

Chapter 16

ADVANCES IN PEANUT FLAVOR QUALITY

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INTRODUCTION

The basis for ultimate product marketing in the peanut industry is the unique flavor of roasted peanuts. Flavor quality of any roasted peanut product is the sum of the effects of genetics, production and handling, storage, and processing factors. Because there are numerous combinations and interactions of these factors, logically there is variation in peanut flavor. The range of flavor variation normally is limited and managed appropriately by modern manufacturing processes. However, there are instances in which environment, seed maturity, handling, or other factors alter the variation so that manufacturing and consumer complaints occur because of an unsatisfactory product. Definition of even consistent interactions has proven to be a challenge for food scientists, chemists, agronomists and other related disciplines; and the title "The Peanut—The Unpredictable Legume" (National Fertilizer Association, 1951) is as appropriate today as it was over 40 years ago.

Various aspects of peanut flavor quality have been discussed in a number of reviews (Pattee and Singleton, 1981; Ahmed and Young, 1982; Pattee *et al.*, 1985; Ahmed and Pattee, 1987; Ory *et al.*, 1992; Sanders *et al.*, 1993). These reviews and associated research publications have increased our understanding and knowledge in the area of chemistry and biochemistry of peanut flavor, but still the specific mechanisms of flavor and off-flavor development remain elusive. Advances in peanut flavor research in the last 10 years have resulted from additional work in a wide range of agricultural commodities including peanuts. Recently, there have been increased efforts to understand the relationships of production, environment, maturation, and handling to flavor and shelf-life quality potentials due to several drought years experienced in the U.S. since 1980. This is borne out even more by advances occurring in the area of descriptive sensory evaluations. Flavor of final products is presently of primary importance to many peanut manufacturing companies and to the scientific community who are developing basic information relating production, handling, and other controllable factors to flavor quality. Our objective in this chapter is to summarize the advances in knowledge about peanut flavor quality which have occurred during the past 10 years.

ADVANCES IN DESCRIPTIVE SENSORY ANALYSIS

Descriptive sensory methods involve the detection and description of both qualitative and quantitative sensory aspects of a product by a trained panel (Meilgaard *et al.*, 1991). A descriptive flavor evaluation separates the overall flavor of the product or a commodity into its components, which are referred to as terms, notes, attributes, or descriptors. These descriptors describe specific, identifiable flavors in the product and have precisely worded definitions which communicate the flavor characteristic to trained panelists. Reference samples which smell or taste much like specific descriptors are used to train panelists. The panelists evaluate intensity of these descriptors using a defined intensity scale. Descriptive panels normally consist of five to 20 members who are able to detect and describe differences in flavor perception and intensity. Panelists should be trained in basic principles of sensory perception, food flavor analysis, descriptor development, and flavor intensity. The most widely used descriptive sensory methods are described in an ASTM manual (Hootman, 1992).

The sensory qualities of roasted peanuts are unique and the present understanding of these qualities has evolved through a number of different sensory methodologies. The Critical Laboratory Evaluation of Roasted Peanuts (CLER) method of evaluating peanuts was the first accepted protocol for quality evaluation by sensory judgment (Holaday, 1971). The CLER method used quality measurement and hedonic responses on a single continuum. Some inherent problems were associated with this method as pointed out by Tiemstra (1973). Fletcher (1987) published a revised CLER method that included descriptors for describing peanut flavor, but intensity was not evaluated.

Lexicons of roasted peanut flavor terms have expanded both in terminology and specificity of definition. The first trained descriptive analysis or flavor profile sensory panel for roasted peanut flavor (from which work was presented) was established in the Department of Food Science, North Carolina State University in 1975. The descriptive terminology consisted of 14 sensory character notes and three categories including aroma, flavor-by-mouth, and aftertaste (Oupadissakoon and Young, 1984). Syarief (1983) and Syarief *et al.* (1985a,b) modified the descriptive terminology by deleting the terms bite, burnt, chemical, green, rancid, stale, oil and adding the terms fruity, oxidized, mold, nose bite, peanut oil, petroleum, throat burn, underroast, and overroast. In 1986, the term fruity was modified to include both the maturity type fruity character as well as the fruity character due to high temperature curing (Pattee *et al.* 1989). Presently, the panel uses the following terms: roasted peanut, underroast, overroast, sweet, bitter, salt, earthy, molasses, tongue/throatburn, painty, fruity, sour, nutty, astringent, woody/hulls/skins, mold, stale, and petroleum. In 1986, USDA, ARS cooperated with flavor and peanut specialists to develop a lexicon which verified and expanded some areas in the peanut flavor lexicon used at North Carolina State University (Johnsen *et al.*, 1988). Sanders *et al.* (1989b) added the term "fruity fermented" to the lexicon which contained the terms roasted

peanutty, raw bean/peanutty, dark roasted peanut, sweet aromatic, woody/hulls/skins, cardboard, painty, burnt, green, earthy, grainy, fishy, chemical/plastic, skunky/mercaptan, sweet, sour, salty, bitter, and the chemical feeling factors astringent and metallic. Although the lexicons appear similar, differences in some areas are substantial. Degree of roast descriptions are different in that Johnsen *et al.* (1988) used the single term "dark roast" with an assumed intensity continuum for roasted flavor while the modified Oupadissakoon and Young (1984) lexicon used the terms "underroast" and "overroast," with an intensity continuum for each. Both lexicons provide freedom to write in new terms, and in common usage, panelists have added several terms. Descriptive terms in a lexicon are used with intensity ratings which must be anchored to specific standards (Meilgaard *et al.*, 1991).

ADVANCES IN INSTRUMENTAL FLAVOR ANALYSIS

Peanut flavor can only be fully perceived by sensory evaluation methods. However, gas chromatographic methods can be used to quantify volatile compounds that may contribute to flavor or indicate flavor problems. Because the exact compounds, the interactions of those compounds, and threshold perceptions constituting peanut flavor are not known, these methods can only approximate sensory methods.

GC and Human Olfactory Bioassay of Roasted Peanut Flavor Volatiles

Several hundred compounds have been identified in roasted peanuts (Walradt *et al.*, 1971; Buckholz *et al.*, 1980; Buckholz and Daun, 1981; Ho *et al.*, 1982). Although an identifiable flavor may result from a single compound, it is more likely that flavor perception is the result of a composite of many chemical species. As an example of this phenomenon, Fischer and Grosch (1982) found that the raw beany, legume flavor in raw peanuts is actually several different compounds. Using solvent extraction and then thin-film distillation of the resulting oil, an aroma concentrate was obtained which exhibited a pea/bean-like flavor. Upon "sniffer port" examination of the gas chromatographic profile of this raw peanut extract, two fractions, green/grassy and green/legume-like odor were found. The first peak contained predominantly n-hexanal and the second peak with green/legume aroma was made up of g-butyrolactone, n-nonanal, benzaldehyde, benzyl alcohol, and 2-methoxy-3-isopropylpyrazine.

