



## Original Research Article

# Simultaneous determination of 31 elements in foodstuffs by ICP-MS after closed-vessel microwave digestion: Method validation based on the accuracy profile



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## ABSTRACT

This article describes an optimisation and validation process for the simultaneous determination of 31 elements – lithium, boron, sodium, magnesium, aluminium, potassium, calcium, titanium, vanadium, chromium, iron, manganese, cobalt, nickel, copper, zinc, gallium, germanium, arsenic, selenium, strontium, molybdenum, silver, cadmium, tin, antimony, tellurium, barium, mercury, lead and uranium – in food samples by inductively coupled plasma-mass spectrometry (ICP-MS) after closed-vessel microwave digestion. In order to improve the limits of quantification of certain elements for risk assessment in the context of the first French Infant Total Diet Study (iTDS), the analytical conditions of the multi-elemental method were optimised. The validation was conducted using the evaluation of several performance criteria such as linearity, specificity, precision under repeatability conditions, intermediate precision reproducibility and limits of quantification (LOQ), with the use of the accuracy profile. Furthermore, the method was supervised by several external quality controls (EQC). Results indicate that this method could be used in the laboratory for the routine determination of these 31 essential and non-essential elements in foodstuffs with acceptable analytical performance. A more sensitive method will be necessary for Hg and Pb to decrease at the lowest the percentage of left-censored data for this iTDS.

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## 1. Introduction

In 2011, following the second French total diet study (TDS) which involved adults and children aged over three years, the French Agency for Food, Environmental and Occupational Health & Safety (ANSES) undertook the first French TDS for infants (iTDS) (<12 months old) and toddlers ( $\geq 12$  months to <36 months old) to estimate exposure to substances of interest present in foods as consumed (prepared for normal consumption), including essential and non-essential elements. Children under 3 years of age are the population that is considered to be the most susceptible in terms of exposure to chemical substances that may be present in diet. As an example, certain chemical contaminants such as aluminium (Al) and mercury (Hg) (Arnich et al., 2012) that produce a risk of exposure in young children require increased monitoring. But minerals such as sodium (Na) also revealed excess for children

over three years in the conclusions of the first French TDS (Leblanc et al., 2005).

The TDS consisted of three major steps: (i) food sampling and sample preparation as consumed, (ii) analysis of the samples, and (iii) dietary exposure assessment by combining occurrence data with national consumption data. The risk characterisation of chemicals in food is based on the comparison between dietary exposure and the relevant health-based guidance value. The estimated intakes are compared to nutritional and toxicological reference values such as the Recommended Daily Intake (RDI) for nutrients and the Provisional Tolerable Weekly Intake (PTWI) for toxic elements (WHO, 1995).

The analytical method may have an excessively high limit of detection (LOD) or quantification (LOQ). This can limit the usefulness of obtained data for dietary exposure assessments. Indeed, according to international guidelines (GEMS/Food-Euro, 1995) for food items with a censoring rate (a level below the LOQ or LOD) of at least 60%, several scenarios were found with regard to the concentrations such as the lower bound assumption (LB), whose non-detected values are estimated to be 0 and the values

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detected, but not quantified, are estimated to be equal to the LOD, and the upper bound assumption (UB), whose non-detected values are estimated to be equal to the LOD and the values detected but not quantified are estimated to be equal to the LOQ. Thus, this process may bring uncertainty in the risk assessment, which increases for larger proportions of censored data.

During the second French TDS, four macromineral elements, sodium (Na), magnesium (Mg), calcium (Ca) and potassium (K), were analysed using a flame atomic absorption spectrometry (FAAS) method (Chekri et al., 2010), 21 elements were analysed using inductively coupled plasma mass spectrometry (ICP-MS) in standard mode (Millour et al., 2011b) and three other elements (chromium (Cr), iron (Fe), selenium (Se)) were also determined by ICP-MS equipped with collision cell technology (CCT) by ICP-MS. To better assess the levels of dietary exposure in children in the context of this first French iTDS, the National Reference Laboratory (NRL) for trace elements aimed to develop and validate one analytical multi-elemental method by ICP-MS enabling simultaneous determination of high mineral content and of low trace element content in foodstuffs, with a suitable LOD and LOQ. Therefore, taking into account the criteria performance obtained in the second French TDS, the requirements of low LOD and LOQ were necessary, in particular for certain trace elements usually found in low levels in food, to decrease the percentage of left-censored data (data < LOQ or < LOD) which can lead bias in the evaluation of exposure. Thus, the ANSES Expert Committee on Contaminants requested that a special effort be made with regard to the following elements in order to reduce, if possible, the limits of quantification of arsenic (As) by a factor of 10 (0.001 mg kg<sup>-1</sup>), cadmium (Cd) and Al by a factor of 2 (Cd: 0.001 mg kg<sup>-1</sup> and Al: 0.209 mg kg<sup>-1</sup>), Hg by a factor of 5 at least (0.002 mg kg<sup>-1</sup>), and lead (Pb) and Se by a factor of 5–10 (Pb: 0.001–0.0005 mg kg<sup>-1</sup> and Se: 0.02–0.01 mg kg<sup>-1</sup>).

