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1

The Fluorine Content in Favorite Foods of Japanese.

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I Introduction

The great majority of work on nutrition of fluorine has dealt with anti-caries and mottled enamel[1]. Since current practice in Japan and the United States favors fluoridation of drinking water and fluoridated dentifrices[1,2], these only have been studied in detail. Though fluorine determination has been made on many common foods as the source of dietary fluorine by many groups of workers in recent years[3,4], fluorine in foodstuffs has been treated lightly for the reason that fluorine is made insoluble by calcium[2], and many efforts in this aspect have not been concerned with fluorine chronic intoxication but with the research of the good source of dietary fluorine[3].

However it is well known that phospatic fertillizer used for the production of foodstuffs contains much fluorine as impurities, and that the occurence of mottled enamel does not always parallel the fluorine content in drinking water[5]. So it is considered that fluorine in foodstuffs as drinking water affects the occurence of mottled enamel.

Besides, according to Schatz (1956)[6], fluorine has abad effect upon the central nervous system. From these reasons further understanding of the roll of fluorine upon nutrition requires an appreciation of the fluorine contents of foodstuffs from the view point of medical geography.

This paper deals with the fluorine content of Japanese foods and daily intake of fluorine by Japanese in 1958 and 1965, reviewing some interesting resluts about the geographical correlation between the fluorine content of rice and "miso", favorite foods of Japanese, and the human mortality with gastric cancer in Japan, reported in our previous papers[7,8].

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II Experimental

Materials: The samples which were obtained at various local markets in 1958-1965 were analyzed for fluorine as soon as possible after delivery. When the experiment could not be done within 1 day after receiving fresh materials, the samples were stored after drying. Rice samples 1960 were given by Food Management Training School in Mikawa-Ichinomiya Aichi-ken.

Mothod of analysis: Samples were ashed with lime suspension at 500°-600°C accordingly to A.O.A.C. method[12], and were distillated by the procedure outlined by Willard and Winter[2]. Fluorine was determined by Zirconium eriochrome cyanin R method[2]; on some materials fluorine was measured by Trium nitrate volumetric method or Aluminum haematoxilin method.

III Results and Discussion

1. Common foods obtained commercially in Japan

Table 1 was compiled from results obtained by determining fluorine in Japanese foods. The results show that mean values of fluorine contents of polished and unpolished rice in 1960, which are Japanese staple foods, were 9.67 and 19.96 ppm respectively.

The fluorine contents of cereals such as rice,

wheat and barley showed a yearly increase from 1958 to 1965. The content of fluorine in vegetables increased, too. Cucumbers in 1965 contained a significantly larger amount of fluorine than did those in 1958, 5.04 ppm as compared with 0.34. The uptake and translocation of fluorine into crops will be affected by temperature, fertilizer, soil and varieties. With respect to fertilizer, the results of a few experiments[9] have shown that increasing the amount used of phosphatic fertilizer, which generally contains fluoride as impurities, caused an increase in the content of fluorine in rice.

On the other hand salted fishes contained much fluorine, the average content being 28.52 ppm on the dry basis. The contents of fluorine in "vegetable" fruits were not more than those of other foods: for instance, a water melon contained 5.04 ppm of fluorine on a fresh basis. But this value is significant, for a water melon is eaten in summer instead of drinking water which generally contains not more than 0.8 ppm of fluorine in Japan. The water soluble fluorine in a sheet of chewing gum was 0.94–925 μg and hundreds ppm of fluorine was found as impurities in eutrophics and other medical supplies.

2. Some materials other than foods

In previous papers[7,10] the contents of fluorine in cigarettes and soot have been reported. As shown in Table 2, the largest value of cigarettes was 640.0 ppm. Soot varied from 83.1 ppm to 128.7 ppm of fluorine. Coal tar and asbesto contained fluorine of 103.0 ppm and 250.3 ppm respectively, and phosphatic fertilizers varied from 0.01 to 9.88% of fluorine.

