Macedonian pharmaceutical bulletin, **69** (2) 47 - 53 (2023)
ISSN: 1857-8969
DOI: 10.33320/maced.pharm.bull.2023.69.02.005
Original scientific paper

Investigation of children's toys quality on the Macedonian market: determination of lead, cadmium and chromium levels

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Received: September 2023; Accepted: November 2023

Abstract

Information about heavy metal contamination in toys is essential to ensure the safety of the children's product. However, 95% of the tested toys come from China, a country known for use of toy materials which are often banned in EU countries. In this paper the results from determination of three heavy metals levels (lead, cadmium and Trivalent chromium), in samples from children's toys available through specialized stores and markets in Republic of North Macedonia (RNM), are presented. The levels of lead, cadmium and Trivalent chromium in 28 samples, divided in three categories according to EN 71-3:2019, were determined using graphite furnace atomic absorption spectrometry (GF-AAS), after suitable sample preparation.

The obtained results showed that the concentration of lead in the toys of the second category (purple slime and tattoo) are significantly higher than the maximum migrated limits (MML). Lead in children's toys originates from toxic lead dyes and can lead to serious health complications of central nervous system, cardiovascular, immune system and kidneys. Toys like finger paints had increased values of cadmium, but insignificantly higher compared to MML. Chromium concentrations did not exceed the MMLin all 28 toys. In summary, 14.3% of the tested toys had higher levels of the examined heavy metals than the regulatory limits. Therefore, improved quality control of children's toys is an emerging issue to ensure consumers are purchasing safe, healthy and environmentally sustainable products.

Key words: children's toys, heavy metals, maximum migrated limits (MML)

Introduction

Children's toys are consumer products or shaped materials designed or intended by the manufacturer for playing of a child 14 years of age or younger, that remain in contact with the body through the oral cavity or skin (Official Gazette of Republic of Macedonia № 149, 2010; № 17, 2012; Directive 2009/48/EC, 2009). Their use is a biological instinct which occurs in all children with normal development and is one of the main ways of communication. Children under the age of six are in a critical developmental stage both physically and cognitively. Therefore, toys should be designed and manufactured in a way that meets hygiene standards, in order to avoid any risk of infection, disease and contamination (Amoruso et al., 2015; Finch et al., 2015).

The greatest danger of using children's toys is the

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presence of toxic substances used during production. Due to the persistent mouthing behavior of children, there is an increased risk of exposure to toxic substances, such as heavy metals from contaminated toys (Al-Qutob et al., 2014). Exposure to heavy metals from consumer's products can have adverse health effects, particularly possible accumulation in children's body may cause damage in the physical and the mental development. Children play and put the toys in their mouths, touch them with their skin and inhale their toxic smells. Some manufacturers use arsenic, cadmium and lead in toys for a variety of reasons. Lead (Pb) can be added as a stabilizer, color enhancer, or anticorrosive agent (Hillyer et al., 2014). Cadmium (Cd) is usually used as a substitute for lead based stabilizers or to make the product look more realistic whereas arsenic use is suspected to be related to the use of certain coloring dyes (Hillyer et al., 2014).

Information about metal content/heavy metal contamination in various children's toys is of prime importance to ensure the safety of the product. The Macedonian market has cheap toys imported from non-EU countries. In Europe, all toys must meet the criteria set by the European Commission Toys Safety Directive and to be marked with CE (*Conformité européenne*) mark. The toy safety European standard (EN 71-3:2019+A1:2021) ensures children's health by minimizing the exposure to potentially hazardous and/or toxic toy products, bearing in mind young children's tendency to mouth objects. Part 3 of the standard – EN71-3+A1:2014-12-entitled "Migration of certain elements" presents the migration limits for 18 elements from various toy's categories. This may be used to assess the compliance of the potentially hazardous substances with legal provisions (Directive 2009/48/EC, 2009; Kratkea et al., 2015; Karaś and Frankowski, 2018).

