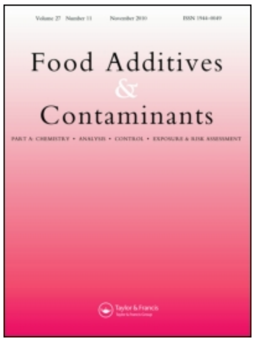

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Tea brewed in traditional metallic teapots as a significant source of lead, nickel and other chemical elements

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An environmental inquiry conducted by the Brussels Inter-communal Laboratory of Chemistry and Bacteriology (BILCB) has revealed that in 2000 a traditional metallic teapot caused in Brussels lead intoxication among a family of Morocco origin. Following this case study of lead poisoning and subsequent preliminary results carried out by the BILCB, which confirmed the dangerousness of this kind of item, samples of traditional metallic teapots were collected from North African groceries in Brussels by the Institute of Public Health (IPH) in collaboration with the BILCB and the Federal Agency for the Safety of the Food Chain (FASFC). Aluminium, copper, iron, nickel, lead and zinc were analysed to identify metals with a potential to migrate into tea solutions. Simulants (natural tea, tea acidified with citric acid and citric acid) were brewed in those teapots in order to identify the leaching potential of migration at boiling point temperature for different contact periods. Multi-elementary analysis was carried out by inductively coupled plasma-atomic emission spectrometry (ICP-AES). It was concluded that the concentrations of those leached metals depend on the nature of the migration liquids, the type of teapots and the contact periods. Most teapots showed a high level of toxic metals in leachates for lead and to a less extent for nickel, which can contribute significantly to the risk of serious poisoning. A comparison of the results with the toxicological reference values was done. The teapots were withdrawn from the market by the FASFC.

Keywords: metals analysis – ICP; food-contact materials; food simulants; migration; toxic elements; beverages

Introduction

It is well known that among Al, Cu, Fe, Ni, Pb and Zn analysed in the present study, Pb is the most toxic metal for the human body, particularly for children (Cleays et al. 2003). Since the time of antiquity lead poisoning has occurred in utensils used in food contact (Milton and Lesser 1988). Nowadays, cases of poisoning by this route are considered to be unusual (Wilson and Card 1986). With the exception of Petit et al. (2003), who reported a case study of Pb poisoning following the use of metallic teapots, most cases are identified for 'mugs' (Zuckerman et al. 1989; Ajmal et al. 1997; Ziegler et al. 2002), earthenware containers (Hellström-Lindberg et al. 2006) and ceramics (Hellström-Lindberg et al. 1997; Phan et al. 1998).

To our knowledge few studies have been dedicated to the origin of the migration process in metallic teapots. We have found in the literature only two studies: one studied Cu (Ni and Li 2008); the older study investigated Zn, Ni, Cu and Sn (Boularbah et al. 1999).

At the request of the Brussels Inter-communal Laboratory of Chemistry and Bacteriology (BILCB), the Scientific Institute of Public Health (IPH) made an environmental inquiry in Brussels where a case of Pb poisoning was reported (Petit et al. 2003) through use of a traditional metallic teapot.

A preliminary study made by the BILCB envisaged the leaching of Pb brewed in those kind of metallic teapots: the concentrations of Pb were in the order of a few hundred $\mu g\,l^{-1}$, but in some cases they could be much higher (up to about $3.5\,mg\,l^{-1}$), which confirmed the potential danger of using those items (Petit et al. 2003).

These items are commonly used by families from the Magreb or countries in Asia Minor. It is also possible to find these products in snack bars, tearooms or oriental restaurants willing to use traditional products when serving customers.

To the extent that they contain substantial amounts of Pb and other metals to known toxicities, the IPH took the initiative to investigate the presence and levels

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of Pb and other metals. These levels were confronted with the toxicological reference values. These results have led to market withdrawals made by the Federal Agency for the Safety of the Food Chain (FASFC).

