Metals in paints on chopsticks: Solubilization in simulated saliva, gastric, and food solutions and implication for human health

Di Zhao\textsuperscript{a}, Albert L. Juhasz\textsuperscript{b}, Jun Luo\textsuperscript{a}, Hong-Bo Li\textsuperscript{a}, Lena Q. Ma\textsuperscript{a,c}

\textsuperscript{a} State Key Laboratory of Pollution Control and Resource Reuse, School of the Environment, Nanjing University, Nanjing 210023, People’s Republic of China
\textsuperscript{b} Future Industries Institute, University of South Australia, Mawson Lakes, South Australia 5095, Australia
\textsuperscript{c} Soil and Water Science Department, University of Florida, Gainesville, FL 32611, United States

\begin{abstract}
Paints are often used on chopsticks, however, a paucity of studies has assessed metals in the paints and the associated health risk. In this study, total Pb, Cd, Cr, Co, and Ni concentrations in paints were assessed for 72 wood and 29 stainless steel chopsticks with different colors while metal solubilization from paints in simulated saliva, 0.07 M HCl, and 1% citric acid solutions was measured for 9 samples having total Pb > 90 mg kg\textsuperscript{-1}, representing exposure scenarios of mouthing, incidental paint ingestion, and metal migration in food. Results showed that Pb (0.12–500,000 mg kg\textsuperscript{-1}), Cd (0.002–120,000 mg kg\textsuperscript{-1}), Cr (2.2–8400 mg kg\textsuperscript{-1}), Co (0.004–2600 mg kg\textsuperscript{-1}), and Ni (0.10–150,000 mg kg\textsuperscript{-1}) concentrations varied considerably among paint samples. Most samples showed low metal concentrations, however, high metal concentrations were observed in red and green paints on stainless steel chopsticks, while paints on wood chopsticks showed lower metal concentrations.

Estimation of daily metal intakes incorporating metal solubilization data in saliva, 0.07 M HCl, and 1% citric acid solutions suggested that the health risk via saliva contact was negligible, while unacceptable health risk was observed for Pb and Cd via their solubilization in 0.07 M HCl and 1% citric acid solutions from paints with high Pb and Cd concentrations. To avoid Pb and Cd exposure, use of metal-based paints on chopsticks should be banned, particularly in countries where food is regularly consumed with chopsticks.

\end{abstract}

1. Introduction

Human exposure to metals including Pb, Cd, Cr, Co, and Ni is associated with health outcomes including anemia, brain damage, osteoporosis, and cancers (ATSDR, 2012; Danadevi et al., 2003; Gallagher et al., 2008; Juric et al., 2017; Navas-Acien et al., 2007). With phasing-out of leaded gasoline and strict control of metals in food and drinking water, increasing attention has been paid to the use of metal-containing paints in various items including toys, jewelry, and interior walls (Guney and Zagury, 2013; Mielke and Gonzales, 2008; Weidenhamer and Clement, 2007). It is known that Pb pigments including 2PbO·PbO\textsubscript{2} (red), PbCO\textsubscript{3}·Pb(OH)\textsubscript{2} (white), PbCrO\textsubscript{4} or PbCrO\textsubscript{4}/PbSO\textsubscript{4} (yellow), and PbCrO\textsubscript{4}/PbSO\textsubscript{4}/FeNH\textsubscript{4}Fe(CN)\textsubscript{6} (green) are commonly used in paints being reported in China (Lin et al., 2009), with Pb concentrations > 150,000 mg kg\textsuperscript{-1} in enamel paint being reported in China (Lin et al., 2009).

