Influence of paint chips on lead concentration in the soil of public playground in Tokyo

Environmental Process Engineering 36646 Michie TAKAOKA

1. INTRODUCTION

Recently, it has been suggested that declined IQ score was found among children with blood Pb level lower than 10 μg/dL, the action level of Center for Disease Control and Prevention (CDC) of the US (*1*,*2*). These results suggest the necessity of lowering Pb exposure level as much as possible. It is well known that soil is an important medium of Pb exposure for children (*3*). As sources of soil Pb, two products, i.e., lead-based paint (LBP) and leaded gasoline, have been widely recognized (*3*). Fortunately, in Japan, phase-out of leaded gasoline was started in early 1975, and market share of non-leaded gasoline reached 100% in 1980 (*4*). Moreover, the LBP has not been used for interior/exterior decoration of houses in Japan. Therefore, Pb exposure level of contemporary Japanese people has long been considered insignificant in the past two decades.

Aung et al.'s estimation revealed that Pb exposure level of Japanese children could reach Provisional Tolerable Weekly Intake set by FAO/WHO, and soil and dust appreciably contributed to the intake (*5*). They found gasoline Pb, that was emitted before 1980s and had been thereafter settled in the surface soil, as one of the sources of Pb in the soil (*6*). Besides gasoline Pb, paint chips can be another significant source of Pb in surface soil of public playground. It is often the case that paint chips peeled off from playing equipments are scattered on the surface soil in the public playground in Japan. Lead concentration in these paints was reported to be high (*7*).

In this study, the effect of paint chips peeled off from the playing equipments to Pb content in the surface soil was evaluated for a number of public playgrounds in Tokyo.

2. MATERIALS AND METHODS

Soil sampling and preparation

Playgrounds where the present samples were taken were located in a ward in Tokyo. Two types of soil sampling were carried out. ***Sampling method 1.*** The area of a playground (12 m × 28 m) was divided into 68 (2 m × 2 m) grids. Surface soil (top 0.5 cm) at the center of each grid was collected. ***Sampling method 2.*** Each of 31 playgrounds was divided into 10 m × 10 m grids. Five sampling points were selected in each grid and soil sample was taken. Soil samples were collected from 91 grids (455 individual points) in 31 playgrounds.

The soil sample was air-dried and sieved through a 150 μm mesh. Five samples collected by sampling method 2 for each grid were mixed in equal weight to represent the grid.

Paint sampling

The paint chips, which were peeled-off from the playing equipments and had scattered on the surface soil, were collected. Sixteen samples were collected from 5 playgrounds; swings (4), slides (2), a sandpit (1), a rail (1), a maze (1), flowerbeds (3), and benches (4).

Analysis of Pb concentration

Approximately 300 mg of soil sample was digested with $HNO_3/HClO_4/HF$ in a Teflon beaker on a hotplate. Around 10 mg of paint chips was digested with HNO_3 using a double vessel digestion bomb for 4 h (*8*). Lead concentrations and isotope ratios of Pb ($^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$) in the samples were determined by ICP-MS.

For analytical quality assurance, NIST SRM 2709 and 2711 were analyzed NIST SRM 981 was used for the calibration of isotope ratios.

Calculation of the degree of peeling-off of paint from surface of playing equipments

Cross-section paper (2 cm × 2 cm per grid) was applied to an appropriate surface of playing equipments and the peeling-off index (POI) was calculated as follows:

$$POI = \{(\text{number of grids all of paint is peeled-off}) \times 1 + (\text{number of grids a part of paint is peeled-off}) \times 0.5\} / \text{total number of grids}$$

When more than 2 playing equipments were present in a playground, POIs for all of the equipments were averaged to represent the playground.

3. RESULTS

Lead concentration in paint chips scattered on surface soil

A maximum of 8.5% of Pb was contained in the paint chips (Table 1). Lead concentration in the paint chips collected from surface soil underneath wooden benches in the playground No.2 was low as compared with those underneath playing equipments made of metals. This result is consistent with my notion that anticorrosive paint is the major contributor to elevated Pb concentration in paint chips scattered on the surface soil.

