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Estimates of Soil Ingestion in a Population of Chinese Children

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Background: China's soil pollution poses serious health risks. However, data regarding the soil ingestion rate (SIR) of the Chinese population, which is critical to assessing associated health risks, are lacking.

Objectives: We estimated soil ingestion of 177 Chinese children from Guangdong, Hubei, and Gansu Provinces.

Methods: We conducted this investigation by employing a tracer mass-balance method. We collected a duplicate of all food consumed and all feces and urine excreted on 1 d ($n = 153$) and over 3 consecutive d ($n = 24$), as well as soil samples from play areas and drinking-water samples. We analyzed concentrations of the tracer elements Al, Ba, Ce, Mn, Sc, Ti, V, and Y in these samples using ICP-AES and ICP-MS and estimated the SIR for each subject.

Results: The estimated SIR data based on each tracer element were characterized by a skewed distribution, as well as higher inter-tracer and inter-subject variation, with several outliers. After removing the outliers, daily SIR median (range) values in milligrams per day were Al, 27.8 (−42.0 to 257.3); Ba, 36.5 (−230.3 to 412.7); Ce, 35.3 (−21.2 to 225.8); Mn, 146.6 (−1259.4 to 1827.7); Sc, 54.8 (−4.5 to 292.0); Ti, 36.7 (−233.7 to 687.0); V, 92.1 (10.4 to 308.0); and Y, 59.1 (−18.4 to 283.0). Daily SIR median/95th percentile (range) values based on the best tracer method (BTM) were 51.7/216.6 (−9.5 to 297.6) mg/d.

Conclusions: Based on the BTM, recommended SIR values for the general population of Chinese children (2.5 to 12 years old) are 52 mg/d for the central tendency and 217 mg/d for the upper percentile. We did not differentiate between outside soil and indoor dust. Considering the lower concentration of tracer elements in indoor dust than outside soil, actual soil and dust ingestion rates could be higher. https://doi.org/10.1289/EHP930

Introduction

It is widely recognized that soil contamination in China is of serious concern (Chen et al. 2014; Larson 2014; Yang et al. 2014). Unintentional soil ingestion is a potentially important route of exposure to soil contaminants, especially for children, who may ingest more soil than adults because of their frequent hand-to-mouth or hand-to-object behaviors (U.S. EPA 2011). Soil ingestion rate (SIR) is a prerequisite for assessing the health risk of contaminated sites to children (U.S. EPA 2011). However, a reliable SIR for Chinese children has yet to be determined.

Doyle et al. (2010), the U.S. EPA (2011), and Moya and Phillips (2014) comprehensively reviewed studies on SIR, showing that the metric was established in the 1970s. Early studies tried to quantify soil ingestion by employing qualitative or semi-quantitative methods, combining hand-to-mouth frequency with soil loadings on the hand (Day et al. 1975; Doyle et al. 2010; Duggan and Williams 1977; Hawley 1985; Kimbrough et al. 1984; Lepow et al.1974).

Since the 1980s, scientists have used the tracer mass-balance method to quantify SIR. Binder et al. (1986) modified the methods used in estimating the amount of soil ingested by ruminants to measure soil ingested by children, neglecting the tracers in food. Subsequent studies (Calabrese et al. 1989; Davis et al. 1990) improved the tracer method of Binder et al. (1986) to estimate a child's SIR by including tracers in food and by increasing the number of tracers. The best tracer method (BTM) was further developed to estimate a child's SIR (Calabrese and Stanek 1993; Calabrese et al. 1997; Stanek and Calabrese 1995). First, the BTM ranks tracer elements according to their food-to-soil (F/S) ratios, defined as the mass of the tracer element ingested from food over a 1-d period divided by the mass of the tracer element in 1 g of soil. Then, the SIR for each subject over the study period is calculated using the median of the SIRs estimated by four tracers with the lowest F/S ratios. Recently, Doyle and colleagues used a higher number of tracers, including radionuclides, to quantify a child's SIR with the tracer mass-balance method (Doyle et al. 2012a; Irvine et al. 2014).

The biokinetic model comparison method (BMC) has also been used to estimate a child's SIR. The model compares direct measurements of a biomarker with predictions from a biokinetic model (U.S. EPA 2011). Hogan et al. (1998) used lead BMC to estimate a child's SIR in three historic lead-smelting communities. Similarly, von Lindern et al. (2003; 2016) compared predicted and measured blood lead levels, showing that the default model soil and dust ingestion rates and lead bioavailability value over predicted blood lead levels.

To the best of our knowledge, available research on SIR is limited, with investigations in children mostly drawn from western Europe and the United States. The SIR of a child from western Europe or the United States is unlikely to be globally representative, due to differences in lifestyle and habits.

Motivated by serious soil contamination in China, we initiated an investigation of Chinese children's SIR, financed by the Ministry of Environmental Protection of China. Two hundred ten children were selected from southeastern to northwestern China (Guangdong, Hubei, and Gansu Provinces). The SIR for each subject was quantified using analogous BTM. This paper reports the results of this study and presents recommended SIR values for Chinese children.