3. City water

As many cities have service water supplied insteed of well water now, it is very necessary to determine the fluorine content of city water, which is almost never fluoridated in Japan excluding the service water of Yamashina (Kyoto). In Table 1 the maximum and minimum contents of fluorine in service water of 109 cities in Japan are recorded. The water sample taken in Osaka showed the maximum content of 0.54 ppm.

Tablel 1 The Content of Fluorine in Common Foods

Food	Producing disrict	Date	Number of samples tested	Fluorine (on the fresh basis)
				ppm
Rice, lowland nonglutionous unpolished	Aichi	1958	97	5.73
	Aichi	1960	6	13.95
	Aichi	1962	7	13.89
	Aichi	1964	7	14.50
Rice, lowland glutinous unpolished	Aichi	1958	83	6.19
	Aichi	1960	6	9.98
Rice, lowland nonglutinous polished	Aichi	1957	10	4.92
	Aichi	1958	17	4.73
Rice, lowland nonglutinous unpolished	Japan	1960	279	19.96
Rice, lowland glutinous unpolished	Japan	1960	171	22.95
Rice, upland nonglutinous unpolished	Japan	1960	63	13.33
Rice, upland glutinous unpolished	Japan	1960	65	23.01
Rice, lowland nonglutinous polished	Japan	1960	78	9.67
Barley	Aichi	1962	5	8.95
	Aichi	1965	4	10.01
Wheat	Aichi	1958	12	5.17
	Aichi	1962	2	6.90
	Aichi	1965	8	8.50
Spinach	Aichi	1958	11	1.97
	A,T.*	1965	5	13.31
"Komatsuna"	Aichi	1958	11	1.82
	A,T.*	1965	5	10.55
Chinese cabbage	Aichi	1958	10	0.87
	A,T.*	1965	5	2.01
Cabbage	Aichi	1958	9	1.12
	A,T.*	1965	5	5.89
Trefoil	Aichi	1958	11	3.47
	A,T.*	1965	4	19.25
Welsh onion	Aichi	1958	9	1.89
	A,T.*	1965	4	5.62
Onion	Aichi	1958	7	0.79
	A,T.*	1965	6	0.97
Shallot	Aichi	1958	7	1.54
	A,T.*	1965	1	3.69

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ıichi	1958	7	1.54
ı,T.*	1965	1	3.69

Food	Producing district	Date	Number of samples tested	Fluorine (on the fresh basis) ppm
Japanese parsley	Aichi	1958	8	2.07
	A,T.*	1965	5	6.72
Radish	Aichi	1958	9	0.96
	A,T.*	1965	4	7.50
Carrot	Aichi	1958	8	2.13
	A,T.*	1965	7	12.05
Burdock	Aichi	1958	11	2.33
	A,T.*	1965	8	8.19
Potato	Aichi	1958	11	1.54
	A,T.*	1965	4	13.89
Sweet potato	Aichi	1958	10	3.74
	A,T.*	1965	3	3.72
Taro	Aichi	1958	10	1.76
	A,T.*	1965	3	7.46
Egg apple	Aichi	1958	11	0.68
	A,T.*	1965	6	5.73
Pumpkin	Aichi	1958	10	1.10
	Aichi	1965	1	5.82
Water melon	Aichi	1958	4	0.48
	Aichi	1965	2	4.47
Cucumber	Aichi	1958	10	0.34
	A,T.*	1965	7	5.04
Lotus rhizome	Aichi	1958	2	1.72
	Aichi	1965	1	18.51
Soy bean	Aichi	1958	10	6.11
	A,T.*	1965	3	8.33
Field pea	Aichi	1958	6	1.60
	A,T.*	1965	2	12.55
Kidney bean	Aichi	1958	11	1.04
	A,T.*	1965	3	10.84
Broad bean	Aichi	1958	11	1.40
Sesame seed	Aichi	1960	1	31.32
	Aichi	1965	1	8.14
Butterbur	Aichi	1958	11	1.01
	Aichi	1965	2	1.82
Flower of the butterbur	Aichi	1965	1	13.18
Apple	Yamagata	1965	1	9.92
Japanese persimmon	Aichi	1958	3	2.27
	Gifu	1965	1	4.16
Pear	Nagano	1965	1	3.54
Peach	Aichi	1965	1	1.77
Grape	Gifu	1965	1	2.37

