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Heavy Metal Content of Canned Orange Juice as Determined by Direct Current Plasma Atomic Emission Spectrophotometry (DCPAES)

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ABSTRACT

Fifty canned orange juice samples representing five brands and three types of container were analyzed for heavy metals. The containers were tin cans, paperboard boxes and laminate pouches. Tin, lead, iron, zinc and copper were determined in the orange juice without prior preparation using plasma atomic emission spectrophotometry. The tin-canned orange juice was found to have higher heavy metal contents that juice kept in a paperboard box or laminate pouch. Except for one canned sample, all other samples showed heavy metal contents below the recommended limits of the FAO/WHO Codex Alimentarius Commission. The heavy metal content of all orange juice samples accorded with those limits adopted by the Egyptian Standards of Specifications (ESS).

INTRODUCTION

Metallic contamination of canned foods is an interesting subject for the food analyst. The significance of trace metal determination in foods has progressively increased as a consequence of rapid development of economy, industry and the expansion of urbanization projects, which contribute to environmental pollution. Extensive packaging of liquid foods in tin plate containers is due to the many advantages of this type of container; namely, ease of packaging, sterilizing, handling and transportation and minimization of the loss of vitamin potency in the foodstuff because of the anaerobic environment of the sealed can. Cans are made from plain carbon steel plates with a thin coating of tin (American Can Company, 1973).

During the preservation of acid food products, such as canned citrus juices, an interaction occurs between the components of the canned food and the material of the can. Corrosion in canned acid products is influenced by the chemical composition of the product, characteristics of the tin plate and the presence of corrosion accelerators such as sulphites, sulphur dioxide, nitrates or oxygen (Nagy *et al.*, 1980).

Extensive studies have been conducted on the heavy metal content of canned foods, especially fruit juices, and on effects of acidity, headspace, storage conditions (duration and temperature), can size, nitrates and sulphites on metal contamination, especially from lead, tin and iron (Nagy *et al.*, 1980; Nagy & Rouseff, 1981; Bache & Lisk, 1984; Khattab *et al.*, 1984; NoirFalise & Collinge, 1984; Seow *et al.*, 1984; Henriksen *et al.*, 1985).

Different analytical methods have been applied to determination of metals in canned foods. These include direct current plasma atomic emission spectroscopy (DCPAES), inductively coupled plasma atomic emission spectroscopy (ICPAES), flame atomic fluorescence spectrometry (FAFS), flame atomic absorption spectrometry (FAAS) and differential polarographic techniques (McHard *et al.*, 1979; Khattab *et al.*, 1984; Dabeka *et al.*, 1985).

In Egypt, until ten years ago, all processed fruit juices were packed in tin cans. Recently, food processors have also started using paperboard boxes and laminate pouches. This work was therefore conducted to determine the heavy metal contents of canned orange juice as affected by type of container, and to assess their compliance with the Egyptian and international recommendations.

MATERIALS AND METHODS

Materials

Fifty samples of Egyptian canned orange juice were collected from supermarkets in Egypt to represent all the existing brands and types of containers on the market (Table 1).

Methods

All the chemicals used in preparing standard solutions for analysis were of highest grade and obtained from BDH and Merck Chemical Companies.

TABLE 1
Orange Juice Samples by Brand and Container Type

Brand	*Type of container*		*Number of samples*
A	Tin can		10
B	Tin can		10
C	Paperboard box		10
D	Paperboard box		10
E	Laminate pouch (Plastic–aluminium–plastic)		10
		Total:	50

Tin (Sn), lead (Pb), iron (Fe), zinc (Zn) and copper (Cu) were directly determined in orange juices in a Beckman Spectraspan V Plasma Atomic Emission Spectrometer by a direct method adopted by Soliman (1986). Analytical parameters were adjusted as follows:

Nebuliser pressure	20 psi
Sleeve pressure	50 psi
Integration time	3×10 s
Entrance slit	100×500 u
Exit slit	50×500 u

The standard solutions used for the calibration were prepared from 1000 mg/litre stock solutions. Analysis of such elements at its corresponding line of choice (see Table 2) was done using standard start-up, peaking and autoranging.