Materials and Methods

Tracer Mass-Balance Method

A tracer mass-balance approach was employed to investigate daily SIR. The general algorithm used to calculate the quantity of ingested soil per day for each child is as follows:

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$$\mathrm{SIR} = \frac{(W_{feces} \times C_{feces} + V_{urine} \times C_{urine}) - (W_{food} \times C_{food} + V_{water} \times C_{water})}{C_{soil}} \quad [1]$$

where, for each child, SIR is the daily soil ingestion rate (mg/d) over the study period, W_{feces} is feces dry weight (g/d), C_{feces} is the concentration of tracer elements in feces (μg/g), V_{urine} is urine volume (mL/d), C_{urine} is the concentration of tracer elements in urine (μg/mL), W_{food} is ingested food weight (g/d), C_{food} is the concentration of tracer elements in food (μg/g), and V_{water} is drinking-water volume (mL/d). C_{water} is the concentration of tracer elements in drinking water (μg/mL) (median concentration for all samples of the three cities was used), and C_{soil} is the concentration of tracer elements in soil (μg/mg) (the median concentrations for each city were used).

Subject Selection

The study was designed to select a population sample of 210 children from southeastern to northwestern China, including 60 children from the Shenzhen area (urban and suburban) of Guangdong Province (southeast China), 90 children from the Wuhan area (urban and suburban) of Hubei Province (central China), and 60 children from the Lanzhou area (urban and suburban) of Gansu Province (northwest China). The distribution and basic statistics of age, height, and weight of the population sample are shown in Figure S1. The children's ages ranged from 2.5 to 11.9 years old, with a median of 5.6 years. The median child's weight was 24.0 kg (12.0–59.0 kg), and the median child's height was 115.0 cm (75–162 cm). Shenzhen, Wuhan, and Lanzhou are located in southeastern, central, and northwestern China, respectively, with high, middle, and low gross domestic product (GDP) levels. Therefore, we hypothesized that SIR levels in the child population from these urban and suburban areas would accurately represent the general SIR of Chinese children. All participants were asked not to take any medicine during the sampling period.

Consent forms were signed by the participants' parents or guardians, and support from teachers and parents was obtained before study initiation. Children, their parents or guardians, and teachers were trained to collect feces, urine, and food samples. Twenty-four children and 186 children participated for 3 consecutive d and 1 d, respectively. A 28-h lag time is generally assumed between ingestion of food and soil and the resulting fecal and urinary output (U.S. EPA 2011). Therefore, sample collection for 1-d participants lasted 52 h (food collection in 24 h and feces and urine collection in the following 28 h; Figure 1a. For the 3-consecutive-d participants, sample collection lasted 100 h (food collection in the first 24-h (1 d), second 24-h (1 d), and third 24-h (1 d) periods and feces and urine collection in the second 24-h (1 d), third 24-h (1 d), and fourth 24-h (1 d) periods, and the following 4 h (Figure 1b).

Food Collection and Analysis

All food samples, including breakfast, lunch, and dinner, were collected for each child using the "duplicate plate" method. After being weighed, all daily food samples were homogenized with a stainless steel blade to form a composite sample for each subject. A 1-g subsample of the composite sample was digested with concentrated nitric acid and hydrogen peroxide (HNO_3-H_2O_2) in a microwave oven. Aluminum (Al), barium (Ba), cerium (Ce), manganese (Mn), scandium (Sc), titanium (Ti), vanadium (V), and yttrium (Y) concentrations in the supernatant were determined by high resolution inductively coupled plasma mass spectrometry (HR-ICP-MS). The concentrations of Sc and V in the supernatant were generally lower than their detection limits

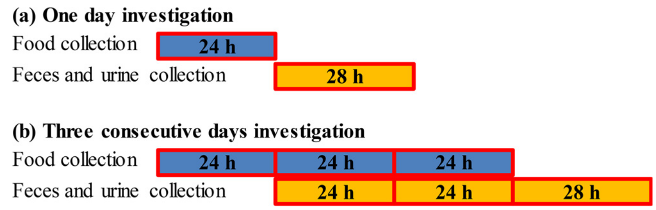


Figure 1. Schematic time processes of food and feces and urine collection. 28-h lag time was recommended by U.S. EPA (2011) [e.g., food collection from d-1 morning (about 0700 hours, including breakfast) to d-2 morning (but not including d-2 breakfast); excreta collection from d-2 morning (about 0700 hours) to d-3 noon (about 1100 hours am)].

(0.005 ng/mL and 0.015 ng/mL, respectively); therefore, their concentrations were assumed to be half their detection limits. The median food wet weights (ww) for each child were 1013.5 g/d, ranging from 267.0 to 1758.7 g/d (see Figure S2a). The median (minimum to maximum) concentrations in nanograms per gram (ww) of tracers in food were 959.1 (244.3 to 5,359.7) for Al, 62.06 (26.83 to 145.38) for Ba, 0.55 (0.02 to 3.05) for Ce, 299.8 (42.4 to 892.0) for Mn, 0.06 for Sc, 343.5 (170.8 to 1,320.3) for Ti, 0.18 for V, and 0.125 (0.001 to 0.959) for Y (see Table S1).

Excreta Collection and Analysis

All feces and urine for each subject were collected daily using pre-labelled and pre-weighed portable sample containers. After measuring urine volume and feces weight, all daily feces and urine samples were evenly mixed to form composite feces and urine samples for each subject. All samples were transported to the laboratory in an ice box. Feces were oven-dried for approximately 72 h at 60°C. The dry weight of the feces samples for each subject was measured. The median and mean weights of dry feces samples were 17.0 and 19.9 g/d, respectively, ranging from 2.4 to 63.8 g/d (see Figure S2b). Notably, 33 subjects did not provide feces samples as required. Median and mean volumes of urine for each subject were 560.0 and 620.4 mL/d, respectively, ranging from 25.0 to 2,143 mL/d (see Figure S2c).

