Environmental Pollution 240 (2018) 599–606

Contents lists available at ScienceDirect

Environmental Pollution

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

Environmental tin exposure in a nationally representative sample of U.S. adults and children: The National Health and Nutrition Examination Survey 2011–2014☆

Hans-Joachim Lehmler [a], Manuel Gadogbe [a], Buyun Liu [b], Wei Bao [b, *]

[a] Department of Occupational & Environmental Health, College of Public Health, University of Iowa, Iowa City, IA, United States
[b] Department of Epidemiology, College of Public Health, University of Iowa, Iowa City, IA, United States

ARTICLE INFO

Article history:

Keywords:
Urinary tin
Human exposure
U.S. population

ABSTRACT

Tin is a naturally occurring heavy metal that occurs in the environment in both inorganic and organic forms. Human exposure to tin is almost ubiquitous; however, surprisingly little is known about factors affecting environmental tin exposure in humans. This study analyzed demographic, socioeconomic and lifestyle factors associated with total urinary tin levels in adults (N = 3522) and children (N = 1641) participating in the National Health and Nutrition Examination Survey (NHANES) 2011–2014, a nationally representative health survey in the United States. Urinary tin levels, a commonly used biomarker of environmental tin exposure, were determined by inductively coupled plasma mass spectrometry (ICP-MS). Detection frequencies of tin were 87.05% in adults and 91.29% in children. Median and geometric mean levels of urinary tin in the adult population were 0.42 μg/L and 0.49 μg/L, respectively. For children, median and geometric mean levels of urinary tin were 0.60 μg/L and 0.66 μg/L, respectively. Age was identified as an important factor associated with urinary tin levels. Median tin levels in the ≥60 year age group were almost 2-fold higher than the 20–39 year age group. Tin levels in children were 2-fold higher than in adolescents. Race/ethnicity and household income were associated with tin levels in both adults and children. In addition, physical activity was inversely associated with urinary tin levels in adults. These results demonstrate that total tin exposures vary across different segments of the general U.S. population. Because the present study does not distinguish between organic and inorganic forms of tin, further studies are needed to better characterize modifiable factors associated with exposures to specific tin compounds, with the goal of reducing the overall exposure of the U.S. population.

1. Introduction

Tin is a naturally occurring element that is found in both inorganic and organic forms in the environment. It is classified by the WHO as potentially toxic (WHO, n.d.-a). Acute health effects associated with exposure to inorganic tin, in particular the consumption of food products contaminated with high levels of tin, include acute gastrointestinal illnesses (reviewed in: Blunden and Wallace, 2003; Ostrakhovitch, 2014). In humans, the severity of adverse gastrointestinal effects are not necessarily associated with the tin levels, but with the chemical form of tin present in foodstuff (Boogaard et al., 2003). Studies investigating a link between chronic, low level exposures to inorganic tin and adverse human health outcomes are limited. A cross-sectional study reported a positive but non-significant association between urinary tin concentration and diabetes (Feng et al., 2015). In coke oven workers in China, urinary tin levels were associated with elevated fasting plasma glucose levels (Liu et al., 2016). Further studies are therefore needed to assess if low level, chronic exposures to inorganic forms of tin are associated to adverse outcomes in humans. In contrast to inorganic tin, organotin compounds are considered more toxic, and several organotin compounds, in particular tributyltin, have been identified by laboratory studies as environmental obesogens (Grün, 2010; Heindel et al., 2017; Zuo et al., 2011). Epidemiological studies of adverse outcomes associated with exposure to organotin compounds are also limited, and suggest a link between organotin

☆ This paper has been recommended for acceptance by Dr. Chen Da.

* Corresponding author. Department of Epidemiology, College of Public Health, University of Iowa, 145 N, Riverside Drive, Room S431 CPHB, Iowa City, IA, 52242, USA.
E-mail address: wei-bao@uiowa.edu (W. Bao).

https://doi.org/10.1016/j.envpol.2018.05.019
0269-7491/

compounds and adverse developmental outcomes (Rantakokko et al., 2013, 2014).

The total global production of tin reached 265,000 tons in 2010, and slightly decreased to 253,000 tons in 2011 (U.S. Geological Survey, 2010). A 2017 survey estimates that 422,900 tons of tin were used globally in 2016 (ITRI, 2017). Tin is used in a range of industrial and consumer applications, including tin-plated food and beverage cans, containers, electrical equipment, and transportation and construction materials. Inorganic tin compounds are also used in the glass and ceramic industries, and for the formulation of soaps and perfumes. Stannous chloride ($SnCl_2$) is the most frequently used form of inorganic tin, and it is listed by the United States Food and Drugs Administration as a Generally Regarded as Safe (GRAS) food additive for human consumption (FDA, n.d.). Organotin compounds were used as stabilizers for polyvinyl chloride (PVC), biocides, and catalysts for the synthesis of polyurethanes and silicones (ENVISA, n.d.); however, the production and use of organotin compounds for many applications, such as anti-fouling paints used on ships, are now banned globally (ENVISA, n.d.; IMO, n.d.).

