standard or spiking standards were added with microliter syringes; the vial was capped with a crimp seal containing a Teflon-lined septum; the vial was shaken and placed in a water bath at 90 °C for 1 h; 1–5 mL of headspace was injected into the gas chromatograph.

Spiking standards were added to the vials for quantitation by standard additions or when preparing external standards. Mixed standards were used whenever possible.

Generally, a 2-mL injection of headspace vapor was used. The conditions for the automated injection of headspace are described under Gas Chromatography. For manual injection with a gas-tight syringe, the heated syringe was filled twice with headspace, the contents were discharged back into the vial each time, the syringe was filled a third time, and the syringe valve was closed. All but the desired volume was discharged immediately before injection, and the remainder was introduced into the gas chromatograph. Gloves were worn when handling the heated syringe. After injection the syringe was dismantled, and air was drawn through the needle and barrel to remove traces of VHCs.

**Gas Chromatography.** Three GC columns were used: (1) 15% OV-17 on 80–100-mesh Chromosorb W HP in a 3.7 m × 2.1 mm i.d. stainless steel column operated isothermally at 85 °C with a 20 mL/min carrier flow; (2) 20% SP-2100/0.1% Carbowax 1500 on 100–120-mesh Supelcoport in a 3 m × 2.1 mm i.d. stainless steel column operated isothermally at 100 °C with a 20 mL/min carrier flow; (3) Perkin-Elmer 4X glass capillary column, 50 m × 0.27 mm i.d. coated with OV-101 (film thickness approximately 0.9 μm), with a 3.5:1 split ratio at the injection port, with a head pressure of 2.1 bar giving approximately a 1 mL/min flow rate at 80 °C, and operated by using the program of hold at 80 °C for 8 min, program at 6 °C/min to 116 °C, and hold for 2 min.

The capillary column was used only on the Perkin-Elmer automated headspace analyzer. On both instruments the detector was operated at 280–300 °C, and the injection port was maintained at 220 °C. Argon–methane (95:5) was the carrier gas and was used on the headspace analyzer as a

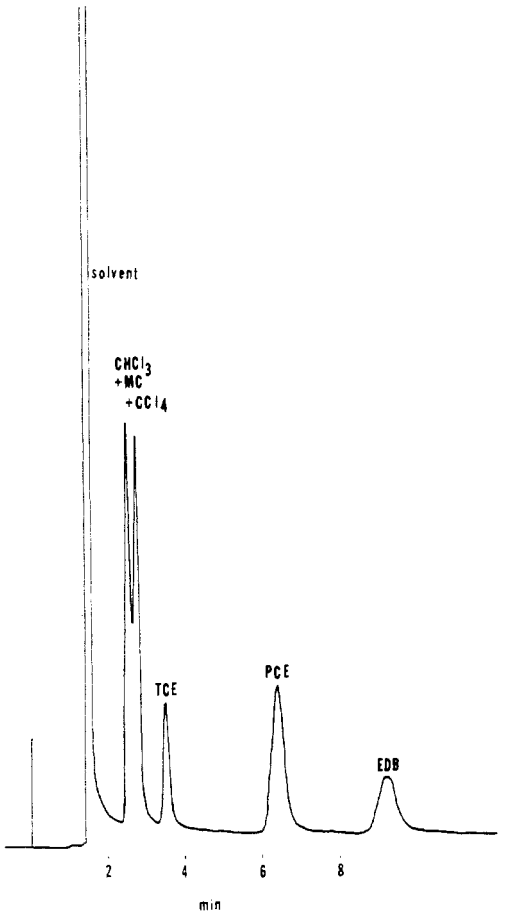


**Figure 1.** Retention of VHCs on column 1 (15% OV-17; 3.7 m; 85 °C). Injection of VHC standards in 2-propanol. Varian 3700 gas chromatograph; ECD, $10^{-11}$ A/mV; attenuation, 1024. $CHCl_3$, 4.2 ng; MC, 1.1 ng; $CCl_4$, 0.7 ng; TCE, 2.3 ng; PCE, 1.0 ng; EDB, 3.8 ng.

makeup gas at a flow rate of 60 mL/min. The headspace analyzer was set for a 9-s injection time for packed columns and 8 s for the capillary column so that approximately 2 mL of headspace was injected into the gas chromatograph at a head pressure of 2–3 bar on the packed columns.