Table 1. Lead concentration in paint chips

Park No.	source	Pb(%)	Park No.	source	Pb(%)
No.1	bench	1.66±0.03	No.3	swing	5.69±0.06
No.1	bench	2.58±0.05	No.4	swing	3.12±0.01
No.1	Slide	4.58±0.09	No.4	sandpit	0.45±0.01
No.1	swing	4.61±0.08	No.4	flowerbed	8.49±0.10
No.2	bench	0.003±0.000	No.4	flowerbed	6.85±0.08
No.2	bench	0.003±0.001	No.4	flowerbed	5.51±0.08
No.2	Slide	0.77±0.01	No.5	swing	0.26±0.01
No.2	maze	3.00±0.03	No.5	rail	2.90±0.03

Average ± SD, n=3

The Pb concentration distribution in playground

Figure 1 shows profile of surface soil Pb distribution within playground A. Lead concentration in the 2 m × 2 m grid ranged from 15.9 mg/kg to 104 mg/kg (mean: 52.3 ± 19.4 mg/kg). As clearly shown in this figure, Pb concentration was higher in the soil of the grid with painted playing equipment (Fig. 1).

The mean Pb concentration each of the 31 playgrounds was 55.5 mg/kg (15.2-237 mg/kg) indicating that soil in some of the playgrounds is contaminated with Pb because natural soil Pb level in Japan was reported to be 20 mg/kg (*9*).

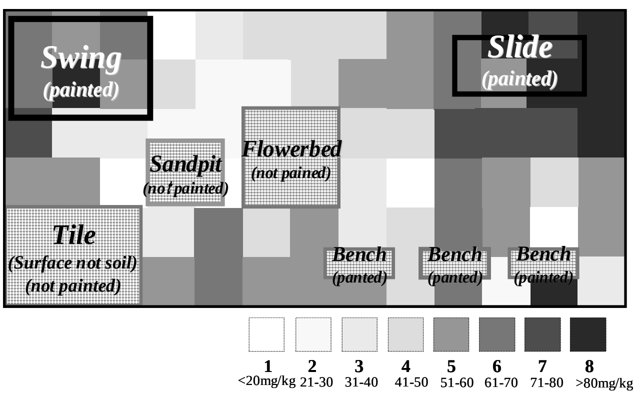

Fig. 1 Lead distribution profile of surface soil within playground A

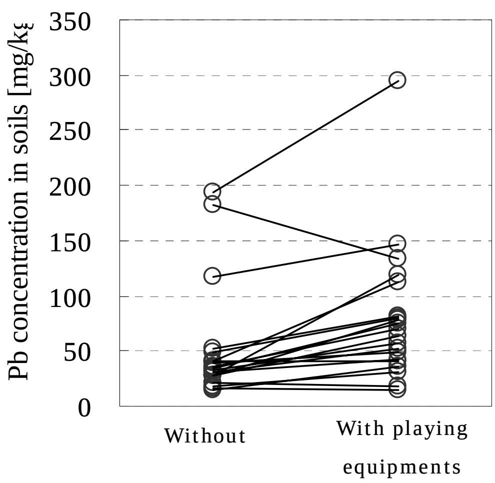

Fig. 2 Comparison of Pb concentration in grids with/without playing equipments for 20 playgrounds in Tokyo

In 17 playgrounds out of 20, where grids (10 m × 10 m) with and without painted playing equipments were present, Pb concentration was higher in the grid with painted playing equipment than that without it (Z = -3.136, p = 0.002, Wilcoxon's matched pair signed rank test) (Fig. 2).

Correlation with the Pb concentration in soil and the degree of peeled-off of paint

Figure 3 shows the correlation between POI and soil Pb concentration for 31 playgrounds. This positive correlation was statistically significant (Spearman rank-correlation coefficient, r = 0.368, p = 0.041).