Food	Producing district	Date	Number of samples tested	Fluorine (on the fresh basis) ppm
Fig	Aichi	1958	3	1.24
Green tea	Aicci	1958	3	88.75
	Aichi	1962	8	599.50
Powdered tea	Kyoto	1958	1	76.00
	Aichi	1962	3	1272.00
Quick coffee	Aichi	1965	2	11.33
Sugar cane	Aichi	1961	1	30.12
Refined sugar	Aichi	1958	9	0.31–21.88
Table salt	Aichi	1960	2	8.33
Japanese pepper	Aichi	1960	1	36.21
Pepper	Aichi	1960	1	22.39
Water cress	Aichi	1960	1	26.49
Capsicum fruits	Aichi	1960	1	41.04
Ginger	Aichi	1960	1	46.70
Garlic	Aichi	1960	1	2.05
"Shichimi" capsicum fruits	Aichi	1960	1	28.26
Milk	Aichi	1958	3	1.64
White of an egg	Aichi	1958	3	1.27
Yolk	Aichi	1958	2	5.36
Eel	Aichi	1958	1	3.54
Crucian	Aichi	1958	2	4.33
Salted fish	Aichi	1960	14	** 6.99–24.37
"Tsukudani" or preserved food boiled down in soy	Aichi	1960–1965	31	2.38–38.10
"Takuan" pickles	Aichi	1960	5	12.09
"Soy"	Aichi	1960	8	8.19–18.22
"Miso" or bean paste	Japan	1960–1963	84	3.3–20.00
Vinegar	Aichi	1965	1	0.098
Rape oil	Aichi	1962	1	25.00
Rape seed	Aichi	1962	1	58.75
Chemical condiments	Aichi	1962	1	26.45
Eutrophics a	Aichi	1962	1	28.94
b	Aichi	1962	1	503.77
c	Aichi	1962	1	259.25
City water	Japan	1965	109	0.02–0.54
Chewing gum	Aichi	1965	75	(water soluble fluorine) 0.94–925 μg/fluorine

* "Aichi and Tokyo" was abbreviated to A,T.

** On the dry basis, the average content was 28.52 ppm.

4. Daily intake of fluorine from the diet of a village in 1958.

In Table 3 the daily intake of fluorine by the individual from the very plain but ordinary diet[11] in villages of the central Japan in 1958 was estimated. Much fluorine was given by the diet for winer. In Shippo village the fluorine intake of a person from the diet for winter in 1958 was 4.38 mg daily, in Yatomi 3.10 mg. In 1965 the average intake of fluorine from the same diet reached 11.13 mg.

5. The geographical correlations between the fluorine contents of rice and "miso" or bean paste and the human mortality with gastric cancer in Japan.[7,8]

The content of rice was correlated with the human mortality with gastric cancer in Japan at the level of significance of 0.1% with coefficiants of + 0.615 for the male death rate and + 0.554 for the female death rate respectively (Figs.1&2).

The fluorine content of "miso" or bean paste, which is of vital importance in Japan, was positively correlated with the death rate, too (Figs. 3 & 4).

6. Geographical and yearly correlations between the mortality with gastric cancer and the amounts of phosphatic fertilizer applied per 10 a.