A 1-g proportion of dry feces was digested with concentrated nitric acid, hydrogen fluoride, and perchloric acid (HNO_3−HF−$HClO_4$) on a heating plate. The concentrations of Al, Ba, Ce, Mn, Sc, Ti, V, and Y in the supernatant were determined by HR-ICP-MS. The median (minimum to maximum) concentrations in micrograms per gram dry weight (dw) of these tracers in feces were 178.3 (24.1 to 1,677) for Al, 4.58 (0.31 to 11.03) for Ba, 0.219 (0.016 to 0.806) for Ce, 24.01 (1.30 to 54.78) for Mn, 0.039 (0.002 to 0.151) for Sc, 28.17 (2.05 to 150.45) for Ti, 0.371 (0.033 to 0.829) for V, and 0.101 (0.005 to 0.395) for Y (see Table S2).

Fifteen milliliters of urine was added to a pre-cleaned digestion tube, and subsequently 3 mL of concentrated HNO_3 and 2 mL of H_2O_2 were added. Then, the tube was placed in a microwave digestion reactor. The digestion program was as follows. The temperature was gradually increased to 120°C and maintained for 5 min, then increased to 160°C and maintained for 5 min, and finally increased to 180°C and maintained for 15 min. After digestion, the solutions were diluted to a final volume of 30 mL with ultrapure water and then stored at 4°C for measuring Al, Ba, Ce, Mn, Sc, Ti, V, and Y by HR-ICP-MS. The median (minimum to maximum) concentrations in micrograms per liter of these tracers in urine were 88.6 (5.0 to 709.1) for Al, 20.55 (2.94 to 38.64) for Ba, 0.34 (0.02 to 0.59) for Ce, 3.95 (1.66 to 9.01) for Mn, 0.08 (0.01 to 0.20) for Sc, 7.87 (1.02 to 35.34) for

Ti, 1.54 (0.20 to 2.49) for V, and 0.08 (0.01 to 0.21) for Y (see Table S3).

Soil Sampling and Analysis

Topsoil samples were collected from places where children generally play (campuses and green spaces near the child's home) by scraping the surface layer of soil from a 10 cm by 10 cm area to a depth of 2 cm. In total, 52, 88, and 78 soil samples were collected in Shenzhen, Wuhan, and Lanzhou, respectively. In the laboratory, soil samples were air-dried, crushed with a ceramic mortar and pestle, and passed through a 0.25-mm sieve. A 0.5-g proportion of the crushed soil samples was digested with concentrated $HNO_3-HF-HClO_4$ on a heating plate. The concentrations of Al, Ba, Mn, Ti, and V in the supernatant were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES), whereas Ce, Sc, and Y were determined by ICP-MS (see Tables S4–S6). The median tracer concentration in the soil of each city was used to calculate the SIR for children from that city because of soil heterogeneity and uncertainty regarding the area of activity of each child.

Drinking-Water Sampling and Analysis

Drinking water was sampled from schools and families, with a total of 85 samples taken from three cities. In the laboratory, drinking-water samples were filtrated through a 0.45-μm filter and measured for Al, Ba, Ce, Mn, Sc, Ti, V, and Y by HR-ICP-MS (see Table S7). Because the difference in tracer concentrations in drinking water was small among the three cities, the median tracer concentration of all samples was used to calculate SIR by Equation 1.

Quality Control

The accuracy of the method for analysis of tracers in soil and feces was tested by analyzing the same elements in certified reference materials (GSS11, GSS13, GSS17, GSS21, GSS22, GSS25, and GSS26; see Table S8). The average (minimum to maximum) recovery of tracer elements was 100.7% (86.1% to 111.0%) for Al, 100.7% (93.0% to 111.4%) for Ba, 97.6% (84.0% to 109.5%) for Ce, 103.5% (97.1% to 110.4%) for Mn, 100.6% (89.1% to 115.8%) for Sc, 101.7% (92.0% to 113.1%) for Ti, 101.2% (96.0% to 107.4%) for V, and 104.7% (81.1% to 116.2%) for Y.

The accuracy of the method for analysis of tracers in food and urine was tested by spiking a known amount of these elements in urine samples. The average (minimum to maximum) recovery of tracer elements was 98.4% (87.1% to 110.5%) for Al, 100.4% (92.5% to 110.1%) for Ba, 99.8% (90.1% to 106.7%) for Ce, 106.9% (96.1% to 119.6%) for Mn, 97.8% (83.5% to 115.9%) for Sc, 97.3% (87.2% to 113.9%) for Ti, 99.9% (91.5% to 114.3%) for V, and 96.1% (82.2% to 108.4%) for Y (see Table S9).

Analysis precision was assessed by replicate measurements for some samples. The average coefficient of variation for replicate measurements on 20 soil samples ranged from 1.7% for Al and Mn to 4.6% for Ce, with a minimum of 0.0% for Al and a maximum of 8.5% for Ce (see Table S10). The average coefficient of variation for replicate measurements on 28 feces samples ranged from 4.6% for Al and V to 6.6% for Sc, with a minimum of 0.0% for Y and maximum of 22.4% for Y (see Table S11). The average coefficient of variation for replicate measurements on 39 urine samples ranged from 2.9% for Ba to 13.9% for Y, with a minimum of 0.0% for Al and a maximum of 47.1% for Sc (see Table S12). The average coefficient of variation for replicate measurements on 15 drinking-water samples ranged from 3.1% for V to 8.6% for Ti, with a minimum of 0.0% for Ce and a maximum of 16.6% for Ce (see Table S13). The average coefficient of variation for replicate measurements on 31 food samples ranged from 9.8% for Mn to 19.2% for Ce, with a minimum of 0.2% for Mn and a maximum of 36.9% for Ce (see Table S14). The concentrations of Sc and V in food were usually lower than their detection limits.

