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## **INTRODUCTION**

In recent years there has been an increased awareness of the existence of environmental and processing contaminants in food and food products prepared for human consumption (1). State and federal regulations designed to monitor the presence of chemicals in commercially-produced foods have resulted in the maintenance of safe concentrations of these contaminants. There are, however, food and natural products processed and sold by individuals locally that are not regulated by testing programs to determine the presence of toxic substances.

Honey is an example of this kind of product that is produced and marketed in Connecticut. Contamination of honey can arise from a variety of sources. Metals from industry and automobile pollution are prevalent in the environment. Pesticides from agricultural applications can contaminate the flowers from which the nectar is drawn. Honeybees may be exposed to a variety of hazardous contaminants and transmit these chemicals to the honey. Toxic substances can enter the hive and contaminate the contents. Processing and canning techniques can introduce metals and other toxic substances into the honey. Any one or a combination of these sources could result in a honey product that has been contaminated with toxic chemicals.

The purpose of this study was to determine if any of these chemicals existed in hazardous concentrations in honey produced and marketed in Connecticut. Samples collected from representative locations throughout the state were analyzed for the presence of metals and pesticides.

## EXPERIMENTAL

**Sample Collection:** One or two pound bottles of honey were collected from roadside stores representing the geographical areas of Connecticut that are listed on the map in Fig. 1. All samples, except samples A and L, were collected in duplicate in September and October of 1977.

**Metal Analysis:** Each sample of honey was heated at 40° in the original container until all solids appeared to become dissolved. The container was repeatedly inverted to insure the homogeneity of the honey. A 25.0 g aliquot of the honey was weighed into a 500 mL erlenmeyer flask and 10.0 mL of nitric acid (Ultrex) was added. The solution was heated at approximately 80° until a few mL of solution remained. The solution was cooled and 2.0 mL of hydrogen peroxide (Ultrex) was added. The solution was heated at 80° until the hydrogen peroxide was evaporated. Nitric acid and hydrogen peroxide were then alternately added with intermittent heating and cooling until the solution became clear or a faint yellow color.

The digested samples were dissolved in 0.125 mL of nitric acid and quantitatively transferred to a 25 mL volumetric flask using reagent grade water. Each sample was then diluted to volume with reagent grade water. Required dilutions of the digested samples were prepared with 0.5% nitric acid and analyzed for Cu, Fe, Zn, Mg and Mn by flame atomic absorption. Flameless atomic absorption using a graphite furnace (Perkin-Elmer Model 2200) was used to analyze the samples for Pb. Mercury and arsenic were analyzed as elemental mercury and arsine, respectively, using a Perkin-Elmer MHS-10 Mercury/Hydride System (2). Sodium borohydride was used for the reduction of mercury and arsenic. The analyses were performed on a Perkin-Elmer Model 403 atomic absorption spectrometer and the parameters used were optimized around those recommended in the instrument analytical manual (3).

**Screen for Halogenated Hydrocarbons:** The solids in the honey samples were dissolved and thoroughly mixed as described in the metal analysis preparation. A 1.0 g aliquot of the honey samples was weighted into a 20 mL culture tube. Ten mL of acetonitrile (Burdick and Jackson), 1.0 mL reagent grade water and 1.5 mL of a saturated Na<sub>2</sub>SO<sub>4</sub> solution were added to the sample and the total was mixed on a rotary type mixer for 10 minutes. Following centrifugation, a 5.0 mL aliquot of the acetonitrile phase was transferred to a culture tube containing 10.0 mL 0.5% Na<sub>2</sub>SO<sub>4</sub> and 4.0 mL hexane (Burdick and Jackson). This solution was mixed on the rotary mixer for 10 minutes and then centrifuged. Three mL of the hexane layer were applied to a 0.5 g florasil column. The column was eluted with 2.0 mL of 5%

