

Rapid communication

# Aluminium levels of fish fillets baked and grilled in aluminium foil

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## Abstract

During storage and preparation of food, aluminium foil is often used for wrapping heat-sensitive raw food for protection against direct heat, e.g. grilled or baked fish fillets. In this investigation the aluminium contents of grilled and baked fish fillets with and without ingredients wrapped in aluminium foil were determined. The selected fish species were mackerel (*Scomber scombrus* L.), a fatty species, ocean perch (*Sebastes* spp.), a medium fatty species, and cod (*Gadus morhua*) and saithe (*Pollachius virens*), two lean fish species. The wrapped fillets were baked for approximately 20 min at 200°C in an oven, either without ingredients or, alternatively, with vinegar and sodium chloride added. In another experiment one part of a fillet was grilled over charcoal without ingredients and the other part was grilled with onion rings and mixed spices added. All aluminium concentrations of both baked and grilled fillets wrapped in aluminium foil increased during heating. The increase in aluminium concentration ranged from a factor of 2 (baked saithe fillets without ingredients from 0.10 up to 0.21 mg/kg) to a factor of 68 (grilled mackerel fillets with ingredients from 0.07 up to 5.04 mg/kg). The aluminium contents of grilled fillets were higher than those of baked fillets. Presumably two factors were responsible for the higher aluminium contents in grilled fillets: first the higher temperature of preparation when grilling fillets and second the high aluminium content of mixed spices (63.5 mg Al/kg), which may be taken up in part by the grilled fillets. All results clearly showed that some aluminium migrated from the aluminium foil into the food. The aluminium migration seems to depend on several factors, e.g. the duration and the temperature of heating, the composition and the pH-value of food, the presence of any other substances (such as organic acids and salt) and complexing reactions that result in dissolution of the complexed metal. Considering the present state of knowledge and the suggested provisional tolerable daily intake of 1 mg Al/kg bodyweight per day of the [World Health Organisation, 1989], no risk to health of the consumer would be expected from eating meals prepared in aluminium foil. © 2001 Elsevier Science Ltd. All rights reserved.

**Keywords:** Aluminium levels; Fish fillets; Grilling; Baking; Aluminium foil

## 1. Introduction

Aluminium is the third most abundant element in the lithosphere and the aluminium resources are considerable. During the last 100 years of industrialisation the importance of aluminium has strongly increased. Because of its favourable physical characteristics, aluminium has become indispensable in many industrial areas, e.g. the packaging industry, as well as in daily life. Aluminium has therefore been used extensively as a packaging material. This is not surprising considering its properties:

1. effective barrier against gases, moisture, flavours, grease, oil, light, non-degenerating;
2. tasteless and corrosion-resistant;
3. excellent thermal and electrical conductivity, no static charges;
4. good mechanical properties at low and elevated temperatures, no change as a function of time;
5. good formability at ambient temperature, deep drawing, stamping, dead-folding;
6. easy combination with other materials, e.g. paper and plastics;
7. decorative appearance and compatible with printing processes;
8. low specific weight (2.7 g/cm<sup>3</sup>); and
9. good recycling properties.

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Aluminium is now widely used as a household foil for packaging and storing different types of food products. One such use is for protecting food against direct heat effects, e.g. during grilling or baking of fish fillets.

