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RESEARCH ARTICLE

# Study of controlled migration of cadmium and lead into foods from plastic utensils for children

**Elizeu Chiodi Pereira[1] · Isabelle Nogueira Leroux[1] · Maciel Santos Luz[2] · Bruno Lemos Batista[3] · Kelly Polido Kaneshiro Olympio[1]**

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## Abstract

Lead (Pb) is a highly neurotoxic chemical element known for reducing intelligence quotient (IQ) and promoting antisocial behavior in children and adolescents, while cadmium (Cd) is a carcinogenic bioaccumulative element. Both these metals are included in the priority pollutant list of the United States Environmental Protection Agency and in the WHO List of Chemicals of Major Public Health Concern, where contaminated foods and beverages are the most common pathways of exposure. The objective of this study was to determine total Cd and Pb levels in colored plastic utensils (cups, mugs, bowls, feeding bottles, and plates) for use by children and to measure the specific migration of these elements into beverages and foods. Total contaminant levels were determined using a handheld X-ray fluorescence analyzer. Specific migration tests were conducted using the simulant solutions acetic acid 3% (m/v) and water. Migration levels were determined by ICP-MS. Specific migration tests for Pb were also performed on commercially available samples (cola soft drink, orange juice, vinegar, and milk), with levels determined by graphite-furnace atomic absorption spectrometry (GF-AAS). A total of 674 utensils were analyzed in loco at major commercial centers in Greater São Paulo, of which 87 were purchased for containing Cd and Pb concentrations above permitted limits. Mean concentrations of the metals detected in the purchased utensils were 1110 ppm for Pb and 338 ppm for Cd. For specific migration assays, Pb levels were 187, 13, and 380 times above the permitted limit (0.01 mg.kg -1) for acetic acid, water, and orange juice, respectively. Cd levels were 50 and 2.4 times above the maximum permitted limit (0.005 mg.kg -1) for acetic acid and water, respectively. The districts where the utensils were purchased were grouped according to their social vulnerability index and compared using ANOVA. Pb levels were different between low and medium/high social vulnerability groups ($p = 0.006$). The findings corroborate the initial hypothesis that these utensils constitute a major source of exposure to PTEs such as Cd and Pb, pointing to the need for stricter regulation and inspection by the Brazilian regulatory agencies.

**Keywords** Lead · Cadmium · Plastic utensils · Specific migration · Children · Potentially toxic metals

Responsible Editor: Lotfi Aleya

✉ Kelly Polido Kaneshiro Olympio
kellypko@usp.br

Elizeu Chiodi Pereira
elizeu.chiodi@usp.br

Isabelle Nogueira Leroux
isabelle.leroux@usp.br

Maciel Santos Luz
macielluz@ipt.br

Bruno Lemos Batista
bruno.lemos@ufabc.edu.br

[1] Departamento de Saúde Ambiental, Faculdade de Saúde Pública, Universidade de São Paulo, Avenida Dr. Arnaldo 715, Cerqueira Cesar, SP 01246-904 São Paulo, Brasil

[2] Laboratório de Processos Metalúrgicos, Materiais Avançados, Instituto de Pesquisas Tecnológicas do Estado de São Paulo, Av. Prof. Almeida Prado, 532, Cidade Universitária, SP 05508-501 São Paulo, Brasil

[3] Centro de Ciências Naturais e Humanas, Universidade Federal do ABC, Avenida dos Estados, 5001 Bairro Santa Terezinha, SP 09210-580 Santo André, Brasil

## Introduction

Testing for the presence of potentially toxic elements (PTEs), such as lead, in ceramic domestic utensils dates back to the 1960s (Huzl et al. 1960). Demont et al. (2012) found that the solubility of metals, and their respective formation of complexes, plays a key role in the lixiviation of metal ions from the surface of materials, whereby changes in pH and temperature influence the specific migration of these ions into foods and beverages upon surface contact with utensils.

Human production of plastic materials began around the 1960s, and to date, about 9 billion tons of plastics have been produced, with a large portion of these materials intended for use as packaging (Geyer et al. 2017). Contamination from plastic toys is one of the most important potential routes of exposure to toxic metals such as cadmium (Cd) and lead (Pb). Greenway and Gerstenberger (2010), in an evaluation of plastic toys from daycare centers in Las Vegas (USA), found that 5.4% of the toys tested had Pb concentrations above levels permitted by local legislation, particularly yellow items. In Colombia, researchers detected levels of Pb in brown, orange, and yellow paints from plastic toys that exceeded permitted limits (Mateus-García and Ramos-Bonilla 2014a, b).

In Brazil, environmental exposure of children aged 1–4 years was evaluated both in households and daycare centers in São Paulo. The results of the study found that over 50 toys contained Pb concentrations exceeding those allowed by Brazilian law, which stipulates a limit of 600 mg.kg$^{-1}$ for lead (Silva et al 2018; Brasil 2008). Similarly, the presence of these and other toxic metals in plastic toys has been reported in many countries, including Kazakhstan (Akimzhanova et al 2020), China (Kang and Zhu 2015; Cui et al. 2015), and the UK (Turner 2018).

