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

Journal:	<i>Food Additives and Contaminants</i>
Manuscript ID:	TFAC-2010-514.R1
Manuscript Type:	Original Research Paper
Date Submitted by the Author:	28-Mar-2011
Complete List of Authors:	BOLLE, Fabien; Scientific Institute Of Public Health, Food Brian, Wendy; Scientific Institute Of Public Health, Food Petit, Daniel; Free University of Brussels, Department of Chemistry, Faculty of Science BOUTAKHRIT, Khalid; Scientific Institute Of Public Health, FOOD Feraille, Guillaume; Scientific Institute Of Public Health, Food Van Loco, Joris; Scientific Institute Of Public Health, FOOD
Methods/Techniques:	Metals analysis - ICP
Additives/Contaminants:	Food contact materials, Food simulants, Migration, Toxic elements
Food Types:	Beverages
Abstract:	An environmental inquiry conducted by the Brussels Inter-communal Laboratory of Chemistry and Bacteriology (BILCB) has revealed that a traditional metallic teapot provoked in the year 2000 in Brussels a 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). Al, Cu, Fe, Ni, Pb and Zn were analysed to identify metals with a potential to migrate in tea solutions. Simulants (nature 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-

	<p>elementary analysis was carried out by ICP-AES. We 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 of 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 to the toxicological reference values was done. The teapots were withdrawn from the market by the FASFC.</p>

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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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Key words: teapot, food contact, migration, ICP-AES, exposure, metals

Abstract

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). Al, Cu, Fe, Ni, Pb and Zn were analysed to identify metals with a potential to migrate in tea solutions. Simulants (nature 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 ICP-AES. We 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 of 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 to the toxicological reference values was done. The teapots were withdrawn from the market by the FASFC.

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INTRODUCTION

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

To our knowledge, few studies were dedicated to the origin of migration process in metallic teapots. We have found in the literature only two studies, one which studied copper, Lixiao et al., (2008) and an older one which investigated Zinc, Nickel, Copper and Tin, Boularbah et al., (1999).

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

A preliminary study made by the BILCB, has envisaged the leaching of lead brewed in those kind of metallic teapots: the concentrations of lead were on the order of a few hundred of µg/l but could be in some cases much higher (up to about 3.5 mg/l), which confirmed the potential danger of using those items (Petit et al., 2003).

These items were very commonly used in families coming from Magrebbian or Minor Asia countries. It is also possible to find these products in snacks, tearooms or oriental restaurants willing to bring a touch of traditional products they serve their customers.

To the extent that they contain substantial amounts of lead and other metals to the known toxicity, IPH has taken the initiative to investigate the levels of presence and confronted with the toxicological reference values. These results have led to market withdrawals made by the 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 to 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 9 items have been purchased. It is sometimes difficult to determine the origin of these products, but these shops are specialized in products from North Africa. Samples code as "1RB1" were collected in three identical copies in order to make a comparison between migration tests.

Other series of 11 teapots was provided by the FASFC, those items, bought also in North African groceries were originated-from India and Morocco.

Migration procedure

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

Three simulants were used as leaching medium: tea infusions (prepared by boiling 6 bags in 1 litre Milli-Q water; pH 4.86), citric acid (99.5% (m/m), Merck) by dissolving 1 g in 1 L of Milli-Q water (pH 2.7) and the mixture of tea nature and citric acid (1 g/L).

Teapots were filled with the leaching solution up to 1 cm from the top and placed on a hotplate for heating; aliquots of 2 mL were taken after a period of 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. It is therefore that the leaching media were used in place of acetic acid that is generally chosen as standard for example in ceramic materials in contact with food ([European Committee for Standardization, 1995](#)). It was decided because it is more realistic to simulating the lemon action used in making some of our tea infusions. A comparison (for three identical pieces) from three migration solutions include: citric acid, 1g / L with pH 2.7 similar to the commercial lyophilized tea (pH of 2.95), tea and tea acidified with citric acid 1 g / L was also performed.

