Exposure to lead and cadmium of the Belgian consumers from ceramic food contact articles

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\textbf{A B S T R A C T}

This study was aimed at estimating the intake of lead and cadmium by the Belgian consumers due to the use of ceramic ware. We adopted refined exposure scenarios with respect to migrant concentration, ceramic ware usage, and metal release characteristics. A deterministic estimation was initially performed, followed by probabilistic estimation, if the deterministic estimate exceeds toxicological reference values. Based on the reference doses established by the European Food Safety Authority (EFSA), the risk of lead and cadmium exposure was characterized by the margin of exposure (MOE) and the tolerable weekly intake (TWI), respectively. The probabilistic median and 95th percentile intake of lead were 0.02 and 5.77 μg/kg b.w. per day for adults, and 0.07 and 17.3 μg/kg b.w. per day for children. The MOEs for neurotoxicity, nephrotoxicity and cardiovascular effects were 7.1, 27 and 64 for average consumers, and 0.02, 0.1 and 0.3 for high consumers. The deterministic mean and 95th percentile intake of cadmium were 0.026 (7% TWI) and 0.03 (8% TWI) μg/kg b.w. per day for adults, and 0.08 (22 % TWI) and 0.09 (25 % TWI) μg/kg b.w. per day for children. Considering the exposure of the Belgian population from foodstuffs resulted in the exceedance of the TWI by as much as 20-fold. The risk of exposure to lead and cadmium of the Belgian consumers suggests measures, such as lowering the migration limits for ceramic ware, should be taken to minimize the risk.

1. Introduction

A release of metals occurs when ceramic particles come into contact with food and beverage, and ceramic ware can be an important source of intake of toxic metals \cite{1-4}. The most reported case is the release of lead from glazed ceramic ware which raised blood lead levels in Mexico \cite{5-7}.

The risk of exposure to lead and cadmium has been evaluated by national and international authorities. Lead impacts neurological, cardiovascular, nephritic, gastrointestinal and hematological systems \cite{8-10}. Children are especially susceptible to the neurotoxicity of lead, and even low levels of exposure can result in the damage to developing brain \cite{11,12}. The Joint FAO/WHO Expert Committee on Food Additives (JECFA) has withdrawn the provisional tolerable weekly intake (PTWI) of lead of 25 μg/kg b.w. due to the absence of the threshold for the negative physiological effects \cite{13}. As a consequence, the European Food Safety Authority (EFSA) Panel on Contaminants in the Food Chain (CONTAM) has proposed the use of benchmark dose (BMD) approach to characterize the dose-response relationship \cite{9}. The benchmark dose lower confidence limits (BMDLs) dietary intake values proposed by the EFSA are respectively 0.50 μg/kg b.w. per day for developmental neurotoxicity, 0.63 μg/kg b.w. per day for nephrotoxicity and 1.50 μg/kg b.w. per day for cardiovascular effects \cite{9}.

Cadmium is toxic to a wide range of organs, and chronic exposure to cadmium exerts toxic effects mainly on kidneys and bones \cite{14}. The International Agency for Research on Cancer (IARC) identifies cadmium as carcinogenic to humans (group 1), with sufficient evidence for lung cancer and limited evidence for kidney, liver and prostate cancer (IARC, 1993). The JECFA establishes the provisional tolerable monthly Intake (PTMI) for cadmium of 25 μg/kg b.w. \cite{13}, and the EFSA sets the tolerable weekly intake (TWI) at 2.5 μg/kg b.w. \cite{14}.

As a source of contamination, the release of lead and cadmium from ceramic ware is monitored by the majority of countries. In the EU the Directive 84/500/EEC (referring to Directive in the following text) sets...
the requirements of ceramics intended to come into contact with food-stuffs; it regulates the permissible limits of lead and cadmium and the testing method [15]. The Directive is currently being revised by the European Commission, and the revision concentrates on lowering the permissible limits and optimizing testing conditions [16].

Limited studies have estimated the intake of lead and cadmium derived from the use of ceramic food contact articles. The German Federal Institute for Risk Assessment (BfR) reported that the intake of lead and cadmium exceeded the PTWI more than 10-fold by assuming that ceramic articles released lead and cadmium at the permissible migration levels set by the Directive, and that a plate with a diameter of 20 cm and a depth of 1.5 cm (4 square decimeters, dm² surface area) containing a volume of 471 mL was used by consumer either once a day or once a week [17]. In 2020, the BfR has characterized the risk of exposure to lead and cadmium using the reference values set by the EFSA [18]. The BfR assumes a daily one-off dietary intake of toddlers from one ceramic plate with a surface area of 3 dm², which also results in the exceedance of reference doses. The Norwegian Scientific Committee for Food Safety (VKM) estimated the exposure to lead and cadmium migrating from ceramic articles based on the assumption that the Norwegians consume 1 L of liquid via a ceramic article of 5 dm² surface per day [19]. Both the highest migration levels from monitoring samples and the Directive permissible levels are used in the estimation. The estimated intakes of lead and cadmium exceed the corresponding PTWI many times. Rebenik et al. [20] and Mania et al. [21] have assessed the risk of exposure to Polish lead and cadmium from ceramic articles applying the permissible limit of the Polish regulation on the release of lead and cadmium from ceramic articles. The intake of lead exceeds the BMDL, by respectively 3-fold and 30-fold for cardiovascular effects and neurotoxicity, and the intake of cadmium exceeds the TWI by 1–4 folds.

