

Study of a Commercial Liquid Smoke Flavoring by Means of Gas Chromatography/Mass Spectrometry and Fourier Transform Infrared Spectroscopy

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A commercial aqueous smoke preparation was extracted exhaustively using dichloromethane as solvent, until the carrier totally lost its smoky odor. Qualitative, and quantitative, characterizations of the extract were performed by means of gas chromatography/mass spectrometry and gas chromatography with flame ionization detection, respectively. Some of the compounds identified, to the best of our knowledge, had not been detected before in wood smoke. Neither benzo[*a*]pyrene nor other polycyclic aromatic compounds with carcinogenic properties were found in this liquid smoke. Alkyl and carbonyl derivatives of syringol and guaiacol constituted approximately 65% of the total volatile fraction of the dichloromethane extract. Syringol derivatives were present in a higher concentration than guaiacol derivatives (ratio 3:1). The Fourier transform infrared spectroscopy technique was tested to study its usefulness for this kind of sample. From the IR spectrum can be obtained qualitative and quantitative information about the main functional groups as well as about the dominant aromatic substitution pattern of the mixture without previous fractionations or separations and with a great saving of time.

Keywords: *Commercial liquid smoke; gas chromatography/mass spectrometry; Fourier transform infrared spectroscopy*

INTRODUCTION

Commercial smoke flavorings used in the food industry have different organoleptic features, due to their different compositions. Many factors exercise an important influence in the composition of these mixtures. Smoke condensates coming from the same wood and the same smoke generation conditions can be fairly different depending on the process followed in the manufacture.

Commercial smoke flavorings have been identified by several techniques. Fractionation of a smoke condensate ether extract with basic, acid, phenolic, carbonyl, and non-carbonyl fractions has been carried out (Fuji-maki et al., 1974). The use of preparative gas-liquid chromatography (Radecki et al., 1976; Radecki and Grzybowski, 1981) or steam distillation followed by extraction with ethyl ether (Radecki et al., 1977) has been reported. Likewise, acetone (Fiddler et al., 1970a) and freon 11 (Potthast and Eigner, 1988) have been used to extract aromatic volatile components of smoke condensates. Separation of the phenol fraction, by treatment with a sodium borate solution of smoke aroma preparations followed by GC/MS study, has also been reported (Wittkowski et al., 1981). Recently, the use of a headspace technique has allowed the identification of aliphatic alcohols, aldehydes, ketones, esters, furan derivatives, alkyl-substituted benzene derivatives, and guaiacol among the volatile components of a commercial smoke condensate (Wittkowski et al., 1990). In most of these papers, no quantitative data about the identified compounds were given.

The use of liquid smoke flavorings does not always give satisfactory results. For this reason, knowledge of their composition is important. The characterization of commercial smoke flavorings is necessary not only for the study of relationships between their composition and their organoleptic properties but also to help improve procedures for the preparation of smoke condensates.

From the health point of view it is essential to study the presence of benzo[*a*]pyrene and other carcinogenic polycyclic aromatic compounds in commercial preparations. For these reasons, in this paper we report the qualitative and quantitative characterization by gas chromatography/mass spectrometry (GC/MS) and by gas chromatography (GC), respectively, of the fraction soluble in CH₂Cl₂ of a commercial liquid smoke. Also, this smoke flavoring was studied by Fourier transform infrared spectroscopy (FTIR) to test the utility of this technique for characterizing this type of sample. IR has been previously used only to identify components of the smoke flavoring, obtained by steam distillation or preparative chromatography (Radecki et al., 1976; Radecki and Grzybowski, 1981; Fiddler et al., 1970b) but not for the characterization of the overall aromatic fraction.