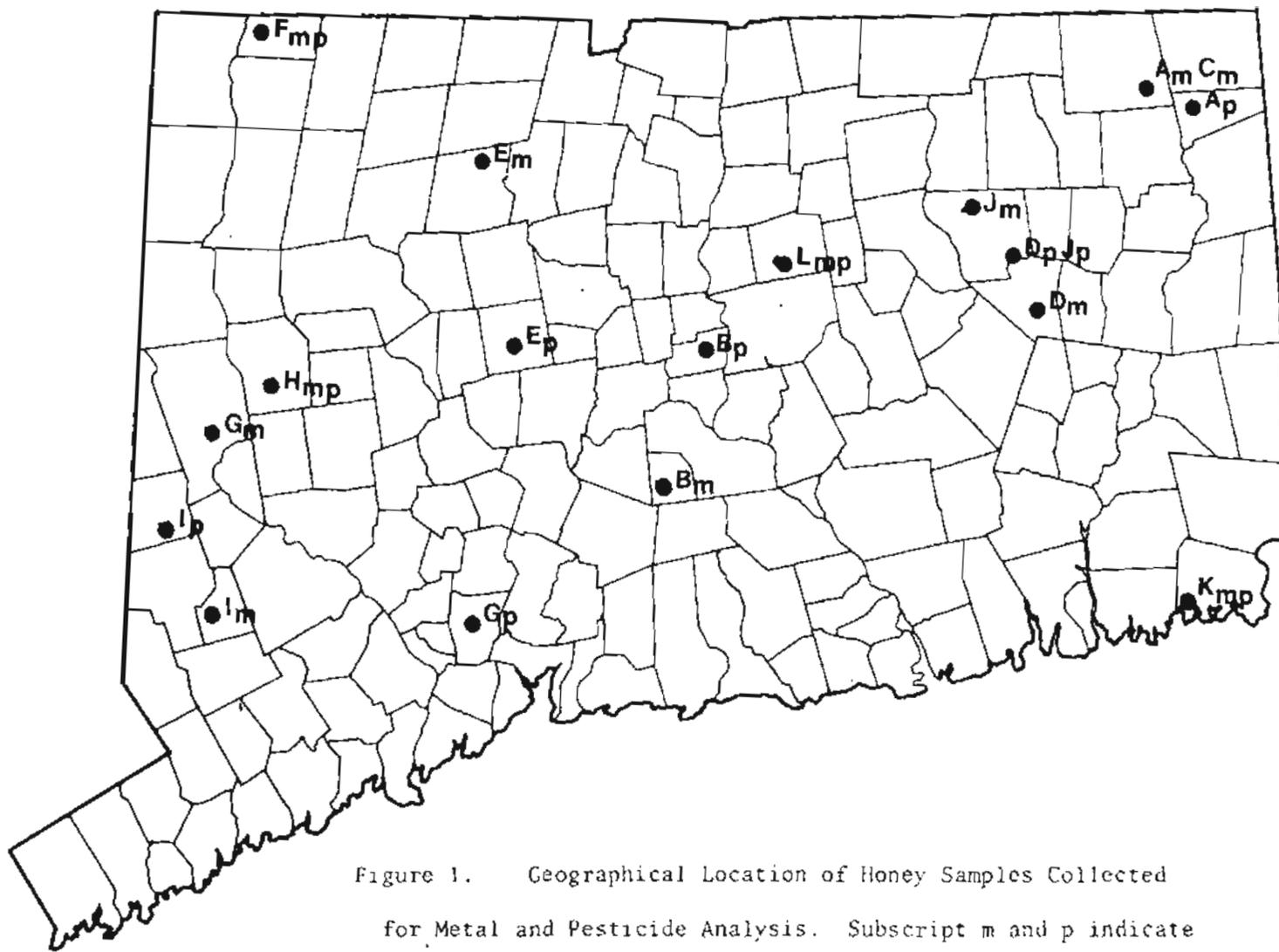


Figure 1. Geographical Location of Honey Samples Collected for Metal and Pesticide Analysis. Subscript m and p indicate location where honey was marketed and produced, respectively.

methanol/hexane. The total effluents were collected in a conical tube and were evaporated just to dryness at room temperature under a stream of nitrogen. The residue was dissolved in 50  $\mu$ l of hexane and 2  $\mu$ L of this solution along with 2  $\mu$ L of 424 ng/mL aldrin (internal standard) were analyzed by gas chromatography. The parameters for the gas chromatographic analysis were: column — 6' x .08" glass containing 10% DC200 on 100/120 mesh supelcoport, column temperature— 215°, injector temperature — 230°, electron capture detector at 275° and a carrier gas of 5% methane/argon at a flow rate of 70 mL/min. The attenuation of the electrometer was adjusted so that 848 pg of aldrin gave a one-half full scale deflection. These conditions insured that a one-tenth deflection in the sample's chromatogram was equivalent to an aldrin concentration of approximately 10 ng/mL. Peaks of less than one-tenth scale deflection were considered trace amounts (4).

## RESULTS AND DISCUSSIONS

The map, Figure I, shows the locations where the honey samples analyzed in this study were produced and marketed. Duplicate samples were collected at each market place except samples A and L, which were single samples. Sample C was the only sample produced outside of Connecticut (Johnston, R.I.). It was however, marketed in Connecticut.

The average concentrations of metals found in honey collected from 12 sites in Connecticut are recorded in Table I. Arsenic and mercury concentrations were less than 0.010  $\mu$ g/g and 0.005  $\mu$ g/g, respectively, in all samples.