The accessible data with respect to the intake of lead and cadmium from ceramic articles has shown concerns; however, it is noted that simple assumptions have been used in previous estimations. A high estimate of exposure arises if assuming the release of lead and cadmium at the levels of the Directive limits, whereas the intake of lead and cadmium is very likely to be underestimated if assuming a ceramic article of 4 dm² or 471 mL is used by consumer (BfR assumption). Furthermore, these estimations neglect the real migration process of ceramic ware. The release of lead and cadmium decreases over the repeated use of ceramic ware [22–25], so the assumption that ceramic ware releases lead and cadmium at constant levels results in considerably high estimates.

No published articles have reported the intake of lead and cadmium of the Belgian population derived from the use of ceramic ware. This study was aimed at estimating the exposure of the Belgian consumers to lead and cadmium via the use of ceramic ware. The understanding of real exposure to lead and cadmium from ceramic ware also provides scientific evidence on the revision of the Directive, particularly regarding the limits of lead and cadmium. To refine the assessment, the present estimation (a) employed realistic data on migrant concentration and ceramic usage, and (b) modeled the migration process of metals from ceramic ware. Initially, a deterministic estimation was performed, followed by a probabilistic estimation, if the deterministic estimate exceeds toxicological reference values. We also discuss contributing parameters to the exposure through qualitative and quantitative sensitivity analyses.

2. Materials and methods

2.1. Deterministic approach

2.1.1. Exposure models

The exposure to chemicals from food contact materials can be described by a combination of the amount of contact materials used containing the concerned chemical and the migration level of the chemical, as shown in the following equation [26]:

\[
\text{Exposure} = \text{Migration} \times \text{Contact Material Usage}
\]  

where Migration Migration represents the concentration of the chemical migrating into food. Contact Material Usage represents the area of materials containing the migrating chemical in contact with food.

According to the Directive, ceramic ware is basically divided into two categories: hollowware (with the capacity less or more than 3 L) and flatware. The Eq. (1) is thus adapted to the Eq. (2), which separately characterizes the exposure of chemicals from hollowware and flatware. Exposure (mg per day per person) = Hollowware Migration (mg/L) × Hollowware Usage (L/day/person) + Flatware Migration (mg/dm²) × Flatware Usage (dm²/day/person) reflect the usage volume of hollowware and the usage area of flatware, respectively. Applying the Eq. (2), the following model inputs were required.

\[
\text{Exposure} = \text{Hollowware Migration} \times \text{Hollowware Usage} + \text{Flatware Migration} \times \text{Flatware Usage}
\]

2.1.2. Model inputs (parameters)

2.1.2.1. Migrant concentrations (hollowware migration & flatware migration).

The safety of ceramic articles on the Belgian market is monitored. Ceramic articles available to consumers were taken from the market by the Federal Agency for the Safety of the Food Chain (FASFC) and analyzed by the Belgian Reference Laboratory for Food Contact Materials of the Sciensano. Sampling was statistically distributed to accurately reflect the market situation. The migration test was performed under the test condition described in the Directive: four replicates were filled with 4% acetic acid at the temperature of 22 ± 2 °C for 24 ± 0.5 h. During the period of 2008–2016, roughly 1400 pieces (4 replicates for each piece) of ceramics have been collected and analyzed including various types of plates, cups and containers. In total, the concentration of lead and cadmium leaching from 1203 ceramic articles was recorded, and it would be used in this exposure assessment. The concentration was expressed as mg/L and mg/dm² for hollowware and flatware, respectively. In the case where the concentration was below the limit of quantification (LOQ), a “<LOQ” was recorded.

2.1.2.2. Ceramic ware usage (Hollowware usage and & flatware usage).

Surveys regarding the usage of ceramic ware are rarely conducted, and thereby this study applied industrial data and transformed industrial data into ceramic ware usage which can be used directly in exposure calculation.

Consumption quantities

The consumption quantity of ceramic ware was retrieved from the Eurostat database, which contains the statistics of manufactured goods. The database covers the industrial data of ceramic ware of Belgium from 2009 to 2016, depicted by indexes with respect to production, import, and export. These indexes were used to calculate consumption quantity with the following formula [42] (Williams, 2008):

\[
CQ = PQ + IQ + EQ
\]

where CQ is the quantity of ceramic ware consumed per year (kg/year), PQ is the quantity of ceramic ware manufactured per year (kg/year), IQ is the quantity of ceramic ware imported per year (kg/year), and EQ is the quantity of ceramic ware exported per year (kg/year). To describe the individual Belgian consumption, another indicator—consumption quantity per capita—was introduced. Dividing the annual consumption quantity by the corresponding annual Belgian population gives the
consumption quantity per capita.

