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Scale and causes of lead contamination in Chinese tea

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32% of Chinese tea samples exceeded the national maximum permissible concentration for Pb.

Abstract

We investigated the scale and causes of Pb contamination in Chinese tea. Lead concentrations in 1225 tea samples collected nationally between 1999 and 2001 varied from <0.2 to 97.9 mg kg^{-1} dry weight (DW), with 32% of the samples exceeding the national maximum permissible concentration (MPC) of 2.0 mg kg^{-1} DW and a significant difference between tea types. There was an increasing trend in tea Pb concentration from 1989 to 2000. Proximity to highway and surface dust contamination were found to cause elevated Pb concentrations in tea leaves. Furthermore, Pb concentration in tea leaves correlated significantly and positively with soil extractable Pb, and negatively with soil pH, suggesting that root uptake of Pb from soils also contributed to Pb accumulation in tea. Potential contributions to human Pb intake from drinking tea were small at the median or national MPC Pb values, but considerable at the highest concentration found in the study.

Keywords: Pb contamination; Tea; Human intake; Atmospheric deposition

1. Introduction

Lead (Pb) is a physiological and neurological toxin that can affect almost every organ and system in the human body (Agency for Toxic Substances and Disease Registry (ATSDR), 1999; Joint FAO/WHO Expert Committee on Food Additives, 2000). The most sensitive part of the body is the central nervous system, particularly in children. The most critical effect of Pb at low concentrations is reduced cognitive development and intellectual performance in children. Lead also damages kidneys and the reproductive system. Lead poisoning, especially in young children, is an environmental and public hazard of global proportions (Adriano, 2001). The main exposure to Pb of the general non-smoking adult population is from food and water, whilst for infants and children, food, air, water and dust or soil are the main potential sources of exposure (Joint FAO/WHO Expert Committee on Food Additives, 2000). Foods may contain Pb from the environment (e.g. uptake by plant roots or foliage) or from food processing and storage (e.g. containers).

The Joint FAO/WHO Expert Committee on Food Additives established a provisional tolerable weekly intake (PTWI) of Pb at 25 µg kg^{-1} body weight for infants and children, and later extended this value to people of all age groups (Joint FAO/WHO Expert Committee on Food Additives, 1993). Human Pb intake in developed countries has declined significantly during the last two decades as a result of the ban on the use of

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Pb in gasoline and other legislations on the use of Pb in water supply systems, soldered cans and paints. In most countries in Western Europe and North America, adult weekly Pb intake is estimated to be less than 5 μg kg^{-1} body weight. Lead intake may be higher in the populations in developing countries. For example, average weekly intake of Pb is estimated at 10.1 and 24.4 μg kg^{-1} body weight for Chinese adults and children, respectively (Joint FAO/WHO Expert Committee on Food Additives, 2000). Excessive blood Pb levels (>10 μg l^{-1}) have been reported in a large proportion of children living in Chinese cities (Washam, 2002).

Drinking tea is a popular refreshment in China and many other countries, but the potential Pb intake from drinking tea was not included in the above dietary intake assessments. Tea plants are normally grown in highly acidic soils, where Pb is potentially more bioavailable for root uptake. Furthermore, tea plants have a large leaf area, a characteristic that is conducive to foliar deposition and/or uptake of Pb from the atmosphere. Rapid industrialization in China over the last two decades has resulted in an increased Pb burden to the environment. There are an increasing number of cases of tea products exceeding the maximum permissible concentration (MPC) for Pb of 2 mg kg^{-1} dry weight set by the Chinese Ministry of Health (Chinese Ministry of Health, 1988). This paper presents results from our recent studies into the extent and causes of Pb contamination in Chinese tea. This information is needed for devising strategies for combating the Pb contamination problem. In addition, potential contributions to human Pb intake from drinking tea are estimated.

2. Materials and methods

2.1. National surveys

In the period 1999–2001, 1225 tea samples were collected from 17 tea producing provinces in China (Zhejiang, Jiangsu, Anhui, Fujian, Guangdong, Yunnan, Sichuan, Jiangxi, Hunan, Hubei, Guangxi, Guizhou, Shangdong, Shanxi, Hainan, Henan and Chongaing). Samples were collected in two ways: (1) representative tea producers including tea farms and tea processing companies were pre-selected and requested to send their samples for analysis; and (2) tea producers sent in their samples for quality checking prior to sale or export. The samples included four main types of tea produced in China: green, black, Oolong and scented teas. The first three types differ in the varieties of tea plants and the way tea leaves are plucked and processed. Green and scented teas are similar, except that the latter is blended with jasmine flowers during processing.

