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Fluoride content of selected human food, pet food and related materials

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Summary

A survey was made of the fluoride content of selected human foods, some animal feeds, and related materials, with the purpose of a better characterization of products whose fluoride may contribute to the intake of fluoride by man as well as by some kinds of animals. Uptake of fluoride, especially from food with or without a foregoing food chain, was found to be more widely spread than has been documented previously. The major source of fluoride is represented by marine organisms, regardless of the ways by which man or animals are exposed to fluoride. As a consequence of the detailed analytical data given, recommendations would extend towards i) a more wide-spread analytical control of foods and feeds for fluoride, ii) an intensified assessment of fluoride bioavailabilities from foods and feeds, and iii) the consideration of such data in the evaluation of fluoride supplementations for optimized intakes, as recommended.

Zusammenfassung

In ausgewählten Lebensmitteln, Tierfuttermitteln und einer Reihe weiterer Materialien wurde die Fluoridkonzentration ermittelt, um solche Quellen besser charakterisieren zu können, aus denen direkt oder indirekt Mensch und Tier Fluorid aufnehmen. Fluorid ist, im wesentlichen durch die Nahrung mit oder ohne vorgeschaltete Nahrungskette, weiter verbreitet als bisher bekannt. Die Hauptquelle des Fluorids wird durch Meereslebewesen repräsentiert, unabhängig davon, auf welchen Wegen Mensch oder Tier Fluorid aufnehmen. Als Konsequenz der detaillierten Analysenwerte muß empfohlen werden, 1. Lebensmittel und Tierfutter umfangreicher auf ihren Fluoridgehalt zu kontrollieren; 2. verstärkt die Bioverfügbarkeit von Fluorid aus der Nahrung des Menschen und dem Tierfutter zu ermitteln und 3. die Ergebnisse solcher Studien bei der Festlegung der Auffülldosis von Fluorid entsprechend den Empfehlungen für eine optimale Zufuhr zur Kariesprophylaxe zu berücksichtigen.

Key words: fluoride, human food, health food, pet food

Introduction

The fluoride concentrations in human foods are in most cases very low (for review see (8)) with one major exception, when raw materials stem from the marine environment. Sea water throughout the world contains 1.3 mg F/l and thus provides a relatively fluoride-rich biotope for all marine life. Some organisms like the small antarctic krill shrimp, however, concentrate fluoride by several orders of magnitude (10). In addition, some organs of marine animals are well known to accumulate fluoride, especially bones and skin, scales included (16). Two major sources contribute to the fluoride content of animal feeds, both raw materials of marine origin like fish meal, and environmental contaminations on a very much localized scale, e.g. in the vicinity of alumina smelters (6, 7). Thus animals raised for human consumption may well contain higher fluoride levels, which has led to legal regulations (3). Furthermore, pet animals may receive, through their rations, more fluoride than anticipated. Since marine plants such as algae serve as raw materials for a great number of purposes (5), their indigenous fluoride content may be spread into a number of products used by man.

For the above reasons and as a preparatory step for studies of the bioavailability of dietary fluoride (15) a limited survey of the fluoride contents of human food, animal feeds, and some related products has been made, the results of which are presented in this paper.

Material and methods

The analyzed substrates and their origins are listed in Tables 2–5 in the results section.

All reagents were of the highest commercially available purity and supplied, unless stated otherwise, by Merck AG, Darmstadt, FRG.

In preparing the materials for analysis they were, if necessary, dried at 105 °C to constant weight. If required, the coarsely chopped substrate was extracted in a Soxhlet apparatus for 24 hrs with acetone for the removal of lipids. These steps were applied mainly to meat, fish products and some pet foods. The dry material was ground in an agate lined ball mill (MM2, Retsch & Co, Haan, FRG). The powders were kept in tightly closed plastic bottles at room temperature till they were analyzed. Analyses were carried out at least in duplicate with independent extracts of the analytical substrates.

Fluoride was extracted from 100–250 mg aliquots of the substances into 10 ml 1-M HCl and magnetically stirred vigorously for 20–30 min (1). The cleared extract was brought to 20 ml in a volumetric flask.