**Conversion factors and allocation**

The consumption quantity from statistics is not a parameter which can be directly used in the exposure model. It is, therefore, converted into consumption volume and/or consumption area by conversion factors. To obtain conversion factors, we conducted a pilot study which investigated the weight, volume and surface area of ceramic ware used by the Belgian consumers.

Samples in the pilot study, consisting of 53 types of hollowware and 53 types of flatware, were from three origins: the European Reference Laboratory for Food Contact Materials (EURL-FCM), FASFC and the Belgian market. For hollowware, the weight and volume were measured; for flatware, the weight, volume, and diameter were measured. The conversion factors—volume-to-weight ratio for hollowware and area-to-weight ratio for flatware—were calculated using the following equations.

\[
VWR = \frac{VH}{WF} \quad (4)
\]

\[
AWR = \frac{AF}{WF} \quad (5)
\]

where VWR is the volume-to-weight ratio (L/kg), AWR is the area-to-weight ratio (dm²/kg), VH is the volume of hollowware (L), AF is the surface area of flatware (dm²), WH is the weight of hollowware (kg), WF is the weight of flatware (kg).

The consumption quantity obtained from industrial data aggregated the quantity for hollowware and flatware, so allocation of consumption quantity between hollowware and flatware was made based on the average weight of hollowware and flatware derived from the pilot study.

### 2.1.3. Exposure scenarios

Apart from parameter inputs, exposure calculation also requires a combination of parameter inputs, so scenarios in relation to each parameter used in the estimation of exposure are defined herein. Table 1 summarizes exposure scenarios with respect to the main parameters. The exposure model can then be expressed as follows:

\[
\text{Exposure} = \sum DF_i \times (\text{MH} \times CQ \times AH \times VWR \times EF + MF \times CQ \times AF \times AWR \times EF) \times \frac{BW \times ED}{BW \times ED} \quad (6)
\]

where \(DF_i\) is the decreasing factor of the \(i\)th migration, MH is the migrant concentration from hollowware (mg/L), CQ is the consumption quantity of ceramic ware (kg/year/person), AH is the allocation for hollowware, AF is the allocation for flatware, EF is exposure frequency of ceramic ware (times/day), MF is the migrant concentration from flatware (mg/dm³), BW is the body weight (kg), ED is the exposure duration in day.

#### 2.1.3.1. Exposure duration, exposure frequency of ceramic ware, body weights and high consumers

The exposure duration was set at one year. Consumers were assumed to use ceramic ware on a daily basis. A Body weight of 60 kg for an adult and a body weight of 20 kg for a child were adopted in the exposure calculation, which is consistent with the body weight used for deriving toxicological reference values. The average consumers in the deterministic estimation are defined as the population who consume the mean quantity of ceramic ware, which is calculated using the mean migrant concentration and the mean consumption quantity of ceramic ware. The higher consumers of the deterministic estimate are defined as the population who consume the 95th percentile quantity of ceramic ware, calculated with the mean migrant concentration and the 95th percentile consumption quantity of ceramic ware.

#### 2.1.3.2. Allocation for hollowware and flatware

The allocation for hollowware and flatware was assumed proportional to the weight of hollowware and flatware. Apart from the allocation derived from the pilot study, additional allocation scenarios including half hollowware (0.5) and half flatware (0.5), complete hollowware (1), and complete flatware (1), were applied to examine the influence of allocation (i.e., the influence of the weight of hollowware and flatware) on exposure.

#### 2.1.3.3. Substitution of censored data

The refinement of migrant concentration takes into account censored data (concentrations below the LOQ) and the migration characteristics of ceramic ware. In the treatment of censored data, a substitution method was applied, where censored data were substituted by the half of the LOQ (medium bound, MB). Moreover, the LOQ (upper bound, UB) and zero (lower bound, LB) substitution scenarios were also used to study the influence of the substitution of censored data on exposure.

#### 2.1.3.4. Decreasing factors

To adequately reflect the release of lead and cadmium after consecutive use, decreasing factors were applied. Beldi et al. [23] showed the release of lead from 14 types of ceramic samples over three repeated migration tests under the Directive test conditions. The percentage of the third migration respect to the first one i.e., the ratio of the concentration of lead from the third migration to the concentration of lead from the first one, varied from 6% to 35%, with the mean value of 20%. Li [25] has reported that the concentration of metals in the second migration test declines over 50% and that the concentration of metals declines over 80% after three migration tests. A decreasing factor of 50% for the second use and a decreasing factor of 20% for the third and the following use were implemented in the estimation. Considering exposure frequency and duration, the initial two migrations have a negligible influence on the outcome of daily use, so an average decreasing factor of 20% to the original concentration would be sufficient.