Although "sniffer port" analysis is not a new technology (Acree *et al.*, 1976, 1984), application in the definition of compounds contributing to peanut flavor is relatively new. The technique involves concentration of the volatiles and GC analysis with a split column set-up providing portions of column effluents to the GC detector and to a port at which effluents may be smelled or "sniffed". Examination of olfactory attributes and GC/Mass Spectrometry (MS) identification of aroma compounds constitute the remainder of the analysis. A high roasted flavor quality "aromagram" generated from 14 repetitions of gas chromatographic and olfactory analysis is presented in Fig. 1.

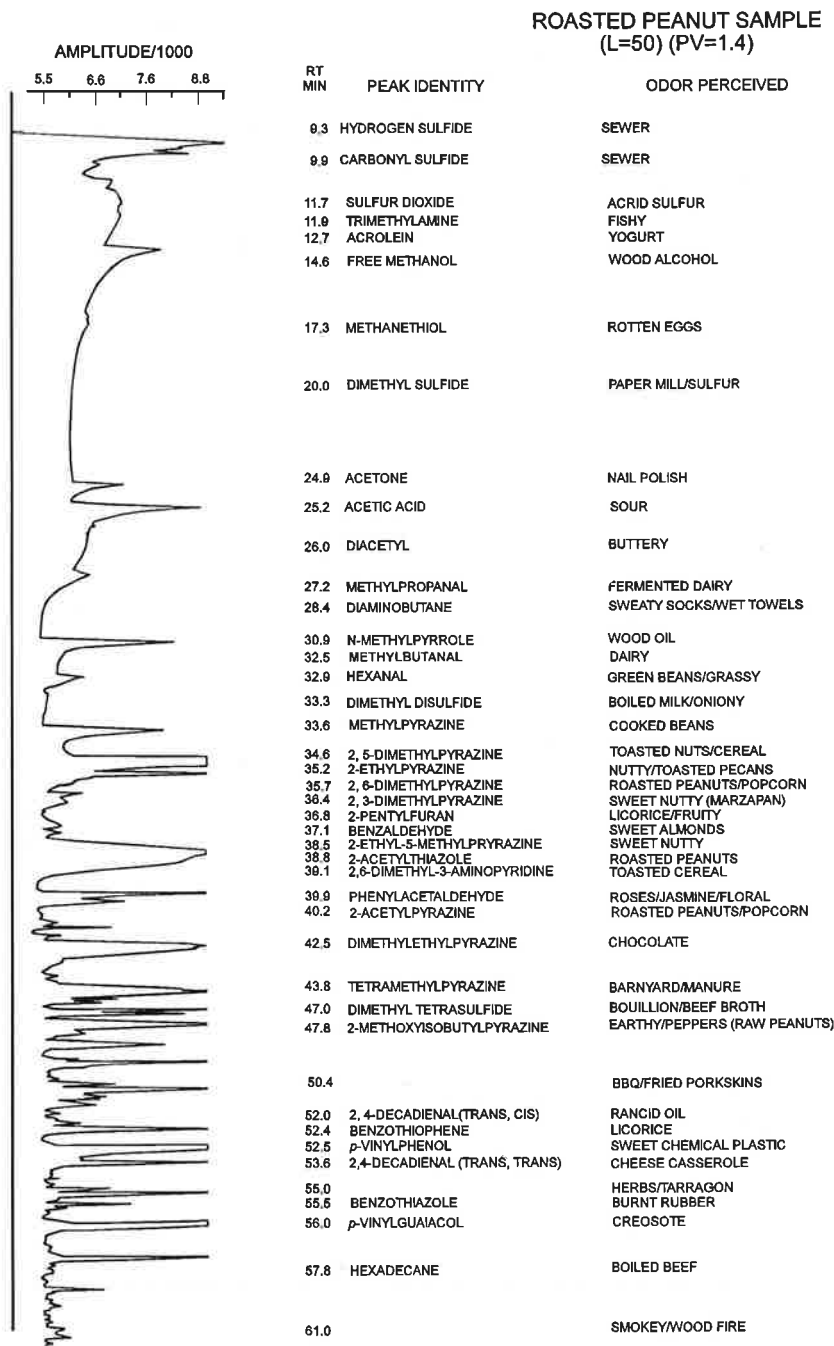


Fig. 1. Aromagram of Florunner peanuts roasted at 162 C to a color of L = 50 [reprinted from Vercellotti *et al.* (1992); copyright 1992 Amer. Chem. Soc.].

Aromagrams of key volatiles and their olfactory attributes have been generated for high quality roasted peanuts as well as seed of varying degrees of rancidity (Vercellotti *et al.*, 1992b). Many compounds identified by MS have also been characterized for their odor quality by GC "sniffer port" sampling. Aroma compounds included pyranones, furans, aldehydes, alcohols, phenols, acetophenone, substituted benzenes, and benzofurans, in addition to the Maillard *N*, *S*, and *O* heterocycles (Vernin *et al.*, 1988, 1992; Ledl and Schleicher, 1990; Grosch and Schieberle, 1991). Analysis of volatiles by direct GC and olfactory "sniffer port" has been improved by use of the external closed inlet device (ECID) with a wide bore glass capillary column as both trap and separation medium (Vercellotti *et al.*, 1992b). The ECID is a workable tool for introduction of peanut flavor volatiles onto a packed or capillary column and permits direct thermal desorption of the concentrated volatiles from foods, vegetable oils, packaging materials or from solid supports used to concentrate volatiles.

GC Volatiles Analysis

As reviewed by Ory *et al.* (1992), gas chromatographic volatile patterns of both raw and roasted peanuts provide data relative to ultimate product quality. Fore *et al.* (1976), Dupuy *et al.* (1983), and Young and Hovis (1990) developed procedures for determining volatiles in peanuts which can be used as indicators of inherent quality for many products. Parliment (1986) and Takeoka *et al.* (1986) considered the relative efficiencies of food flavor concentration and chromatographic analysis. These references are worthwhile points of comparison for the peanut flavor volatiles methods reviewed here.

Vercellotti *et al.* (1992a) reported techniques developed for analysis of peanut butter volatiles by direct gas chromatography at two purge temperatures (60 and 127 C) with flame ionization (FID) or sulfur specific flame photometric detection (FPD). Some 18 FID active compounds were identifiable as markers using this methodology in a study of volatiles from peanuts roasted to various degrees. Peak identification was essentially that listed previously by Fore *et al.* (1979). These basic techniques have been used to examine the relationships of GC volatiles with degree of roast and sensory flavor analysis (Bett and Boylston, 1992; Crippen *et al.*, 1992).

Many off-flavor compounds produced by Maillard reactions of sulfur amino acids and carbohydrates have intense sulfide or sulfur heterocyclic off-flavors. Although many of these compounds by themselves have undesirable characters, in proper proportion they add considerably to the positive blend of desirable flavors (Teranishi *et al.*, 1974). Hydrogen sulfide, carbonyl sulfide, methanethiol, dimethylsulfide, carbon disulfide, propanethiol, diethylsulfide, and dimethyl disulfide were identified by FPD and used as peanut volatile marker compounds by Vercellotti *et al.* (1992a). Nine unidentified sulfur compounds also are present in peanut seed. Watkins (1987) and Watkins and Young (1987) examined sulfur containing compounds in peanut butter and reported a similar list of compounds. Most sulfur-containing compounds have aromatic thresholds of perception that are less

than 1 ppb in water, and thus are likely to play a critical role in roasted peanut flavor.