A yearly increase in the amounts of phosphatic fertilizer applied per 10 a was correlated with the increase in mortality with gastric cancer in Japan, the correlaton coefficient being + 0.998 (Fig. 6). As reported in an earlier paper,[7] there

Table 2 The Content of Fluorine of Materials Other Than Food

Samples	Number of samples tested	Fluorine
		ppm
Cigarette, (domestic) 1960	10	14.4–103.8
Cigarette, (domestic) 1962[10]	16	42.7–640.0
(imported) 1962[10]	19	35.6–462.4
Soot, made from firewood	1	123.51
Soot, made from coal	1	120.27
Soot, made from heavy oil	2	83.10
Soot, made from lignite	1	128.71
Coal tar a	1	82.55
b	1	103.00
Asbesto	1	250.33
$CaCO_3$ (reagent) a	1	10.00
b	1	112.00
MgO (reagent)	2	317.00
CaO (reagent) a	1	450.00
b	1	365.00
c	1	5.00
Phosphatic fertilizer[7]		0.01–9.88%

Table 3 Daily Intake of Fluorine by the Individual From the Rural Diet[12] in Aichi-ken

	1958								1965			
	Shippo-mura* in Aichi				Yatomi-cho in Aichi				The average in Aichi			
	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter
	mg	mg	mg	mg	mg	mg	mg	mg	mg	mg	mg	mg
The mornning meal	0.572	0.238	0.625	0.664	0.546	0.218	0.563	0.612	2.950	2.295	2.748	3.232
The midday meal	0.370	0.060	0.245	1.310	0.308	0.052	0.250	1.252	3.322	2.282	2.928	4.098
The evening meal	0.217	1.030	1.430	0.358	0.158	1.052	1.338	0.309	2.452	1.792	2.632	3.545
Drinking water	2.050	2.050	2.050	2.050	0.925	0.925	0.925	0.925	0.250	0.250	0.250	0.250
(Tea)	(0.0004)	(0.0004)	(0.0004)	(0.0004)	(0.0004)	(0.0004)	(0.0004)	(0.0004)	(0.0004)	(0.0004)	(0.0004)	(0.0004)
Total	3.209	3.378	4.350	4.382	1.937	2.247	3.076	3.098	8.974	6.619	8.558	11.125

* Shippo-mura is an area of mottled enamel.

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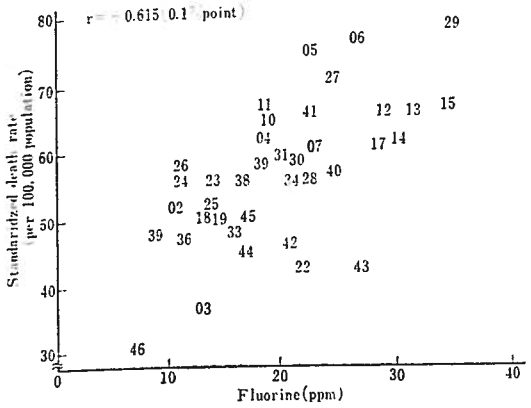
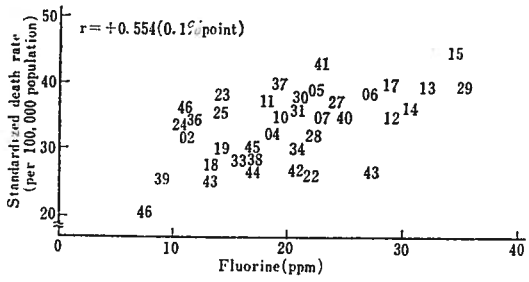
	1965			
	The average in Aichi			
er	Spring	Summer	Autumn	Winter
ıg	mg	mg	mg	mg
.2	2.950	2.295	2.748	3.232
52	3.322	2.282	2.928	4.098
09	2.452	1.792	2.632	3.545
25	0.250	0.250	0.250	0.250
04	(0.0004)	(0.0004)	(0.0004)	(0.0004)
98	8.974	6.619	8.558	11.125

Fig. 1 The Geographical Correlation between the Fluorine Content of Rice and the Male Standardized Death ♯ rate with Gastric Cancer in 1960[7]



* Prefecture is denoted by the serial number given in Fig. 5.