MATERIALS AND METHODS

Sample, Solvents, and Standard Compounds. The sample studied was a water-based smoke flavoring used in the Spanish food industry. The extraction of the volatile compounds was carried out with CH₂Cl₂. This organic solvent was selected for its high effectiveness to extract polycyclic aromatic compounds (Guillén et al., 1991; Guillén, 1994) and aromatic compounds in general, its high volatility, and its absorption in infrared spectroscopy only at 954 and 714-680 cm⁻¹. As standard compounds for identification of some components and for the gas chromatographic quantification we used 2-methoxyphenol, 2,6-dimethoxyphenol, 4-hydroxy-3,5-dimethoxybenzaldehyde, 1,4-dimethoxybenzene, 1,2,3-trimethoxybenzene, 1,3,5-trimethoxybenzene, 2-methyl-2-cyclopenten-1-one, 3-methyl-2-cyclopenten-1-one, 3-methyl-1,2-cyclopentanedione, 2-furancarboxaldehyde, and 5-methyl-2-furancarboxaldehyde (from Aldrich and Fluka).

Extraction, GC/MS, and GC. A sample of 20.1 g of liquid smoke was exhaustively extracted with dichloromethane until the carrier totally lost its smoky odor. The solvent was

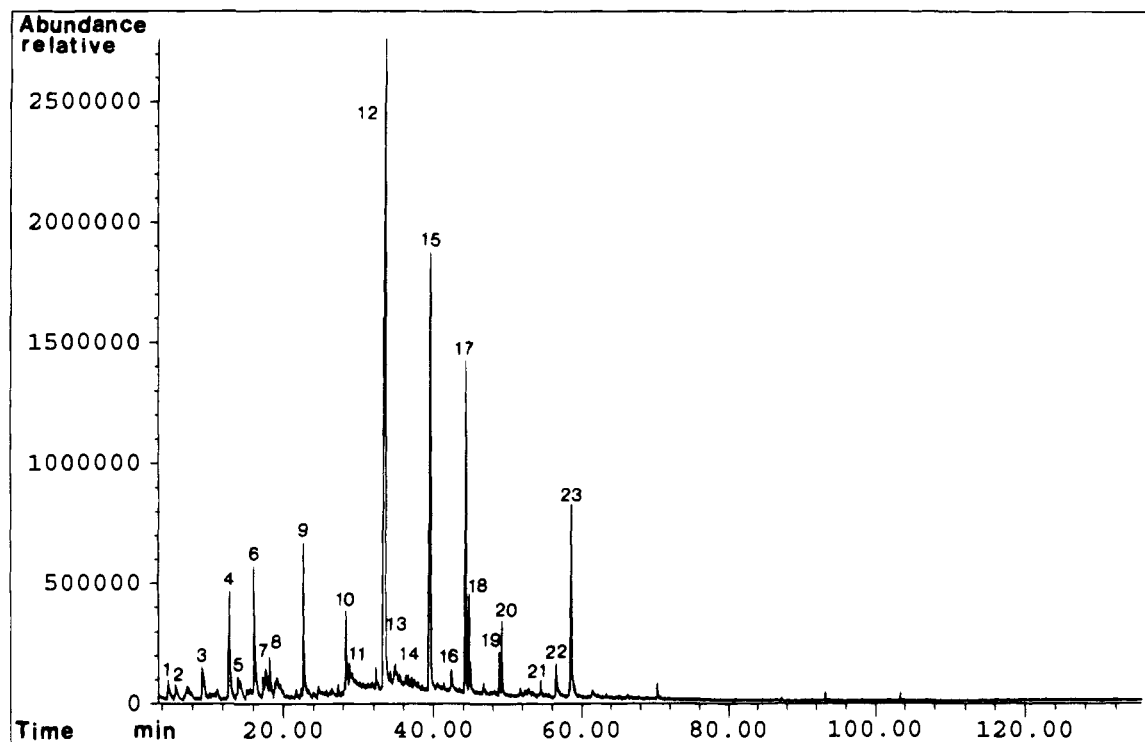


Figure 1. Total ion chromatogram of the soluble fraction in CH_2Cl_2 of a commercial liquid smoke.

partially evaporated under vacuum in a rotary apparatus, using mild and careful conditions to avoid the loss of very volatile compounds, until a volume of 10 mL was reached, and this was kept in a refrigerator. Components with higher or similar volatility to CH_2Cl_2 were not analyzed.