### 2.2. Probabilistic approach

Intake which exceeds toxicological reference values is further refined with probabilistic estimation. The probabilistic estimation adopted the same exposure model, scenario and input data as the deterministic estimation did. But the average consumers and the high consumers in the probabilistic estimate were different from those in the deterministic estimate. Defined from the perspective of intake distribution, the average consumers represent the population exposed at the median level of intake distribution, and the high consumers correspond to the population exposed at the 95th percentile of intake distribution. Statistical analyses were performed using the R (version 3.4.3, Foundation for Statistical Company, Vienna, Austria) and the package EnvStats was applied.

Input parameters including consumption quantities, conversion factors, and migrant concentrations were modeled following a number of processes: computing summary statistics, assessing goodness-of-fit with the quantile-quantile plot, estimating distribution parameters and testing for goodness-of-fit. The level of statistical significance of the goodness-of-fit was set at 0.01. In the case where more than one distribution fit the input data, the Akaike information criterion (AIC) was
calculated, and the distribution with the smallest AIC value was the best fitted one [27]. Censored migrant concentrations were treated with the maximum likelihood estimation method, which estimates the best fit distribution of the observed values above the quantification limit, compatibly with the percentage of data below the limit [10], rather than the substitution method in the deterministic estimation.

Using the fitted distribution of input variables, the estimation of exposure was repeated numerous times, during which a random number would be taken from the individual distribution of the variable. Each repetition or iteration would create one estimate of intake, and these estimates were subsequently combined to derive the distribution of intake. The calculation of exposure applied the Monte Carlo simulation approach. The simulation was run using the Latin Hypercube sampling method for each variable, with 10,000 iterations.

In order to determine the contribution of input parameters to exposure, the sensitivity of inputs was analyzed by bootstrapping 1000 new combinations of the consumption quantity, conversion factor and migrant concentration from the exposure output generated by the simulation. Correlations coefficients between inputs and estimated exposure were used to assess the levels of contribution of parameters.

3. Results and discussion

3.1. Migrant concentrations

The 1203 records included 664 records of hollowware and 539 records of flatware. As shown in Table 2, among hollowware lead was detected in 100 specimens, and cadmium was detected in 54 specimens. In the case of flatware, lead was detected in 180 specimens, and cadmium was detected in 101 specimens. 84 % hollowware and 81 % flatware released cadmium below the LOQ. For records below the LOQ, a value equal to the half of LOQ (medium bound, MB) was assigned. The mean concentrations of lead were 0.019 mg/L for hollowware and 0.016 mg/dm$^2$ for flatware. The mean concentrations of cadmium were 0.003 mg/L and 0.0007 mg/dm$^2$ for hollowware and flatware, much lower than the concentrations of lead.

Table 2 represents the mean concentrations for different scenarios as well. For the UB scenario, the mean concentration of lead was 0.024 mg/L in hollowware and 0.017 mg/dm$^2$ in flatware, and the mean concentration of cadmium was 0.005 mg/L in hollowware and 0.0008 mg/dm$^2$ in flatware. In the case of LB scenario, the mean concentration of lead was 0.015 mg/L in hollowware and 0.016 mg/dm$^2$ in flatware, and 0.002 mg/L in hollowware and 0.0007 mg/dm$^2$ in flatware.

3.2. Ceramic ware usage

3.2.1. Consumption quantities

The mean and 95th percentile consumption quantities of ceramic ware of the Belgian consumers at the period of 2009–2016 are listed in Table 3. To better evaluate the consumption of ceramic ware in Belgium, it is also compared with the consumption of ceramic ware in the European (EU) representing consumption quantity per EU consumer (Dividing the total consumption quantity in the EU by the total population in the EU gives the consumption quantity per EU consumer). The mean consumption quantity from 2009 to 2016 was 1.54 kg per person, ranging from 1.33 kg per person to 1.86 kg per person. There is no difference between the Belgian consumption and the EU consumption (1.57 kg per person).

3.2.2. Conversion factors and allocation

The measured parameters and conversion factors are listed in Table 4. The weights of hollowware and flatware measured in the pilot study varied from 0.11 to 0.60 kg and from 0.14 to 0.77 kg, respectively. The mean VWR was 1.3 ± 0.3 L/kg for hollowware, and the mean AWR was 8.5 ± 1.4 dm$^2$/kg for flatware. The mean weight was respectively 0.27 ± 0.11 kg and 0.40 ± 0.15 kg for hollowware and flatware, so the allocation of consumption quantity was 0.4 (0.27/0.67) for hollowware and 0.6 (0.40/0.67) for flatware.

3.2.3. Hollowware usage volume and flatware usage area

The consumption quantity of ceramic articles of 1.54 kg per person equates to approx. 2.3 pieces of hollowware (1.54 kg × 0.4/0.27 kg) and 2.3 pieces of flatware (1.54 kg × 0.6/0.40 kg). Consumers may have much more ceramic items in their cupboard, actually, only several pieces are used for one meal or one day. We assumed that these quantities of ceramic articles (1.54 kg) were used by the Belgian consumers on a daily basis. The consumption of ceramic ware was therefore equal to the usage of ceramic ware. Applying conversion factors and allocation for hollowware and flatware, consumption quantities were transformed into consumption volume and consumption area. On average, the usage of ceramic ware for a Belgian consumer was of 0.80 ± 0.19 L in volume and of 7.8 ± 1.5 dm$^2$ in surface area.