Despite the particularly dangerous nature of these products, the European Commission has not established, and does not produce, specific guidelines on the subject. Therefore, the Council of Europe has taken the initiative to initiate a resolution on metals and alloys. This could therefore provide a legal basis for consolidated operations of public authorities.

The objective of this investigation was to study the migration of toxic metals from metallic teapots by using different simulants and to assess the related risks by comparing the results with the established toxicological safety limits.

Materials and methods

Sampling

Teapots samples (Figure 1) were collected from two places in Brussels well known for the sale of these products. Two shops were visited in each area and one sample of each model of teapots was bought (codes AS and RB). A total of nine items were purchased. It is sometimes difficult to determine the origin of these products, but these shops specialize in products from North Africa. Samples coded '1RB1' were collected in three identical copies in order to make a comparison between migration tests.

Another series of 11 teapots was provided by FASFC; those items, also bought in North African groceries, originated from India and Morocco.

Migration procedure

Teapots were cleaned before contact with different leaching media, washed with a detergent and rinsed with Milli-Q water, then dried at room temperature.

Three simulants were used as the leaching medium: tea infusions (prepared by boiling six bags in 1 L Milli-Q water; pH 4.86), citric acid (99.5% (m/m), Merck) prepared by dissolving 1 g in 1 L of Milli-Q water (pH 2.7); and the mixture of natural tea and citric acid ($1\,g\,l^{-1}$).

Teapots were filled with the leaching solution up to 1 cm from the top and placed on a hotplate to heat them. Aliquots of 2 ml were taken after 15, 30, 45 and 60 min of contact. These aliquots were then diluted to 10 ml. Before analysis, acidification or dilution with nitric acid 2% (v/v) was performed.

The aim of the choice of citric acid was made in the context of worst-case conditions. Therefore, the leaching media were used in place of acetic acid, which is generally chosen as a standard, for example in ceramic materials in contact with food (European Committee for Standardization 1995). This was decided because it is more realistic when simulating the action of lemon used when making some of our tea infusions. A comparison (for three identical pieces) from three migration solutions included: citric acid, $1\,g\,l^{-1}$ at pH 2.7, similar to commercial lyophilized tea (pH 2.95), tea, and tea acidified with citric acid, $1\,g\,l^{-1}$.

The use of citric acid helps standardize the acidity of the juice and corresponds to a current consumption pattern. This standardization would not be possible using real lemon. Citric acid helps, moreover, to explore the possibility of using a standard simulant in the laboratory and within the logic a worst-case scenario that prevails in Europe when establishing the compliance of a material or article intended for food contact (Official Journal of the European Union 2004). This simulant also avoids having to work with significant levels of blank when testing the migration. The concentration of some elements in tea also likely distorts the evaluation of results.

Digestion procedures

The teapots were first drilled with a drilling machine. Approximately 0.05 g of metal powder was weighed in a PFA digestion tube, then 3 ml HCl plus 1 ml HNO_3 were added. Afterwards, digestion was carried out at 180°C in an oven and evaporated to about 0.5 ml.

Figure 1. Teapots samples collected from the market in Brussels.

Each digested sample was transferred quantitatively into a 50 ml calibrated tube and set to volume with Milli-Q water.

ICP-AES determination of metals

A routine method for the determination of metals in migration liquid with inductively coupled plasma-atomic emission spectrometry (ICP-AES) was validated and accredited for several elements. The limit of detection (LOD), defined as three times the standard deviation of the blank, was less than $10\,\mu g\,l^{-1}$ for all elements analysed; within-laboratory reproducibility was less than 5%; and expanded measurement uncertainty was less than 15% for all element measured. An external calibration curve was carried out with the following standards: 25, 50, 100 and $200\,\mu g\,l^{-1}$; all standards were prepared by weighing solutions ($1\,g\,l^{-1}$; Fluka) in 2% (v/v) nitric acid (70% (w/w); J. T. Baker). The sample solutions were measured in triplicate (instrument programmed) by ICP-AES (Perkin-Elmer Optima 4300DV, Norwalk, CT, USA). Samples were diluted in nitric acid 2% (v/v) and re-analysed when the concentration appeared to exceed the highest concentration of the calibration curve. An internal control during sample analysis was performed; a check of the slope values and the correlation coefficient, analyse of the half-scale standard ($100\,\mu g\,l^{-1}$) after each ten samples measurement.