Fig. 2 The Geographical Correlation between the Fluorine Content of Rice and the Female Standardized Death Rate with Gastric Cancr in 1960[7]

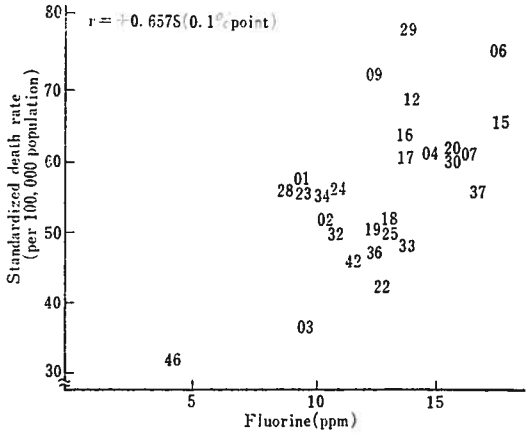


* Prefecture is denoted ♯ by the serial number given in Fig. 5.

was a geographical correlation between the amounts of phosphatic fertilizer applied per 10 a of paddy field and the mortality with gastric cancer in Japan, too.

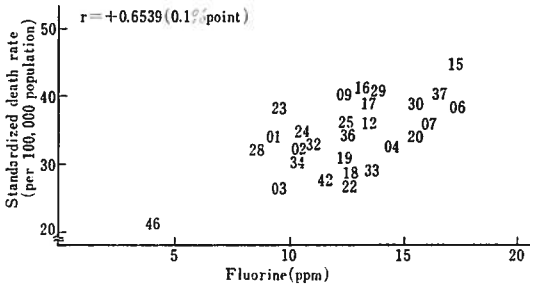
From the above-mentioned results the authors conclude that the daily fluorine intake into the human body is to be investigated with different districts and different crop years from the view point of medical geography or epidemiology, and that further experiments should be done on

Fig. 3 The Geographical Correlation between the Fluorine Content of "Miso" and the Male ♯ Standardized Death Rate with Gastric Cancer in 1960[8]



* Prefecture is denoted by the serial number given in Fig. 5.

Fig. 4 The Geographical Correlation between the Fluorine Content of "Miso" and the Female Standardized Death ♯ Rate with Gastric Cancer in 1960[8]



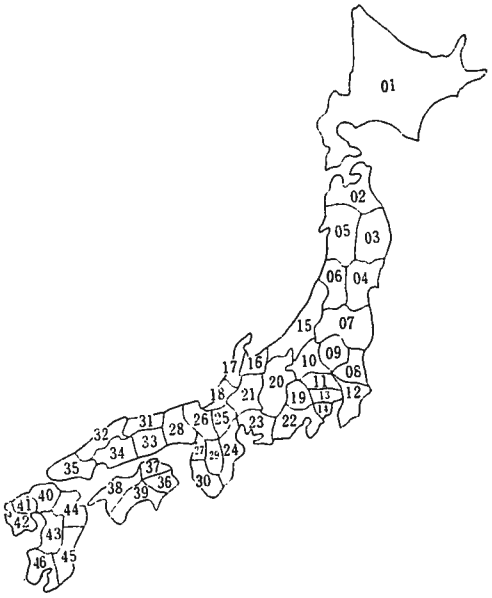
* Prefecture is denoted by the serial number given in Fig. 5.

fluoride, of which no role on cancer is known, from many aspects such as the effects on virus, the relation to other components and the so-called paradoxical effects of fluoride.

Ⅳ Summary

The fluorine content of Japanese foods was determined and the daily intake of fluorine by

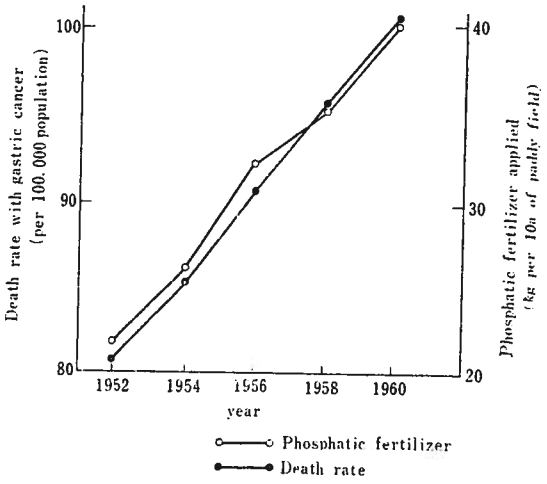
Fig. 5 Map of Japan



The serial numbers given here denote prefectures. Prefecture is an administrative area over cities, towns and villages.

