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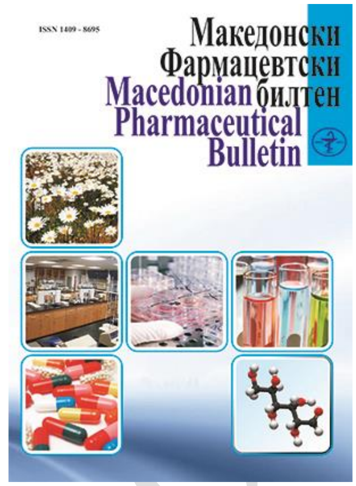

Title: Evaluation of selected heavy metals in food packaging materials

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Evaluation of selected heavy metals in food packaging materials

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Abstract

Heavy metal pollution is a major environmental challenge due to rapid industrial growth and human urban activities. Nowadays, attractive packaging materials containing different chemical substances are widely used. Since various types of packaging materials may contain toxic heavy metals above the permissible limits, it is mandatory their levels to be tested in order to protect public health.

The aim of this study was to analyze and control the concentrations of copper (Cu), zinc (Zn), manganese (Mn) and iron (Fe), as well as the complete migration into plastic materials that come in contact with food. In November 2021, thirty-three plastic materials which come into contact with food were supplied from local markets in the Kumanovo region. The samples were analyzed by AAS using the method MKC EN ISO 1186-9: 2010. The results have shown that plastic items with low density polyethylene (LDPE) and high-density polyethylene (HDPE) contained higher concentrations of manganese (0.018 mg/L and 0.012 mg/L, respectively), plastic items with polypropylene (PP) + polypropylene random copolymer (PPR) and polypropylene contained higher concentrations of iron (0.0250 mg/L and 0.0073 mg/L), while higher concentrations of copper and zinc were identified in PP plastic items. All examined heavy metals were found at the lowest concentration in the plastic items with polyethylene terephthalate (PET). The analysis of metals Cu, Zn, Mn and Fe in plastic food contact materials has confirmed their concentration to be below the detection limit (LOD). All analyzed materials were in accordance to the special requirements of the Rulebook for safety of plastic materials and products that come in contact with food.

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Keywords: specific migration (SML), heavy metals, plastics, full migration (OML)

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Introduction

General use items are all materials and items that come in contact with food as well as consumer items. Plastic materials are widely used for food packaging due to their excellent properties, especially when combined with multilayer packaging through co-extrusion or lamination processes (Júnior et al., 2020). Other favorable characteristics include flexibility and variable sizes and shapes, relatively small weight, stability, barrier properties, breaking resistance and high quality (Ardic et al., 2015). However, food packaging materials that come in direct contact with food can cause food contamination with toxic substances and elements that migrate into the food. Heavy metal contamination of plastic packaging materials is one of the major food safety concerns (Li et al., 2014). Metals are important as trace elements for living organisms, but they can accumulate in living organisms above a certain concentration and cause toxic effects or they can be converted into other compounds in the environment (Dökmeci, 2020) and cause toxic effects, such as disorders in mental function, kidneys, nervous system, respiratory system and many other physiological activities of body cells and other organs. The term heavy metal refers to a group of metals and metalloids with an atomic density greater than 4 g/cm^3, or at least five times the density of water (Onakpa et al., 2018).

The migration of chemicals into plastic packaging for food or medicine is another example of unwanted migration because some migrating substances can be toxic or give off an unpleasant taste to food or eventually destroy the drug or improve the breakdown of active substances in the drug (Danish Ministry of the Environment, Environmental Protection Agency, 2014).

Sustainable plastics, with an estimated lifespan of hundreds of years in marine degradation, can be degraded into micro - and nanoplastics in shorter time frames, facilitating their uptake from marine biota through the food chain. These polymers may contain chemical additives and contaminants, including some known endocrine disruptors that can be harmful at extremely low concentrations to marine biota, posing a potential risk to marine ecosystems, biodiversity and availability of food (Wagner et al., 2014).

In general, migration from food packaging material depends on the migrant's initial concentration, the migrant diffusion coefficient in the packaging material, and the interaction between the packaging and the food simulator (Stoffers et al., 2007).