There was a considerable range between the individual metal concentrations in samples collected at different locations within Connecticut. Manganese exhibited the greatest variation with a high concentration that was 135 times the low concentration. The differences in metal concentrations did not appear to correlate to the region of the state where the honey was produced. This was illustrated by a comparison of samples D and J which were produced at the same geographic location by different producers. Magnesium was found in sample D in a concentration 14 times that found in sample J. Sample D had a manganese concentration 106 times that of sample J. Studies conducted across the United States and in various foreign countries have also reported large variations in metal concentrations in honey (5,6). Table II shows a comparison of these data with the data collected from this study. The average concentrations of Cu, Mg and Mn

TABLE I. Concentrations of Metals in Honey Marketed in Connecticut

Sample	Date Collected	Metal Concentration ( $\mu\text{g/g}$ ) <sup>a</sup>					
		Cu	Fe	Zn	Mg	Mn	Pb
A <sup>b</sup>	9/03/77	0.31	5.21	7.24	13.4	0.52	0.200
B	9/13/77	1.70	3.02	1.53	247	37.8	0.144
C	9/20/77	0.20	3.20	0.86	16.4	0.63	0.161
D	9/20/77	1.06	2.96	0.40	198	34.1	0.102
E	9/27/77	0.67	10.8	10.10	38.5	4.33	0.149
F	9/27/77	0.35	1.17	0.17	15.6	0.28	0.055
G	9/27/77	0.82	11.0	3.65	46.3	3.14	0.140
H	9/27/77	0.68	5.83	2.50	95.9	5.29	0.156
I	9/28/77	0.22	2.56	0.73	23.0	0.77	0.056
J	9/28/77	0.26	1.62	1.34	14.3	0.32	0.060
K	10/10/77	0.44	1.58	1.08	45.4	11.8	0.074
L <sup>b</sup>	10/12/77	0.52	2.42	0.98	68.4	3.34	0.022
Average		0.60	4.28	2.55	68.5	8.53	0.110
Standard Deviation		0.43	3.38	3.07	76.8	13.24	0.056

<sup>a</sup>Each value represents the average concentration of all samples collected at that location

<sup>b</sup>Single samples

TABLE II. Comparison of Metals in Connecticut Honey with Those Reported in United States Honey

Metal	United States (5,6)		Connecticut	
	Range ( $\mu\text{g/g}$ )	Average ( $\mu\text{g/g}$ )	Range ( $\mu\text{g/g}$ )	Average ( $\mu\text{g/g}$ )
Cu	0.14-1.04	0.40	0.20-1.70	0.60
Fe	0.70-33.50	5.1	1.17-11.00	4.3
Mg	7-126	29	14.3-247	69
Mn	0.17-9.53	2.2	0.28-37.8	8.5

were slightly higher in Connecticut honey than those reported in honey collected throughout the United States. Considering the variability in the range of concentrations, these differences may be considered insignificant.

Average concentrations of zinc and lead were not reported for honey produced in the United States. Values for these metals were found in studies of Australian and Japanese honey (7). The average concentration of zinc in honey produced in Australia (2.0  $\mu\text{g/g}$ ) was similar to the average concentration of zinc found in honey collected for this study (2.6  $\mu\text{g/g}$ ). Concentrations of lead in honey from Hungary, Australia and Japan ranged from 0.05 to 6.3  $\mu\text{g/g}$ . The range of lead concentrations in Connecticut honey was 0.022  $\mu\text{g/g}$  to 0.16  $\mu\text{g/g}$ .

Table III lists some foods which contain high or low concentrations of some of the essential elements analyzed in this study. A comparison of these values with the averages shown in Table I indicated that Connecticut honey was a poor or below average source of these metals.

Daily per capita tolerance levels of mercury and lead were listed by the World Health Organization (8). Total daily dietary tolerance of lead and mercury were 429  $\mu\text{g/person/day}$  and 42.9  $\mu\text{g/person/day}$  respectively. Based on an average honey consumption in North America in 1970 of 1.92 g/person/day (7), the average lead intake as a result of eating honey produced in Connecticut would be 0.21  $\mu\text{g/day}$  or 0.05% of the daily tolerance. The values for mercury would be  $< 0.0096 \mu\text{g/day}$  or  $< 0.02\%$  of the daily tolerance levels.

In a total dietary survey conducted by the FDA in 1973, arsenic intake was listed at an average of 3.8  $\mu\text{g/person/day}$  (8). Less than 0.019  $\mu\text{g}$  of As/day or less than 0.5% of the average total daily intake of arsenic would be consumed from eating honey produced in Connecticut.

A screen for the presence of halogenated hydrocarbons was performed on the samples listed in Table I. Concentrations of these compounds were not found in any of the samples above the limit of detection (approx. 10 ng/g).

TABLE III. High and Low Dietary Concentrations of Essential Elements<sup>a</sup>

Metal	Rich Source		Poor Source	
	Food	Conc. ( $\mu\text{g/g}$ )	Food	Conc. ( $\mu\text{g/g}$ )
Cu	Liver	44	Milk	0.2
Fe	Liver	80	Apples	1.0
Mg				<250
Mn	Nuts	200	Citrus Fruits	0.3
Zn	Shellfish	90	Apples	0.3

<sup>a</sup>Upshaw et al (9)

## CONCLUSION

The concentrations of metals found in a representative set of honey samples collected in Connecticut were found to be comparable to values reported internationally. Connecticut-produced honey was shown to contain concentrations of heavy metals far below hazardous amounts. The concentrations of all metals analyzed in this study may be considered within normal ranges. The honey samples were also found to be free of hazardous concentrations of halogenated hydrocarbons.

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