Journal of Applied and Natural Science
14(2), 631 - 639 (2022)
ISSN : 0974-9411 (Print), 2231-5209 (Online)
journals.ansfoundation.org

Research Article

Heavy metals and aluminium intake from stored canned tomato, sardines and tuna in Algeria

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Article Info
https://doi.org/10.31018/jans.v14i2.3435
Received: April 8, 2022
Revised: June 4, 2022
Accepted: June 10, 2022

How to Cite

Terbeche, R. *et al.* (2022). Heavy metals and aluminium intake from stored canned tomato, sardines and tuna in Algeria. *Journal of Applied and Natural Science*, 14(2), 631 - 639. https://doi.org/10.31018/jans.v14i2.3435

Abstract
This study assessed heavy metals intake and their impacts on healthcare in Algeria. Peculiar attention was given to heavy metals found in largely consumed canned foods in Algeria such as double concentrated tomato, tuna crumbs and sardines. Chemical analyses of the metal and aluminium containers (foil, tray) were performed by X-Ray Fluorescence (XRF) and EDS spectrometry (Energy Dispersive Spectrometry). The determination of the trace metal content in canned food (element metal trace EMT) was achieved by Atomic Absorption Spectrometry (AAS). The approach proposed in this study aimed to highlight the interaction of the product and packaging material, and thus to identify and quantify heavy metals traces that were able to undergo specific or overall migration to food. The morphology of the contact surface food / packaging was observed by Scanning Electron Microscope (SEM) and showed a slight degradation of the base metal (Black Iron). There was an obvious increase in EMTs during tuna, sardines and tomatoes storage and artificial aging. A special interest was given to cooking modes using aluminium foil and trays. The respective dosages of aluminium, in baked food (fresh tuna) were obtained. Specific Aluminium migration was noticed and was strong for the flame cooking mode.

Keywords: Atomic Absorption Spectrometry (AAS), Canned food, contamination, element metal trace (EMT), Energy Dispersive Spectrometry (EDS), heavy metals, migration, Scanning Electron Microscope (SEM), X-Ray Fluorescence (XRF)

INTRODUCTION

As far as canned concentrated tomatoes consumption is concerned, Algeria is ranked at the 16th position producing 300.116 tons/per year and 9.599 kg per inhabitant. Canned fish is widely consumed in Algeria. Tuna represents 86.8%, whereas sardines represent 12.8% of the total. Canned foods are practical and handy to use. Foodstuff transformation and packing facilitates manipulation and storage (Robertson *et al.*, 2016). Since industrialization, heavy metals have contaminated land and air. They can get increasingly concentrated in human bodies through food chain. They might generate severe diseases such as fibromyalgia, Alzheimer disease, and autism (Langguth Cueva, 2021).

The increasing dependency of humans on canned foodstuff might bring undesirable heavy metals to the organism (Codex Alimentarius Commission, 1995 and Council of Europe, 2004). Human body intoxication by metals might come from elements like aluminium, arsenic, chrome, cobalt, copper, manganese, molybdenum, nickel, zinc and other toxic metals such as cadmium, mercury or lead (Picot 2013) .Human activities like chemical industries generate heavy metal pollution. Metals are particularly dangerous because they are not biodegradable. They can reach very high levels in human bodies through activities like fish cooking and consumption (Gu *et al.*, 2015 and Food and Agriculture Organization of the United Nations FAO, 1995) . Arsenic, cadmium, mercury and lead are ubiquitous elements and would inevitably contaminate nutrients. They can induce toxic effects through weak exposition *via* food dairy (Boisset, 2017). In fish, metals like Hg, Ni, Cu, Pb, Cr and Zn concentration levels are linked to their availability in the aqueous environment (Liu *et al.,* 2012). Canned tuna might not be a healthy food if contaminated with toxic elements to the nervous system such as mercury and arsenic (Hygie, 2021).

Metals are very convenient for packaging. They provide though physical and barrier protection properties. Metals are generally abundant in the earth's crust. Aluminium and white iron are cheap, malleable, transformable and recyclables. Moreover, they have attractive ergonomic and decorative potential for consumers (Gaurav *et al.,* 2020). These two metals are heavily used in packaging and are the main focus of the present study. The objective of this study was to examine the intake of metals in general and heavy metals in particular through the packaging and storage of foods intended for human consumption.