All participants provided their written informed consent to engage in the SIR protocols. This study was approved by the Ethics Committee, Institute for Environmental Health and Related Product Safety, Chinese Centers for Disease Control and Prevention.

Table 1. Estimated daily soil ingestion rate (parameter, mg/d) (pre-processed original data) based on Al, Ba, Ce, Mn, Sc, Ti, V, and Y for 177 children.

Parameter	Al	Ba	Ce	Mn	Sc	Ti	V	Y
Max	3205.0	1125.1	1502.8	4040.4	1321.6	2977.2	1335.6	1922.3
99.5%	1881.8	1040.8	838.2	3670.6	1320.5	2372.1	830.0	1220.2
97.5%	780.1	728.4	610.4	2360.9	643.9	1706.2	515.4	597.1
95.0%	426.1	545.1	342.7	2055.2	496.5	1143.3	382.7	528.5
90.0%	214.7	356.9	175.7	1460.9	269.0	735.8	280.1	256.8
75.0%	101.0	160.6	103.1	698.7	135.0	260.5	158.5	132.5
Median	33.7	52.6	39.7	162.8	62.3	51.2	100.6	68.7
25.0%	8.8	−27.3	18.2	−132.5	28.5	−25.0	59.8	29.6
10.0%	−3.4	−62.9	5.3	−500.1	13.6	−81.3	35.8	14.1
5.0%	33.7	52.6	39.7	162.8	62.3	51.2	100.6	68.7
2.5%	−14.6	−115.2	−2.9	−1128.3	7.9	−171.6	20.3	2.7
0.5%	−28.8	−202.2	−14.7	−1945.3	2.1	−227.3	14.4	−78.5
Min	−42.0	−230.3	−21.2	−3514.8	−4.5	−233.7	10.4	−518.8
Mean	115.1	106.2	91.8	332.2	124.4	215.0	139.0	125.3
SD	313.0	207.7	167.1	906.8	192.2	468.1	147.2	215.2
CV%	271.9	195.5	181.9	272.9	154.4	217.7	105.9	171.8
n	177	177	177	177	177	177	177	177

Results

Soil Ingestion Rate

The SIR for each child was calculated for 177 children with Equation 1 using tracers Al, Ba, Ce, Mn, Sc, Ti, V, and Y separately (Table 1). SIR means were higher than medians for all eight tracers. The ratios of SIR mean to median ranged from 1.4 for V to 4.2 for Ti. This shows that the distribution of SIR based on each tracer was highly skewed. In addition, the SIR coefficient of variation based on each tracer was high, ranging from 105.9% for V to 272.9% for Mn. Frequency distribution histograms show some outliers, predominantly on the high-value end (Figure 2a and Figure S3). In short, SIR data based on each tracer element were characterized by a skewed distribution, high variation, and some outliers.

Statistically Processed Soil Ingestion Rate

It is generally assumed that outliers are due to experimental errors such as transit time misalignment, measurement error, and source error (Doyle et al. 2010). On the other hand, pica behavior in children might also lead to high outliers (U.S. EPA 2011). To obtain a statistically representative SIR for the large typical portion of the population of children, high outliers, some of which may have been from children who had pica, were removed (in Figure 2a for Al and Figure S3) per outlier box plots. The remaining SIR values are shown in Figure 2b for Al and in Figure S4. Although the remaining data included some new outliers, these new outliers are close to the "whiskers" and may represent high SIR for children, and thus are retained in the data set. According to mass-balance theory, SIR cannot be negative. However, the negative values obtained are not outliers according to the frequency distribution histograms, so were not removed. If these negative SIR values were removed as outliers, the SIR for a population of

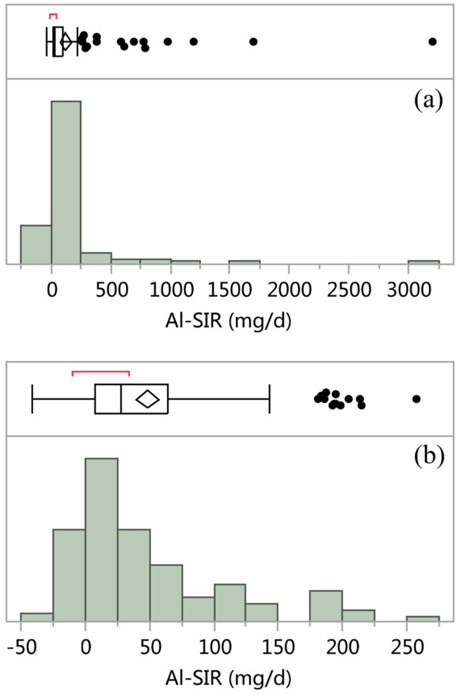


Figure 2. Frequency distribution histogram and outlier box plot of soil ingestion rate (SIR) based on tracer Al: (*a*) all SIR data and (*b*) remaining SIR data after removing outliers in (*a*). The vertical line within the box represents the median sample value. The ends of the box represent the 25th and 75th quantiles. The box has lines that extend from each end, sometimes called whiskers. The whiskers extend from the ends of the box to the outermost data point that falls within the distances computed as follows: first quartile $-1.5*$ (interquartile range) and third quartile $+1.5*$ (interquartile range). If the data points do not reach the computed ranges, then the whiskers are determined by the upper and lower data point values (not including outliers). The data points not in the range of the whiskers are considered outliers.

children might be overestimated. Basic parameters of these statistically processed SIR are shown in Table 2. It should be noted that the frequency distributions of SIR after removing outliers are still skewed; thus, median values may more accurately represent the central tendency of the SIR for a population of children than mean values. However, mean SIR values were also calculated and are shown in Table 2 to provide comparisons with previous studies (Calabrese et al. 1989; 1996; 1997; Davis et al. 1990; Doyle et al. 2012a; Irvine et al. 2014). SIR values for a population of Chinese children are recommended according to median and 95th percentile values.