Inorganic tin is released into the environment from both natural and anthropogenic sources. In contrast, organotin compounds are mainly released from anthropogenic sources (ATSDR, n.d.; Carlin Jr, n.d.; Ostrakhovitch, 2014). Inorganic tin is considered relatively immobile in the environment according to the Agency for Toxic Substances and Disease Registry (ATSDR, n.d.). Human exposures can occur by inhalation, ingestion, or dermal absorption (WHO, n.d.-c). Consumption of food, in particular canned food and beverages, is considered to be a major source of exposure of humans to inorganic forms of tin (Blunden and Wallace, 2003; Ostrakhovitch, 2014; WHO, n.d.-a). Dietary intake levels of inorganic tin from canned foods can vary widely because tin levels in canned food are, for example, affected by the use of lacquered vs. unlacquered cans, the acidity of the food, storage conditions, and the presence of agents that influence the extent of tin dissolution from the tin coating (Greig and Pennington, n.d.; IPCS, n.d.). Interestingly, several studies reported that the consumption of canned food is not associated with urinary tin levels (Hayashi et al., 1991; Shimbo et al., 2007, 2013; Yang et al., 2015). Diet is also a major route of exposure to organotin compounds. Seafood accounts for a majority of intake of organotin compounds (Airaksinen et al., 2010; Choi et al., 2012; Filipkowska et al., 2016; Guérin et al., 2007); however, a recent analysis of diet samples from Portugal demonstrates that for the majority of samples the estimated daily intakes of organotin compounds were below the tolerable daily intake of 100 ng/kg/d (Sousa et al., 2017).

Tin was detected with a 36% frequency in a small, recent survey of treated drinking water samples from the United States (Glassmeyer et al., 2017), with median and maximum tin levels of 6.4 μg/L and 15.9 μg/L, respectively. These levels are comparable to tin levels reported for drinking water in earlier studies and suggest that drinking water is not a significant source of tin exposure in the general population (Schroeder et al., 1964; WHO, n.d.-c). Similarly, air makes only a small contribution to the daily intake of tin in the general population (Schroeder et al., 1964; WHO, n.d.-c). Occupational exposures can result in an increased exposure to tin, as assessed using urinary tin levels (Julião et al., 2007; Liu et al., 2016). Inorganic tin is poorly adsorbed by humans, with only 3% of inorganic tin being absorbed from a diet supplemented with 50 mg tin per day (Johnson and Greger, 1982). Studies in rats and human subjects show that feces is the major route of excretion of inorganic tin following oral exposure, mostly because of the poor absorption of tin in the gastrointestinal tract (Benoy et al., 1971; Calloway and McMullen, 1966; Hiles, 1974; Winship, 1988). However, based on animal studies, inorganic tin is primarily eliminated from the systemic circulation *via* the urine (Hiles, 1974; Mance et al., 1988). Several factors, such as the route of administration, the dose and the form of tin, may explain differences in the fecal *vs.* urinary excretion of tin observed in animal studies (Hiles, 1974). Like inorganic forms of tin, organotin compounds can also be absorbed in the gastrointestinal tract and are eliminated with the feces, bile or urine in a manner that is highly compound specific.

Overall, there is contemporary exposure of humans to both inorganic and organic forms of tin. Dietary intake of tin is influenced by several factors, such as age and the consumption of food and beverages contaminated with tin compounds. However, factors influencing the urinary elimination of both inorganic and organic forms of tin, and thus the toxicologically relevant internal tin dose, have not been investigated previously in the general U.S. population. Here, we analyzed associations between potential demographic, socioeconomic and lifestyle factors and urinary levels of total tin determined by ICP-MS in U.S. adults and children participating in NHANES 2011–2014, a nationally representative health survey in the United States.

2. Materials and methods

2.1. NHANES survey

NHANES is a nationally representative survey of the non-institutionalized U.S. population that is administered by the National Center for Health Statistics at the Centers for Disease Control and Prevention (CDC) (CDC, n.d.-a). The survey collects a broad range of information, for example on the demographics, socioeconomic status, diet, lifestyle, and medical conditions; performs extensive health examinations; and collects specimens for laboratory tests from study participants. NHANES data are released in 2-year cycles. NHANES has been approved by the National Center for Health Statistics Ethics Review Board and written informed consent was obtained from all participants. Additional information about NHANES is available elsewhere (CDC, n.d.-a). For this study, we analyzed data from the NHANES 2011–2012 and 2013–2014 cycles, because tin levels were only measured in these two cycles. The final study population consisted of 5163 participants (children and adults) who had data available on urinary tin concentrations.

2.2. Total tin analysis

Concentrations of total tin in urine samples were measured using ICP-MS at the Inorganic and Radiation Analytical Toxicology Division of Laboratory Sciences, National Center for Environmental Health, CDC (CDC, n.d.-b). Briefly, all urine samples as well as blanks, calibrators, and QC samples were diluted 1:9 (*v*/*v*) with a diluent containing 10 μg /L of the internal standards iridium and rhodium for multi-internal standardization and 500 μg/L gold in 2% (*v*/*v*) nitric acid prior to analysis. The analysis of total tin in the urine samples was performed in the standard mode using an ELAN DRC II ICP-MS equipped with an ESI SC4-DX auto sampler. The lower limit of detection (LLOD) of the urinary tin analysis was 0.090 μg/L (CDC, n.d.-b). For urinary tin levels below the LLOD, an imputed fill value was assigned by CDC staff as the LLOD divided by square root of 2.

2.3. Potential factors influencing urinary tin levels

Information on age, gender, race/ethnicity, education, family income, smoking status, alcohol intake, physical activity, and medical conditions was collected by NHANES using standardized questionnaires. Race/ethnicity was categorized as non-Hispanic white, non-Hispanic black, Hispanic (Mexican and non-Mexican Hispanic), and other race/ethnicity. Education was grouped as less than high school, high school, and college or higher. Family income-