The concentrations of heavy metals were determined in each leaching solution, regarded as a procedural blank (the leaching medium was placed in a Pyrex baker), before the migration test. In the citric acid simulant all metal contents were less than the LOD of the method ($10\,\mu g\,l^{-1}$). For the natural tea used as a leaching solution, the predominant metals present were ($mg\,l^{-1}$): Al (4.2 ± 0.3), Zn (1.4 ± 0.2), Mn (0.93 ± 0.09), and Cu (0.050 ± 0.003). These concentrations were subtracted from the migrated values in the subsequent migration test with natural tea.

Table 1. Major composition of the teapots.

Teapot codes	Cu (%)	Zn (%)	Pb (%)	Fe (%)	Ni (%)	Al (%)
1 AS 1	53	38	4.9	2.1	1.4	0.2
1 AS 2	55	38	4.4	1.3	0.5	0.2
2 AS 1	54	38	5.5	1.2	0.8	0.9
2 AS 2	64	35	–	0.4	0.7	0.1
1 RB 1 a	64	35	0.1	–	0.2	
2 RB 2	66	33	–	0.6	0.1	–
2 RB 3	55	38	5.3	1.2	0.2	0.4
No. 1	52	37.1	6.7	1.7	0.5	0.4
No. 2	61	38	0.1	0.1	0.2	–
No. 3	54	35	4.5	4.3	1.1	0.3
No. 4	55	38	6.0	1.0	0.1	0.2
No. 5	54	39	4.8	1.7	0.4	0.3
No. 6	65	35	–	–	0.4	–
No. 7	55	37	4.6	2.7	0.4	0.3
No. 8	64	36	–	–	0.4	–
No. 9	63	37	–	–	0.1	–
No. 10	64	36	–	–	0.3	–
No. 11	60	34	5.7	–	0.8	–

Results and discussion

Major composition of the teapots

The composition of teapots was studied by dissolving a quantity of the alloys using aqua regia. A multi-elementary analysis was carried out by ICP-AES. This revealed that all teapots are made of brass alloy, mainly 59% Cu and 36% Zn, and that a majority of teapots were from leaded brass types because Pb is known for its excellent ductility. The presence of others metals such as Fe, Al and Ni at low contents was also noticed. Al and Ni are added for their corrosion resistance properties (Table 1). On the basis of these results, the migration of Cu and Zn followed by Pb, then Fe, Al and Ni was suspected in certain cases.

Comparison between simulants

Three identical teapots (codes 1RB1 a, b, c) were used for this comparison. Three simulants were used for the migration test: natural tea, citric acid, and a mixture of tea and citric acid for different contact periods (Table 2).

The effect of pH is an important factor in the conduct of a migration study. The addition of citric acid was chosen because it is the main acid that can be found in 'lemon tea' and its concentration corresponds to the concentration of citric acid that should be expected by adding a slice of lemon to a cup of tea. In this study we considered that one slice, corresponding to 3.3 ml of juice, is equivalent to approximately one-tenth of a lemon ($80\,g\,l^{-1}$ of citric acid) in a cup of 250 ml.

Leaching solutions, maintained at a constant temperature, were taken at different contact periods (15, 30, 45 and 60 min) in order to assess the migration kinetics with time.

Table 2. Pb, Ni and Zn concentrations in various migration liquids from a teapot for a contact time of 30 min at boiling temperature.