Naturally, food and beverage contact with contaminated plastic utensils can result in specific migrations of these potentially toxic elements. The presence of these elements in plastic has been related to plastic packaging for storing foods (Kiyataka et al 2014). Inthorn et al. (2002), through specific migration tests on plastic utensils and specific for use in microwave appliances, showed that Pb migrated in orders of up to 0.7 μg.L$^{-1}$ depending on pH, temperature, and food contact time with the plastic types.

Regarding Pb, the toxic effects of this metal in children have been associated with learning difficulties, attention deficit, low intelligence quotient (IQ), and antisocial behavior (Bellinger 2004; Needleman 2004; Lanphear et al 2005; Hornung et al. 2009; Mazumdar et al. 2011; Dickerson et al. 2016; Blackowicz et al. 2016; Wagner et al. 2017, Olympio et al. 2009; Olympio et al. 2010; Olympio et al. 2017). Lead contamination can occur during the food production process through specific migration from packaging into the foods contained (Kiyataka et al. 2014). With regard to Cd, the main toxicological effects include renal damage, hypertension, emphysema, malformation, and impaired reproductive function (Magna et al. 2014). Exposure to Cd can take place through occupational activities involving contact with the metal (OSHA 2013, Salles et al. 2021) and by ingesting foods contaminated by this element (Rahimzadeh et al. 2017).

Child exposure to these metals is a global health problem, where children are especially vulnerable both because of their hand-to-mouth behavior and the fact their gastrointestinal and nervous systems are undergoing development (Olympio et al. 2009; Li et al. 2016; Tamayo et al. 2016). Ingestion is therefore the main route of exposure to Cd and Pb where, in the case of Pb, food ingestion accounts for 80% of exposure to the element (Li et al. 2016). Moreover, Cd and Pb both feature in the list of 10 chemicals of public health concern (World Health Organization (WHO). 2011), due to their toxic effects on the body, even at low levels.

Considering, therefore, the enormous production of plastics, specifically their use for food packaging, as well as the toxicological effects of Pb and Cd, the importance of evaluating these plastic utensils as a potential source of exposure to Cd and Pb is evidenced. Thus, the objective of this study was to determine Cd and Pb concentrations in colored plastic utensils for use by children during feeding and to measure the levels which migrate from the utensils into foods and beverages.

## Materials and methods

### Description of materials, target population, elements chosen for chemical analysis, and specific migration tests

The objects of study were plastic utensils including cups, mugs, bowls, and feeding bottles intended for use by children for being colorful, containing superhero stickers or famous characters appreciated by children. In addition, children represent one of the most important population groups in order to avoid chemical exposure, since the toxicological effects of Pb in Cd are pretty harmful to this group, considering their underdevelopment of biological systems, such as gastrointestinal and neurological ones.

The materials were purchased between March and June 2018 at major commercial centers located in the districts of Lapa, Pinheiros, Capão Redondo, São Miguel, and Pari in the cities of São Paulo and Osasco, Greater São Paulo, Brazil. All of the utensils purchased were low-cost (≤ $1 USD).

Pb and Cd were the elements that had the highest values when X-ray fluorescence analyses were performed on the colored plastic utensils. Other elements, such as mercury, arsenic, and chromium had mass fractions below the detection limits of the equipment or specific legislations. Considering that only Cd and Pb had the highest averages, only these elements were selected for specific controlled migration tests.

Specific migration tests are chemical tests that use simulant solutions (such as water, ethanol, and acetic acid) to estimate the degree of contamination of a given substance in a sample (Freire et al 1998). The tests are conducted according to the recommendations from regulatory agencies, and the parameters, such as permissible threshold concentrations and methods of analysis, may change from country to country.

## Analysis using X-ray fluorescence spectrometry (XRF)

Cd and Pb in the utensils were quantified in loco using a handheld analyzer of metal alloys with coupled XRF (model Niton XL2 Analyzer, Thermo Scientific). The device was calibrated after each start-up employing a certified calibration standard based on the EN-71–3:2013, in accordance with the manufacturer's specifications. The arithmetic mean was calculated using $LOD/\sqrt{2}$ for values below the LOD (10 and 20 $mg.kg^{-1}$ for Pb and Cd, respectively).

All utensils exhibiting values exceeding 90 $mg.kg^{-1}$ and 75 $mg.kg^{-1}$ (U.S Consumer Product Safety Commission 1997; World Health Organization (WHO). 2011) for lead and cadmium, respectively, were purchased. The limit established in Brazilian law is 600 $mg.kg^{-1}$ for Pb (Brasil 2008). Since no values have been defined in Brazil for Cd limits, US legislation was adopted to define these parameters for the purposes of comparing the study results.

Materials containing Cd and Pb above the limits allowed under US law were sent to the laboratory. The materials were first washed with Extran detergent to remove any soot layer and then rinsed in deionized water. After the cleaning procedure, the items were analyzed by XRF in triplicate, with measurements taken at different points of the utensil to improve determinations of contamination by these elements.

## Sample preparation for chemical analysis and specific migration tests for Cd and Pb

The specific migration assays were carried out on a subsample of 11 cups and mugs containing the highest mass fractions of Cd and Pb, observing EN 1186–1 (2002a) and EN 1186–3 (2002b) recommended by Resolution 52/10 of the Brazilian Health Surveillance Agency – ANVISA (Brazil 2010b).

