

# Distribution of elements in honeys and effect of a thermoelectric power plant on the element contents

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Lead, cadmium, iron, copper, zinc, manganese, calcium, magnesium and potassium contents of Turkish honeys were investigated. Most of the elements were more abundant in honeydew honeys than in flower honeys. But calcium content of honeydew honeys was smaller than in flower honeys. Considerable temporal fluctuation of elements was observed in honeydew honeys produced in the same area and from the same hives during autumn 1992, 1993 and 1994. Element contents of Turkish honeys showed high coefficients of variation, and the coefficients of variation decreased with increasing amounts of total minerals in honeys. The thermoelectric power plant which was 15–18 km away from the hives did not cause any pollution in honeys. Turkish honeys did not show any contamination of lead, cadmium, iron or zinc, and amounts of these elements were found well below the permitted limits. © 1998 Elsevier Science Ltd. All rights reserved

## INTRODUCTION

Apiculture in Turkey is in a state of expansion. In 1993, there were 3 686 000 hives, representing a yearly increase of 4.1%. Honey production in that year was 59 207 metric tons (Anon., 1995). Honey is mainly produced in the middle and west of Turkey. Vegetation cover in these areas is characterized by citrus, olive tree, pine, cotton plant, sunflower, apple, peach, plum, apricot, almond, agnus-castus, thyme, broom, heather and abundant pasture. However, total number of plant species in Turkey is nearly 10 000 (Anon., 1994).

Sugar, ash and moisture contents of Turkish honeys were determined previously (Temiz and Şengonca, 1981; Kurt and Yamankaradeniz, 1982). Mineral content was studied only briefly by Sevimli *et al.* (1992). They determined K, Br, As, Sb, Fe, Zn, Cr and Co contents of five different honeys. As the mineral content has been largely ignored until now, it seemed of interest to determine this parameter in Turkish honeys.

In addition, honey has been considered a suitable material for monitoring environmental contamination. Honeybees may forage in a variety of environments and hence they effectively sample their surroundings for the constituents in or on the forage plants, the soil and the atmosphere of the area. Some data on the mineral contents of honeys have been reported. Tatsuno *et al.* (1968) determined ash, copper and lead contents of 21

commercial honeys and three collected honeys. Copper and lead contents of the honeys were within the ranges of 0–2.6 ppm and 0.2–6.3 ppm, respectively. Varju (1970) investigated mineral composition of acacia honeys and its correlation with those of the plant and the soil. He found that the honeys of acacia trees grown on soils rich in Ca and Fe were richer in these elements than the others. Borukh and Panchenko (1973) analysed mineral components of honeys of the mountain-forest zone of the Ukrainian Carpathians. Makarochkin (1974) did spectral analysis of the mineral contents of honeys and reported that trace element contents of the honeys depended on the geographical area.

Tong *et al.* (1975) analysed 19 samples of honey taken from grocery shelves, near zinc mines, adjacent to an industrial area, and near a major highway, and one sample collected in 1899. They determined 47 elements in the honeys and reported that certain samples of honey produced in the vicinity of the New York State Thruway appeared to contain elevated levels of certain elements known to be emitted by the traffic, such as aluminium, barium, calcium, copper, magnesium, nickel, palladium and silicon. Honey that had come into contact with metal equipment in processing contained the greatest amounts of zinc and tin.

Crane (1975) determined the concentrations of K, Na, Ca, Mg, Fe, Cu, Mn and other minerals in flower honeys and honeydew honeys. Mean values of minerals

as ppm for flower honeys were: K 205, Na 18, Ca 49, Mg 19, Fe 2.4, Cu 0.29, Mn 0.30. The mean values for honeydew honeys were: K 1676, Na 76, Ca 51, Mg 35, Fe 9.4, Cu 0.56, Mn 4.1. Honeydew honeys contained rather elevated levels of the above minerals except calcium which remained nearly constant.

Gilbert and Lisk (1978) investigated the use of honey as an environmental indicator of radionuclide contamination. They detected radioactive elements in honey, but at lower concentrations than those present in bees.