INTERRELATIONSHIPS AMONG QUALITY, HANDLING, AND COMPOSITION

Factors Influencing Flavor and Quality

Peanut quality arises as a result of a complex interaction of genetic, physiological and biochemistry processes that produce the chemical composition of the peanut seed. The extent to which this interaction reaches its full potential can be referred to as degree of maturation or maturity. After digging, peanuts are subjected to a series of processes which will influence ultimate flavor quality. These processes, in general, are windrow curing, combining, bulk curing, and storage. For this chapter we have chosen to combine both curing processes for discussion purposes, and because of a lack of information, the topic of combining effects on peanut chemistry will not be addressed.

Genetic and Environmental Factors Related to Flavor

Flavor of roasted peanuts is an important characteristic influencing consumer acceptance, and enhancement of roasted peanut flavor has been a long standing objective of the peanut industry. However, there has been little reported research into the genetic and environmental factors influencing roasted flavor. Various aspects of roasted peanut flavor and other parameters which affect it have been previously reviewed (Ahmed and Young, 1982; Pattee *et al.*, 1985; Ahmed and Pattee, 1987).

Only recently has it become known that the roasted peanut attribute is an inherited trait. Pattee and coworkers (Pattee and Giesbrecht, 1990; Pattee *et al.*, 1993) reported broad-sense heritability estimates of 0.26 and 0.31 for the roasted peanut attribute, 0.05 and 0.37 for the nutty attribute, and 0.14 and 0.68 for the sweet attribute. None of the remaining attributes evaluated were reported to have broad-sense heritability.

In the U.S., runner-, virginia-, and spanish-type peanuts are produced. The ancestors of cultivars in the runner and virginia market types are predominantly members of *Arachis hypogaea* ssp. *hypogaea* var. *hypogaea*, but in both types there has been substantial introgression of germplasm from spanish (*A. hypogaea* ssp. *fastigiata* Waldron var. *vulgaris* Harz) ancestors (Isleib and Wynne, 1992). Claims of superiority in flavor for one or another market type are often expressed, but comparisons of flavor between market types commonly have been confounded by production region and associated environment. Limited information on effect of location on roasted peanut intensity was initially published for peanuts grown within the Virginia-Carolina area (Pattee *et al.*, 1993), but differences among the three peanut-growing regions could not be measured. Using data accumulated over 5 years, Pattee and Giesbrecht (1994) did not find a significant genotype x region interaction; however, their study was not an orthogonally balanced

design. Recently, Pattee *et al.* (1994) used subsets from their 5-year data set, which were orthogonal for cultivar and environment and showed environmental variation to be highly significant. Differences among years was the largest component of the variation. Variation across production regions (i.e., Southeast, Southwest, and Virginia-Carolina) was not significant, but it was significant across locations within years and regions. Further, cultivar-by-environment interaction was significant in most subsets and the main component of interaction was cultivar-by-location interaction within years and regions. They indicated that field experiments designed to compare genotypic means should emphasize replications across year-location combinations rather than within combinations. For accurate estimation of means, replication across several years are necessary but, for comparison of genotypes, additional locations should be substituted for years.

When conducting flavor studies on genetic resources one must be cognizant of the sources of variation in the attribute of interest. Within roasted peanut flavor, the sensory attributes overroast, underroast, and fruity have been identified as sources of variation on the attribute roasted peanut (Pattee *et al.*, 1990, 1991), and appropriate covariant adjustments have been proposed (Pattee and Giesbrecht, 1994). They suggested that unadjusted roasted peanut sensory data be statistically examined for the effects of these relationships on the roasted peanut sensory attribute value before proceeding with most additional statistical analyses when the maximum response value is desired. Improperly roasted peanut samples, which produce undesirable values for overroast and underroast, are indicated by CIELAB L^o color values <56 or >62 of the roasted peanut paste (Pattee *et al.*, 1991). These observations on a maximum roasted peanut attribute response in relation to roast color have since been confirmed by Crippen *et al.* (1992). The fruity sensory attribute (Pattee *et al.*, 1989, 1990; Sanders *et al.*, 1989a) can only be quantitatively determined by sensory evaluation, although a relatively constant relationship exists between intensity and concentration of ethanol in the sample. Fruity attribute intensities of 2 or above, based on a 14-point scale, are consistently related to a reduction in the roasted peanut attribute response (Pattee and Giesbrecht, 1990; Pattee *et al.*, 1990, 1993).

How (1984) compared the flavor acceptance of virginia, runner, spanish, and valencia market types. He found no significant differences in the means of panelists' scores for acceptance. Pattee and Giesbrecht (1990) used a 3-year data set on flavor to evaluate 71 peanut genotypes and found 22% of the runner genotypes had intensities of roasted peanut flavor higher than Florunner; 31% of the virginia genotypes were higher than Florigiant, which was the industry standard at that time. Applying a recently developed covariant adjustment technique for analyzing flavor data obtained from samples outside the acceptable roast color and fruity limits (Pattee and Giesbrecht, 1994) plus guidelines for the minimum number of observations (Pattee *et al.*, 1994), Pattee, Giesbrecht, and Isleib (unpubl. data, 1994) examined 61 genotypes from a 5-year study. In this adjusted data comparison of the virginia types, eight of the 37 genotypes had significantly higher roasted flavor intensity than the NC 7 control. Of the 19 runner types tested,

none were significantly better than the Florunner control and four were significantly lower in intensity. The eight significantly higher virginia genotypes were not different in intensity from Florunner.

Maturity and Harvesting

Maturity in peanuts is a complex concept because, as a botanically indeterminate plant, flowering and initiation of peanut development occur over an extended period of time. The complexity arises in that all mature peanuts are not large and all immature peanuts are not small. The interrelationship of size and maturity within sized lots has been discussed by Sanders (1989). He and others (Coffelt *et al.*, 1975; Pattee *et al.*, 1981b; Williams *et al.*, 1987) reported that peanut size and maturity are positively, but not absolutely, correlated. Thus, peanuts of different physiological maturity are found consistently in all commercial sizes. This interrelationship is confounded even more in lots that are marketed on a count per weight basis wherein large and small seed may be mixed together to give the same count as seed of consistent uniform size. Davidson *et al.* (1978) and Sanders (1989) indicated that peanut seed size distribution is affected by environment in predictable ways. Variation in irrigation, harvest date, and soil temperature generally produced significant differences among percentages of individual maturity classes in each commercial size. Increasing soil temperature tended to decrease seed size, although hull scrape maturity profiles indicated more rapid maturation; conversely, cooler temperatures shifted the mean size upward and tended to delay maturation. The distributions of maturity within commercial seed sizes were sufficiently different to suggest that flavor, roast color, shelf-life and other quality factors would be affected in final roasted products (Sanders *et al.*, 1989b). These differences are related to variations among carbohydrate, amino acid, protein, moisture and other components. Further, these components are also responsible for differential responses of maturity classes to high temperature curing and freeze damage. Development of production and handling methodologies producing a more consistent maturity distribution at the in-shell, shelled or roasted whole-nut level should result in improvements in consistent flavor and shelf-life.