The selected plastic utensils with the highest Cd and Pb mass fraction were taken to the laboratory, where they were washed with Extran detergent and deionized water. After that, using a diamond-tipped band saw, 1 $cm^2$ fragments of each of the 11 utensils were removed and subsequently rewashed. Simulant solutions used were 15 mL of water and 15 mL of acetic acid 3% (m/v). The fragments were submerged separately in these solutions for controlled migration assays during 10 days at room temperature (25 ºC controlled in clean room – ISO Class 8) using 50-mL conical polypropylene tubes. Analytical blanks were prepared simulating the same conditions as the migration assays and all sample preparations were also carried out in the clean room. Figure 1 shows the methods adopted to perform chemical analysis both in loco and in the laboratory.

After the contact period had elapsed, Cd and Pb concentrations in the simulant solutions were determined in triplicate by inductively coupled plasma mass spectrometry (ICP-MS Agilent 7900, Hachioji, Japan). Total mass fractions of these elements in the plastic utensils were calculated based on the ratio of total surface area of the cups and mugs to their respective fragments.

## Specific migration tests for commercial beverages

Additionally, specific migration assays for Pb were performed using commercially available beverages from supermarket outlets for 4 of the utensils exhibiting the highest lead values, following the sampling procedures described in the previous topic.

Solutions for specific migration were cola soft drink (Coca-Cola™), vinegar (Castelo), processed orange juice (Xandô), and milk (Italac). The tests were conducted according to the procedures outlined earlier. The concentrations of Pb in the solutions were analyzed by graphite-furnace atomic absorption spectrometry (GF-AAS) (Model Varian GTA120 AA 240FS).

## Statistical analyses

For statistical treatment, the Paulista Social Vulnerability Index (IPVS) for each district where utensils were purchased was referenced. According to the Foundation State System of Data Analysis (FUNDAÇÃO SEADE 2010), the IPVS conveys information on the highest and lowest social vulnerability conditions to which a population is exposed. The index comprises 5 classes of vulnerability, as depicted in Fig. 2.

As shown in Fig. 2, the IPVS was 1 (extremely low social vulnerability) for the districts of Pinheiros and Lapa, 2 (low social vulnerability) for the district of Pari and city of Osasco, 4 (medium social vulnerability) for the district of São Miguel, and 5 (high social vulnerability) for the district of Capão Redondo. These results were pooled into 3 groups: group 1 (IPVS = 1), group 2 (IPVS = 2), and group 3 (IPVS = 4 + IPVS = 5). For group 3, the districts of São