The GC/MS employed a Hewlett-Packard chromatograph, Model 5890 Series II, equipped with a mass spectrometer selective detector 5971 (MS), and a Hewlett-Packard Vectra 486/66U computer. A fused-silica capillary column (30 m long, 0.25 mm diameter), coated with a nonpolar stationary phase (Hewlett-Packard 5, cross-linked 5% phenyl methyl silicone) was used. The temperature program began at 50 °C (0.5 min) and increased 2 °C/min until 290 °C was reached (10 min). Helium was used as gas carrier. Injector and detector temperatures were 250 and 280 °C, respectively. The injection technique used was splitless. The volume of sample injected was close to 1 μL . Mass spectra were recorded at an ionization energy of 70 eV. Components were identified by their retention times, by their mass spectra, by comparing their mass spectra with those in a commercial library (Wiley 138K, Mass Spectral Database, 1990), and in some cases by using standards, as in our previous studies (Blanco et al., 1991; Guillén et al., 1992a).

A Hewlett-Packard gas chromatograph Model 5890 Series II, equipped with a flame ionization detector (FID) and a Hewlett-Packard 3395 integrator, was used for the quantitative study. A fused-silica capillary column (30 m long, 0.32 mm i.d.), coated with a nonpolar stationary phase (Hewlett-Packard 5), was used. The temperature of the detector was 300 °C. The other conditions of the chromatographic run were the same as those used in GC/MS. As external standards for the quantification 3-methyl-1,2-cyclopentanedione, 2-methoxyphenol, 2,6-dimethoxyphenol, 4-hydroxy-3,5-dimethoxybenzaldehyde, and 5-methyl-2-furancarboxaldehyde were used.

Infrared Spectra. FTIR spectra were obtained of the liquid smoke soluble fraction in CH_2Cl_2 using a NaCl demountable liquid cell, with a path length of 0.15 mm. FTIR spectra were recorded on a Nicolet Magna 550 spectrometer with a DTGS KBr detector, co-adding 32 interferograms obtained at a resolution of 4 cm^{-1} . The spectrometer was operated with Nicolet OMNIC software, and the spectra were corrected for scattering.

Each stage of this experimental section was performed several times to obtain accurate results.

RESULTS AND DISCUSSION

The liquid-liquid extraction was exhaustively performed until the water lost its smoky odor; for this reason, the volatile fraction studied can be considered representative of the overall aromatic fraction.

GC/MS and GC Characterization. Taking into account that the flavoring here studied was a commercial sample of unknown composition, the first task was the identification of its volatile flavoring components. For this reason, the GC/MS technique was selected. Because the mass spectrometer detector is not very sensitive in mode scan (10 ng), injections at several concentrations were carried out. However, injections of slightly concentrated samples give rise to changes in the elution order of some compounds of this complex mixture. On the other hand, GC/MS provides the identification of compounds even when the chromatographic separation is not sufficient to afford an accurate quantification (Wittkowski et al., 1981). Figure 1 shows the total ion chromatogram of the liquid smoke soluble fraction in CH_2Cl_2 . Table 1 lists the compounds tentatively identified, grouped in families. The number corresponding to each peak in Figure 1 corresponds to the peak numbers given in Table 1. Components were identified by their retention times, by their mass spectra, by comparing their mass spectra with those in a commercial library (Wiley 138K, Mass Spectral Database, 1990), and in some cases by using standards. Concentration in the flavoring (grams per kilogram) of the major compounds is given in Table 1.