The use of citric acid helps standardize acidity of the juice and corresponds to a current consumption pattern. This standardization would not be possible using a real lemon. Citric acid helps, moreover, to explore the possibility of using a standard simulant in the laboratory and within the logic "Worst case" that prevails in Europe, in establishing the compliance of a material or article intended for food contact ([Regulation 1935/2004/EC](#)). This simulant also avoids having to work with significant levels of blank in testing migration. The concentration of some elements in tea is likely distorting the evaluation results.

Digestion procedures

First, the teapots were drilled with a drilling machine. Approximately 0.05 g of metal powder was weighed in PFA digestion Tube, then, 3 mL HCl + 1 mL HNO₃ were added. Afterwards, digestion was carried out at 180°C in the oven and evaporated to about 0.5 mL. 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 LOD, defined as three times the standard deviation of the blank, were less than 10 µg/L for all elements analysed, the within-laboratory reproducibility was less than 5% and the expanded measurement uncertainty were less than 15% for all element measured. An external calibration curve was carried out with the following standards: 25, 50, 100 and 200 µg/L, all standards were prepared by weighing a solutions (1 g/L, Fluka) in 2% (v/v) nitric acid (70% (w/w), J.T. Baker). The samples solutions were measured in triplicate (instrument programmed) with ICP-AES (Perkin-Elmer Optima 4300DV, Norwalk, CT, USA). Samples were diluted in nitric acid 2% (v/v) and reanalyzed when concentration appeared to exceed the highest concentration of the calibration curve. An internal control during the samples analysis was performed; check of the slope values and the coefficient of the correlation, analyse of one standard each ten samples measurement.

The concentrations of heavy metals were determined in each leaching solution, regarded as procedural blank (the leaching medium were placed in a Pyrex baker), before the migration test. In the citric acid simulant, all metals content were less than the LOD of the method (10 µg/L). For the tea nature used as leaching solution, the predominant present metals were (in mg/L): Al (4.2 ± 0.3), Zn (1.4 ± 0.2), Mn (0.93 ± 0.09), and Cu (0.050 ± 0.003). These concentrations values must be subtracted from the migrated values in subsequent migration test with tea nature.

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 were carried out by ICP-AES, this revealed that all teapots are made up by Brass alloy, meanly 59% Cu and 36% Zn and a majority of teapots are from leaded brass types because lead is known for its excellent ductility. We notice also the presence of others metals such as Fe, Al and Ni at low contents. Al and Ni are added for their corrosion resistance properties (Table 1). On the basis of these results, we suspect the migration of Cu and Zn followed by Pb, then Fe, Al and Ni in certain cases.

Comparison between simulants

Three identical teapots (codes 1RB1 a, b, c) were used for this comparison. Three simulants were used for migration test; tea nature, 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 a "lemon tea" and the concentration corresponds to the concentration of citric acid that we should expect to gain 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 1/10 of lemon (80 g / L of citric acid) in a cup of 250 ml.

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

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

It is already established that the use of these teapots "1RB1" leads to high concentrations of Lead, Nickel and Zinc, and this even after 15 minutes of contact. The use of lemon juice actually leads to a significant increase in migration levels: up more than 8 to 10 times in the case of Lead.

The kinetics of migration

Kinetic migration studies were carried out with teapots bodies, filled up by leaching media as tea infusion 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) have been illustrated in figure 2 and 3. For lead and nickel, we observe similar behaviour, for almost teapot, both in tea leaching solution and citric acid; the release rate is higher at the first migration period of approximately 15 min and less than 50% of metals have been leached, after the rate of release decreases significantly with time. This can be explained partly by a gradual metals enrichment of the passive surface oxide layer, except for example teapots n° 6 in case of Ni and n° 11 in case of Pb where the migration increased with time. Comparing the simulants, it appears that high concentration of metals have been released with citric acid which can be explained by 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 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 but they did not show any real interest at this stage of the study due to their negligible migration or to their absence in teapots composition and to their high level in tea infusion. The suggestion made before about the metals at high content (high migration of Cu and Zn) likely to migrate from teapots is not confirmed except for the following teapots n°2, n°11 for copper and n°2RB2, n°8 and n°10 for zinc. All of the teapot leachates examined contained high concentrations of Pb and Ni then Zn or Fe and Cu, the presence of other elements as Al depends on the type of teapot and its highest leached concentrations was found for teapot reference 2RB2. Teapot (1RB1a) released the highest concentration of lead and teapot (1AS1) for the nickel in spite of their low content in these teapots. From these results, we noticed that migration of metals did not match well with the content of these metals in teapot except for the cases quoted above. The migration phenomena of metals depends on electrochemical properties of elements, in spite of their low redox potentials (Al; -1.66V, Zn; -0.762V, Fe; -0.44V), the corrosion of these elements is less than that of Pb and Ni. These can be explained by the formation of a