Numerous studies have measured metal concentrations in paints used on toys, jewelry, and house walls (Cui et al., 2015; Greenway and Gerstenberger, 2010; Kumar and Gottesfeld, 2008; Mateus-Garcia and Ramos-Bonilla, 2014), showing Pb concentration up to 390,000 mg kg\textsuperscript{-1} in paint fragments from exterior walls in England (Turner and Sogo, 2012; Turner et al., 2016). However, a paucity of studies has assessed whether the paints on chopsticks contain harmful metals and pose risk to human health during their daily use. Chopsticks are traditional utensils to eat food in East Asia that have been used for > 6000 years. Most Chinese people use chopsticks 3 times a day during meals. Due to recent popularity of Chinese food, chopsticks are becoming increasingly popular in many countries including Japan, Korea, Thailand, Vietnam, and even Europe and America. Chopsticks are often made of wood, bamboo, stainless steel, and plastic. To provide decoration, reduce corrosion, facilitate drying, and inhibit bacterial growth, paints are often used on chopsticks. However, paints on chopsticks could directly contact with foods during eating, leading to metal migration from chopstick paints into foods, thereby contributing to human metal exposure. Since Pb-based paints are still available in many countries (Clark et al., 2015; Shu et al., 2015), it is possible that paints used on chopsticks contain toxic metals, posing potential health risk to humans
During daily use of chopsticks.

Determination of total metal concentrations in chopstick paints provides a rough assessment of health risk associated with their use, as not all metals in chopstick paints can enter into human body and be absorbed. For those who use chopsticks, one important exposure scenario is that metals may migrate from chopstick paints into food solution during chopsticks use, resulting in metal exposure via food consumption. There have been procedures measuring metal migration from tableware into simulated food simulants (Demont et al., 2012; Dong et al., 2014; BS EN 13130-1:2004). Acid food simulants have been used to test metal migration from ceramic food packaging materials into food, e.g., 1% citric acid (Dong et al., 2014). Other exposure scenarios include saliva contact with chopstick paints and incidental ingestion of paint fragments. Simulated saliva and gastrointestinal fluid extraction have been used to refine health risk assessment of metals in solid matrices, representing mouthing and ingestion pathways (Wragg et al., 2011). However, metal solubilization from paints on chopsticks into saliva, gastric, and food simulants has not been reported, which is critical to understand the health risk of using chopsticks with colorful paints.

Therefore, the aim of this study was to assess whether metals in paints on chopsticks could pose a risk to human health during their daily use. Two types of chopsticks were purchased from the Chinese market, i.e., wood and stainless-steel chopsticks. The specific objectives were to: (1) determine total concentrations of Pb, Cd, Cr, Co, and Ni in paints on 101 chopsticks varying in types (wood and steel) and paint colors; (2) measure metal migration potential in simulated saliva, gastric, and food simulants from chopstick paints; and (3) characterize health risk of the chopsticks with incorporation of metal solubilization data. To our best knowledge, this is the first comprehensive study to investigate the health risk of metals in chopstick paints. Results of this study provide scientific guidelines for the general populations to select and use safe chopsticks.

2. Materials and methods

2.1. Chopstick samples and total metal concentration analyses

To investigate the health risk associated with metals in paints coated on chopsticks, 29 samples of stainless steel and 72 samples of wood chopsticks painted with different colors of paints were purchased from retail stores across China via the Internet (Table S1). All the samples were produced in China, from different location including Guangdong (Dongguan, Guangzhou, Shenzhen, and Jieyang Cities), Jiangxi (Yichun), Zhejiang (Yiwu, Lishui), and Shandong (Baqiao) Provinces. For each sample, at least 4 pairs were obtained with each pair being used for analyses of total metal concentration in paints and metal solubilization from paints in simulated saliva, gastric, and food solutions. These chopstick samples represented those commonly used by the general public of China. Average length of the chopsticks was ~ 23 cm, with a total surface area of ~ 53 cm². Paints were often coated on the chopsticks held in the hand (~ 1/3 of total surface area), however, there were also chopstick samples with all surface being covered with paints.