Teapot codes	Simulants	Pb ($mg\,l^{-1}$)	Ni ($mg\,l^{-1}$)	Zn ($mg\,l^{-1}$)
1 RB 1 a	Citric acid	71	18	3.3
1 RB 1 b	Tea	8	5.8	2.0
1 RB 1 c	Tea plus citric acid	45	13	3.8

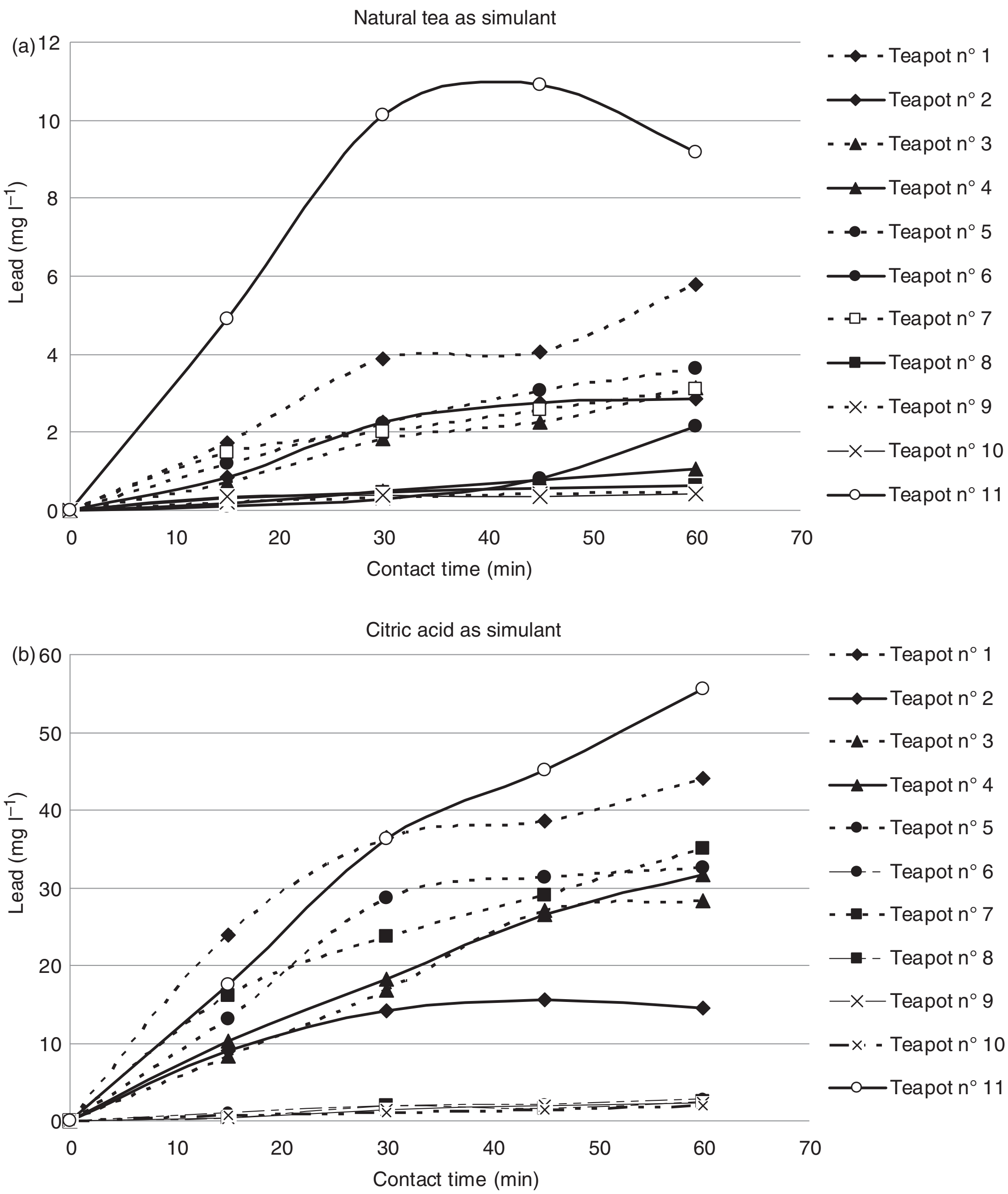

Figure 2. (a) Evolution of lead migration, from various teapots, in natural tea migration liquid with the contact time. (b) Evolution of lead migration, from various teapots, in citric acid simulant with the contact period.

