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Original Article

Exposure Assessment of Lead among Japanese Children

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Abstract

Objective: Lead intake from possible exposure routes among children residing in the Tokyo Metropolitan Area was estimated.

Methods: Lead concentrations in house dust samples collected from the houses of the children and those in 24-h duplicate diet samples of the children were determined. The daily lead intake was estimated by multiplying the lead concentrations in the house dust, diet, soil and ambient air (the latter two were from the literatures) by the corresponding intake and/or inhalation rates, and summing all of the products. Bioaccessibility tests were performed on the house dust, soil and diet samples to determine the lead uptake level.

Results: Children residing in the Tokyo Metropolitan Area were estimated to be exposed to 21.5 µg of lead on a daily basis, with a maximum intake of up to 70.4 µg. The average weekly intake per kg body weight for a 5-year-old Japanese child was found to be 8.0 µg/kg bw/wk, which is below the Provisional Tolerable Weekly Intake (PTWI) of 25 µg/kg bw/wk. However, the maximum weekly intake was found to be 26 µg/kg bw/wk.

Conclusions: House dust and soil ingestion can be the predominant routes of exposure to lead among children in Japan, and the source(s) of lead in such media must be specified to reduce the lead intake level of the children.

Key words: lead intake, soil, house dust, diet, estimated blood lead

Introduction

Human exposure to low levels of environmental lead is inevitable, since lead is ubiquitous and one of the most widely dispersed contaminants. Among the population groups, children are the subpopulation of concern for lead exposure. They are exposed to more lead than adults due to their behaviors such as playing on the floor or outdoors, sucking on objects, and hand-to-mouth activity, which is normal developmental activity. The high gastrointestinal absorption of lead of children also causes higher uptake of lead to the system, which may lead to irreversible damage to a susceptible nervous system during its developing stage. Although classical lead poisoning is a rare occurrence worldwide these days, developmental effects in children caused by low level exposure to lead are well acknowledged (1–6).

Children in Japan are considered to have a lower risk of lead, as a result of the complete phasing out of leaded gasoline as early as the mid-1980s (7). Moreover, the lead level in food in Japan has been found to be in the lowest range in the world (8). Consequently, the blood lead level in Japanese children, measured in 1993 (9), was relatively low compared to those of other countries (10–14).

Contaminant(s) levels in the children's environment are measured to adequately assess multimedia exposure to the contaminant(s), with the intention to ensure a safe and healthy environment for children (15). To our knowledge, however, systematic studies for assessing lead levels in the children's environment in Japan have been limited.

Our aim was to determine the lead levels in the Japanese children's environment, and to estimate the lead intake from every possible route of exposure. This paper describes the estimation of the daily lead intake via the ingestion of soil, house dust and diet and via the inhalation of ambient air. The bioaccessible fraction of lead, which is dissolved in synthetic gastric juice, was determined for house dust, soil and diet, to compare the risk associated with ingestion of soil and house dust with that associated with dietary intake.

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Materials and Methods

We used house dust, soil, diet, and ambient air as the sources of exposure to lead for the children in this study. House dust samples were collected from households with a child or children, and lead contents were determined. Lead intake via soil ingestion was estimated from the lead concentration data of our previous study (16). Dietary lead intake was also based on our multi-elemental analysis of 24-h duplicate diets. Inhalation lead exposure was calculated from data for lead in ambient air available in the literature, and included in the total lead intake estimation.

House dust sampling and analysis

Vacuum cleaner dust filter bags from the households with a child/children were collected in 2003. A total of 21 vacuum cleaner dust bags were collected from households in the Tokyo Metropolitan Area. The period of time the subject households collected the house dust, i.e., the time elapsed from the previous replacement to a new filter bag, could not be specified for all of the subject households.