Historically, many additives and catalysts used in plastics have been based on toxic metal compounds. Despite the subsequent restrictions, hazardous additives remain in the plastic (Turner and Filella, 2021). Plastic production rates have increased 20-fold since 1964,

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resulting in an estimated 311 million tons of plastic in the oceans as of 2014. Additional estimates show that with the current rates of plastic production, by 2050, the total mass of plastic will exceed biomass of fish (Munier and Bendell, 2018).

There is already scientific evidence and sufficient facts to support the activities of scientific institutions, industry, politics and civil society communities to prevent the ongoing fight against plastics and toxic chemicals trapped in the environment (Gallo et al., 2018).

The migration test is aimed at determining and estimating the actual or maximum concentration of a migrating food packaging compound for a particular packaging method (Schmid and Welle, 2020). There are two main categories of plastics, including thermoplastics and thermostats (Fadlalla, 2010). Plastics contain not only polymers or copolymers but also chemicals that are intentionally added for some functionality (plasticizers, stabilizers, additives, color pigments, etc.) (Murphy, 2001). Because raw plastics are brittle, hard chemicals are added during the manufacturing process to obtain specific properties (such as elasticity, stability, color density, transparency) and thus to be adapted to the final product for specific use. Some additives may prevent or finally slow down the decomposition of plastics (Letcher, 2020).

Plastics can also be made from microorganisms that create macromolecular structures from starting materials by fermentation processes. Potential health risk may arise from the migration of incompletely reactive starting substances, intermediates or by-products of the fermentation process (Commission Regulation (EU), 2011). An important factor which is not taken enough into account when choosing the type of plastic material for a particular type of food (acidic, fatty and neutral) is the inertia of the polymer, on which will depend the transfer (diffusion) of food substances. The more inert the material is and the lower the chemical danger in terms of migration of toxic substances in food (Kiroska-Petreska et al., 2014).

In this study, plastic food containers of polypropylene (PP), polystyrene (PS), low density polyethylene (LDPE), high density polyethylene (HDPE), PP + PPR (polypropylene random copolymer) and polyethylene terephthalate (PET) were analyzed. The main purpose of the study was to investigate the concentration of toxic metals such as manganese (Mn), iron (Fe), copper (Cu) and zinc (Zn) in plastic containers.

Materials and methods

Thirty-three (33) plastic food materials were purchased from the markets in the region of Kumanovo. The samples were brought to the Center for Public Health in Skopje to analyze

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complete migration of non-volatile substances, as well as to confirm initially measured concentrations of heavy metals at the Center of Public Health in Kumanovo.

Chemicals

Reagents used include 3% aqueous solution of glacial acetic acid (used as a simulation solution), high quality redistilled water, reference standards for manganese, iron, copper, zinc (Merck, Carl Roth), and modifiers.

Sample preparation and AAS analysis

The AAS analysis was performed based on the method MKC EN ISO 1186-9: 2010 (method with European standard that specifies the methods of testing plastic products, which are intended to come into contact with food products). This method was standardized by the Institute for Standardization in the Republic of North Macedonia in 2010. The samples were analyzed for specific metal migration limit (SML-specific migration limit) and complete migration of non-volatile substances (OML-overall migration limit). Thirty-three plastic samples were washed with detergent and warm water, then washed with distilled water to remove any possibility of dirt. According to the regulations for special safety requirements for plastic materials and products that come in contact with food, the simulator B is used for all items, because it is the simulator used for the worst predictable conditions (Commision Regulation (EU), 2020/1245). This simulator is used when materials that can also come in contact with sour, dairy or alcoholic foods, are aimed to be analyzed.

Thirty-three samples were divided into two groups:

First group: plastic samples that are intended to come into multiple contacts with food. Herein were samples numbered 1, 4, 7, 8, 10, 13, 15, 23, 24, 25 and 26. These samples were placed in a sterilizer (Thermostat Binder KB 400 E6 GmbH®) at a temperature of 70 °C for a period of 2 hours.