From chopstick surfaces, paints were peeled off using an acid-cleaned ceramic knife and digested using concentrated HNO₃ and H₂O₂ based on USEPA Method 3050B. Concentrations of Pb, Cd, Cr, Co, and Ni in digests were determined using inductively coupled plasma mass spectrometry (ICP-MS, NexION300X, PerkinElmer, USA) following dilution of the digests using 0.1 M high-purity HNO₃. These elements are among the widely reported metals in paints (Clark et al., 2015; Turner et al., 2016).

2.2. Metal dissolution in simulated saliva fluid

To assess the health risks, solubilization of metals from paints into simulated saliva was assessed for 9 chopstick samples having total paint Pb > 90 mg kg⁻¹ (Table 1), representing the possible exposure scenario of chopstick contact with mouth. Saliva extraction was proposed by the US Consumer Product Safety Commission to simulate exposure via mouthing behavior (U.S. CPSC, 1997). Among the 9 samples, 8 were stainless-steel and one was wood chopstick. For the wood chopstick, the mouth end was painted with red paint, while for the 8 stainless-steel chopsticks, the mouth end was varnish. However, the painted section of the stainless-steel chopsticks was also assessed for metal solubilization in saliva since children may suck the chopsticks during eating, leading to metal exposure via saliva contact.

Briefly, simulated saliva (pH 6.5 ± 0.05) was prepared following the UBM method (Wragg et al., 2011). The assay is a physiologically-based test that mimics mouth-stomach-intestine conditions. For the procedure, 40 mL of saliva fluid was added into a 50-mL centrifuge, then the painted section of a chopstick (length: 7.5 cm, surface area: 17.3 cm²) was immersed in the saliva fluid. The extraction was kept at 37 °C for 6 h (U.S. CPSC, 1997; Weidenhamer et al., 2011). At the end of extraction, the solution was centrifuged, filtered (0.45 µm), and diluted with 0.1 M high-purity HNO₃ prior to analysis using ICP-MS. The long extraction time of 6 h was recommended by U.S. CPSC (1997) for metal-containing jewelry. In addition, metal solubilization during the one-time, long extraction period could be regarded as the potential maximum mass of metals that could be solubilized when chopsticks are repeatedly used, providing a conservative estimate of health risk. Also, this could overcome the difficulties in measuring metal solubilization from chopsticks when they are repeatedly used.

2.3. Metal dissolution in simulated gastric fluid

During chopstick use, paint may peel off from chopsticks and therefore be incidentally ingested, particularly for children who may suck the chopsticks during eating. In this case, metals in paint may be dissolved in acidic gastric fluid and become bioavailable. Therefore,

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| Table 1 Mean values of total metal concentrations in paints (mg kg⁻¹) and dissolved metal concentrations (mg L⁻¹) in 40 mL of artificial saliva and 0.07 M HCl solutions from paints for selected chopsticks with total Pb concentrations in paints exceeding 90 mg kg⁻¹. Standard deviations of the data are provided in the Supplementary data.
metals in paints of the 9 chopstick samples with elevated metal concentrations were also extracted with 0.07 M HCl (Table 1), a simulated gastric fluid (pH = 1.8). It is a protocol recommended by the European Standard on Safety of Toys for migration of elements (EN 71-3) (Güney and Zagury, 2014). In addition, it is a Chinese standard method GB/T 23994-2009 to determine bioaccessible metals from the coating of consumer products.

Briefly, painted section (length: 7.5 cm, surface area: 17.3 cm²) of a chopstick was immersed into 40 mL of 0.07 M HCl in 50-mL centrifuge tubes, which were maintained at 37 °C for 6 h after which metal concentrations in extracts were determined using ICP-MS. The 6 h of extraction was used to provide a conservative assessment of metal release in simulated gastric fluid (U.S. CPSC, 1997; Weidenhamer et al., 2011).

### 2.4. Metal migration in acid food simulant

Two chopsticks, i.e., #96 and #101 that showed the highest Pb concentration in paints among wood and stainless-steel chopsticks (9300 and 50,000 mg kg⁻¹) were assessed for metal migration from paint in simulated acid food composition, i.e., 1% citric acid (Dong et al., 2014).

