

Reiner Ranau · Jörg Oehlenschläger · Hans Steinhart

Aluminium content in edible parts of seafood

Received: 26 July 2000 / Revised version: 4 October 2000

Abstract Samples of the edible parts of different fish species and of crustacean and molluscan shellfish were collected in the North Sea, the Barents Sea, the Baltic Sea, the Northeast Atlantic, and in Greenland waters. Most of the aluminium concentrations in fillets of lean and fatty fish were lower than 0.2 mg Al/kg wet weight. Exceptions were the aluminium concentrations in fillets of fish caught near an aluminium smelting plant (up to 1 mg Al/kg wet weight). Presumably a connection between the aluminium content in sea water and the fish fillets is responsible for this. The investigations of fillets of saithe, haddock, and cod with different lengths (age) showed that the aluminium levels decreased with increasing length, however, the decrease was not significant. An aluminium accumulation in muscle tissue with increasing age could not be detected. A comparison between fillets and different organs of cod showed higher aluminium concentrations in organs, especially in gills. The aluminium intake via gills, which are in continuous contact with the ambient water, is responsible for this. In the edible part of crustacean and molluscan shellfish higher aluminium concentrations (up to 5 mg Al/kg wet weight) were detected. The different feed spectrum and metabolism of these species seem to be responsible for

the higher aluminium accumulation in marine crustacean and molluscan shellfish.

Keywords Aluminium contents · Seafood · Fish fillets

Introduction

The high level of industrialisation and some of its negative consequences, such as acid rains, leads to an increase in the aluminium concentration in natural waters and biological systems. Aluminium has a low bioavailability, therefore it was until recently common opinion that aluminium does not present a hazard to health. However, the elevated concentration of dissolved aluminium-ions (Al^{3+}) in water may lead to uptake by plants and marine species and thereby entry of aluminium into the food chain.

Recent reports associated aluminium with several skeletal (osteomalacia: [1–3]) and neurological disorders (Alzheimer's disease: [8–13]) in humans. On account of the findings from dialysed uremic patients, who receive large amounts of orally administered aluminium-containing phosphate-binding gels or have been exposed to aluminium-contaminated dialysate for controlling serum phosphorus levels, it seems that aluminium is responsible for another neurological disorder: encephalopathy or dialysis dementia, [4–7]. Therefore, in uremic patients the total body aluminium concentration is greatly increased and brain aluminium levels have increased, similarly.

Therefore, the public interest for aluminium has been increased in the last decades. Seafood (fish, molluscan and crustacean shellfish) are regarded as being a healthy and tasty food for humans. The human consumption of seafood in Europe has increased in the past years. Therefore, more knowledge about the contents of aluminium in different marine species is necessary; particularly since the available literature does not

Reiner Ranau (✉) · J. Oehlenschläger
Federal Research Centre for Fisheries,
Institute of Biochemistry and Technology, Palmallee 9,
22767 Hamburg, Germany
e-mail: ranau.ibt@bfa-fisch.de
Tel.: +49-40-38905271
Fax: +49-40-38905262

H. Steinhart (✉)
Institute of Biochemistry and Food Chemistry,
University of Hamburg, Grindelallee 117,
20146 Hamburg, Germany
e-mail: steinhart@lc.chemie.uni-hamburg.de
Tel.: +49-40-428384356
Fax: +49-40-428384342

contain much information about aluminium levels in marine food. An analytical method for the determination of aluminium in fish tissues and muscle of molluscan and crustacean shellfish was developed using graphite furnace atomic absorption spectrometry (GFAAS), [14, 16, 18–21, 26].

Table 1a Instrument settings for aluminium determination by AAS

AAS-Instrument	Perkin Elmer AAS 4100ZL, THGA, AS 70
Al-hollow cathode lamp	current 25 mA
Wavelength	309.6 nm
Bandwidth	0.7 nm
Sample and standard volume	20 µl
Measurement area	0–60 µg Al/L
Matrix modifier	1. 5 µg Pd(NO ₃) ₂ 2. 3 µg Mg(NO ₃) ₂
Detection limit (3σ)	1 µg Al/kg
Characteristic mass	15 pg/0.0044 A s
Sensitivity	30 µg Al/L ≅ A s 0.180
Signal	Peak area
Background compensation	with inverse longitudinal Zeeman-effect

Materials and methods

Samples of different marine species were collected onboard the German fishery research vessel “Walter Herwig III” in different areas of the Northeast Atlantic, North Sea, Baltic Sea, Barents Sea, Greenland waters, and in the coastal waters of Norway near Stavanger. The edible parts of fish (muscle tissue), of crustaceans (abdominal muscle), and of cephalopods (tubes and tentacles)

Table 1b Furnace programme for aluminium determination by AAS

Temperature programme				
Step	T (°C)	ramp (s)	hold (s)	gas flow (mL/min)
1a. Dry	110	1	20	250
1b. Dry	130	5	30	250
2a. Ash	800	5	10	250
2b. Ash	1500	5	15	250
3a. cool down	1010	1	10	250
3b. cool down	20	1	10	250
4. Atomize	2300	0	5	0 (read)
5. Cleaning	2400	1	3	250
Purge gas		Argon		