With the advent of the hull scrape maturity method (Williams and Drexler, 1981) significant improvements have been made in harvesting peanuts at the optimum time. General practice has shown that harvest at optimum yield results in the best overall quality because of the important role of peanut maturation in changes of biochemical components related to quality. Sanders and Bett (1993) reported sensory differences in medium-grade size Florunner peanuts harvested weekly over a 6-week period. The percentage of mature peanuts as determined by hull scrape classification in the medium grade size increased progressively until about 1 week before optimum harvest date and then remained relatively constant. Earlier harvested peanuts had lower roasted peanut flavor impact and more bitter taste. Sanders *et al.* (1989a) reported trends toward increased intensity of flavor descriptors such as roasted peanutty and sweet aromatic in progressive hull scrape maturity classes. Intensity attributes such as "painty" and "fruity"

fermented" were higher in immature peanuts. The term "painty" is related to the common term rancid, and indicates the propensity of oil components in immature peanuts to undergo enzymatic or autoxidative changes which decrease shelf-life. Sanders *et al.* (1989a) also found that immature peanuts of the same commercial size roast darker than mature peanuts when subjected to the same roast conditions. Sanders and Greene (1989) demonstrated that methylpropanal is higher in immature peanuts. The relationship of decreased sensory acceptability in immature peanuts to high curing temperature has been demonstrated in several studies (Whitaker and Dickens, 1964; Pattee *et al.*, 1965, 1989, 1990; Sanders *et al.*, 1990). Sanders (unpubl. data, 1990) classified individual high-temperature-cured, roasted peanuts by roast color and found the darkest colors (immature) contained fruity fermented and other off-flavors whereas the lighter colors (mature) did not contain any off-flavors.

Curing and Flavor Relationships

Curing of peanuts sets into motion a number of metabolic processes which have been minimally explored with regards to impact on flavor quality. The initiation of degradative processes and other enzymatic and nonenzymatic reactions is also involved in the curing process of peanuts following harvest. One of the best documented degradative processes in peanuts is the degradation of carotenoids during slow curing in the windrow (Holley and Young, 1963; Beasley and Dickens, 1968; Emery and Gupton, 1968; Pattee *et al.*, 1968). The broad range of maturity levels and concurrent moisture levels combine to produce a very sensitive balance between high and reduced flavor quality due to the production of off-flavor constituents and possible loss of roasted peanut attribute precursors. During this thermodynamic process the concentration of peanut metabolites increases and they are in a highly reactive state. Little is known about the loss of roasted peanut flavor potential during curing, and previous reviews have summarized the known biological processes which lead to flavor quality deterioration and the conditions to be used if high quality is to be maintained (Sanders *et al.*, 1993).

Although extraneous and objectionable flavors in raw peanuts may arise from many sources, a major source of off-flavors is improper curing. This causes anaerobic respiration to become the predominant metabolic process as a result of an insufficiency of diffused oxygen to meet the increased respiration demands resulting from increased curing temperatures (Whitaker and Dickens, 1964). The relationship between off-flavors and increased curing temperatures was first documented by Dickens (1957). A relationship between increased head-space volatiles and off-flavors was first documented by Pattee *et al.* (1965) and subsequently confirmed by a number of researchers (Singleton *et al.*, 1971; Brown *et al.*, 1977; Lovegren *et al.*, 1982; St. Angelo *et al.*, 1984). The off-flavor was first reported as "high-temperature-curing off-flavor" but recently the descriptive terms "fruity" or "fruity fermented" were developed. All these studies have concluded that increased levels of alcohols and aldehydes in raw peanut samples are indicative of the presence of off-flavors. Although gas chromatographic profiling of peanut volatiles has

been suggested as a method of ascertaining the quality of raw peanuts, this method has not been generally accepted because of high cost and time factors (Sanders *et al.*, 1982b).

Pattee (1984) developed a rapid chemical colorimetric test to measure the concentration of organic volatiles in the head-space over raw, ground peanuts; but the use of caustic reagents limited its use. This advancement led to the development of an electronic meter for measuring organic volatiles over raw, ground peanuts which is sensitive, has a relatively low cost, and is simple to operate (Dickens *et al.*, 1987). This meter has undergone several additional improvements and is now commercially marketed. Interrelationships between head-space volatile concentration (HSVC), market grade (or seed-size), and roasted flavor in runner-type (Pattee *et al.*, 1990) and virginia-type (Pattee *et al.*, 1989) peanuts have been studied using various models of the electronic meter. Descriptive sensory profiling of a roasted peanut paste made from either market-grade or selected seed-size samples of runner-type or virginia-type peanuts and HSVC levels indicated that the fruity attribute was most characteristic of the off-flavor associated with increasing HSVC values. Low intensity levels of this fruity attribute were characterized as a sweet fruity character and higher levels of intensity as an alcohol-fermented fruity character. Whether peanuts were segregated by market-grade or seed-size, the larger seed were more flavorful and resistant to high-temperature curing characteristics.

Sanders *et al.* (1989a) investigated the effect of curing temperature on flavor quality of medium grade size Florunner peanuts from pods which had been segregated by color-based maturity classes before curing. They found intensity ratings of the descriptors roasted peanutty and sweet aromatic were lowest and ratings for fruity fermented, painty, sour, and bitter were highest for medium grade size immature peanuts which were cured at the higher temperatures. Additionally, Sanders *et al.* (1990) evaluated flavor and curing temperature relationships in commercial grade sizes and indicated that immature peanuts in the various market grades were responsible for fruity fermented off-flavor.

Freezing temperatures occurring while peanuts are still in the windrow also may result in an off-flavor described as fruity fermented. Singleton and Pattee (1989, 1991, 1992) reported that small, immature, high moisture peanut seed are very susceptible to freeze damage. They reported increases of specific and total volatiles and greater conductivity in leachates from small seed which had been subjected to freezing temperatures. These components were highly related to seed moisture content. Descriptive flavor data for peanuts where ambient temperature was monitored indicates freezing temperatures affect #1 size peanuts much more than medium or jumbo size peanuts (Bett, unpubl. data, 1988).

Basha *et al.* (1991) studied the effect of curing temperature on composition of peanut seed and roasted peanuts. They showed that seed cured at high temperatures contained greater amounts of α -amino nitrogen than peanuts cured at ambient temperatures. The temperature effect was more pronounced in less mature seed. In all seed maturity classes, curing temperature had no

effect on soluble and insoluble sugars and total protein content. However, peanuts cured at higher temperatures contained very little arachin polymer and also lost three arachin polypeptides.