Due to different eating habits, individuals may vary in the time used to eat. Therefore, to determine the influence of contact time on metal migration from chopstick paints in foods, kinetics of metal migration in 1% citric acid was determined. Briefly, acid food simulant of 1% citric acid solutions was prepared, with 40 mL being added into a 50-mL centrifuge tube. Then the painted section (length: 7.5 cm, surface area: 17.3 cm²) of chopsticks #96 and #101 was immersed in the food. The extraction was kept at 40 °C over a period of 360 min to study the metal release kinetics. At the time intervals of 5, 10, 20, 30, 60, and 360 min, 1 mL of extract was pipetted, centrifuged, filtered, and diluted prior to analyses using ICP-MS.

In addition to the different extraction time, food temperature may have a significant influence on metal migration from chopstick paints. Therefore, metal migration from paint of chopsticks #96 and #101 in 40 mL of 1% citric acid at 20, 40, and 60 °C over 20 min was also investigated. The extraction period of 20 min was used since it was close to the average time for meals. Extracts were processed and analyzed similarly as above.

### 2.5. Health risk assessment

Following assessment of metal dissolution in simulated saliva, gastric, and food solutions, daily intake values (DI) of Pb, Cd, Cr, Co, and Ni associated with chopstick use were calculated for adults with body weight (bw) of 60 kg under 3 scenarios, i.e., mouthing (DImouth), ingestion (DIngest), and metal migration in citric acid solution (DIconc). However, it should be noted that being different from the one-time dissolution assessed by this study, metals in chopstick paints might be actually gradually released and exposed to humans during the whole serving life of chopsticks. Therefore, it was assumed that the amount of metals dissolved in the simulated saliva, gastric, and food solutions assessed would be gradually exposed to adults over the service life of chopsticks.

\[
\text{DI}_{\text{saliva}} = \frac{\text{DM}_{\text{saliva}} \times 2}{\text{LL} \times \text{BW}} \\
\text{DI}_{\text{ingestion}} = \frac{\text{BA}_{\text{HCI}} \times 2}{\text{LL} \times \text{BW}} \\
\text{DI}_{\text{citric}} = \frac{\text{DM}_{\text{citric}} \times 2}{\text{LL} \times \text{BW}}
\]

where, DMsaliva, DMHCl, DMcitric (μg) are the dissolved metal mass solubilized from chopstick paints in 40 mL of saliva, 0.07 M HCl, and 1% citric acid fluids, respectively. LL is the life length of chopsticks, and BW is the mean body weight of adults at 60 kg (China Environmental Press, 2013). A parameter of “2” was included because chopsticks are used in pair. In this study, it was assumed that the service life of chopsticks was 3 months, i.e., 90 d. For DIcalculation, metal release at 40 °C at the time interval of 360 min was used, proving a conservative estimation.

The calculated DI were compared to provisional tolerable daily intake values (PTDIs) of 1.4, 0.5, 0.6, 5.0, and 10.0 μg kg⁻¹ d⁻¹ for Co, Cd, Pb, Cr, and Ni (RIVM, 2001; JECFA, 2011).