Direct sieving of the vacuum cleaner filter pack contents with 149-μm mesh was performed. A mechanical shaker and alumina balls were employed for efficient sieving. The house dust samples were digested with nitric acid, perchloric acid and hydrofluoric acid (16). Lead concentrations in digested house dust samples were determined by inductively coupled plasma mass spectrometry (ICPMS) using an HP 4500 instrument (Yokogawa Analytical Systems Inc., Japan).

Throughout the digestion and analysis of the house dust samples, a rigorous data quality control procedure was followed. For contamination correction and accuracy checks, procedure blanks and certified reference material (CRM) from the National Institute of Standards and Technology (SRM 2709 San Joaquin Soil) were incorporated. The lead concentration determined in our study [19.1±0.7 μg/g (n=3)] was in good agreement with the certified value (18.9±0.5 μg/g).

Playground Soil

A total of 46 sand and surface soil samples were collected from 23 playgrounds and sieved with 149-μm mesh, and the lead contents were analyzed. The details of the sampling, digestion and analysis are described elsewhere (16).

Duplicate diet

The 24-h duplicate diet samples, including the snacks, beverages and drinking water of the children, were collected over 7 consecutive days from 33 volunteer households in the Tokyo Metropolitan Area in November and December of 2000. The Ministry of Environment, Government of Japan, undertook this sampling as a part of a survey on the soil ingestion rate of the Japanese (17). The subjects included 33 children (19 boys and 14 girls) and their mean age was 5.1±0.9 years old. The survey staff visited the subjects' households daily to collect the duplicated diet. They weighed the sample and compared it with a food list filled in by the caregiver (most often the mother) of the subject child. When there was a missing sample, the survey staff asked for the approximate size of the missing portion and estimated the missed weight. The missed weight was then recorded and added to the total weight of the 7-day composite. The weight of the 7-day composite diet for each child, including correction for missing samples, varied from 5520 to 11230 g/week. The duplicated diet samples for the 7 days thus collected were mixed and homogenized to make a composite for each child from the household.

A portion (5 g) of the composite was freeze-dried, and an aliquot (100 mg) of the dried sample was digested with nitric acid by the Teflon double digestion bomb method (18). Multi-element analysis of the diet samples was performed using ICPMS (Agilent 7500, Yokogawa Analytical Systems Co., Ltd., Tokyo). In this report, only the lead data is presented: the detailed methods and results other than those for lead will be presented elsewhere. The accuracy of our diet analysis was validated through the analysis of Typical Japanese Diet CRM from the National Institute for Environmental Studies, Japan. Our result [0.59±0.01 mg/kg (n=8)] was in good agreement with the reference value (0.62 mg/kg) of the CRM for lead.

Bioaccessibility of house dusts, playground soils and diets

The bioaccessibility of house dust, soil and diet was determined to approximate the lead uptake by the body of children. An *in vitro* soil oral bioaccessibility test developed by the Solubility/Bioavailability Research Consortium (SBRC) (19) of the United States was employed for the determination of lead bioaccessibility of house dust, playground soil and diet. This method, which simulates only the stomach condition of humans, was validated through swine *in vivo* tests (20), indicating that the leached concentration of lead under the test conditions is predictive of oral bioavailability in an animal model. In this method, the lead concentration extracted into the simulated gastric juice (acidified glycine) out of the total content is measured, and that fraction is termed the bioaccessible fraction. The bioaccessibility in this report was calculated by the following equation (21):

Bioaccessibility (%)=[lead mobilized from house dust/soil/diet during extraction (μg)]/[Total lead present in house dust/soil/diet (μg)]×100

One gram of dry house dust, soil or diet was added to 100 mL of 0.4 M glycine solution, which had been adjusted to pH 1.5 with HCl. The mixture was end-over-end rotated at 37°C at 30 rpm for 1 hr, and then filtered through a 0.45-μm cellulose acetate disk filter (Sartrius, Germany). After confirming that the pH of the filtrate was between 1.0 and 2.0, the lead concentration was determined by ICPMS with bismuth as an internal standard. The quality control and quality assurance of the bioaccessibility tests of soil and house dust were described in our previous study (22).

