

Dietary tin intake and association with canned food consumption in Japanese preschool children

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Abstract

Objectives Dietary intake of tin has seldom been studied in children although they probably have a high intake. This study was initiated to investigate dietary tin intake (Sn-D) of children in Japan.

Methods In this study, 24-h food duplicate samples were collected from 296 preschool children in Miyagi prefecture, Japan. Sn in the samples were analyzed by inductively coupled-plasma mass spectrometry, after homogenization and wet digestion.

Results Sn-D by the children was low, with 4.2 µg/day as a median. The distribution was however wide, from 0.4 µg/day up to >3 µg/day. Canned foods were the major dietary Sn source, whereas rice contributed essentially little. Sn-D among canned food consumers was 30.2 µg/day as a geometric mean (10.6 µg/day as a median), whereas Sn-D among the non-consumers of canned foods was distributed log-normally, with 3.3 µg/day as a geometric mean (2.5 µg/day as a median). Sn levels in urine did not

differ between children who consumed canned foods on the day previous to urine collection and those who did not. The Sn-D was far below the provisional tolerable weekly intake (14 mg/kg body weight/week) set by the 2001 Joint FAO/WHO Expert Committee. Nevertheless, children took more Sn than adults when compared on a body-weight basis.

Conclusions Canned foods were the major source of dietary Sn intake for preschool children studied. Thus, median Sn-D was higher for the canned food consumers (10.6 µg/day) than for non-consumers of canned foods (2.5 µg/day). Sn-D by canned food-consuming children was, however, substantially lower than the provisional tolerable weekly intake. No difference was detected in Sn levels in urine between canned food-consuming and non-consuming children.

Keywords Canned foods · Children · Dietary intake · Inductively coupled plasma-mass spectrometry · Japan · Preschoolers · Tin · Urine

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Introduction

Inorganic tin (Sn) is a unique metal in the sense that it has been considered least toxic to humans [1], possibly because absorption from the gastrointestinal (GI) tract is limited and passage through the tract is rapid [2] and accumulation in the tissue is low [3], although the half life is substantially long, e.g., 29 days in mice [4]. Sn may, however, cause acute GI tract problems, such as abdominal distension, pain and vomiting both in experimental animals [5] and in humans [6] when ingested at a high dose [7]. Accordingly, a provisional tolerable weekly intake was set at 14 mg/kg body weight/week by the Joint FAO/WHO Expert Committee in 1989, and the provisional tolerable weekly intake

was kept unchanged when re-evaluated in 2001 [8], whereas the rationale for the provisional tolerable weekly intake was questioned in a later meeting [2]. Possibly because of such low toxicity, Sn has been extensively employed for inside as well as outside coating of food cans for packaging and preservation of various foods [6].

Thus, Sn use may result in direct contact with foods (including liquid foods such as drinks and soup) for general populations. The migration of Sn from the inside wall of cans to edible contents may take place subject to acidity of the liquid inside the can [6]. Subsequently, a number of studies has been carried out in various industrialized (and therefore canned food consuming) countries to examine the extent of Sn ingestion through daily foods [9–13]. The daily Sn intake appeared to be not in excess of the provisional tolerable daily intake in any populations studied, as will be discussed later. The existing results are, however, all on healthy adult populations and have no focus on possible sensitive populations such as children, although it might be the case that children ingest more Sn on a body weight basis than adult people as, for example, children would take more canned drinks [7].

The present study was initiated to quantify dietary Sn intake of children in Japan by use of food duplicate sample collections [14], the method which allows instrumental determination of Sn in collected samples. Attention was given to possible contribution of intakes of canned foods (canned juice in particular) and that of boiled rice as a staple food. Levels of daily dietary tin intake (Sn-D) were compared between canned food consumers and non-consumers of canned foods. Comparison on Sn intake was also made to adult populations [12] in the same country of Japan.

Materials and methods

Ethical issues

The protocol of this study was approved by the Ethics Committee of Miyagi University. A guardian of each child (a mother in most cases) provided an informed consent in writing on behalf of her (his) child.