passivation layers or to the own structure of the brass, indeed, the alloy Cu-Zn present a high corrosion resistance and only the added elements as Pb and Ni are very affected by the corrosion due perhaps to their non dissolution in the brass matrix.

Comparison of the results to the toxicological reference values

The objective is to confront the obtained values to the toxicological reference values (TRV expressed in mg/day: Cu: 5; Fe: 17; Pb: 0.214; Ni: 0.7; 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 which consider a person of 60 kg weight (Draft resolution on metals and alloys 2010: 2, is not publicly available). The assumption would be a consumption of 4 cups, with a volume of 200 mL each one, of tea or nature tea with lemon (citric acid simulant), namely 800 mL per day. Here, it only takes a contact of 15 min.

For Copper, the comparison of the ingestion of copper compared to the toxicological reference value reveal that no surplus compared to TRV. But in the case of lemon tea, 4 of 11 teapots induce a surplus of up to 468%.

For Iron, both in the case of tea nature or lemon tea, there are no surplus compared to TRV.

In case of Lead (Table 4), the comparison of the ingested lead, from nature tea, to the toxicological reference value shows a surplus in 8 cases per 11 teapots, and some of them go up to 6.5 times of the TRV. In the case of lemon tea ingestion, we exceeded the TRV with the 11 teapots and up to 90 times.

For Nickel, the comparison of ingested nickel to the toxicological reference values in the case of nature tea shows an excess in 8 cases per 11 teapots, and some of them go up to 4 times the TRV. In case of lemon tea, the ingested nickel exceeded the TRV in 10 per 11 teapots with and goes up to 15 times TRV.

In the case of Zinc, both for nature tea or lemon tea there are no surplus in all teapots.

CONCLUSIONS

It should be noted the potential risk of lead and nickel poisoning induce for the general population by some traditional metallic teapots which were currently found on the Belgian market. Consequently, exceed of the toxicological reference values (TRV) for lead and the nickel are very worrying and the asked assumptions are meant but not out. Factors such as a longer stay (migration up to 9 times higher after 1 hour rather than 15 min.) could exacerbate the exposure. Use lemon is also aggravating since migration levels may be more than 10 times larger than with a nature tea. Also note in the case of lead, the sustained attention to have for populations most at risk as children (including cases cited above) since these, given the greater assimilation, are more susceptible than adults to lead poisoning. The other metals (Al, Cu, Fe and Zn) envisaged in this study were not so problematic, they all fallen clearly below the TRV.

Acknowledgments

This work has benefited from the help of Dr. Petit from Inter-communal Laboratory of Chemistry and Bacteriology (BILCB) and the help of the Provincial unit of Control (UPC) in Brussels from the Federal Agency for the Safety of the Food Chain (FASFC) who by their kind collaboration allowed the culmination of this work.

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Fig. 1 : Teapots samples collected from the market in Brussels

Fig. 2a : The evolution of lead migration, from various teapots, in *nature tea* migration liquid with the contact time.

Fig. 2b : The evolution of lead migration, from various teapots, in *citric acid* simulant with the contact period.

Fig. 3a : The evolution of Nickel migration, from various teapots, in *nature tea* migration liquid with the contact time.

Fig. 3b : The evolution of Nickel migration, from various teapots, in *citric acid* simulant with the contact period.