### 2.6. Quality assurance and quality control

Total metal concentration and metal solubilization from chopstick paints in simulated saliva, 0.07 M HCl, and 1% citric acid were determined in replicates (n = 2) using a pair of chopsticks. During measurement of metal concentrations in chopstick paints, a paint standard reference material (SRM NIST 2581, Pb-based powdered paint) was included. The accuracy of the digestion method USEPA 3050B was ascertained by consistency between measured (4272 ± 64.1 mg kg⁻¹, n = 3) and the certified Pb concentration (4490 ± 110 mg kg⁻¹) in the SRM. Besides Pb-based powdered paint, quality assurance and control were conducted using soil (SRM 2711a, NIST). The accuracy of metals was assessed with measured Cd, Cr, Co, and Ni concentrations of 52.7 ± 2.3, 53.9 ± 1.1, 9.72 ± 0.11, and 22.1 ± 0.7 mg kg⁻¹ in NIST 2711a (54.1 ± 0.5, 52.3 ± 2.9, 9.89 ± 0.18, and 21.7 ± 0.7 mg kg⁻¹). The ICP-MS detection limits of Pb, Cd, Cr, Co, and Ni calculated as 3 times the standard deviation of the blank values were 0.001–0.071 μg L⁻¹. Metal concentrations in blanks from both the digestion and extraction experiments were below or close to detection limit. For saliva, gastric, and citric acid extraction, Pb, Cd, Cr, Co, and Ni (10 μg L⁻¹) were spiked into the extraction blank solutions, producing spike recovery rates of 95–102%. In addition, during analyses using ICP-MS, spike and check standards (1–10 μg L⁻¹) were included every 20 samples, with recoveries being 102 ± 3.9% and 98 ± 9.1%, respectively (n = 30). Indium (m/z 115) was added into the samples, calibration standards, and blanks and used as internal standards to compensate for matrix effects and long-term signal drift produced by matrix components. The recovery of internal standards was within 90–110%. To avoid interference of extraction matrix, all extraction solutions were diluted with 0.1 M HNO₃ several times before metal concentration determination, so that the matrix effects could be neglected.

All data are presented as mean and standard deviation of 2 replicate analyses, with 2 significant figures. Statistical differences in metal concentrations between paints on wood and steel chopsticks and variation in metal concentration with paint color were assessed using variance analysis based on Tukey’s multiple comparisons using SAS (version 9.1.3). All graphs were created using SigmaPlot (version 12.5, Systat Software Inc., San Jose, CA, USA).

### 3. Results and discussion

#### 3.1. Metal concentrations in paints on chopsticks

In this study, 101 chopstick samples (72 wood and 29 stainless steel) were collected. The samples are numbered #1–101 based on increasing Pb concentration in paint (Table S1). Lead, Cd, Cr, Co, and Ni were detected in paints of all chopstick samples. However, concentrations of Pb (0.12–500.000 mg kg⁻¹), Cd (0.002–120.000 mg kg⁻¹), Cr (2.2–8500 mg kg⁻¹), Co (0.004–2600 mg kg⁻¹), and Ni (0.10–150,000 mg kg⁻¹) showed considerable variation across several orders of magnitude among samples (Figs. 1A and Table S1), suggesting varied quality of paints used on chopsticks in the market. In addition, large variations in metal concentrations in paints on chopsticks were observed between replicates (Table S2). This was related to the fact that paints used for metal measurements were peeled off from one pair of the chopstick, which may differ considerably in metal composition among different colors.
Paints on steel chopsticks showed (p < 0.05) higher metal concentrations compared to wood chopsticks (Fig. 2A). Lead concentrations in paints from the 72 wood chopsticks were 0.12–9300 mg kg\(^{-1}\), with a median value of 1.5 mg kg\(^{-1}\). However, Pb concentrations in paints from the 29 steel chopsticks were 2.7–500,000 mg kg\(^{-1}\), with a median value of 20 mg kg\(^{-1}\). Similarly, Cd, Cr, Co, and Ni concentrations in paint from steel chopsticks (median of 1.1, 1400, 21, and 170 mg kg\(^{-1}\), respectively) were (p < 0.05) higher than wood chopsticks (median of 0.18, 4.4, 0.04, and 0.36 mg kg\(^{-1}\), respectively). The data suggested that metal-containing paints were used more often on stainless steel than wood chopsticks. The reason behind this was unclear. However, using cheaper paints to lower the cost of stainless steel chopsticks may be a possibility. Notwithstanding, due to the higher metal concentrations in paints from steel chopsticks, higher health risk would be postulated from the use of stainless steel chopsticks.