In 1989, 1992, 1998, 2000 and 2001, premium grade tea samples were collected. These were the samples submitted by tea producers for an annual competition in the national agricultural exhibition, and subsamples were taken for Pb analysis.

2.2. Influence of highways on Pb concentration in tea leaves

Three tea plantations in the proximity of national highways were selected: Jingjialing, near Hangzhou city, Yongchang in Fuyang county, and Shangyu tea farm, all in Zhejiang province. The Hangzhou and Fuyang tea plantations are near the GD-320 national highway, and the Shangyu tea farm near the GD-104 national highway. Selected soil properties for the three sites are shown in Table 1. In May 2002, tea leaves were plucked at the distances of 10, 30, 60, 100, 150 and 200 m from the highway. Two types of samples were collected, each in triplicate: shoots comprising the unopened leaf buds plus two youngest open leaves (bud + 2 leaves), and old leaves left over from the growth in the previous year (old leaves). Samples were dried in an oven at 80 °C for 24 h.

2.3. Leaf washing experiment

This experiment was carried out to investigate the influence of surface dust contamination on the concentration of Pb in tea leaves. In April 2002, tea leaves were collected from a tea garden in the experimental field of the Tea Research Institute, near Hangzhou. Soil properties are shown in Table 1. Leaves were plucked as bud + 1 leaf, bud + 2 leaves, bud + 3 leaves, bud + 4 leaves, bud + 5 leaves, and old leaves, respectively. Four replicates were collected for each leaf type, two of which were unwashed and the other two washed with distilled water for 20 min. During washing, each leaf was brushed gently by fingers to remove dust on the leaf surface. After washing, the samples were dried in an oven at 80 °C for 24 h.

Table 1
Selected properties of soils used for the influence of highway and leaf washing experiments

Site	pH (in H_2O)	Organic matter (%)	Total Pb (mg kg^{-1})	Extractable Pb (mg kg^{-1})
Hangzhou Jingjialing[a]	4.85	2.34	80.0	4.0
Huyang Yongchang[a]	4.39	2.12	71.2	4.2
Shangyu tea farm[a]	3.54	4.73	87.2	4.6
Tea Research Institute[b]	3.17	5.60	51.9	6.9

[a] Used for the influence of highway experiment.
[b] Used for the leaf washing experiment.

2.4. Relationship with soil Pb

To determine whether Pb concentrations in tea leaves correlate with soil Pb and pH, 224 paired soil and tea leaf samples were collected from different tea gardens in Zhejiang province in April and May 2002. The fields selected for sampling covered a wide range of tea plantations, from low to high yielding, hilly to flat lands, young to old tea bushes, and from highly acidic soils to limed soils. Tea samples were one bud + 2 leaves, unwashed, and dried at 80 °C for 24 h. Soils were sampled from the depths of 0–20 cm and 20–40 cm, respectively, and air-dried.

2.5. Tea infusion experiment

To determine the dissolution rate of tea Pb into the infusion, 3 g ungrounded tea was placed in 50 ml of boiling distilled water in a flask. The flask was maintained at about 100 °C for 10 min in a boiling water bath. The infusion was then filtered and Pb concentration in the filtrate was determined by ICP-AES.