Table 2 Aluminium contents in fillets of different fish species caught in open North Sea: in mg Al/kg wet weight, categorised in lean, fatty and flat fish

	n	Aluminium content (mg Al/kg wet weight)		
		mean ± SD	min.	max.
lean fish (fat content <1.5%)				
Cod (<i>Gadus morhua</i>)	10 ^[a] + 10 ^[b]	0.076 ± 0.038	0.033	0.192
Pollack (<i>Pollachius pollachius</i>)	5 ^[a] + 1 ^[b]	0.046 ± 0.024	0.021	0.089
Saithe (<i>Pollachius virens</i>)	5 ^[a] + 10 ^[b]	0.099 ± 0.040	0.032	0.155
Haddock (<i>Melanogrammus aeglefinus</i>)	5 ^[a] + 8 ^[b]	0.162 ± 0.099	0.034	0.349
Whiting (<i>Merlangius merlangus</i>)	2 ^[a]	0.069 ± 0.002	0.067	0.070
Ling (<i>Molva molva</i>)	8 ^[a] + 1 ^[b]	0.067 ± 0.044	0.037	0.180
Hake (<i>Merluccius merluccius</i>)	9 ^[a]	0.056 ± 0.023	0.025	0.088
Anglerfish (<i>Lophius piscatorius</i>)	5 ^[a] + 2 ^[b]	0.078 ± 0.037	0.032	0.131
John Dory (<i>Zeus faber</i> L.)	1 ^[a]	0.101		
fatty fish (fat content >1.5%)				
Spurdog, greyfish (<i>Squalus acanthias</i> L.) fillet	6 ^[a]	0.114 ± 0.054	0.055	0.189
Spurdog, greyfish (<i>Squalus acanthias</i> L.) rolled strips	3 ^[a]	0.166 ± 0.064	0.106	0.233
Mackerel (<i>Scomber scombrus</i> L.)	4 ^[a] + 6 ^[b]	0.074 ± 0.024	0.040	0.102
Scad (<i>Trachurus trachurus</i> L.)	10 ^[a]	0.102 ± 0.031	0.057	0.149
Herring (<i>Clupea harengus</i>) dry matter 20.7%	4 ^[b]	0.278 ± 0.017	0.265	0.301
Herring (<i>Clupea harengus</i>) dry matter 30.0%	5 ^[a] + 5 ^[b]	0.098 ± 0.026	0.064	0.148
Anchovy (<i>Engraulis encrasicolus</i> L.)	1 ^[b]	0.159		
Greater sandeel (<i>Hyperoplus lanceolatus</i>)	1 ^[b]	0.086		
Conger (<i>Conger conger</i>)	5 ^[a]	0.080 ± 0.016	0.064	0.105
(Striped) catfish (<i>Anarhichas lupus</i>)	4 ^[a]	0.052 ± 0.028	0.033	0.094
flat fish				
Plaice (<i>Pleuronectes platessa</i>)	15 ^[a] + 2 ^[b]	0.102 ± 0.044	0.048	0.176
Dab (<i>Limanda limanda</i>)	5 ^[b]	0.110 ± 0.046	0.061	0.176
Flounder (<i>Platichthys flesus</i>)	2 ^[a]	0.101 ± 0.018	0.088	0.113
Lemon sole (<i>Microstomus kitt</i>)	8 ^[a] + 1 ^[b]	0.064 ± 0.024	0.040	0.111
Sole (<i>Solea vulgaris</i>)	2 ^[a]	0.065 ± 0.012	0.056	0.073
Witch (<i>Glyptocephalus cynoglossus</i>)	1 ^[a]	0.108		
Turbot (<i>Psetta maxima</i>)	6 ^[a]	0.088 ± 0.057	0.042	0.177
Halibut (<i>Hippoglossus hippoglossus</i>)	1 ^[a]	0.095		

^[a]Singlesamples.

^[b]Pooled samples: n = 5–10 fishes of similar length (± 3 cm), all samples determined in duplicate.

were prepared from freshly slaughtered specimens, deep-frozen onboard and stored at -30°C until further treated in the laboratory on land. In the laboratory, the deep-frozen samples were freeze-dried without further treatment. The freeze-dried samples were finely ground in a ball mill and kept in high-density polyethylene bags at room temperature and at low humidity until analysed. Up to 1 g (weight depending on the aluminium concentration) of lyophilised sample portions was weighed into silica petri dishes. The petri dishes were put in the chamber of a plasma asher for mineralisation. The decomposition method used mineralised the samples in a microwave-activated oxygen plasma under vacuum without the addition of chemicals. After the complete mineralisation the remaining ash was dissolved with dilute nitric acid and transferred quantitatively into a polypropylene volumetric flask (volume depending on the aluminium concentration). For the measurement of the aluminium concentration, 20 μL aliquots of diluted ash solutions were injected into the graphite tubes of the electrothermal graphite furnace atomic absorption spectrometer.