Teapots codes	Cu (%)	Zn (%)	Pb (%)	Fe (%)	Ni (%)	Al (%)	Formatted Table
1 AS 1	53	38	4.9	2.1	1.4	0.2	Deleted: Prevalent heavy metal content
1 AS 2	55	38	4.4	1.3	0.5	0.2	Formatted: Centered
2 AS 1	54	38	5.5	1.2	0.8	0.9	Deleted: Cu (53), Zn (38), Pb (4.9), Fe (2.1), Ni (1.4), Al (0.2)
2 AS 2	64	35	-	0.4	0.7	0.1	Formatted: Centered
1 RB 1 a	64	35	0.1	-	0.2	-	Deleted: Cu (55), Zn (38), Pb (4.4), Fe (1.3), Ni (0.5), Al (0.2)
2 RB 2	66	33	-	0.6	0.1	-	Formatted: Centered
2 RB 3	55	38	5.3	1.2	0.2	0.4	Deleted: Cu (54), Zn (38), Pb (5.5), Fe (1.2), Al (0.9), Ni (0.8)
n° 1	52	37.1	6.7	1.7	0.5	0.4	Formatted: Centered
n° 2	61	38	0.1	0.1	0.2	-	Deleted: Cu (54), Zn (38), Pb (5.5), Fe (1.2), Al (0.9), Ni (0.8)
n° 3	54	35	4.5	4.3	1.1	0.3	Formatted: Centered
n° 4	55	38	6.0	1.0	0.1	0.2	Deleted: Cu (64), Zn (35), Ni (0.7), Fe (0.4), Al (0.1)
n° 5	54	39	4.8	1.7	0.4	0.3	Formatted: Centered
n° 6	65	35	-	-	0.4	-	Deleted: Cu (64), Zn (35), Ni (0.2), Pb (0.1)
n° 7	55	37	4.6	2.7	0.4	0.3	Formatted: Centered
n° 8	64	36	-	-	0.4	-	Deleted: Cu (64), Zn (35), Ni (0.2), Pb (0.1)
n° 9	63	37	-	-	0.1	-	Formatted: Centered
n° 10	64	36	-	-	0.3	-	Deleted: Cu (66), Zn (33), Fe (0.6), Ni (0.1)
n° 11	60	34	5.7	-	0.8	0.9	Formatted: Centered

Table 1 : Major composition of the teapots.

Teapots codes	Simulants	Pb (mg/L)	Ni (mg/L)	Zn (mg/L)
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 + citric acid	45	13	3.8

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

Teapots 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	-	-
n° 1	0.31	0.15	3.82	36.5	10.6	2.2
n° 2	0.19	17.1	0.52	14.1	4.1	14.9
n° 3	0.16	13.9	2.01	16.7	1.2	8.1
n° 4	0.07	2.2	1.05	18.2	8.7	1.7
n° 5	0.19	0.33	4.35	28.7	9.9	2.1
n° 6	0.10	1.1	0.31	2.0	16.6	1.5
n° 7	0.31	0.41	3.3	23.7	10.7	3.7
n° 8	0.52	8.05	0.4	2.0	13.6	30.0
n° 9	0.37	1.85	0.13	1.3	15.6	9.6
n° 10	0.35	18.0	0.2	1.1	9.9	19.8
n° 11	1.1	40.8	0.62	36.2	18.6	24.1

Table 3 : Metals concentrations (mg/L) in leachates of citric acid from various teapots after a contact period of 30 min.

Lead (hypothesis of 4 tea cups/day [800 mL])				
Teapots n°	Migration liquid	mg/L	mg/day	% TRV*
n°1	Nature tea	1,74	1,4	650
n°2	Nature tea	0,84	0,7	314
n°3	Nature tea	0,71	0,6	264
n°4	Nature tea	0,18	0,1	66
n°5	Nature tea	1,19	1,0	444
n°6	Nature tea	0,10	0,1	36
n°7	Nature tea	1,49	1,2	557
n°8	Nature tea	0,32	0,3	118
n°9	Nature tea	0,20	0,2	73
n°10	Nature tea	0,34	0,3	129
n°11	Nature tea	4,90	3,9	1830
n°1	Citric acid	23,97	19,2	8961
n°2	Citric acid	9,10	7,3	3400
n°3	Citric acid	8,30	6,6	3101
n°4	Citric acid	10,21	8,2	3816
n°5	Citric acid	13,12	10,5	4905
n°6	Citric acid	1,07	0,9	399
n°7	Citric acid	16,12	12,9	6027
n°8	Citric acid	0,38	0,3	142
n°9	Citric acid	0,42	0,3	157
n°10	Citric acid	0,65	0,5	243
n°11	Citric acid	17,60	14,1	6578

*: Toxicological reference values

Table 4 : Comparison of the ingested lead, from nature tea and citric acid after a contact period of 15 min, to the toxicological reference values.