Second group: plastic items that are intended to come into contact with food over a long period of time. To this group belonged the samples numbered 2, 3, 5, 6, 9, 11, 12, 14, 16, 17, 18, 19, 20, 21, 22, 27, 28, 29, 30, 31, 32 and 33. Testing for 10 days at 40 °C has covered the total storage time under refrigeration and freezing conditions, including preheating and/or heating conditions up to 70 °C. These items were placed in a sterilizer (Thermostat Binder KB 400 E6 GmbH®) at a temperature of 40 °C for period of 10 days.

Table 1 presents the composition of the tested plastic materials as well as their abbreviated designation.

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Table 1

Method for determining complete migration of non-volatile substances (OML)

The tests were performed according to the method EN ISO 1186-3 (method for testing the overall migration in liquid food simulators with total immersin). Laboratory cups marked with the ordinal number were measured empty. Then, the cups were fill with 100 mL of the extract obtained after preparation, under the given conditions, and the extract evaporated to dryness. The residue in the cups has been measured and the total migration has been calculated. The transfer of the total amount of substances from the packaging to the food is called complete migration. The term specific migration refers to the migration of a specific (identified) toxic substance (inorganic or organic).

Method for determination of metals with GF – AAS

Thirty-three numbered simulators were transferred to polyester GFAAS cups in a graphite furnace with the appropriate standards according to which the value is read. Each sample was analyzed in triplicate for greater accuracy.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Results and discussion

According to regulation established specific rules for plastic materials, products that come in contact with food must not release higher amounts of copper (Cu) than 5 mg/kg, iron (Fe) 48 mg/kg, manganese (Mn) 0.6 mg/kg and zinc (Zn) 25 mg/kg to food or food simulant, and total migration must not exceed 10 mg/dm^2. Concentrations of the investigated heavy metals such as manganese, iron, copper and zinc have been determined to be lower than LOD in all thirty-three plastic containers purchased from regional supermarkets, before to come into contact with food.

Table 2

Fig. 5

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From the results presented in Table 2, it can be observed that the highest concentration of manganese was determined in the first sample P31 (0.00180 mg/L), LDPE according to the polymer composition, while in other samples no traces of manganese were found, especially in most of the samples which according to the composition are PP and PET. More significant variation was noticed in the results obtained for iron. Namely, the lowest concentration of iron was detected in the samples that according to the composition of the polymer belong to PET and HDPE, while the highest concentration of iron (0.110 mg/L) was measured in the sample P31, LDPE according to the composition of the polymer, followed by sample P3 (0.00730 mg/L) and P4 (0.00570 mg/L) (PP according to the composition of the polymer).

Similar variations as for iron concentration were found for zinc, with the lowest zinc concentration found in PET samples and the highest zinc concentration found in samples P32 (0.0190 mg/L) and P30 (0.0150 mg/L), both belonging to the LDPE material according to polymer composition followed by samples of the PP group, P33 (0.0130 mg/L) and P3 (0.01270 mg/L).

The obtained results showed that the concentration of copper in all the analyzed materials was lower than detection limit of the method. Materials that belong to group LDPE and PET according to the composition of the polymer were free of copper or contain only traces, while the maximum concentration of copper was determined in the sample P10 (0.0040 mg/L) designated as PP according to the composition of the material.

On the other hand, the obtained results showed that plastic items intended to come into multiple contacts with food contained higher concentration of zinc and manganese, while in most plastic samples that are intended to come into contact with food for a longer period of time higher concentration of iron and copper was found. Regarding the composition of the polymer, the results showed that plastic items with LDPE and HDPE contained higher concentration of manganese (0.0180 mg/L and 0.012 mg/L, respectively), plastic items with PP + PPR and PP contained higher concentration of iron (0.0250 mg/L and 0.00730 mg/L, respectively), while higher concentrations of copper and zinc have been identified in PP plastic items. From the obtained results, it can be concluded that the lowest concentration of all examined heavy metals, manganese, zinc, iron and copper was identified in the plastic materials with PET.

Complete migration values (mg/dm^2) of non-volatile substances of plastic items intended to come into multiple contacts with food and plastic materials intended to come into contact with food over a long period of time (measured by gravimetric method, analytical scale Sartorius BP 110S) are shown in Table 3.