Equipment

Plasma asher (Plasma Prozessor 200-G, Technics Plasma GmbH, München, Germany)

The mineralisation of the samples was performed in a closed, low-temperature microwave oxygen plasma processor system equipped with a high performance pump.

Atomic absorption spectrometry (Perkin Elmer Bodenseewerk, Überlingen, Germany)

A Perkin Elmer Model 4100ZL atomic absorption spectrometer with Zeeman-background correction equipped with a transversely heated graphite atomiser (THGA) was used for the aluminium determination. Pyrolytically coated graphite tubes fitted with a pyrolytic graphite platform were used. The instrument settings and furnace programmes for analysis of aluminium are described in Tables 1a and 1b.

The two drying steps and extended drying times ensured the complete drying prior to the ashing steps. Further the conditions of the time-temperature programme ensured a long lifetime of the graphite tube (about 800 firings) and the programme resulted in an optimal peak area signal. Samples and standards were diluted with two modifiers (Table 1a) using the AS-70 autosampler. The use of the modifiers and the two ashing steps ensured complete removal of interfering compounds and stabilised the aluminium ions (Pd-Al). The light source was a single element hollow cathode lamp, whose operating parameters (current and spectral bandwidth) were those recommended by the manufacturer. Argon was employed in the graphite furnace as external and internal gas, and the flow of the latter was interrupted during atomisation. For more details, see reference [14].

Reagents/Chemicals

Standard

Aluminium solutions in 0.2% (w/w) nitric acid containing 0 (blank), 5, 10, 20, 30, 40, and 60 μg Al/L were prepared from a Titrisol concentrate containing 1000 mg Al/L (Merck, Darmstadt,

Table 3 Aluminium contents in fillets of different fish species caught in open Northeast Atlantic: in mg Al/kg wet weight, categorised in lean, fatty and flat fish

	n	Aluminium content (mg Al/kg wet weight)		
		mean \pm SD	min.	max.
lean fish (fat content <1.5%)				
Pollack (<i>Pollachius pollachius</i>)	1 ^[a]	0.042		
Saithe (<i>Pollachius virens</i>)	5 ^[a]	0.058 \pm 0.024	0.026	0.093
Haddock (<i>Melanogrammus aeglefinus</i>)	1 ^[b]	0.132		
Whiting (<i>Merlangius merlangus</i>)	5 ^[a]	0.088 \pm 0.051	0.033	0.164
Blue whiting (<i>Micromesistius poutassou</i>)	5 ^[a]	0.058 \pm 0.008	0.051	0.071
Blue ling (<i>Molva dipterygia</i>)	8 ^[a]	0.051 \pm 0.011	0.041	0.076
Torsk, tusk (<i>Brosme brosme</i>)	5 ^[a]	0.100 \pm 0.023	0.084	0.140
Forkbeard (<i>Phycis blennoides</i>)	1 ^[a] + 1 ^[b]	0.055 \pm 0.005	0.051	0.058
Hake (<i>Merluccius merluccius</i>)	5 ^[a]	0.064 \pm 0.022	0.042	0.091
Roundhad rat tail (<i>Coryphaenoides rupestris</i>)	5 ^[a]	0.127 \pm 0.040	0.090	0.178
Anglerfish (<i>Lophius piscatorius</i>)	5 ^[a]	0.052 \pm 0.006	0.047	0.062
Greater argentine (<i>Argentina silus</i>)	5 ^[a]	0.091 \pm 0.016	0.075	0.114
Rat fish (<i>Chimaera monstrosa</i>)	1 ^[b]	0.083		
fatty fish (fat content >1.5%)				
Spurdog, greyfish (<i>Squalus acanthias</i> L.) fillet	3 ^[a]	0.087 \pm 0.028	0.055	0.105
Herring (<i>Clupea harengus</i>) dry matter 31.9%	3 ^[a]	0.132 \pm 0.038	0.098	0.173
Redfish, ocean perch (<i>Sebastes</i> spp.)	5 ^[a] + 2 ^[b]	0.096 \pm 0.022	0.065	0.125
Rockfish (<i>Helicolenus dactylopterus</i>)	1 ^[b]	0.062		
flat fish				
Plaice (<i>Pleuronectes platessa</i>)	4 ^[a]	0.069 \pm 0.019	0.045	0.089
Dab (<i>Limanda limanda</i>)	4 ^[a]	0.073 \pm 0.013	0.054	0.083
Megrim (<i>Lepidorhombus whiffiagonis</i>)	5 ^[a]	0.087 \pm 0.014	0.064	0.100
Lemon sole (<i>Microstomus kitt</i>)	4 ^[a]	0.054 \pm 0.015	0.035	0.067
Witch (<i>Glyptocephalus cynoglossus</i>)	4 ^[a]	0.085 \pm 0.046	0.039	0.131
Long rough dab (<i>Hippoglossoides platessoides</i>)	1 ^[a]	0.074		
Greenland halibut (<i>Reinhardtius hippoglossoides</i>)	5 ^[a] + 2 ^[b]	0.138 \pm 0.072	0.058	0.272
Halibut (<i>Hippoglossus hippoglossus</i>)	5 ^[a]	0.030 \pm 0.006	0.021	0.036

^[a]Single samples.