The median (minimum to maximum) SIR was 27.8 (-42.0 to 257.4) mg/d for Al, 36.5 (-230.3 to 412.7) mg/d for Ba, 35.3 (-21.2 to 225.8) mg/d for Ce, 146.6 (-1259.4 to 1827.7) mg/d for Mn, 54.8 (-4.5 to 292.0) mg/d for Sc, 36.7 (-233.7 to 687.0) mg/d for Ti, 92.1 (10.4 to 308.0) mg/d for V, and 59.1 (-18.4 to 283.0) mg/d for Y. The process of removing outliers decreased the SIR coefficient of variation for Al, Ce, Sc, V, and Y, but only slightly changed it for Ba, Mn, and Ti due to a roughly synchronous decrease in mean values and relative standard deviations. Nevertheless, the process of removing outliers shortened the ranges between the minimum and maximum values as well as the differences between the median and mean values.

Soil Ingestion Rate Based on the Best Tracer Method (BTM)

Soil ingestion estimates are presented in a tracer-specific manner. As each tracer estimates the same soil ingestion behavior, high inter-tracer consistency in soil ingestion values would be expected

Table 2. Statistically processed daily soil ingestion rate (parameter, mg/d) based on Al, Ba, Ce, Mn, Sc, Ti, V, and Y for 177 children.

Parameter	Al	Ba	Ce	Mn	Sc	Ti	V	Y
Max	257.3	412.7	225.8	1827.7	292.0	687.0	308.0	283.0
99.5%	223.4	411.7	220.0	1807.1	288.5	628.9	303.2	266.5
97.5%	199.1	373.3	180.1	1686.8	260.1	542.5	270.6	249.7
95.0%	192.2	308.8	161.6	1434.2	221.2	473.4	224.2	238.5
90.0%	136.7	218.8	131.8	1083.9	180.7	343.5	193.6	173.9
75.0%	63.6	137.7	85.4	537.9	114.0	145.3	143.8	110.6
Median	27.8	36.5	35.3	146.6	54.8	36.7	92.1	59.1
25.0%	7.4	−29.6	16.9	−138.1	26.9	−29.9	56.6	28.0
10.0%	−4.8	−63.1	4.9	−479.3	11.9	−89.3	34.1	13.5
5.0%	−9.7	−101.2	0.6	−625.2	8.9	−116.9	24.3	7.2
2.5%	−15.1	−115.3	−3.6	−878.6	7.9	−174.2	19.6	3.1
0.5%	−30.0	−204.1	−15.2	−1157.3	1.6	−228.0	14.1	−11.3
Min	−42.0	−230.3	−21.2	−1259.4	−4.5	−233.7	10.4	−18.4
Mean	47.7	63.1	55.4	230.8	77.7	81.9	106.4	79.8
SD	59.8	125.9	52.0	617.6	68.8	177.6	64.6	68.3
CV%	125.3	199.4	93.8	267.6	88.5	216.8	60.7	85.7
Range	299.4	643.0	247.0	3087.1	296.5	920.7	297.6	301.4
n	161	165	165	163	162	158	163	161

if each tracer reliably estimated soil ingestion (Calabrese et al. 1997). However, the results in Table 2 display relatively high inter-tracer variability in soil ingestion estimations. This variability could be caused by transit time misalignment, measurement error, or source error (Doyle et al. 2010). The lack of inter-tracer consistency presents a significant problem concerning interpretation of accurate soil ingestion. BTM has been used to improve the accuracy of soil ingestion estimates (Calabrese and Stanek 1993; Calabrese et al. 1997; Stanek and Calabrese 1995). For BTM, the F/S ratio was used to identify suitable tracer elements. A low F/S ratio generally shows high suitability of a tracer element. The average F/S ratios in this study were 0.002 for V, 0.005 for Y, 0.006 for Sc, 0.008 for Ce, 0.017 for Al, 0.088 for Ti, 0.122 for Ba, and 0.431 for Mn. Therefore, V, Y, Sc, Ce, and Al were more suitable tracer elements than Ti, Ba, and Mn. However, estimates of soil ingestion based on these five tracer elements were still different from one another (Figure 3). The mean SIR for V was significantly higher than that for Y, Sc, Ce, and Al; the mean SIRs for Y and Sc were significantly higher than those for Ce and Al. However, the differences in SIRs between Y and Sc and between Ce and Al were not statistically significant.