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Cu (52), Zn (37.1), Pb (6.7), Fe (1.7), Ni (0.5), Al (0.4)		
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Cu (61), Zn (38), Ni (0.2), Pb (0.1), Fe (0.1)		
Page 1: [3] Deleted	KBoutakhrit	3/22/2011 10:46:00 AM
Cu (54), Zn (35), Pb (4.5), Fe (4.3), Ni (1.1), Al (0.3)		
Page 1: [4] Deleted	KBoutakhrit	3/22/2011 10:46:00 AM
Cu (55), Zn (38), Pb (6.0), Fe (1.0), Al (0.2) Ni (0.1)		
Page 1: [5] Deleted	KBoutakhrit	3/22/2011 10:46:00 AM
Cu (54), Zn (39), Pb (4.8), Fe (1.7), Ni (0.4), Al (0.3)		
Page 1: [6] Deleted	KBoutakhrit	3/22/2011 10:46:00 AM
Cu (55), Zn (37), Pb (4.6), Fe (2.7), Ni (0.4), Al (0.3)		
Page 1: [7] Deleted	KBoutakhrit	3/22/2011 10:46:00 AM
Cu (60), Zn (34), Pb (5.7), Ni (0.8)		
Page 1: [8] Deleted	KBoutakhrit	3/22/2011 10:41:00 AM