^[b]pooled samples: n = 5–10 fishes of similar length (± 3 cm), all samples determined in duplicate.

Germany). Nitric acid (65% w/w, Suprapur, Merck) of highest purity was used for the preparation of the standards and for the digestion, while for labware cleaning, analytical reagent grade nitric acid (65% w/w, pro analysi, Merck) was used. De-ionized water ($\geq 318 \text{ M}\Omega \text{ cm}^{-1}$ resistance) used for the preparation of all solutions was obtained from a NANOpure II water purification system (Sybron/Barnstead, Boston, Massachusetts, USA) and contained no detectable aluminium. The used matrix modifiers were: 1) palladium nitrate solution [$c(\text{Pd}) = 10.0 \pm 0.2 \text{ g/L Pd}(\text{NO}_3)_2$ in nitric acid (15% w/w), Merck] and 2) magnesium nitrate solution [$c(\text{Mg}) = 10.0 \text{ g/L}$ in nitric acid, Perkin Elmer] prepared by dissolving in de-ionized water (1:10) and stored in pre-cleaned polypropylene containers.

Cleaning

To avoid contamination from the containers, polypropylene vessels (flask, volumetric flask), high density polyethylene (HDPE) bottles, and further plastic equipment (beakers, autosampler cups, spoon, removable tips, tweezers) were used. They were cleaned by soaking in 2% (w/w) nitric acid (pro analysis grade) for >24 h followed by soaking in de-ionized water for >24 hours. After this cleaning procedure all cleaned vessels and equipment were dried and kept in aluminium-free containers until use. This avoided an aluminium contamination through room dust.

Table 4 Aluminium contents in fillets of different fish species caught in open Barents Sea, Greenland waters and Baltic Sea: in mg Al/kg wet weight, categorised in lean, fatty and flat fish

	n	Aluminium content (mg Al/kg wet weigh)		
		mean \pm SD	min.	max.
lean fish (fat content <1.5%)				
Barents Sea				
Cod (<i>Gadus morhua</i>)	5 ^[a]	0.061 \pm 0.010	0.048	0.075
Arctic cod (<i>Boreogadus saida</i>)	1 ^[a]	0.191		
Haddock (<i>Melanogrammus aeglefinus</i>)	1 ^[a] + 1 ^[b]	0.051 \pm 0.018	0.038	0.063
Blue whiting (<i>Micromesistius pouassou</i>)	1 ^[a] + 1 ^[b]	0.102 \pm 0.011	0.094	0.110
Greenland waters				
Blue ling (<i>Molva dipterygia</i>)	2 ^[a]	0.043 \pm 0.008	0.037	0.048
Torsk, tusk (<i>Brosme brosme</i>)	1 ^[a]	0.056		
Baltic Sea West of Bornholm				
Cod (<i>Gadus morhua</i>)	9 ^[a]	0.111 \pm 0.052	0.055	0.195
Whiting (<i>Merlangius merlangus</i>)	5 ^[a]	0.144 \pm 0.032	0.115	0.183
Baltic Sea East of Bornholm				
Cod (<i>Gadus morhua</i>)	4 ^[a]	0.295 \pm 0.044	0.236	0.330
fatty fish (fat content >1.5%)				
Barents Sea				
Capeline (<i>Mallotus villosus</i>)	2 ^[b]	0.272 \pm 0.055	0.233	0.311
Redfish, ocean perch (<i>Sebastes</i> spp.)	4 ^[a]	0.115 \pm 0.038	0.059	0.147
(Striped) catfish (<i>Anarhichas lupus</i>)	7 ^[a]	0.094 \pm 0.036	0.050	0.135
(Spotted) catfish (<i>Anarhichas minor</i>)	7 ^[a]	0.055 \pm 0.023	0.031	0.097
Greenland waters				
Redfish, ocean perch (<i>Sebastes</i> spp.)	13 ^[a] + 6 ^[b]	0.069 \pm 0.025	0.041	0.136
(Striped) catfish (<i>Anarhichas lupus</i>)	5 ^[a]	0.053 \pm 0.009	0.044	0.067
(Spotted) catfish (<i>Anarhichas minor</i>)	4 ^[a]	0.064 \pm 0.007	0.057	0.070
Baltic Sea West of Bornholm				
Mackerel (<i>Scomber scombrus</i> L.)	5 ^[a]	0.117 \pm 0.026	0.094	0.155
Herring (<i>Clupea harengus</i>)	5 ^[a] + 5 ^[b]	0.110 \pm 0.026	0.072	0.147
Anchovy (<i>Engraulis encrasicolus</i> L.)	1 ^[a]	0.182		
Sprat (<i>Sprattus sprattus</i>)	5 ^[b]	0.173 \pm 0.043	0.114	0.212
Baltic Sea East of Bornholm				
Sprat (<i>Sprattus sprattus</i>)	3 ^[b]	0.113 \pm 0.020	0.096	0.135
flat fish				
Barents Sea				
Long rough dab (<i>Hippoglossoides platessoides</i>)	3 ^[b]	0.076 \pm 0.024	0.049	0.095
Greenland waters				
Greenland halibut (<i>Reinhardtius hippoglossoides</i>)	4 ^[a]	0.118 \pm 0.024	0.092	0.150
Halibut (<i>Hippoglossus hippoglossus</i>)	6 ^[a]	0.062 \pm 0.020	0.037	0.083
Baltic Sea West of Bornholm				
Dab (<i>Limanda limanda</i>)	5 ^[a]	0.166 \pm 0.057	0.110	0.250
Flounder (<i>Platichthys flesus</i>)	4 ^[a]	0.158 \pm 0.069	0.088	0.251
Turbot (<i>Psetta maxima</i>)	4 ^[a]	0.170 \pm 0.025	0.142	0.191
Baltic Sea East of Bornholm				
Plaice (<i>Pleuronectes platessa</i>)	4 ^[a]	0.128 \pm 0.012	0.115	0.141