With respect to paint color, metal concentrations varied substantially (Fig. 2B). Although Pb concentration varied considerably among samples having the same color, high Pb concentrations > 10,000 mg kg\(^{-1}\) were observed for samples with red and green paints, while samples with black, pink, white, and blue paints showed low Pb concentrations of < 20 mg kg\(^{-1}\). Similarly, high Cd concentrations (> 10,000 mg kg\(^{-1}\)) were observed in samples with red and green paints, while samples with black, pink, white, and blue paints showed Cd concentrations < 10 mg kg\(^{-1}\). Similar with Pb and Cd, the highest concentrations of Cr, Co, and Ni were also detected in red and green paints. This is consistent with a report that among 337 new household enamel paints from 11 countries, white paints contained the lowest Pb concentration while yellow, red, and green paints exhibited the highest concentration (Clark et al., 2009). Lin et al. (2009) showed higher Pb concentration in yellow, green, and red paints than those of other colors in 58 new house paint samples. The variation in metal concentrations with paint color on chopsticks may suggest the preferential use of metal pigments with red and green colors in paints on chopsticks. Different colors are observed for different Pb pigments including 2PbO·PbO\(_2\) (red), PbCO\(_3\)/Pb(OH)\(_2\) (white), PbCrO\(_4\) (yellow), and PbCrO\(_4\)/PbSO\(_4\)/Fe(NH\(_2\))\(_2\)CN\(_2\) (green) (Turner et al., 2016). Based on metal concentrations, chopsticks with red and green paints deserve more attention of potential exposure and health risk.

Our study was the first systematic report of metal concentrations in paints on chopsticks. Studies showed Pb concentration exceeding 10% in paint fragments from both interior and exterior walls in urban environments (Clark et al., 2015; Turner et al., 2016). A recent study showed that Pb was detected in 139 cases out of 197 analyses in decorated glassware for beverage consumption, with Pb concentration ranging from 0.04 to 400,000 mg kg\(^{-1}\) (Turner, 2018). It is known that Pb compounds such as PbCrO\(_4\), PbO, and PbCO\(_3\) are used as pigments in paints for desired colors (Kumar and Gottesfeld, 2008; Yost and Weidenhamer, 2008). In addition, Pb compounds are added to paint for durability. Compared to Pb-free paint, Pb-based paint is cheaper so its use is popular (Clark et al., 2014; Turner et al., 2016).

Previous studies have shown that Pb in paint fragments from outdoor structures in England was due to addition of PbCrO\(_4\) based on the molar ratio of Pb to Cr being ~ 1 (Turner et al., 2016). Our study on lipstick samples with Pb concentration > 7500 mg kg\(^{-1}\) also shows that Pb is present as PbCrO\(_4\) based on X-ray absorption spectroscopy (Zhao et al., 2016). However, in this study, the molar ratio of Pb to Cr in chopstick paints varied considerably from 0.01 to 100 (Fig. S3). Most chopsticks showed molar ratios of Pb to Cr < 1, while the ratios being ~ 20–100 in the chopsticks samples (#97–101) having Pb concentrations of 91,000–500,000 mg kg\(^{-1}\). The data suggested that Pb in chopstick paints was not from addition of PbCrO\(_4\). Other Pb pigments might be used, which require further investigation.

Beside Pb, Cd in paints is also a potential hazard, which needs more attention. A recent paper focusing on metals on decorated glassware found that Cd was detected in ~ 70% of all enamelled surfaces of 72 products, with the maximum Cd concentration being 70,000 mg kg\(^{-1}\).
In addition, high levels of Cd (> 10,000 mg kg\(^{-1}\)) were found in inexpensive jewelry items by X-ray fluorescence (Weidenhamer et al., 2011). Cadmium sulphoselenides and CdO are popular Cd pigments to produce a variety of colors including red, orange, and yellow (Lehman, 2002). Thus, it is becoming increasingly apparent that Pb and Cd are being used in a variety of consumer goods, posing health risks to people.