In the study of Morse and Lisk (1980), 16 elements were determined in honeys from the United States, Mexico, El Salvador and China to observe possible significant differences in their elemental compositions. They reported rather high levels of some trace elements. Concentration ranges of these elements as ppm were: Cd 0.102–0.267, Cr 0.843–2.67, Co 0.456–0.770, Fe 5.80–183, Pb 1.79–3.19, Ni 0.304–1.25, Zn 2.00–172. The source of these metals was considered as steel or galvanized containers used in processing, shipping or storage.

D'Ambrosio and Marchesini (1982) determined heavy metal contamination of seven honey samples. Samples from polluted areas showed high heavy metal levels. In addition, in one polluted area, autumn honey contained 13.8 ppm Fe, 2.25 ppm Mn and 2.37 ppm Pb, as compared to 8.2, 0.50 and 0.083 ppm, respectively, in spring honey. Cimino *et al.* (1984) studied the effect of volcanic activity on honey. Samples of the honey produced by bees in the vicinity of Mount Etna volcano, in Sicily, were analysed to determine the concentrations of 24 elements. The samples appeared to contain, in relation to others from unpolluted areas, elevated levels of elements such as As, Ba, Co, K, Li, Mn, Sr and Zn. The metal enrichment was correlated to element contents of the Mount Etna lava ashes. The other reports on the elemental analysis of honey are: analysis of trace metals in Polish and foreign honeys, in which high levels of Zn attributable to unsuitable processing and packaging were observed (Gajewska *et al.*, 1984); determination of trace elements in honey from the Sumadija region in Yugoslavia, in which possible uptake of metals from containers and equipment used for honey collection and processing was evaluated, and a high level of Zn was observed (Darmati *et al.*, 1985); the heavy metal contents of Swiss and foreign honeys, in which a high level of Pb in the Swiss honey in polluted areas was evaluated (Bogdanov *et al.*, 1985); analysis of lead, cadmium and manganese in German honeys, in which it was found that the level of metal contamination increased from spring to autumn (Stein and Umland, 1986); the analysis of birch sap and honey to predicate the metal-loading over a large area in Münster and its surroundings in Germany, in which dependence of the metal concentrations in birch sap and honey on the pH-value and the metal contents of the soil was established (Stein and Umland, 1987); metallic impurities in honeys imported into Poland, in which the highest degree of Zn, Fe and

Pb in honeys imported from China and North Korea was established (Gajek *et al.*, 1987).

Jones (1987) investigated the potential use of honey as an indicator in mineral prospecting and environmental contamination. Silver, Cd, Cu and Pb levels were reported in honeys collected throughout the UK. The element contents of honeys were investigated in relation to those in the soils collected from within the foraging area. Considerable spatial and seasonal fluctuations were apparent. No correlations were observed between honey and soil concentrations for either Cu or Pb. It was concluded that the low concentrations of heavy metals in honey and their inherent variability (due to differences in floral source, foraging area, atmospheric particulates on the flower, etc.) detract from the reliable use of honey as a monitoring tool. Consequently honey appeared as a less sensitive indicator than honeybees. It is also possible to see additional reports on the elemental analysis of honey, its utilization as an indicator of environmental pollution and relationship between the mineral composition of honey and its geographical origin (Serra-Bonvehi, 1989; Favaro *et al.*, 1990; Rodriguez-Otero *et al.*, 1994).

This work was undertaken to determine distribution of elements in honeys. The aim was also to investigate the effect of a thermoelectric power plant on the environmental pollution, using honey as a contamination indicator.

## MATERIALS AND METHODS

### Samples

Samples were 74 liquid honeys. Eleven samples were flower honeys collected from various areas in Turkey during 1993 and 1994. Fourteen of them were honeydew honeys taken from grocery shelves over the years 1993 and 1994. Sixteen samples were honeydew honeys collected from various locations during 1993 and 1994. The remaining 33 samples were honeydew honeys produced in the vicinity of a thermoelectric power plant (Yatağan thermoelectric power plant in the south-west of Turkey). Twelve of these honeys were produced in autumn 1992, twelve samples in autumn 1993 and nine samples in autumn 1994.