The ceramic usage obtained from the transformation is evaluated by comparing with the results reported by Fekete et al. [28]. A survey, involved 89 Belgian adults, investigated the surface area of different materials used for food preparation. The mean usage area of ceramic ware for Belgian consumers was reported to be 13.62 dm$^2$ per day. For the purpose of comparison, we adopted the complete flatware scenario, and it resulted in the mean usage area of 12.0 ± 2.5 dm$^2$ per day if ceramic articles were used on daily basis. It shows no statistical difference between the usage area of the survey and the usage area of the daily use scenario. Hence, the estimated usage of ceramic ware of the Belgian consumers in this study can to a large extent reflect the real usage of ceramic ware, and it can be used to calculate exposure.

Table 2

| Elements | Distribution of samples (No.) | Concentration (mg/L or mg/dm$^2$) |
|----------|-------------------------------|----------------------------------|
|          | Below LOQ | Above LOQ | Above permissible levels | Mean MB | Mean UB | Mean LB |
| Hollowware (664) | 564 | 100 | 0 | 0.019 | 0.024 | 0.015 |
| Pb | 610 | 54 | 0 | 0.003 | 0.005 | 0.002 |
| Cd | 539 | 180 | 6 | 0.016 | 0.017 | 0.016 |
| Flatware (539) | 438 | 101 | 3 | 0.0007 | 0.0008 | 0.0007 |

MB: medium bound, equal to the half of the LOQ; UB: upper bound, equal to the LOQ; LB: lower bound, equal to zero.

Table 3

| Years | Belgian consumer | EU consumer |
|-------|------------------|-------------|
| 2009  | 1.53             | 1.70        |
| 2010  | 1.86             | 1.86        |
| 2011  | 1.45             | 1.76        |
| 2012  | 1.64             | 1.68        |
| 2013  | 1.33             | 1.31        |
| 2014  | 1.68             | 1.39        |
| 2015  | 1.36             | 1.40        |
| 2016  | 1.47             | 1.48        |
| Mean  | 1.54             | 1.57        |
| P95   | 1.80             | 1.82        |
3.3. Intakes of lead and cadmium from ceramic ware

The deterministic intakes of lead from ceramic articles in relation to various exposure scenarios are listed in Table 5. The estimated mean (calculated with mean migrant concentration and mean consumption quantity of ceramic ware) and 95th percentile (calculated with mean migrant concentration and the 95th percentile consumption quantity of ceramic ware) intake of lead were 28 μg per day and 33 μg per day, respectively. They correspond to the mean and 95th percentile intake of 0.47 μg/kg b.w. per day and 0.54 μg/kg b.w. per day for a 60 kg Belgian adult, and of 1.40 μg/kg b.w. per day and 1.62 μg/kg b.w. per day for a 20 kg Belgian child.

The BfR [17] reported the intake of lead of the Germans ranged from 0.71 to 3.2 mg per day, much higher than the intake of lead of our study. Using release data of lead and cadmium from plates from 2016 to 2017, the BfR [18] showed that the median intake of lead of the Germans was 164 μg per day, lower than the estimate of BfR in 2005, but higher than the estimate of this study. The VKM [19] represented that the Norwegian population had the intake of lead from 4 to 30 mg per day and Rebeniak et al., [20] estimated the intake of lead of the Polish varying from 2.0–2.4 mg per day, both of which largely exceed the results of this study. It indicates that simple assumptions adopted in these studies overestimate the intake of lead.

The estimated distributions and parameters for variables are shown in Table 6. Regulatory migration data, annual consumption quantities in various years, and conversion factors from the pilot study were used to fit the corresponding distribution model. The uniform distribution fitted the allocation for ceramic ware. In contrast, every possible value from the distribution of input parameters (for example a combination of the 95th percentile migrant concentration, the 95th percentile consumption quantity of ceramic ware and the 95th percentile of conversion factors) can be adopted in the probabilistic estimate, i.e., the “95th percentile probabilistic estimate” tends to represent the worst case scenario.

As shown in Table 7, the estimated mean (calculated with mean migrant concentration and mean consumption quantity of ceramic ware) cadmium intake was 1.6 μg per day, which corresponds to 0.026 μg/kg b.w. per day for a 60 kg Belgian adult and 0.08 μg/kg b.w. per day for a 20 kg Belgian child. The estimated 95th percentile (calculated with mean migrant concentration and the 95th percentile consumption quantity of ceramic ware) cadmium intake was 1.8 μg per day, 10 times higher than that of the deterministic estimate. The significant exceedance arises from different definitions of the 95th percentile lead intake. The “deterministic 95th percentile estimate” was calculated using the mean migrant concentration and the 95th percentile of the consumption quantity of ceramic ware. In contrast, every possible value from the distribution of input parameters (for example a combination of the 95th percentile migrant concentration, the 95th percentile consumption quantity of ceramic ware and the 95th percentile of conversion factors) can be adopted in the probabilistic estimate, i.e., the “95th percentile probabilistic estimate” tends to represent the worst case scenario.