^[a]Single samples.

^[b]Pooled samples: n = 5–10 fishes of similar length ($\pm 3 \text{ cm}$); all samples determined in duplicate.

Results and discussion

The aluminium contents in fillets of different fish species, caught in the open North Sea, are presented in Table 2. The fish species were subdivided in three categories: lean, fatty and flat fish species. Each sample was analysed at least in duplicate. All values are given in mg Al/kg wet weight.

The aluminium levels in individual species varied within a considerable range. The majority of fish (lean, fatty, flat) species contained aluminium contents close to 0.1 mg Al/kg wet weight. The aluminium concentrations in fillets of different fish species caught in different locations (Northeast Atlantic, Barents Sea, Greenland waters, Baltic Sea) were of the same order (Tables 3 and 4).

All figures, given in Tables 2–4, are of comparable magnitude. The fat content or the differing trophic levels or the different mode of life (e.g., ground or pelagic fish) of fish species seem to have no effects on the aluminium levels. At first sight the catch locations also have no influence on the aluminium levels; however, some fish fillets as shown in Table 5 contained significantly more aluminium. These specimens were caught in the coastal waters of Stavanger/Norway near Skudeneshavn. The fish with the highest aluminium levels were caught approximately 40 km North of Skudeneshavn near an aluminium smelting plant.

Aluminium levels of fish caught near that aluminium smelting plant were significant higher; especially, the aluminium contents of haddock (*Melanogrammus aeglefinus*) was high (up to 0.9 mg Al/kg wet weight). Obviously a connection between the aluminium content in sea water and fish fillets is present. Possibly the different mode of life (pelagic or ground fish) and different feeding spectra of these fish species caught near that aluminium smelting plant seem to influence the aluminium contents, especially the aluminium contents of ground fishes such as cod (*Gadus morhua*), pollack (*Pollachius pollachius*), haddock (*Melanogrammus aeglefinus*), and ling (*Molva molva*).

The correlation between the aluminium contents and the length (age) of saithe (*Pollachius virens*), haddock (*Melanogrammus aeglefinus*), and cod (*Gadus morhua*) caught in the North Sea were investigated.

The highest aluminium levels were found in fillets of haddock, ranging from 0.14 (>55 cm) to 0.35 (27–30 cm) mg Al/kg wet weight. The aluminium levels of cod fillets ranged from 0.06 (80–89 cm) to 0.19 (40–49 cm) mg Al/kg wet weight and the lowest aluminium contents were found in fillets of saithe, ranging from 0.10 (69–72 cm) to 0.15 (37–40 cm) mg Al/kg wet weight. The aluminium contents of all fish fillets scattered around the calculated graph considerably. The aluminium contents generally decreased with increasing length and the slopes of the linear functions were negative. Therefore, it seems that there is no alumi-

