JOURNAL OF AGRICULTURAL SCIENCE IN FINLAND
Maataloustieteellinen Aikakauskirja
Vol. 59: 379—385, 1987

The fluoride content of Finnish honey

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Abstract. The content of fluoride was determined with an ion-specific electrode on 59 samples of honey from 47 localities in Finland. The concentrations ranged from 25 to 550 μg/kg and the mean for all the localities was 85.7 μg F^-/kg fresh weight honey. The concentrations were lowest in eastern and northern Finland, where the amount of fluoride in the soil is small, and higher on the coast and areas where more fluoride occurs in the groundwater and bedrock. However, the highest levels of fluoride in honey were detected in the vicinity of Helsinki and some other places where the amount in the groundwater is not especially high. The higher levels in some honey may be due to fluorides introduced into the environment by the activity of man. The effects of fluoride on human health are discussed.

Index words: honey, fluoride, groundwater

Introduction

Fluorine is an element which occurs in compounds everywhere in the lithosphere, but only small amounts are generally found in the biosphere. Fluorine is an important constituent of teeth and bones, and has been considered essential to animal life (WHO 1984). The best known of its biological functions are connected with the use of fluoride in the prevention of dental caries (MURRAY 1986). Especially at higher concentrations, compounds of fluorine are harmful to living organisms and can occur as environmental pollutants (LILLIE 1970, WEINSTEIN 1977, WHO 1984).

The total daily fluoride intake of adult man ranges from about 0.5 to 5.0 mg, mainly depending on its amount in drinking water (WHO 1984). In Finland the daily intake of fluoride in food, excluding drinking water, was estimated to be 0.56 mg (KOIVISTOINEN 1980). As regards the caries-inhibiting effect, the optimal concentration of fluoride in drinking water in a temperate climate is approximately 1 mg/litre, and in many areas water is artificially fluoridated to attain the optimal level (WHO 1984, MURRAY 1986). In Finland the fluoride content in groundwater varies between 0.01 and 6.0 mg, and the average is 0.1 mg/litre (VUORINEN et al. 1986).

Recently it has been established that the caries-preventive effect of fluoride is greatest when it is consumed together with cariogenic sugar products (LUOMA 1985). As honey is a sugary product, its fluoride content can affect its cariogenic potential. Hardly anything is known, however, about the normal levels of fluoride in honey. For example, the comprehensive handbook of honey (CRANE 1975) does not mention fluoride among the over 180 substances so far detected in honey. The aim of the present study was to determine fluoride in honey samples collected from different areas of Finland, and to compare the results with the fluoride content of the groundwater. This study is a part of the investigation of Finnish honey commenced by VARIS et al. (1982, 1983).

Materials and methods

Honey samples

The bulk of the honey originated from the years 1977 and 1978 and has been described in more detail in connection with the analysis of other constituents (VARIS et al. 1982, 1983). The honey samples were stored in a deep freeze until analysed. The samples for the determination of fluoride were selected to be representative with respect to geographical distribution and the occurrence of fluorine in Finnish soils. In several cases samples from the same locality were pooled before the analysis. The Espoo and Helsinki honey included samples from the years 1985 and 1986. The water content of the samples averaged 17.6 % (VARIS et al. 1983).

Analysis for fluoride

The determinations of fluoride were performed by means of a fluoride ion-specific electrode, Orion 94-09, connected with a 901 ionalyzer and 90-01-00 reference electrode (Orion Research Incorporated, Cambridge, Massachusetts 02139). The measurements were made on 20 % (w/v) solutions of honey in the presence of 0.2 M sodium citrate/HCl buffer pH 5.6 at room temperature (20 ± 1°C) under continuous magnetic stirring (KAURANEN 1977). The electrode response was recorded until the change was no more than 0.1 mV/min, which generally took from 10 to 30 min. Standards containing 0.005—0.5 ppm F^- as sodium fluoride were measured in the same conditions and the fluoride concentrations in the samples were evaluated from a standard curve on semilogarithmic paper. All the samples were measured at least twice. The reliability of the method was also checked by addition of a known amount of sodium fluoride to a measured sample. The concentrations of fluoride in melted snow were determined similarly. The reagents were pro analysi grade (E. Merck, Darmstadt) and the solutions were made with double distilled water. The solutions were prepared and stored in polyethylene containers.