Some materials (Table 1) were ashed in a peroxide bomb (IKA-Universalbombe, IKA-Werk, Staufen/Br., FRG) with Na_2O_2 according to Wurzschmitt (17) by mixing about 100 mg substrate with 6–8 g Na_2O_2 and a few drops of ethyleneglycol. After reaction and cooling, the reaction mixture was dissolved in de-ionized water, neutralized with acetic acid, and brought to 200 ml in a volumetric flask with de-ionized water.

Fluoride was determined by means of gas chromatography (2) after derivation as follows: in special tubes for derivation (Serva GmbH, Heidelberg, FRG) 1 ml aliquots were mixed with 0.5 ml 25 % HCl (w/v) and 1 ml benzene reagent consisting of 760 µl trimethylchlorosilane and 5.6 µl isopentane (W. Günther, Griesheim/Hess., FRG) in 1000 ml benzene. The mixture was shaken vigorously for 30 min (IKA-Schüttler S50, IKA-Werk, Staufen/Br., FRG). After centrifugation for 5 min at 4000 rpm, 3 µl of the supernatant benzene layer was injected into the gas chromatograph (Hewlett-Packard, Frankfurt, FRG, 5840A with flame ionization detector; 2 m steel column of 3 mm external diameter; 20 % silicon oil DC 200/50 on Chromosorb P; N_2 stream at 20 ml/min; oven at 60 °C, injection block at 130 °C; detector at 250 °C).

All glassware was kept prior to use in concentrated sulphuric acid for 24 hrs, and after thorough rinsing used exclusively in these analyses.

Data of the fluoride contents of all substrates are given for the samples as purchased unless stated otherwise.

Results

In some instances, direct comparisons were made of the two digestion methods for fluoride prior to derivation and gas chromatography (table 1). Obviously, the digestion method of choice depended on the substrate to be analyzed: dental impression materials, but to a greater extent, siliceous earths and certain marine algae needed oxidative digestion whereas fish bone meal and mammalian bone meal gave essentially identical fluoride values with both methods.

It should be mentioned that Taves (13) did not find any differences in the fluoride contents of 95 food items analyzing ashed and unashed samples with the exception of two cereal products and black pepper. Wright and Davison (16), too, found a good coincidence comparing the fluoride contents of marine organisms obtained after acid extraction or potassium hydroxide fusion without ashing.

Table 2 presents data on fluoride in selected human food items. Elevated concentration of fluoride in meat products (normal range 0.1–0.3 mg/kg; (6)) most probably resulted from admixture of osseous material due to partial use of mechanically deboned meat (so-called separator meat). For this reason, some samples of bone meal were also analyzed (table 2), showing a fluoride content of 200–300 mg/kg and thus indicating a possible content of 0.4 % of finely ground osseous material in these meat products at the maximum.

Marine fish, as consumed by man, normally contains very low concentrations of fluoride (0.2–0.3 mg/kg fresh weight (6)). However, fish normally eaten with bones and skin like sardines in oil or smoked sprat, contained 16–20 mg F/kg. For the purpose of comparison, a number of fish bone samples were analyzed which gave fluoride concentrations between 580 and about 1000 mg/kg (table 2), thus easily explaining the fluoride values in, for example, whole sardines.

Table 1. Comparison of peroxide bomb ashing and HCl extraction for fluoride determination by gas-chromatography.

Substrate	Relative fluoride content (%) by bomb ashing $\bar{x} \pm$ S.D.	n	by HCl extraction $\bar{x} \pm$ S.D.	n
Dental impression materials (Zelgan, Palgate Quick, Tissutex)	100 ± 2.7	8	84 ± 4.8	6
Siliceous earth and Kieselgur (3 samples)	100 ± 4.7	8	65 ± 26.0	10
Lithothamnion algal powder (1 sample)	100 ± 5.4	6	4.5	2
Fish meal (3 samples)	100 ± 6.3	9	97 ± 6.2	6
Mammalian bone meal (3 samples)	100 ± 7.1	8	107 ± 8.6	6

n = number of independent analyses.