The results listed in Table 2 concern only the prevalent leached metals such as Pb, Ni and Zn after a 30-min contact period with the leaching medium. Pb migrated more easily in these liquids compared with the other elements, and this migration is stronger in the presence of citric acid, which confirms that pH has a crucial part to play. It can be concluded that citric acid alone can be used as simulant in a worst-case scenario. Moreover, the obtained concentrations for the metals listed above are not lower than those obtained after contact with tea–citric acid, except for Zn. Furthermore, overvaluation (case of Pb and Ni) or underestimation (as in the case of Zn) does not lead to unrealistic estimates and does not exclude the results being beyond a range of 50%.

It has already been established that the use of these teapots (1RB1) leads to high concentrations of Pb, Ni and Zn, and this even after 15 min of contact. The use of lemon juice actually leads to a significant increase in migration levels, up more than 8–10 times in the case of Pb.

Kinetics of migration

Kinetic migration studies were carried out with teapot bodies, filled by leaching media such as tea infusion

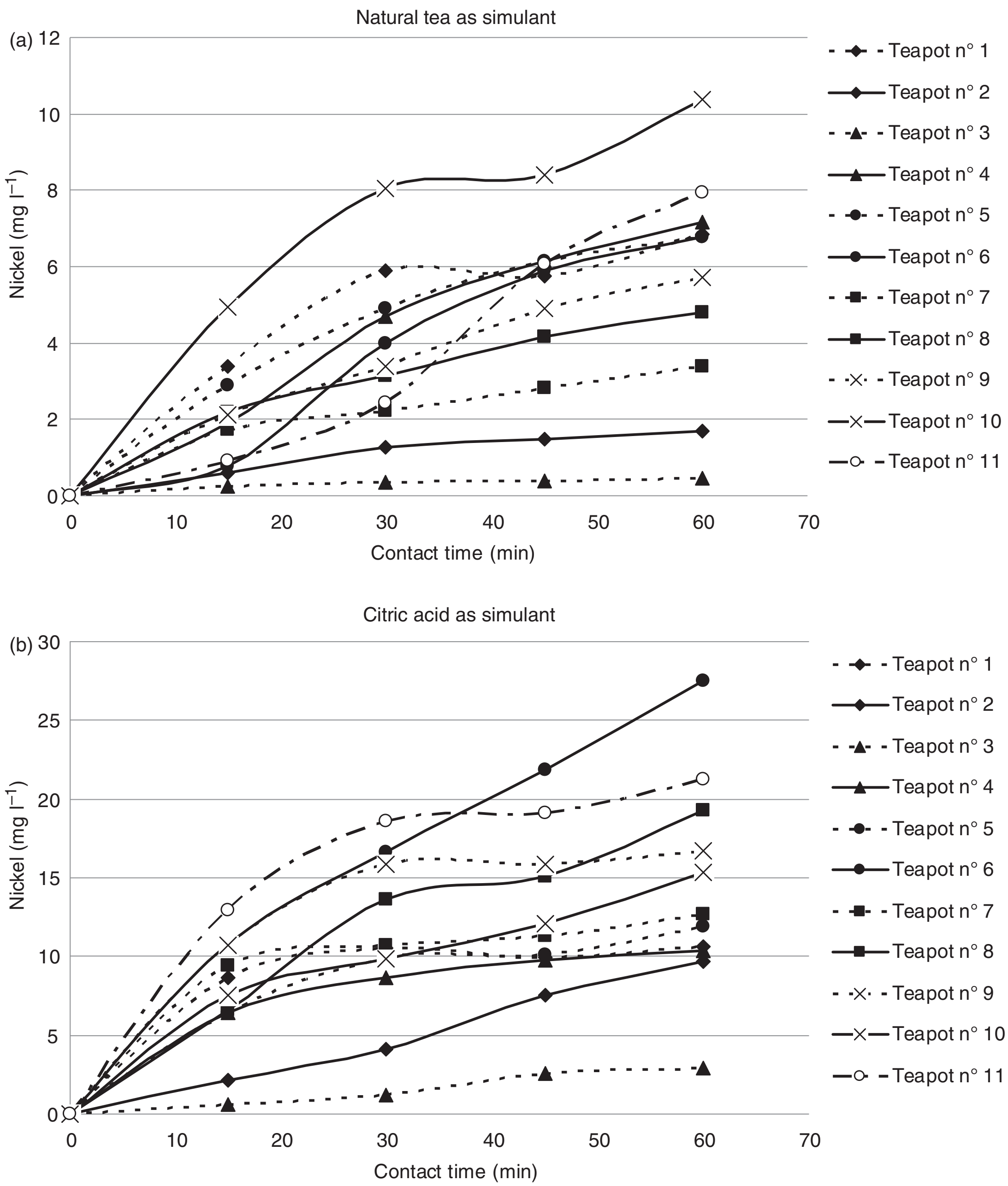

Figure 3. (a) Evolution of nickel migration, from various teapots, in natural tea migration liquid with the contact time. (b) Evolution of nickel migration, from various teapots, in citric acid simulant with the contact period.