Results and discussion

The concentrations of fluoride in 59 samples of honey from 47 localities in Finland varied from 25 to 550 μg/kg and the mean for all localities was 85.7 μg/kg fresh weight honey (Table 1, Fig. 1). The concentrations were lowest in eastern and northern Finland, where the amount of fluoride in the bedrock is low, and higher in localities where more fluoride occurs in the soil. However, the highest concentrations of fluoride in honey were detected in the vicinity of Helsinki and some other places where its amount in the groundwater is not particularly high (Table 1, Figs. 2 and 3). The reasons for these higher values are not completely clear, but evidently fluoride from other sources than the local bedrock has affected its amount in the honey samples. In some cases the great local variation in the fluoride content of the soil can also have affected the results. On average the fluoride content of the honey was about 20 % of that of the groundwater. No correlation was found between the fluoride content and the pollen spectrum of the honey.

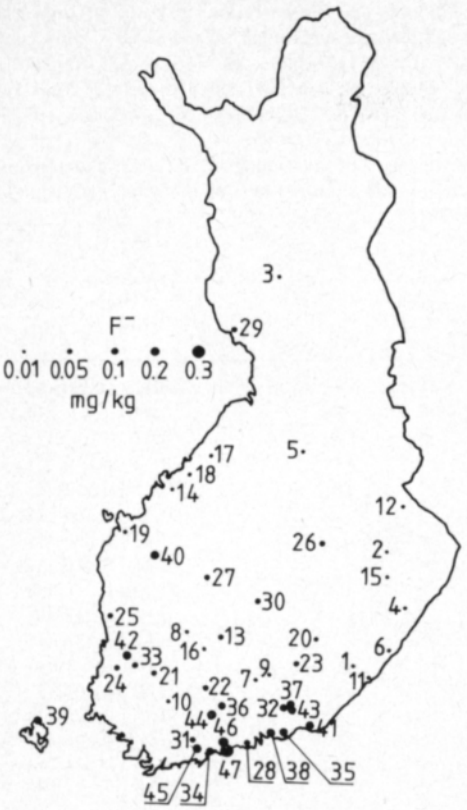

Fig. 1. The content of fluoride in Finnish honey. The numbers refer to the localities in Table 1.

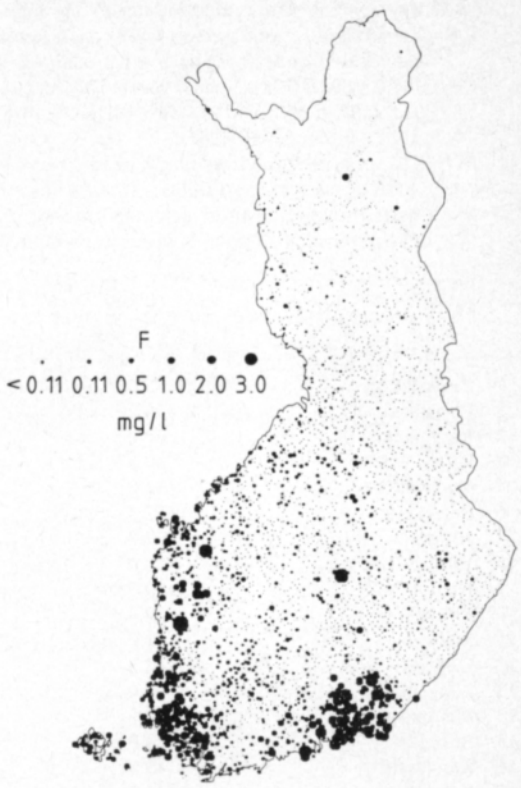

Fig. 2. The content of fluoride in Finnish groundwater according to the investigation of the Geological Survey of Finland (Vuorinen et al. 1986).

So far only a few investigations have been made of the amount of fluorine in honey, and these were mainly made on honey subject to environmental pollution. Tong et al. (1975), who used spark source mass spectrometry to study the effects of pollution on mineral elements in honey in the USA, reported, without closer specification, that the concentrations of fluorine ranged from < 1 to 8900 μg/kg fresh weight. Mayer et al. (1986) have found up to 1200 μg F^- in honey from a fluoride pollution area in Puyallup Valley northwestern USA, and 300 μg/kg in honey from control colonies outside the area. At these levels fluoride did not affect the colony vigor or honey production.

