Environment International xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Exposure of children to metals via tap water ingestion at home: Contamination and exposure data from a nationwide survey in France☆

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ARTICLE INFO

Article history:
Received 1 February 2016
Received in revised form 31 May 2016
Accepted 8 June 2016
Available online xxxx

Keywords:
Inorganic
Drinking-water
Metal
Metalloid
Environmental exposure

ABSTRACT

29 inorganic compounds (Al, As, B, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cu, Fe, Gd, K, Mg, Mn, Mo, Na, Nd, Ni, Pb, Sb, Se, Sr, Tl, U, V and Zn) were measured in the tap water of 484 representative homes of children aged 6 months to 6 years in metropolitan France in 2008–2009. Parents were asked whether their children consumed tap water. Sampling design and sampling weights were taken into account to estimate element concentrations in tap water supplied to the 3,581,991 homes of 4,923,058 children aged 6 months to 6 years. Median and 95th percentiles of concentrations in tap water were in μg/L: Al: <10, 48.3, As: 0.2, 2.1; B: <100, 100; Ba: 30.7, 149.4; Ca: 85,000, 121,700; Cd: <0.5, <0.5; Ce: <0.5, <0.5; Co: <0.5, 0.8; Cr: <5, <5; Cu: 70, 720; K: 2210, 6740; Fe: <20, 46; Mn: <5, <5; Mo: <0.5, 1.5; Na: 14,500, 66,800; Ni: <2, 10.2; Mg: 6500, 21,200; Pb: <1, 5.4; Sb: <0.5, <0.5; Se: <1, 6.7; Sr: 256.9, 1004; Tl: <0.5, <0.5; U: <0.5, 2.4; V: <1, 1; Zn: 53, 208. Of the 2,977,123 young children drinking tap water in France, some were drinking water having concentrations above the 2011 World Health Organization drinking-water quality guidelines: respectively 498 (CI 95%: 0–1484) over 700 μg/L of Ba; 121,581 (CI 95%: 7091–236,070) over 50 mg/L of Na; 2044 (CI 95%: 0–6132) over 70 μg/L of Ni, and 78,466 (17,171–139,761) over 10 μg/L of Pb. Since it is representative, this tap water contamination data can be used for integrated exposure assessment, in conjunction with diet and environmental (dust and soil) exposure data.

1. Introduction

Water is sourced for potabilization from various supplies; its quality depends on water resource origin and possible contamination, as well as on subsequent water treatment processes (such as filtration, aeration, and chemical precipitation) and interaction with distribution system equipment (Dinelli et al., 2012). Drinking-water is essential to human health and French child consumers drink, on average, 250 mL of tap water per day (Fantino and Gourmet, 2008), (Lioret et al., 2010). Tap water is a source of trace elements that are essential to life, such as Ca, Mg, Fe, and Zn – yet can also be contaminated by heavy metals or metalloids such as As, Pb, Tl, or U, which are recognized to be toxic (Villaescusa and Bollinger, 2008), (Zietz et al., 2010), (Peter and Viraraghavan, 2005). Heavy metals can be toxic at very low levels of concentration. Ingested metals can accumulate in the body, affecting the nervous system (especially in children during the period of maximum brain growth) disrupting the normal functioning of internal organs and acting as cofactors in other diseases. Certain heavy metals have recently been associated with infertility (Giaccio et al., 2012). Assessment of human exposure to heavy metals is increasingly being investigated in all environmental media - such as dust and soil (Ibanez et al., 2010) (Mielke et al., 2010), (Glorennec et al., 2012), and water (Dinelli et al., 2012), (Nahar and Zhang, 2012), (Stalder et al., 2012) in order to evaluate the impact of environmental pollution on public health, particularly in children. Twelve metals or metalloids are regularly quantified in water to verify compliance with the concentration limit imposed by the EU Drinking Water Directive (European Directive 1998/83EC, 1998). Assessing health risks linked to metal ingestion via water requires local contamination data (in addition to values for ingested quantities) with a water sampling strategy that is adapted to end-consumer exposure.