The aim of the present work was to develop and validate an ICP-MS method after closed-vessel microwave digestion for the determination of 31 essential and non-essential trace elements (lithium (Li), boron (B), Na, Mg, Al, K, Ca, titanium (Ti), vanadium (V), Cr, Fe, manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), gallium (Ga), germanium (Ge), As, Se, strontium (Sr), molybdenum (Mo), silver (Ag), Cd, tin (Sn), antimony (Sb), tellurium (Te), barium (Ba), Hg, Pb and uranium (U)), and to demonstrate that this method could be applied to about 300 food matrices mainly consumed by French infants and toddlers, while obtaining adequate analytical performances. Thus, the choice of isotopes, analysis mode, specific internal standards by element and range of calibration were optimised. The validation was based mainly on the accuracy profile procedure (AFNOR, 1998, 2010). A comparison of the LOQs was made between the accredited multi-elemental method used for the second French TDS (21 elements) (Millour et al., 2011b) and the developed method here. The results obtained with external quality controls (EQC) during this study have also been presented.

## 2. Material and methods

### 2.1. Reagents and gas

All solutions were prepared with analytical reagent grade chemicals and ultrapure water (18 MΩ cm) obtained by purifying distilled water with the Milli-QTM PLUS system associated with an Elix 5 pre-system (Millipore S.A., St Quentin-en-Yvelines, France).

- Nitric acid: Suprapur HNO<sub>3</sub> (67% v/v) and Rectapur HNO<sub>3</sub> (54% v/v) was purchased from VWR (Fontenay-sous-Bois, France).
- Standard solutions: Standard stock solutions containing 1000 mg L<sup>-1</sup> of each element and 2500 mg L<sup>-1</sup> of Ca, Na, K

and Mg were purchased from Analytika (Prague, Czech Republic) or from Ultra Scientific (North Kingstown, USA) and were used to prepare calibration standards. Working standards were prepared daily in 6% of HNO<sub>3</sub> (67%, v/v) and were used without further purification.

- Tuning solution: a 10 mg L<sup>-1</sup> multi-element solution (Agilent Technologies, Courtaboeuf, France) was used to prepare a tuning solution containing several elements such as Li, yttrium (Y), thallium (Tl), Co, cerium (Ce), that made it possible to cover a wide range of masses.
- Factor P/A solution: to obtain a linear response of the detector between pulsed and analogue modes, a 2.5 mg L<sup>-1</sup> to 20 mg L<sup>-1</sup> factor P/A multi-element solution (Agilent Technologies, Courtaboeuf) was used.
- Internal standard solutions and gold (Au): 1000 mg L<sup>-1</sup> standard stock solutions of scandium (Sc), Y, indium (In), rhenium (Re), bismuth (Bi) and Au were purchased from Analytika (Prague, Czech Republic) or from Ultra Scientific (North Kingstown, USA). To avoid the mercury memory effect, gold chloride was added to the internal standard solutions to maintain Hg as Hg<sup>2+</sup> in solution (Lo and Wai, 1975). The internal standard solutions were added to all samples, with calibration standards and blanks at the same concentration, to obtain information on changes in sensitivity in different mass regions.
- Ultrapure grade carrier (99.9995% pure argon (Ar) and helium (He)) was supplied by Linde Gas (Montereau-Fault-Yonne, France).

### 2.2. Reference materials and samples

The certified reference materials (CRMs) used to validate the method were all purchased from LGC Standards (Molsheim, France) or from Courtage Analyses Services (Mont-Saint-Aignan, France). The CRMs analysed were: SRM 1548a (Typical diet) and SRM 1566b (Oyster tissue) from the National Institute of Standards and Technology, TORT-2 (Pancreas lobster) from the National Research Council Canada, NIM-GBW 10014 (Cabbage) from the Institute of Geophysical and Geochemical Exploration of China and IAEA 155 (Whey powder) from the International Atomic Energy Agency.

The method was optimised and validated by analysing various types of matrices from different foodstuffs of animal and plant origin, beverages and diverse infant food matrices (honey, pork, tuna, milk, infant formula, breakfast cereals, yoghurt, baby juice, pot vegetable meat or fish, beef, shrimp, carrot, etc.). The food matrices used were chosen among the 293 composite food samples from the first French iTDS.

### 2.3. Sample digestion procedure

Samples were digested using a Multiwave 3000 microwave digestion system (Anton-Paar, Courtaboeuf, France), equipped with a rotor for eight 80 mL quartz vessels (operating pressure, 80 bar). The sample digestion procedure was performed according to the NF EN 13805 standard (AFNOR, 2002b) and had previously been optimised (Noël et al., 2003). Before use, the quartz vessels were decontaminated with 6 mL of 50% HNO<sub>3</sub> (54%, v/v) in the microwave digestion system, then rinsed with ultrapure water and dried in a 40 °C oven. From 0.3 to 0.5 g (for dry food, i.e. powdered milk, freeze-dried food...) and from 0.5 to 2.0 g (for fresh diet, i.e. yoghurt, pot vegetable meat...) of dietary samples were weighed precisely in the quartz digestion vessels and wet-oxidised with 3 mL ultrapure water and 3 mL suprapur HNO<sub>3</sub> (67%) in the microwave digestion system. One randomly selected vessel was filled with reagents only and taken through the entire procedure as a blank. After cooling to room temperature, sample solutions

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