A mixed sample of Finnish honey was analysed in connection with the investigation of the mineral element composition of Finnish foods (Koivistoinen 1980). The fluoride content in this sample, as determined with an ion-specific electrode after ashing, was found to be 800 μg/kg, or about 10 times as high as the average concentration in the present study. The higher level of fluoride in the ashed sample may indicate that only a part of the fluorine in honey is directly detectable with the electrode. As the result is based on only one determination, however, the possibility of an experimental error cannot be excluded. Ashing was also tried in the present study, but proved to be laborious and apt to cause contamination by extraneous fluoride.

The principal source of the mineral elements of honey is the nectar of flowers, and the mineral content of honey is thus dependent on

Table 1. The content of fluoride in honey and groundwater in Finland. The concentrations in honey are in most cases based on one pooled sample. If more than one sample was analysed from the same locality, the results are means of the determinations: No. 11 25.0 (1977), 57.0 (1978); No. 30 47.5, 80.5, 71.0 (1977), 165.0, 100.0 (1978); No. 34 95.0 (1978), and another apiary 109.5 (1985), 120.0 (1986); No. 35 128.5 (1977), 91.5 (1978); No. 43 145.0 (1977), 180.0 (1978); No. 46 230.0 (1977), 210.0 (1978); No. 47 550.0 (1977), 218.5 (1985), 305.0 (1986).
The concentrations in groundwater are based on the investigations of the Geological Survey and are means ± SE of the content of fluoride in wells or springs located not more than 10 km from the apiary. In groundwater the lower limit of determination was 0.1 mg F^-/litre.
The number of samples is given in parentheses.

No.	Locality	Honey µg F^-/kg		Year		Groundwater mg F^-/litre	
				1977	1978		
1	Ruokolahti	25.0			x	0.11 ± 0.010	(8)
2	Polvijärvi	26.2		x		0.24	(1)
3	Rovaniemi mlk	28.0			x	0.12	(1)
4	Kitee	30.3			x	0.12 ± 0.014	(5)
5	Vuolijoki	32.0		x		0.15 ± 0.052	(4)
6	Parikkala	32.5		x		0.61 ± 0.169	(21)
7	Asikkala	37.0		x		0.17 ± 0.066	(5)
8	Ylöjärvi/Kuru	38.0		x		0.31 ± 0.086	(16)
9	Heinola mlk	39.5		x		0.10 ± 0	(3)
10	Ypäjä	39.5		x	x	0.50 ± 0.218	(9)
11	Imatra	41.0	(2)	x	x	0.41 ± 0.186	(10)
12	Lieksa	42.5			x	0.10 ± 0	(8)
13	Längelmäki	43.0		x	x	0.90 ± 0.800	(2)
14	Pietarsaari mlk	43.5			x	0.70 ± 0.070	(3)
15	Liperi	44.0		x		0.20 ± 0.090	(4)
16	Kangasala	45.0		x	x	0.22 ± 0.055	(15)
17	Kalajoki	47.5		x		0.10 ±	(2)
18	Kälviä	49.0		x		0.26 ± 0.165	(2)
19	Mustasaari	49.0		x		0.47	(1)
20	Mikkeli mlk	54.5		x		0.13 ± 0.025	(5)
21	Huittinen	59.5		x		0.10 ± 0	(2)
22	Hattula	60.5		x	x	0.17 ± 0.057	(10)
23	Mäntyharju	61.5		x	x	0.24 ± 0.118	(6)
24	Eurajoki	64.2		x		0.85 ± 0.257	(8)
25	Merikarvia	68.5		x		0.12 ± 0.015	(2)
26	Siilinjärvi	75.5		x	x	0.14 ± 0.037	(3)
27	Ähtäri	82.5			x	0.10 ± 0	(7)
28	Porvoo mlk	85.5		x	x	0.15 ± 0.055	(2)
29	Keminmaa	89.4		x		0.14 ± 0.040	(2)
30	Jyväskylä	92.8	(5)	x	x	0.10	(1)
31	Lohja	93.0		x		0.12 ± 0.022	(4)
32	Elimäki	95.5		x		1.40 ± 0.184	(13)
33	Kokemäki	100.5		x	x	0.13 ± 0.019	(8)
34	Espoo	108.2	(3)		x	0.43 ± 0.178	(6)
35	Pyhtää	110.0	(2)	x	x	2.67 ± 0.592	(3)
36	Hausjärvi	117.5		x	x	0.51 ± 0.140	(13)
37	Valkeala	119.0		x		1.54 ± 0.178	(14)
38	Loviisa	120.0		x	x	1.28 ± 0.361	(4)
39	Geta	121.0			x	1.72 ± 0.313	(6)
40	Ilmajoki	125.0		x	x	0.10 ± 0	(3)
41	Vehkalahti	134.0			x	2.45 ± 0.355	(17)
42	Nakkila	154.0		x		0.24 ± 0.058	(12)
43	Anjalankoski	162.5	(2)	x	x	1.74 ± 0.198	(10)
44	Loppi	167.0		x		0.19 ± 0.092	(4)
45	Siuntio	195.0			x	0.13 ± 0.021	(4)
46	Vantaa (Tikkurila)	220.0	(2)	x	x	0.17 ± 0.044	(3)
47	Helsinki (Viikki)	357.8	(3)		x	0.39 ± 0.145	(3)
	Mean ± SE	85.68 ± 9.08				0.495 ± 0.091	