The best estimate for SIR was calculated for each subject by taking the median of the best five tracer element (V, Y, Sc, Ce, and Al) estimates. Figure 4. shows the frequency distribution and basic statistical parameters of SIR based on the best tracer method. The frequency distribution for the best estimated SIR is also skewed. The SIR range for 24 children investigated for 3 consecutive d was 3.4–132.5 mg/d, similar to the control (15.3–144.0 mg/d for 16 children investigated for 1 d at the same site). The best estimate of the SIR for 177 Chinese children ranged from -9.5 to 297.6 mg/d, with median, mean, and 95th percentile values of 51.7, 73.5, and 216.6 mg/d, respectively. Reference intervals (RIs) are those values that lie between the lower (LRL) and upper (URL) reference limits (CLSI/IFCC Guideline C28-A3; CLSI 2008). The LRL is defined as the lower limit of the 90% confidence interval (CI) of the 2.5 percentile (P2.5), whereas the URL is defined as the upper limit of the 90% CI of the 97.5 percentile (P97.5) (CLSI 2008). The physical lower limit of the 90% CI of P2.5 for the SIR of Chinese children is assumed to be 0 mg/d, whereas its statistical estimation is -5.7 mg/d. Therefore, the SIR RI for Chinese children is 0–253.0 mg/d.

Discussion

Comparisons among soil ingestion studies are shown in Table 3. Al-based SIR means for various studies range from 2.0 mg/d

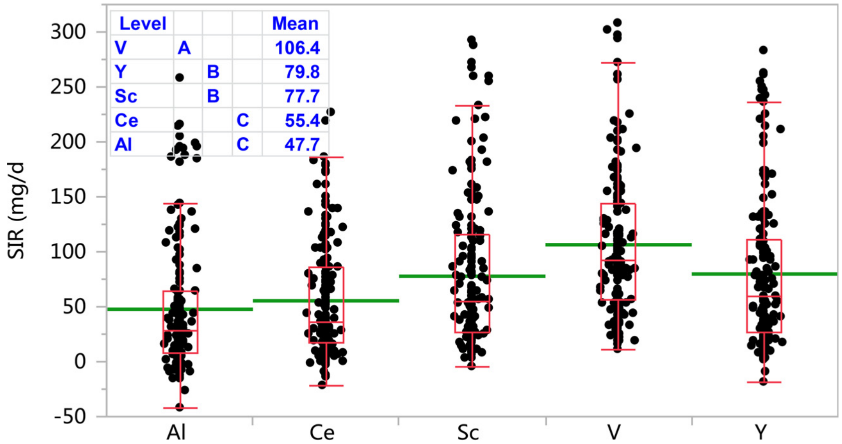


Figure 3. Comparisons of estimated soil ingestion rates based on Al, Ce, Sc, V, and Y. The outlier box plot, mean, and connecting letters report [mean values that are not sharing a letter (i.e., A, B, or C) are significantly different at the $p = 0.05$ level] are shown in the figure.

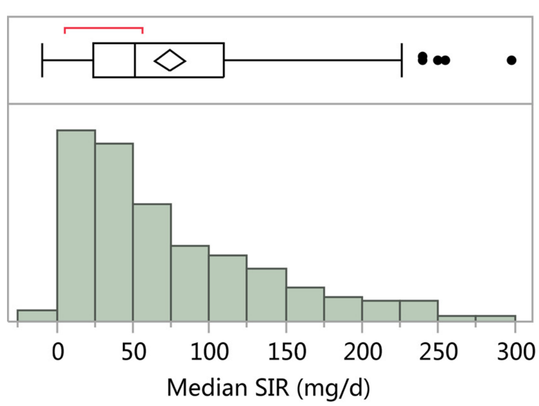
(Calabrese et al. 1996) to 154.0 mg/d (Calabrese et al. 1989), whereas Al-based SIR medians range from −3.3 mg/d (Calabrese et al. 1997) to 33.3 mg/d (Doyle et al. 2012b). The Al-based SIR minimum for various studies is −279 mg/d (Davis et al. 1990), whereas the Al-based maximum value is 4929.0 mg/d (Calabrese et al. 1989). The mean and median values for our Al-based SIR are similar to those observed by Davis et al. (1990) and Davis and Mirick (2006).

Ti-based SIR means for various studies range from −544.4 mg/d (Calabrese et al. 1997) to 3215.0 mg/d (Irvine et al. 2014). However, Ti-based SIR medians range from 3.0 mg/d (Calabrese et al. 1996) to 759.0 mg/d (Irvine et al. 2014). The Ti-based SIR minimum from other studies is −15,736 mg/d (Calabrese et al. 1997), whereas the Ti-based SIR maximum is 16459.0 mg/d (Irvine et al. 2014). Our Ti-based SIR median is similar to that found by Calabrese et al. (1989). Ti has not been found to be a very reliable tracer.

Ba- and Mn-based SIR means and medians observed in our study are higher than those observed by Calabrese et al. (1989), but much lower than those in Irvine et al. (2014). Our Ce-, V-, and Y-based SIR means and medians are similar to those found by Calabrese et al. (1989; 1996). The BTM-based SIR mean and median for our study (73.5 and 51.7 mg/d) are higher than those found by Calabrese et al. (1997; 6.8 and −2.4 mg/d) and are similar to those observed by Doyle et al. (2012b; 74.9 and 50.0 mg/d).

Using results from four mass-balance soil ingestion studies (Calabrese et al. 1989; Calabrese et al. 1997; Davis et al. 1990; Davis and Mirick 2006), Stanek et al. (2012b) developed a stochastic model to estimate the accuracy of soil ingestion estimators for different tracer elements. Their results suggest that soil ingestion estimates based on Ti, Ba, and Mn are unreliable and of limited value in estimating soil ingestion. In addition, a meta-analysis by Stanek et al. (2012a) of SIR values obtained from data on 216 children using Al and Si as tracer elements were 25.5 and 32.6 mg/d for the mean and median values; whereas the 95th percentile was estimated as 79.4 mg/d (Table 3).