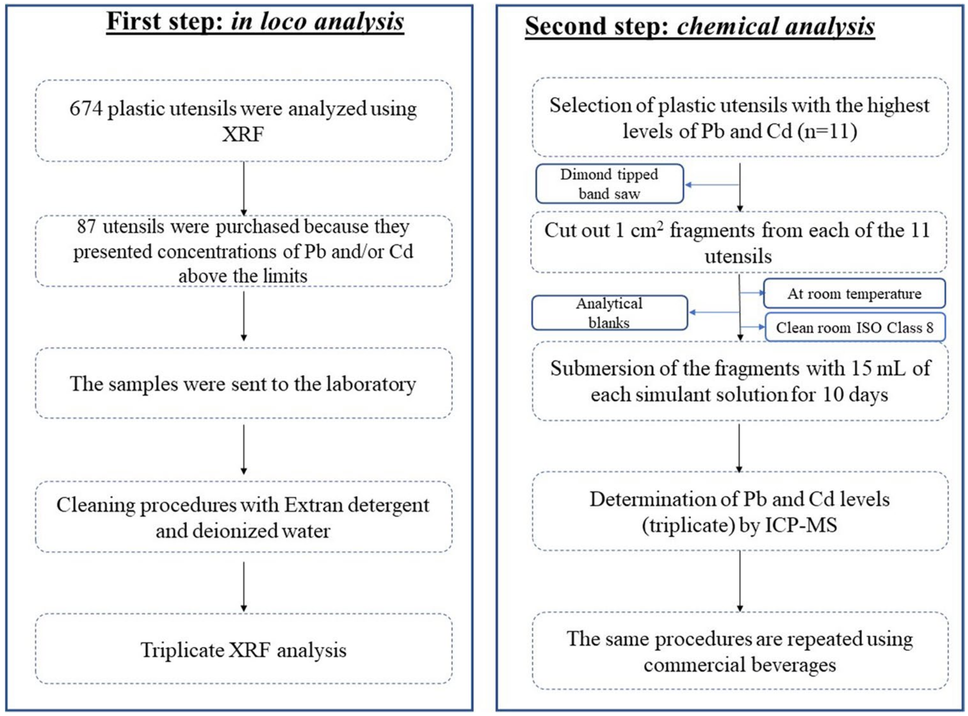


**Fig. 1** Flowchart of the methods carried out for the samples

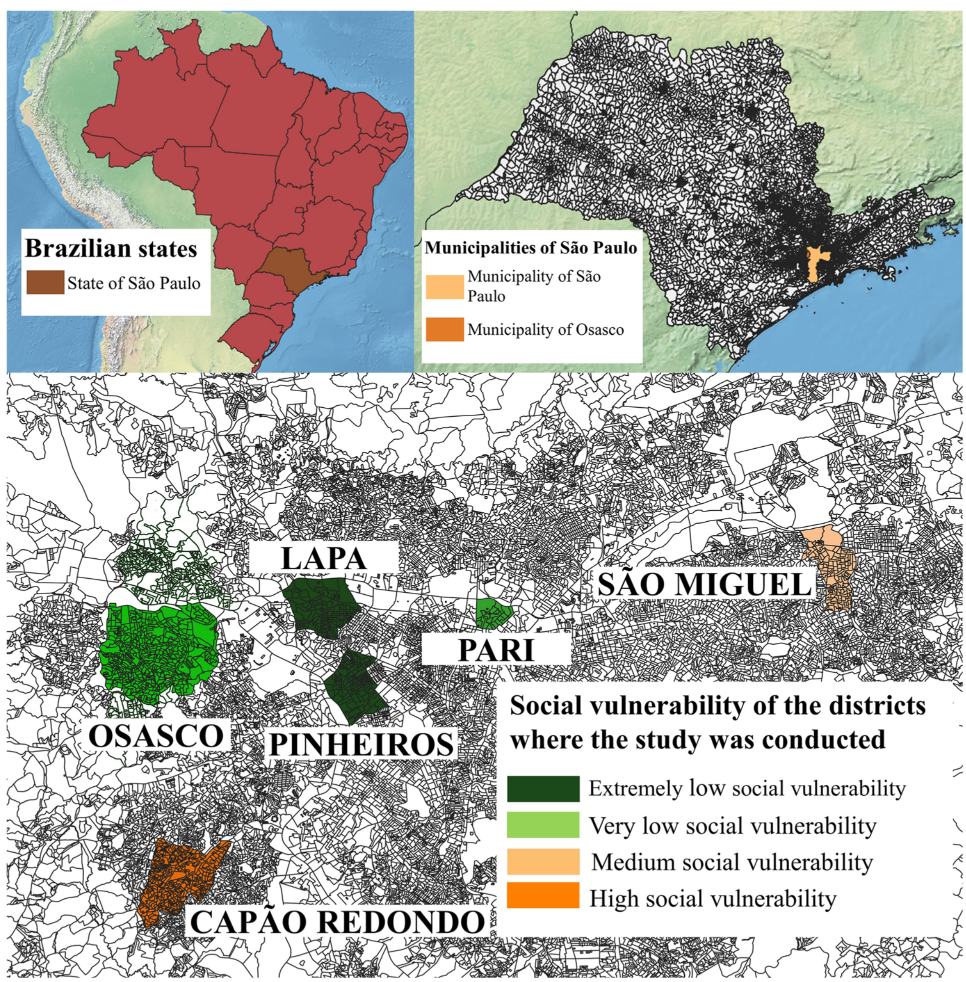


**Fig. 2** Distribution of IPVS for districts where utensils were purchased

Miguel and Capão Redondo were pooled together because their socioeconomic characteristics were similar.

The analysis of variance (ANOVA) test (95% confidence interval) was performed to compare the mean of Pb and Cd mass fractions measured in plastic utensils purchased for each region and group. The Stata 14 software package was employed for all statistical analyses.

## Results

### Analyses by XRF

A total of 674 plastic utensils were analyzed using XRF as per the procedures described in Methods. The Cd and Pb values in most of the utensils tested proved below the limit of detection. The distribution of utensils and percentage with detectable levels by type are presented in Table 1 below.

Of the total 674 utensils analyzed, 87 were purchased for exhibiting Pb and/or Cd mass fractions above the limit permitted by US law (90 $mg.kg^{-1}$ and 75 $mg.kg^{-1}$, respectively). The means, standard deviations and minimum and maximum values for Pb in the plastic utensils, by type and color, are shown in Table 2.

The overall mean Pb for the utensils purchased ($n = 87$) was 1110 $mg.kg^{-1}$. Based on the 600 $mg.kg^{-1}$ limit for Pb established under Brazilian law (Brasil 2008), 34 utensils exceeded the limit, exhibiting a mean Pb concentration of 2443 $mg.kg^{-1}$.

Of the 87 plastic utensils purchased, 15 contained Cd exceeding the limit allowed by US law (U.S Consumer Product Safety Commission 1997; World Health Organization (WHO). 2011). The means, standard deviations, and minimum and maximum values for Cd mass fractions in the plastic utensils, by type and color, are shown in Table 3.

The mean and number of utensils containing Pb and Cd mass fractions above legal limits for commercial centers visited in Greater São Paulo are given in Table 4.

Of the regions visited, the district of Pari had the highest percentage (22.8%) of utensils exhibiting Pb above-recommended levels by US law. By contrast, in the district of Pinheiros, only 7.2% of utensils contained values exceeding this limit. The highest percentage of utensils exceeding legal limits for Pb was those purchased in the city of Osasco,

**Table 1** Distribution and percentage of plastic utensils by type

| | **Total samples analysed *N* (%)** | **Samples with detectable concentrations *N* (%)** | **Pb N (%)** | **Cd N (%)** |
|---|---|---|---|---|
| Cups | 270 (40.06) | 122 (42.51) | 120 (43.01) | 27 (47.37) |
| Mugs | 174 (25.81) | 65(22.65) | 61(21.86) | 10 (17.54) |
| Plates | 107 (15.87) | 52 (18.12) | 51(18.28) | 15 (26.32) |
| Bowls | 103(15.28) | 38 (13.24) | 37 (13.26) | 5 (8.80) |
| Feeding bottles | 20 (2.97) | 10 (3.48) | 10 (3.58) | - |
| **Total** | **674 (100)** | **287 (100)** | **279 (100)** | **57 (100)** |