The objective of this study is to determine the concentrations of the metals and other elements to which children are exposed through drinking tap water at home in France. 29 elements were measured (Al, As, B, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cu, Fe, Gd, K, Mg, Mn, Mo, Na, Nd, Ni, Pb, Sb, Se, Sr, Tl, U, V and Zn) within a perspective of representative and integrative chronic exposure assessment.

☆ Funding: This research was funded by the French ministries in charge of health, ecology, and housing. Funding bodies were not involved in study design, data analysis or interpretation.

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http://dx.doi.org/10.1016/j.envint.2016.06.009
0160-4120/

2. Material and methods

2.1. Study design

Our dwellings sample was derived from a national lead poisoning survey (n = 3623 children) (Etchevers et al., 2014), using a sampling design allowing for the provision of population-based estimates. The design of the initial lead poisoning survey was a two-stage sampling, stratified at the first stage: the primary sampling units were hospitals within which the children were the second sampling units. Detailed inclusion procedures and representativeness have been described and discussed by Etchevers et al. (2014). A subsample of 484 children and corresponding housing units was then selected and investigated. Homes were main residences (as opposed to second homes) in metropolitan France inhabited by at least one child aged 6 months–6 years located as described (Fig. 1). The design and sampling weights were taken into account in a design-based analysis (Lumley, 2010a) to ultimately achieve population-based estimates of concentrations of metals and elements in the tap water consumed by the whole population. A post-stratification on age, location and type (single or multi-units) was performed on the sampling weights to increase the precision of estimators, as described in details by Lucas et al. (2014).

Environmental investigations (measurements and questionnaire) were carried out at the 484 homes between October 2008 and August 2009. For statistical calculations, values below the Limit of Quantification (LOQ) were assigned to the value of LOQ/2. Statistical analyses were performed using the "survey" R® 2.9.0 software package (Lumley, 2010b).

An individual written report on the results was sent to each family. Prior authorisation (Authorisation No. 908326) was obtained from the French Data Protection Authority (Commission Nationale de l'Informatique et des Libertés). Although already published elsewhere, results concerning lead concentration in water (Lucas et al., 2012), are also partially reported here to ensure consistency.

2.2. Tap water use as drinking-water

A questionnaire was used to collect information about child behaviour. In particular, parents were asked whether or not their child consumed tap water.

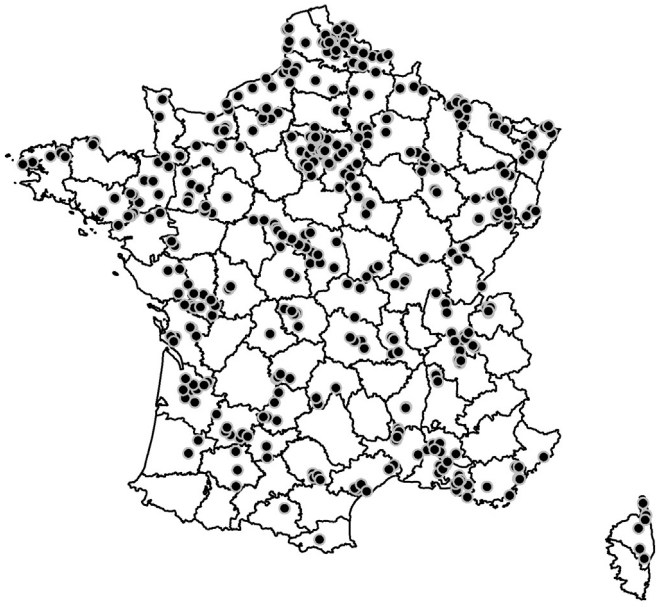

Fig. 1. Location of the 484 tap water samples, France 2008–2009.

2.3. Tap water measurement

2.3.1. Drinking-water sampling

Tap water was sampled in the kitchen at random times of day, from Monday to Friday. Taps were long-flushed. Following a controlled 30 min stagnation time, a 2 L sample was taken, using a high density polyethylene (HDPE) bottle, homogenized by shaking, and 250 mL was immediately transferred into another HDPE bottle containing 1% HNO_3 (65–70%) (Agence Française de Normalisation, 2004).