hemical analysis of prevalent heavy metals content in teapots components



Fig. 1

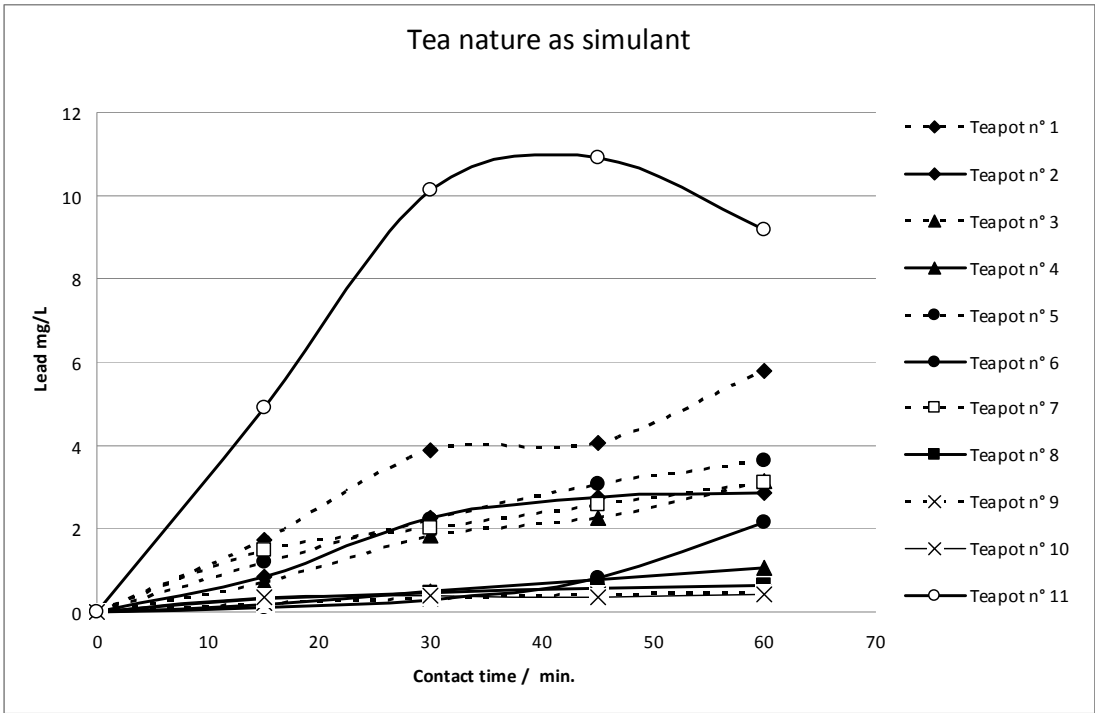


Fig. 2a