The objective of this study was to determine the heavy metal levels (Pb, Cd and Cr) in children's toys selected from the market in RNM and to assess their safety, based on valid Macedonian and EU regulations.

Material and methods

Chemicals, reagents and apparatus

Certified reference standard for Pb (1000 mg/L) was supplied by Merck, Germany, whereas certified reference standards for Cd (1000 mg/L) and Cr (1000 mg/L) were purchased from CPA Chem., Bulgaria. For the pH determination, pH meter (ino Lab pH 730 WTV Germany) was used. The ultrasonic treatment of the samples was conducted using DC-250H MRC ultrasonic bath.

Sample collection

Twenty-eight toy samples, purchased from specialized children's toy stores and markets throughout the territory of RNM, were included in the study. All samples were verified according to their declaration and classified in one of the following three categories: category I (brittle, powder-like or pliable materials); category II (liquid or sticky materials) and category III (coatings and scraped-off materials) according to the safety standard for toy materials and parts (Directive 2009/48/EC, 2009).

Determination of heavy metal levels

Toy's surface material/paint was removed by clean razor blades. If the toy consisted of different material, samples were taken from each part separately. Sample mass was 100 mg. To avoid cross-contamination, fresh razor blades, new gloves and new underlying paper on the lab bench for each new toy were used.

Samples were placed in suitable Erlenmeyer flasks and HCI solution (c = 0.07 moL/L) was added with a volume 50 times greater than the mass of scraped material (V = 50 x m). For the Cd determination, Erlenmeyer flasks containing the samples were covered to be protected from light. The pH value of all sample solutions was determined. The flasks were treated at 37 ± 2 °C for 1 h using ultrasonic bath and then let stand for another 1 h at 37 ± 2 °C, without ultrasonic treatment. At the end of the sample treatment period, the pH value was determined again because it should not be outside the limits of pH = 1.1 to 1.2. If the pH is greater than 1.2, 6 moL/L HCl is added to obtain pH within the requested limits. The number of drops was calculated and adjusted to the immersion volume (EN 71-3:2019). The levels of Pb, Cd and Cr (III) in the toy samples were analyzed in triplicate on graphite furnace atomic absorption spectrometer (PerkinElmer Analyst 600). Calibration curves were made in the range 0 - 100 ppb for Pb, 0 - 10 ppb for Cd and 0 - 25 ppb for Cr.

The levels for each analyte in the sample were calculated according to the following formula:

$$\text{Migration (mg/kg)} = C \times V \times 1000 / A \qquad \text{(Eq. 1)}$$

where:

Migration (mg/kg) - element released from the sample, expressed in mg/kg;

C - concentration of the element (metal) in the migration solution (mg/L);

V - volume of added simulant (mL) + volume of added drops for pH adjustment

A – weight of the sample (mg).

Results and discussion

The appearance/description, the laboratory code and the category of the investigated children's toys collected from the Macedonian market are given in Table 1.

Levels of Pb, Cd and Cr in all samples are presented in Fig. 1. The determined levels of the examined heavy metals were compared with the MML for each metal considering the toy's category. MMLs of Pb are 13.5, 3.4 and 160 mg/kg and of Cd are 1.9, 0.5 and 23 mg/kg for category I, II and III, respectively. Respective MMLs of Cr

Table 1. Description of examined toys according to the toy's category (Directive 2009/48/EC)

Toy label/sample label	Toy description	Toy's category
24	Dark coloured play-doh	I
25	Medium coloured play-doh	I
26	Bright coloured play-doh	I
21	Dark finger-paint	II
22	Medium colored finger-paint	II
23	Bright colored finger paints	II
27	Slime jelly	II
28	Metalic tattos	II
1	Plastic gun	III
2	Plastic remote control car	III
3	Sports ball	III
4	Baby toy, plastic	III
5	Rubic's cube	III
6	Plastic pyramid cups	III
7	Plastic music instrument	III
8	Plastic gun	III
9	Painting toy set	III
10	Plastic bath toys	III
11	Plastic police car	III
12	Plastic gun	III
13	Plastic dinosaur	III
14	Set of wooden stacking rings	III
15	Glass marbles	III
16	Stuffed toys	III
17	Stuffed toys	III
18	Paper puzzle	III
19	Wooden puzzle	III
20	Children's picture book	III

for category I, II and III are 37.5, 9.4 and 460 mg/kg (EN 71-3:2019).