2.3.2. Analysis

The toxic metals and metalloids that could be analysed together, and whose performance level was high enough to be quantifiable, were analysed in tap water using an inductively-coupled plasma mass spectrometer (ICP-MS Agilent Technology 7500ce equipped with a quadrupole mass filter and an octopole reaction cell) by ICPMS. The elements were Al, As, B, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cu, Fe, Gd, K, Mg, Mn, Mo, Na, Nd, Ni, Pb, Sb Se, Sr, Tl, U, V, and Zn. The system is equipped with an autosampler (CETAC ASX-510), a micro nebulizer (microflow) and a Scott chamber. The plasma is energized by radio frequency (27.12 MHz). ICP/MS settings were as follows: sample feed rate of 0.3 mL/min, RF power of 1550 W and plasma gas flow rate of 15 L/min. The makeup gas flow rate was 0.2 L/min, and the carrier gas flow rate was 0.8 L/min. The mass spectrometer interface included a nickel sampling cone and a nickel skimmer cone.

29 elements were analysed simultaneously. Total duration of data acquisition was 202 s per sample. The acquisition parameters for each element were isotopic mass in atomic mass unit (amu), gas mode and acquisition time u(s): Be (9, no-gas mode, 0.3); B (10, no-gas mode, 0.3); Na (23, He mode, 0.3); Mg (24 and 26, He mode, 0.3); Al (27, He mode, 0.9); K (39, He mode, 0.3); Ca (43 and 44, no-gas mode, 0.3); V (51, He mode, 0.3); Cr (52 and 53, He mode, 0.3); Fe (54, 56 and 57, He mode, 0.3); Mn (55, He mode, 0.3); Co (59, He mode, 0.3); Ni (60 and 62, He mode, 0.3); Cu (65, He mode, 0.3); Zn (66, He mode, 0.3); Zn (67 and 68, He mode, 0.9); As (75, He mode, 1.5); Se (78, H_2 mode, 1.5); Sr (86 and 88, no-gas mode, 0.3); Mo (95 and 97, no-gas mode, 0.3); Cd (111 and 114, no-gas mode, 0.9); Sb (121 and 123, no-gas mode, 0.3); Ba (135 and 137, no-gas mode, 0.3); Ce (140, no-gas mode, 0.3); Nd (143 and 146, no-gas mode, 0.3); Gd (157, no-gas mode, 0.3); Tl (203 and 205, no-gas mode, 0.3); Pb (206, 207 and 208, no-gas mode, 0.3); Bi (209, no-gas mode, 0.9); U (238, no-gas mode; 0.3). In order to suppress inter-element interference, we used corrected equations: one for Fe quantification (the isotope 54 amu was corrected by isotope 52 amu of Cr), one for Cd quantification (the isotope 114 amu was corrected by isotope 118 amu of Sn) and for In quantification (the isotope 115 amu was corrected by isotope 118 amu of Sn (equations are not shown).

To ensure identical operating conditions, both an external calibration series and an internal standard [Ge (72 amu), Sc (45 amu), Rh (103 amu), In (115 amu) or Ir (193 amu)] were used for each sample sequence. The internal standard for each metal was chosen in relation to closed mass and ionization potential. The calibration series was prepared using the same acids as were used for samples. Calibration curves were obtained using at least five points. Some of the samples had to be diluted since they fell outside the range of the calibration. The quantification limits (LOQ) for each element are shown in Table 1, with the exception of Bi, Gd and Nd: 0.1 μg/L, 0.1 μg/L and 0.5 μg/L respectively.

2.3.3. Quality control

Analytical blanks and National Institute of Standards and Technology NIST 1643 were used for water analysis quality control. These control samples were inserted into all analysis series, and elements were determined in the same manner as for samples. No contamination of Sb by flask was observed. The laboratory has had French accreditation (Comité Français d'accréditation (COFRAC), 2013) for the 29 elements

Please cite this article as: Le Bot, B., et al., Exposure of children to metals via tap water ingestion at home: Contamination and exposure data from a nationwide survey in France, Environ Int (2016), http://dx.doi.org/10.1016/j.envint.2016.06.009