2.6. Analytical methods

Dried tea samples were ground into a powder with a stainless steel grinder. Tea samples collected in 1998–2002 were digested in a microwave digestion oven (CEM Mars5, CEM Corp, Matthews, NC). Approximately 0.5 g of each sample was weighed into a Teflon® coated tube and 5 ml of concentrated HNO_3 (high purity grade) was added. The samples were allowed to predigest for 15 min at room temperature and then transferred to the microwave oven. Digestion was carried out at 170 °C for 10 min at a maximal pressure of 800 psi. Samples were cooled to room temperature and diluted to 25 ml with high quality deionized water (>18 MΩ specific resistance). Lead concentration was determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES) (IRIS/AP, Thermo Jarrell Ash Corporation, Franklin, USA). The detection limit for Pb was 0.004 mg l^{-1}, equivalent to 0.2 mg kg^{-1} in tea samples. Blanks and a reference tea material (GB07605-GSV-4, Institute of Geophysical and Geochemical Sciences, Chinese Academy of Geological Sciences, Langfang, Hubei, China) with a certified Pb concentration (4.4 ± 0.2 mg Pb kg^{-1}) were included for quality control. Repeated analysis of the reference material gave a mean Pb concentration of 4.4 mg kg^{-1}, with a standard deviation of 0.08 mg kg^{-1} ($n = 5$). The recovery rate for externally added Pb was 99.1–102.0%. Tea samples collected in 1989 and 1992 were analyzed for Pb using flame atomic absorption spectrometry (AAS, Shimazu AA-646, Japan). Tea samples weighing approximately 5.0 g were ashed at 500 °C for 6 h, and the ash was dissolved with 2 ml 6 M HCl. Lead in the solution was pre-concentrated using 0.01% (w:v) dithizone dissolved in *n*-butyl acetate before the AAS measurement (Institute of Tea Research, 1989). The method gave a mean Pb concentration of 1.01 mg kg^{-1} for the reference tea material GBW 08505, which has a certified Pb concentration of 1.06 ± 0.10 mg kg^{-1}. To ensure that the two methods of Pb analysis were comparable, we re-analyzed five samples from the 1989 collection using the microwave digestion/ICP-AES method and obtained an excellent agreement between the two methods ($y = 1.04x - 0.02$, $R^2 = 0.999$; where y and x are Pb concentrations by the ICP-AES and AAS methods, respectively).

Soil samples were ground to <0.8 mm for the determination of extractable Pb, pH and organic matter, and to <0.016 mm for total Pb determination. For the determination of soil total Pb, soils were digested with a solution of $HF-HClO_4-HNO_3$ (Liu, 1996), and Pb determined by ICP-AES. Soil extractable Pb was extracted with 0.1 M HCl (1:5 soil to solution, w:v) for 1.5 h in a reciprocal shaker (Xin et al., 1990), and Pb determined by ICP-AES. Soil pH was determined using a combined glass electrode in 1:1 (w:v) soil-water suspensions. Soil organic matter was analyzed by the dichromate method (Liu, 1996).

Data are expressed on a dry weight (DW) basis. Statistical analyses, including multiple linear regression and analysis of variance (ANOVA) were performed using Genstat® 5 for Windows® (Numerical Algorithms Group, 1998). Where appropriate, data that showed skewed distributions were log-transformed prior to statistical testing to stabilize variance. For samples having Pb concentrations below the analytical detection limit (<0.2 mg kg^{-1} DW), concentrations equal to half the value of the detection limit were used in the subsequent statistical analyses.

3. Results and discussion

3.1. Pb concentrations in Chinese teas: a nationwide survey in 1999–2001

The concentration of Pb in the 1225 tea samples collected in 1999–2001 from major tea growing areas in China varied from below the detection limit (0.2 mg kg^{-1} DW) to 97.9 mg kg^{-1} DW, with a mean and median of 2.7 and 1.4 mg kg^{-1} DW, respectively. Fig. 1 shows the frequency distributions and box plots for tea Pb concentrations in different tea types. ANOVA on log-transformed data showed significant ($p < 0.001$) differences in the Pb concentrations between different tea types. Green and scented teas had a lower median and mean Pb concentrations than black and Oolong teas. Many samples exceeded the Chinese MPC of 2.0 mg kg^{-1} DW, accounting for 24, 32, 59 and 53% of