Food duplicate sample collection, and analysis for Sn in the duplicates, boiled rice and urine

The food duplicate collections were conducted in winter seasons (December–March) in 2001–2004 in Miyagi prefecture on the Pacific coast in northeast Japan, the prefecture where there has been no known environmental pollution with toxic metals such as cadmium or lead. In total, 296 preschool children (159 boys and 137 girls) aged of 3–6 years in 15 kindergartens participated in the study

by offering 24-h food duplicate samples [14, 15]. In addition, a majority (217 children) offered first-morning urine samples on the next day of survey. Participation of kindergartens was on a voluntary basis, whereas all children in the volunteering kindergartens joined the survey.

The procedures for collecting 24-h food duplicate samples and pre-treatment for instrumental analysis were previously described in detail [12, 16–18]. In short, each mother of the children was asked to prepare a food duplicate sample by cooking for a hypothetical child in her family. The same food items at the same portions her child consumed on the entire study day (i.e., three meals and any snacks including energy-free tea, water and other drinks) were saved in metal leakage-free plastic containers.

After manual separation for each food item (by use of metal leakage-free utensils) and weighing of separated food items (weight being recorded), a total homogenate (including a major portion of boiled rice) was prepared, and a portion of the homogenate was subjected to Sn analysis after wet-digestion by heating in presence of extra-pure mineral acids. A minimum portion (a few gram) of the boiled rice was digested separately for determination of Sn in boiled rice. Energy intake was estimated from the food weight by use of the 2010 version of food composition tables [19]. The child's guardian who prepared the food duplicate sample (his/her mother in most cases) was asked to submit food menus of the day and to report if her child took canned foods (including canned drinks) in three meals or snack.

Analyses for Sn in food homogenate, boiled rice and in urine samples were conducted by inductively coupled-plasma mass spectrometry under the conditions previously detailed [12]. The material limits of determination (LOD) were 0.05 $\mu\text{g Sn/l}$ for wet digests of the homogenate and boiled rice, and 0.03 $\mu\text{g Sn/l}$ for urine wet digests, which were equivalent to 0.05 $\mu\text{g Sn/day}$ for food duplicate, 0.01 $\mu\text{g Sn/kg}$ for boiled rice, and 0.03 $\mu\text{g Sn/l}$ for urine. Daily dietary intake of Sn (Sn-D) was calculated from Sn in the unit weight of each homogenate and a total weight of the homogenate.

Creatinine (CR) and a specific gravity of urine (SG) were measured by colorimetry and refractometry, respectively. Sn in urine as observed (Sn- U_{ob}) was further corrected for CR (Sn- U_{cr}) [20] or for SG (SG of 1.016 was taken as a standard) (Sn- U_{sg}) [21], respectively.

Statistical analysis

A preliminary statistical analysis of Sn-D showed that the distribution of Sn-D as a whole was markedly skewed, and was not normal or log-normal. Accordingly, a median was taken as a representative parameter in general, and Mann–Whitney test in addition to *t*-test was employed for detection of possible difference in distributions. An arithmetic

mean (AM) and a geometric mean (GM) were also calculated when adequate. When Sn-D levels in some cases were remarkably deviated from Sn-D in other samples, Smirnov test for extreme values was applied; the cases were identified as extremely deviated with $p < 0.05$, and such cases were excluded from further statistical evaluation.

Results

Demographic characters of participating children and adequacy of 24-h collection of food duplicate samples

The participating subjects were 296 children in total (159 boys and 137 girls) at the ages of 3–6 years, as previously described in detail [22]. The average energy intake estimated from the food duplicate samples was 1382 (3 year-olds) to 1529 kcal/day (6 year-olds) for boys, and 1140 (3 years) to 1332 kcal/day (6 years) for the girls. When compared with reference intake of foods for 3–6 year-old Japanese children [23], the observed energy intake was 91.8–109.7 % of recommended daily allowance [22]. Thus, food duplicate samples were considered as adequately collected.

Dietary Sn intake (Sn-D)

The 296 cases were classified into two groups by intake of canned foods, i.e., 132 cases with reported consumption of canned foods (including drinks or juice) and 164 cases with no report of such consumption. Among the 164 cases with no report, Sn levels in several cases were found to be markedly higher than others. Application of Smirnov test after logarithmic conversion gave p values smaller than 0.05 for 9 cases (with Sn-D of 77.5–1113.6 $\mu\text{g}/\text{day}$). Thus, the 9 cases were excluded, and 155 cases were identified as non-consumers.