Aluminium ions in the human diet are non-bioavailable from the small intestine, because the hydrated charged ions are not able to penetrate the lipid protein membranes of the duodenal mucosa. But it can be assumed that at least part of the human dietary aluminium intake is in the form of chelates with natural food components such as citric and lactic acids (Shuping, 1996). In the human body, aluminium ions could inhibit different metabolism processes caused by competition reactions between aluminium and other ions such as calcium, magnesium or iron (Macdonald & Martin, 1988). Therefore, in recent reports, aluminium has been associated with several skeletal (osteomalacia: Blumenthal & Posner, 1984; Boyce et al., 1982; Bushinsky, Sprague, Hallegot, Girod, Chabala, & Levi-Setti, 1995) and neurological disorders (Alzheimer's disease: Armstrong, Winsper, & Blair, 1996; Johnstone, 1992; Lamb, 1995; Martyn, 1990; Perl, 1985; Zapatero, Garcia de Jalon, Pascual, Calvo, Escanero, & Marro, 1995) in humans. Although the causality between aluminium uptake via food and these diseases is not established, public interest in effects of aluminium on human health has increased in recent years. On the other hand, it seems certain that aluminium is responsible for another neurological disorder: encephalopathy or dialysis dementia (Alfrey, 1997; Alfrey, Legendre, & Kaehny, 1976; Ganrot, 1986; Meiri, Banin, Roll, & Rousseau, 1992) in uremic patients on dialysis.

Many possible sources of aluminium intake by the human body exist, e.g. drugs, foods, drinking water, industrial exposure. Food is the main route by which the normal daily intake of aluminium occurs. The total aluminium content of foods comprises naturally present aluminium, aluminium from food additives and, to a great extent, aluminium from food containers such as cans, cookware, utensils and food wrappings. It has been established that cooking of acidic and low acidic foods in aluminium saucepans or foil causes leaching of the metal (Baxter, Burrell, Crews, & Massey, 1989; Brunner, Arnold, & Stolle, 1999; Greger, 1985; Greger, Goetz, & Sullivan, 1985; Lione, 1984; Liukkonen-Lilja & Piepponen, 1992; Moody, Southam, Buchan, & Farmer, 1990; Pennington, 1987; Rajwanshi et al., 1999; Takeda, Kawamura, & Yamada, 1998a, b, 1999; Tenakone, Wickramanayake, & Fernando, 1988; Treptow & Askar, 1987; Watanabe & Dawes, 1988). In some publications it was reported that foods, which were manufactured and stored in aluminium cans, showed an increased aluminium content (Aikoh & Nishio, 1996; Jagannatha Rao & Radhakrishnamurty, 1990; Rajwanshi, Singh, Gupta, & Dass, 1997; Šeruga, Grgić, & Mandić, 1994; Šeruga, Grgić, Grgić, & Šeruga, 1997; Severus, 1989).

The extent of the increase of aluminium is strongly dependent on factors such as temperature, pH-value, duration of contact or heating, presence of sugar, organic acids, salt and other ions.

Grilled and baked meals are widely consumed in Germany and are often prepared in aluminium trays or aluminium foil. Therefore, in relation to the above-mentioned possible association of excessive aluminium intake with some disorders, it is of great interest to measure the aluminium content of these dishes.

An analytical method for the determination of aluminium in grilled and baked fish fillets was developed using graphite furnace atomic absorption spectrometry (GFAAS; Delves, Suchak, & Fellows, 1989; Hewitt et al., 1990; Liukkonen-Lilja & Piepponen, 1992; Lopez, Cabrera, Lorenzo, & Lopez, 2000; Magalhaes, Lima, Krug, & Arruda, 1999; Müller, Anke, & Illing-Günther, 1998; Ranau, Oehlschläger, & Steinhart, 1999; Seruga et al., 1994, 1997; Sullivan, Kehoe, & Smith, 1987; Tahan, Sanchez, Granadillo, Cubillan, & Romero, 1995; Xiao-quan, Shen, & Zhe-ming, 1988; Yang, Pennickx, & Smeyers-Verbeke, 1994).