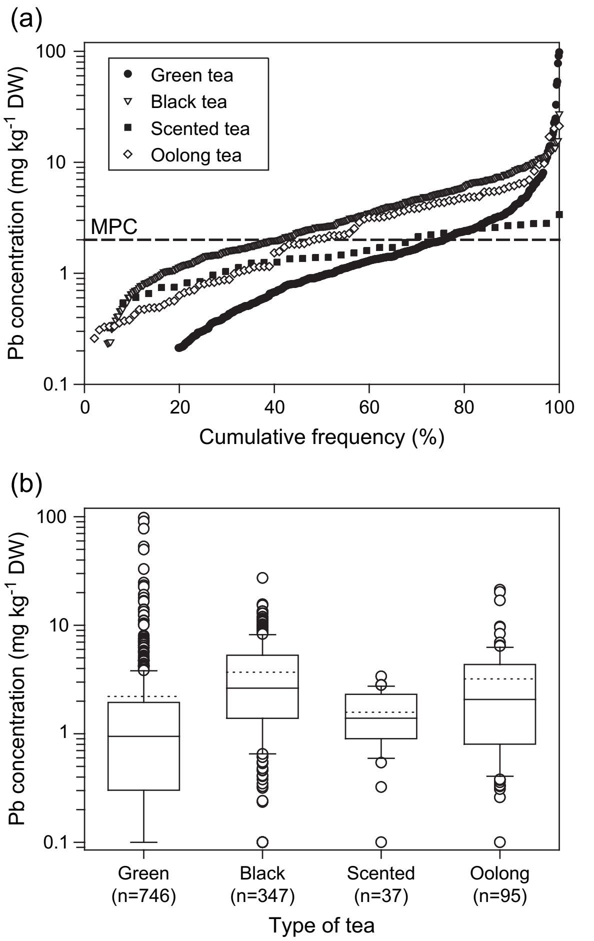

Fig. 1. Frequency distributions (a) and box plots (b) of Pb concentration in different tea types in the 1999–2001 survey. Samples with Pb concentrations lower than the detection limit (0.2 mg kg^{-1} DW) were assigned a value of 0.1 mg kg^{-1} DW for the construction of frequency distribution and box plot. The central solid line within each box is the median, and the bottom and top of each box represent the 25th and 75th percentiles, respectively. The dotted line is the mean. The whiskers represent the 10th and 90th percentiles, respectively, and values outside this range are plotted as individual outliers (○).

the total numbers of samples analyzed for green, scented, black and Oolong teas, respectively. Overall, 32% of the samples exceeded the MPC. This is the most comprehensive survey of tea Pb concentrations in China, and highlights the scale of Pb contamination. The higher exceedance for black and Oolong teas than for green and scented teas can be explained by the different types of leaves that are picked for different teas; more and older leaves are harvested for producing black and Oolong teas than for green and scented teas. Older leaves tend to contain higher concentrations of Pb than younger leaves (see below, also (Ramakrishna and Palmakumbura, 1987; Natesan and Ranganathan, 1990)). The concentrations of Pb in the majority of the tea samples analyzed were markedly higher than those reported world-wide for any other food categories (mostly <0.5 mg kg^{-1} DW) (Wolnik et al., 1983; Wiersma et al., 1986; Ward and Savage, 1994; Joint FAO/WHO Expert Committee on Food Additives, 2000; Zhao et al., 2004). For comparison, a previous report showed Pb concentrations in 139 green teas produced in Japan varying from 0.11 to 1.93 mg kg^{-1} (Tsushida and Takeo, 1977).

3.2. Differences in tea Pb concentration between normal and premium grades

The main difference between the normal and premium grades is in the number of leaves per shoot picked for tea processing. For green tea, normal grade tea is usually made from shoots having the unopened leaf bud and three to four leaves, whereas to make premium grade tea only one to two leaves and the unopened leaf bud per shoot are used. This difference could contribute to the difference in Pb concentration because it tends to increase with leaf age (see below). Of the 118 premium and 184 normal grade green teas sampled from Zhejiang province between 1999 and 2001, the median and mean Pb concentrations were about 40% and 60%, respectively, lower in the premium tea than in the normal tea (Fig. 2). ANOVA on log-transformed data showed a significant ($p<0.001$) difference in Pb concentration between the two quality grades. Furthermore, only 15% of the premium grade tea exceeded the MPC, as compared to 37% of the normal grade tea.