Vercellotti *et al.* (1994) used GC/MS methodology to define and characterize changes during curing in carbon pools or carbohydrate precursors of peanut flavor for mature versus immature peanuts. They found low molecular weight sugar alcohols and reductones and other sugars to be the main components of carbohydrate turnover during peanut curing. These metabolites decrease to a stable concentration at curing equilibrium in both windrow/wagon-dried peanuts as well as in longer term stackpole drying.

Storage

Pattee *et al.* (1981a,b) stored sized peanuts in simulated commercial conditions (4 C, 65% relative humidity) and found that concentrations of amino acids decreased with seed size. Significant changes occurred in the free amino acid fractions in storage periods up to 9 months. These free amino acids were probably subjected to oxygen-centered free radical attack and, like the lipid oxidation processes in raw or roasted peanuts, the amino acids combined with other molecules or underwent scission reactions to lower molecular weight species. The carbohydrate components, fructose, glucose, inositol, sucrose, and stachyose were significantly affected by storage time while raffinose and ribose were not changed. Storage produced no significant effect on color or hedonic score of roasted flavor of peanut paste from selected seed size categories (Pattee *et al.*, 1982). Oxidative stability in raw peanuts decreased with storage time but not in a uniform manner across seed size. Iodine values of raw peanuts were not affected by storage time (Pattee *et al.*, 1982). How (1984), studying the effect of variety and gas composition on the flavor acceptance of peanuts, found no difference in flavor scores between various packaging gases. There were marked decreases in flavor acceptance for all the varieties of peanuts after 10 months of storage.

Bett and Boylston (1992) found that 2,5-dimethylpyrazine, ethylpyrazine, 2,3-dimethylpyrazine, 2-ethyl-5-methylpyrazine, trimethylpyrazine, 2-ethyl-3,6-dimethylpyrazine, 2,5-diethylpyrazine, 2-isobutyl-3-methylpyrazine, and 2-methylfurfural decreased in roasted peanuts during storage. Hexanal, 2-heptanone, 2-octanone, 3-octen-2-one, 2-nonanone, and 2-pentylfuran increased during storage. Sensory analysis of these samples showed roasted peanutty intensity decreased and painty and cardboardy increased with storage time.

Moisture Effects

The variation of flavor-related characteristics of peanuts during roasting as affected by initial moisture contents was reported by Chiou *et al.* (1991a). Total carbohydrate, glucose, and free amino acid contents were increased in peanuts in the early stage of roasting. Changes in specific amino acid content depended upon time of roasting and initial moisture content. Chiou *et al.* (1993) combined partially defatted peanut meal with raw peanut oil and found that the addition of 10-20% moisture was essential for the formation of peanutty flavor. Significant antioxidative activities were observed in

peanut oils roasted with partially defatted peanut meal under carbon dioxide, helium, and nitrogen.

Roasting

Roasted flavor and color formation during peanut roasting are part of a process involving heterocyclic condensations, chromophore formation, and polymerization of browning products (Buckholz and Daun, 1981). The optimization of the roasting process is a minimizing of both the underroast and overroast characteristics to form a maximum in roasted peanut flavor intensity (Pattee *et al.*, 1991). The concentration of free amino acids in peanuts declines during the course of roasting whereas sugars remain at a stable concentration while the targeted optimum roasted flavor and color are achieved (Moss, 1987). When the targeted roast is achieved, prolonged heating results in rapid consumption of sugars through polymerization and caramelization reactions that result in burnt or dark roast off-flavors. Burnt flavors are commonly associated with the reaction products of caramelization since phenolics are produced from this pyrolytic treatment of peanuts (Cammarn *et al.*, 1990). The peanut industry has traditionally used the "color" approach to explore the kinetics of the roasting reaction since there is a well established relationship between final roast flavor and color (Pattee *et al.*, 1991; Crippen *et al.*, 1992).

Chemistry of Roasting

Carbohydrates and other carbonyls are deemed important to peanut flavor because they react with amino acids to form flavor compounds during roasting (Heath and Reineccius, 1986a,b). In order for the reaction to occur, the carbonyl needs to be a reducing sugar, a polyhydroxycarbonyl compound, or a nucleophilic displacement product of a non-reducing sugar such as sucrose (Jezo, 1963b; Manley-Harris *et al.*, 1980; Richards, 1989). These compounds can then form a Schiff base with an amino acid, which may undergo many kinds of retroaldol reactions to form highly reactive reductones of two or more carbon atoms (Vernin *et al.*, 1988, 1992). Reductones are strong electron donor molecules, which may give rise to many hundreds of possible volatile flavor compounds by condensations and ring closures with nitrogen, sulfur, and oxygen intermediates (Paulsen and Pflughaupt, 1980; Ledl and Schleicher, 1990). Simultaneously with formation of volatile condensation products of the nitrogen, sulfur, and oxygen heterocycles in peanuts, nonvolatile polyhydroxy substituted imidazoles and pyrazines are generated. These compounds then condense into browning polymers which visually have many shades of yellow, orange, red, brown, and black (Ledl, 1986). Although sucrose, the predominant carbohydrate in peanuts is a non-reducing sugar, its importance in the aspect of browning reactions and polymer formation cannot be neglected (Del Pilar Buera *et al.*, 1987a,b,c). As a ketal, sucrose is unstable to dilute acid hydrolysis at or below physiological pH. When sugar solutions are concentrated at elevated temperature, as happens during peanut roasting, residues of carboxylic acids catalyze hydrolytic reactions of sucrose into glucose and fructose, both of which are reducing sugars and can participate in the Maillard reaction to form flavor compounds

(Ledl, 1986). The aminolysis of sucrose has been studied in model systems (Jezo *et al.*, 1963a), and demonstrated to involve a kind of nucleophilic displacement reaction either directly by the basic amino group or by neighboring group participation of a dissociated hydroxyl function to split the molecule (Manley-Harris and Richards, 1980; Del Pilar Buera *et al.*, 1987a,b,c). Other oligosaccharides of sucrose such as raffinose, stachyose, etc., as well as soluble polysaccharides in the peanut matrix also are substrates for such aminolysis reactions. It is during maturation and curing that the precursors for these reactions are brought to their optimum level. Reactivation of these processes by rehydration of the seed, through humidity increases or direct moisture addition during storage, may alter the balance of flavor precursors and thus the generation of various flavor compounds (Chiou *et al.*, 1991a).