Table 5 Aluminium contents in fillets of different fish species caught in coastal waters of Stavanger/Norway: in mg Al/kg wet weight, categorised in lean, fatty and flat fish

	n	Aluminium content (mg Al/kg wet weight)		
		Mean ± SD	min.	max.
near Skudeneshavn				
lean fish (fat content <1.5%)				
Cod (<i>Gadus morhua</i>)	2 ^[a]	0.131 ± 0.003	0.129	0.133
Pollack (<i>Pollachius pollachius</i>)	2 ^[a]	0.091 ± 0.013	0.082	0.100
Saithe (<i>Pollachius virens</i>)	2 ^[b]	0.082 ± 0.019	0.068	0.095
Haddock (<i>Melanogrammus aeglefinus</i>)	7 ^[a]	0.151 ± 0.050	0.070	0.226
Whiting (<i>Merlangius merlangus</i>)	5 ^[a]	0.134 ± 0.056	0.074	0.221
Ling (<i>Molva molva</i>)	5 ^[a]	0.099 ± 0.016	0.081	0.118
Torsk, tusk (<i>Brosme brosme</i>)	6 ^[a]	0.108 ± 0.019	0.081	0.128
fatty fish (fat content >1.5%)				
Mackerel (<i>Scomber scombrus</i> L.)	8 ^[a]	0.101 ± 0.062	0.035	0.200
(Striped) catfish (<i>Anarhichas lupus</i>)	1 ^[a]	0.225		
flat fish				
Dab (<i>Limanda limanda</i>)	2 ^[b]	0.128 ± 0.025	0.110	0.145
Akrehamn (app. 50 km north of Skudeneshavn)				
lean fish (fat content <1.5%)				
Cod (<i>Gadus morhua</i>)	3 ^[a]	0.295 ± 0.093	0.199	0.385
Pollack (<i>Pollachius pollachius</i>)	3 ^[a]	0.281 ± 0.143	0.195	0.446
Haddock (<i>Melanogrammus aeglefinus</i>)	4 ^[a] + 2 ^[b]	0.936 ± 0.196	0.664	1.232
Ling (<i>Molva molva</i>)	3 ^[a]	0.285 ± 0.017	0.271	0.304
fatty fish (fat content >1.5%)				
Mackerel (<i>Scomber scombrus</i> L.)	2 ^[a]	0.078 ± 0.029	0.057	0.098
Horneel (<i>Belone belone</i>)	2 ^[b]	0.161 ± 0.002	0.159	0.162

^[a]Single samples.

^[b]Pooled samples: n = 5–10 fishes of similar length (± 3 cm), all samples determined in duplicate.

nium accumulation in fillets with age. The linear function (aluminium content as a function of length) for cod had the greatest negative slope (-0.0233). The negative slope of the linear function of haddock was -0.0142 and the lowest negative slope (-0.0022) was found for the linear function for saithe. The trends (confidence area 95%) were significant; however, the significance's were on a low level.

Investigations of different organs of cod caught in the North Sea showed higher aluminium contents in organs (Table 6) compared to fillets. The highest aluminium levels were found in gills, which are in continuous contact with the ambient water. Meinelt et al. [24] reported similar results for fish (*Phoxinus phoxinus*) exposed to elevated aluminium concentrations. The aluminium levels were increased in liver, kidneys and especially in gills (up to a factor of 10).

While all fillets of lean and fatty fish investigated showed aluminium contents lower than 1 mg Al/kg wet weight, the amount of aluminium in the muscle of other marine species such as crustacean and molluscan shell-

fish was significantly higher. In Table 7 aluminium contents of different marine shellfish species caught in different locations (Barents Sea, Greenland Waters, North Sea, Northeast Atlantic) are reported.

It is known that in muscles of marine species like mussels or snails higher concentrations of metals (e.g., copper, zinc) are found than in fish fillets. Probably the other trophic level and metabolism of these species is responsible for the higher metal accumulation in marine shellfish species.

In Table 8 some aluminium levels of fish and other marine species reported in the literature are summarised.

The aluminium levels of Baltic herring, cod, mackerel and prawns reported by Jorhem and Haegglund [16] are in good agreement with the aluminium levels presented here. But the majority of the other reported aluminium levels were higher than the present levels. The information contained in the corresponding references was not detailed enough (information about the catch location was lacking or insufficient, results were based

Table 6 Aluminium contents in different organs of cod: in mg Al/kg wet weight

Organs	n	Aluminium content (mg Al/kg wet weight)		
		mean \pm SD	min.	max.
Liver	2 ^[a]	0.215 \pm 0.012	0.206	0.223
Gill	2 ^[b]	0.628 \pm 0.009	0.621	0.634
Heart	2 ^[b]	0.448 \pm 0.130	0.356	0.540
Spleen	1 ^[b]	0.290		
Kidney	1 ^[b]	0.355		
Brain	2 ^[b]	0.443 \pm 0.229	0.281	0.605
Gonad (female)	6 ^[a]	0.063 \pm 0.042	0.030	0.132

^[a]Single samples.

^[b]Pooled samples: n=5–10 fishes of similar length (± 3 cm), all samples determined in duplicate.