**Table 2** Mean, standard deviation, and minimum and maximum Pb levels, in $mg.kg^{-1}$, by utensil type and color

| | **N** | **%** | **Mean** | **SD*** | **Minimum** | **Maximum** |
|---|---|---|---|---|---|---|
| *Utensil color* | | | | | | |
| Red | 30 | 34 | 942 | 1288 | 185 | 5989 |
| Yellow | 9 | 10 | 1930 | 2685 | 91 | 8166 |
| Orange | 8 | 9 | 2296 | 2050 | 260 | 5601 |
| Green | 32 | 37 | 943 | 1526 | 104 | 7017 |
| Black | 5 | 6 | 291 | 280.5 | 125 | 783 |
| Other colors | 3 | 3 | 323 | 177 | 167 | 515 |
| **Total** | **87** | **100** | **1110** | **1642** | **91** | **8166** |
| *Utensil type* | | | | | | |
| Cups | 33 | 38 | 1235 | 1751 | 91 | 7107 |
| Mugs | 19 | 22 | 758 | 992 | 130 | 4027 |
| Plates | 18 | 21 | 776 | 1856 | 107 | 8166 |
| Bowls | 11 | 13 | 2048 | 1943 | 173 | 5989 |
| Feeding bottles | 6 | 7 | 821 | 981 | 171 | 2579 |
| **Total** | **87** | **100** | **1110** | **1642** | **91** | **8166** |

*Standard deviation

**Table 3** Mean, standard deviation, and minimum and maximum Cd levels, in mg.kg$^{-1}$, by utensil type and color

| | N | % | Mean | SD* | Minimum | Maximum |
|---|---|---|---|---|---|---|
| *Utensil color* | | | | | | |
| Red | 8 | 53 | 224 | 120 | 81 | 352 |
| Yellow | 2 | 13 | 198 | 70 | 148 | 247 |
| Green | 4 | 27 | 596 | 552 | 83 | 1195 |
| Pink | 1 | 7 | 195 | - | - | - |
| **Total** | **15** | **100** | **318** | **321** | **81** | **1195** |
| *Utensil type* | | | | | | |
| Cups | 7 | 47 | 336 | 384 | 83 | 1995 |
| Mugs | 5 | 33 | 260 | 119 | 81 | 352 |
| Plates | 3 | 20 | 370 | 488 | 88 | 993 |
| **Total** | **15** | **100** | **318** | **321** | **81** | **1195** |

*Standard deviation

**Table 4** Distribution of mean Cd and Pb levels, in mg.kg$^{-1}$, and the number of utensils, according to their mass fractions, by geographic area in which they were purchased

| *Districts* | *No. of items analyzed* | *Mean Pb ± SD* | *% items with Pb > 90* mg.kg$^{-1}$ | *% items with Pb > 600* mg.kg$^{-1}$ | *Mean Cd ± SD* | *% items with Cd > 75* mg.kg$^{-1}$ |
|---|---|---|---|---|---|---|
| *Lapa* | 82 | 162.0 ± *685.4* | 7.3 | 4.9 | 18.5 ± *39.1* | 1.2 |
| *Pinheiros* | 69 | 142.5 ± *768.7* | 7.2 | 5.8 | 18.5 ± *39.1* | 1.4 |
| *Osasco* | 165 | 298.8 ± *1021.2* | 17.6 | 9.7 | 20.9 ± *34.7* | 3.6 |
| *Capão Redondo* | 106 | 35.7 ± *119.2* | 7.5 | 0.9 | 23.5 ± *88.9* | 0.9 |
| *São Miguel* | 138 | 72.8 ± *209.3* | 15.2 | 4.3 | 37.7 ± *143.9* | 5.1 |
| *Pari* | 114 | 135.6 ± *461.8* | 22.8 | 4.4 | 20.5 ± *24.5* | 1.8 |

the region with the highest mean Pb level. The mean of Cd was very similar across districts and remained within legal limits. For Pb, however, only the districts of Capão Redondo and São Miguel had mean levels below 90 mg.kg$^{-1}$ for the element.

Group 1 (IPVS = 1, Pinheiros and Lapa) had mean Pb of 153.09 ± 722.21 and mean Cd of 18.57 ± 38.35; group 2 (IPVS = 2, Osasco and Pari) mean Pb of 232.10 ± 176.54 and mean Cd of 20.73 ± 30.93; and group 3 (IPVS = 4 + IPVS = 5, São Miguel and Capão Redondo) had a mean Pb of 56.65 ± 176.54 and mean Cd of 31.51 ± 123.03 mg.kg$^{-1}$.

Comparison of the groups showed no statistically significant difference for Cd, whereas a significant difference was found among the groups for Pb ($p = 0.0089$), particularly groups 2 and 3 ($p = 0.06$) on ANOVA. There was a major disparity between these groups for income, where the value for group 2 was 3 times higher than for group 3.