Volatiles of Roasting

Various volatile compounds are produced during roasting of peanuts. Koehler and Odell (1970), following earlier work by Koehler *et al.* (1969), demonstrated that below 100 C little formation of pyrazines occurs in peanuts or model systems of sugars and amino acids. However, above 115 C thermal decomposition of certain pyrazines becomes significant, and loss of these flavor components occurs. Lovegren and St. Angelo (1981) analyzed volatiles produced by heating ground, raw peanuts at various temperatures in the direct GC inlet to effect roasting under inert gas sweep. They recommended using a sweep temperature of no higher than 130 C to avoid producing additional peanut volatiles in the sweep gas. Above 154 C, the temperature at which physical browning in raw peanut samples begins, dimethylpyrazine, dimethylethylpyrazine, and benzeneacetaldehyde form in increasingly larger quantities. Buckholz *et al.* (1980) and Buckholz and Daun (1981) indicated that roasting peanuts causes a decrease in carbonyls and an increase in pyrazines. How (1984) found that overroast and underroast attributes were more intense in air-roasted peanuts than in oil-roasted peanuts. This may have been due to less uniform roasting in air-roasted peanuts. How (1984) also found the degree of roast is often related to methylpropanal and methylbutanal, along with *N*-methylpyrrole and the pyrazines group. Buckholz *et al.* (1980) and Buckholz and Daun (1981) demonstrated a high correlation between the intensity of benzeneacetaldehyde and desirable flavor in roasted peanuts. The relative quantity of benzeneacetaldehyde is highest at medium roast (Hunter L = 50.5). Further, Pattee and Giesbrecht (1994) found the optimum roasted peanut intensity occurs at L° 58.3, which is only 0.8 actual color units different from the Hunter L value. Vinylphenol, which possess a strong chemical plastic odor, increases even in the darkest roasts; and for all the range of roasts it is the predominant flavor volatile when volatiles are collected at 127 C (Vercellotti *et al.*, 1992a). Lovegren *et al.* (1987) defined relative concentrations of each of the above gas chromatographic peaks using response factors from external standards. Vercellotti *et al.* (1992a) found balanced peanut roast characteristics are present in Florunner peanuts roasted to a Hunter L of 49 to 50, at

which color volatile roasted peanut marker compounds are found in a rather narrow range of intensities. Crippen *et al.* (1992) found methylpyrazine, dimethylpyrazine, methylethylpyrazine, *N*-methylpyrrole, benzeneacetaldehyde, vinylphenol, methylbutanal and methylpropanal levels to be highly correlated with the dark roasted descriptor and also the sulfur-containing compounds methanethiol, carbon disulfide, propanethiol, diethylsulfide, dimethyldisulfide, and several unknown sulfur compounds. They also found free methanol, thermally produced methanol, pentane/acetone, methylbutanol, and two unidentified compounds vary with the degree of roast. The descriptor woody/hulls/skins was correlated with *N*-methylpyrrole, methylbutanal, methylpropanal, vinylphenol, some pyrazines, methanethiol and an unknown. Methanol and ethanol correlated with raw/beany. The two desirable descriptors, roasted peanutty and sweet aromatic, did not correlate with any compound monitored. Free methanol decreased with increased roast time. The larger quantities of alkaloidal pyrazine compounds in very dark roasts might be responsible for some of the bitter and dark roast off-flavors involved (Belitz and Wieser, 1985). There are many Maillard reaction products in the roasted peanut volatiles mixture. A composite of Maillard reaction pathways elucidated by Paulsen and Pflughaupt (1980), Ledl (1986), and Ledl and Schleicher (1990) is illustrated in Fig. 2. Roasted peanut Maillard products have been extensively studied by Mason and Waller (1964), Mason *et al.* (1966), Walradt *et al.* (1971), Waller *et al.* (1979), Buckholz *et al.* (1980), Buckholz and Daun (1981), and Ho *et al.* (1982). The many heterocyclic nitrogen compounds such as pyrazines, thiazoles, thiophenes, and other aromatics or sulfides suggest a role in the total flavor impact. Mason and Waller (1964), Newell *et al.* (1967), and Ahmed and Young (1982) reviewed the role of precursors as well as enumerated the collective heteroatomic compounds produced in peanut flavor.

Compositional Changes

Basha and Young (1985) attempted to identify the protein/peptide source of amino acids potentially involved in formation of flavor compounds by extracting and examining peanut seed proteinaceous components before and after oil roasting. The studies indicated that roasting caused a decrease in methionine-rich proteins, aggregation of arachin proteins, and gradual decrease in a 70,000 mol wt polypeptide.

The effects of maturity and precursors of roasted flavor were studied by Rodriguez *et al.* (1989). They found protein and carbohydrate contents of the raw and roasted samples to be similar; however, the α -amino nitrogen content was lower in the roasted samples. Protein and carbohydrates from different maturity samples responded similarly to roasting, but the α -amino nitrogen decreased more in the mature (38-55%) than in the immature seed (25-28%). Roasting significantly lowered the amounts of the 90,000, 70,000, 50,000, and 32,000 mol wt polypeptides.

In a comparison of quality in raw and roasted sound and shriveled peanut seed from sized commercial lots, Chiou *et al.* (1992) found that shriveled seed underwent greater decreases in total and free amino acids, less increase in sucrose, and more protein denaturation during roasting. Chiou *et al.*

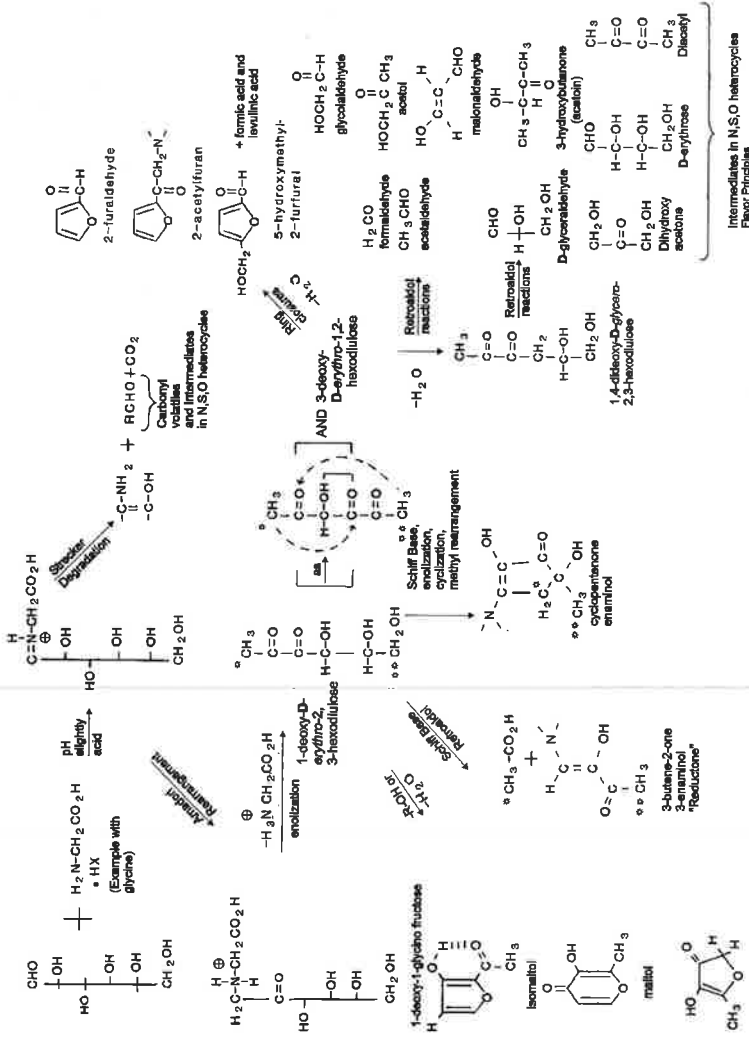


Fig. 2. A composite of Maillard reaction products as reported by Paulsen and Pflughaup (1980), Ledl (1986), and Ledl and Schlecter (1990).