Among the substances tentatively identified are compounds arising from the thermal degradation of the three main classes of wood components, i.e. carbonyls, furans, and pyran derivatives, as well as some compounds that could mainly arise from cellulose and hemicellulose pyrolysis (Fiddler et al., 1967). Phenol, guaiacol, and syringol derivatives and other compounds could principally arise from the pyrolysis of lignin (Kim et al., 1974). Figure 2 shows structures of four signifi-

Table 1. Compounds Tentatively Identified in the Soluble Fraction of the Liquid Smoke in CH₂Cl₂, Together with the Peak Number in Figure 1 and Concentration in the Flavoring of the Principal Compounds

Family: compound (peak number) (g/kg)
phenol derivatives: phenol, 2-methylphenol, 3-methylphenol, 4-methylphenol, 2-ethylphenol, 3-ethylphenol, 4-ethylphenol, 2,3-dimethylphenol, 2,4-dimethylphenol, 2,5-dimethylphenol, 3,4-dimethylphenol, 2-methoxyphenol (guaiacol) (6) (0.07), 2-methoxy-3-methylphenol, 2-methoxy-4-methylphenol (9) (0.06), 2-methoxy-4-ethylphenol (10) (0.08), 2-methoxy-4-propylphenol (13) (0.01), 4-hydroxy-3-methoxybenzaldehyde (vanillin) (14) (0.01), 1-(4-hydroxy-3-methoxyphenyl)ethanone (16) (0.02), 1-(4-hydroxy-3-methoxyphenyl)-1-propanone (0.01), 1-(4-hydroxy-3-methoxyphenyl)-2-propanone* (18) (0.02), 2-hydroxy-5-methoxybenzaldehyde*, 2,6-dimethoxyphenol (syringol) (12) (0.48), 2,6-dimethoxy-4-methylphenol (15) (0.18), 2,6-dimethoxy-4-ethylphenol (17) (0.08), 2,6-dimethoxy-4-(1-propenyl)phenol (19) (0.01), 2,6-dimethoxy-4-(1-propenyl)phenol (isomer) (0.01), 2,6-dimethoxy-4-propylphenol (20) (0.01), 4-hydroxy-3,5-dimethoxybenzaldehyde (21) (0.03), 1-(4-hydroxy-3,5-dimethoxyphenyl)ethanone (22) (0.03), 1-(4-hydroxy-3,5-dimethoxyphenyl)-1-propanone (0.02), 1-(4-hydroxy-3,5-dimethoxyphenyl)-2-propanone* (23) (0.02), 3,4-dimethoxyphenol, 4,5-dimethoxy-2-methylphenol*, 2-methyl-1,3-dihydroxybenzene*, 3-methyl-1,2-dihydroxybenzene, 2-methoxy-1,4-dihydroxybenzene*, 3-methoxy-1,2-dihydroxybenzene (11) (0.05)
uran and pyran derivatives: 2-furancarboxaldehyde (1), 1-(2-furanyl)ethanone, 5-methyl-2-furancarboxaldehyde (3), 3-hydroxy-2-methyl-4H-pyran-4-one (maltol) (7) (0.03), 3-hydroxy-2,6-dimethyl-4H-pyran-4-one*
ketones: 2-cyclopenten-1-one, 4-hydroxy-4-methyl-2-pentanone (1), 1-(acetyloxy)-2-propanone (2) (0.01), 2-methyl-2-cyclopenten-1-one, 3-methyl-2-cyclopenten-1-one** (3), 1-cyclopentylethanone*, 3-ethyl-2-cyclopenten-1-one, 5-ethyl-2,5-dimethyl-2-cyclopenten-1-one, 5,5-dimethyl-3-(1-methylethyl)-2-cyclohexen-1-one*
lactones: dihydro-2(3H)-furanone, 2(5H)-furanone, 3-methyl-2(5H)-furanone*
diketones: 3-methyl-2,4-pentanedione, 3-methyl-1,2-cyclopentanedione (cyclotene) (4) (0.15), 3,5-dimethyl-1,2-cyclopentanedione* (5) (0.04), 5,5-dimethyl-1,2-cyclopentanedione*, 3-ethyl-1,2-cyclopentanedione (8) (0.02), 5-ethyl-3-methyl-1,2-cyclopentanedione*, 2-ethyl-2,5-dimethyl-1,2-cyclopentanedione*, 5-ethyl-5-methyl-1,2-cyclopentanedione*
alkyl aryl ethers: 1,4-dimethoxybenzene, 1,2,3-trimethoxybenzene**, 1,3,5-trimethoxybenzene**, 3,4-dimethoxytoluene, 1,2,3-trimethoxy-5-methylbenzene*
hydrocarbons: toluene

cant compounds found in the sample: 3-methyl-1,2-cyclopentanedione or cyclotene (I), 3-hydroxy-2-methyl-4H-pyran-4-one or maltol (II), 2-methoxyphenol or guaiacol (III), and 2,6-dimethoxyphenol or syringol (IV).