3.3. Changes over time in tea Pb concentration

In addition to the large survey conducted in 1999–2001, we also analyzed tea samples of the premium grade collected in 1989, 1992, 1998, 2000 and 2001 to assess the trend of tea Pb concentrations over time. There is a clear increasing trend in the Pb concentration

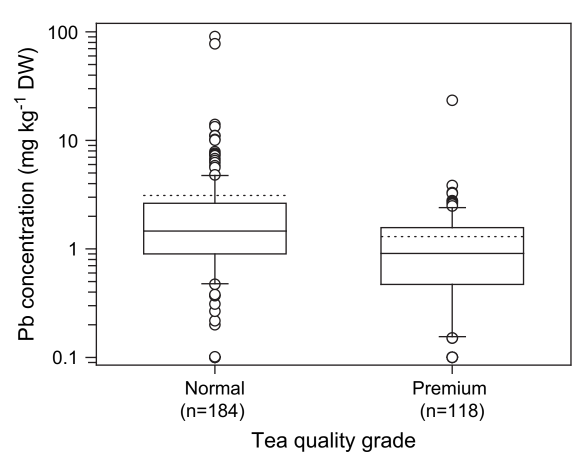

Fig. 2. Box plot of Pb concentration in normal and premium grade green teas collected in 1999–2001 from Zhejiang provinces. See caption for Fig. 1 for explanation of box plot.

in the premium grade teas from 1989 to the more recent years of 1998–2001 (Fig. 3). Median values were 0.44, 0.49, 0.69, 0.76 and 0.66 for the years 1989, 1992, 1998, 2000 and 2001, respectively. ANOVA on log-transformed data showed significant ($p<0.001$) differences in Pb concentrations between different years. The percentage of samples exceeding the MPC also increased, from 4.8% in 1989 to 9.3% in 2001. The increasing trend may be attributed to the rapid industrialization in China over the last decade, and in particular, the growth of traffic and the consumption of fuel. For example, the number of vehicles in Hangzhou, the capital city of Zhejiang province, increased by 5.5-fold between 1989 and 2001. Unleaded gasoline was first introduced in Beijing and Shanghai in 1993, and gradually in other large cities in China. The whole country has stopped producing and selling leaded gasoline since mid-2000. The small downward shift in the median Pb concentration in 2001, compared to that in 2000, may be the very first indication of a decreasing trend of Pb emissions and deposition since the ban of leaded gasoline.

3.4. *Influence of highways on Pb concentrations in tea leaves*

Before the introduction of unleaded gasoline, automotive emissions had a major influence on Pb concentrations in roadside soil and vegetation (Ward et al., 1974, 1977; Kelly et al., 1996). To evaluate the impact of highways on Pb concentrations in tea leaves, we selected three plantations in Zhejiang province which were in close proximity to major highways. Tea leaves were picked at locations from 10 to 200 m away from the highway. On average, old leaves had 2–2.5-fold more Pb than young leaves (one bud + 2 leaves). Fig. 4 shows that, in both old and young tea leaves, Pb concentration