Recently, several studies have used other methodologies to estimate SIRs in children based on the time/activity pattern modeling approach. Özkaynak et al. (2011) used a specific version of the Stochastic Human Exposure and Dose Simulation Model for multimedia pollutants (SHEDS Multimedia) to estimate soil and dust ingestion exposures for young children between 3 and 6 y old. The study showed mean and 95th percentile total ingestion rates of soil and dust of 68 and 224 mg/d, respectively. Soil and dust ingestion rates were calculated using the following measures: particle loading to indoor surfaces, fraction transferred to the hands, hand surface area, fraction of hand surface area that may be mouthed, frequency of hand-to-mouth events, amount dissolved by saliva, and exposure time. Another mechanistic time–activity pattern-based modeling approach by Wilson et al. (2013) showed mean (95% percentile) total ingestion rates of



Parameter	SIR	95% CI
Max	**297.6**	
99.5%	261.2	
97.5%	239.5	215.8-253.0
95.0%	216.6	
90.0%	163.3	
75.0%	108.9	
Median	**51.7**	
25.0%	24.6	
10.0%	12.5	
5.0%	7.5	
2.5%	3.7	-5.7-7.5
0.5%	-8.0	
Min	**-9.5**	
Mean	**73.5**	
SD	63.7	
CV%	86.6	
Range	307.1	
n	169	

Figure 4. Frequency distribution histogram and basic statistical parameters of SIR based on the best tracer method (the median of the best five tracer elements (V, Y, Sc, Ce, and Al) estimates). 95% confidence interval (CI) of 97.5 and 2.5 percentiles are listed.

Table 3. Soil ingestion rate (SIR) comparisons for previous studies and this study.

Tracer element	Age (y)	Mean (mg/d)	Median (mg/d)	SD (mg/d)	Minimum (mg/d)	Maximum (mg/d)	n	Reference
Al	1–4	154.0	30.0	629.0		4929.0	64	Calabrese et al. 1989
Al	2–7	38.9	25.3		−279.0	904.5	101	Davis et al. 1990
Al	3–7	36.7	33.3			107.9	19	Davis and Mirick 2006
Al	1–4	2.7	−3.3	95.8	−202.8	461.1	64	Calabrese et al. 1997
Al	1–4	2.0	0.0	14.0	−46.0	58.0	62	Calabrese et al. 1996
Al	2.5–11.9	47.7	27.8	59.8	−42.0	257.3	161	This study
Al	2.5–6.8	45.6	24.1	61.7	−27.0	257.3	96	This study
Al	Adult	36.9	31.0	51.9		177.0	43	Doyle et al. 2012b
Al	Adult	36.0	7.0	117.0		268.0	87	Irvine et al. 2014
Ti	1–4	170.0	30.0	691.0		3597.0	64	Calabrese et al. 1989
Ti	2–7	245.5	81.3		−5820.8	6182.7	101	Davis et al. 1990
Ti	3–7	206.9	46.7			808.3	19	Davis and Mirick 2006
Ti	1–4	−544.4	11.9	2509.0	−15736.0	4066.6	64	Calabrese et al. 1997
Ti	1–4	6.0	3.0	442.0	−2156.0	1442.0	62	Calabrese et al. 1996
Ti	2.5–11.9	81.9	36.7	177.6	−233.7	687.0	158	This study
Ti	2.5–6.8	72.1	32.3	176.5	−233.7	687.0	95	This study
Ti	Adult	3215.0	759.0	5622.0		16459.0	87	Irvine et al. 2014
Ba	1–4	29.0	−19.0	868.0		5626.0	64	Calabrese et al. 1989
Ba	2.5–11.9	63.1	36.5	125.9	−230.3	412.7	165	This study
Ba	2.5–6.8	42.6	17.7	113.1	−198.4	353.7	96	This study
Ba	Adult	318.0	467.0	1622.0		2405.0	87	Irvine et al. 2014
Ce	1–4	75.0	28.0	121.0	−111.0	597.0	62	Calabrese et al. 1996
Ce	2.5–11.9	53.5	34.8	48.8	−21.2	186.0	163	This study
Ce	2.5–6.8	45.4	29.0	47.1	−21.2	186.0	98	This study
Ce	Adult	72.2	51.0	179.5		516.0	43	Doyle et al. 2012b
Ce	Adult	12.0	−4.0	72.0		132.0	87	Irvine et al. 2014
La	1 − 4	8.6	84.5	1377.2	−10673.0	1089.7	64	Calabrese et al. 1997
La	1–4	−16.0	39.0	946.0	−7249.0	626.0	62	Calabrese et al. 1996
La	Adult	132.6	104.0	158.6		683.0	43	Doyle et al. 2012b
La	Adult	12.0	−2.0	78.0		156.0	87	Irvine et al. 2014
Mn	1–4	−496.0	−340.0	1974.0		4189.0	64	Calabrese et al. 1989
Mn	2.5–11.9	230.8	146.6	617.6	−1259.4	1827.7	163	This study
Mn	2.5–6.8	118.7	70.0	534.5	−1259.4	1827.7	98	This study
Mn	Adult	1998.0	1034.0	10107.0		18226.0	87	Irvine et al. 2014
Sc	2.5–11.9	77.7	54.8	68.8	−4.5	292.0	162	This study
Sc	2.5–6.8	70.4	40.8	70.2	−4.5	287.7	98	This study
V	1–4	456.0	123.0	1013.0		6736.0	62	Calabrese et al. 1989
V	2.5–11.9	106.4	92.1	64.6	10.4	308.0	163	This study
V	2.5–6.8	91.4	84.2	54.2	10.4	302.0	95	This study
V	Adult	−183.0	−185.0	238.0		169.0	87	Irvine et al. 2014
Y	1–4	65.0	11.0	717.0		5269.0	62	Calabrese et al. 1989
Y	2.5–11.9	79.8	59.1	68.3	−18.4	283.0	161	
Y	2.5–6.8	65.2	47.1	60.4	−18.4	260.6	96	This study
Y	Adult	−7.0	−17.0	145.0		230.0	87	Irvine et al. 2014
BTM	1–4	6.8	−2.4	74.5	−101.3	380.2	64	Calabrese et al. 1997
BTM	2.5–11.9	73.5	51.7	63.7	−9.5	297.6	169	This study
BTM	2.5–11.9	60.8	41.4	57.0	−9.5	254.3	99	This study
Al, Si	1– < 8	25.5	32.6				216	Stanek et al. 2012a
Al, Ce, La, Si	Adult	74.9	50.0	119.5		683.0	159	Doyle et al. 2012b
Al, Ce, La, Si	Adult	32.0	18.0	88.0		152.0	261	Irvine et al. 2014