Results

Lead concentrations in house dust and diet samples

The lead concentrations in the 21 house dust samples varied between 18 mg/kg and 359 mg/kg on a dry weight basis; the mean lead concentration was 117 mg/kg. The lead concentrations in the diet samples varied from 0.0023 to 0.0059 mg/kg wet weight, with the mean being 0.0044 mg/kg on a wet weight basis.

Table 1 Lead concentrations in environmental exposure media and daily lead intake estimates

Media	N	Pb concentration[a]	Daily Pb intake (μg/day)[b]	
			Mean-based	Maximum-based
Soil	46	46.4 (11.7–248) mg/kg	4.64	24.8
House dust	21	117 (17.7–359) mg/kg	11.7	35.9
Diet	33	0.0044 (0.0023–0.0059) mg/kg	4.79	8.86
Air	—	0.045 (0.016–0.094) μg/m^3	0.39	0.82
Total			21.5	70.4

[a] Lead concentration is expressed on a dry weight basis for soil and house dust and on a wet weight basis for diet.
[b] The ingestion rate was assumed to be 100 mg/day for soil and house dust based on the default soil ingestion rate [200 mg, (17)] and a soil:dust=1:1 assumption (24). The ingestion rate for diet was based on the measured weight for each child. The inhalation rate of 5-years-old child was assumed to be 8.75 m^3/day [see text and (25)].

Estimated daily lead intake

Table 1 presents the mean concentration of lead (with the min-max range) in each exposure medium, and the daily intake of lead from each exposure route. The lead concentration in the soil, 46.4 mg/kg (11.7–248 mg/kg), was recapitulated from our previous study (16), and that in ambient air, 0.045 μg/m^3 (0.016–0.094 μg/m^3), was cited from the Tokyo Metropolitan Government monitoring data of 2000 (23).

The ingestion rate for soil and house dust was set at 100 mg/day each: this was based on the default soil ingestion rate (200 mg/day) set by the Ministry of Environment of Japan in 2000 (17), and the assumption of 1:1 ingestion of soil and house dust reported by Stanek and Calabrese for American children (24). The inhalation rate for children was set at 8.75 m^3/day for the calculation of inhalation exposure to ambient lead. This inhalation rate was calculated by assuming 12 h of rest (1.944 m^3/8 h: mean of boys and girls) and 12 h of light activities (3.888 m^3/8 hrs: mean of boys and girls) (25). The weekly dietary lead intake was calculated for each child by multiplying the lead concentration in the 7-day diet composite of the child with the corresponding weight of food consumed over the period. The daily dietary lead intake was calculated from the estimated weekly intake. The mean daily dietary intake of the 33 children and the maximum intake are presented in Table 1.

The daily lead intake from all of the sources amounted to 21.5 μg/day when the calculation was based on the means, and it was 70.4 μg/day when based on the maximum concentrations observed. House dust and soil contributed to the total daily intake of lead by 54 and 22%, respectively, for the mean-based estimate. The figures for the maximum-based estimate amounted to 51 and 35%, respectively. Dietary intake contributed 22 and 13% to the mean- and maximum-based estimates, respectively. Inhalation of ambient air negligibly contributed to daily lead intake. Thus, the non-dietary oral route constituted 77–86% of the daily intake.

When the soil:house dust ratio varied from 190 mg:10 mg to 10 mg:190 mg instead of 100 mg:100 mg as shown in Table 1, the lead intake from soil and house dust varied from 9.99 to 22.7 μg/day accordingly for the mean-based estimate. Thus, the mean-based estimate can have a ±6 μg/day uncertainty depending on the assumption of soil:dust ingestion (while the total amount is fixed at 200 mg/day). Similarly, the maximum-based estimate has a ±10 μg/day uncertainty.

In Table 2, the bioaccessibilities of lead in soil, house dust and diet, as determined in the present study, are shown. The mean bioaccessibility ranged from 43 to 57%.