The results obtained from the tested samples revealed that Pb levels (sample number 27 and 28) were significantly higher than MML according to the Directive 2009/48/EC. Samples number 27 and 28 are sticky colored objects that come in direct contact with the skin. The report written by Piccinini (2014) for the adverse health effect of tattoos in general emphasized the potential for long term effects of exposure to the chemicals in the inks that might become critical with time due to the high number of tattooed people. Samples labeled 21 and 22 (finger paints, category II) showed increased levels of cadmium. For the other tested toys from category, I and III, Cd levels did not

exceed the regulatory limits (Directive 2009/48/EC, 2009). Cr (III) levels in all tested samples were within the permissible limits. The results for heavy metal levels in children's toys belonging to category III (sample labels 1-20), divided according to the material they are made of polymer (1 -13), wooden (14, 19), glass (15), textile (16,17) and paper (18, 20) showed compliance of the investigated toys with the regulatory requirements (Official Gazette of Republic of Macedonia № 149, 2010; № 17, 2012; Directive 2009/48/EC, 2009).

Heavy metals in children's toys are concentrated in the surface coating (usually rich in colors) and they come in contact and migrate into the human body mostly through mouthing behavior of young children. One of the most commonly encountered metals in toys is Pb because it is used in the production of various pigments. Pb, both in paints as well as children's polyvinyl (PVC) toys is a major health care concern which has attracted the attention of the international community (Njati and Maguta, 2019; Shein and Hou, 2018). Compared to adults, children are much more vulnerable to lead's harmful effects, particularly when exposed repeatedly. Lower IQ and cognitive decline are connected to lead exposure in young children due to Pb's interference with cell signaling and nerve conduction (Muscat, 2020). Although no level of lead exposure is considered safe, 90 mg/kg is a permissible limit in lead-based paints based on the UN Environment "Model Law and Guidance for Regulating Lead Paint" introduced in September 2017 (Njati and Maguta, 2019). The U.S. EPA, the EU, as well as many other countries throughout the world, including China, accepted this restriction, presuming that the exterior section of children's toys should have no more than 90 mg/kg lead concentration. Cd compounds used as toy's stabilizers can readily leach out endangering health. There should be no level of cadmium in the blood that is considered safe for children, similar to lead, because a buildup of Cd in the kidney may result in kidney disorders and carcinogenic effects (Turner, 2019). Turner (2019) emphasized that the Cd migration in certain items is an order of magnitude higher than the Toy Safety Directive limit of 17 mg/kg, presenting serious danger to children. Pb and Cd are the primary research focus within toys safety investigations due to their respective neurotoxic and nephrotoxic characteristics. Additional metals including Cr, Ni, Zn, Cu, and Mn are essential issues regarding contamination of toys. Cr metal and Cr (III) compounds are not generally considered harmful, while Cr (VI) is a very active carcinogen at the cellular level (Shin et al., 2023). Recently, Sawicka et al. (2021) reported incomplete current knowledge for transformation of chromium in the body, unexplained mechanism of genotoxic and mutagenic action of Cr (VI) and doubts about the safety use of both trivalent and hexavalent Cr. Since skin contact with chromium can produce serious allergic reactions including skin redness, swelling, and ulceration (Shin et al., 2023), Cr is subject to a limit in toy surface coatings and parts regardless of the type of chromium in the product.