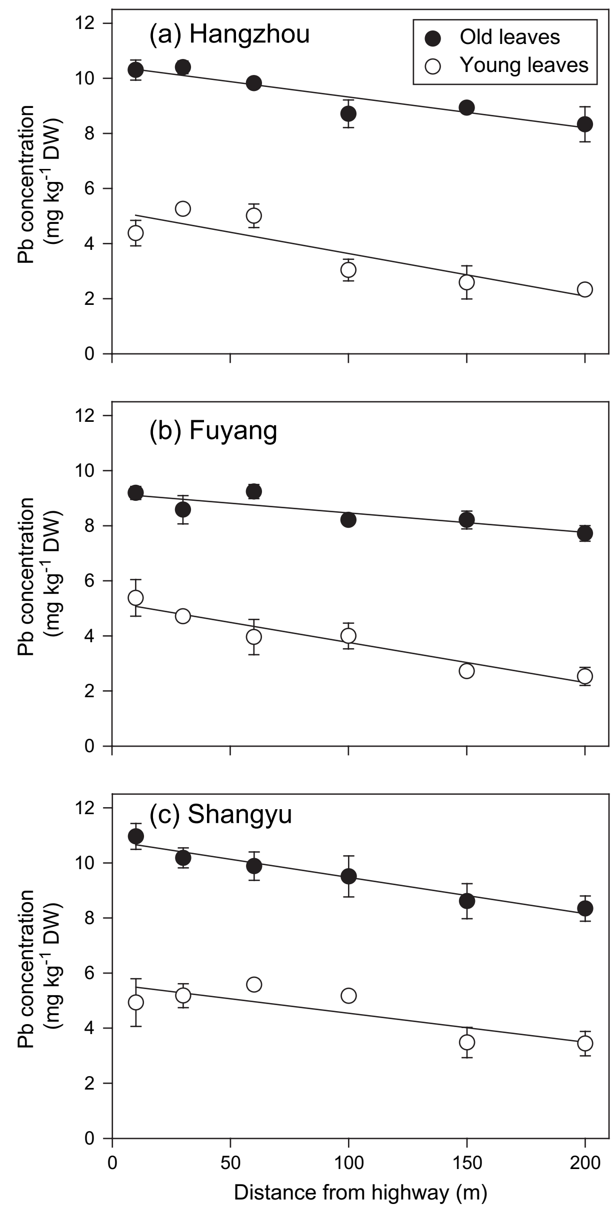

Fig. 4. The influence of the proximity to highway on the concentration of Pb in tea leaves at three locations in Zhejiang province: (a) Hangzhou; (b) Fuyang; and (c) Shangyu.

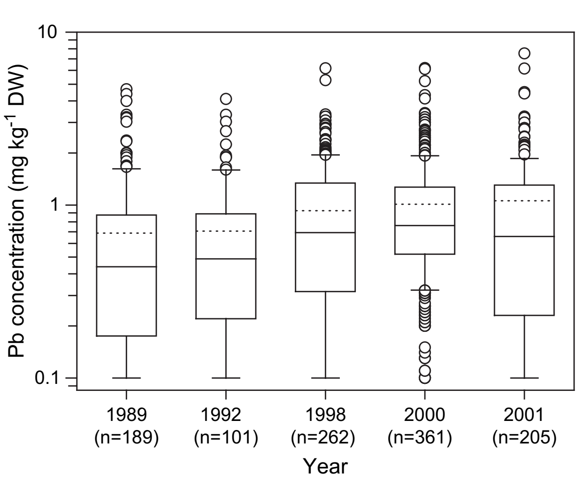

Fig. 3. Box plot of Pb concentration in premium grade teas collected nationwide in 1989, 1992, 1998, 2000 and 2001. See caption for Fig. 1 for explanation of box plot.

decreased linearly with increasing distance from the highway in all three plantations, with linear slopes varying from −0.007 to −0.015 mg Pb kg^{-1} per m distance ($p<0.001$). The results demonstrate a clear influence of highway traffic on Pb concentration in tea leaves, even though unleaded gasoline had been introduced 1.5 years before this work was conducted. Because tea leaves were unwashed in this experiment, it is likely that samples collected closer to the highway would have more soil dust on the leaf surface, which would have an effect of increasing Pb concentration in the samples because of rather high concentrations of total Pb in the soils (Table 1).

3.5. Effect of washing on Pb concentration in tea leaves

To determine the contribution of surface contamination (e.g. dust) to the accumulation of Pb in leaves, freshly plucked leaves were washed with distilled water for 20 min. Lead concentrations increased markedly with leaf age in both washed and unwashed leaves (Fig. 5). Washing had little effect on Pb concentration in the samples containing the unopened leaf bud and 1 or 2 young leaves. However, washing removed considerable amounts, up to 23%, of Pb from older leaves (Fig. 5), indicating that surface contamination did contribute to the accumulation of Pb in tea. Similarly, Tsushida and Takeo (1977) reported that washing removed up to 50% of Pb in tea leaves. It is possible that a brief washing with distilled water only would not remove all surface contamination. If so, the extent of surface contaminated Pb could be greater than shown in Fig. 5. However, even after washing, older leaves still contained markedly more Pb than younger leaves, suggesting that either root uptake and/or penetration of surface deposited Pb across leaf cuticles may contribute to Pb accumulation. Although washing can reduce tea Pb concentration, this is not practical for large scale tea processing. However, tea leaves collected after rainfall may be expected to have a lower Pb concentration.