3.2. Metal migration from chopstick paints into saliva and 0.07 M HCl

Based on the 9 samples selected for assessing metal migration, solubilization of Pb, Cd, Cr, Co, and Ni in 40 mL of saliva was 0.001–0.13, < 0.001–0.25, 0.003–0.004, < 0.001–0.008, and < 0.001–0.002 mg L\(^{-1}\), being significantly lower than that of 0.001–760, < 0.001–130, 0.05–24, < 0.001–2.1, and 0.003–3.1 mg L\(^{-1}\) in 40 mL of 0.07 M HCl (Tables 1 and S2). This could be explained by higher potential of metal release in acidic solution from paints than in the pH neutral saliva, suggesting that compared to incidental ingestion, saliva contact may be less important for metals in paints on chopsticks. However, it should be noted that an acidic drink would considerably reduce the pH of saliva, leading a greater metal solubilization extent.

Compared to the guideline values of 0.01, 0.005, and 0.05 mg L\(^{-1}\) for Pb, Cd, and Cr in drinking water recommended by the Chinese standards (GB 5749-2006, 2006), metals dissolved into saliva and HCl solutions exceeded the limit for chopsticks with high metals including #98, #99, #100, and #101. Taken chopstick #99 for example, Pb concentration in saliva was 0.13 mg L\(^{-1}\), being ~ 13 times higher than the limit in drinking water. Whereas when HCl solution was used, dissolved Pb in the extraction solution was 760 mg L\(^{-1}\), about 5 orders of magnitude higher than the drinking water limit, suggesting unacceptable health risk.

Overall, metal solubilization in both saliva and 0.07 M HCl varied among samples depending on metal concentrations (Table 1). For example, Pb solubilization in 0.07 M HCl was 0.001–0.54 mg L\(^{-1}\) for the 5 samples (#89, 92, 94, 96, and 97) with Pb concentration of 100–91,000 mg kg\(^{-1}\) in paints, being significantly lower than that of 23–770 mg L\(^{-1}\) for the 4 samples (#98–101) with Pb concentration of 280,000–500,000 mg kg\(^{-1}\). This was also true for Cd solubilization in 0.07 M HCl, i.e., < 0.001–0.04 mg L\(^{-1}\) for samples #89, 92, 94, 96, and 97 and 2.4–130 mg L\(^{-1}\) for samples #98–101, suggesting that the major factor influencing metal solubilization in saliva and gastric fluid was total metal concentrations in paints.

However, when metal solubilization was compared between samples, factors other than metal concentration influencing metal solubilization were observed. For example, although chopstick sample #101 showed significantly higher total Pb concentration in paints than #99, Pb solubilization in both saliva and 0.07 M HCl solutions (0.11 and 440 mg L\(^{-1}\)) were significantly lower than that of 0.13 and 760 mg L\(^{-1}\) from #99 (Table 1). By assuming that the paint mass was ~ 0.1 g per chopstick, bioaccessible Pb in percentage in samples #96–98 based on HCl extraction was 0.24–3.3%, while for samples #99–101, bioaccessible Pb was significantly higher (36–86%). The inconsistency was more obvious when total Cr and Cr solubilization in 0.07 M HCl were compared between samples #96 and #101. Although...
sample #96 showed ~ 2-fold higher total Cr concentration (8500 vs. 3900 mg kg\(^{-1}\)) than #101, Cr solubilization in 0.07 M HCl from #96 was two magnitudes lower than that from #101 (0.20 vs. 24 mg L\(^{-1}\)). The inconsistence suggested that metal concentration in paints was not the only one factor influencing metal solubilization. Different species of metal compounds might be incorporated into paints coated on different chopsticks, leading to metal solubilization independent on total metal concentrations. This also highlighted the importance of determining specific metal solubilization to accurately assess the health risk of chopsticks.