(1991b) roasted peanuts under various gas environments for various times and found the best flavor was obtained by roasting in nitrogen or carbon dioxide for 18 minutes, while roasting in oxygen for 25 minutes resulted in greater stability during initial stages of an oven test at 62 C. Decreases of total α -amino acid nitrogen, glucose, sucrose, and conarachin contents as well as specific lipoxygenase activity were dependent on the extent of heat treatment (roasting time) rather than the gas environment.

Moss (1987) studied peanut roasting using a forced air, batch pilot plant roaster (4-5 kg capacity). The effects of various roasting temperatures, roasting times, relative humidities of the roasting chamber and initial peanut temperatures were evaluated by moisture content, color, and peroxide value analysis. After estimating drying curves of the peanuts during roasting, a correlation between the yellow-blue (b^*) color scale and moisture content was suggested. Increased relative humidities in the roaster were found to enhance rate of oxidation of the product as measured by shelf-life peroxide values. Moderate initial temperature fluctuations had no effect on the final roast character of the peanuts. Within the temperature limits tested, longer roasts at lower temperatures resulted in a superior quality product.

Composition and Flavor Relationships

The various components in both raw and roasted peanuts determine flavor and shelf-life. Peanut composition studies are generally in relation to market-type, cultivar, growing area or maturity and are usually divided into lipid, carbohydrate or protein components or fractions that tend to affect one of these components. The implications of differences in particular fractions or components have not been generally reported since the consequence of those differences often are not known. One exception is the role of lipids and specific lipid fractions as they relate to the shelf-life of peanuts through the development of off-flavor compounds from oxidized lipids (Fore *et al.*, 1953; Sanders 1980a,b; Sanders *et al.*, 1982a). The term "shelf-life" here is used interchangeably with flavor since the time to occurrence of unwanted or unacceptable changes in flavor generally constitutes the shelf-life potential of a raw or roasted product.

Lipids

The high oil value of peanuts varies somewhat with cultivar, maturity, and production conditions. In the four major market-types (runner, virginia, valencia, and spanish) total oil content varies from 44-56% (Cobb and Johnson, 1973; Holaday and Pearson, 1974; Ahmed and Young, 1982) while average protein content is about 25% and ranges from 22-33% (Ahmed and Young, 1982). Holaday and Pearson (1974) reported a significant correlation between production location and total oil content and protein. In maturity studies of cultivars NC 2 and Florunner, total oil (as a percentage of dry weight) increased significantly and then decreased slightly (Pattee *et al.*, 1974; Sanders *et al.*, 1982a). The most rapid changes in oil percentage occurred in early maturity stages and corresponded to the time of very rapid increases in seed dry weight. No reports of a correlation between the percentage of oil in peanuts and shelf-life were found. However, the above

reports indicated a significant positive correlation of oil content with maturity, which is highly related to flavor and shelf-life potential.

Peanut oil composition varies with maturity and the concentration of some oil components has been shown to influence oxidative stability and shelf-life (Fore *et al.*, 1953; Sanders, 1980a,b). As peanuts mature, total oil, triacylglycerol, and oleic/linoleic acid (O/L) ratio increase, while free fatty acids, polar lipids, monoacylglycerols, and diacylglycerols decrease (Pattee *et al.*, 1974; Sanders *et al.*, 1982a). In Florunner peanuts, the relative weight percentage of free fatty acids was shown to decrease from 4.5 to 0.7 % as peanut seed matured (Sanders, 1980a). The consistent relationship of these components to shelf-life leaves little doubt as to the relationship of maturity and shelf-life. Mzingo *et al.* (1985) reported that percent stearic, oleic and arachidic acids increased and palmitic, linoleic, eicosenoic, behenic and lignoceric acids decreased with increasing commercial screen size in five virginia cultivars grown at two locations for 3 years. Significant increases in O/L ratio were recorded for the larger seed sizes. Norden *et al.* (1987) examined the range of oleic and linoleic acid distributions found in over 500 genotypes and reported that oleic acid content ranged from 37-80% and linoleic acid ranged from 2-43%.

Lipid Oxidation

The influence of water activity on the rate of autoxidation of raw peanut oil was investigated by Gopalakrishna and Prabhakar (1983). They reported that the rate of peroxide formation decreased significantly with water activity (a_w) greater than 0.67. The formation of free fatty acids was not affected by a_w . The authors concluded that components present in the raw oil, possibly phospholipids, protected the oil against autoxidation when the moisture content of the seed was high. Descriptive sensory analysis (Bett and Boylston, 1992) indicated that when roasted peanuts undergo autoxidation, positive attributes (such as roasted peanutty) decrease while negative attributes such as cardboard and painty increase. A large number of lipid oxidation products as well as heterocycles are involved in these changes (Vercellotti *et al.*, 1992b). During lipid oxidation, volatiles rose dramatically while off-flavors intensified in the olfactory portion of the aromagram; simultaneously, positive olfactory attributes became imperceptible as many heterocycles or thio-derivatives (ppb level) disappeared at high peroxide values.

A number of questions about peanut flavor persistence exist concerning the relationships of sensory changes and the physical versus perceptual loss of pyrazines and other high impact heterocycles during storage of roasted peanuts. It has alternatively been suggested (Bett and Boylston, 1992) that the roasted peanut flavor compounds are not destroyed by free radical propagation, but rather are adsorbed into the hydrocolloid matrix of the seed.

Antioxidants and Oxidants

The concentration of endogenous nonenzymatic antioxidants in raw seed varies according to the same genotypic and cultivation factors which govern

many seed components. Peanuts contain approximately 26-60 mg tocopherols/100 g (of which 12-25 mg is α -tocopherol), 6 mg ascorbic acid, and trace amounts of carotenes (Cobb and Johnson, 1973). α -Tocopherol is an excellent chain-breaking antioxidant, almost 100 times more effective against peroxy radicals than BHT (Burton and Traber, 1990). Pattee *et al.* (1968) suggested that the degradation of carotenoids during curing probably involved lipoxygenase being activated during the harvesting process. Such a process probably involves the carotenoids as antioxidants, and their loss early in the curing process could predispose the peanut seed to increased potential for oxidative damage later during storage. Hokes (1977) determined tocopherol content and fatty acid profiles in 31 cultivars for 4 years. A multiple regression equation for the prediction of stability of cold pressed oil revealed that 87% of the stability could be correlated with total tocopherol/% linoleic acid. Hashim *et al.* (1993) reported that α tocopherol content decreased with seed size as did O/L ratio in Florunner peanuts.