Among the main components of this flavoring are the well-known alkyl derivatives of syringol and guaiacol. Thus, 2,6-dimethoxyphenol (0.48 g/kg), 2,6-dimethoxy-4-methylphenol (0.18 g/kg), 2,6-dimethoxy-4-ethylphenol (0.08 g/kg), 2,6-dimethoxy-4-propylphenol (0.01 g/kg), *cis* and *trans* isomers of 2,6-dimethoxy-4-(1-propenyl)phenol (0.03 g/kg) were found. All of these derivatives have been considered as key compounds responsible for the antioxidant properties of liquid smoke (Maga, 1988). 2-Methoxyphenol (0.07 g/kg), 2-methoxy-4-methylphenol (0.06 g/kg), 2-methoxy-4-ethylphenol (0.08 g/kg), and 2-methoxy-4-propylphenol (0.01 g/kg) were also present. The concentration of syringol and its alkyl derivatives was 3 times higher than that of guaiacol and its alkyl derivatives. From these and previous studies (Baltes et al., 1981; Potthast, 1988), it can be assumed that the source of this liquid smoke was hardwood.

Carbonyl compounds derived from guaiacol and syringol are also present in the sample, and they have been well separated in the chromatographic run. 1-(4-Hydroxy-3-methoxyphenyl)ethanone (0.02 g/kg), 1-(4-hydroxy-3-methoxyphenyl)-2-propanone (0.02 g/kg), 1-(4-hydroxy-3-methoxyphenyl)-1-propanone (0.01 g/kg), 1-(4-hydroxy-3,5-dimethoxyphenyl)ethanone (0.03 g/kg), 1-(4-hydroxy-3,5-dimethoxyphenyl)-2-propanone (0.02 g/kg), and 1-(4-hydroxy-3,5-dimethoxyphenyl)-1-propanone (0.01 g/kg) as well as vanillin (0.01 g/kg) and syringaldehyde (0.03 g/kg) have been identified. Syringol and guaiacol

derivatives represent approximately 65% of the total weight of the volatile fraction of the sample.

In addition to alkyl derivatives of phenol, which are present in not very high concentrations, another group of compounds, which are interesting because of their strong antioxidant effects, are dihydroxybenzene derivatives, which are also present in small concentrations.

A large number of aliphatic compounds containing carbonyl groups are constituents of this liquid smoke, even though mostly in small concentrations. In this group of derivatives are compounds arising from monosaccharide thermal reactions, such as 2-furancarboxaldehyde, 3-hydroxy-2-methyl-4H-pyran-4-one, and 3-methyl-1,2-cyclopentanedione. Compounds having two carbonyl groups can occur not only in the dicarbonyl form but also in the keto-enol form. Among these compounds, cyclotene, which is present in a concentration of 0.15 g/kg, has been considered to have significant organoleptic and preserving properties (Radecki et al., 1976).

Some alkyl aryl ethers occur in the sample, and only one hydrocarbon was detected. Noteworthy is the presence of some derivatives not previously detected in wood smoke (Maga, 1988), as is the almost total absence of hydrocarbons. Compounds not previously detected are asterisked in Table 1. Compounds with two asterisks were identified by using commercially available standards. Even though carcinogenic polycyclic aromatic hydrocarbons have been detected in several commercial liquid smoke preparations, even in high concentrations (Yabiku et al., 1993; Gomaa et al., 1993), in the liquid smoke studied here this kind of compounds has not been detected.