Statistical parameters for Sn-D distribution were calculated for total 296 cases, 132 canned food consumers and 155 non-consumers, as summarized in Table 1. For visual

Table 1 Distribution of Sn by canned food intake

	Total cases	Those who consumed	
		Canned foods	No canned foods ^a
No. of cases	296	132	155
Sn intake ($\mu\text{g}/\text{day}$)			
Arithmetic mean	149.9	292.0	5.4
Geometric mean	10.3	30.2	3.3
Minimum	0.4	0.9	0.4
Median	4.2	10.6	2.5
Maximum	3255	3255	48

^a Nine cases were excluded after Smirnov test for extreme values

understanding, the distributions are presented as histograms after logarithmic conversion in Fig. 1. It was clear that the distribution of total cases (Fig. 1a) did not follow a normal distribution even when logarithmic conversion was applied; there was a larger compilation around $\log \text{Sn-D}$ ($\mu\text{g}/\text{day}$) = 0.5 [=log 3.16 ($\mu\text{g}/\text{day}$)] and a smaller accumulation with less remarkable peak formation at 3.0 [=log 1000 ($\mu\text{g}/\text{day}$)]. Analysis with 132 canned food consumer cases (Fig. 1b) showed less clear accumulations, but the peak at 0.5 (which was observed also in Fig. 1a) became

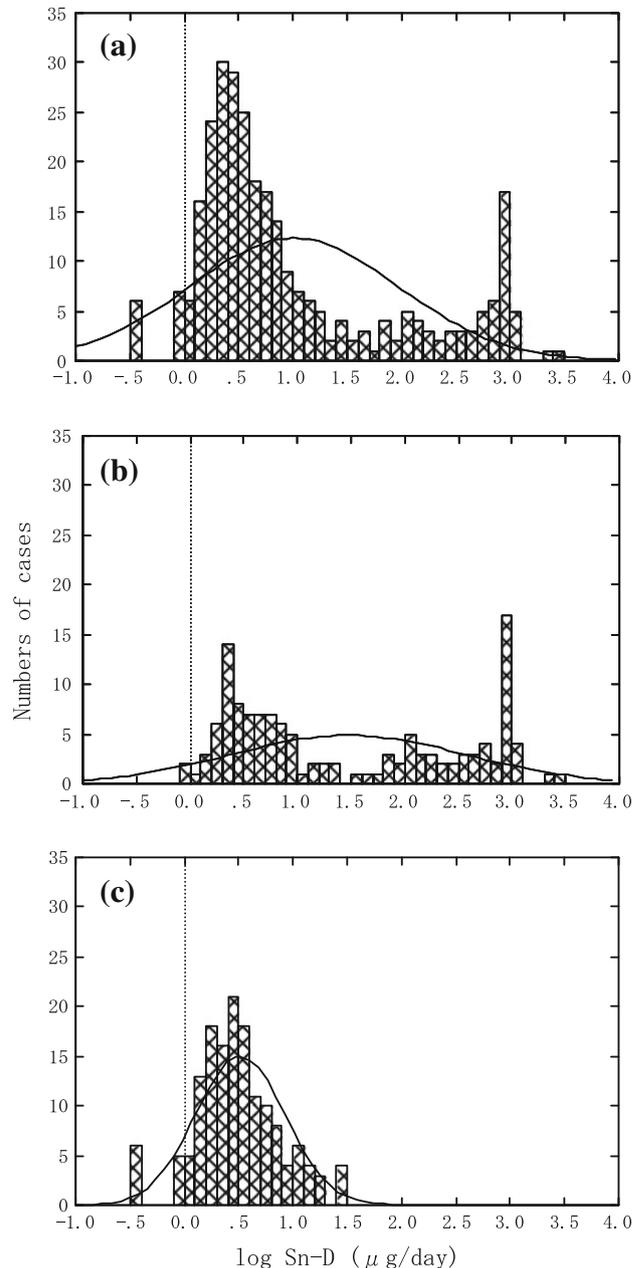


Fig. 1 Distribution histograms for daily Sn intake. **a** Total cases. **b** Cases of canned food consumers. **c** Cases of non-consumers of canned foods. The curve in each figure indicates a normal distribution

