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REVIEW

Pyridines in Foods

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Pyridines represent a class of compounds that can significantly contribute to the organoleptic properties of foods. This review summarizes the pyridine compounds identified in various food systems, their flavor properties, and their formation in model systems.

Numerous types of heterocyclic compounds have been associated with the flavor properties of food. However, one such class that has received comparatively little attention is the pyridines. In fact, no flavor research publication has been devoted exclusively to their investigation. For the most part they have been found in the basic fraction of food volatiles which is usually dominated by pyrazines. In addition, aside from several patent claims, their flavor properties have not been extensively reported.

Thus, the primary objectives of this review are to summarize pyridine compound food occurrences, to report on their flavor properties, and to discuss their formation in model systems relative to the literature.

FOOD OCCURRENCES

Compared to other important classes of compounds associated with food flavor, relatively few pyridines have been identified as being naturally occurring. Likewise, they have been found in a relatively small number of foods including meat and milk products, alcoholic and non-alcoholic beverages, nuts, cereals, vegetables, and several miscellaneous products. A detailed listing of the food and

related systems in which pyridines have been identified is summarized in Table I. In reviewing the foods in which pyridines have been identified, all have either undergone some degree of microbial activity or thermal treatment, which would indicate that pyridines may not exist in unprocessed foods.

FLAVOR PROPERTIES

Since no extensive investigations have been devoted to food-associated pyridines, little specific organoleptic data are available. The only exception appears in the patents of Winter et al. (1972, 1975, 1976a,b) in which various pyridine derivatives were included among the 38 compound classes reported to have potential flavor applications. The descriptive terminologies summarized in Table II resulted when the compounds in question, at concentrations varying from 0.005 to 5 g/100 L, were added to either a 65% sucrose solution or a 1.35% instant coffee. As can be seen, most of the compounds evaluated had green, bitter, astringent, roasted, or burnt properties, although some had more pleasant and characteristic sensations.

The practical application of pyridines as food flavorings can perhaps best be appreciated by viewing the data presented in Table III. This represents a summation of the pyridines on the Food and Drug Administration (FDA)

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Table I. Food and Related Systems in Which Pyridines Have Been Identified

system	reference
artichoke	Buttery et al. (1978)
asparagus	Tressl et al. (1977)
barley	Wang et al. (1969); Shimizu et al. (1970); Collins (1971); Harding et al. (1978)
beans	Buttery et al. (1975)
beef	Watanabe and Sato (1971); Coppock and MacLeod (1977); MacLeod and Coppock (1976, 1977)
beer	Harding et al. (1977)
casein	Ferretti et al. (1970); Ferretti and Flanagan (1971)
cheese	Dumont et al. (1974a-c, 1975); Dumont and Adda (1978)
chicken	Horvat (1976)
cocoa	Vitzthum et al. (1975a)
coffee	Viani et al. (1965); Gianturco et al. (1966); Reymond et al. (1966); Goldman et al. (1967); Stoffelsma et al. (1968)
eggs	Kato et al. (1978)
filberts	Kinlin et al. (1972)
jasmin	Toyoda et al. (1978)
lamb	Buttery et al. (1977)
orange flowers	Sakurai et al. (1979)
peanuts	Shu and Waller (1971); Walradt et al. (1971)
pecans	Wang and Odell (1972)
peppermint oil	Takahashi et al. (1980)
popcorn	Walradt et al. (1970)
potato chips	Deck and Chang (1965); Buttery et al. (1971)
rice	Tsugita et al. (1978); Withycombe et al. (1978); Yajima et al. (1978)
rum	Wobben et al. (1971)
soy	Nunomura et al. (1978); Liardon and Ledermann (1980)
tea	Vitzthum et al. (1975b)
tomato	Dirinck et al. (1977)
whiskey	Wobben et al. (1971)

Generally Recognized As Safe (GRAS) list as utilized by the Flavor and Extract Manufacturers' Association (FEMA). As can be seen, all are incorporated into flavorings in the low ppm range, thus indicating that they possess potent organoleptic properties.