The honey samples were stored in glass jars. Each jar contained about 500 g of honey and was fitted with an iron lid plated with lac. The honey samples were kept at room temperature until analysis.

### Lead and cadmium analyses

Lead and cadmium contents of honeys were determined as described previously (Anon., 1981) with some modifications. Twenty grams of honey sample were placed in a 100 ml Pyrex beaker, and 2 g of  $Mg(NO_3)_2 \cdot 6H_2O$  were added as ashing aid. Samples were dried at 105°C for

2 days, followed by ashing at 450°C. Ashing was completed with a little nitric acid. The ash was extracted with HCl + HNO<sub>3</sub> mixture and the extract was buffered to a pH of 4 with CH<sub>3</sub>COONa. Lead and cadmium were extracted into methyl isobutyl ketone as their ammonium pyrrolidine-dithiocarbamate complexes and then determined by flame atomic absorption spectroscopy at 283.3 nm and 228.8 nm, respectively, by using an air-acetylene flame. Measurements were made without delay after the solvent extraction. Cadmium was determined immediately and lead within 2 h.

Lead and cadmium concentrations were determined as three replicates with a Pye Unicam SP-9 atomic absorption spectrophotometer. To eliminate the errors derived from matrix effect and volatilization of lead at 450°C, a standard addition method was used instead of plotting a calibration curve (Skoog and West, 1981). To apply the standard addition technique, 20 g of honey sample was taken and 1 ml of Pb-Cd working solution was added which contained 1 ppm of Pb and 0.1 ppm of Cd. A standard-added sample was analysed in the same way as those without standard addition. The number of replicates were also three for the standard-added samples. Before applying the standard addition technique, a calibration curve was obtained to see the linear relationship between absorbance and lead concentration in the concentration range being worked. A similar curve was plotted for the linear relationship between absorbance and cadmium concentration.

Cd(NO<sub>3</sub>)<sub>2</sub>·4H<sub>2</sub>O is a volatile compound with a boiling point of 132°C (Weast, 1974), which may allow cadmium losses during the ashing procedure. To prevent the formation of this compound, moisture was completely evaporated at 105°C before ignition.

#### **Iron, copper, zinc, manganese, calcium, magnesium and potassium analyses**

Ten grams of honey sample was transferred into a 50 ml Pyrex beaker. After drying at 105°C for 2 days, the sample was ashed at 600°C. The ash was extracted with HCl + HNO<sub>3</sub> mixture, and the extract was diluted to 50.0 ml. Mineral concentrations were determined according to Whiteside (1976) using a calibration curve. Mineral materials were analysed as three replicates.

Iron, copper, zinc and manganese were determined by atomic absorption spectroscopy at 248.3, 324.8, 213.9 and 279.5 nm, respectively, using an air-acetylene flame, after dilution of 8 ml of the ash solution with 2 ml of LaCl<sub>3</sub> solution. The LaCl<sub>3</sub> solution (2 g LaCl<sub>3</sub>·7H<sub>2</sub>O/l) was added to eliminate anion interference (Skoog and West, 1981).

Calcium and magnesium were also determined by atomic absorption spectroscopy at 422.7 and 285.2 nm, respectively, using an air-acetylene flame, after dilution of 1 ml of solution, which had been used in the previous step, with 9 ml of distilled water.

Three ml of 400 ppm Na<sup>+</sup> solution was mixed with 1 ml of the above solution, which had been used in calcium, magnesium determination, and diluted to 50.0 ml by distilled water. Potassium concentration of this solution was determined by emission spectroscopy at 766.5 nm using air-acetylene flame. The addition of the Na<sup>+</sup> solution was needed to eliminate ionization interference (Skoog and West, 1981).

#### **Statistical analysis**

The Kruskal-Wallis test followed by the Mann-Whitney test was used to compare element contents of flower honeys, grocery shelf honeys and honeydew honeys from unpolluted areas. Comparison of element contents of honeydew honeys produced in the vicinity of the thermoelectric power plant during autumn 1992, 1993 and 1994 was accomplished in the same way.