smaller, whereas the incomplete peak at 3.0 stayed essentially unchanged. In cases of 155 non-consumers (Fig. 1c), there was a single peak at around 0.5 and the distribution was essentially log-normal. The median and GM Sn-D were 2.5 and 3.3 µg/day, respectively (log 2.5 = 0.40 and log 3.3 = 0.52). Statistical comparison between the canned food consumer and non-consumer groups by Mann–Whitney test showed that Sn-D for the consumer group (10.6 µg/day as a median) was significantly ($p < 0.01$) higher than that for the non-consumer group (2.5 µg/day).

Possible role of boiled rice as a Sn source

The amount of boiled rice was 206 g/day on an average which accounted for 15.6 % (maximum; 33.1 %) of total foods (1312 g/day) by weight. Thus, it was considered possible that boiled rice might play a role as a dietary source of Sn. Sn level in boiled rice was, however, rather low (0.27 µg/kg as a median) and accounted for only a very minor portion in daily Sn-D (i.e., 0.047 µg/day as a median).

A multiple regression analysis was conducted taking daily Sn intake as a dependent variable, and sex (1 for boys and 0 for girls), age, daily consumption of total foods (by weight), boiled rice (by weight) and energy, together with canned food intake (1 for YES and 0 for NO) as independent variables ($R^2 = 0.263$, $p < 0.01$) (Table 2). It turned out that standardized regression coefficients were negative (i.e., <0), although significant, both for total foods ($p < 0.01$) and for boiled rice

Table 2 Multiple regression analysis taking dietary Sn intake (µg/day) as a dependent variable

Independent variable		SRC	<i>p</i>
Item	Unit		
Sex ^a		−0.074	0.164
Age	Years	0.042	0.413
Total foods	g/day	−0.579	<0.01
Boiled rice	g/day	−0.130	0.023
Energy intake	kcal/day	0.278	<0.01
Canned food intake ^b		0.330	<0.01

SRC standardized regression coefficient

^a 1 for boys and 0 for girls

^b 1 for Yes and 0 for No

Table 3 Possible contribution of juice intake in dietary Sn intake

^a Sn-D is dietary intake of Sn
^b $p > 0.10$ for both coefficients

Parameter	Unit	AM	Median	Minimum	Maximum	Correlation coefficient ^b	
						Pearson's	Spearman's
Sn-D ^a	µg/day	144.7	5.26	1.9	1200		
Juice intake	g/day	144.6	130.9	5.0	308.2	−0.056	0.173

($p < 0.05$). Taking together the previous observation that the account for Sn from boiled rice (0.047 µg/day) in total foods (4.2 µg/day) was very small (i.e., 1 %), the contribution of boiled rice as a dietary Sn source was considered to be very limited. The coefficient was positive and significant ($p < 0.01$) for energy intake. Canned food intake had a positive and significant ($p < 0.01$) coefficient of 0.330, as expected. Other independent variables of sex and age were insignificant.

Lack of association of canned juice intake with Sn-D

Reported food intakes showed that a number of children consumed canned juice (mostly tomato juice) on several occasions on the survey day. Accordingly, cases were selected for those who consumed tomato juice. In practice, the consumption was confirmed in 27 cases, and the amounts were in a range from 5.0 to 308.2 g/day (Table 3).

Analysis for Pearson's correlation gave a negative and insignificant coefficient. Analysis for non-parametric Spearman's correlation also resulted in an insignificant coefficient, suggesting that no correlation existed between Sn-D and canned juice intake.

Sn in urine of canned food-consumers and non-consumers

Complete sets of Sn, CR and SG for the first morning urine sample were available for 217 children. The Sn-U levels of the 217 cases as a whole were distributed almost log-normally with GM of 2.69 µg/l for Sn-U_{ob}, 3.83 µg/g cr for Sn-U_{cr}, and 1.92 µg/l for Sn-U_{sg} (Table 4). For visual understanding, the distribution of the observed (uncorrected) Sn-U values is depicted in Fig. 2. When Sn-U values were classified by the consumption of canned foods (i.e., 97 consumers and 120 non-consumers), no significant difference ($p > 0.10$) was detected either by unpaired *t*-test (after logarithmic conversion) or non-parametric Mann–Whitney test (Table 4). It should be noted that a significant difference in Sn-D was re-confirmed for the 217 cases, as for total 296 cases.