**Table 5** Types of plastic utensils by color and mean Pb and Cd mass fraction (mg.kg$^{-1}$) for 11 utensils used in specific migrations assays

| Utensil type | Color | Pb mass fraction | Cd mass fraction |
|---|---|---|---|
| Mugs ($n = 6$) | Orange | 4535 | < LOD |
| | Pink | < LOD | 195.0 |
| | Green | 4027 | < LOD |
| | Red | 188.0 | 352.0 |
| | Red | 2579 | < LOD |
| | Red | 965.0 | < LOD |
| Cups ($n = 5$) | Orange | 4662 | < LOD |
| | Orange | 5901 | < LOD |
| | Green | 7107 | < LOD |
| | Red | 559.0 | 263.0 |
| | Red | 440.0 | 244.0 |
| **Mean and SD*** | | 2815.3 ± 2545.3 | 101.3 ± 133.5 |

*Standard deviation

## Specific migration tests for simulant solutions

The types of plastic utensils ($n = 11$) used in the migration assays, along with the overall mean and standard deviation for mass fractions of the metals, are given in Table 5. The values shown were derived from the XFR analyses of these utensils.

The mean Pb values as measured by XRF analyses shown in Table 5 are almost 32 times over the limit allowed by US law, whereas Cd is 1.4 times greater.

For specific migration of these metals, Brazilian law stipulates limits of 0.005 $mg.kg^{-1}$ and 0.01 $mg.kg^{-1}$ for Cd and Pb, respectively (Brasil 2010a, b).

For Pb, mass fractions migrating to simulant solutions were 187 times greater than the legal limit for acetic acid and 13 times greater for water. The levels of Cd exceeded permitted limits by 50.2 and 2.4 times for acetic acid and water, respectively. The results for the specific migration tests are shown in Table 6.

Even when water was employed as the simulant solution, over 90% of samples exhibited specific migration values that exceeded the legal limit for Pb. For cadmium, migration values exceeded the limit in 54.55% of samples.

### Specific migration tests for Pb—commercial beverages

The results of specific migration tests for lead in cola, vinegar, orange juice, and milk are given in Table 7 below.

Based on the area of samples (1 $cm^2$), for orange juice, Pb migration proved 28.74% higher than the legal limit. For the total surface area of the utensil, this value represents a migration level 380 times greater than permitted limits. The specific migration levels of Pb were above permitted limits for all the beverages tested.

**Table 6** Cd and Pb mass fraction ($mg.kg^{-1}$) for specific migration tests in plastic utensils exhibiting highest levels in loco ($n = 11$)

| | Acetic acid | Water |
|---|---|---|
| ***Pb*** | | |
| Mean | 1.87 | 0.13 |
| Minimum | < LOD | < LOD |
| Maximum | 5.46 | 0.44 |
| Migration limit | 0.01 | 0.01 |
| % samples > migration limit | 90.90 | 90.90 |
| ***Cd*** | | |
| Mean | 0.251 | 0.012 |
| Minimum | 0.004 | < LOD |
| Maximum | 1.505 | 0.041 |
| Migration limit | 0.005 | 0.005 |
| % samples > migration limit | 90.91 | 54.55 |

*LOD < 0.001 $ug.kg^{-1}$

## Discussion

To the best of our knowledge, this is the first study investigating colored plastic food utensils intent to use by children as a potential source of Cd and Pb exposure. In the present study, the results showed that high mass fractions of these metals determined by XRF had major potential for specific migration into simulant solutions and commercial beverages. Similar studies have been conducted involving plastic packaging for storing foods. Kiyataka et al. (2014) detected Pb levels of 30–40 $mg.kg^{-1}$ using specific migrations tests, whereas cadmium concentrations proved below the limit of detection for the method. Furthermore, Hurip and Yeni (2021) performed chemical analyses for Pb and Cd in three different polypropylene samples and did not detect specific migration of these elements as a result of the methods adopted, even using a more concentrated acetic acid (4%) simulant solution, which highlights the importance of this type of study.

Of the 674 items analyzed in the present study, 287 contained detectable values of the metals (Table 1) and, of this total, 87 had levels which exceeded permitted limits. It is important to emphasize, however, that while levels in some items tested were within allowed limits, this does not rule out the possibility of samples containing some degree of contamination by Cd, Pb, or other PTE not measured in this study.

The XRF results (Tables 1–4) were cause for concern, revealing the mean Cd and Pb levels far exceeding limits allowed by US and Brazilian law (Brasil 2008; U.S Consumer Product Safety Commission 1997; World Health Organization (WHO). 2011). A number of studies have been carried out on plastic toys as a source of exposure. Greenway and Gerstenberger (2010) evaluated lead contamination using XRF in toys from 50 daycare centers in the USA. The results showed around 5.4% contained concentrations that exceeded legally allowed limits, with yellow items exhibiting the highest lead levels. The present study found the highest Cd mass fractions in green items (Table 3) and the