MATERIALS AND METHODS

In this study double concentrated tomato, tuna crumbs in tomato sauce and sardines in tomato sauce were analysed with their respective metal packaging. Can's aging effect and cooking impact were taken into account.

White iron characterisation

Three canned food were studied: double concentrated tomato, tuna crumbs and sardines. For each food two cans were studied. One fresh or recent and one aged. The capacity and the production year were crucial, decisive and mentioned. Although the cans manufacturing was Algerian but the origin of the metal remained unknown.

After rinsing by clean water, 2 small metal samples were cut from each can (20x30mm, 3g weight) and heat dried at 55°C for 19days. From each can, the first small metal sample was observed by the Scanning Electron Microscopy (SEM) (Azad *et al.,* 2018) in order to reveal the interior contact surface morphology. The second sample was stripped from its interior coating by a silicon carbide abrasive of different grades. Then it was mirror polished by a micrographic polisher. All these samples were subjected to X-Ray Fluorescence (XRF) (Temitope D.T.O, 2018) and Energy Dispersive Spectrometry (EDS) (Azad *et al*., 2018). The basic black steel content was characterised in the samples.

Aluminium paper and tray characterisation

Two different cooking times for tuna (frozen -18°C), after defrosting in the open air and seasoning (oil, lemon, salt), were carried out in the oven (60°C/2h) and over a flame (blue). However, two tuna samples were wrapped in Aluminium foil and two others were put in Aluminium tray with aluminium lid. After cooking and clean water rinsing, four samples (10x20mm) were obtained:

Aluminium foil: (1 oven cooked + 1flame cooked)

Aluminium tray: (1 oven cooked + 1flame cooked)

In each case, 2 witness samples (10x20mm) were analysed under the same conditions and were used as interpretation references.

The SEM Observation and EDS chemical analysis were carried out to study the morphological alteration of the contact surface Aluminium-food.

Nutrients analysis in white iron contact

The first nutrients group is conserved in white iron cans: Tuna crumbs and Sardine in tomato sauce, and Double concentrated tomato. Each food was conserved in two cans: One recent and one aged. All sample preparations and cooking modes were done according to the Technical Centre “AFAK CONTROL” guidance. This laboratory was created in Algeria in 1997 under the ISO 17025 standard.

Tuna crumbs in tomato sauce

Two cooking methods were available:

Method I: crude product canning followed by cooking inside the opened can

Method II: Product cooking, cooling, cleaning and canning, then Appertization (105°C)

In this study Method II was used. The tuna was cooked in brine and then cleaned. The skin, red and black muscles and the bones were removed in order to obtain tuna crumbs. As a remainder fresh defrosted tuna was used to prepare canned tuna. Under this section, fresh tuna means freshly canned tuna. One can was artificially aged (named can tuna, 55°C heat dry, 47 days)

Sardines in tomato sauce

Two samples were under consideration. Sample 1: canned sardines in tomato sauce, room temperature witness. Sample 2: canned sardines in tomato sauce, kept at 55°C for 19 days for artificial aging.

The following guidance was used:

Sample preparation (crush a trial)

A trial (5 to 10g) was put in a 250ml beaker; 50ml 1M HCL was added.

It was put in a bain-marie set up at 80°C, on a magnetic stirrer, for 1hour.Then it was left to decant for a few minutes. Using a 0.45µm porosity filter, it was filtrated in a 100ml class A flask. Finally, it was adjusted to 100ml with 1M HCL

Under the same conditions as above, a blank test was set up and the atomic absorption was carried out.

From the red absorbance and the curve absorbance/ quantities, the concentration of the element was deduced easily (Carapelle, 2013 and Farré *et al.,* 2008).

Double concentrated tomato

In this study, stability tests were carried out. The pH of witness samples and the pH of heat-dried samples were measured. Different temperatures and various periods were used, simulating accelerated artificial aging of the concentrated tomato. These tests were carried out on different concentrated tomato sauces.