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Table 3

Fig. 6

The above presented results (Table 3) show that the highest values of complete migration (mg/dm^2) of non-volatile substances were obtained for sample P7 (3.200 mg/dm^2) followed by samples P10 and P4 (2.500 mg/dm^2 and 2.340 mg/dm^2). All these plastic materials are intended to come into multiple contacts with food. According to the composition of the polymer the sample P7 is PS, while the samples P10 and P4 are PP. As for the plastic materials that are intended to come into contact with food for a longer period of time, there are large variations in the obtained values for complete migration of non-volatile substances depending on the composition of the polymer. It is important to potentiate that complete migration in plastic samples which are intended to come into contact with food over a long period of time is significantly smaller, compared to the complete migration of plastic items that are intended to come into multiple contacts with food. Complete migration values have shown variations depending on the composition of the polymer. No non-volatile substances migration of plastic items with HDPE and PET was observed.

Data published in an article coming from India show deviations of heavy metal concentrations from the recommended national norms. Thus, examination of toxic heavy metals in plastic food containers from different parts of India revealed toxic heavy metals Cu (1.61 ppm) and Zn (1.02 ppm) where the concentration of Cu and Zn was above the permissible limit (Khan and Khan, 2015). Opposite, according to Li et al. (2014) the determined quantities of 14 investigated metal elements were lower than the limit levels defined by Regulation (EU) no. 10/2011 as it was shown in this study.

Conclusion

The obtained levels from the analysis of heavy metals (copper (Cu), zinc (Zn), manganese (Mn) and iron (Fe)) in plastic materials that come in contact with food (purchased from markets in the Kumanovo region) were lower than maximum allowed limits stated by the national legislation Rulebook on special requirements for safety of plastic materials and products that come in contact with food. Regarding the composition of the polymer, the results showed that plastic items with LDPE and HDPE contained higher concentration of manganese, plastic items with PP + PPR and PP contained higher concentration of iron, while plastic items

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with PP had higher concentration of copper and zinc. In terms of all examined heavy metals, the lowest concentration was observed in PET plastic materials.

The complete migration of non-volatile substances of the plastic materials depends on the composition of the polymer as well as the time of contact between the food and plastic materials. Plastic materials that are intended to come into contact with food over a long period of time have shown significantly lower complete migration of non-volatile substances compared to plastic items that are intended to come into multiple contacts with food.

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Резиме

Анализа на некои тешки метали во материјали што доаѓаат во контакт со храната

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Клучни зборови: специфична миграција (SML), тешки метали, пластика, целосна миграција (OML)

Загадувањето со тешки метали е голем еколошки предизвик поради брзиот индустриски раст и човечките урбани активности. Во денешно време, широко се користат атрактивни материјали за пакување кои содржат различни хемиски супстанци. Бидејќи различни видови материјали за пакување може да содржат токсични тешки метали над дозволените граници, задолжително е нивното ниво да се тестира за да се заштити јавното здравје. Целта на оваа студија беше да се анализираат и контролираат концентрациите на бакар (Cu), цинк (Zn), манган (Mn) и железо (Fe) и целосната миграција во пластични материјали кои доаѓаат во контакт со храната. Во ноември 2021 година, од локалните пазари во Кумановско земени се триесет и три (33) пластични материјали кои доаѓаат во контакт со храна. Примероците беа анализирани со AAS користејќи го методот МКС EN ISO 1186-9: 2010. Резултатите покажаа дека пластичните предмети со полиетилен со мала густина (LDPE) и полиетилен со висока густина (HDPE) содржат повисоки концентрации на манган (0,0180 mg/L и 0,012 mg/L, соодветно), пластичните предмети со полипропилен (PP) + полипропилен случаен кополимер (PPR) и полипропилен содржат повисоки концентрации на железо (0,0250 mg/L и 0,00730 mg/L), додека повисоки концентрации на бакар и цинк беа детектирани во PP пластични

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предмети. Во пластичните предмети со полиетилен терефталат (PET) сите испитани тешки метали се пронајдени во најниска концентрација. Анализата на металите Cu, Zn, Mn и Fe во пластичните материјали што доаѓаат во контакт со храната потврди дека нивната концентрација е под границата за откривање на применетиот метод. Сите анализирани материјали беа во согласност со посебните барања од Правилникот за безбедност на пластичните материјали и производи кои доаѓаат во контакт со храната.