**Table 7** Pb migration mass fraction ($mg.kg^{-1}$) into simulants cola, vinegar, orange juice, and milk

| Sample | Migration mass fraction into simulant for sample area | Standard deviation | Migration mass fraction into simulant for total area of utensil |
|---|---|---|---|
| Coca-Cola | 0.0131 | 0.0003 | 1.43 |
| Vinegar | 0.0144 | 0.0003 | 1.57 |
| Orange juice | 0.0348 | 0.0020 | 3.80 |
| Milk | 0.0058 | 0.0002 | 0.63 |

highest Pb levels in orange and yellow utensils (Table 2). According to the National Institute of Metrology, Quality and Technology (INMETRO 2015), inorganic lead-based pigment can be used to confer paints yellow, orange, and red hues. These pigments represent a low-cost option compared to other types of pigmentation, perhaps explaining the high concentrations of these metals in colored utensils. Otherwise, according to the European Chemical Agency, cadmium pigments have been used in almost 70% of plastics found in Europe since the 1980s. These pigments (cadmium zinc sulfide yellow, cadmium sulfoselenide red, and cadmium sulfoselenide orange) are responsible for the bright and long-lasting coloration seen in the materials (European Chemical Agency, 2012). Another important result is about the source of pigments that may provide the high standard deviations observed in Table 4. One hypothesis for this variation may be related to the manufacturing methods of these plastic utensils or even be related to recycling processes. In these cases, higher concentrations can be observed in colors such as orange as a result of the mixture between pigments referring to the colors yellow and red that may present higher concentrations of these PTEs. Furthermore, utensils made of nonpigmented plastics (e.g., transparent plastic) and white plastic have lower concentrations not only of Pb but also of other toxic metals such as Cd and Hg than colored ones as shown in previous studies (Kiyataka et al 2014; Hurip and Yeni 2021).

Although the highest percentage of utensils exceeding the legal limit for lead (22.8%) were found in the district of Pari, the highest mean Pb was found in the city of Osasco, where both these regions had similar socioeconomic characteristics according to their IPVS. The ANOVA results showed a statistically significant difference between groups 2 and 3 ($p = 0.006$). This finding was expected, given that mean Pb values for the group 2 regions of Osasco and Pari were 4 times higher than those detected in the group 3 districts of São Miguel and Capão Redondo (group 3), despite the higher IPVS of the latter. Other factors to take into account include the low cost of the utensils and the location in which they were purchased, constituting major commercial hubs of Greater São Paulo. Socioeconomic aspects such as educational level, income bracket, besides other factors, are known to be directly associated with a host of chemical exposure scenarios (Carvalho et al 2017). Shoppers often travel to these commercial centers to buy this type of product and thus exposure to these Pb- and Cd-contaminated items extends beyond the local population.

The highest migrations were detected for acetic acid 3% solution (m/v), with 99.9% of utensils exhibiting specific migration that exceeded permitted limits for both Cd and Pb (Table 6). Whitt et al. (2016) investigated a specific migration of heavy metals from recycled polyethylene terephthalate using deionized water as the simulant solution. The levels of Cd and Pb which migrated into the simulant proved below the limit of detection of the method. In the present study, levels of migration of Cd exceeded allowed limits for 54.55% of the utensils tested, with an even higher proportion for Pb, at around 91% of items tested. All of the migration test results using simulant solutions exceeded the allowed levels established by Brazilian law of 0.005 mg.kg$^{-1}$ and 0.01 mg.kg$^{-1}$ for Cd and Pb, respectively. No contamination was detected in the analytical blanks, thus confirming the major potential bioaccessibility of the metals previously quantified in loco using XRF analyses (Table 5).

The migration tests on consumable beverages were performed for Pb only. The results found for these tests were even more alarming (Table 7). For orange juice, migration values were 360 times higher than the legally allowed limit, while all the other beverages also exceeded migration limits on tests for sample fragment area and overall area of both cups and mugs. These beverages were chosen because they are routinely consumed by children using the utensils in both home and school environments, where children can spend up to 10 h a day at daycare centers (Olympio et al. 2018). Notably, the analyses of commercial beverages showed they contained Pb and Cd levels which were below the limit of detection.

Many authors have implicated plastic objects as potential sources of exposure. Turner (2018) analyzed around 200 toys made of recycled plastics in the European Union and found high levels of PTEs in the base material. In a study conducted in Kazakhstan, Akimzhanova et al. (2020) found levels of lead contamination in children's plastic jewelry of 50 mg.kg$^{-1}$. Guney and Zagury (2013) investigated the bioaccessibility of both Pb and Cd in plastic toys and found values in the 642–647 mg.kg$^{-1}$ range for Pb and 1.31–1.34 mg.kg$^{-1}$ for Cd. In a Chinese study of specific migration of lead in plastic toys, Kang and Zhu (2015) found a level of 95 mg.kg$^{-1}$. Cui et al. (2015), in a study of bioaccessibility of Pb in toys from the Chinese market, found levels of 3.19 mg.kg$^{-1}$ for Pb and 0.86 mg.kg$^{-1}$ for Cd. All of the above-cited studies, consistent with the present investigation, confirmed plastic toys and jewelry as a potential source of Cd and Pb exposure, as well as domestic utensils routinely used for feeding children.