Statistical analysis is affected as described by Snedecor & Cochran (1974).

TABLE 2
Wavelength used for Analysis of Test Elements

Element	*Wavelength* ($A°$)
Tin	3 034·1
Lead	2 883·0
Iron	2 559·4
Zinc	2 138·6
Copper	3 247·5

RESULTS AND DISCUSSION

Ingestion of food products containing excessive amounts of tin (400 ppm) has been responsible for several physiological disorders; namely, nausea, vomiting, diarrhoea, fever and headache (Nagy *et al.*, 1980). More recent studies have also indicated that excessive levels of tin in the diet can cause reduced retention of calcium, copper and zinc in rat tissues (Gregor & Johnson, 1981). Because of potential tin intoxication from ingestion of a canned food, several international studies have been conducted to determine the level of tin in canned citrus juices and to establish maximum levels for tin in these juices (Bielig *et al.*, 1978; McHard *et al.*, 1979). The World Food Codex (FAO /WHO Codex Alimentarius Commission, 1978) had established a tentative tolerance of 250 mg kg^{-1} for tin in canned orange juice. The Egyptian Standards of Specifications (ESS, 1976) of canned orange juice recommended the same maximum limit (250 mg kg^{-1}). Table 3 shows the tin content of orange juice samples as affected by type of container. All the samples examined were markedly below the maximum

TABLE 3
Tin Content of Orange Juice (mg kg^{-1}) as Affected by Type of Container

Sample No.	*Brand*				
	A Tin can	*B* Tin can	*C* Paperboard box	*D* Paperboard box	*E* Laminate pouch
1	139	22·7	0·456	0·234	0·410
2	78·1	29·1	0·538	0·070	0·424
3	78·4	23·7	0·550	0·325	0·424
4	87·1	23·8	0·327	0·368	0·424
5	63·6	32·1	0·374	0·424	0·439
6	58·3	45·8	0·644	0·424	0·467
7	60·1	71·5	0·491	0·424	0·382
8	59·3	34·1	0·503	0·439	0·453
9	38·2	29·6	0·409	0·368	0·467
10	96·9	25·6	0·187	0·453	0·467
Average	38·2–139	22·7–71·5	0·187–0·644	0·070–0·453	0·382–0·467
Mean	75·9	33·8	0·448	0·353	0·436

LSD at $P > 0{\cdot}05 = 20{\cdot}16$.
F calculated = 55·73**
F Table at $P > 0{\cdot}05 = 2{\cdot}58$
at $P > 0{\cdot}01 = 3{\cdot}78$

** Statistically highly significant.

limit set by the Codex Alimentarius Commission and the ESS. Highly significant differences were found between content of tin in canned orange juice (brands A and B) and in paperboard boxes or laminate pouches. It was also found that tin can samples (brand A) contained significantly higher tin contents than samples of brand B. The high tin content of orange juices packed in tin cans was due to the detinning of the cans during and after processing. The high temperature of the juice, coupled with the presence of a small amount of dissolved oxygen in the juice, causes an initial rapid detinning of the can. Oxygen dissolved within the juice, and entrapped in the headspace of the can, readily reacts with tin to form stannous ions. After dissolved oxygen and headspace oxygen are exhausted, hydrogen gas is produced in the acidic juice medium (Nagy *et al.*, 1980).

Since oxygen accelerates the corrosion of tin coating and is detrimental to many important nutrients (Veldhuis, 1971), it is no wonder that current industrial practice is to keep oxygen levels as low as possible during the processing of citrus juices or to use other types of containers such as paperboard boxes, laminate pouches or glass bottles.