Two samples were used:

Sample 1 (or can 1): concentrated tomato (Production date: 01/05/2016),140g , concentrated at 28%, pH= 4.05

Sample 2 (or can 2): Concentrated tomato (Production date: 2011; a real aging).400g , concentrated at 28%, pH=3.50

Nutrients analysis in aluminium contact

The second nutrient group used Aluminium foil (Al pap in figures) and trays for cooking. As mentioned above, following defrosting at room temperature and seasoning (oil, lemon, salt), tuna (frozen at -18°C) was cooked at 60°c for 2 hours in the oven and on a blue flame (1200°C).

The results of all the different assays mentioned above were obtained by Atomic Absorption Spectrometry (AAS) (Lars, 2019).

RESULTS AND DISCUSSION

The chemical composition of cans of white iron and the acceptable threshold values are indicated in Table1. Metal samples had higher rates of Mn, Ti, Si than the normal threshold. The other values were within an acceptable range of values. This is due to the cans manufacturer norms. Table 2 shows the chemical analysis of Aluminium foil and tray by EDS. Al values were followed in witnesses and in foil and trays used in cooking (oven, flame). These values remained more or less constant. It could be explained by the fact that the Al quantities transferred during cooking were infinitesimal. Tuna crumbs in tomato sauce elements metal traces are shown in Table 3.It is noteworthy that, for comparison purposes, only elements metal trace (EMT) were fully exploited from quantitation of the two products (fresh and canned tuna) (Table 1 and Fig 2). There was an obvious increase in Al, Fe; Pb, Cr, Ni, Sn during storage and artificial aging (55°C, 47 days). This is in accordance with de Lima *et al.* (2021), who revealed the accumulation of the majority of heavy metal elements. The overall percentage repartition of these EMTs did not vary a lot during aging. Suppose an error of 20% on the quantitation made in bad conditions with poor laboratory decontamination is taken into account. In that case, it should be deduced that fresh tuna relatively kept its residual contamination (Fig. 1) within a range of ±20% (Fig. 3).

EMTs results of Sardine in tomato sauce are shown in Table 4 and Fig. 4. There was an obvious slight increase in Al, Fe; Pb, Ni, Sn ,Mn ,Zn during storage and artificial aging (55°C, 19 days). However Cr decreased and Sn remained stable during aging. The overall percentage repartition of other EMTs (Mn, Zn, Fe) did not vary a lot during aging. Suppose an error of 20% on the quantitation made in bad conditions with poor laboratory decontamination is taken into account. In that case, it should be deduced that sardines relatively kept their residual contamination (Fig.5) within a range of ±20% (Fig.6).

Table 1. Chemical composition of cans white iron and the acceptable threshold values* (%).

	Mn	Co	Ti	Sn	P	AL	Zn	Cu	Fe
Tomatoe Can (2011)	3.00	0.60	0.63	0.86	0.05	0.62	0.26	0.10	98.25
Tomatoe Can (2016)	1.60	/	0.43	0.78	0.06	0.43	0.19	0.15	98.16
Recent Tunacan	2.10	/	0.58	0.42	0.01	0.54	/	0.06	98.47
Recent Sardinecan	2.17	/	0.46	0.542	0.02	0.59	/	0.16	98.35
Max/ treshold*	0.10		0.30	0.1	0.10	1		0.4	

Table 2. Chemical analysis of aluminium foil and tray by EDS

Aluminium foil	1) Witness							
	ChemicalElement	O	Al	Cu	Se	Mo	Sn	Total
	Mass %	0.52	97.21	0.39	1.81	0.07		100
	Atom %	0.89	98.3	0.17	0.63	0.02		100
	2) Oven							
	ChemicalElement	O	Na	Al	Cl	Mn	Sn	Total
	Mass %	1.99	0.05	97.95	0.01			100
	Atom %	3.3	0.06	96.36	0.01			100
	3) Flame							
	ChemicalElement	O	Na	Al	Cl	Mn	Sn	Total
	Mass %	0.6	0.01	99.34	0.02	0.03		100
	Atom %	1	0.01	98.95	0.02	0.01		100
Aluminium tray	1) Witness							
	ChemicalElement	O	Al	Ni	Cu	As	Se	Total
	Masse %	2.27	95.81	0.05	0.04	0.03	1.8	100
	Atome %	3.82	95.51	0.03	0.02	0.01	0.61	100
	2) Oven							
	ChemicalElement	O	Na	Al	Cl	Mn	Sn	Total
	Masse %	1.05	0.07	98.33	0.03	0.44	0.08	100
	Atome %	1.77	0.08	97.9	0.02	0.21	0.02	100
	3) Flame							
	ChemicalElement	O	Na	Al	Cl	Mn	Sn	Total
	Mass %	1.21	0.11	97.94	0.04	0.51	0.19	100
	Atom %	2.03	0.13	97.51	0.03	0.25	0.04	100