VWR: Volume-to-weight ratio; AWR: Area-to-weight ratio; SD: Standard deviation.

Table 4
Measured parameters and conversion factors of ceramic ware from pilot study.

| Ceramic ware category | Weight | Volume | Surface area | VWR | AWR | Allocation | Consumption volume | Consumption area |
|-----------------------|--------|--------|--------------|------|-----|------------|--------------------|-----------------|
| Hollowware            | kg     | L      | dm³          | L/kg | dm³/kg| L         | 0.4                | 0.80 ± 0.19     |
|                       | 0.27 ± 0.11 | 0.36 ± 0.22 | 1.3 ± 0.3 | 0.4  | 0.80 ± 0.19 | –          | ± 1.5              |
| Flatware              | 0.40 ± 0.15 | 0.41 ± 0.19 | 3.4 ± 1.1 | –    | –    | 8.5 ± 1.4 | 0.6                | –               |

Table 5
The intakes of lead (μg/day/person) in relation to various scenarios.

| Scenarios | Allocation for hollowware | Allocation for flatware | Substitution | Intakes of lead |
|-----------|---------------------------|------------------------|--------------|-----------------|
| Case 1    | 0.4                       | 0.6                    | MB           | 28              |
| Case 2    | 0.5                       | 0.5                    | MB           | 20              |
| Case 3    | 1                         | 0                      | MB           | 7.6             |
| Case 4    | 0                         | 1                      | MB           | 299             |
| Case 5    | 0.4                       | 0.6                    | UB           | 30              |
| Case 6    | 0.4                       | 0.6                    | LB           | 27              |

Table 6
The fitted distributions and parameters for inputs.

| Variables             | Distributions | Parameters        |
|-----------------------|---------------|-------------------|
| Lead concentration in hollowware (mg/L) | Log-normal | Mean = 0.0760, SD = 26.97 |
| Lead concentration in flatware (mg/dm³) | Log-normal | Mean = 0.4738, SD = 633.4 |
| Consumption quantity (kg) | Uniform | Min = 1.13, Max = 1.86 |
| VWR (L/kg) | Log-normal | Mean = 1.12, SD = 0.34 |
| AWR (dm³/kg) | Log-normal | Mean = 8.54, SD = 12.29 |

Table 7
The intakes of cadmium (μg/day/person) in relation to various scenarios.

| Scenarios | Allocation for hollowware | Allocation for flatware | Substitution | Intakes of cadmium |
|-----------|---------------------------|------------------------|--------------|--------------------|
| Case 1    | 0.4                       | 0.6                    | MB           | 1.6               |
| Case 2    | 0.5                       | 0.5                    | MB           | 1.5               |
| Case 3    | 1                         | 0                      | MB           | 1.2               |
| Case 4    | 0                         | 1                      | MB           | 1.8               |
| Case 5    | 0.4                       | 0.6                    | UB           | 2.1               |
| Case 6    | 0.4                       | 0.6                    | LB           | 1.6               |

MB: medium bound, equal to the half of the LOQ; LB: lower bound, equal to zero; UB: upper bound, equal to the LOQ;
corresponding to 0.03 μg/kg b.w. per day and 0.09 μg/kg b.w. per day for a 60 kg Belgian adult and a 20 kg Belgian child, respectively. The intake of cadmium of the Germans ranged from 0.047 to 0.28 mg per day [17]. The Norwegian population had the intake of cadmium varying from 0.23 to 0.3 mg per day [19]. Rebeniak et al. [20], showed the intake of lead of the Polish ranging from 0.15 to 0.21 mg per day. All the reported cadmium intakes were higher than that of this study.

3.4. Sensitivity analysis

The influence of allocation of consumption quantity and substitution of censored data were analyzed, as shown in Table 5. The increase of flatware consumption resulted in a growing intake of lead, and the increase of mean lead intake was circa 8 μg per day over the 10 % rise in flatware consumption. Allocating consumption quantities completely to hollowware resulted in an intake of lead of 7.6 per day; allocating consumption quantities completely to flatware gave rise to an intake of lead of 209 per day, exceeding the entire hollowware scenario by a factor of 30. It indicates that the weight of hollowware and flatware large influence the exposure to lead. Adopting the LB and UB substitution scenarios, the mean intakes of lead were respectively 27 per day and 30 per day. The substitution of censored data with LB, MB, and UB caused a gradual increase in lead intake. The substitution of censored data influences the intake of lead, but not as much as allocation does. The contribution of input parameters on the intake of lead was quantitatively analyzed (Fig. 2). The concentration of lead of flatware contributes most to lead intake, followed by the concentration of lead of hollowware. Since the migration of flatware and hollowware are significant influencing factors, lowering the migration limit of lead for flatware and hollowware, particularly the limit for flatware, is a feasible measure for the reduction of lead intake from ceramic ware.