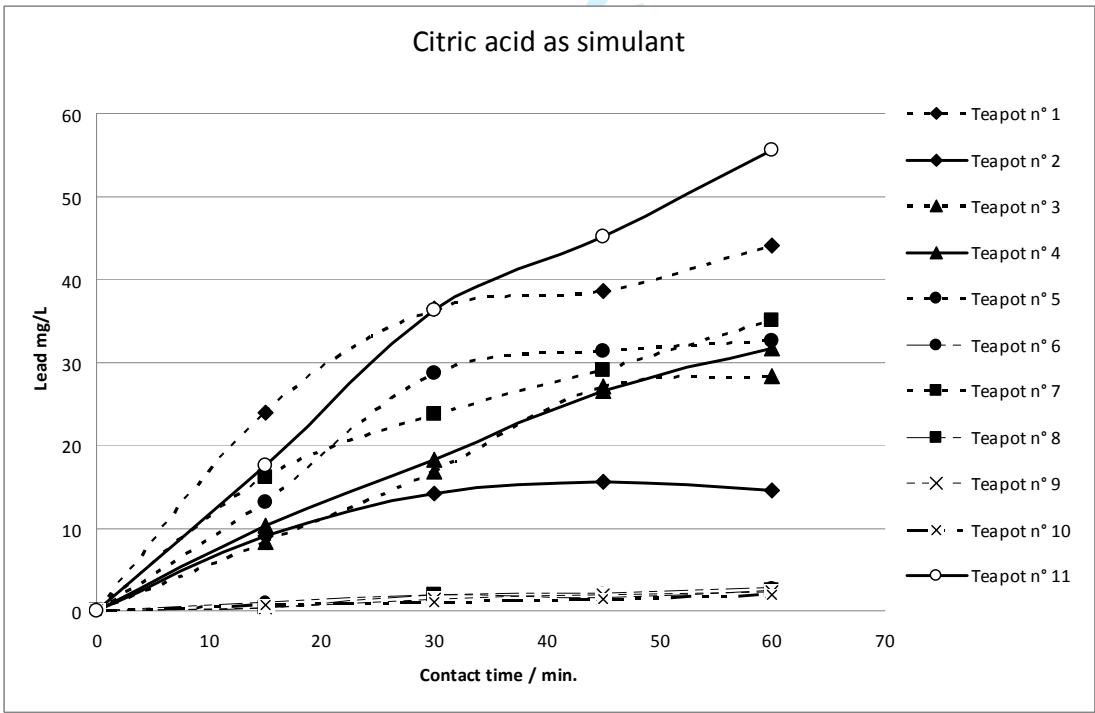


Fig. 2b

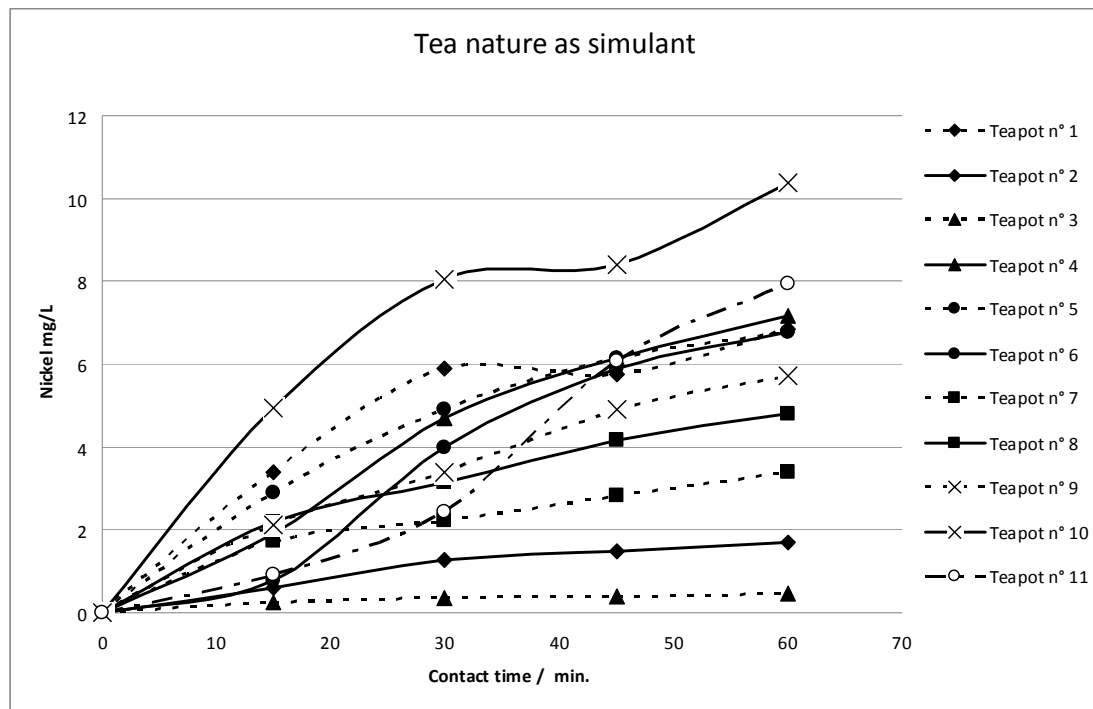


Fig. 3a

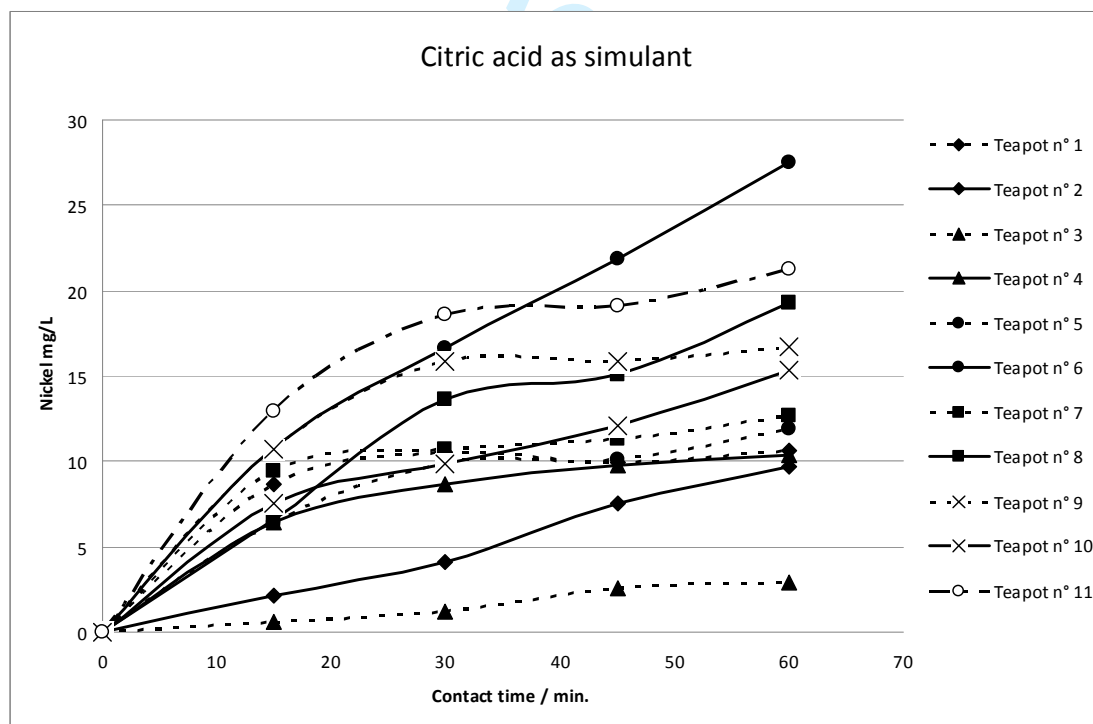


Fig. 3b