According to Swarnalatha *et al.* (2017), EMTs are naturally and anthropologically ubiquitous. The best way to avoid tinning removal is to use an interior varnish (Enrique *et al.,* 2015). Accelerated aging and acidity trigger an average of 15% increase in different EMTs (Fig. 6) . Moreover, the interior surface state morphology (tinning and varnish) shows a slight degradation, such as black points (holes in the varnish or the tinning) and so the basis metal disaggregation (the black iron). It was noticed that varnish coagulated in some places under the appearance of white spots due eventually to acidification (Fig. 7).

PHs of double concentrated tomatoes measured on 30 samples (concentrated at 28%) are given in Table 5. Stability tests show the correlation between the product quality from one side and the storage time and environmental conditions from the other side (temperature and moisture), thus allowing us to fix optimal storage time and conditions. They are carried out in thermostatically controlled ovens following special norms. It is noteworthy that a correlation between artificial and natural aging exists (one month of heat drying equals 18 months in real life) (Table 6).Therefore this artificial aging is an important parameter in our study. Tomatoes cans with optimal pH stability (pH around 4) were chosen. The present study noticed that element metal traces (EMT) are retained and revealed in the 2 food cans. The contents of the second older can (can N°2) are more affected. (Fig. 8, 9). It is noteworthy to mention that during sampling and after checking out the cans under-study, no rust or damage was present inside or outside. No swelling was visible and thus, the product was still stable. Storage material in contact with food plays a major role in nutrient conservation. Innovative and intelligent material is constantly improved (Gheid *et al.,* 2021). Normally, inside the can, a medium hermetically sealed and oxygen-free, nutrients can produce complex agents with tin. The electrochemical behaviour of the tin-iron battery is modified. At the reverse of what is predicted by the standards potential values of redox couples, tin acts, in general, as a sacrificial anode ensuring a cathodic protection of iron . However, this is sometimes associated with the potential migration of packing chemical products towards nutrients. Tin, glass, ceramics and plastics do liberate infinitesimal quantities of chemicals for food (Al Ghoul *et al.,* 2020). Even with tin scratches, iron remains protected from oxidation while tin oxidises. In addition, the interior protection of cans is strengthened by the presence of varnish; the most common is epoxy phenolic with a 5 µm thickness. Zinc, a major compound used to prevent iron acid corrosion, can migrate towards nutrients (Noureddine *et al.,* 2019 and Buculei *et al.,* 2012). However, in the case of carbonated acid nutrients, the Sn-Fe battery works in the usual sense and steel is rapidly attacked. The obtained results confirm EMT migration towards food. In the hypothesis that the original tomato and its concentrate were fine and not contaminated, we should conclude that contamination might originate from the cans' interior varnish. Damages may come from the paint quality or mishandling. After warm water cleaning, it was noticed that the interior varnish layer was damaged or completely taken off (can N°1, Fig 10 and can N°2, Fig. 11 and 12). In Fig. 12, holes in the varnish coat allowed direct contact with iron and food. Packages and food

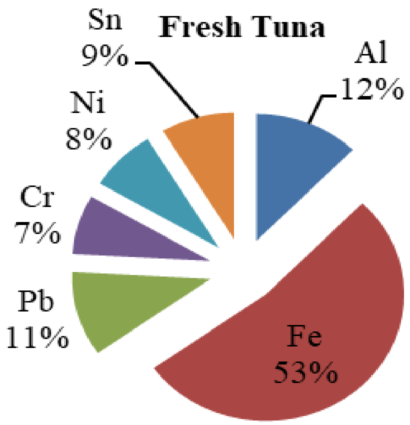

Fig. 1. *Comparison in % between fresh tuna (recent can) and can's tuna (aged can) based on EMT quantitation*