## 2. Materials and methods

Sampling of different fish species was performed on the German fishery research vessel "Walther Herwig III" in the North Sea. Fillets of mackerel (*Scorpaenidae* L.), ocean perch (*Sebastes* spp.), saithe (*Pollachius virens*), and cod (*Gadus morhua*) were deep-frozen onboard and stored at  $-30^{\circ}\text{C}$  until treated further in the laboratory on land. One part of the thawed fillet was wrapped in aluminium foil without ingredients and grilled over charcoal until cooked. Onion rings and mixed spices (approx. 1–2 g) were added to the other part of fillet which was then grilled under the same conditions. In a second series of experiments the fillets were baked in an oven for approximately 25 min at  $200^{\circ}\text{C}$  without ingredients and with the addition of vinegar/acetic acid (10%, 5 mL) and 1 g sodium chloride. The aluminium levels of all samples were determined using GFAAS. The grilled and baked samples were lyophilised directly after being deep-frozen in the aluminium foil. The freeze-dried samples were carefully unwrapped and the fish part was finely ground in a ball mill and kept in high-density polyethylene bags at room-temperature and at low humidity until analysed. Up to 500 mg 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 mineralised the samples in a microwave-activated oxygen plasma under vacuum at low temperatures ( $<300^{\circ}\text{C}$ ) without the addition of chemicals. After mineralisation was fully complete, the ash was dissolved

in diluted nitric acid and transferred quantitatively into a polypropylene volumetric flask (volume depending on the aluminium concentration). For the measurement of the aluminium concentration, 20  $\mu\text{L}$  aliquots of diluted ash-solutions were injected into the graphite tubes of the electrothermal graphite furnace atomic absorption spectrometer, with Zeeman-background correction, equipped with a transversely heated graphite atomiser. The instrument settings and furnace programmes for analysis of aluminium are described in Table 1.

The two drying steps and the extended drying times ensured 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 1) 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 (Ranau et al., 1999).

To avoid contamination, suprapure chemicals and polypropylene vessels (flask, volumetric flask), high density polyethylene bottles and other plastic equipment (beakers, autosampler cups, spoon, removable tips,

tweezers) were used for analysis. 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 h. After this cleaning procedure, all cleaned vessels and equipment were dried and kept in aluminium-free containers until use. This avoided aluminium contamination through room dust.

### 3. Results and discussion

In these investigations, the aluminium contents of cod, saithe, ocean perch, and mackerel fillets baked and grilled in aluminium foil were determined. Table 2 presents the aluminium contents of raw materials, of fillets baked and grilled in aluminium foil with or without ingredients. All values are given in mg Al/kg wet weight. Additionally, the proportional increases are given in Table 2.

The ingredients [baked fillets: vinegar (5 mL, 10%) and 1 g sodium chloride; grilled fillets: onion rings and approximately 1–2 g mixed spice] were added in order to get a lower pH-value (e.g. vinegar) and in order to simulate the normal human eating habits (onion rings and mixed spice). The increase of aluminium in all fillets after preparation (baked and grilled) was considerable (Table 2). Compared with raw materials, the increase in the aluminium contents in the baked fillets without ingredients ranged from factors of 2 (saithe) to 6 (mackerel). The increases in aluminium contents in the

Table 1  
Instrument settings and furnace programme for aluminium determination by atomic absorption spectrometry (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 $\mu\text{l}$			
Measurement area	0–60 $\mu\text{g Al/L}$			
Matrix modifier	5 $\mu\text{g Pd}(\text{NO}_3)_2$ 3 $\mu\text{g Mg}(\text{NO}_3)_2$			
Detection limit ( $3\sigma$ )	1 $\mu\text{g Al/kg}$			
Characteristic mass	15 $\text{pg}/0.0044 \text{ A s}$			
Sensitivity	30 $\mu\text{g Al/l} \cong \text{A s } 0.180$			
Signal	Peak area			
Background compensation	With inverse longitudinal Zeeman-effect			
<i>Temperature programme</i>				
Step	T ( $^{\circ}\text{C}$ )	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 of aluminium foil-grilled and -baked fillets with and without ingredients and of raw materials: in mg Al/kg wet weight