Several studies dealing with heavy metal level determination in children's toys can be found in literature. In the study performed in several laboratories from the EU countries, for determination of migration of heavy metals, 5% of all examined toy samples did not comply with the regulatory requirements and all of them were manufactured in China (Igweze et al., 2020). Also, a study performed in countries of the former USSR in 2012 revealed that 30% of the examined toys had higher levels of heavy metals according to the national standards. From all the tested samples, 18% had higher levels of Pb, 2% had higher levels of cadmium and 6% of the examined samples had higher levels of chromium (Balbanova et al., 2004). A recent study conducted in 2018 in some Asian countries showed that 62% of the examined toys had higher levels of all organic as well as inorganic toxicants, and 19% had elevated levels of heavy metals (Ismail et al., 2017).

The levels of heavy metals in toys, tested in different laboratories are usually found in varying amounts. In general, the levels of Pb are higher than those of Cr, and the levels of Cd in most of the cases are lower. Pb and Cd usually have the highest levels in black, then in red- and yellow-colored children's toys (Ismail et al., 2017). Kindi and Ali (2020) have reported Pb, Cd and Cr were found in varying amounts in thirty-two investigated samples (children's toys with black, red and yellow coverings). Further, in their study SPSS program was used to make correlations between amount of Pb, Ni and C and revealed the black color had the highest heavy metal concentrations, then red and yellow. Pb amounts were also found to be related to toy price, age group, color, and sales volume in the study of Shen et al. (2018) who explored whether Pb-based paints are an issue for toys sold on Chinese online shopping platforms as well as influencing factors which may be related to the lead amounts in toys. It was noticed that toys sold by unorganized sellers and cheaper ones contained higher Pb amounts compared to organized sellers, such toys were mostly intended for infant and toddlers, while toys with large sales volume tended to have lower Pb concentrations. Although limited research was applied on lead-based paints and their use in toys, such toys may pose health risk to children, particularly vulnerable children (Shen et al., 2018). On the contrary, manufacturers of children's toys are fighting to make as much profit as possible, offering various products, some of which are of dubious quality and pose a potential danger to children's health. The levels of investigated heavy metals in toys produced in China are higher than those produced in EU countries (Ismail, et al., 2017).

Also, due to new legal regulations, which only require checking the declaration upon importation of the product, the number of samples that are taken in for testing to the laboratories was again decreased. Thus, future efforts must be made to adjust regulatory requirements and each sample to be subjected to analysis, in order to ensure children's health.