In relation to sensory properties, phenols of pungent cresolic, sweet, smoky, and burnt odors (Kim et al., 1974; Baltes et al., 1981; Toth and Potthast, 1984) are present, and compounds with odors described as sweet (2-furancarboxaldehyde and 5-methyl-2-furancarboxaldehyde) or grassy, sweet-spicy (3-methyl-2-cyclopenten-1-one) (Kim et al., 1974) are also present and contribute to the overall aroma.

In this context, this liquid smoke contains the majority of the components identified in another commercial

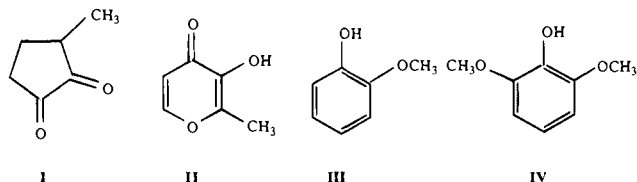


Figure 2. Structures of four significant compounds found in the sample: 3-methyl-1,2-cyclopentanedione or cyclotene (I); 3-hydroxy-2-methyl-4H-pyran-4-one or maltol (II); 2-methoxyphenol or guaiacol (III); and 2,6-dimethoxyphenol or syringol (IV).

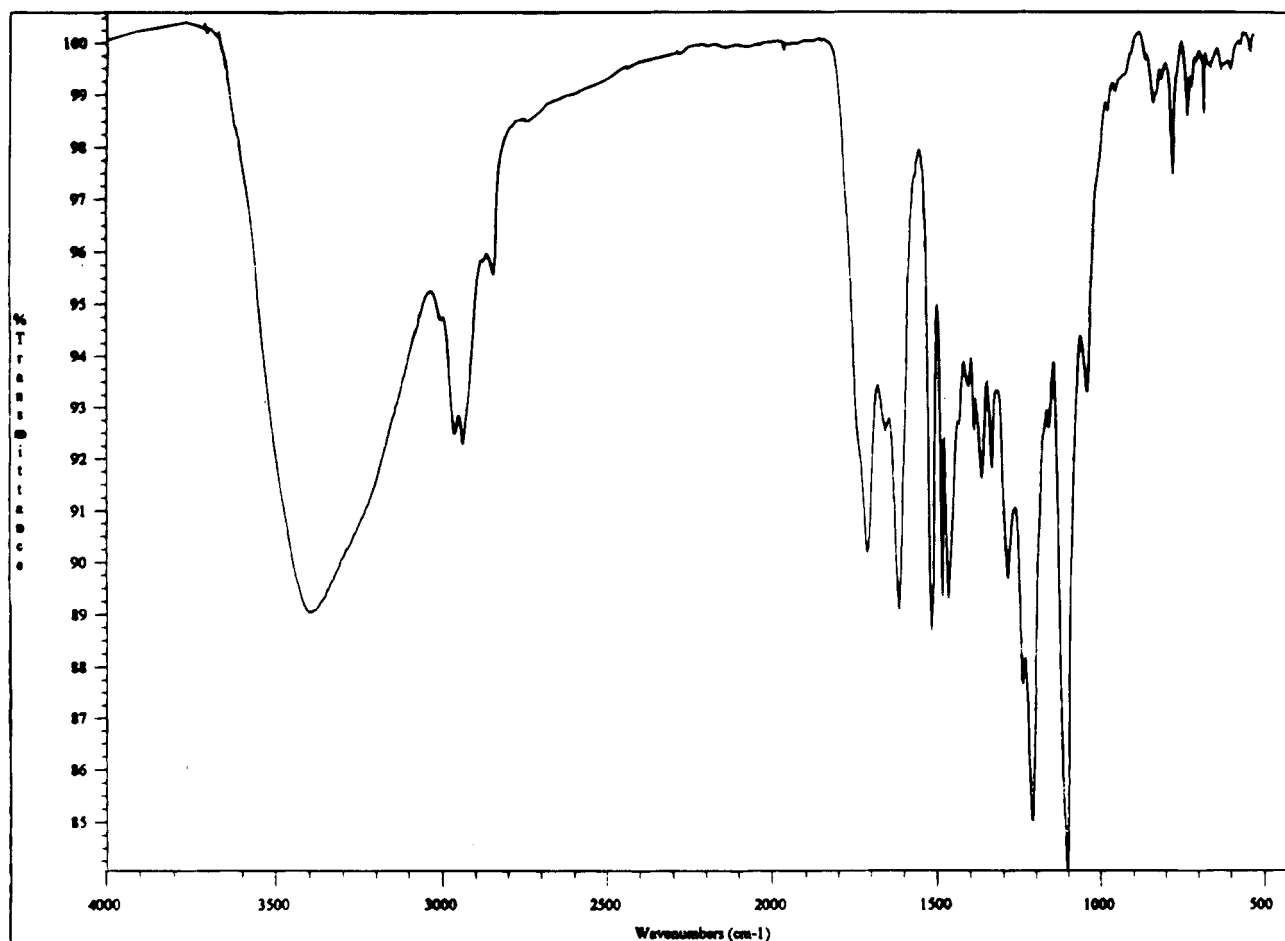


Figure 3. Infrared spectrum from 4000 to 500 cm^{-1} of the soluble fraction in CH_2Cl_2 of the liquid smoke.

smoke flavoring, highly appreciated owing to its organoleptic features (Radecki et al., 1977), and also contains the majority of the derivatives identified in a fraction considered to contain the essential compounds of smoke flavor (Fiddler et al., 1970).

Fourier Transform Infrared Spectroscopy Study.

It is well accepted that the characteristic smoky flavor does not appear to be limited to one class of compounds but is due to the blend of compounds (Fujimaki et al., 1974; Olsen, 1976). Carbonyl compounds contribute to modify the harsh phenol flavor, and the characteristic aroma of each mixture is due to the proportion of the compounds contained in it (Fiddler et al., 1970). Fourier transform infrared spectroscopy provides information about the occurrence of functional groups in simple substances or in mixtures of products and is a very versatile technique that has been little used to obtain information about smoke flavorings.