Table 2 Bioaccessibility of soil, house dust and diet

	n	Mean (%)	Range (%)
House dust	20	57	44–67
Soil	44	43	8–73
Diet	6	52	43–62

[n] number of samples.

Discussion

One of the major findings of our estimation of daily lead intake of Japanese children was the level of lead intake itself, since the exposure assessment of lead among children from all of the possible exposure sources has not been performed in Japan. The mean-based estimate was 21.5 μg/day (Table 1), and this can be compared with the recently reported values from other countries with a similar estimation approach, such as 40.82 μg/day in the Philippines (26) and 29.5 μg/day in Canada (27). The mean-based estimate can be converted to 8.0 μg/kg/week by considering the average body weight of a 5-year-old Japanese child (18.7 kg) (28). This is less than the Provisional Tolerable Weekly Intake (PTWI) set by the Joint FAO/WHO Expert Committee for Food Additives (JECFA), 25 μg/kg/week (29), indicating that the children in Tokyo Metropolitan Area are not excessively exposed to lead if they have only contact with environmental media with average lead content. However, our maximum-based estimate (70.4 μg/day), the worst-case scenario, corresponds to 26 μg/kg/week, thus exceeding the PTWI, though the probability of reaching this maximum-based estimate is not high.

Estimating the blood lead level of Japanese children with the daily lead intake estimated in this study may be interesting, because the dose-effect relationship of lead is established on the blood lead level. The JECFA estimated that lead intake at the PTWI, 25 μg/kg/week, corresponds to 5.7 μg/dL in blood (30). Based on this estimate, 21.5 μg/day intake from all sources (8.0 μg/kg/week) corresponds to 1.8 μg/dL, and the maximum-based estimate (26 μg/kg/day) to 5.9 μg/dL. Even the maximum-based estimate is well below the current action level (10 μg/dL) the US Center for Disease Control and Prevention (CDC) has set, however, it must be pointed out that a recent study indicated cognitive defects in reading, mathematics, short-term memory, and visual construction skills at blood lead of 2.5 μg/dL (31).

Another major finding was the great contribution of soil and house dust, the non-dietary oral source, to the daily lead intake of Japanese children. The significance of soil and house dust as a lead intake source has been noted in previous studies conducted in other countries. The present results indicated that the same held true for Japanese children, in spite of the great differences in life styles compared to those of other countries, e.g., taking shoes off at the house entrance, and the lack of interior lead paint in Japan, which may be related to lead abundance in some exposure media. Moreover, the bioaccessibility results (Table 2) indicated that these non-dietary sources have bioaccessibility similar to that of diet, and thus the significance of these sources not only on an intake basis, but also on an uptake basis, has been demonstrated.

In the present results, the high lead concentration in the house dust, and consequently the great contribution to daily lead intake, was particularly notable. The high lead concentration in house dust, in general, poses a higher lead exposure risk for young children who spend more of their time indoors. Although, to date, the predominant source of lead in house dust has not been clarified in Japan, fine particles of tracked-in soil, and airborne particles may be responsible (32). Further research is needed to understand the source and pathway of lead in the indoor environment in Japan, taking the possible relevance of lead in house dust into consideration.

The present study indicated that the most effective means for reducing children's lead exposure level is reducing the lead level in the soil in the children's environment, since soil can contribute to daily lead intake via direct ingestion and indirectly via house dust. Lindern et al. demonstrated that an overall decrease of 0.5–0.6 mg lead/kg of house dust per 1.0 mg lead/kg soil reduction was observed during the 10-year soil remediation in the yards of homes occupied by children in the Bunker Hill Superfund Site, in northern Idaho (33).

Despite its versatility, lead poses a human risk especially to children, when inhaled or ingested. In many countries, actions have been taken to reduce the risk from lead exposure, depending on the significance of the sources of lead. The present research indicated that lead in house dust and playground soil deserves attention when considering lead exposure among children in Japan.

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