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Table 1. Designation, description and composition of food plastic materials intended for testing of complete migration of non-volatile substances and specific migration of metals

Sample Designation	Sample/ Description	Composition of polymer
1	Red plastic tray	PP
2	Yellow plastic mustard bottle	LDPE
3	Blue plastic container with ice molds	PP
4	Transparent plastic shovel	PP
5	Transparent disposable plastic box	PET
6	Transparent plastic cup of 300 mL	PET
7	Disposable Food Box	PS
8	White plastic chopping board	PP
9	Plastic bottle for vinegar of 250 mL	PP
10	Gray plastic funnel	PP
11	Transparent plastic bottle of 1.5 L	PET
12	Pink plastic storage box	PP
13	Pink plastic spoon for honey	PP
14	Transparent plastic container with purple lid	PP
15	White bottle of 200 mL	PP
16	Transparent four-layer plastic ice cream molds	PP
17	Light brown ice cream cone lid in the shape of a bear	PP+PPR
18	White ice cream dish lid in the shape of a bear	PP+PPR
19	Yellow dish ice cream lid in the shape of a bear	PP+PPR
20	Blue ice cream dish lid in the shape of a bear	PP+PPR
21	Jar lid (inside)	HDPE
22	White plastic bottle cap	PP
23	Green plastic canape sticks	LDPE
24	Dark blue plastic canape stick	LDPE
25	Pink plastic canape stick	LDPE
26	Orange plastic canape stick	LDPE
27	Blue preform for PET bottle (wholesale)	PET

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28	Transparent PET bottle preforms	PET
29	Blue cap for a large bottle	HDPE
30	Grand Coffee foil 200 mg	LDPE
31	Bar Coffee Classic foil 100 mg	LDPE
32	Gold Coffee foil 500 mg	LDPE
33	Transparent foil for Minas Coffee	PP

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Table 2. Selected heavy metal concentrations (mg/L) in food plastic materials

		Heavy metals (mg/L)			
Sample	Polymer composition	Manganese (Mn)	Iron (Fe)	Copper (Cu)	Zinc (Zn)
Blind test 3% CH_3COOH (2 h/70 °C)		0.0	0.00110	0.00010	0.00200
Blind test 3% CH_3COOH (10 days/40 °C)		0.0	0.00260	0.00008	0.00300
1	PP	0.00054	0.00560	0.00090	0.01040
2	LDPE	0.00000	0.00000	0.00000	0.00000
3	PP	0.00002	0.00730	0.00110	0.01270
4	PP	0.00006	0.00570	0.00050	0.00930
5	PET	0.00003	0.00052	0.00000	0.00000
6	PET	0.00000	0.00120	0.00220	0.00000
7	PS	0.00010	0.00230	0.00160	0.00720
8	PP	0.00000	0.00110	0.00080	0.00200
9	PP	0.00000	0.00150	0.00000	0.00560
10	PP	0.00022	0.00400	0.00400	0.00830
11	PET	0.00000	0.00200	0.00020	0.00000
12	PP	0.00000	0.00150	0.00020	0.00400
13	PP	0.00000	0.00200	0.00030	0.00570
14	PP	0.00000	0.00000	0.00000	0.00000
15	PP	0.00000	0.00320	0.00060	0.00590
16	PP	0.00000	0.00400	0.00130	0.00400
17	PP+PPR	0.00001	0.00300	0.00220	0.00690
18	PP+PPR	0.00000	0.00600	0.00030	0.00730
19	PP+PPR	0.00000	0.00300	0.00060	0.00890
20	PP+PPR	0.00000	0.02500	0.00100	0.00900
21	HDPE	0.00120	0.00300	0.00100	0.01110
22	PP	0.00001	0.00000	0.00010	0.00610
23	LDPE	0.00000	0.00030	0.00004	0.00830
24	LDPE	0.00000	0.00000	0.00020	0.00250
25	LDPE	0.00000	0.00220	0.00040	0.00840

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26	LDPE	0.00000	0.00000	0.00040	0.00720
27	PET	0.00025	0.01500	0.00090	0.00930
28	PET	0.00000	0.00000	0.00000	0.00200
29	HDPE	0.00000	0.00000	0.00210	0.00900
30	LDPE	0.00000	0.00080	0.00040	0.01500
31	LDPE	0.00180	0.11000	0.00000	0.00200
32	LDPE	0.00000	0.00800	0.00030	0.01900
33	PP	0.00012	0.00310	0.00020	0.01300