**Quantitation.** VHC residues were quantitated by using external standards or by standard additions. External standards were made by fortifying vials containing uncontaminated foods identical with the sample. When uncontaminated foods were not available, quantitation by standard additions was necessary. The quantity of VHCs present was first estimated to 1 order of magnitude by comparing the response of the sample to that of an aqueous standard of VHCs. An additional, better estimate was derived from a single-point standard addition experiment, using as the standard twice the amount estimated by the preliminary analysis. For quantitation by standard additions, 0.25, 0.5, and 1.0 times the estimated amount of analyte were added individually to separate vials, each containing the same amount of the sample.

An internal standard was added to each vial to correct for variation in injection volume. 1,1,1,3-Tetrachlorotetrafluoropropane was the most useful compound for this purpose. Dibromomethane ($CH_2Br_2$) and 1,2-dibromoethane (EDB) were also used as internal standards; however, these compounds often coeluted with naturally derived sample components or contaminants of interest.

## RESULTS AND DISCUSSION

Headspace analysis was chosen for the determination of VHCs because the technique provided a degree of sample cleanup without loss due to volatilization. Concentration of the sample was not necessary for volatile hydrophobic compounds such as VHCs, because there was appreciable partitioning into the headspace for most matrices. Generally 50% or more of the analyte was contained in the headspace over a 1–2-g aqueous sample under the conditions described.

The use of the two GC columns was necessary to resolve certain pairs of compounds. Table I lists the retention ratios for selected VHCs. Columns 1 and 2 or 1 and 3 are complementary; that is, all the compounds listed can be separated on one or the other column. Figures 1–3 illustrate the chromatography of selected VHCs. Columns 2 and 3 give good separation of these compounds, while column 1 does not separate the first three. Column 1 was

Table I. Retention Ratios for Selected VHCs[a]

| compound | column 1 | column 2 | column 3 |
|---|---|---|---|
| chloroform | 1.00 | 1.00 | 1.00 |
| 1,1,1-trichloroethane | 1.05 | 1.20 | 1.15 |
| carbon tetrachloride | 1.10 | 1.35 | 1.27 |
| 1,2-dichloroethane | 1.35 | 1.13 | 1.07 |
| trichloroethylene | 1.41 | 1.57 | 1.44 |
| bromodichloromethane | 1.63 | 1.54 | 1.43 |
| dibromomethane | 1.89 | 1.52 | 1.38 |
| 1,1,1,3-tetrachlorotetrafluoropropane | | 1.67 | 1.84 |
| tetrachloroethylene | 2.56 | 3.02 | 2.25 |
| 1,2-dibromoethane | 2.42 | 2.79 | 2.11 |

[a] Column 1: 15% OV-17; 85 °C. Column 2: 20% SP-2100/0.1% Carbowax 1500; 100 °C. Column 3: 4X capillary OV-101; 80–116 °C. Retention times for $CHCl_3$ on columns 1-3 were 2.42, 2.53, and 7.33 min, respectively.