01	Hokkaido	24	Mie
02	Aomori	25	Shiga
03	Iwate	26	Kyoto
04	Miyagi	27	Osaka
05	Akita	28	Hyogo
06	Yamagata	29	Nara
07	Fukushima	30	Wakayama
08	Ibaraki	31	Tottori
09	Tochigi	32	Shimane
10	Gumma	33	Okayama
11	Saitama	34	Hiroshima
12	Chiba	35	Yamaguchi
13	Tokyo	36	Tokushima
14	Kanagawa	37	Kagawa
15	Niigata	38	Ehime
16	Toyama	39	Kochi
17	Ishikawa	40	Fukuoka
18	Fukui	41	Saga
19	Yamanashi	42	Nagasaki
20	Nagano	43	Kumamoto
21	Gifu	44	Oita
22	Shizuoka	45	Miyazaki
23	Aichi	46	Kagoshima

Fig. 6 Amounts of Phosphatic Fertilizer Applied to Paddy Field and Death Rate with Gastric Cancer in Japan



the individual from the rural diet in the central Japan was estimated with different crop years. In 1958 the fluorine intake of a person was 4.38 mg daily in winter, and in 1965 was 11.13 mg. This increase was due to the yearly increase of the fluorine content of Japanese foods. The contents of fluorine of rice and "miso", which are the mainstay of the diet in Japan, showed geographical correlations with human mortality with gastric cancer.

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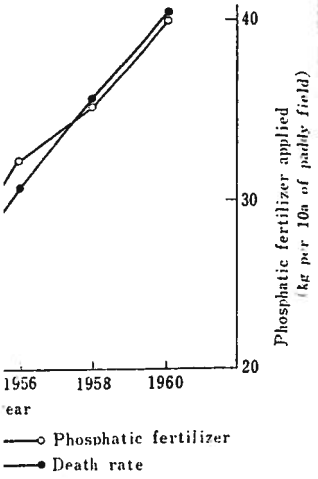
f Phosphatic Fertilizer Paddy Field and Death Gastric Cancer in Japan



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日本人の好む食品のフッ素含有量

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日本食品のフッ素含有量を，AOAC法による灰化，Willard Winter 法による過塩素酸からの水蒸気蒸溜，ジルコニウムエリオクロシアニンR法による比色定量によつて測定し，日本人1人1日あたりのフッ素摂取量を1958年の愛知県1農村の食餌献立にしたがつて計算したところ，フッ素含有量のもつとも多い冬の献立では3～4 mg のフッ素を水および食品中より摂取していることがわかつた。同じ献立をもちい，1965年の食品フッ素含有量にしたがつて計算すると，フッ素摂取量は11.13mgとなり，約3倍の増加をしめした。これは穀類および野菜類のフッ素量が近年増加の傾向にあるためであり，肥料ことにリン酸質肥料の原料であるリン鉱石中のフッ化物が，施肥量の増加とともにより多く農産物中に移行したためであろう。一方日本食品のフッ素量を医学地理的に検討した結果，日本人の主食としての水稲うるち玄米および代表的な副食品としての味噌のフッ素量が胃ガン死亡率と地理的な順相関をしめすことがわかつた。飲料水のみならず，食品のフッ素量を衛科学的に重要視することの必要性はいうまでもないが，ガンとフッ素との関係は未知の事柄であるから，ガンとウイルス，ウイルスとフッ素との相互関係を知ることのみによつても，フッ素の直接ないしは間接の栄養的役割があきらかになるのではなかろうか。