Table 2. Fluoride contents of human food.

Food item	Remarks/origin*		mg F/kg as purchased	mg F/kg dry weight	n
Meat and meat products					
Ground meat	pork and beef	purchased 7. 12. 81	0.32	2.33	2
Ground meat	pork and beef	purchased 8. 12. 81	0.22	1.33	2
Ground meat	pork and beef	purchased 29. 9. 82	0.29	1.53	3
Ground meat	beef		0.23	1.58	2
Mettwurst		purchased 29. 9. 82	0.21	1.45	3
Mettwurst		purchased 30. 9. 82	0.22	1.47	2
Gelbwurst			0.22	1.66	5
Big Mac	MacDonald		0.30	1.11	6
Delta-Würstchen with Lactoprotein and 6 % chicken			0.39	2.33	6
Stadtwurst			0.44	2.51	3
Hamburger	MacDonald		0.47	2.25	2
Fleischkäse			0.48	3.50	6
Baby-Salami			0.52	1.84	3
Fleischwurst			0.74	5.16	3
Mini-Salami (Bifi)			1.00	2.43	3
Unox-Würstchen in own skin, vacuum packed			1.03	7.51	4
Fish and fish meals					
Canned sardines	Portuguese product	without skin and bones	4.6	8.4	6
Canned sardines	Greek product	with skin and bones	15.8	52.6	6
Canned sardines	Italian product	with skin and bones	15.9	61.6	9
Canned sardines	Moroccan product	with skin and bones Jan. 80	18.3	57.5	6
Canned sardines	Moroccan product	with skin and bones Oct. 80	19.5	63.1	6
Smoked sprats			18.9	55.7	6
Fish meal			133	–	2

Table 2. Fluoride contents of human food (continuation).

Food item	Remarks/origin*		mg F/kg as purchased	mg F/kg dry weight	n
Fish meal	mackerel	Nordsee – Deutsche Hochseefischerei Bremerhaven, FRG	165	–	2
Fish meal	red perch + herring		190	–	2
Fish meal	red perch + herring		440	–	3
Fish bone meal	cod		580	–	2
Fish bone meal	red perch		800	–	2
Fish bone meal	saithe		1025	–	3
Mammalian bone meal					
Bone meal	pork and beef	local slaughter-house	207	–	2
Bone meal	pork and beef		293	–	2
Bone meal	extracted with acetone	H. Nagel, Hamburg, FRG	541	–	4
Bone meal	glue-removed		828	–	4
Whole body meal	about 50 % bones	local slaughter-house	308	–	3
Chicken bone meal	laboratory preparation		152	–	4
			mg F/litre		
Tea-infusion	1.5 g leaves/100 ml water, brewing time:		3 min	5 min	
Ceylon Assam			1.16	1.26	4
Maracuja			1.45	1.60	4
Crown of India			1.80	2.08	4
Ceylon broken Orange Pekoe			2.53	2.73	4
Indian broken Darjeeling			3.10	3.40	4

* All food items were purchased from local shops unless otherwise stated.
n = number of independent analyses.

Table 3. Fluoride contents of health foods and salt for human consumption and of dental impression materials.