Figure 3 shows the FTIR spectrum of the liquid smoke soluble fraction in CH_2Cl_2 . This shows a broad band absorption with a maximum at 3407 cm^{-1} due to the intermolecular hydrogen bonded OH stretching vibration of phenol groups. This band can include absorptions of the O-H bonds of carbonyl compounds, which can undergo keto-enol tautomerism. However, this contribution is small because of the small concentration of these compounds.

Aromatic C-H stretching vibration is observed as a shoulder at 3010 cm^{-1} , showing the small concentration of aromatic C-H bonds in the sample. The C-H olefinic stretching vibration could also contribute to this absorption, but this contribution is negligible due to the small

proportion of this type of bond in the mixture. Both the methyl (methylene) asymmetrical stretching band at 2963 cm^{-1} (2939 cm^{-1}) and the methyl (methylene) symmetrical stretching band at 2872 cm^{-1} (2850 cm^{-1}) are clearly shown. Methyl groups of ether compounds absorb at about 2850 cm^{-1} , while at 2734 cm^{-1} there is the aldehydic C-H stretching band due to the Fermi resonance (Günzler and Böck, 1975).

At 1712 cm^{-1} appears a pronounced stretching band of carbonyl groups in aldehydes, ketones, and α -diketones. Shoulders at 1737 and 1721 cm^{-1} are also present and can be assigned to the C=O group of furancarboxaldehyde and 5-methylfurancarboxaldehyde. The small proportion of lactones, which show bands above 1740 cm^{-1} due to C=O stretching, could explain the absence of peaks at this frequency.

Absorptions at 1615, 1516, and 1483 cm^{-1} are mainly assigned to carbon-carbon ring stretching vibration of aromatic compounds (Günzler and Böck, 1975). However, the band at 1464 cm^{-1} could be due to asymmetrical methyl bending and/or scissoring methylene bending vibrations, while the absorption observed at 1364 cm^{-1} could be due to symmetrical methyl bending vibration. The region of the spectrum between 1037 and 1331 cm^{-1} is not helpful.