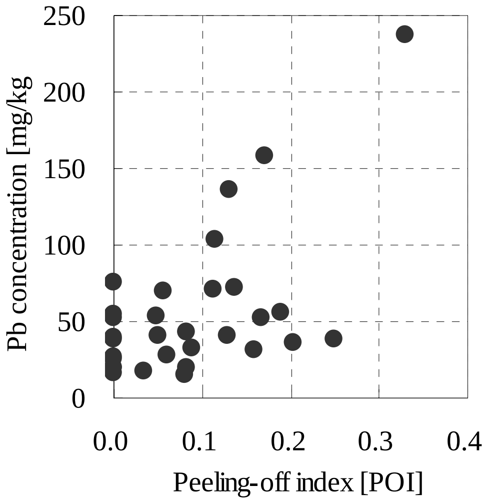

Fig. 3 Correlation between Pb concentration and POI

Lead isotope composition of the playground soils

Figure 4 shows Pb isotope ratios of surface soils of the playgrounds. Lead isotopic composition of the playground soil distributed with rather wide range. Although the predominant sources of Pb in surface soil can be assumed to be native soil and atmospheric fallout, Pb isotopic compositions of some soil samples were distinct from these sources indicating contribution from gasoline Pb of the past. Figure 5 shows the correlation between Pb concentration and isotopic ratio. This positive correlation was statistically significant (Spearman rank-correlation coefficient, $^{207}Pb/^{206}Pb$ and Pb conc. r = 0.536, p = 0.002; $^{208}Pb/^{206}Pb$ and Pb conc., r = 0.600, p < 0.001).

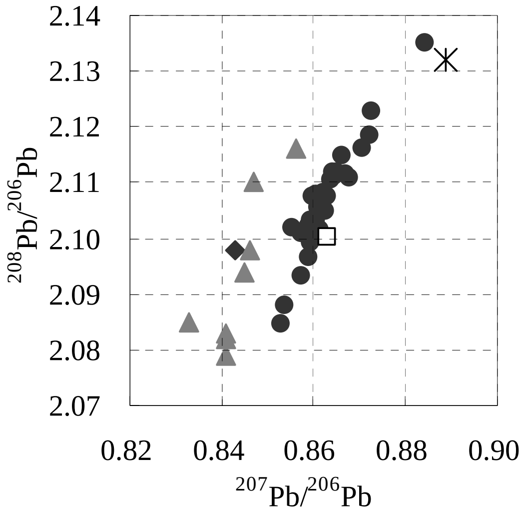

Fig. 4 Plot of Pb isotope ratios in playground soil and other environmental samples

※●This study, □Airborne particulate matter (*11*), ▲Ore (*12*),◆Bed rock and soil (*13*), ＊Leaded gasoline (*11*)

4. DISCUSSION

The results shown in Fig. 1 and Fig. 2 indicated that paint chips peeled off from the playing equipments contributed to the elevated Pb concentrations observed in playground soils in Tokyo. It is possible that the paint chips are reduced to smaller fragments and particles by weathering and the fragments/particles will eventually be mixed into the soil particles.

However, the correlation between the Pb concentrations in playground soil (the average of all grids in playground) and POI of the playing equipments in the playground was only marginally significant (Fig. 3). I can assume that there are technical inadequacies in this data analysis that could have led to marginal significance (e.g., repainting). Besides these technical problems, large playground-to-playground variation in soil Pb concentration can also be the reason of the marginal significance of POI-Pb concentration relationship. In fact, Pb concentrations of the surface soils of the grids in playgrounds without any painted equipments (n = 37 grids from 31 playgrounds) ranged from 11.8 to 229 mg/kg. Thus the larger variation of Pb concentration in surface soil might have masked relatively smaller variation caused by paint chips when multiple playgrounds are involved in the comparison.

Lead isotopic composition of our playground soil samples distributed with rather wide range (Fig. 4). According to the list of Pb isotope ratios in a number of environmental media in Japan (*10*), no other sources than gasoline Pb distribute to significantly less radiogenic direction beyond the atmospheric particles. Thus it can be reasonable to assume that playground soil with isotope ratios close to gasoline Pb does contain gasoline Pb. In addition, there was a significantly positive correlation between the isotopic ratios and Pb concentration showing that playground with gasoline signature had higher Pb concentration. Thus my isotopic result was consistent with Aung et al.'s in that gasoline Pb emitted in the past was the major source of excess Pb in the surface soil of playgrounds in Tokyo (*6*).