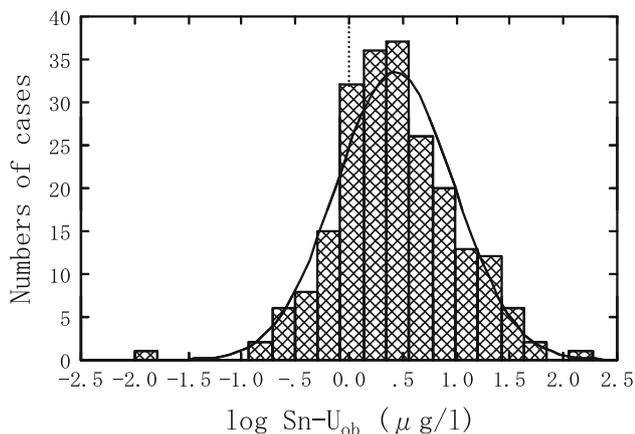
When possible correlation was examined with total 296 cases between Sn-D and Sn-U (as observed, as corrected for CR, and as corrected for SG) before and after logarithmic conversion (Table 5), Pearson's and Spearman's correlation coefficients were all insignificant ($p > 0.10$).

Table 4 Sn in urine (Sn-U) in selected children by canned food consumption

	Sn-D ($\mu\text{g}/\text{day}$)			Sn-U _{ob} ($\mu\text{g}/\text{l}$)			Sn-U _{cr} ($\mu\text{g}/\text{g cr}$)			Sn-U _{sg} ($\mu\text{g}/\text{l}$)		
	Total case	Consumers	Non-consumers	Total case	Consumers	Non-consumers	Total case	Consumers	Non-consumers	Total case	Consumers	Non-consumers
No. of cases	217	97	120	217	97	120	217	97	120	217	97	120
GM	9.86	34.11	3.61	2.69	2.95	2.49	3.83	4.18	3.56	1.92	2.15	1.76
GSD	10.35	13.61	4.32	3.55	3.37	3.71	3.59	3.38	3.77	3.63	3.41	3.80
Minimum	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD						
Median	3.5	18.9	2.6	2.4	2.5	2.3	3.7	4.0	3.5	1.7	2.0	1.6
Maximum	3254.8	3254.8	1113.6	185.9	49.4	185.9	250.6	79.2	250.6	198.3	49.4	198.3

No significant difference ($p > 0.10$) was detected between consumers and non-consumers in urine by unpaired *t*-test (after logarithmic conversion), as well as Mann-Whitney test irrespective of correction of urine for creatinine (U_{cr}), urine specific gravity (1.016) (U_{sg}) or none (i.e., as observed; U_{ob}). Difference in Sn-D was significant ($p < 0.01$) by Mann-Whitney test

217 children for whom Sn-U, CR, SG were available in urine

**Fig. 2** Distribution of Sn levels in urine (observed uncorrected values). The curve in the figure shows a normal distribution**Table 5** Possible correlation of Sn in urine with dietary Sn intake

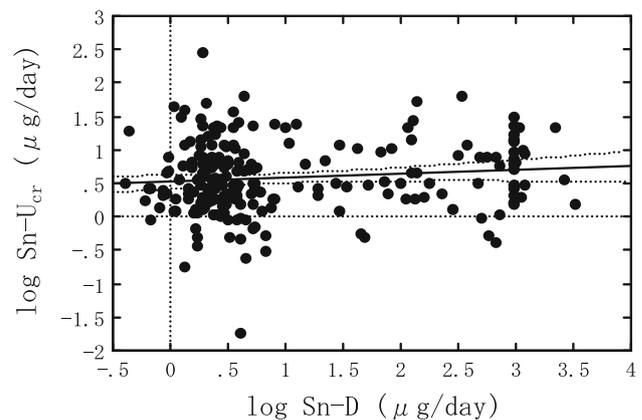
X ^c	Y ^c	Intercept (α) ^c	Slope (β) ^c	Correlation coefficient ^d	
				Pearson's	Spearman's
Sn-D ^a	Sn-U _{ob} ^b	5.948	0.001	0.032	0.159
	Sn-U _{cr} ^b	6.672	<0.001	<0.001	0.231
	Sn-U _{sg} ^b	4.371	<0.001	<0.001	0.241
Log Sn-D	Log Sn-U _{ob}	0.342	0.079	0.148	
	Log Sn-U _{cr}	0.520	0.059	0.110	
	Log Sn-U _{sg}	0.217	0.061	0.114	