### 3.3. Metal migration from chopsticks paints in 1% citric acid solution

In addition to saliva contact and incidental ingestion, metal release from chopstick paints to foods could occur during chopstick use. Therefore, the two samples, #96 and #101, which showed the highest Pb concentration in paints among wood and stainless-steel chopsticks, were selected for further analyses of metal migration in simulated food solution, i.e., 1% citric acid (Fig. 3). For both #96 and #101, the most soluble metal was Pb, followed by Cd, while release of other metals was quite low. In addition, significantly higher Pb and Cd migrations were observed from chopsticks #101 than #96, due to significantly higher concentrations of Pb and Cd in chopsticks #101 (500,000 and 120,000 mg kg\(^{-1}\)) than #96 (9300 and 220 mg kg\(^{-1}\)) (Table 1).

Extraction time was an important factor influencing metal solubilization in 1% citric acid. With increasing extraction time from 5 to 360 min, Pb dissolution at 40 °C from paints on #101 and #96 increased significantly from 140 to 1600 and from 0.07 to 0.95\(^{-1}\), while Cd dissolution increased significantly from 32 to 180 and from 0.005 to 0.17 mg L\(^{-1}\) (Fig. 3A, Table S3). This suggested increased health risk when chopsticks were in food solutions over a longer period.

In addition to extraction time, food temperature also significantly affected metal solubilization from paints on chopsticks. With the exception of Cr dissolution from #96, migration of Pb, Cd, Cr, Co, and Ni in 1% citric acid from the two chopsticks increased significantly with increasing extraction temperature (Fig. 3B, Table S3). For example, with increasing temperature from 20 °C to 60 °C, Pb dissolution from paints on #101 and #96 over 20 min increased from 240 to 1000 and from 0.11 to 0.24 mg L\(^{-1}\). This could be the result of lower diffusion activation energy and higher diffusion coefficient caused by high temperature, making metal dissolution from paint being faster, suggesting that chopstick contact with hot food could increase the associated health risk.

### 3.4. Health risk assessment

Metal migration data suggested that metals in chopstick paints may be released into saliva, gastric, and food compositions, possibly resulting in human exposure for adults who use chopsticks on a daily basis. Therefore, based on metal dissolution data, daily metal intake was calculated for adults with body weight of 60 kg under 3 scenarios, i.e., mouthing, ingestion, and metal migration into food (Fig. 4, Table S4). For all tested samples, solubilization of Pb, Cd, Cr, Co, and Ni in saliva resulted in metal daily intake several magnitudes lower than that in 0.07 M HCl and 1% citric acid, suggesting that compared to the exposure scenarios of incidental paint ingestion and metal solubilization...
in paints of all collected chopsticks samples, varying considerably among individual samples with most samples showing low metal concentrations. However, extremely high Pb, Cd, Cr, Co, and Ni concentrations up to 500,000, 120,000, 8500, 2600, and 150,000 mg kg\(^{-1}\) were observed in paints on some chopsticks, posing a potential health risk to humans. Estimation of daily metal intakes following assessing metal solubilization in saliva, 0.07 M HCl, and 1% citric acid suggested that Cr, Co, and Ni were not a health risk, while the extremely high Pb and Cd concentrations (360,000–500,000 and 38,000–120,000 mg kg\(^{-1}\)) in paints of several chopstick samples could lead to daily Pb and Cd intakes well above the PTDIs for adults. Interestingly, the extremely high Pb and Cd concentration was mainly observed from stainless steel chopsticks with red and green colors, while paints in wood chopsticks contained significantly lower metal concentrations. Therefore, the adverse health risk associated with Pb and Cd could be avoided with the recommendation of using painted wood chopsticks rather than the painted stainless-steel chopsticks. In addition, painted chopsticks should not be used in contact with hot food solutions over a long period, which could facilitate metal solubilization in food and result in higher metal exposure.

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### Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.envres.2018.07.036.

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