Table 7 Aluminium contents in the edible part of different marine shellfish species: in mg Al/kg wet weight

	n	Aluminium content (mg Al/kg wet weight)		
		mean \pm SD	min.	max.
Marine species				
Barents Sea				
Deep sea shrimps (<i>Pandalus borealis</i>) ^[c]	2 ^[b]	1.013 \pm 0.344	0.769	1.256
Octopus (<i>Eledone cirrosa</i> Lam.)	2 ^[a]	1.312 \pm 0.181	1.184	1.440
Greenland waters				
Deep sea shrimps (<i>Pandalus borealis</i>) ^[c]	3 ^[b]	1.021 \pm 0.061	0.960	1.082
North Sea				
Norway lobster (<i>Nephrops norvegicus</i>) ^[c]	11 ^[b]	2.410 \pm 0.757	1.538	3.730
Squid (<i>Loligo</i> spp.)	6 ^[a]	0.205 \pm 0.087	0.111	0.352
Snail (<i>Buccinum undatum</i>)	6 ^[a]	4.067 \pm 1.533	2.023	6.202
Mussel (<i>Mytilus edulis</i>)	2 ^[a]	4.950 \pm 1.636	3.793	6.107
Crab (<i>Cancer pagurus</i> L.) claw	4 ^[a]	3.392 \pm 1.456	2.162	5.434
Northeast Atlantic				
Deep sea shrimps (<i>Pandalus borealis</i>) ^[c]	2 ^[b]	1.253 \pm 0.441	0.941	1.565
Squid (<i>Loligo</i> spp.)	1 ^[a]	0.210		
Octopus (<i>Eledone cirrosa</i> Lam.)	1 ^[a]	0.247		

^[a]Single samples.

^[b]pooled samples: n=5–10 marine species.

^[c]Guts removed quantitatively, all samples determined in duplicate.

on only a few samples). Therefore a comparison between the results reported here and those presented in literature is difficult and the reliability of the results published earlier is not sufficient because of the few samples investigated.

Due to the progressive improvement of analytical techniques (mineralisation, GFAAS or ICP-MS) and

the laboratory equipment (clean-room with dust filter, Teflon or plastic equipment and Suprapur reagents) from year to year, generally, a tendency can be established that the recently reported aluminium contents are lower than the majority of the older reported aluminium contents.

Table 8 Aluminium contents of fish and other marine species in different references: in mg Al/kg wet weight, except Ref. [15] (values on dry weight basis)

Ref.	marine species	n	Aluminium content (mg Al/kg wet weight)		
			mean \pm SD	min.	max.
[15]	Codfish		6.99 \pm 0.66		
[15]	Flounder		3.53 \pm 0.78		
[15]	Lobster		7.80 \pm 0.64		
[15]	Oyster		606 \pm 9		
[15]	Sea scallops		23.0 \pm 1.9		
[15]	Shrimp		151 \pm 1		
[15]	Squid		3.95 \pm 0.37		
[16]	Baltic herring	1	0.085		
[16]	Cod	1	0.086		
[16]	Mackerel	1	0.027		
[16]	Prawns	1	1.3		
[17]	Perch (<i>Perca fluviatilis</i>)		<2		
[17]	Pikeperch (<i>Lucioperca lucioperca</i>)		<2		
[17]	Baltic herring fillets (<i>Clupea harengus membras</i>)		<2		
[17]	Baltic herring with bones (<i>Clupea harengus memb.</i>)		<2		
[17]	Sprat (<i>Sprattus sprattus</i>)		<2		
[17]	Salmon (<i>Salmo salar</i>)		4		
[17]	Rainbow trout (<i>Salmo gairdnerii</i>)		3		
[17]	Whitefish (<i>Coregonus</i> sp.)		<2		
[17]	Bream (<i>Abramis</i> sp.)		<2		
[17]	Pike (<i>Exos lucius</i>)		<2		
[17]	Eel (<i>Anguilla</i> sp.)		2		
[17]	Cod (<i>Gadus morhua</i>)		<2		
[17]	Flounder (<i>Platichthys flesus</i>)		<2		
[17]	Redfish frozen (<i>Sebastes</i> sp.)		2		
[17]	Shrimp canned	2	<2		
[17]	Mussels canned in water		60		
[18]	Cod	6	0.35 \pm 0.06	<0.30	0.44
[18]	Flounder	12	0.55 \pm 0.20	0.32	0.84
[19]	Sea shrimp	1	14.930		
[20]	Rosefish, redfish		1.2 \pm 0.5	0.7	1.9
[20]	Herring fillet (1991)		2.5 \pm 1.8	0.9	5.6
[20]	Herring fillet (1988)		4.9 \pm 1.7	2.1	7.6
[20]	Trout fresh		3.5 \pm 1.6	1.5	5.8
[20]	Mackerel fillet		4.2 \pm 1.6	2.4	7.3
[20]	Sardines		5.5 \pm 1.8	2.2	7.2
[21]	Herring (baked)	2		0.1	0.5
[21]	Salmon (cooked)	2		0.1	0.2
[21]	Trout (baked)	2		0.1	0.1
[21]	River eel (baked)	2		0.6	0.6
[21]	Sea eel (baked)	2		0.1	0.2
[21]	Cod (cooked)	2		0.1	0.5
[21]	Turbot (baked)	2		0.4	0.5
[21]	Plaice (baked)	2		<0.1	0.1
[21]	Sole (raw)	1	0.2		
[21]	Sole (baked)	3		0.1	0.4
[22]	Cod (cooked)		0.4		
[23]	Cod (frozen)	3	0.35	<0.30	0.44
[23]	Haddock (frozen)	1	<0.1		
[23]	Flounder (raw)	6	0.55	0.32	0.84
[23]	Sole (frozen)	1	<0.1		
[23]	Snail (raw)	1	98.9		
[23]	Crayfish	1	23.9		