As for cadmium, a growth of 10 % flatware consumption increased cadmium intake of 0.1 μg per day (Table 7). Allocating consumption quantities completely to flatware resulted in the intake of cadmium of 1.8 μg per day. Allocating consumption quantities to hollowware resulted in the intake of cadmium of 1.2 μg per day, as much as the entire flatware scenario. In the case of LB substitution, the mean cadmium intake was 1.4 μg per day; in the case of the UB substitution, the mean cadmium intake was 2.1 μg per day. Comparing with the intake of lead, the substitution of censored data has pronounced effects on cadmium intake. The difference arises from the low occurrence of cadmium in the monitoring samples: the percentage of censored data of cadmium is extremely high, and thereby the substitution of censored data markedly affects the estimate of cadmium. The uncertainty of cadmium intake can be largely reduced by lowering the quantification limits.

3.5. Risk characterization of lead and cadmium

Apart from the exposure derived from the use of ceramic ware, the exposure from other dietary sources is taken into account. The FASFC has estimated the exposure of the Belgian population to lead and cadmium from foodstuffs [29,30]. As shown in Table 8, the estimated mean lead intake of the Belgian adults and children were 7.7 and 8.3 μg per day, respectively, which were approx. 4 times lower than those from ceramic ware. The estimated mean intake of cadmium was 76 μg per day for a Belgian adult and 80 μg per day for a Belgian child, exceeding the intake from ceramic ware by about 50-fold. It indicates that ceramic ware considerably contributes to the dietary lead intake, whereas foodstuffs are the dominant source of the dietary cadmium intake.

The risk of exposure to lead was characterized by the margin of exposure (MOE) recommended by the EFSA CONTAM Panel. The MOE is calculated as the ratio of the BMDLs to exposure for critical effects at the systolic blood pressure chronic disease, chronic kidney disease and scores of intelligent quotient [9]. The CONTAM Panel considers a MOE of 10 or greater of negligible public concern [9]. At lower MOE, but greater than one, the risk for cardiovascular effects and nephrotoxicity is considered very low, whereas for neurotoxicity the risk is assumed to be low, but not as low as of no potential concern [9].

The MOEs in relation to deterministic and probabilistic estimates were calculated, as summarized in Table 9. In the case of deterministic estimates, the MOEs for neurotoxicity were 0.4 and 0.3 in the average and high consumers, and the MOEs for nephrotoxicity and cardiovascular effects were 1.3 and 3.2 in the average consumers, and 1.1 and 2.7 in the high consumers. In the case of probabilistic estimates, the MOEs for neurotoxicity, nephrotoxicity and cardiovascular effects were 7.1, 27, and 64 in the average consumers, and were 0.02, 0.1 and 0.3 in the high consumers. As discussed in Section 3.1, the large variation of the MOE between the deterministic and probabilistic estimates is attributable to different definitions of the 95th percentile lead intake. On the ground of the EFSA criteria, the risk of neurotoxicity for children, particularly for the high consumption subgroup, needs to be concerned. The risk of nephrotoxicity and cardiovascular effects in the general population is negligible; however, the risk for high consumers cannot be excluded. Measures should be taken to reduce the exposure to lead. As ceramic ware is a significant source of lead exposure, the exposure from ceramic ware should be substantially reduced.

Fierens et al. [31] have reported that the blood lead level of the Belgian children aged 2–6 years is 16.6 μg/L (ranging from 3 to 68 μg/L) and that the blood lead levels of the Belgian men and women are respectively 31.7 μg/L (ranging from 12 to 118 μg/L) and 21.4 μg/L (ranging from 4 to 93 μg/L). The respective BMDLs derived from blood lead levels are 12 μg/L for neurotoxicity, 15 μg/L for nephrotoxicity, and 36 μg/L for cardiovascular effects [9]. The risk of exposure to lead of the Belgian children needs to be concerned, and the risk of exposure to lead of the Belgian adults is low but not as low as of no concern. Our findings are consistent with the results of biomonitoring data.