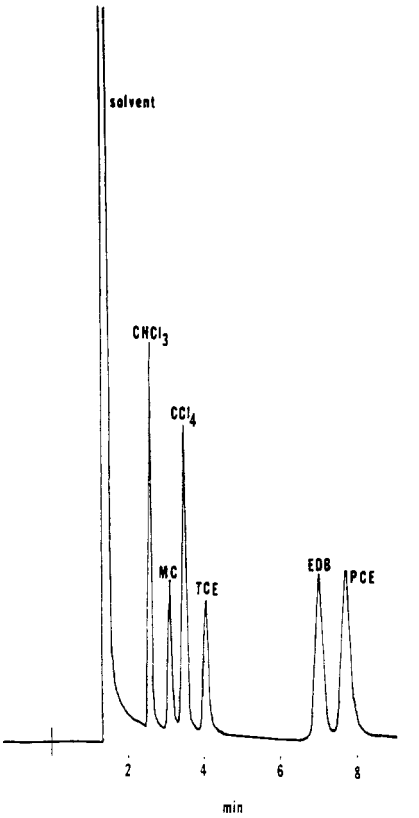


**Figure 2.** Retention of VHCs on column 2 (20% SP–2100/0.1% Carbowax 1500; 3 m; 100 °C). Injection of VHC standards in 2-propanol. Varian 3700 gas chromatograph; ECD, $10^{-11}$ A/mV; attenuation, 1024. $CHCl_3$, 3.0 ng; MC, 0.8 ng; $CCl_4$, 0.5 ng; TCE, 1.6 ng; EDB, 2.8 ng; PCE, 0.8 ng.

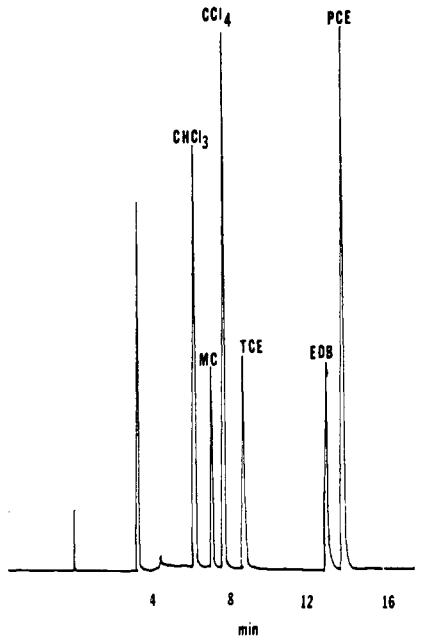


**Figure 3.** Retention of VHCs on column 3 (OV-101; 4X capillary; 50 m; 80–116 °C program). Headspace above 2 g of fortified noncola soft drink. Perkin-Elmer F-42 gas chromatograph; ECD, 3 nA; attenuation, 32; 8-s injection at 2.1 bar; 3.5:1 split ratio. $CHCl_3$, 815 ppb; MC, 192 ppb; $CCl_4$, 81 ppb; TCE, 362 ppb; EDB, 826 ppb; PCE, 154 ppb.

used for added evidence of the identity of VHCs and to separate certain compounds which coelute on columns 2 and 3. TCE, bromodichloromethane, and $CH_2Br_2$ can only be separated on column 1. In addition, some sample components, from butter, for example, interfere with the determination of TCE and $CCl_4$ on columns 2 and 3 but do not coelute with these compounds on column 1.

The maximum attainable temperature of the circulating water bath is 90 °C; this temperature was chosen for the equilibration of the samples and headspace to maximize the partitioning of VHCs into the headspace. Equilibrium was established in less than 1 h at this temperature for diluted, digested, or neat samples as indicated by no observed changes in the amount of analyte determined over periods ranging up to several hours.

Detection limits for the VHCs depend on the ECD response, the vapor pressure and water solubility of the analyte, and the nature of the food matrix. In general, compounds with boiling points less than 150 °C can be determined. Partitioning of the nonpolar VHCs into the headspace is not as favorable from fatty foods as from those containing little or no lipid. Partition coefficients (analyte concentration in headspace/analyte concentration in sample) decrease by factors of 5–20 for samples with high lipid contents (e.g., butter and corn oil) compared to aqueous samples. Detection limits for $CHCl_3$, MC, $CCl_4$, $CH_2Br_2$, TCE, and PCE in water were 0.19, 0.07, 0.02, 0.14, 0.23, and 0.04 ppb, respectively.

Detection limits can be improved by adding more sample to the vial. Improvements of up to about 3-fold are possible for undiluted samples by using up to 20 g of sample in a headspace vial rather than the usual 1–2 g. For samples requiring digestion, there is a practical limit of 5 g. One gram per vial is considered the minimum amount necessary for analysis.