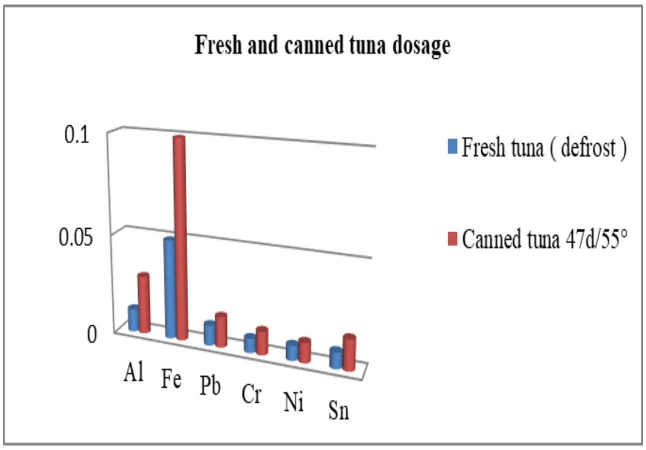

Fig. 2. *Comparison of EMTs in fresh and canned tuna*

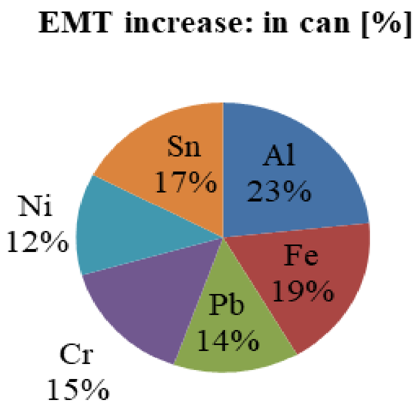

Fig. 3. *Comparison of EMTs in tuna*

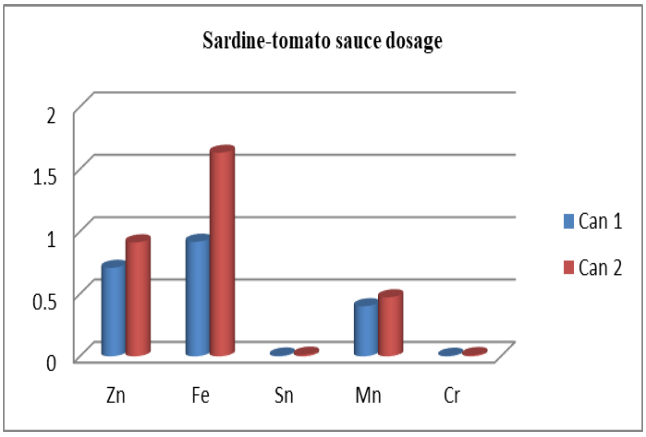

Fig. 4. *Sardines in tomato sauce and elements metal trace (can 1 I recent. can 2 is aged)*

Fig. 5. *Comparison in % between the two sardines cans based on EMT*

may transfer chemicals in both directions (Bassioni *et al.,* 2012). The best way to avoid or reduce cans detinning in contact with aggressive nutrients is to coat cans with an interior varnish. The thickness of the coating is highly linked to the cans' performances. The aggressive product conditioning, such as concentrated tomato sauce needs an 8-12 µm thickness in order to prevent the interaction between the can and its contents (De Bruce *et al.,*1986). From a toxicological point of view, bad manufacturing abilities and/ or a long and incorrect

Table 3. Tuna crumbs in tomato sauce and Elements Metal Traces EMT

Elements [mg.kg -1]	Fresh tuna	Can's tuna (47d/55°C)
Al	0.0115	0.0293
Fe	0.0495	0.0993
Pb	0.0101	0.0153
Cr	0.0071	0.0117
Ni	0.0074	0.0099
Sn	0.0081	0.0153

Table 4. EMTs results of Sardine in tomato sauce.

Eléments [mg.kg -1]	Can 1	Can 2
Al	0.0815	0.0999
Fe	0.1759	0.2845
Pb	0.0571	0.0874
Cr	0.0664	0.0191
Ni	0.0093	0.0131
Sn	0.0287	0.0801

Table 5. Average pH and stability of 30 samples.