soil and dust of 61 (204) mg/d for 7-mo- to 4-y-old children and 55 (185) mg/d for children 5–11 y of age. In addition, von Lindern et al. (2016) estimated children's soil and dust ingestion rates using the lead biokinetic modeling method. They concluded that soil and dust ingestion rates at the Bunker Hill Superfund Site in Idaho averaged 66 mg/d (95% CI: 57, 75 mg/d) for children 6 mo–9 y old. SIRs from the studies above are broadly similar to our results. Nevertheless, children's mouthing behaviors may differ between different countries or populations with different ethnic or lifestyle characteristics (Tsou et al. 2015), thereby influencing children's SIRs between different countries or populations. As shown by Chien et al. (2017), the SIR for children under 3 years of age in Taiwan based on the silicon (Si) tracer mass-balance technique showed SIRs to be lower than values found in the United States.

The recommended values for child daily soil and dust ingestion by the U.S. EPA are 100 mg/d (central tendency) and 200 mg/d (95th percentile) (U.S. EPA 2011). The BTM-based SIR mean, median and 95th percentile values for Chinese children are approximately 73.5, 51.7, and 216.6 mg/d, respectively.

Our study has some limitations. We did not distinguish whether and to what extent the residual fecal tracers (excluding tracer amounts from food and drinking water) were of outdoor soil or indoor dust origin. Previous studies have indicated that the ratio of the average concentration of tracer elements in outdoor soil to that in indoor soil was approximately 2 to 3 (Calabrese et al. 1997; Davis et al. 1990). Additionally, approximately 50% of residual fecal tracers were estimated to be of outdoor soil origin (Stanek and Calabrese 1992). Considering the low concentration of tracer elements in indoor dust and the high proportion of children's waking time spent indoors, the actual soil (outdoor soil and indoor dust) ingestion rate would increase by 1.5–2 times after including dust values (Davis et al. 1990). Limitations related to the mass-balance method itself include transit time misalignment, measure errors, and source errors. In addition, the 1-d investigation period for most participants may have led to relatively high misalignment of transit time. Moreover, SIRs for the 1-d snapshot may not completely represent longitudinal SIRs over a year. Swallowing toothpaste was not considered because of a lack of data on amount swallowed, although toothpaste contains some tracer elements. The median concentration of tracers in soil for each city was used because of the high heterogeneity of soil. BTM-based SIR estimates statistically overcame the high inter-tracer inconsistency or variability of SIRs based on each tracer SIR estimate. The reasons for this need to be investigated further. Si, frequently used as a tracer in previous studies, was not included in this study because its measurement method is different from that of other tracers. Although SIR data estimated by each tracer and BTM were skewed, mean SIR values were calculated in order to provide comparison with previous studies. In addition, SIR values should not be negative, but some estimated negative SIR values were not statistically removed. Finally, this study did not differentiate between outside soil ingestion and indoor dust ingestion, and the number of children in this study was small relative to the Chinese population.

Conclusions

To the best of our knowledge, this paper presents the first large-scale study of SIR for 177 Chinese children (153 for 1 d and 24 for 3 consecutive d) from Guangdong, Hubei, and Gansu Provinces, employing the tracer mass-balance method. Estimated SIR values were characterized by relatively high inter-subject and inter-tracer variation, mainly due to transit time misalignment, source error, or measurement error. Al, Ce, Sc, V, and Y were more suitable tracer elements than Ba, Mn, and Ti due to their low F/S ratios. The estimated SIR median and 95th percentile values were 27.8 and 192.2 mg/d for Al, 35.3 and 161.6 mg/d for Ce, 54.8 and 221.2 mg/d for Sc, 59.1 and 238.5 mg/d for Y, and 92.1 and 238.5 mg/d for V. BTM-based SIR median, mean, and 95th percentile values were 51.7, 73.5, and 216.2 mg/d, respectively. In addition, SIR RI for a population of Chinese children was 0–253 mg/d. Our estimates of Chinese children's SIR are similar to those of previous studies and also the value recommended by the U.S. EPA, but with low inter-tracer and inter-subject variation. This investigation of soil ingestion over 1 d on a relatively large child population obtained similar statistical estimates of children's SIR to other studies. Nonetheless, longitudinal SIRs over multiple time periods warrant further investigation.

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