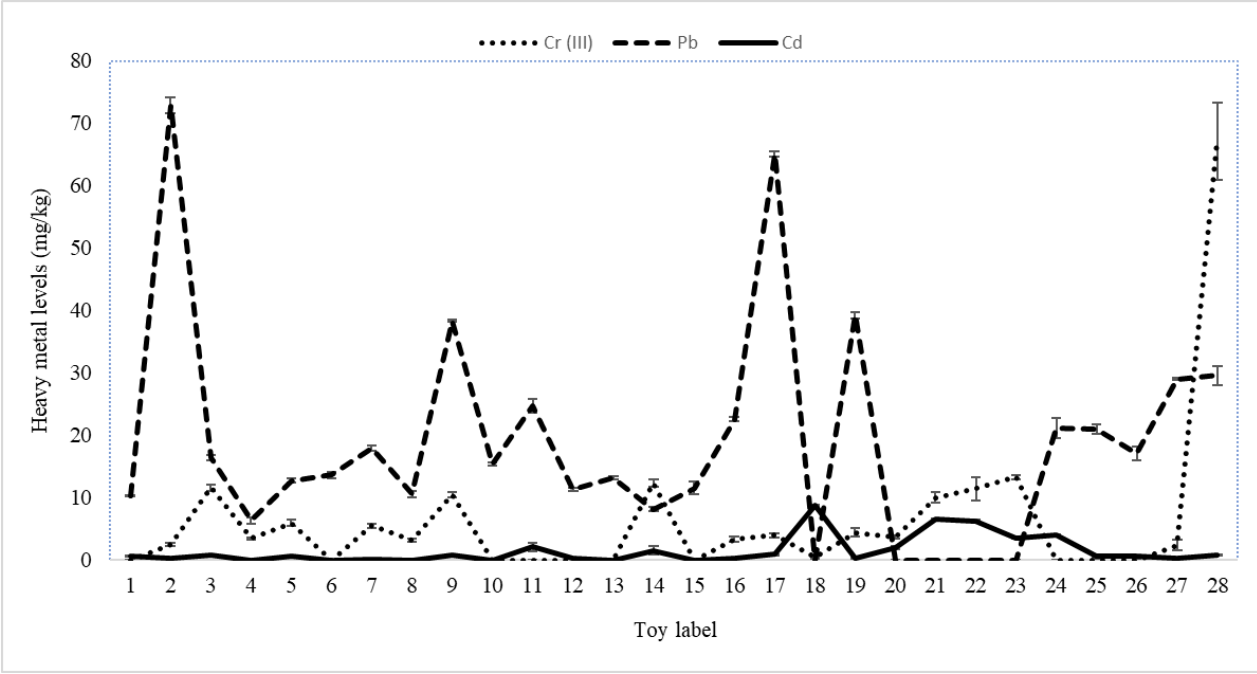

Fig. 1. Levels of Pb, Cd and Cr (III) in the examined toys; Toy label is given in Table 1.

Conclusion

According to the obtained results, four tested toys or 14.3% were not adequate for children's use, because they can endanger children's health, particularly if they come into contact with them or swallow them. Toys of the second category show higher values of lead considering national and international regulatory requirements. The toy - tattoo for children (category II), showed an increased concentration of lead, which is expected in this type of samples. Toys, so-called finger paints, showed slightly increased values of cadmium (category II). Toys from the I and III category were in a compliance with regulatory requirements. In order to improve children's healthcare regular delivery of toys samples available on the market to specialized laboratories for quality control is needed. Apart from this, it is necessary for parents and others to be very careful when buying children's toys, at the same time to take into account the adequacy of the toy in relation to the age of the child.

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Резиме

Истражување на квалитетот на детските играчки на македонскиот пазар: определување на нивоата на олово, кадмиум и хром

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Клучни зборови: детски играчки, тешкиа метали, максимално дозволено ниво на миграција

Информациите за контаминација со тешки метали во играчките за деца се неопходни за да се осигура нивната безбедност. Сепак, 95% од тестираните играчки доаѓаат од Кина, земја позната по употреба на материјали што често се забранети во земјите на ЕУ. Во овој труд се прикажани резултатите од определувањето на три тешки метали (олово, кадмиум и тривалентен хром), во примероци од детски играчки достапни преку специјализирани продавници и маркети во Република Северна Македонија. Концентрациите на олово, кадмиум и тривалентен хром во 28 примероци, поделени во три категории, според EN71-3:2019, беа определени со помош на атомска апсорпциона спектрометрија со графитна печка (GF-AAS), по соодветна подготовка на примерокот.

Добиените резултати покажаа дека концентрацијата на Pb во играчките од втора категорија (виолетов слајм и тетоважа) е значително повисока од максималното дозволено ниво на миграција (МДК). Оловото во детските играчки потекнува од токсичните оловни бои и може да доведе до сериозни здравствени компликации на централниот нервен систем, кардиоваскуларниот, имунолошкиот систем и бубрезите. Во некои играчки како боја за прсти беше забележана повисока вредност за кадмиум во споредба со останатите примероци, но незначително повисока од МДК. Концентрациите на Cr не го надминуваат МДК во сите 28 играчки.

Накратко, 14,3% од тестираните играчки имаат повисоки нивоа на испитани тешки метали од регулаторните граници. Затоа, подобрената контрола на квалитетот на детските играчки е неопходна за да се обезбеди сигурноста на потрошувачите дека купуваат безбедни, здрави и еколошки одржливи производи.