The International Agency for Research on Cancer (IARC 1993) classifies Cd as a substance carcinogenic for humans whose childhood exposure is associated with adverse health effects in adult life, such as renal problems, osteoporosis, and lung cancer and, also, to the immunosuppressive effects (Schoeters et al. 2006). Moreover, the United States Centers for Disease Control (CDC) has established a blood lead reference value of 3.5 μg.dL$^{-1}$, calculated according to the 97.5th percentile based on the National Health and Nutrition Examination Survey (NHANES) (Advisory Committee for Childhood Lead Poisoning Prevention 2012). However, the toxic effects of lead can be seen in children at concentrations lower than this reference, with toxicological effects related to learning difficulties, attention deficit, low intelligence quotient, and antisocial behavior (Bellinger 2004; Needleman 2004; Lanphear et al 2005; Hornung et al. 2009; Mazumdar et al. 2011; Dickerson et al. 2016; Blackowicz et al. 2016; Wagner et al. 2017, Olympio et al. 2009; Olympio et al. 2010; Olympio et al. 2017). According to Paulson and Brown (2019), this value is in the process of being revised to the lower limit of 3.5 $\mu g.dL^{-1}$. Plastic utensils are one of many sources and routes of exposure for children. In a study of 50 daycare centers with 2397 children, the authors found blood lead levels of around 2.16 $\mu g.dL^{-1}$ and a 97.5th percentile of 13.9 $\mu g.dL^{-1}$ (Olympio et al. 2015, 2018). In a later investigation involving a subsample of the Olympio et al. (2018) study, Silva et al. (2018) explored potential sources of lead exposure, identifying sources that included paint coatings of walls, floors, doors, and windows, as well as plastic toys and utensils handled by children at daycare centers and within their homes. Other studies have investigated the associations of blood lead levels with exposure of children aged 1–4 years by analyzing 24 h diet in school and household settings. The results showed Pb levels of 2.71 $\mu g.dL^{-1}$ in blood, 1.61–2.24 $\mu g.kg^{-1}$ in the diet, and bioaccessibility for Pb, Cd, and As of $0.18 \pm 0.11$ $\mu g.kg^{-1}$, $0.08 \pm 0.04$ $\mu g.kg^{-1}$, and $0.61 \pm 0.41$ $\mu g.kg^{-1}$, respectively (Leroux et al. 2018a, 2018b). In addition, another route of exposure to these metals occurs in the form of informal home-based work. For example, in the production of jewelry and fashion jewelry in the city of Limeira, São Paulo state, children or other members of the household are exposed to elements such as Pb, Cd, and As, among others (Ferrreira et al. 2019; Pereira et al. 2020; Salles et al. 2021).

Therefore, there is a clear need for further studies investigating plastic utensils for use by children as a source of exposure not only to Cd and Pb but also to other potentially toxic elements (PTEs) these materials may contain. Moreover, as outlined previously, contamination of these materials by Cd and Pb is not a problem specific to Brazil but an issue documented in many other countries including the USA, China, and Kazakhstan (Guney and Zagury 2013; Kang and Zhu 2015; Cui et al. 2015; Turner 2018; Akimzhanova et al. 2020). In the present study, a large proportion of the plastic utensils tested were imported, predominantly from China, a major trading partner for Brazil.

As a limitation of this study, only the simulant solutions of water and acetic acid 3% (m/v) were employed in the present investigation. Official Brazilian guidelines recommend that, for fatty foodstuffs such as milk, ethanol-based solutions be used at the appropriate mass fractions according to the type of food under analysis. It is important to point out, however, that the regulatory body states that the use of acetic acid 3% (m/v) as a simulant solution suffices for performing migration tests. Another important point is related to the different concentrations of Pb and Cd within the districts themselves, which may bring interesting and more in-depth results regarding the chemical exposure for these contaminants. The temperature at which experiments are conducted is crucial and the suggested temperatures are provided by the legislation and EN standards cited. Another limitation of this study was that only Pb and Cd were considered as contaminants of these plastic utensils and future chemical analysis should be performed considering other PTEs such as arsenic, chromium, mercury, and organic compounds.

## Conclusions

The Cd and Pb values determined are cause for concern, given the vast majority of plastic utensils assessed contained levels exceeding permitted limits. Furthermore, the results of the migration tests evidenced the major potential bioaccessibility of these elements in foods and beverage, potentially leading to harmful health effects even at low levels, especially when children are being considered the target group for exposure to these PTEs, as their biological systems are developing. It is important to note, however, that the general population is also exposed to these sources of Cd and Pb, since they also make use of these plastic utensils.

Taken together, these results underscore the urgent need for stricter regulations and inspections governing the production of these materials, from the manufacturing process through to their use, where orange, yellow, and green pigments were associated with the highest levels of Cd and Pb, probably due to the pigments used to give the paints color, brightness, and durability. Therefore, the use of colorless/transparent utensils is recommended as a means of mitigating exposure to these elements via this source, in that these elements are invariably employed to bind color to the plastic material.

As shown, the level of social vulnerability of the districts where utensils were purchased suggests a relationship between price and levels of Cd and Pb in the utensils. However, it is important to bear in mind shoppers tend to gravitate to commercial centers to buy these utensils and goods at low prices. Therefore, it stands to reason those locations which are a source of high concentrations of these elements may also pose a risk to populations living outside these districts.

Lastly, future studies systematically assessing other PTEs such as arsenic, a highly toxic substance with a range of health effects, using controlled migration tests are warranted. These other contaminants are also part of the WHO list of 10 chemicals of major public health concern.

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**Data availability** All data generated or analyzed during this study are included in this published article (and its supplementary information files).

## Declarations

**Ethics approval and consent to participate** Not applicable, as this study did not include people or animals.

**Consent for publication** Not applicable.

**Competing interests** The authors declare no competing interests.

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