Product	Origin	mg F/kg	n
Health food			
Bone Meal Tablets	Nutrition Headquarters Inc., Carbondale, USA	848	4
Rational Bone Meal Tablets	G. R. Lane Health Products Ltd., Gloucester, GB	684	2
Bone Meal Plus	Healthcrafts, Associated Health Food, purchased 20. 7. 82	522	3
Bone Meal Plus	Byfleet, Surrey, GB, purchased 20. 9. 82	589	3
Aminochel Bone Meal Tablets	Healthcrafts, Godalming, Surrey, GB	286	4
Super Kelp Tablets	Healthcrafts, Associated Health Food, Byfleet, Surrey, GB	338	4
Dolomite Tablets	Nutrition Headquarters Inc., Carbondale, USA	163	3
Dolomit Tabletten	Salus-Haus, Bruckmühl (Obb.), FRG	126	3
Kieselgur N	Macherey, Nagel & Co., Düren, FRG	283	2*
Flügge Kieselerde Tabletten	Flügge Diät, Stuttgart, FRG	116	3*
Flügge Kieselerde Pulver	Flügge Diät, Stuttgart, FRG	46.6	3
Flügge Kieselerde + Calcium Kapseln	Flügge Diät, Stuttgart, FRG	17.9	3*
Sanddorn Kalktabletten	Natura-Werk Hannover, FRG purchased 5. 10. 83	99	3
Sanddorn Kalktabletten	Natura-Werk Hannover, FRG purchased 8. 10. 83	102	3
Grano Vita Mineralstoffe	De-Vau-Ge Gesundkostwerk, Lüneburg, FRG	16.4	3
Calcium Sandoz Brausetabletten	Sandoz A.G., Nürnberg, FRG	6.0	3
Formula 3+6 Capsules	The Food Supplement Co., Godalming, Surrey, GB	4.0	3
Basica Mineralstoffpräparat	Klopfer Nährmittel GmbH., München, FRG	2.9	3
Aktiv-Kalk Pulver	Dr. Metz KG., Kelkheim, FRG purchased 4. 6. 82	2.0	3
Aktiv-Kalk Pulver	Dr. Metz KG., Kelkheim, FRG purchased 8. 10. 83	2.2	3
Weleda Aufbaukalk 1	Weleda A.G., Schwäbisch Gmünd, FRG	1.4	3
Weleda Aufbaukalk 2	Weleda A.G., Schwäbisch Gmünd, FRG	1.5	3
Merfluan Zahnsalz	Menadier GmbH., Hamburg, FRG	8,8	3

Table 3. Fluoride contents of health foods and salt for human consumption and of dental impression materials (continuation).

Product	Origin	mg F/kg	n
Table salt			
Bad Reichenhaller Markensalz	Saline Bad Reichenhall, FRG	1.1	3
Titro Salz	Nordmark Werke, Hamburg, FRG	0,95	3
Sea Salt			
Dr. Ritters Südsalz	Dr. Ritter GmbH., Köln, FRG	0.66	3
Schneekoppe Meersalz	Schneekoppe Reform GmbH., purchased 23. 1. 82	0.80	3
Schneekoppe Meersalz	Gelsenkirchen, FRG, purchased 12. 6. 82	1.80	3
Golden Harvest	Natural Sales Company, Pittsburgh, USA	0.95	3
Sea Salt	Co. des Salines du Midi, Montpellier, France	1.43	3
HMW-Meersalz	Hafen-Mühlen-Werke, Bremen, FRG	4.6	3
DANGA-Vollmeersalz	Meersalzsiederei U. Sabarth, Hage/Ostfriesland, FRG	6.8	3
Dental impression materials			
Zelgan	De Trey, Wiesbaden, FRG	14,550	2*
Tissutex	The Dental Manufacturing Co., Frankfurt, FRG	12,600	3*
Palgate Quick	ESPE, Seefeld (Obb.), FRG	11,500	3*

American products were purchased from the health food section in supermarkets in Maryland, Texas and New York except for bone meal tablets which were obtained through mail order. British products were purchased in three health food shops in London, the French product in Lille and the German products locally and in Berlin from health food shops.
n = number of independent analyses; * = analysed after peroxide bomb ashing.

The relatively high fluoride content in tea is well known (8, 12); the tea samples shown in table 2 were analyzed for investigation of the fluoride bioavailability from tea (15) and indicate that on average 3 min of brewing time released 91 % of the fluoride obtained after 5 min, a period which is said not to be exceeded normally.

Amongst health foods and preparations for self-medication numerous products are concerned with bone formation and calcium supply besides general roborant effects; a part of these is prepared from bones and petrous raw material. A short-list of such preparations was analyzed for fluoride (table 3) and revealed for several products revealed concentrations which were unexpectedly high. It should be mentioned, however, that no declaration of the high fluoride levels was given for any product, though intake of the recommended doses of products with > 100 mg F/kg would result in fluoride uptake in the order of 1–3 mg/day. On the other hand, quite a number of products are so low in fluoride that their intake would never affect total intake of fluoride per day by man.