Buttery et al. (1977) have reported that in general pyridines have less pleasant odors than pyrazines and thus suggested that possibly the series of alkylpyridines they found associated with roasted lamb is the reason for its flavor rejection by some consumers. They reported that 2-pentylpyridine, in dilute solutions, had a fatty- or tallo-ly-like odor and possessed an odor threshold in water of 0.6 ppm.

Harding et al. (1977) evaluated the sensory properties of English beer to which several pyridines, which were also found to be present in beer at low concentrations, were added, and as seen in Table IV, most of the resulting sensations would have to be considered objectionable. The rationale for the addition levels utilized was not reported. Of the three pyridines found naturally, 3-acetylpyridine was the most abundant, and as seen in Table IV, its addition resulted in an objectionable product. Thus, in beer perhaps pyridines are not entirely desirable and perhaps their formation should be discouraged. Apparently at least a portion of them can be derived from barley.

The compound 2-acetylpyridine has been described by Buttery et al. (1971) as possessing a cracker-type aroma. They postulated that it could result from the decomposition of 2-acetyl-1,4,5,6-tetrahydropyridine, which has been described by Hunter et al. (1969) as having an aroma reminiscent of freshly baked soda crackers.

Table II. Flavor Properties of Various Pyridine Derivatives^a

pyridine derivative	flavor properties
2-methylpyridine	astringent, hazelnut
3-methylpyridine	green, earthy, hazelnut
3-ethylpyridine	buttery, green, caramel
2-allylpyridine	green, melon-like
4-isobutylpyridine	fatty
2-propen-1-ylpyridine	green, burnt, coffee-like
4-propen-1-ylpyridine	green, fruity, strawberry-like
2-methyl-5-ethylpyridine	fatty, green
2,3-dimethylpyridine	roasted, rubbery
2,4-dimethylpyridine	green
2,5-dimethylpyridine	roasted, green, earthy
2,6-dimethylpyridine	green
3,4-dimethylpyridine	green, almond-like
3,5-dimethylpyridine	green, fatty, roasted
2-vinylpyridine	green
4-methylpyridine	green
2-methyl-6-vinylpyridine	bitter, earthy, roasted
4-phenylpyridine	bitter, caramel
4-benzylpyridine	bitter
2-benzylpyridine	green, astringent
2-methyl-5-methylpyridine	winey, buttery, cereal-like, caramel
2-methoxypyridine	burnt, mustard-like
2-butoxypyridine	leathery, green
2-(methoxymethyl)pyridine	caramel
2-(methoxymethyl)-6-methylpyridine	roasted, hazelnut-like
2-(hydroxymethyl)pyridine	roasted, moldy
3-(hydroxymethyl)pyridine	bitter, green
4-(hydroxymethyl)pyridine	hazelnut-like, coffee-like
2-(hydroxymethyl)-6-methylpyridine	bitter, burnt, coffee-like
2-(2-hydroxyethyl)pyridine	green, roasted
2-(2-ethoxyethyl)pyridine	bitter, astringent
2-acetylpyridine	roasted, coffee-like
6-methyl-2-acetylpyridine	chocolate-like
3-acetylpyridine	burnt, toasted
4-acetylpyridine	burnt, coffee-like
pyridine-2-carboxaldehyde	caramel, fatty
pyridine-3-carboxaldehyde	bitter, astringent
pyridine-4-carboxaldehyde	fruity
6-methylpyridine-2-carboxaldehyde	caramel, fruity
2-benzoylpyridine	bitter, musty
3-benzoylpyridine	green, woody, fruity
4-benzoylpyridine	astringent, bitter, musty
2,6-diacetylpyridine	bitter, coffee-like
4-(γ -pyridyl)butan-2-one	earthy
methylpyrid-2-yl acetate	astringent, bitter
pyrid-2-ylmethanethiol	popcorn, nutty, caramel, cereal
2-mercaptopyridine	burnt
2-(methylthio)pyridine	phenolic
2-(ethylthio)pyridine	green, bitter
pyrid-2-ylthiol acetate	roasted
dipyrid-2-yl sulfide	weak
2-pyrid-2-ylethanethiol	roasted, astringent, earthy
2-pyrid-2-ylethyl methyl sulfide	mushroom, bitter, green
2-pyrid-2-ylethyl ethyl sulfide	astringent, fatty, green
2-pyrid-2-ylethanethiol acetate	astringent, roasted
2-pyrid-2-ylethyl furfuryl sulfide	astringent, green
pyrid-2-ylmethyl methyl sulfide	bitter, green, earthy
pyrid-2-ylmethyl ethyl sulfide	metallic
pyrid-2-ylmethanethiol acetate	bitter, fatty