The influence of transition metals, particularly iron and copper, on flavor and the shelf-life stability of peanut products is potentially a critical one because of their participation in the oxidative rancidity process. Iron and copper are very common transition metals which exhibit a variety of oxidation states and therefore play key roles in the autoxidation and enzyme-catalyzed oxidation of membrane and storage lipids. They readily transfer electrons making them useful cofactors for reactive oxygen species which do not effectively initiate lipid oxidation. Virtually all lipid and other biomolecule oxidations are advanced by metal ions. The key enzymes important in the direct or indirect oxidation of lipids and the generation of radical species are iron-containing proteins such as lipoxygenases, peroxidases, xanthine oxidase, and others. Transition metals can promote the oxidation of lipids by producing a fatty acid radical via hydrogen abstraction of an unsaturated fatty acid, by indirectly generating reactive oxygen species during the oxidation of certain cofactors, and by reacting with ozone to generate superoxide (Kanner *et al.*, 1987). Gaines and Hammons (1981) reported little variation in the concentration of micronutrients including the pro-oxidants copper and iron in four cultivars grown in six locations in the U.S. Conkerton *et al.* (1989) reported that oil and copper contents decreased consistently in eight genotypes in response to midseason drought stress.

Sanders *et al.* (1992) found that tocopherol content was consistently different in peanuts from various origins. Peanuts produced in the U.S. had a consistently higher tocopherol content than peanuts produced in China or Argentina. Additionally, copper content was significantly lower in U.S. peanuts and iron content was generally lower. These factors were related to higher O/L ratios, and oil oven stability studies indicated the overall longer shelf-life potential of the U.S.-produced peanuts. Thus, the study confirmed the close relationship of oil quality factors such as free fatty acids, O/L ratio, peroxide value, total carbonyls, tocopherols, copper and iron to most measures of shelf-life stability. It was suggested also that inappropriate levels of these compounds will then contribute to a shorter shelf-life of the resulting products. Bett *et al.* (1994) reported descriptive sensory analysis of the same

peanuts evaluated by Sanders *et al.* (1992) and flavor scores were closely aligned with quality factor indicators.

Hydrolytic stabilization of peanuts can be achieved through humidity and temperature control; however, oxidative processes such as lipoxygenase activation at sites of cell membrane disruption may still occur (Ahmed and Young, 1982; Ory *et al.*, 1992). The lipoxygenases activate oxygen to produce hydroperoxides at allylic sites of the polyunsaturated fatty acids present in the seed. These hydroperoxides are subsequently rearranged to conjugated dienes and may undergo retroaldol reactions to saturated and unsaturated carbonyl compounds. These compounds, or condensation products of other low molecular weight reactants such as amines or thiols, are the origin of many kinds of flavor compounds contributing positive or negative characters to roasted peanut products (Ory *et al.*, 1992).

Transition metal-catalyzed activation of oxygen leads to many kinds of free radical reactions including the formation of hydroperoxides (Stadtman and Oliver, 1991; Kanner, 1992). Reductones or electron donors from reducing sugar reactions and Maillard browning processes are potent free radical scavengers, protecting both raw and roasted peanuts from damage during storage (Bailey and Um, 1992). These free radicals also attack proteins, nucleic acids, polysaccharides, and membrane phospholipids (Stadtman and Oliver, 1991). Such reactions may result in the loss of desirable roasted peanut flavor by destruction of low molecular weight volatile flavor compounds that are present at very low human olfactory threshold levels (ppb concentrations) as well as the generation of volatile off-flavor carbonyls (hexanal, heptadienals, nonanal, decadienals, etc.) in the high ppm range (Vercellotti *et al.*, 1992b). The loss of key roast-quality flavor compounds along with generation of the carbonyl rancidity factors results in a serious peanut shelf-life stability problem.

Cross linking of proteins by the free radical reaction combined with aldol condensation or imine bridging destroys physical properties of the polymeric matrix of the peanut. Thus, texture, gel-strength, water-holding ability, and milling properties deteriorate, either through poor storage conditions of raw peanuts or exposure of roasted products to oxidizing conditions (McWatters and Cherry, 1982; Kim *et al.*, 1992). Damage to peanut cell membranes during any postharvest treatment may initiate these free radical events by mixing metal catalysts with oxygen and exposing membrane surfaces to activated oxygen species (Kanner, 1992; Vercellotti *et al.*, 1992b).

Proteins and Carbohydrates

Musingo *et al.* (1989) examined the effects of drought and temperature stress on several of the protein factors of peanuts. They reported variable changes in α -amino nitrogen and total protein content with seed size, but found a consistent increase in a 70,000 mol wt polypeptide. Although the specific amino acids or peptides involved in flavor development and, possibly shelf-life stability, have not been conclusively described, changes in amino acid quality and quantity do occur among years. Holaday and Pearson (1974) reported significant production location effects on total protein from three

genotypes of the three most prevalent market types. However, they did not find a significant relationship with temperature, fertilization, or rainfall at the locations tested.

McMeans *et al.* (1990) reported that soil temperature increases result in decreased concentrations of the free carbohydrates fructose, glucose, sucrose, raffinose, and stachyose in Florunner peanuts. This decrease was observable in comparisons of sized seed and hull scrape maturity stages from each soil temperature plot. Musingo *et al.* (1989) reported that total carbohydrates in sized seed increased with temperature stress. Drought has two effects in that dry conditions affect the normal physiology of the plant (and thus the physiological/biochemical composition of the developing peanuts) and higher temperature conditions generally associated with drought change development, maturation, and biochemical reactions and reactants in peanuts. As in other studies of changes of biochemical constituents, the specific effect on flavor and shelf-life of these differences and those reported due to maturity are presently undetermined. Flavor relevant interactions between protein and carbohydrates have been published but new information on the potential interactions of all three via free radical mechanisms provides necessary impetus for continued compositional investigations.

Protein Degradation

Protein degradation in stored peanuts is less well documented than that of other organic molecules. However, during the oxidative processes involved in roasting peanuts, it has been shown that the disappearance of whole classes of polypeptides involving deterioration of peptides and storage proteins occurs (Neucere *et al.*, 1969, 1972; Young *et al.*, 1974). How these protein and peptide reactive events contribute to the generation of flavor molecules is not known. However, Stadtman and Oliver (1991) recently published a general review on the degradation or modification of proteins by metal-catalyzed oxidations. An interesting comment in that review was that, once the proteins are cleaved by the free radical reactions, they become much more susceptible to proteolysis and considerable damage is done to the biochemical architecture of cells or structural proteins by these oxidations. Although not thoroughly studied in the peanut, such an oxidative protein degradation model could be extrapolated to protein storage. This model would entail proteolytic degradation, leading to peptides reactive in Maillard reactions, and the subsequent production of off-flavors or protein cross-linking.

FLAVOR QUALITY OF NEW PRODUCTS

Several products have been developed that incorporate peanut seed or flour. Holt *et al.* (1992) determined that wheat flour can be replaced with 24% cowpea and 46% defatted peanut flours in tortillas without altering baking characteristics. The characteristic beany flavor of the legumes was a problem. Lee and Beuchat (1991) replaced buttermilk with peanut milk in salad dressing. A 25% replacement can occur without a loss in sensory quality. Santos *et al.* (1989) formulated an imitation cheese spread with

peanut paste. Some panelists commented that a peanut flavor was detected in the cheese spread.

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