^a Sn-D is dietary intake of Sn in $\mu\text{g}/\text{day}$

^b Sn-U_{ob} (in $\mu\text{g}/\text{l}$), Sn-U_{cr} (in $\mu\text{g}/\text{g creatinine}$) and Sn-U_{sg} (in $\mu\text{g}/\text{l}$) are Sn in urine as observed, after correction for creatinine concentration [20], and after correction for a specific gravity of urine (taking 1.016 as a standard) [21], respectively

^c α and β are the intercept and the slope in a regression equation $Y = \alpha + \beta X$, where X is Sn-D or log Sn-D, and Y is Sn-U_{ob}, Sn-U_{cr}, Sn-U_{sg}, log Sn-U_{ob}, log Sn-U_{cr} or log Sn-U_{sg}, respectively

^d All correlation coefficients are statistically insignificant ($p > 0.10$)

**Fig. 3** Relationship of Sn in urine with dietary Sn intake after logarithmic conversion. Each dot indicates one case and the solid line in the middle is a calculated regression line. Two broken curves on both sides of the line indicate the 95 % range for the regression line. Note that the range of distribution is wide and that the regression line is almost in parallel to a horizontal line

The relation of log Sn-D and log Sn-U_{cr} is presented in Fig. 3 as an example.

Discussion

The present analyses on Sn-D of pre-school children in Miyagi prefecture, Japan, gave very similar results with the previous observation on adult populations in four regions in Japan. Median Sn-D for the populations studied were 4.2 $\mu\text{g}/\text{day}$ for children (Table 1) and 5.6 $\mu\text{g}/\text{day}$ for adults [12]. Sn-D was higher for canned food consumers than for non-consumers, both in children (Table 1) and in adults [12]. The finding of higher Sn-D for canned food consumers than for non-consumers of canned foods is in a close agreement with the observation that Sn in canned foods was much higher than that in fresh foods of the same kind, and the consumption of canned foods was associated with high dietary Sn intake [9]. Among non-consumers of

canned foods, Sn-D was distributed log-normally (Fig. 1c); medians were 2.5 and 4.5 $\mu\text{g}/\text{day}$ for children (Table 1) and adults [12], respectively. These distributions may indicate background Sn intake via foods, although the minimum–maximum range was rather wide (Table 1) suggesting substantial variation in Sn contents in natural foods.

While the medians for Sn-D on a daily consumption basis were larger for adults than for children, the reverse was the case when evaluated on a body weight basis. Boys in the present study were 16.6 (at 3 years) to 23.2 kg (at 6 years) heavy and girls were 14.2 (at 3 years) to 20.7 kg (at 6 years), or 18.7 kg on average for boys and girls in combination. Typical Japanese adult (i.e., >20 years of age) men and women weigh 65.3 and 52.7 kg [24] or about 59 kg. Thus, adults may be 3 times heavier than 3–6-year-old children. As a result, the Sn-D was 0.225 μg Sn/kg body weight/day for children whereas it was 0.095 μg Sn/kg body weight/day in case of adult people. Thus, it was clear that children took substantially more Sn-D ($0.225/0.095 = 2.4$ times) than adults (e.g., their parents) on a body weight basis.

The present survey made it clear that children will take 4.2 μg Sn/day as a median although the maximum value observed was as high as 3255 $\mu\text{g}/\text{day}$. In 1989, the Joint FAO/WHO Expert Committee [7] converted previously established provisional maximum tolerable daily intake of 2 mg/kg body weight/day to provisional tolerable weekly intake of 14 mg/kg body weight/week, and kept the same value after re-evaluation in 2001 [8]. The intake of 4.2 μg Sn/day (or 3255 μg Sn/day as the maximum) (Table 1) by a 18.7 kg heavy child will be equivalent to 0.0016 (or 1.218) mg Sn/week. The current intake level should be taken as far less than the provisional tolerable weekly intake.