^a From Winter et al. (1972, 1975, 1976a,b).

Table III. FDA GRAS Approved Pyridine Compounds^a

compound	FEMA no.	av usage level, ppm
pyridine	2966	0.4-1.0
2-pyridinemethanethiol	3232	2.0
2-acetylpyridine	3251	3.0-5.0
2-(2-methylpropyl)pyridine	3370	0.2-0.5
3-(2-methylpropyl)pyridine	3371	0.1-0.5
2-pentylpyridine	3383	0.1-1.0
3-ethylpyridine	3394	0.05-0.06
3-acetylpyridine	3424	2.0-3.0
2,6-dimethylpyridine	3540	3.0-10.0
5-ethyl-2-methylpyridine	3546	0.5-1.5
quinoline	3470	0.1-0.5
6-methylquinoline	2744	0.1-0.5

^a From Hall and Oser (1965, 1970), Oser and Hall (1972), and Oser and Ford (1973a,b, 1974, 1978).

Table IV. Influence of Pyridine Compound Additions on the Flavor Properties of Beer

compound added	level added, ppb	aroma	flavor	aftertaste
2-methylpyridine	200	estery, creamy	cardboard, creamy	soapy, full
2-acetylpyridine	100	grainy, creamy, biscuity	burnt, grainy, oxidized	malty
3-acetylpyridine	500		harsh	harsh

In comparing the odor qualities of nine odiferous chemicals, Dravnieks et al. (1979) described the odor properties associated with pyridine as pyridine-like, pungent, solvent, and fishy, none of which can be considered as being very desirable. Thus, it would appear that not all pyridine derivatives can have widespread application as positive flavoring materials. In fact, it is entirely possible that the off-flavors associated with certain foods may be associated with the present of pyridines. This area deserves further scientific investigation.

However, it should be noted that whether pyridine or a substituted pyridine plays a positive or negative role in a particular food flavor cannot be judged adequately by sole reliance on an odor/flavor description of the pure compound. Concentration of the component in the food item, occurrence, and concentration of other volatile and nonvolatile compounds will all have an important bearing on the perception of the compound in question within the food product.

MODEL SYSTEM STUDIES

The pyrolysis products of pyridine and pyridine derivatives have been reported. Early work by Roth (1886) indicated that when pyridine was passed through a red-hot glass tube, hydrogen cyanide and bipyridines resulted. This was also confirmed by Ruhemann (1929). Later, Meyer and Hofmann-Meyer (1921) reported the reaction formed 2,2'-bipyridine as well as the 2,3' and 2,4' isomers. Similar results were obtained by Krumholz (1949).