Sample [mg.kg -1]	Witness pH	pH (37° C)	pH (55° C)
1	4.12	3.92	3.81
2	4.09	3.90	3.8
3	4.07	3.89	3.8
4	4.10	3.90	3.78
5	4.04	3.91	3.84
6	4.05	3.90	3.8
7	4.12	3.94	3.8
8	4.02	3.91	3.83
9	4.05	3.90	3.81
10	4.04	3.91	3.85
11	4.02	3.92	3.84
12	4.03	3.92	3.85
13	4.13	3.88	3.85
14	4.01	3.94	3.91
15	4.07	3.91	3.87
16	4.05	3.92	3.9
17	4.12	3.92	3.97
18	4.04	3.91	3.9
19	4.22	3.94	3.89
20	4.23	3.94	3.94
21	4.12	3.94	3.9
22	4.11	3.92	3.9
23	4.04	3.90	3.86
24	4.05	3.94	3.92
25	4.04	3.94	3.89
26	4.14	3.90	3.89
27	4.11	3.90	3.89
28	4.11	3.94	3.88
29	4.20	3.92	3.9
30	4.23	3.92	3.9
pH	4.09	3.92	3.87
	Room temperature	21 days	7 Days

storage time could lead to significant contamination of canned food through tin dissolution.

Nutrients in contact with aluminium paper and trays

EMTs total concentrations measured in tuna samples are shown in Table 7. The absolute input of Al in all kind of cooking (Tray/ foil; oven/ flame) is higher than 0.017. The product EMTs global contamination and leaching are linked to cooking modes. It is in accordance with previous work showing that leached Aluminium quantity is related to the cooking time and that the cooking method Flame (Paper or tray) is more contaminant (Dordevic *et al.,* 2019). The mean value (0.0098) of Al in fresh tuna (not cleaned + cleaned divided by 2) was calculated and was deducted from different calculations in order to obtain the absolute quantity in relation to the cooking mode. In fact, the mean value (0.0098) is the residual dose of Al element in the fresh tuna. This Al dose could be explained by storage and previous manipulations. Al Zubaidy *et al.* (2019) showed that a unique cooked meal wrapped in Aluminium foil could leach as much as 400mg of Aluminium. The leaching is proportional to the cooking temperature. Aluminium foil should be avoided in cooking while using tomato, lemon juice and spices. Outside, the aluminium is at high temperature (blue flame ~ 1200°C). It triggers partial aluminium evaporation (fusion at 660.3° C) and aluminium salts formation. Inside in contact with tuna at high temperature and acidity, aluminium is soluble and able to migrate to nutrients. Aluminium migration towards tuna during different modes of cooking is shown in Figs.13, 14 and 15.

Conclusion

While ferrous metals (White iron) and non-ferrous metals (Aluminium) are used as storage tools, EMT contamination could happen. Migration was related to contact time and quality. In general, the migration was higher in the case of a liquid nutrient and was positively linked to the wetting capacity of the nutrient (ex, fatty liquid). It could be accelerated by heat. The present

Table 6. Double concentrated tomato and Elements metals traces

Elements [mg.kg -1]	Can 1	Can 2
Al	0.0815	0.0999
Fe	0.1759	0.2845
Pb	0.0571	0.0874
Cr	0.0664	0.0191
Ni	0.0093	0.0131
Sn	0.0287	0.0801

Table 7. Aluminium quantitation for a different mode of cooking

1) EMT quantitation				2) Al quantitation			
EMT [mg.kg -1]	**FreshTuna**	**FreshTuna cleaned**	**Fresh Tuna (moy)**	**Tray. Oven**	**Trayflame**	**Foil. oven**	**Foil. Flame**
Al	0.0115	0.0081	0.0098	0.0268	0.0461	0.0391	0.0753
				- 0.0098	- 0.0098	- -0.0098	- 0.0098
		Al in absolute input :		0.017	0.0363	0.0293	0.0655
Fe	0.00495						
Pb	0.0101						
Cr	0.0071						
Ni	0.0074						
Sn	0.0081						