and citric acid, at boiling temperature for 15, 30, 45 and 60-min contact period. Only the results concerning the migration of two toxic metals (Pb and Ni) are illustrated in Figures 2 and 3. For Pb and Ni, we observed a similar behaviour, for almost every teapot, both in tea-leaching solution and citric acid; the release rate is higher at the first migration period of approximately 15 min and fewer than 50% of metals were leached; thereafter the rate of release decreases significantly with time. This can be explained partly by gradual metal enrichment of the passive surface oxide layer, except, for example, for teapot no. 6 in the case of Ni and no. 11 in the case of Pb where the migration increased with time. Comparing the simulants, it appears that a high concentration of metals has been released with citric acid, which can be explained by the pH difference.

Metals analysis

Table 3 displays the Al, Cu, Fe, Pb, Ni and Zn concentrations found in teapot leachates obtained by using citric acid as a simulant for a contact period of 30 min. Others elements as Ag, As, B, Ba, Be, Bi, Cd, Co, Cr, Mn, Mo, Sb, Se, Sr and V were also analysed,

Table 3. Metals concentrations (mg l^{-1}) in leachates of citric acid from various teapots after a contact period of 30 min.

Teapot codes	Al	Cu	Fe	Pb	Ni	Zn
1 AS 1	0.23	0.36	2.3	34	35	7.7
1 AS 2	0.43	0.36	2.2	42	10	8.6
2 AS 1	–	–	0.74	18	10	0.9
2 AS 2	–	–	0.54	62	15	3.5
1 RB 1 a	–	–	0.6	71	18	3.3
2 RB 2	3.5	11	4.7	11	5.8	40
2 RB 3	0.33	–	–	42	–	–
No. 1	0.31	0.15	3.82	36.5	10.6	2.2
No. 2	0.19	17.1	0.52	14.1	4.1	14.9
No. 3	0.16	13.9	2.01	16.7	1.2	8.1
No. 4	0.07	2.2	1.05	18.2	8.7	1.7
No. 5	0.19	0.33	4.35	28.7	9.9	2.1
No. 6	0.10	1.1	0.31	2.0	16.6	1.5
No. 7	0.31	0.41	3.3	23.7	10.7	3.7
No. 8	0.52	8.05	0.4	2.0	13.6	30.0
No. 9	0.37	1.85	0.13	1.3	15.6	9.6
No. 10	0.35	18.0	0.2	1.1	9.9	19.8
No. 11	1.1	40.8	0.62	36.2	18.6	24.1