It should be considered in addition that the Committee noted in a later meeting that the basis for the provisional tolerable weekly intake was unclear as the value might be based on acute effects and studies on possible health effects after chronic exposures should be desirable [2]. It would be the Sn concentration and not the amount that induces the acute effects. Furthermore, the effects may be different depending on valencies of Sn and co-presence or absence of Sn-adsorbing solid materials [25, 26].

International comparison was intended to evaluate the Sn intake by Japanese children. In winter in 1984–1985, Smart et al. [27] studied 24–27-month-old children UK and observed that Sn intake was 1.16 mg/day (1.16) as GM (geometric standard deviation; GSD). The method employed for analysis was flame atomic absorption spectrometry [28], the high values observed in the study of Smart et al. [27] might be attributable to the difference in the method of analyses i.e., flame atomic absorption

spectrometry (Smart et al. [27]) versus inductively coupled plasma-mass spectrometry (the present study). It should also be noted that no information on canned food consumption was given unfortunately. No other report was available on Sn intake by children elsewhere.

The previous survey [12] on adult populations revealed that the Sn intake by the Japanese population (0.04 mg/day) was substantially lower than the levels reported for other populations, e.g., people in the UK (2.4 mg/day by Ysart et al. [13]) and France (2.3 mg/day by Noël et al. [11]), whereas nearly equal to that for Spanish population (0.035 mg/day by Bocio et al. [10]). It is quite conceivable that the situation is similar for children to that for adult people.

With regard to Sn-U, GM among the children studied (canned food consumers and non-consumers together) was 3.8 $\mu\text{g}/\text{g}$ cr. Hayashi et al. [29] reported similar GM values of 2.1 and 5.3 $\mu\text{g}/\text{g}$ cr for adult Japanese men and women, respectively (the GM values were estimated from AM and arithmetic standard deviation [ASD] by use of the moment method [30] for comparison). Hayashi et al. [29] also found no difference in Sn-U between canned food consumers and non-consumers. Pascal et al. [31] observed 3.1 $\mu\text{g}/\text{g}$ cr for general US population. Later, Shimbo et al. [12] obtained 2.0 $\mu\text{g}/\text{g}$ cr for adult Japanese people. Over-all, it appears likely that there will be no substantial difference in Sn-U among the populations studied.

The absence of correlation between dietary Sn-D and Sn-U is as previously observed by this study group [12]. The absence is expected as the absorption of Sn via the GI tract is low [3] and the half-life is long [4], so that Sn-U will not reflect Sn-D on the previous day. In addition, urinary excretion is a minor route for Sn elimination [32].

The lack of association between the quantity of canned juice ingestion and Sn-D on the day is rather contrary to the expectation. This point may deserve further study. It might be the case that cans for liquid foods such as tomato juice were well lacquered to minimize Sn migration from cans, or the pH of the juice was not low [25].

There are several limitations in the present study. First, concern remains with regard to the accuracy of the information on intake of canned foods. The exclusion of nine high-Sn cases after the Smirnov test may indicate that the information given by guardians may not be complete, but some cases of the canned food consumption may remain unreported. In this respect, it should be noted that the visual inspection of collected samples [33] may not be sensitive enough to identify canned foods which are the leading source of Sn in the daily foods. Sn concentration ingested was out of the study target due to technical reasons in the present study. It was also not possible to carry out Sn valency analysis despite the discussion described above.

In conclusion, the present study made it clear that, whereas dietary Sn intake by preschool children distributed in a wide range up to >3 mg/day, the median was as low as 4.2 µg/day. The increase in Sn intake was primarily associated with canned food consumption. The role of boiled rice as a dietary Sn source is apparently limited.

The level of Sn intake even by canned food consuming Japanese children was far below the provisional tolerable weekly intake maintained by the 2001 Joint FAO/WHO Expert Committee. Thus, the risk of excess dietary Sn intake should be very remote for children in Japan. The Sn levels in urine were similar between children who consumed canned foods on the previous day and those who did not.

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Conflict of interest The authors declare that they have no conflicts of interest.

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