The risk of cadmium exposure was characterized by comparing with the TWI (2.5 μg/kg b.w. per week) derived from the EFSA. The estimated mean cadmium intake was 0.18 μg/kg b.w. per week for adults and 0.52 μg/kg b.w. per week for children, accounting for circa 22 % TWI for children and 7% TWI for adults. It indicates that the risk of cadmium exposure from ceramic ware is low. However, combining the cadmium intake of the Belgian consumers from foodstuffs resulted in the exceedance of the TWI by as much as 20-fold for high consumers. The risk of cadmium exposure cannot be excluded. Fierens et al. [31] showed that the urinary cadmium level of the Belgian children aged 7–11 years was 0.06 μg/kg creatinine (varying from 0.01 to 0.22 μg/kg creatinine).
and that the urinary cadmium levels of the Belgian men and women aged 40–60 years were respectively 0.21 μg/kg creatinine (varying from 0.04 to 0.87 μg/kg creatinine) and 0.25 μg/kg creatinine (varying from 0.04 to 1.0 μg/kg creatinine). The critical urinary cadmium concentration for the derivation of TWI is 1 μg/kg creatinine [14]. The risk of cadmium is therefore negligible. Considering the long half-life of cadmium, the risk of exposure to cadmium, particularly for the old generation, cannot be fully excluded. The risk assessment of this study is basically subject to the results of the biomonitoring data. As a dominant source of cadmium exposure, the intake of ceramic derived from foodstuffs should be controlled. Although ceramic ware is not the major source of cadmium exposure, the migration limit of cadmium can be lowered based on the principle of as low as reasonably achievable (ALARA). It is acknowledged that nephrotoxicity is a critical effect for both lead and cadmium, which can result in cumulative effects of a mixture of metals [9,14,32]. The cumulative effects of lead and cadmium were not assessed in this study, and this might underestimate the real risk from multiple exposure to chemicals [33].

### 3.6. Uncertainty analysis

Overall, the intakes of lead and cadmium have been overestimated. The overestimation may arise from the following sources.

#### 3.6.1. Exposure models

The estimation employs the generally accepted model for the exposure to migrant from food contact materials. However, due to the absence of direct data for the estimation of exposure from ceramic food contact materials, the application of the exposure model causes uncertainties. First, the use of the regulatory migration data of lead and cadmium in food simulants, rather than the migration data from the food itself, gives rise to uncertainty. The direction and magnitude of the uncertainty depend on how ceramic articles are used in food preparation, serving, and storage. The amounts of metals leaching into foodstuffs rely on a number of factors such as the pH of food, contact duration with food and contact temperature [34–36]. In most cases, the release of metals from the regulatory migration tests is higher than that from the contact with real foodstuffs, according to studies on migration from ceramic articles [2–4,35,37]. Hence, the use of monitoring data on the compliance test of ceramic ware is very likely to cause an overestimation. Besides, the use of industrial data also resulted in uncertainties. Since uncertainties in relation to the usage of ceramic articles are closely related to exposure scenarios and parameter inputs, they are discussed in the following parameter and scenario uncertainty section.

#### 3.6.2. Parameter inputs

The use of industrial data from the Eurostat database gives rise to uncertainties, since the database is not designed for exposure assessment. Specifically, ceramic items recorded in the database are not exclusive for ceramic food contact articles. Indeed, it also includes other household ceramic articles such as ashtrays, soap dishes, and toothbrush holders, but these items take up less than 2% of the quantity of ceramic articles. The inclusion of other household articles causes an overestimation of exposure less than 1%. The quality of industrial data, for example missing data, confidential data and imprecise data, also has an impact on exposure. The direction and magnitude of uncertainties from these sources are unknown. Furthermore, the amount of ceramic consumption obtained in this study cannot describe the variability at the individual level. The intake of lead and cadmium was considered at the same level of all consumers but only different in body weight, which results in large uncertainties. It is also the limitation of this study. The representativeness of samples influences conversion factors and migrant concentrations. A wide range of ceramic articles with various weights, sizes, and dimensions have been included in the pilot study, which is considered to represent the ceramic ware used in Belgium. The samples involved in the monitoring program are recognized as representative of the real situation of ceramic ware on the Belgian market.

#### 3.6.3. Exposure scenarios

The default body weights used in this study also generate uncertainties. Lebacq [38] reported that the average body weight for the Belgian adults aged 18–64 was 76 kg, and thereby the adoption of default value overestimated the exposure for the Belgian adults. Moreover, the body weights of children were not specified based on age groups, which may cause an underestimation. The EFSA reported a body weight of 5 kg for the European children aged 0–12 months, of 12 kg for the European children aged 1–3 years, and of 23 kg for the European children aged 3–10 years [39]. Children aged 0–3 years rarely use ceramic ware, so the underestimation is not considerable.

The assumption on the usage of ceramic ware has been evaluated by comparing with the usage area reported by another study. The results were compatible, so the uncertainty that originates from this assumption is insignificant. The substitution of censored data can either over- or underestimate the intake of lead and cadmium. Due to the low occurrence of cadmium, uncertainties from substitutions are relatively large for cadmium intake (27%). The application of decreasing factors results in a slight overestimation since relatively conservative decreasing factors are applied.

### 4. Conclusion

This study estimated the intake of lead and cadmium derived from ceramic ware using refined scenarios. The transformation of industrial data is particularly appealing in the absence of real consumption data, which are usually difficult to obtain. The application of decreasing uncertainties...
factors has largely lowered the occurrence, arising from the assumption that ceramic articles constantly release metals at the level of the first migration test over repeated exposure.

The risk of exposure to lead of the Belgian consumers needs to be controlled. Ceramic ware assumption that ceramic articles constantly release metals at the level of below the TWI, its combination with the exposure from foodstuffs factors has largely lowered the overestimation, arising from the ALARA principle, the migration of cadmium for ceramic ware can be lowered.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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