Conclusion

The aluminium levels of all investigated fillets of lean and fatty fish species were lower than 1 mg Al/kg wet weight. Investigations of fillets of saithe, haddock and cod with different lengths (ages) showed that there was no aluminium accumulation in fillet with increasing age. A comparison between fillets and different organs of cod showed higher aluminium concentrations in organs, especially in gills. Probably the main aluminium intake over the gills, which are in continuous contact with the ambient water, plays a significant role in this connection.

In the edible part of crustacean and molluscan shellfish higher aluminium concentrations (up to 5 mg Al/kg wet weight) were detected. The different feed spectrum and metabolism of these species seems to be responsible for the higher aluminium accumulation in marine crustacean and molluscan shellfish.

The majority of aluminium levels reported earlier in literature were higher than the present results. But the results reported were rather old and generally the information about aluminium levels in fresh marine species was poor and not detailed enough. Therefore it is necessary that more investigations are performed in order to get results which are statistically sound.

A comparison with the provisional tolerable daily intake of 1 mg aluminium/kg body weight per day established by WHO in 1989 [25] indicated that the aluminium content of the edible part of aquatic food does not play a significant role in daily intake via food. Thus, a high consumption of fish is not a risk to health.

References

- Blumenthal NC, Posner AS (1984) *Calcif Tissue Int* 36:439–441
- Boyce BF, Elder HY, Elliot HL, Fogelman I, Fell GS, Junors BJ, Beastall G, Boyle IT (1982) *The Lancet* 2:1009–1013
- Bushinsky DA, Sprague SM, Hallegot P, Girod C, Chabala JM, Levi-Setti R (1995) *Journal of Bone and Mineral Research* 10:1988–1997
- Meiri H, Banin E, Roll M, Rousseau A (1992) *Progress in Neurobiology* 40:89–121
- Ganrot PO (1986) *Environmental Health Perspectives* 65:363–441
- Alfrey AC, Legendre GR, Kaehny WD (1976) *The New England Journal of Medicine* 294:184–188
- Alfrey AC (1997) *Dialysis Encephalopathy. Mineral and Metal Neurotoxicology*. In: Yasui M, Strong MJ, Ota K, Verity MA (eds) CRC Press Inc, pp 127–136
- Armstrong RA, Winsper SJ, Blair JA (1996) *Dementia* 7:1–9
- Johnstone T (1992) *Can Med Assoc J* 146:431–432
- Lamb BT (1995) *Nature Genetics* 9:4–6
- Martyn CN (1990) *Environmental Geochemistry and Health* 12:169–171
- Perl DP (1985) *Environmental Health Perspectives* 63:149–153
- Zapatero MD, Garcia de Jalon A, Pascual F, Calvo ML, Escanero J, Marro A (1995) *Biological Trace Element Research* 47:235–240
- Ranau R, Oehlenschläger J, Steinhart H (1999) *Fresenius J Anal Chem* 364:599–604
- Sun D-h, Waters JK, Mawhinney TP (1997) *J Agric Food Chem* 45:2115–2119
- Jorhem L, Haeggglund G (1992) *Z Lebensm Unters Forsch* 194:38–42
- Nuurtamo M, Varo P, Saari E, Koivistoinen P (1980) *Acta Agriculturae Scandinavica Suppl* 22:77–87
- Sullivan DM, Kehoe DF, Smith RL (1987) *J Assoc Off Anal Chem* 70:118–120
- XU GS, Jin RP, Zhang ZW, Zhang WQ, Ren DL, Chen J, Huang GW (1993) *Biomedical and Environmental Sciences* 6:319–325
- Müller M, Anke M, Illing-Günther H (1998) *Food Chem* 61:419–428
- Yang Q, Penninckx W, Smeyers-Verbeke J (1994) *J Agric Food Chem* 42:1948–1953
- Greger JL (1985) *Food Technology* 39:73–78
- Pennington JAT (1987) *Food Additives and Contaminants* 5:161–232
- Meinelt T, Stüber A, Krüger R, Steinberg C (1996) *Fischer & Teichwirt* 1/1996:5–8
- Becker K, Nöllke P, Hermann-Kunz E, Krause C, Schenker D, Schulz C (1990) *Umwelt-Survey 1990/91 Band III: Im Auftrag des Bundesministeriums für Umwelt, Naturschutz und Reaktorsicherheit*
- Delves HT, Suchak B, Fellows CS (1989) *Aluminium in Food and the Environment*. In: Massey R, Taylor D (eds) Royal Society of Chemistry, special publication No. 73, pp 52–67