A special group of preparations consists of sea salt which should, if representing total solids of sea water, contain fluoride at a level between 25 and 37 mg/kg. However, no sample of sea salt was found with more than 7 mg F/kg. This may be explained by the assumption that fractional crystallization of sea water preferentially yielded NaCl, leaving the majority of NaF in the brine.

Dental impression materials, based on alginate, contain a great deal of fluoride (Table 3). These analyses are the basis for bioavailability studies (15).

With the experience gained so far, some pet foods were checked for their fluoride content (table 4). The specimens were mainly selected if their label indicated the presence of osseous material or substances of marine origin like fish. The data of table 4 indeed indicate fluoride concentrations which might merit attention if really wholesome animal feeds are prepared. At the end of table 4 two examples are given as to how fluoride may reach the soil of private gardens when fertilizers containing substances of animal origin are used.

Since marine organisms, both of the animal and plant kingdoms, may play a role in marine food chains, plankton samples were analyzed as well as a number of weed-like large algae which represent raw material for industrial processing (table 5). One would expect that some representatives of the plankton world may well contribute to the fluoride accumulation in marine animals.

In addition, fluoride analyses of a krill meat sample used in rat feeding experiments (9) and in bioavailability studies (14) are given. Finally, seal teeth of two species were analyzed, one feeding mostly on krill, while the other is polyphagous. The difference found of about 25 % more fluoride in the teeth of the krill-eating seal seems to be smaller than expected, provided the polyphagous species does not also ingest a large amount of fluoride, e.g. from fish. Incidentally, a preliminary macroscopic inspection of the teeth did not reveal any symptoms like mottling or enamel cracks which, when observed in man, would indicate some overdosage of fluoride.

Table 4. Fluoride contents of animal food, pet food and fertilizers.

Food item	Origin	mg F/kg	n
Specified for cats			
Brekkies with meat and fish		51.1	3
Kitekat – with liver		34.6	3
Kitekat – with tuna		14.2	3
Kitekat – with offal	EFFEM GmbH, Verden, FRG	8.23	3
Whiskas – with lean meat		8.64	3
Whiskas – with meat and tuna		1.63	3
Katkins with 3 sorts of meat		2.23	3
Friskies – dried sea fish		80.7	2
A-la-Katz – 3 × Fisch		31.1	3
Friskies – with sea fish and mackerel	Friskies Tiernahrung GmbH, Hamburg, FRG	18.0	2
Katzen-Fit		4.20	2
Latzi-Katz-Fisch-Cocktail	Latz Purina GmbH, Euskirchen, FRG	29.6	3
Specified for dogs			
hap – Herzhafte Bissen		101	3
hap – Saft und Kraft		65.1	3
chappi + Rind (beef)	EFFEM GmbH, Verden, FRG	7.30	6
PAL + Rind (beef)		10.5	6
PAL + Leber (liver)		9.25	6
Latz-Biskuits	Latz Purina GmbH, Euskirchen, FRG	45.9	3
Latz – Hundekuchen		28.5	3
bingo – with 5 sorts of fresh meat	W. Hopermann GmbH, Oberhausen, FRG	2.58	6
Specified for birds			
Sittichkuchen		2.95	3
Ei-Biskuits		0.69	2
Pickstein (mineral supplement)	Vitakraft-Werke, Bremen, FRG	136	3
Sepia-Schale (cuttlefish bone)		172	3
Sepia-Schale (cuttlefish bone)		44.8	3
Sepia-Schale (cuttlefish bone)	Product of Thailand	79.9	3
Sepia-Schale (cuttlefish bone)	unknown	58.3	3
Garden fertilizer			
Knochenmehl – Blumendünger	Günther-Cornufera GmbH, Fürth, FRG	397	2
Oscorna – Blumendünger		299	2

All products were purchased from local shops.
n = number of independent analyses.

Table 5. Fluoride contents of some marine organisms.