Detection limits can also be increased by injecting larger volumes of headspace onto the column. This can be achieved by using a gas-tight syringe. Up to 5 mL has been injected on packed columns for confirmatory analysis by GC–MS. However, injection of large volumes can result in poor resolution of analyte peaks. In addition, when larger volumes are injected, the nonhalogenated volatile components in the sample, eluting as a "solvent front", can overload the column or detector and obscure early eluting peaks.

Table II. Determination of VHCs in Fortified Samples (1 g) of Mayonnaise

| compound | added, ng | determined, ng | % theoretical | $\sigma^a$ | $n^b$ |
|---|---|---|---|---|---|
| 1,1,1-trichloroethane | 26.0 | 22.9 | 88.0 | 3.0 | 3 |
| | 78.0 | 76.3 | 98.0 | 3.0 | 6 |
| | 156 | 155 | 99.5 | 0.7 | 2 |
| | 390 | 380 | 97.5 | 3.0 | 2 |
| carbon tetrachloride | 13.5 | 13.2 | 97.5 | 3.0 | 6 |
| | 27.0 | 21.8 | 80.5 | 0.7 | 2 |
| | 67.5 | 69.2 | 102 | 2.0 | 2 |
| trichloroethylene | 132 | 126 | 95.3 | 3.0 | 6 |
| | 264 | 264 | 100 | 3.0 | 2 |
| | 660 | 632 | 95.8 | 1.0 | 2 |
| tetrachloroethylene | 39.0 | 38.5 | 98.6 | 3.0 | 6 |
| | 78.0 | 73.6 | 94.3 | 1.0 | 2 |
| | 195 | 180.5 | 92.5 | 3.0 | 2 |

[a] $\sigma$ = standard deviation of the mean percent theoretical. [b] $n$ = number of replicates.

Problems with quality control of headspace vials, septa, and crimp tops have been noted. Leaks sometimes made quantitation impossible. In the worst cases, roughly 10% of the sealed vials leaked. This occurred particularly when vials, septa, and crimp tops from different suppliers were intermixed.

The reproducibility of injections depended to some extent on the nature of the analyte and sample. The largest variation was observed in aqueous samples. Large variations were also noted in samples for which homogeneity is difficult to obtain. Ten repetitive injections of headspace from vials containing a fortified whole milk sample were made by using the automated headspace analyzer over a 2-day period (five injections per day). Relative standard deviations for the integrated peak areas ranged from 5% for $CHCl_3$ and PCE to 13% for $CCl_4$. In general, relative standard deviations for calculated analyte concentrations were 10–20%.

The accuracy and precision of the method are illustrated for mayonnaise. Samples were independently fortified in this laboratory, sealed in crimp-top vials, and analyzed as unknown samples by using external standards. Table II summarizes the results of the experiment; in general, 90–100% of the theoretical quantity was determined. The small standard deviation of the percent theoretical determined in the analysis of fortified mayonnaise samples (1–3% absolute) appears to be due to the high lipid content in the sample. Larger standard deviations would be expected for fortified aqueous samples, as indicated for the fortified whole milk sample above.

The general technique is adaptable to various types of foods. In some cases, digestion with sulfuric acid was employed to facilitate the release of VHCs from fatty foods. With other foods (e.g., jelly), water was added as a diluent to disperse the sample and assure better reproducibility. When digestion or dilution was not necessary, the sample was analyzed neat. In the latter case, lower detection limits were generally achieved.

Digestion of the sample does not, however, assure homogeneity. The digestion process with sulfuric acid does not affect lipids present in the sample. Protein is digested by the sulfuric acid, and the lipid, though sometimes charred, is found floating in a layer on the digestion medium. No measurable degradation of the VHCs has been