Fish species and ingredients	n <sup>a</sup>	Aluminium content (mg Al/kg wet weight)			
		Mean±S.D.	Min.	Max.	Increase <sup>b</sup> (%)
<i>Baked fillets (20 min at 200°C)</i>					
<i>Cod (Gadus morhua)</i>					
Baked without ingredients	4	0.295±0.074	0.217	0.395	388
Baked with vinegar (10%, 5mL) and 1 g NaCl	4	0.525±0.138	0.405	0.675	691
<i>Saithe (Pollachius virens)</i>					
Baked without ingredients	7	0.206–0.056	0.146	0.312	208
Baked with vinegar (10%, 5mL) and 1 g NaCl	8	0.569±0.151	0.403	0.863	575
<i>Ocean perch<sup>c</sup> (Sebastes spp.)</i>					
Baked without ingredients	4	0.265±0.045	0.218	0.326	276
Baked with vinegar (10%, 5mL) and 1 g NaCl	3	1.599±0.402	1.185	1.988	1666
<i>Mackerel (Scomber scombrus L.)</i>					
Baked without ingredients	7	0.434±0.167	0.224	0.642	586
Baked with vinegar (10%, 5mL) and 1 g NaCl	5	1.140±0.551	0.516	1.871	1541
<i>Grilled fillets (grilled over charcoal)</i>					
<i>Cod (Gadus morhua)</i>					
Grilled without ingredients	3	0.412±0.325	0.144	0.774	542
Grilled with onions rings and mixed spices	4	1.039±0.430	0.499	1.476	1367
<i>Ocean perch<sup>c</sup> (Sebastes spp.)</i>					
Grilled without ingredients	4	1.059±0.184	0.855	1.248	1103
Grilled with onions rings and mixed spices	3	1.089±0.025	1.063	1.113	1134
<i>Mackerel (Scomber scombrus L.)</i>					
Grilled without ingredients	3	0.564±0.203	0.335	0.722	762
Grilled with onions rings and mixed spices	2	5.043±0.428	4.741	5.346	6815
<i>Raw material (fish caught in the open North Sea)</i>					
<i>Cod (Gadus morhua)</i>					
	20	0.076±0.038	0.033	0.192	
<i>Saithe (Pollachius virens)</i>					
	15	0.099±0.040	0.032	0.155	
<i>Mackerel (Scomber scombrus L.)</i>					
	10	0.074±0.024	0.040	0.102	
<i>Ocean perch<sup>c</sup> (Sebastes spp.)</i>					
	7	0.096±0.022	0.065	0.125	
Onion rings	1	0.099			
Mixed spice	1	63.470			

<sup>a</sup> n determined in duplicate.

<sup>b</sup> Increase compared to raw materials.

<sup>c</sup> Caught in open Northeast Atlantic.

baked fillets with ingredients (lower pH-value) were higher and ranged from factors of 6 (saithe) to 17 (ocean perch). The aluminium contents of the baked fillets of lean fish (cod and saithe) were lower than those of fatty fish (ocean perch and mackerel). Obviously the fat content of fillets had an influence on the aluminium increase.

In all grilled fillets, higher aluminium levels than in raw fillets could be detected. The increase in aluminium levels in the grilled fillets, without ingredients, ranged between a factor of 5 (cod) and 11 (ocean perch) compared with the levels in raw materials. In the grilled fillets with ingredients added, these factors ranged from 11 (ocean perch) to 68 (mackerel). With the exception of grilled ocean perch fillets with ingredients, the aluminium contents of grilled fillets were higher than those of baked fillets. One possible reason for this could be that a part of the high aluminium content in the mixed spices (63.5 mg Al/kg) may be taken up by the grilled fillets.

Another possible reason could be that the temperature of preparation of the grilled fillets is substantially higher than the preparation temperature (200°C) of the baked fillets and that the higher temperature of preparation promoted a stronger aluminium migration from the foil into the fillets.

A comparison between the two different sides of aluminium foil (glossy and dull) showed that the differences in the aluminium contents in baked and grilled fillets, which were wrapped, respectively, with the glossy and the dull sides of the aluminium foil inside, were not significant.