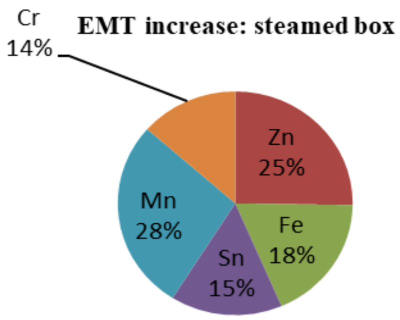

Fig. 6. *Results in % of EMTs in both cans*

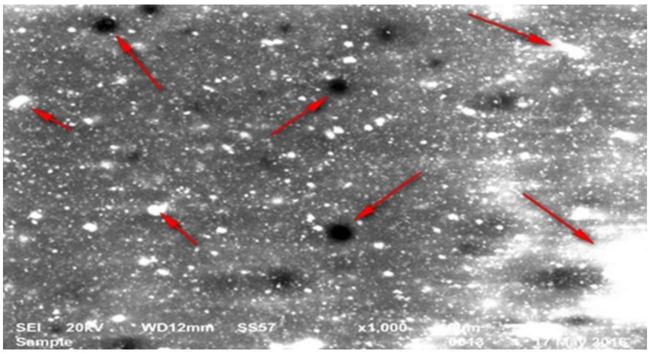

Fig. 7. *On the protective layer (varnish)*

Fig. 8. *Comparison in % of the 2 tomatoes cans on the basis of ETM dosage*

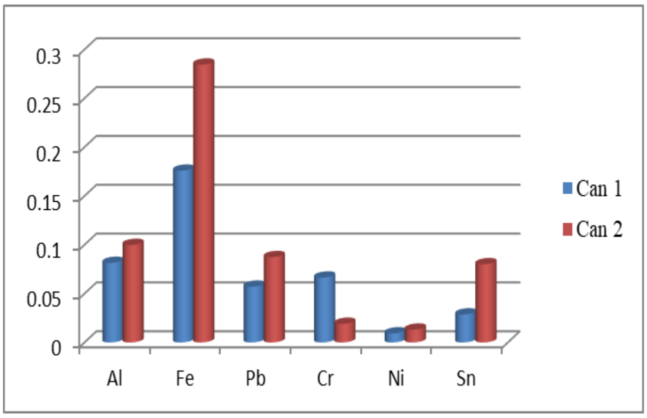

Fig. 9. *Comparison: quantisation of elements metal traces (EMT) in both double concentrated tomatoes cans*

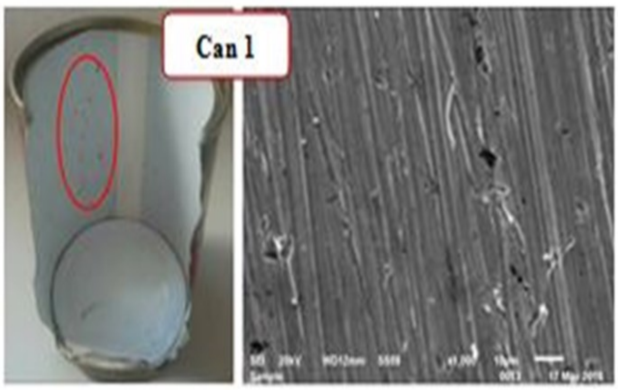

Fig. 10. *Rinsing the can with warm water. On the right the interior surface state protection with epoxyphenilic varnish, 5 µm thickness. Scratches were probably due to poor varnish application quality. Can 1 SEM observation*

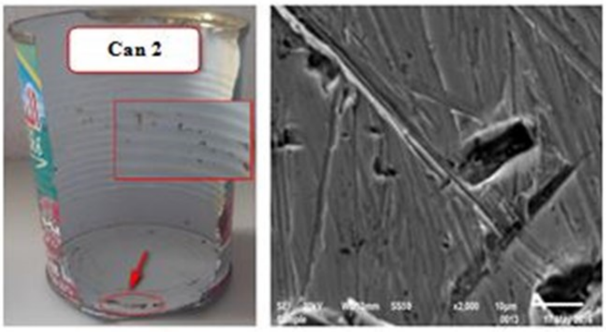

Fig. 11. *Rinsing the can with warm water (scratches). On the right the interior surface state protection with epoxyphenilic varnish, 5 µm thickness. Due to metal oxidation, the varnish layer is completely damaged. Can 2 SEM observation*