3.6. Relationship between soil Pb and Pb concentration in tea leaves

Previous studies have shown that foliage deposition and/or foliar uptake of Pb from the atmosphere is the predominant route of entry to plants, whereas root uptake of Pb contributes little to the accumulation of Pb in the above-ground parts. Using an isotope-labelling

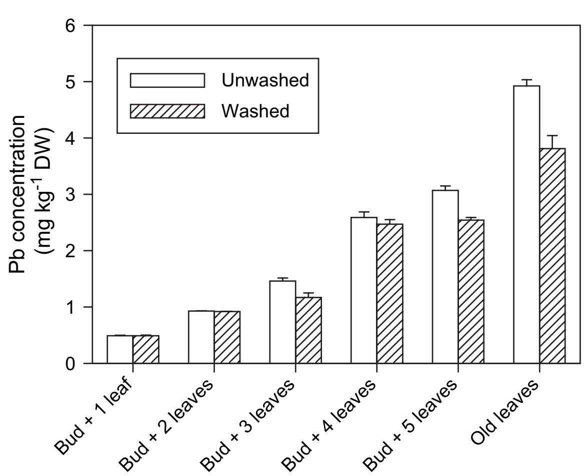

Fig. 5. The effect of washing with distilled water on the concentration of Pb in different ages of tea leaves.

technique, Tjell et al. (1979) showed that between 90 and 99% of total Pb in ryegrass was derived from atmospheric Pb in a relatively uncontaminated area in Denmark. Similarly, Dalenberg and Vandriel (1990) reported that 100% of wheat grain Pb was derived from atmospheric deposition and none from uptake from soil in an experiment in the Netherlands. A recent study of Pb concentration in wheat and barley grain collected nationwide in the UK showed no significant correlation between soil Pb and grain Pb concentrations (Zhao et al., 2004). These findings are not surprising, given that the above studies included soils with pH ranging from slightly acidic to alkaline, and therefore low solubility and bioavailability of soil Pb is expected (Jopony and Young, 1994; Sauvé et al., 1998a; Sauvé et al., 1998b). In contrast, tea is normally grown on acidic soils, where Pb solubility could be much higher.

To investigate the relationship between soil extractable Pb and the concentration of Pb in tea leaves, we collected tea leaves (bud + 2 leaves) and corresponding soils samples from the 0–20 cm and 20–40 cm depths from 224 plantations in Zhejiang province in 2002. Soil pH in the top 20 cm depth ranged from 3.02 to 6.91, with a median of 4.73. The concentration of 0.1 M HCl extractable Pb in the soils varied from 1.1 to 32.2 mg kg^{-1}, with a median of 5.1 mg kg^{-1}. Tea Pb concentration was found to correlate highly significantly with soil extractable Pb in the top 20 cm depth ($r = 0.43$, $p < 0.001$, based on the log transformed data; Fig. 6a). Tea Pb concentration (log transformed) also correlated significantly ($r = 0.30$, $p < 0.001$), but negatively, with soil pH in the 0–20 cm depth (Fig. 6b). Soil extractable Pb and pH together explained 25% of the variation in tea Pb concentration according to the following regression equation:

$$\log(\text{Tea Pb}) = 0.33 + 0.48\log(\text{Soil extractable Pb}) - 0.08\,\text{pH}\left(R^2_{\text{adj}} = 0.25, p < 0.001\right)$$

The coefficients for both soil extractable Pb and pH were highly significant ($p < 0.001$). There was also a significant correlation between tea Pb and soil extractable Pb in the 20–40 cm depth (data not shown), though less close than with the data of the 0–20 cm depth. We conclude from this relationship that root uptake of Pb from soil is an important pathway of Pb entry to tea leaves, which is different from previous studies with other plant species growing on less acidic soils. The results also imply that, on highly acidic soils with elevated Pb concentration, tea Pb concentration would continue to be elevated after a total ban on lead in gasoline. A practical option for dealing with such a problem is to increase soil pH by liming with $CaCO_3$/$MgCO_3$.