Kruskal-Wallis and Mann-Whitney tests were also used to compare the coefficients of variation of elements in honeydew honeys produced in the vicinity of the thermoelectric power plant at different times. Mean values for elements in honeydew honeys in the vicinity of the power plant were compared with values in honeydew honeys from unpolluted areas by using only the Mann-Whitney test.

## **RESULTS AND DISCUSSION**

Element contents of flower honeys, grocery shelf honeys and honeydew honeys from unpolluted areas, and contents of honeydew honeys produced in the vicinity of the thermoelectric power plant at different times, together with the corresponding coefficients of variation, are presented in Table 1. In addition, total amounts of minerals of honeys are tabulated in Table 1, which include iron, calcium, magnesium and chiefly potassium and calculated in millimole kg<sup>-1</sup>. Potassium was the most abundant of the elements determined. Comparison of the element contents of the flower honeys, the grocery shelf honeys and the honeydew honeys from unpolluted areas is shown in Fig. 1. Contents of cadmium, iron, copper, zinc, manganese, magnesium and potassium were significantly higher in the honeydew honeys than in the flower honeys ( $p \leq 0.05$ ). Only calcium content of the honeydew honeys was significantly smaller than the flower honeys ( $p \leq 0.05$ ). The change in the lead content was not significant. Total amount of the minerals was 52.5 mmol kg<sup>-1</sup> in the honeydew honeys, but the flower honeys contained 13.4 mmol kg<sup>-1</sup> total minerals. Element contents of the flower and honeydew honeys are in accordance with the results of Crane (1975), who also reported that levels of most elements were higher in honeydew honeys than in flower honeys, except calcium which remained nearly constant. Elemental analysis of the grocery shelf honeys, which were purchased as honeydew honeys, gave intermediate

**Table 1. Elemental analyses of flower honeys, grocery shelf honeys and honeydew honeys from unpolluted areas and analyses of honeydew honeys produced in the vicinity of a thermoelectric power plant**

Honey	Unpolluted area	Vicinity of power plant	Number of samples	Date removed from hive	Pb	Cd	Fe	Cu	Zn	Mn	Ca	Mg	K	Total minerals mmol kg <sup>-1</sup>
Flower	Yes		11	1993–1994	55.2	4.53	4.90	0.350	0.977	0.309	71.5	21.0	416	13.4
					(0.81)	(1.01)	(0.97)	(0.42)	(0.38)	(0.49)	(0.37)	(0.46)	(0.69)	
Grocery shelf	Yes		14	1993–1994	49.3	5.48	7.68	0.621	1.84	0.541	50.1	38.7	852	24.7
					(1.16)	(0.52)	(0.63)	(0.38)	(0.55)	(0.36)	(0.25)	(0.23)	(0.51)	
Honeydew	Yes		16	1993–1994	35.4	10.8	10.4	1.05	1.54	0.752	28.5	55.0	1926	52.5
					(0.47)	(0.62)	(0.69)	(0.22)	(0.52)	(0.22)	(0.91)	(0.08)	(0.27)	
Honeydew		Yes	12	autumn 1992	19.7	0.450	1.18	0.384	79.0	26.0	281		10.7	
					(0.88)	(0.44)	(0.55)	(0.51)	(0.71)	(0.68)	(1.15)			
Honeydew		Yes	12	autumn 1993	43.3	10.5	8.56	1.24	1.34	0.673	19.5	56.1	2196	59.2
					(0.37)	(0.41)	(0.40)	(0.28)	(0.34)	(0.28)	(0.40)	(0.20)	(0.14)	
Honeydew		Yes	9	autumn 1994	32.9	11.0	7.60	1.05	1.34	0.749	52.7	37.4	1132	31.9
					(0.95)	(0.61)	(0.36)	(0.52)	(0.41)	(0.31)	(0.47)	(0.26)	(0.31)	
Mean values for honeydew honeys in the vicinity of power plant					38.8	10.7	12.3	0.901	1.28	0.589	50.2	40.1	1209	