The zone between 900 and 700 cm^{-1} shows maxima at 825, 767, and 719 cm^{-1} . Bands in this zone can be mainly attributed to aromatic C-H bending vibration out of plane, though some heterocycles also show absorptions in this region, and CH_2Cl_2 has a band at 714–680 cm^{-1} . Taking into account the predominance of benzene derivatives, bands in this range can be useful

to disclose the predominant substitution patterns of the aromatic ring in the mixture. Aromatic rings with five or four neighboring hydrogen atoms show the most intense band at 770–730 cm^{-1} . Compounds of this group are phenol (746 cm^{-1}), guaiacol, and 2-methylphenol (748 and 747 cm^{-1}). The spectrum (Figure 3) only shows a shoulder at 750 cm^{-1} and therefore could explain the small proportion of compounds having this type of substitution pattern. Aromatic rings with three adjacent hydrogen atoms show one intense absorption band between 810 and 750 cm^{-1} . Compounds of this group are syringol (768 cm^{-1}) and 2,6-dimethylphenol (765 cm^{-1}). Figure 3 shows a strong absorption at 767 cm^{-1} , indicating that these compounds are present in agreement with GC/MS results.

Finally, aromatic rings with two adjacent or isolated hydrogen atoms give bands from 800 to 900 cm^{-1} . Aromatic compounds such as 2-methoxy-4-ethylphenol (808–816 cm^{-1}), 2-methoxy-4-(2-propenyl)phenol (817–802 cm^{-1}), 2,6-dimethoxy-4-(2-propenyl)phenol (827–802 cm^{-1}), and 2,6-dimethoxy-4-methylphenol (818–794 cm^{-1}) absorb in this range. The observed band at 825 cm^{-1} (Figure 3) mainly represents this type of substitution pattern in the sample.

FTIR is an interesting technique also from a quantitative point of view. The Beer–Lambert law relates the observed intensity (A) of a given band and the concentration (c) of the corresponding functional group through the equation $A = \sum \rho \epsilon c$, where ϵ is the absorption coefficient of each functional group in the molecule and ρ is the sample thickness. The problem in the quantitative analysis of complex mixtures is the determination of absorption coefficients. The basis for these approximations is that each functional group has its own intensity, which does not vary drastically from molecule to molecule when they have a similar environment (George and McIntyre, 1987; Guillén et al., 1992b).

Taking into account all of the above, the absorptions related to specific functional groups measured through the area of each specific band of several liquid smoke samples give valuable information about the concentration of these functional groups in the sample. That is, semiquantitative concentration data can be obtained of phenol and carbonyl functional groups and aliphatic and aromatic C–H bonds of the liquid smoke here studied. The dominant substitution pattern in the aromatic ring, as well as the predominance of syringol or guaiacol derivatives in the sample, can be deduced with great time saving compared with other techniques. In other words, this technique can give the proportion of the different functional groups in the sample in an approximate way, without either fractionations or separations, in a short period of time.

In conclusion, it is evident that the infrared spectra give valuable information about the commercial liquid smoke studied. The functionalities found by means of FTIR are in agreement with those found by GC/MS, except for compounds at very small concentrations. From the GC/MS and GC studies it can be concluded that the major flavoring components of this liquid smoke are guaiacol and syringol derivatives, together with a small proportion of carbonyl compounds, mainly cyclohexene.

ACKNOWLEDGMENT

This work has been supported by the Comisión Interministerial de Ciencia y Tecnología (CICYT, ALI93-0467 and ALI94-0989). We thank the editor and

referees for useful suggestions and remarks. We also thank E. Peman, APASA, Astigarraga (Guipuzcoa), for a gift of commercial liquid smoke.

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Received for review March 28, 1994. Revised manuscript received October 21, 1994. Accepted November 4, 1994.*

JF940159T

* Abstract published in *Advance ACS Abstracts*, December 15, 1994.