but they did not show any real interest at this stage of the study due to their negligible migration or their absence in teapot composition and to their high level in tea infusion. The suggestion made above about the metals at high content (a high migration of Cu and Zn) likely to migrate from teapots is not confirmed, except for the following teapots: no. 2, no. 11 for Cu, and nos 2RB2, 8 and 10 for zinc. All the teapot leachates examined contained high concentrations of Pb and Ni, then Zn or Fe and Cu; the presence of other elements such as Al depends on the type of teapot, and its highest leached concentrations were found for teapot no. 2RB2. Teapot 1RB1a released the highest concentration of Pb and teapot 1AS1 for Ni, in spite of the low content in these teapots. From these results it is noticed that the migration of metals did not match well with the content of these metals in teapots except for the cases quoted above. The migration phenomena of metals depends on the electrochemical properties of the elements, in spite of their low redox potentials (Al, −1.66 V; Zn, −0.762 V; Fe, −0.44 V); the corrosion of these elements is less than that of Pb and Ni. These can be explained by the formation of passivation layers or by the structure of the brass. Indeed, the alloy Cu–Zn presented a high corrosion resistance and only the added elements such as Pb and Ni are greatly affected by the corrosion due perhaps to their non-dissolution in the brass matrix.

Comparison of the results with toxicological reference values (TRVs)

The objective is to compare the obtained values with TRVs (mg day^{-1}): Cu, 5; Fe, 17; Pb, 0.214; Ni, 0.7; and Zn, 25. These values are those currently adopted by the Council of Europe to draft a resolution in metallic materials and alloys in contact with food for a person of 60 kg weight (Council of Europe 2010). The assumption is a daily consumption of four cups, each with a volume of 200 ml, of tea or natural tea with lemon (citric acid simulant), namely 800 ml day^{-1}. Here it only takes a contact time of 15 min.

Table 4. Comparison of the ingested lead, from natural tea, and citric acid after a contact period of 15 min compared with the toxicological reference values: hypothesis of four tea cups day^{-1} (800 ml).

Teapot number	Migration liquid	mg l^{-1}	mg day^{-1}	% TRV[a]
No. 1	Natural tea	1.74	1.4	650
No. 2	Natural tea	0.84	0.7	314
No. 3	Natural tea	0.71	0.6	264
No. 4	Natural tea	0.18	0.1	66
No. 5	Natural tea	1.19	1.0	444
No. 6	Natural tea	0.10	0.1	36
No. 7	Natural tea	1.49	1.2	557
No. 8	Natural tea	0.32	0.3	118
No. 9	Natural tea	0.20	0.2	73
No. 10	Natural tea	0.34	0.3	129
No. 11	Natural tea	4.90	3.9	1830
No. 1	Citric acid	23.97	19.2	8961
No. 2	Citric acid	9.10	7.3	3400
No. 3	Citric acid	8.30	6.6	3101
No. 4	Citric acid	10.21	8.2	3816
No. 5	Citric acid	13.12	10.5	4905
No. 6	Citric acid	1.07	0.9	399
No. 7	Citric acid	16.12	12.9	6027
No. 8	Citric acid	0.38	0.3	142
No. 9	Citric acid	0.42	0.3	157
No. 10	Citric acid	0.65	0.5	243
No. 11	Citric acid	17.60	14.1	6578

Note: [a]Toxicological reference value.

For Cu the comparison of the ingestion of Cu compared with the TRV reveals the fact that there is no surplus compared with TRV. But in the case of lemon tea four of 11 teapots induced a surplus of up to 468%.

For Fe, both in the cases of natural tea or lemon tea, there was no surplus compared with TRV.

For Pb (Table 4), the comparison of the ingested Pb, from natural tea, with the TRV showed a surplus in eight cases per 11 teapots, and some of them went up to 6.5 times the TRV. In the case of lemon tea ingestion, we exceeded the TRV with the 11 teapots and by up to 90 times.