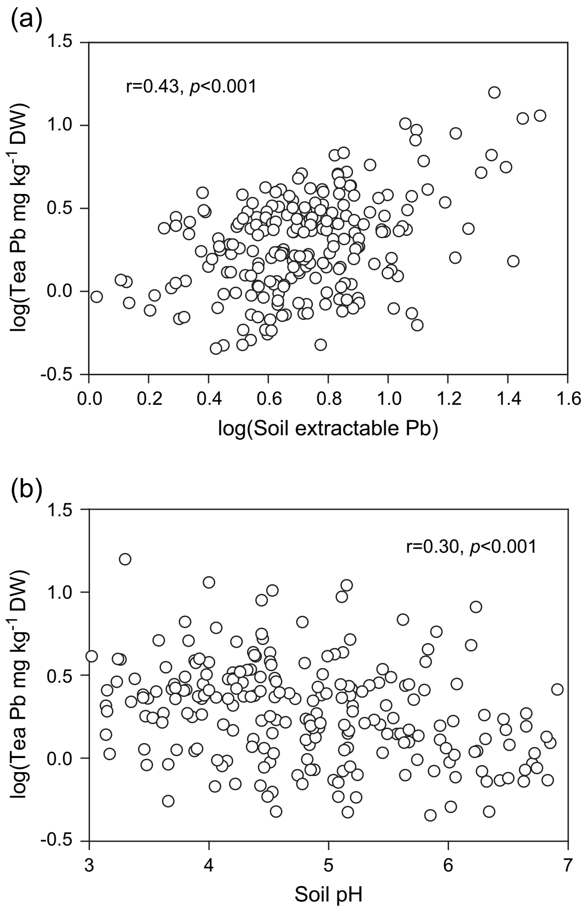

Fig. 6. The relationship between Pb concentration in tea leaves and soil extractable Pb (a) and soil pH (b).

3.7. Implications for Pb intake by humans

Weekly intakes of Pb by adults in Northern America, Australia and most Western European countries were estimated to be less than 5 μg kg^{-1} body weight, and 10.1 μg kg^{-1} body weight for the Chinese population, well below the current PTWI of 25 μg kg^{-1} body weight (Joint FAO/WHO Expert Committee on Food Additives, 2000). However, these estimates did not consider potential contribution from drinking tea to Pb intake, which depends on the amount of tea consumed, the concentration of Pb in tea, and the dissolution rate of tea Pb into the infusion. For the latter, Michie and Dixon (1977) reported a wide range from 5 to 80%, whereas Natesan and Ranganathan (1990) gave a mean value of 43% after a 5 min brewing. Our preliminary test showed that 5.2–27.0% (mean = 17.4%, $n = 10$) of the total tea Pb was dissolved into the infusion. Average annual tea consumption in China is rather low, at 0.36 kg per capita (compared to 2.76 and 2.77 kg per capita in Ireland and Iraq, respectively, the most tea loving countries in the world (International Tea Committee, 2003)). This figure does not take into account that not everybody drinks tea and most children do not drink tea. We assume that a regular drinker in China would consume 2 kg tea annually (equivalent to about 2 tea bags per day). Using the median Pb concentration found in the national survey (1.4 mg kg^{-1} DW) and the average dissolution rate of 17.4%, the weekly Pb intake from drinking tea is estimated to be 9.3 μg per person, equivalent to 0.16 μg kg^{-1} body weight (60 kg per person), which represents 0.6% of the PTWI and is small compared to the average weekly total intake of 10.1 μg kg^{-1} body weight for Chinese adults. With Chinese MPC for tea Pb of 2 mg kg^{-1} DW, the weekly intake of Pb would be 0.22 μg kg^{-1} body weight (= 0.9% of the PTWI), indicating the stringent nature of the MPC value. However, with the maximum Pb concentration in tea recorded in the national survey (97.9 mg kg^{-1} DW), drinking tea alone would contribute 653 μg Pb intake per person per week, equivalent to 10.9 μg kg^{-1} body weight and 44% of the PTWI. In this extreme scenario, total Pb intake would not be far away from the PTWI recommended by the Joint FAO/WHO Expert Committee on Food Additives (Joint FAO/WHO Expert Committee on Food Additives, 1993).

4. Conclusions

The present study shows the extent of Pb contamination in Chinese teas, with a considerable proportion (32%) of the samples exceeding the Chinese MPC. There was an increasing trend in tea Pb concentration from 1989 to 2000, reflecting the rapid industrialization and increased consumption of leaded gasoline in China. The results suggest that both atmospheric deposition and root uptake contributed to Pb accumulation in tea leaves; the latter route was made possible because of the generally acidic nature of the soils in tea plantations. The elevated Pb concentration in tea is more likely to affect trade than human health, because potential contributions to human Pb intake from drinking tea were small at the median or at the national MPC Pb values.

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