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Table 3. Complete migration values (mg/dm^2) of non-volatile substances in food plastic materials

Sample	Composition of polymer	Area (dm^2)	Volume (mL)	Complete migration (mg/dm^2)
Blind test 3% CH_3COOH (2h/70 °C)		/	100 (extract)	0.0002 g
Blind test 3% CH_3COOH (10 days/ 40 °C)		/	100 (extract)	0.0023 g
1	PP	6.21	340	0.164
2	LDPE	1.44	170	0.390
3	PP	1.00	100	0.000
4	PP	0.80	170	2.340
5	PET	2.55	340	0.000
6	PET	1.26	170	0.000
7	PS	1.00	170	3.200
8	PP	0.57	170	0.000
9	PP	1.43	170	0.000
10	PP	1.00	170	2.500
11	PET	1.00	170	0.000
12	PP	3.02	340	0.000
13	PP	1.00	170	0.000
14	PP	1.00	150	0.000
15	PP	1.00	170	0.000
16	PP	1.00	250	0.166
17	PP+PPR	1.00	170	0.000
18	PP+PPR	1.00	170	1.000
19	PP+PPR	1,00	170	0.000
20	PP+PPR	1,00	170	0.000
21	HDPE	1,00	170	0.000
22	PP	1,00	170	0,000

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23	LDPE	1,00	170	0,670
24	LDPE	1,00	170	0,000
25	LDPE	1,00	170	0,166
26	LDPE	1,00	170	0,333
27	PET	1,00	170	0,000
28	PET	1,00	170	0.000
29	HDPE	1.00	170	0.000
30	LDPE	1.00	170	1.330
31	LDPE	1.00	170	0.200
32	LDPE	1.00	170	0.000
33	PP	1.00	170	0.000

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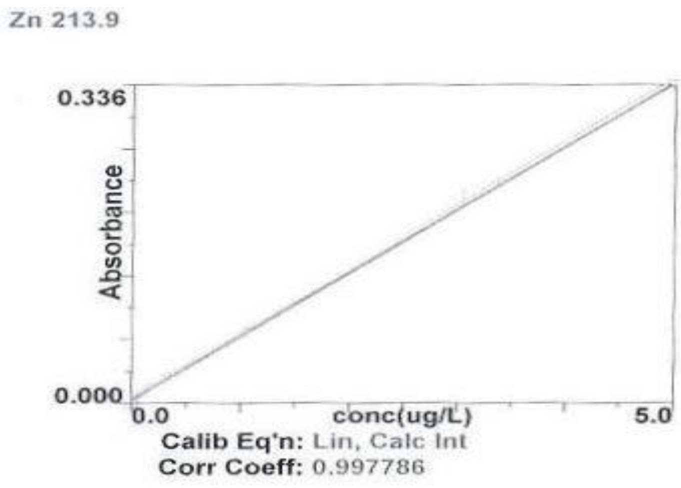

Fig. 1. Calibration curve of standart solution of Zn.

The coefficient of regression (R^2) was 0.997786.

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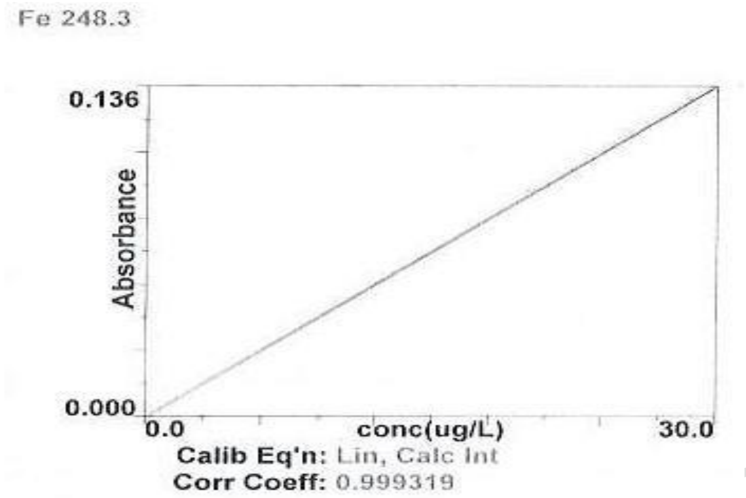

Fig. 2. Calibration curve of standart solution of Fe.