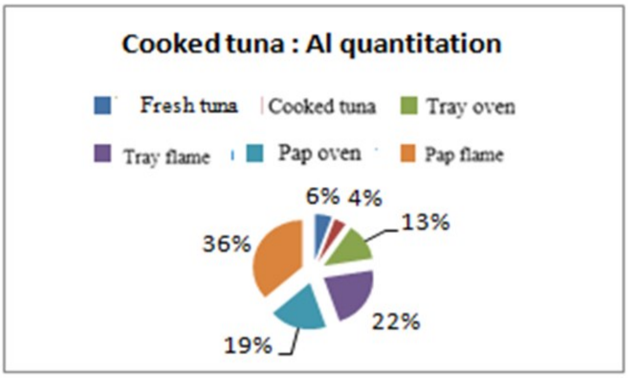

Fig. 13. *Al quantitation ; fresh tuna (previously frozen at -18°C) and cooking modes in aluminium foil (pap) and tray.*

study showed that Fe, Pb, Cr, Ni, Sn, Zn, Mn and particularly Al could reach high concentrations in stored and cooked food. Aluminium atoms effectively diffuse in the nutrients. A specific migration did happen and it was very strong for the flame cooking mode. Therefore, the aluminium foil should be restricted to cold food packaging. The application of polymer nano composites packaging materials in industrial, food and agricultural products is a superior alternative to traditional packaging materials such as glass, paper, and metals

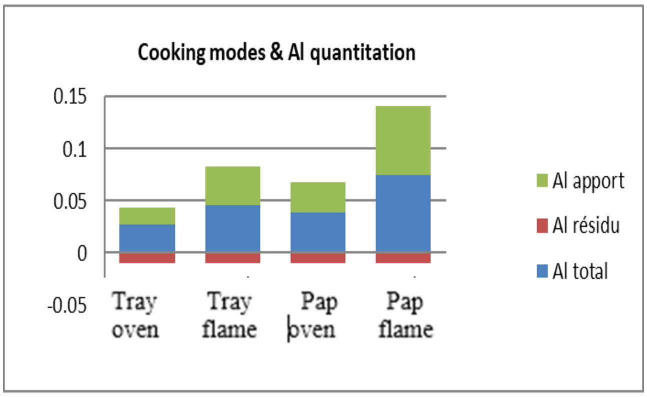

Fig. 15. *Quantitation Fresh tuna (previously frozen at -18) and absolute supply in Al versus cooking modes.*

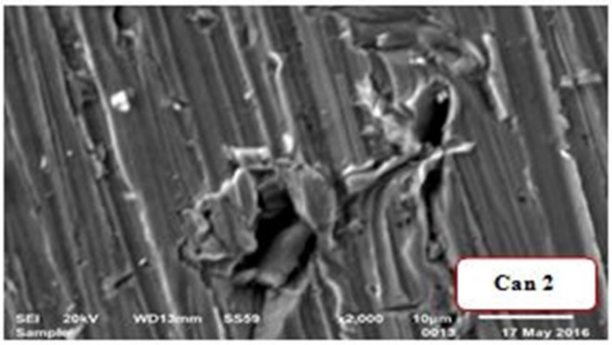

Fig. 12. *SEM observation of the varnish layer in can 2.*

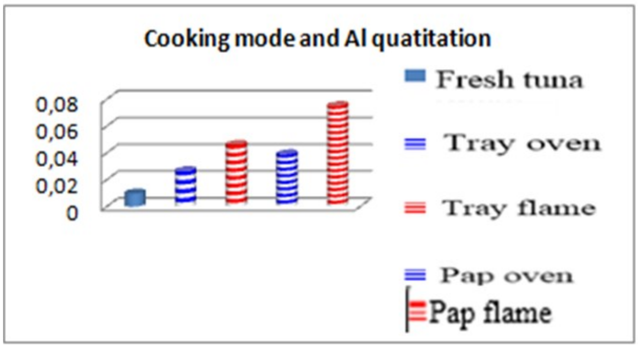

Fig. 14. *Al quantitation; Fresh tuna (previously frozen at -18°C) and relative supply in Al versus cooking modes).*

due to their functionalization, flexibility, and minimal cost for a specific food product, a careful choice of packaging material should be made by considering the end-product components and all their possible interactions as well as the resultant impact on food quality and safety. For any food-packaging selection, the benchmark is compliance with valid legislation and regulations, which may demand measurement of global and specific migration to assess the safety of the packaging material.

Conflict of interest

The authors declare that they have no conflict of interest.

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