Hurd and Simon (1962) pyrolyzed a series of methyl-substituted pyridines at temperatures varying from 700 to 800 °C, and the resulting identified compounds included pyridine, other methylpyridines, benzene, acetonitrile, acrylonitrile, benzonitrile, and quinoline. Higman et al. (1970) pyrolyzed collagen at 840 °C under nitrogen and observed at least 21 compounds. Among the seven identified were pyridine, 2-, 3-, and 4-methylpyridine, and 3-vinylpyridine. Similar compounds were identified during the pyrolysis of tobacco (Schlotzhauer and Schmeltz, 1967). Kang et al. (1980) identified pyridine, methylpyridine, 2-vinylpyridine, dimethylpyridine, isopropenylpyridine,

hydroxypyridine, and a cyanomethylpyridine among the pyrolysis products of chondroitin sulfate.

The thermal decomposition of certain amino acids can also result in pyridines. For example, Lien and Nawar (1974) found 2-methyl-5-ethylpyridine among the 13 compounds identified from the pyrolysis of alanine whereas pyridine, 3-methylpyridine, and 2,3,5-trimethylpyridine were associated with β -alanine decomposition. They postulated that 2-methyl-5-ethylpyridine could be formed either from the aldol condensation of acetaldehyde to form crotonaldehyde followed by dimerization in the presence of ammonia or via the Tschitschibabin reactions whereby an aldehyde and ammonia form an imine which is followed by an aldol-like trimerization in turn followed by a ring-closing reaction resulting in the loss of ammonia and hydrogen. All of the intermediates were also identified in the reaction.

A melanoidin resulting from a glucose-ammonia reaction which had undergone acid hydrolysis was shown to contain small amounts of β -hydroxypyridine (Tsuchida et al., 1973, 1975). Pyrolysis of the same material by Tsuchida et al. (1976) resulted in the identification of pyridine, 2-methylpyridine, 3-hydroxypyridine, and 2-methyl-5-hydroxypyridine. Other specific Amadori compounds were thermally degraded by Shigematsu et al. (1977). In the case of 1-deoxy-1-L-alanine-D-fructose, methylpyridine was one of the resulting 21 compounds observed.

Kato et al. (1973) thermally reacted sulfur-containing amino acids with glucose at 160 °C. In the case of cysteine, pyridine and 2-methyl- and 3-methylpyridine were among the resulting compounds, whereas when cystine was utilized, pyridine, 3-methylpyridine, and 2-methyl-5-ethylpyridine were present. However, when pyruvaldehyde was substituted for glucose, no pyridines were found. The compounds 2-methylpyridine and 2-methyl-5-ethylpyridine were among the 45 isolated from a system of cysteine-cystine-ribose that was refluxed for 24 h at 125 °C (Mulders, 1973). Noller (1957) reported that 2-methyl-5-ethylpyridine is formed when paraldehyde is reacted with ammonia at 200 °C. Buttery et al. (1977) have postulated that 2-pentylpyridine can form from deca-2,4-dienal and ammonia if first condensed to the aldimine followed by ring closure to the 1,2-dihdropyridine which in turn could be oxidized by air to the compound in question. The compound 2-hydroxypyridine was the only pyridine derivative detected among the 51 identified by Shibamoto and Russell (1977) when glucose, hydrogen sulfide, and ammonia were heated at 100 °C for 2 h. A total of 95 compounds were detected by Shibamoto and Bernhard (1978) when rhamnose and ammonia were reacted for 2 h at 100 °C. Pyridines among these included 2-amino-5-methylpyridine and 2-acetylpyridine.

The work of Suyama and Adachi (1979, 1980) represents the most specific investigations dealing with pyridine compound formation in food systems. They reacted alkanals with amino acids under neutral conditions at room temperature and reported (Suyama and Adachi, 1979) that quaternary pyridium betaines were formed. When these intermediates were heated at 180 °C for 2 min, pyridines resulted (Suyama and Adachi, 1980). When starting with glycine and propanal, they found 3,5-dimethyl-2-ethyl-, 2,5-dimethyl-4-ethyl-, and 3,5-dimethylpyridine. In the case of glycine-propanal-crotonal, 2,5-dimethylpyridine and 3,4-dimethylpyridine were identified. In all reactions, other volatiles were detected but not identified.

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