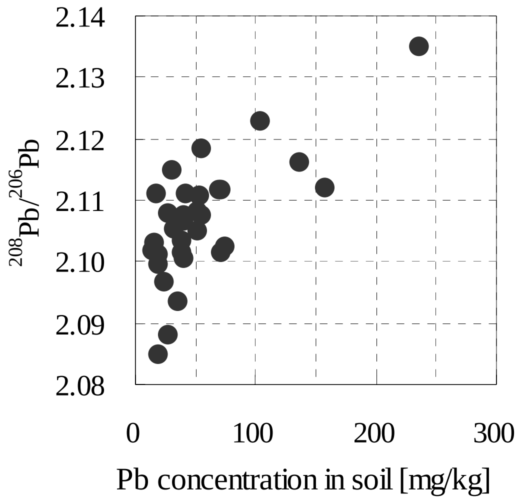

Fig. 5 Correlation with Pb concentration and $^{208}Pb/^{206}Pb$

The correlation coefficients between Pb concentration and isotopic ratios indicate that approximately 30-40% of playground-to-playground variation in Pb concentration can be explained by isotope ratio, i.e., by the contribution from gasoline Pb. On the other hands, paint chips can explain additional 10% of the variation. The playground with greater contribution from gasoline, consequently with higher Pb concentration, was constructed prior to 1970s, when leaded gasoline was still in use in Japan, hence the year of construction of playground was certainly one of the factors of the variation.

5. CONCLUSION

My analysis indicated that there were at least two factors that contributed to variation in the Pb concentration in surface soil of public playground in Tokyo: gasoline Pb and paint chips from playing equipment. The correlation coefficients indicated that gasoline Pb exhausted from automobile before 1980s influenced more to Pb concentration in public playground soil than did paint chips peeled off from playing equipments, the contribution of which was relatively small but still statistically significant. In order to decrease Pb concentration in the surface soil of public playground in Japan, consequently to reduce Pb intake of children, it is necessary to replace surface soil to remove excess Pb due to the past gasoline Pb as well as to use lead-free anticorrosive pigment to playing equipments to reduce Pb from paint chips.

LITERATURE CITED

(1) Lanphear, B. P. ; Dietrich, K. ; Auinger, P. ; Cox, C. *Public Health Reports* **2000**, 115, 21-529.
(2) Canfield, R. L. ; Henderson, C. R. ; Cory-Slechta, S. A. ; Cox, C. ; Jusko, T. A. ; Lanphear, B. P. *N. Engl. J. Med.* **2003**, 348, 1517-1526.
(3) Mieke, H.W. ; Reagan, P. L. *Environ. Health Perspect.* **1998**, 106 (Suppl.1), 217-229.
(4) Yoshinaga, J. Organolead compounds in the environment. In "Organometallic compounds in the environment" Edited by Craig P., Wiley, Chichester 2003, pp. 151-194.
(5) Aung, N. N. ; Yoshinaga, J. ; Takahashi, J. *Environ, Health Prevent. Mad*. **2004**, 9, 257-261.
(6) Aung. N. N. ; Yoshinaga, J ; Tanaka, A. *J. Environ. Chem.* **2004**, 14, 545-553.
(7) Irie, K. ; Maeda, N. ; Yoshida, K. ; Kaniwa, M. *Nihon Kashei Gakkai-shi* **1997**, 48, 1103-1109. [in Japanese]
(8) Okamoto, K. ; Fuwa, K. *Anal. Chem.* **1984**, 56, 1758-1760.
(9) Asami, T. Heavy metal pollution in soils of Japan. Asami T. ed: Japan Scientific Society Press, Tokyo [in Japanese]
(10) Aung, N.N. ; Uryu, T. ; Yoshinaga, J. *Anal. Sci.* **2004**, 20, 195-198.
(11) Mukai, H. ; Furuta, N. ; Fujii, T. ; Ambe, Y. ; Sakamoto, K. ; Hashimoto, Y. *Environ. Sci. Technol.* **1993**, 27, 1347-1356.
(12) Sato, K. *Econ. Geol*. 1975, 70, 800-805.
(13) Murozumi, M. ; Nakamura, S. ; Yochida, K. *Nihon Kagakukai-shi* **1982**, 9, 1479-1484. [in Japanese]