The coefficient of regression (R^2) was 0.999319.

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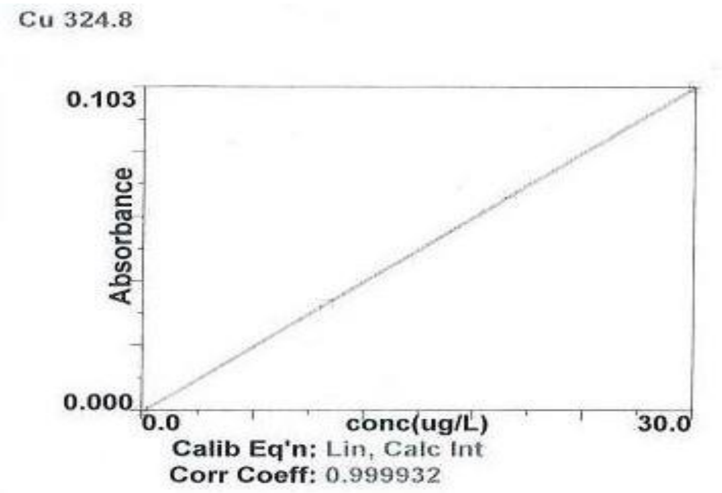

Fig. 3. Calibration curve of standart solution of Cu.

The coefficient of regression (R^2) was 0.999932.

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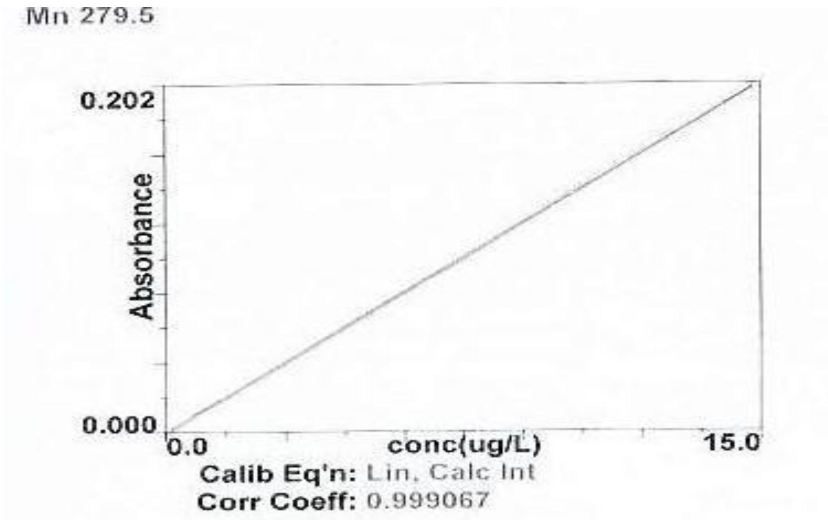

Fig. 4. Calibration curve of standard solution of Mn.

The coefficient of regression (R^2) was 0.999067.