Numerous investigations on the factors affecting corrosion of the tin can were carried out. Cruess (1948) reported that acidity of fruit products accelerates tin plate corrosion. Higher tin contents were found in juice packed in plain cans than in enamel-lined cans shortly after processing (Nagy *et al.*, 1980).

Nagy *et al.* (1980) pointed out that the uptake of tin by canned single strength orange juice was related to storage time and temperature. Storage at temperatures higher than 43·3°C greatly accelerated detinning of canned products. Khattab *et al.* (1984) analyzed a few samples of canned orange juice. The mean value of their samples was 275 mg kg^{-1} which exceeded the maximum recommended limit (250 mg kg^{-1}). In Malaysia, Seow *et al.* (1984) reported that samples of Malaysian canned orange juice contained 20–318 mg kg^{-1} of tin.

Lead is nutritionally a non-essential mineral that shows moderate toxicity. Its toxicity is due to its binding with active sites of important enzymes in cells and some ligands in the cell membrane (Stoewsand, 1980). The sources of lead in food are the natural lead content of food, environmental pollution and food processing activities involving the use of lead (Nagy & Rouseff, 1981). The lead content of canned orange juice samples is given in Table 4. The tin-canned orange juice samples contained higher lead contents than the samples packed in non-tin containers. Orange juice packed in paperboard box (Brand D) had the lowest lead concentration whereas the tin-canned orange juice (Brand B) showed the highest with differences being statistically significant, but insignificant within the tin-canned samples (Brands A and B). Only one tin-canned sample (Brand A)

TABLE 4
Lead Content of Orange Juice ($mg\,kg^{-1}$) as Affected by Type of Container

Sample No.	*Brand*				
	A *Tin can*	*B* *Tin can*	*C* *Paperboard box*	*D* *Paperboard box*	*E* *Laminate pouch*
1	0·118	0·169	0·122	0·054	0·097
2	0·059	0·205	0·128	0·048	0·113
3	0·093	0·207	0·121	0·031	0·114
4	0·143	0·187	0·132	0·031	0·149
5	0·139	0·197	0·130	0·030	0·133
6	0·138	0·227	0·133	0·011	0·114
7	0·195	0·233	0·142	0·014	0·130
8	0·181	0·144	0·161	0·042	0·100
9	0·145	0·154	0·136	0·055	0·112
10	0·322	0·157	0·133	0·040	0·122
Average	0·059–0·322	0·144–0·233	0·121–0·161	0·011–0·055	0·097–0·149
Mean	0·152	0·188	0·134	0·036	0·118

LSD at $P > 0·05 = 0·062$.
F calculated = 16·95**
F Table at $P > 0·05 = 2·58$
at $P > 0·01 = 3·78$

** Statistically highly significant.

exceeded the FAO/WHO recommended limit of lead in canned orange juice (0.3 $mg\,kg^{-1}$). All the orange juice samples had lead contents below the maximum limit adopted by the Egyptian Standards of Specifications.

The important source of lead in canned orange juice is the solder used to seal the side seam. Solders are constructed from 98% lead and 2% tin. Barbieri *et al.* (1980) reported that the solubilized lead came from the side seam and in particular (about 60%) from the seam laps. Aldini *et al.* (1980) found that 40% of the solubilized lead came from the seam laps. Rouseff & Ting (1980) concluded that lead variations in canned citrus juices were primarily due to can-to-can variation which, in turn, meant different amounts of solder surface exposed to juice. They also found that lead concentration was directly proportional to juice acidity. It may be concluded from Table 4 that the variations between the non-tin orange juice samples were due to the different varieties of orange used and/or differences in the processing activities. Moreover, the difference between the two brands of tin-canned orange juice (Brands A and B), may be due to difference in solder splashings and solder seepage through the side seam. Bielig *et al.* (1978) studied some factors affecting lead uptake in canned orange and tomato juices. They found that headspace, nitrates, longer storage time, high temperature and use of lacquered cans increased the lead uptake. Capar (1979) found that canned fruit juice contained at least 60% more lead than glass-packed products. Thus it may be advisable to follow good processing practices and to use other types of containers such as paperboard boxes, laminate pouches or glass bottles.