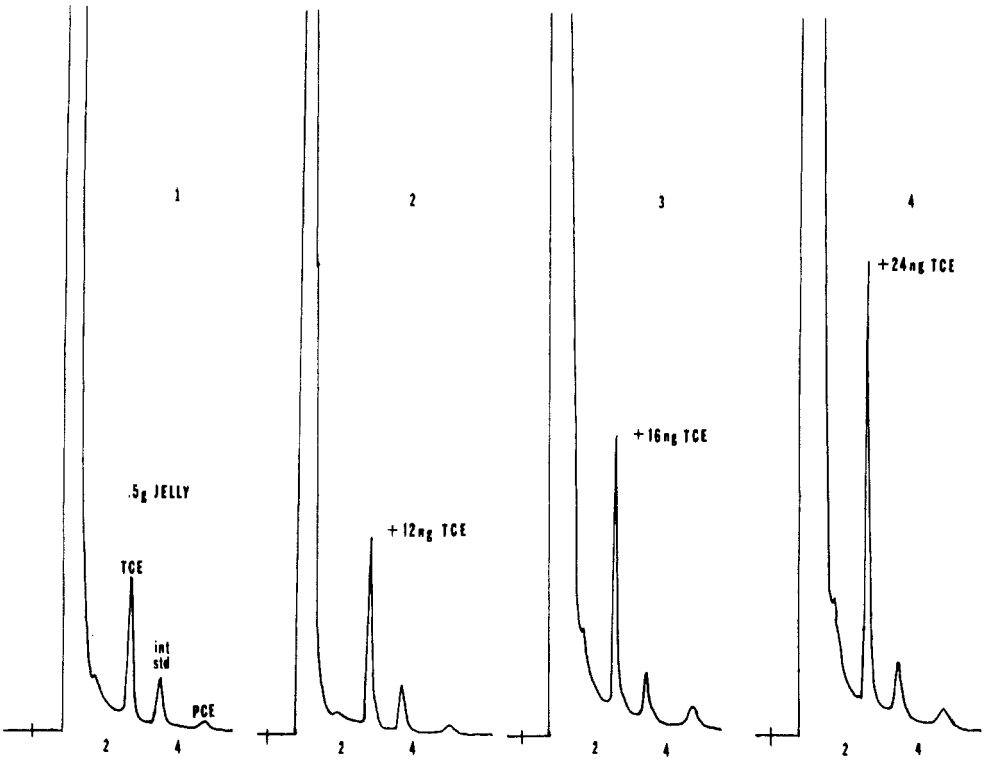


**Figure 4.** Analysis of crab apple jelly using the method of standard additions. Column 1 (15% OV-17; 3.7 m; 90 °C). (1) 0.5 g of jelly in 5 mL of water, (2) 12 ng of TCE added, (3) 16 ng of TCE added, and (4) 24 ng of TCE added.

observed through this process. Equilibrium is established for this multiphase system and quantitative results are obtained. Digestion can also produce volatile nonhalogenated compounds which can interfere with analysis; thus, to lower detection limits, some samples must be analyzed without digestion (e.g., cheese). In practice, the procedure can be adapted to give a compromise among the lowest detection limits, best reproducibility, and least interferences in the determinative step.

Several different types of foods have been analyzed by using this procedure. Fish from various U.S. waterways have been found to contain VHCs. In general, low levels (10–100 ppb) of volatiles such as $CHCl_3$, MC, TCE, and PCE have been detected in raw fish. A few fish samples, which were collected near industrial sites, contained higher levels of PCE (ppb): American eel (Delaware River), 250 and (Newark Bay) 1050; carp (Delaware River), 77; striped bass (Raritan River), 108; spot fish (Houston Ship Channel), 88 plus 220 ppb of $CHCl_3$.

Strawberry and maple walnut ice cream from a Boston food processor contained 10 and 16 ppb of MC, respectively. Plant tap water, Chinese style sauce, quince jelly, crap apple jelly, grape jelly, and chocolate sauce, all from a food processor in Pennsylvania, contained TCE at the following levels (ppb): 68, 28, 40, 25, 20, and 50, respectively. PCE was found in the same samples at the following levels (ppb): 0.4, 2, 2.2, 2.5, 1.6, and 3.6.

GC–MS confirmation was obtained for TCE in the tap water and quince jelly samples from Pennsylvania. Full-scan mass spectra of the TCE component were essentially identical with reference spectra (levels of 100 ppb were necessary).

Figure 4 illustrates the analysis of a crab apple jelly sample using quantitation by standard additions. The concentration of TCE residue was 25 ppb, and the PCE residue, though below accurate quantitation levels, was estimated to be 2.5 ppb.

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