It is established knowledge that most foods which are stored in aluminium cans, or have been cooked in aluminium pans, trays, or foil, accumulate some aluminium. As mentioned above, the amount of aluminium that is accumulated in foods during preparation depends on the pH-value and the time of the cooking. Recently, several investigators have suggested that the use of aluminium foil

can increase the amount of aluminium in foods or dishes (Brunner et al., 1999; Liukkonen-Lilja & Piepponen, 1992; Moody et al., 1990; Pennington, 1987; Rajwanshi et al., 1999; Severus, 1989; Takeda et al., 1998a, b, 1999); however, the significance of this increase in relation to total aluminium intake is questionable. Liukkonen-Lilja and Piepponen, Pennington and Severus reported low increases of aluminium in aluminium foil-wrapped and baked fish fillets (flounder, trout). Brunner et al. tested the migration of aluminium to halibut during baking (180°C, 25 min) in aluminium foils and in roasting tubes. To these halibut fillets 3% salt and 5% lemon juice (pH-value of homogenate 6.5–6.9) were added. Compared to the fillets of halibut prepared in roasting tubes, the aluminium contents of fillets of halibut prepared in aluminium foils were higher by a factor of 16 (from 0.6 up to 9.6 mg Al/kg wet weight). Takeda et al. (1998b) investigated the migration of aluminium from aluminium foil into food-simulating solvents. All parameters, the type of solvent (different pH-value), the temperature and the time, affected the magnitude of aluminium dissolution. The aluminium migration into acidic solvents when heated (30 min at 95°C), was higher than that into tap water and ranged from a factor of 60 for 0.5% aqueous citric acid to a factor of 200 for 4% aqueous acetic acid. Other investigators (Moody et al., 1990; Rajwanshi et al., 1999) have reported that the aluminium migration from aluminium foil into acidic (acetic, citric, oxalic, tartaric acid), fluoride-containing and heated solutions, resulted in high aluminium increase (factors of 20 to >100). Takeda et al. (1998a) have reported aluminium migration from aluminium foil into foods and the effect of food components on the migration. The migration levels in all heated (30 min at 95°C) and acidic foods (including orange and tomato juice, yoghurt and different types of pickles and vinegars) were less than that in 4% acetic acid. Dissolution of aluminium was enhanced by the addition of sodium chloride, but was reduced by the presence of proteins, amino acids, sugar or cholesterol. The decrease of aluminium dissolution from aluminium foil in 4% acetic acid (30 min at 95°C) was up to 10% in the presence of protein or amino acids. Because components of food, such as proteins, amino acids or sugar, decrease the aluminium migration from aluminium foil into foods, Takeda et al. (1999) suggested that migration was depressed by the addition of such foods. Therefore Takeda and co-workers concluded that human intake of aluminium from disposable aluminium foil is extremely low.

#### 4. Conclusion

All aluminium concentrations of fillets baked and grilled in aluminium foil clearly increased. The aluminium contents increased by factors ranging between 2

and 68. Generally, the aluminium contents of grilled fillets were higher than those of baked fillets. In the former case, the higher temperature of preparation when grilling fillets is likely to be responsible for this, as well as the very high aluminium content of mixed spice, which may be taken up in part by the grilled fillets. Considering all results, it can be concluded that, during the preparation of food wrapped in aluminium foil, some aluminium migrates from the foil into the food. Probably the aluminium migration depends on several factors:

1. the chemical composition of the raw food material and the other ingredients;
2. the conditions of preparation: duration and temperature of heating;
3. the pH-value of the food or dishes; and
4. the presence of any other substances (such as organic acids and salt), as complexing reactions result in dissolution of the complexed aluminium.

Regarding the present state of knowledge and the suggested provisional tolerable daily intake of 1 mg Al/kg bodyweight per day of the World Health Organisation (WHO, 1989; Becker, Nöllke, Hermann-Kunz, Krause, Schenker, & Schultz, 1990), there is no evident risk to the health of the consumer from eating meals prepared in aluminium foil.

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