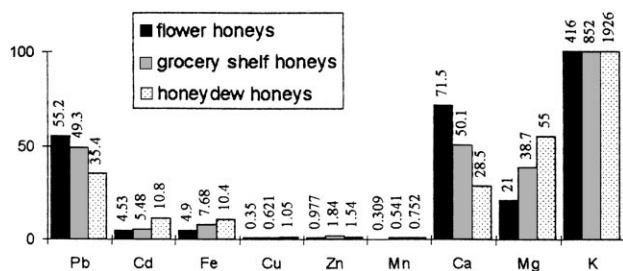
Pb and Cd in ppb, all others ppm; numbers in parentheses are coefficients of variation

values. Iron, copper, zinc, manganese, magnesium and potassium contents of grocery shelf honeys were significantly higher than in the flower honeys ( $p \leq 0.05$ ). Cadmium, copper, manganese, magnesium and potassium contents of the grocery shelf honeys were significantly smaller than those in the honeydew honeys ( $p \leq 0.05$ ). Total amount of the minerals for the grocery shelf honeys was 24.7 mmol kg<sup>-1</sup> which was an intermediate value between those of the flower and honeydew honeys. The grocery shelf honeys might have contained flower honey or other variables (such as spatial or temporal differences) might have caused the decrease in element contents.

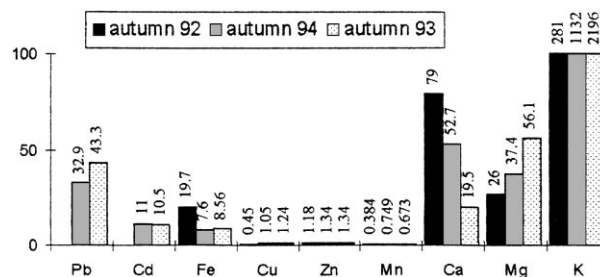
Temporal fluctuation of the elements in honeydew honeys produced in the vicinity of the thermoelectric power plant at different times is seen in Table 1. Although these honeys were produced in the same area and from the same hives, temporal difference caused great fluctuation in the element contents. The element contents of autumn 1992 honeys were the least. Autumn 1993 honeys acquired the greatest amounts of the elements and autumn 1994 product had intermediate values. The amounts of the total minerals were 10.7 mmol kg<sup>-1</sup>, 59.2 mmol kg<sup>-1</sup> and 31.9 mmol kg<sup>-1</sup> for autumn 1992, 1993 and 1994 honeys, respectively. Figure 2 shows that copper, manganese, magnesium and potassium contents of 1993 honeys were higher than

in 1992 honeys ( $p \leq 0.05$ ). Calcium content of 1993 honeys was significantly smaller than in 1992 honeys ( $p \leq 0.05$ ). 1994 honeys showed intermediate values. Copper, manganese and potassium contents of autumn 1994 honeys were significantly higher than in 1992 honeys, magnesium and potassium contents of 1994 honeys were significantly smaller than in 1993 honeys ( $p \leq 0.05$ ). On the other hand, iron content of autumn 1992 honeys was unexpectedly high. This increase in the iron content may be to balance the poor mineral content of honey or may be a result of contamination from containers or equipment.

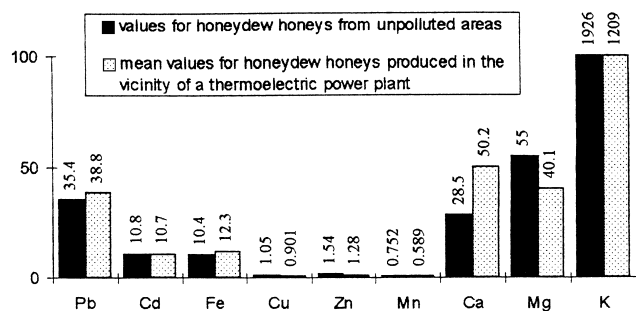
Mean values of the elements for honeydew honeys produced in the vicinity of the thermoelectric power plant at different times are presented in Table 1. In Fig. 3 these values are compared with the values of honeydew honeys from unpolluted areas. Cadmium, copper, zinc, manganese, magnesium and potassium contents of honeydew honeys produced in the vicinity of the power plant were smaller than those from unpolluted areas. Although there was an increase in lead, iron and calcium contents of honeys in the vicinity of the power plant, this increase was not related to any pollution and resulted from poor total mineral content of honeys



**Fig. 1.** Comparison of element contents of flower honeys, grocery shelf honeys and honeydew honeys from unpolluted areas (Pb and Cd in ppb, all others ppm).