Table 5 shows the iron content of orange juice samples examined. As expected, the orange juices packed in tin cans (Brands A and B) contained significantly higher iron contents than those packed in paperboard or laminate pouch. Brand B (tin can) had the highest iron content whereas Brand D (paperboard box) had the lowest. No significant differences between the iron content of orange juice packed in paperboard boxes or laminate pouches (Brands C, D and E) were found. All the orange juice samples had iron contents below the FAO/WHO recommended level of iron

TABLE 5
Iron Content of Orange Juice ($mg\,kg^{-1}$) as Affected by Type of Container

Sample No.	*Brand*				
	A *Tin can*	*B* *Tin can*	*C* *Paperboard box*	*D* *Paperboard box*	*E* *Laminate pouch*
1	5·26	1·74	0·118	0·032	0·388
2	4·09	1·96	0·133	0·040	0·371
3	1·74	2·43	0·128	0·030	0·215
4	1·28	2·36	0·125	0·032	0·215
5	1·72	6·35	0·123	0·032	0·215
6	2·47	2·35	0·169	0·032	0·205
7	3·21	5·71	0·141	0·061	0·246
8	1·52	2·77	0·137	0·048	0·216
9	0·99	1·84	0·303	0·119	0·275
10	3·11	1·50	0·160	0·125	0·272
Average	0·99–5·26	1·5–5·71	0·118–0·303	0·03–0·125	0·205–0·388
Mean	2·54	2·90	0·154	0·055	0·262

LSD at $P > 0{\cdot}05 = 1{\cdot}4$.
F calculated $= 20{\cdot}87$**
F Table at $P > 0{\cdot}05 = 2{\cdot}58$
at $P > 0{\cdot}01 = 3{\cdot}78$

** Statistically highly significant.

in canned orange juice (15 mg kg^{-1}). No suggested limit of iron concentration in canned orange juice was found in the Egyptian Standards of Specifications. The iron content of fourteen samples of canned orange juice ranged from 0·8 to 2·1 mg kg^{-1} (Nagy *et al.*, 1980), whereas six Malaysian tin-canned orange juices contained about 14·6 mg kg^{-1} as a mean value (Soew *et al.*, 1984). Nagi *et al.* (1980) found that the iron values of freshly canned orange juices were similar to those of non-canned orange juice, suggesting that iron dissolution of the base plate was minimal during canning. However, the high temperature and length of storage after processing relatively increased the uptake of iron by canned orange juice. Szarski (1971) and Britton (1975) referred the high levels of iron and tin in canned foods to technical faults and mainly to slow corrosion occurring over prolonged periods of time.

Table 6 shows that tin-canned orange juice (Brand A) had the highest zinc concentration and that packed in laminate pouch (Brand E) had the lowest. A highly significant difference was found between Brand A and other brands

TABLE 6
Zinc Content of Orange Juice (mg kg^{-1}) as Affected by Type of Container

Sample No.	*Brand*				
	A Tin can	*B* Tin can	*C* Paperboard box	*D* Paperboard box	*E* Laminate pouch
1	0·729	0·439	0·065	0·208	0·054
2	2·40	0·543	0·087	0·214	0·060
3	1·81	0·362	0·296	0·214	0·071
4	1·12	0·428	0·043	0·214	0·082
5	1·30	0·763	0·060	0·214	0·082
6	0·983	0·741	0·060	0·214	0·098
7	2·24	0·769	0·054	0·203	0·098
8	2·15	0·043	0·060	0·208	0·104
9	2·02	0·033	0·071	0·159	0·060
10	2·19	0·043	0·061	0·164	0·049
Average	0·729–2·40	0·033–0·769	0·043–0·296	0·159–0·214	0·049–0·104
Mean	1·69	0·416	0·086	0·201	0·076