the plants from which the nectar originates. According to WEINSTEIN (1977), the major site of fluoride accumulation in plants is the leaf, in which the concentration of fluoride generally ranges from 0.5 to 25 mg/kg dry weight. In vegetables in Finland, the fluoride content has been found to vary from 0.02 to about 1 mg/kg fresh weight (KOIVISTOINEN 1980). The amount of fluoride in the vegetation depends on several factors, and often no close relationship has been found between the concentration of fluoride in the soil and that in the plant (WEINSTEIN 1977). Phosphate fertilizers, which generally contain fluoride as an impurity, can increase the amount of fluoride in soil and plants, and there is some evidence that fertilization can also otherwise increase the amount of fluoride in plants (GABOVICH and OVRUTSKIY 1969). Therefore, fertilization could also increase the amount of fluoride in honey produced in intensively cultivated areas.

Although small amounts of fluoride are beneficial to the health and normal development of man and higher animals, larger amounts are toxic to animals and plants, and harmful effects of fluorides have often been observed in the environment. Insects are susceptible to fluoride, and poisoning can occur through fluoride-containing pesticides or industrial emissions (LILLIE 1970, ALSTAD et al. 1982). The toxicity of fluoride to bees depends on several factors, but generally the LD_{50} varies between 5 and 8 μg per bee (MAURIZIO 1960).

Fluoride is not a serious environmental problem in Finland, but fluoride emissions have been observed to affect the vegetation in the vicinity of fertilizer factories at Siilinjärvi and Oulu (KÄRENLAMPI et al. 1982). Symptoms of fluoride intoxication were also noticed in the vicinity of a porcelain factory at Tammisaari in the 1970's. A mixed sample of honey from two apiaries at Siilinjärvi was analysed in the present study, but the amount of fluoride in this sample (No. 26) was not particularly high, evidently because the apiaries were located more than 3 km from the factory (cf. KAURANEN 1978).

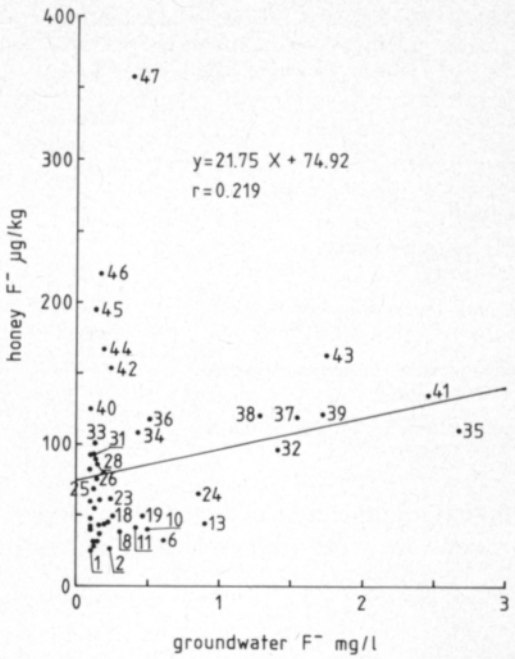

Fig. 3. The correlation between the content of fluoride in honey and groundwater. The numbers refer to the localities in Table 1.

The high level of fluoride in honey from Viikki (No. 47) was unexpected, because the bedrock in the area is not rich in fluoride. The apiary is located near the seashore in the neighbourhood of Helsinki, but no symptoms of pollution are evident in the area. However, determination of fluoride in the snow in spring 1987 indicated that the concentrations were elevated around the city and especially in the vicinity of a porcelain factory located 3 km from the apiary of Viikki (Table 2). At the end of the 1970's the factory started to use kaolin containing less fluoride, which could explain the smaller amounts of fluoride in honey from Viikki in the 1980's.