**Fig. 2.** Temporal change in element contents of honeydew honeys produced in the vicinity of a thermoelectric power plant at different times (Pb and Cd in ppb, all others ppm). Amounts of total minerals are 10.7 mmol kg<sup>-1</sup>, 31.9 mmol kg<sup>-1</sup> and 59.2 mmol kg<sup>-1</sup> for autumn 1992, 1994 and 1993 honeys, respectively.



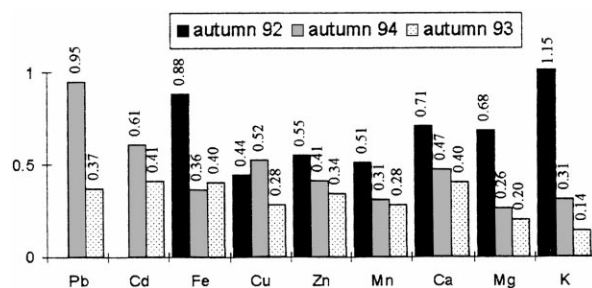
**Fig. 3.** Comparison of element contents of honeydew honeys from unpolluted areas with those in honeydew honeys produced in the vicinity of a thermoelectric power plant (Pb and Cd in ppb, all others ppm).

produced in the vicinity of the power plant. There was no statistically significant difference between values of honeydew honeys from unpolluted areas and mean values for honeydew honeys produced in the vicinity of the power plant. Consequently it appears that the thermoelectric power plant did not cause any pollution in honeydew honeys at a distance of 15–18 km from the hives.

From previous studies, it is seen that industry, mining, automobile exhaust gases, etc., may cause lead and cadmium contamination in honeys (Tatsuno *et al.*, 1968; Morse and Lisk, 1980; D'Ambrosio and Marchesini, 1982; Bogdanov *et al.*, 1985; Stein and Umland, 1986; Gajek *et al.*, 1987). Honey that comes into contact with metal containers or equipment during storage, processing or shipping, may have elevated levels of zinc and iron (Tong *et al.*, 1975; Morse and Lisk, 1980; Gajewska *et al.*, 1984; Darmati *et al.*, 1985; Gajek *et al.*, 1987). The values in Table 1 indicate that there is no contamination of lead, cadmium, iron or zinc in Turkish honeys. Contents of these elements in Turkish honeys are well below the permitted limits of Codex Alimentarius Commission (Anon., 1984).

Element contents of Turkish honeys showed high coefficients of variation (Table 1). High coefficients of variation were also reported in previous studies. Bogdanov *et al.* (1985) calculated coefficient of variation of lead as 1.11 and Rodriguez-Otero *et al.* (1994) found 0.71 for iron. Coefficients of variation of the elements for honeydew honeys produced in the vicinity of the thermoelectric power plant are shown in Fig. 4. It is clearly seen that coefficients of variation of the elements decrease with increasing amount of the total minerals in honey. The use of Kruskal–Wallis and Mann–Whitney tests indicated that coefficients of variation of the elements in autumn 1992 honeys were significantly higher than those in autumn 1994 and 1993 honeys ( $p \leq 0.05$ ).

It is concluded that honeys with low total mineral content and high levels of calcium show high coefficients of variation of the elements. This type of honey may be affected easily by the contamination sources and may have elevated levels of possible contaminants such as lead, iron, etc. If honeys contain high amounts of total



**Fig. 4.** Coefficients of variation of elements for honeydew honeys produced in the vicinity of a thermoelectric power plant at different times. Amounts of total minerals are 10.7 mmol kg<sup>-1</sup>, 31.9 mmol kg<sup>-1</sup> and 59.2 mmol kg<sup>-1</sup> for autumn 1992, 1994 and 1993 honeys, respectively.

minerals, they show a stable elemental composition and hence they are expected to possess a resistance to the effects of contamination sources.

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