For Ni, the comparison of ingested Ni with the TRVs in the case of natural tea showed an excess in eight cases per 11 teapots, and some of them went up to four times the TRV. In the case of lemon tea, the ingested Ni exceeded the TRV in 10 of 11 teapots, with and at 15 times the TRV.

In the case of Zn, both for natural tea or lemon tea, there were no surplus in all teapots.

Conclusions

The potential risk of lead and nickel poisoning for the general population induced by some traditional metallic teapots which were found on the Belgian market should be noted. Consequently, exceeding the toxicological reference values (TRV) for lead and nickel is very worrying. Factors such as a longer brewing time (migration up to nine times higher after 1 h rather than after 15 min) could exacerbate the exposure. The use lemon is also aggravating since migration levels may be more than ten times higher than with a natural tea. In the case of lead, surveillance must be given to those population presenting a high risk of exposition like children (including the cases cited above) since these, given their greater assimilation, are more susceptible than adults to lead poisoning. The other metals (aluminium, copper, iron and zinc) envisaged in this study were not so problematic since they all clearly fell below the TRV.

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References

Ajmal M, Khan A, Nomani AA, Ahmed S. 1997. Heavy metals: leaching from glazed surfaces of tea mugs. Sci Total Environ. 207:49–54.

Boularbah A, Bitton G, Morel JL. 1999. Assessment of metal content and toxicity of leachates from teapots. Sci Total Environ. 227:69–72.

Cleays F, Sykes C, Limbos C, Ducoffre G. 2003. Childhood lead poisoning in Brussels prevalence study and etiological factors. J Phys IV France. 7:1–4.

Council of Europe. 2010. Draft Resolution of Council of Europe on metals and alloys in contact with food, Guidelines project under consideration.

European Committee for Standardization. 1995. EN 1388-1: Material and articles in contact with foodstuffs – Silicate surfaces – Part 1: Determination of the release of lead and cadmium from ceramic ware. Brussels: European Committee for Standardization.

Hellström-Lindberg E, Bjorklund A, Karlson-Stiber C, Harper P. 1997. Extraction of lead, cadmium and zinc from overglaze decorations on ceramic dinnerware by acidic and basic food substances. Sci Total Environ. 197:167–175.

Hellström-Lindberg E, Bjorklund A, Karlson-Stiber C, Harper P, Seldén AI. 2006. Lead poisoning from souvenir earthenware. Int Arch Occup Environ Hlth. 79:165–168.

Milton A, Lessler MA. 1965. Lead and lead poisoning from antiquity to modern times. J Sci. 88:78–88.

Ni L, Li S. 2008. Effects of organic matters coming from Chinese tea on soluble copper release from copper teapot. Sci Total Environ. 389:202–207.

Official Journal of the European Union. 2004. Regulation (EC) No 1935/2004 on materials and articles intended to come in to contact with food and repealing Directives 80/590/EEC and 89/109/EEC. L338/4. Strasbourg: European Parliament and the Council.

Petit D, Claeys F, Sykes C, Noefnet Y. 2003. Lead poisoning from metallic teapots traditionally used by North African populations. J Phys IV France. 107:1053–1056.

Phan TG, Estell J, Duggin G, Beer I, Smith D, Ferson MJ. 1998. Lead poisoning from drinking Kombucha tea brewed in a ceramic pot. Med J Aust. 169:644–646.

Wilson TW, Card RT. 1986. Lead poisoning: unusual manifestation and unusual source. Can Med Assoc J. 135:773–775.

Ziegler S, Wolf C, Salzer-Muhar U, Schaffer A, Konnaris C, Rudiger H, Osterode W. 2002. Acute lead intoxication from a mug with a ceramic inner surface. Am J Med. 112:677–6784.

Zuckerman MA, Savory D, Rayman G. 1989. Lead encephalopathy from an imported Toby mug. Postgrad Med J. 65:307–309.