Air pollution could also explain the higher levels of fluoride in some other honey samples in the present study, e.g. those from Tikkurila, Nakkila and Jyväskylä. The variation in the fluoride content of honey from the same apiary at Jyväskylä (No. 30) could be explained by periodical pollution.

Even the highest levels of fluoride now detected in honey were so low that no effect on

Table 2. The content of fluoride in snow in the vicinity of Helsinki and the apiaries at Viikki and Tikkurila at the end of March 1987.

Location	Fluoride μg F^-/kg snow
The city of Helsinki (Eläintarhanpuisto)	16
The apiary of Viikki, 7 km NE of the city	11
A park near a porcelain factory, 3 km SW of the apiary of Viikki	26
The apiary of Tikkurila, 14 km N of the city	9
Archipelago, 16 km SW of the city	6

human health can be expected. The concentrations were on average around the level found in milk and vegetables in Finland (KOIVISTOINEN 1980). This amount is only 10 % of the optimal concentrations of fluoride in drinking water and 1 % of the level found to afford significant protection against dental caries in sugar products (LUOMA 1985). There is experimental evidence that the cariogenic effect of honey is approximately the same as that of sucrose (BIRKHED et al. 1979; SHANNON et al. 1979).

Acknowledgements. We are indebted to Mr. Heikki Rainio, M.A. for advice on the fluoride analyses performed at the State Institute of Agricultural Chemistry. This study was supported financially by the Juho Vainio foundation.

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Ms received October 14, 1987.

SELOSTUS

Suomalaisen hunajan fluoridipitoisuus

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Fluori on alkuaine, joka luonnossa miltei aina esiintyy yhdisteinään, fluorideina. Fluoridin katsotaan olevan välttämätön luuston normaalikehitykselle ja sillä on myös merkittävä hammaskariesta ehkäisevä vaikutus. Äskettäin on voitu todeta, että tämä vaikutus on suurin jos fluoridia saadaan yhdessä kariesta aiheuttavien sokerituotteiden kanssa. Suuremmissa määrissä fluorideilla on haittavaikutuksia ja monissa tapauksissa fluorin yhdisteet ovat esiintyneet ympäristömyrkkyinä.

Eräiden havaintojen mukaan hunajassa voi esiintyä merkittäviä määriä fluoria. Tiedot hunajan sisältämän fluorin määrästä ovat kuitenkin erittäin puutteelliset. Sen vuoksi on Helsingin yliopiston maatalous- ja metsäeläintieteen laitoksella osana laajemmasta suomalaisen hunajan koostumusta koskevasta tutkimuksesta selvitetty fluoridin määrä 59 hunajanäytteestä 47 paikkakunnalta. Tutkitut hunajat olivat pääosin peräisin vuosilta 1977 ja 1978. Mittaukset tehtiin Valtion maatalouskemian laitoksella fluoridispesifistä elektrodia käyttäen.

Fluoridin määrä tutkituissa näytteissä vaihteli 25:stä 550:een μg/kg ja pakkakunnittain laskettu keskiarvo oli 85.7 μg F^-/kg hunajaa. Pitoisuudet olivat pieniä itä- ja pohjois-Suomessa, missä fluorin määrä maaperässä on vähäinen ja suurempia rannikoilla ja rapakivialueilla, missä fluoria esiintyy enemmän maaperässä ja pohjavedessä. Suurimmat hunajan fluoridipitoisuudet todettiin kuitenkin Helsingin lähistöltä ja eräiltä muilta paikkakunnilta, missä fluorin määrä maaperässä ei luonnostaan ole kovin korkea. Helsingin ympäristössä lumesta keväällä 1987 tehdyt mittaukset viittaavat siihen, että suuremmat fluoridin määrät hunajassa voivat osittain johtua ilman epäpuhtauksista. Fluorin määrää maaperässä voi myös lisätä sen esiintyminen epäpuhtautena fosforilannoitteissa.

Kaikenkaikkiaan fluoridin määrä suomalaisessa hunajassa näyttää olevan niin pieni, ettei sillä ihmisen ravitsemuksen kannalta ole merkitystä. Keskimäärin pitoisuudet hunajassa ovat samansuuruisia kuin maidossa ja vihanneksissa ja suurimmatkin todetut määrät pienempiä kuin fluoridin suositeltu pitoisuus juomavedessä. Näin ollen hunajan sisältämällä fluoridilla ei voi myöskään olla merkittävää hammaskariesta estävää vaikutusta.