LSD at $P > 0·05 = 0·433$.
F calculated = 50·85**
F Table at $P > 0·05 = 2·58$
at $P > 0·01 = 3·78$

** Statistically highly significant.

of canned orange juice. No significant difference existed between samples packed in paperboard boxes or laminate pouches or Brand B tin-canned orange juice. All the orange juice samples had zinc contents lower than the limit set by the Codex Alimentarius Commission (5 mg kg^{-1}). No maximum limit of zinc content of canned orange juice was set by the ESS. To study the effect of storage on the zinc content of canned foods, Branca (1982) analyzed 345 commercially canned food samples and found that zinc content was increased by 32% and 57% after 280 and 365 days of storage, respectively.

Table 7 shows that the two Brands, A and B, of tin-canned orange juice had significantly higher copper contents than those samples packed in other types of containers (paperboard box and laminate pouch). The mean values for zinc of the non-tin orange juice were very close.

All the orange juice samples had copper contents below the limit set by the Egyptian Standards of Specifications (5 mg kg^{-1}). Sanchez *et al.* (1981) found that 190 samples of canned food contained less than 2 mg kg^{-1} copper, which were attributed to the initial concentration of copper in the

TABLE 7

Copper Content of Orange Juice (mg kg^{-1}) as Affected by Type of Container

Sample No.	*Brand*				
	A *Tin can*	*B* *Tin can*	*C* *Paperboard box*	*D* *Paperboard box*	*E* *Laminate pouch*
1	0·093	0·013	0·027	0·030	0·017
2	0·074	0·090	0·033	0·031	0·022
3	0·075	0·131	0·019	0·032	0·031
4	0·067	0·128	0·043	0·031	0·030
5	0·079	0·100	0·027	0·032	0·030
6	0·069	0·130	0·031	0·032	0·017
7	0·114	0·143	0·025	0·032	0·022
8	0·115	0·008	0·020	0·032	0·020
9	0·188	0·007	0·036	0·011	0·046
10	0·093	0·007	0·035	0·020	0·053
Average	0·067–0·188	0·007–0·143	0·019–0·043	0·011–0·032	0·017–0·053
Mean	0·097	0·076	0·030	0·028	0·029

LSD at $P > 0{\cdot}05 = 0{\cdot}046$.
F calculated = 10·04**
F Table at $P > 0{\cdot}05 = 2{\cdot}58$
at $P > 0{\cdot}01 = 3{\cdot}78$

** Statistically highly significant.

TABLE 8
Significance of Simple Correlation Coefficients (r) Among the Heavy Metal Contents of Canned Orange Juice

Heavy metal	*Copper*	*Zinc*	*Iron*	*Lead*	*Tin*
Tin	0·4**	0·68**	0·77**	0·44**	
Lead	0·45**	0·35**	0·54**		
Iron	0·41**	0·48**			
Zinc	0·39**				
Copper					

r Table at $P > 0{\cdot}05 = 0{\cdot}27$
at $P > 0{\cdot}01 = 0{\cdot}35$

** Statistically highly significant.

raw materials. Branca (1980) reported that storage of canned foods for 270 days did not affect their copper contents while iron, zinc and lead contents increased.

The correlation coefficients between the heavy metals of canned orange juice samples are given in Table 8. Highly significant correlations between the five metals were found. This was attributed to natural occurrence of metals in the raw orange juice. The highest significant correlation was observed between tin and iron. It indicates that the dissolution of iron is accompanied by excessive leaching of tin. This confirms results cited by Hall (1979) in his study on the shelf life of canned foods.

In conclusion, packing of orange juice in tin cans causes higher metallic contamination than packing in paperboard or laminate pouches. It is therefore recommended to expand the use of non-tin containers for packing orange juice.

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