Substance	Origin		mg F/kg dry weight	n
Seaweed Flour				
From brown algae	H. Carroux, Hamburg, FRG		18.8	2
From brown algae	H. Carroux, Hamburg, FRG	second sample	9.7	2
Lithothamnion powder	Biomaris, Bremen, FRG		1,097	2*
Algae				
Lessonia flavicans	Alginate Industries, Hamburg, FRG		85.5	2
Ecklinia maxima	Alginate Industries, Hamburg, FRG		35.8	2
Ascophyllum nodosum	Alginate Industries, Hamburg, FRG		25.5	2
Laminaria hyperborea	Alginate Industries, Hamburg, FRG		22.6	2
Macrocystis spec.	Alginate Industries, Hamburg, FRG		12.2	2
Kilkieran spec.	Alginate Industries, Hamburg, FRG		11.8	2
Lessonia nigrescens	Alginate Industries, Hamburg, FRG		9.3	2
Durvillea spec.	Alginate Industries, Hamburg, FRG		7.6	2
Laminaria hyperborea	Dr. K. Lünig, Biologische Anstalt Helgoland, FRG		25.7	2
Laminaria saccharina	Dr. K. Lünig, Biologische Anstalt Helgoland, FRG		23.1	2
Fucus serratus	Dr. K. Lünig, Biologische Anstalt Helgoland, FRG		14.7	2
Fucus vesiculosus	Dr. K. Lünig, Biologische Anstalt Helgoland, FRG		6.1	2
Fucus spiralis	Dr. K. Lünig, Biologische Anstalt Helgoland, FRG		5.9	2
Plankton (North Sea)**				
Biddulphia sinensis			24.1	2
Biddulphia sinensis	Dr. E. Hagmeier,	second sample	17.4	2
Thalassiosira nordenskioeldii	Biologische Anstalt		16.3	2
Coscinodiscus concinnus	Helgoland, FRG		6.2	2
Mixed		sample 1. 4. 82	17.6	2
Mixed		sample 5. 4. 82	12.3	2

Table 5. Fluoride contents of some marine organisms (continuation).

Substance	Origin	mg F/kg dry weight	n
Plankton (antarctic)** ***			
Sample M8101-35		28.2	2
Sample M8101-37		22.5	2
Sample M8101-40		15.2	2
Sample M8101-39	Prof. E. Suess,	13.4	2
Sample M8101-36	Corvallis, Oregon, USA	12.9	2
Sample M8101-21		7.5	2
Sample M8101-41		7.0	2
Sample M8101-43		5.1	2
Krill (Euphausia superba Dana)			
Krill meat	Bundesforschungsanstalt für Fischerei, Hamburg, FRG	970	3
Seal teeth			
Krabbenfresserrobbe		3,370	3
(Lobodon carcinophaga)	Prof. D. Adelung, Kiel, FRG		
Wedellrobbe		2,700	3
(Leptonychotes wedelli)			

n = Number of independent analyses.
* Fluoride extraction with peroxide bomb ashing.
** Small sample sizes allowed HCl extraction of F only.
*** Designations M8101 – refer to the antarctic sampling site determined in sea maps.

Discussion

In rather wide scatter, surprisingly high contents of fluoride were observed in human, health, and pet foods. Certain really extreme values occurring in nature, e.g. in sponges on coral reefs south of Tasmania (4) with >100,000 mg F/kg or in teeth of perciform fishes (11) with about 50,000 mg F/kg, were not observed in the present study.

Bone meals of mammalian or fish origin were analyzed rather thoroughly because they are held to be a suitable model of fluoride in meat or fish products, thus enabling by their high fluoride content an assessment of bioavailabilities. It should be stressed here that the analytical figures of this paper represent, if the source of fluoride is eaten or fed, intakes but not bioavailabilities; this latter point will be dealt with in a forthcoming paper (15).

The data presented in this paper certainly suggest that more attention be paid, in certain areas of human life, to the possible occurrence of fluoride in the environment of man. In view of the extremely well established protective role of fluoride against tooth decay (8), another consequence of these data would be a reappraisal of fluoride supplementation in caries prevention programmes as soon as bioavailabilities of important sources of dietary fluoride are known, with the purpose of obtaining optimal protection by optimized intakes.

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