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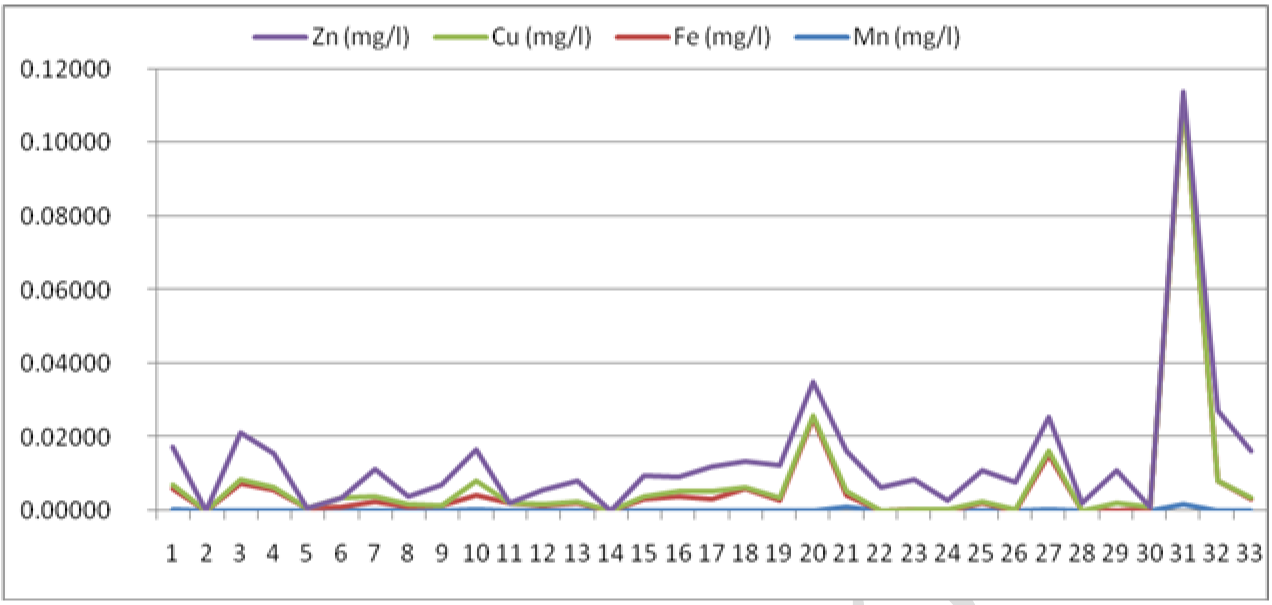

Fig. 5. Comparative presentation of selected heavy metal concentrations (mg/L) in food plastic materials: zinc (Zn), copper (Cu), iron (Fe) and manganese (Mn).

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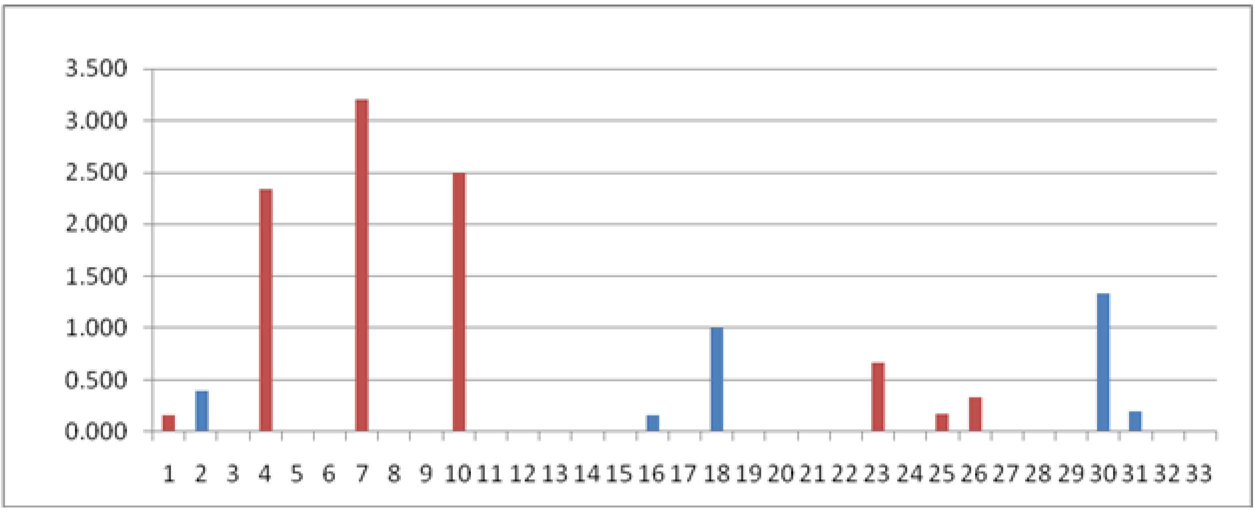

Fig. 6. Complete migration values (mg/dm^2) of non-volatile substances in food plastic materials: plastic materials that are intended to come in multiple contacts with the food are red colored, while plastic materials that are intended to come in contact with the food in a longer period are blue colored.

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